Thermodynamics of the Hard Tetrahedron System

by

Amir Haji Akbari Balou

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Doctoral Committee:

Professor Sharon C. Glotzer, Chair
Professor Jeffrey Lagarias
Professor Michael J. Solomon
Professor Robert M. Ziff

To my parents, Zhila and Zakarya

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LIST OF ABBREVIATIONS

app approximant

bcc body-centered cubic

DA degenerate approximant

DC degenerate crystal

DQC degenerate quasicrystal

EOS equation of state

fcc face-centered cubic

FEP Free Energy Perturbation

FK Frank-Kasper

hcp hexagonally close packed

HS hard sphere

ico icosahedron

MC Monte Carlo

MD Molecular Dynamics

NLP nonlinear programming

NP nanoparticle

OOP orientational order parameter

PD pentagonal dipyramid

PDC periodic divide and concur

PDMS poly(dimethylsiloxane)

PMMA poly(methyl methacrylate

PRINT Particle Replication in Nonwetting Templates

QC quasicrystal

rdf radial distribution function

SOC strong orientational coordinate

TBP triangular bipyramid

TCP tetrahedrally close packed

TI thermodynamic integration

tTBP truncated triangular bipyramid

UV ultraviolet

ABSTRACT

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Chair: Sharon C Glotzer

The self-assembly of nanoparticles into ordered structures is governed by interaction and shape anisotropy. Recent advancements in the synthesis of faceted nanoparticles and colloids have spurred interest in the exclusive effect of shape anisotropy, which manifests itself in phase behavior of polyhedral shapes. Among them, hard regular tetrahedra have attracted particular attention because they prefer local symmetries that are incompatible with periodicity. We study their thermodynamics using Monte Carlo simulations and observe that they self-assemble into a a dodecagonal quasicrystal, which is the first reported quasicrystal in hard particle systems. The quasicrystal and its approximants pack very efficiently and can be compressed to unusually high packing fractions. The densest quasicrystal-like phase is the $(3.4.3^2.4)$ approximant, which can be compressed to a packing fraction of 85.03%. Using free energy calculations, we confirm that this approximant is more stable than the densest known packing of regular tetrahedra, a simple triclinic crystal with four particles in a unit cell, at intermediate packing fractions. The solid-solid transition from the approximant to the dimer crystal occurs at extremely high packing fractions, and involves no symmetry breaking, which is unusual in hard particle systems. The superior stability

of the approximant at intermediate densities can be partly attributed to correlated motion of particles that gives it some 'fluid-like' character. We also show that the quasicrystal is robust to building block polydispersity and forms at polydisprsities as large as 12%.

We also study the thermodynamics of hard triangular bipyramids– i.e. dimers of tetrahedra– and observe the formation of a degenerate quasicrystal at $\phi \geq 54\%$. The quasicrystal is similar to the quasicrystal formed by hard tetrahedra in the monomer level but degenerate in pairing of tetrahedra into dimers. This pairing degeneracy has never been observed for any quasicrystal before, and should not be confused with the well-known notion of degeneracy that is associated with random tilings and phason flips. We also construct the $(3.4.3^2.4)$ approximant of the degenerate quasicrystal and compare it with the densest packing of dimers using free energy calculations. Like the hard tetrahedron system, the quasicrystal approximant is more stable at densities below 79.7%.

We also study the thermodynamics and dense packings of hard truncated triangular bipyramids and observe that the degenerate quasicrystal forms for truncations as high as 0.4. No other ordered structure forms in simulations of systems with larger truncations except for a stretched hexagonally close packed phase that self-assembles in systems with truncations $\frac{2}{3}$ to 0.80. In isobaric simulations of small systems– i.e. systems with as many as four particles in a box– we observe eight distinct families of dense packings with two to four particles in the fundamental domain. Only one of them forms in self-assembly simulations i.e. for truncations $\frac{2}{3} - 0.8$.

Inspired by the possibility that hard tetrahedra form liquid crystalline phases as most other hard particles do, we propose a simplified procedure for deriving suitable order parameters for identifying and quantifying orientationally ordered structures of particles with nontrivial symmetries. This procedure is based on identifying orbits of a particle and forming a suitable symmetric tensor from them. There is an

isomorphism between these symmetric tensors- which we call strong orientational co-ordinates (SOCs)- and distinct orientations of the symmetric particle. Orientationally ordered structures are then described as idealized probability distributions in terms of SOCs and the quantification of order is formulated as a constrained nonlinear opti-mization problem. These SOCs can also be used to average rotational fluctuations in inherent structure calculations as well as to quantify temporal and spatial correlations of orientational order.

CHAPTER I

Introduction

What are the building blocks of the universe and how do they come together to make different forms of matter? These are among the most fundamental questions that have intrigued humanity for millennia, and yet they are far from being fully addressed despite all the groundbreaking advances in the natural sciences. Some prominent Greek philosophers such as *Plato* and *Democritus* believed that matter is composed of discrete indivisible entities i.e. atoms, but it was not until the early nineteenth century that the atomic theory of matter was fully confirmed in experiment. Yet, this knowledge was far from sufficient for making sense of different states of matter, their physical properties and how and why they transform into one another. Thermodynamics and statistical mechanics were founded by giants such as *Gibbs* and *Boltzmann* partly to address these questions.

Statistical mechanics was a huge success in bridging the microscopic world of atoms and molecules to the macroscopic forms of matter. But like any other successful endeavor in science, it posed more questions than it managed to answer. In the early years of statistical mechanics, van der Wáals produced an accurate equation of state for gaseous and liquid states of matter by using a very simple (nonspecific) attractive potential [1]. This led some to conjecture that the types of phases formed by a particular system hardly depend on the details of the inter-particle potential, and are instead determined by its general features such as the range and anisotropy of interaction and depths and number of minima in the potential. If this is indeed true, it is natural to ask:

1. What types of structures can arise from a given intermolecular potential?

2. What is the simplest potential that can give rise to a particular structure?

The history of theoretical and computational statistical mechanics is full of trial-and-error-based attempts to answer these questions by exploring different types of

potentials and structures, yet a comprehensive theoretical framework for addressing these questions is just being envisioned [2].

These questions are of more practical relevance today thanks to breakthroughs in colloidal sciences and nanotechnology that has made possible the synthesis of a wide range of nano- and colloidal particles of different shapes, sizes and surface patterns [3–7]. The level of control that can be exerted on the way these particles interact (e.g. range, directionality and specificity of interaction) is also unprecedented and cannot be easily achieved at the Angstrom scales of atomic and molecular systems. Geometric and interactional anisotropy of these new building blocks makes them suitable for assembling complicated structures with interesting mechanical, optical and electric properties [8].

The types of phases that could emerge in a system not only depends on features of the inter-particle potential, but also on the shape of the building blocks. Hard particles are suitable model systems for investigating the exclusive effect of building block geometry on phase behavior. For a long time it was believed that hard particles are not capable of forming any ordered structure since a decrease in entropy due to ordering cannot be compensated by a decrease in internal energy due to the absence of attractive interactions. It is therefore not surprising that Onsager's prediction of an isotropic-to-nematic transition in a system of infinitely thin hard rods [9], and Kirkwood's prediction of hard sphere crystallization [10] were calls for controversy at their times; this issue was not settled until these phenomena were observed in simulation [11–13] and even then there were skeptics. Since then, several ordered phases have been observed in computational studies of several hard particle systems [14–21].

Studying hard particle systems is also interesting because of the intimate relationship between the question of thermodynamic stability in hard particle systems and the problem of packing in geometry. It follows from simple thermodynamic reasoning that an arrangement of hard particles occupying the largest fraction of space is thermodynamically favored in the limit of infinite pressure. Solving the packing problem for a given building block can therefore partially elucidate the phase diagram of the corresponding hard particle system. In the meantime, thermodynamic simulations of hard particles can give mathematicians hints on how to solve the otherwise difficult packing problems.

The packing problem has dazzled the simplest and the most complicated minds alike since the dawn of civilization. From the peasants trying to fit as many apples in a box to great thinkers of all ages have attacked this problem from different perspectives. Written accounts of studies of the face-centered cubic (f.c.c.) packings of spheres

can be found in Sanskrit works of 499 BCE [22]. Plato associated the five basic elements of universe (earth, water, air, fire and cosmos) with the five regular polyhedra i.e. Platonic solids that, according to him, pack together to form different forms of matter [23]. As early as the seventeenth century, prominent thinkers such as Kepler and Newton were in pursuit of the densest arrangements of congruent spheres. It is however a prohibitively nontrivial task to solve packing problems for objects not tiling the Euclidean space. The seemingly intuitive Kepler's conjecture that an f.c.c. arrangement of spheres is the densest was only proven recently [24]. Even the simpler case of the hexagonal lattice packing of disks in two dimensions was not proven to be optimal until 1892 [25]. It is thus not surprising that the problem of identifying the densest packings of solids was included as a part of Hilbert's 18th problem posed in the beginning of the 20th century [26]. In the wake of resurgent interest in hard particle systems, several unresolved packing problems are currently revisited mostly for convex shapes such as ellipsoids, cylinders and tetrahedra [18, 23, 27–34]. No mathematical theory exists for studying packings of concave objects.

The regular tetrahedron is the simplest of the five Platonic solids, and the only Platonic solid that lacks central symmetry. Plato associated it with the element *fire* and his student Aristotle erroneously thought that it tiles the Euclidean space [23]. It almost took eighteen centuries for this assertion to be proven untrue [35]. Despite a few studies of lattice packings of regular tetrahedra in the 1960's [36, 37], the problem received little attention until recently when a number of surprisingly dense packinsg of tetrahedra were introduced in a relatively short period of time [18, 27–32].

This drama started in 2006 with a paper by Conway and Torquato [27], who constructed dense packings of tetrahedra with packing fractions as high 72%. Their failure to surpass the sphere packing fraction of $\pi/\sqrt{18} \approx 74.04\%$ led them to question *Ulam's conjecture* that all convex objects in $\mathbb{R}^3$ pack better than spheres. Their speculation was debunked by Chen in 2008 [28] after she analytically constructed a crystal with a 77.86% packing fraction.

In 2009, we observed that hard tetrahedra can spontaneously assemble into a dodecagonal quasicrystal, which could be compressed to a packing fraction of 83.24%. We also constructed a periodic approximant of the quasicrystal and compressed it to a packing density of 85.03% [18]. The last two months of 2009 were the climax of this drama, and the competition would become so intense that records would change in a matter of days. After the discovery of the simple 'dimer crystal' by Kallus [30], the densest known packing of tetrahedra was finally introduced by Chen *et al.* [31], who obtained it by generalizing and optimizing the structure discovered by Kallus.

The whole drama was chronicled in an article entitled "Packing Tetrahedrons, and Closing In on a Perfect Fit" in *The New York Times* on January 4, 2010.

This thesis is devoted to the study of the thermodynamics of hard regular tetrahedra and related systems using Monte Carlo simulations and free energy calculations, and is organized as follows. In Chapter II a thorough overview of the relevant literature is presented. The basics and implementation details of the simulation methods used in this dissertation are presented in Chapter III. Chapter IV outlines a general scheme for identifying and quantifying orientational order in systems of symmetric building blocks including tetrahedra. We present our self-assembly simulations of the hard tetrahedron system, including the structural description of the quasicrystal, in Chapter V while the thermodynamics and the phase diagram of the hard tetrahedron system– including comparing the stability of the quasicrystal and the dimer crystal– is thoroughly covered in Chapter VI. In Chapter VII, the thermodynamics of a related system i.e. hard triangular bipyramids is thoroughly studied. Concluding remarks and an overview for further research is given in Chapter VIII.

CHAPTER II

Background

This chapter is organized as follows. In Section 2.1, we present a historic overview of the advancements in the synthesis of non-spherical nano- and colloidal particles and discuss the key techniques in their fabrication. Section 2.2 is devoted to the thermodynamics of hard particle systems. Key results in computational studies of non-spherical hard particles are reviewed in Section 2.2.1. An overview of the packing problem is presented in Section 2.3. The well-publicized example of the sphere packing problem and the Kepler conjecture is presented in Section 2.3.1 while Section 2.3.2 is reserved for an overview of the regular tetrahedron packing problem, which is the main subject of this dissertation.

2.1 Synthesis of Non-spherical Nano- and Colloidal Particles

Nanotechnology is as old as human history. Recent studies on the *Lycurgus Cup*[1], a glassy Roman artifact of 4th century AD, reveal that its exotic optical dichroism is due to the presence of gold and silver nanoparticles that have been unintentionally dissolved into its constituent glass [38]. The first scientific description of unusual properties of nanometer-sized particles is given by the British scientist Michael Faraday [39], who is considered as the founder of the colloidal sciences. Here is how he

---

[1]The Website of British museum reads: "This extraordinary cup is the only complete example of a very special type of glass, known as dichroic, which changes colour when held up to the light. The opaque green cup turns to a glowing translucent red when light is shone through it... The scene on the cup depicts an episode from the myth of *Lycurgus*, a king of the Thracians (around 800 BC). A man of violent temper, he attacked Dionysos and one of his maenads, Ambrosia. Ambrosia called out to Mother Earth, who transformed her into a vine. She then coiled herself about the king, and held him captive. The cup shows this moment when Lycurgus is entrapped by the branches of the vine, while Dionysos, a Pan and a satyr torment him for his evil behaviour. It has been thought that the theme of this myth - the triumph of Dionysos over Lycurgus - might have been chosen to refer to a contemporary .political event, the defeat of the emperor Licinius (reigned 308-24 AD) by Constantine in 324 AD."

describes the phenomenon that we now know as the *Tyndall effect* in his famous *Bakerian Lecture* [40] in 1857:

> "...*The latter, when in their finest state, often remain unchanged for many months, and have all the appearance of solutions. But they never are such, containing in fact no dissolved, but only diffused gold. The particles are easily rendered evident, by gathering the rays of the sun (or a lamp) into a cone by a lens, and sending the part of the cone near the focus into the fluid; the cone becomes visible, and though the illuminated particles cannot be distinguished because of their minuteness, yet the light they reflect is golden in character, and is seen to be abundant in proportion to the quantity of solid gold present.*"

Reports of nanoparticle synthesis can be traced back to as early as the 1960's [41]. However, the resulting spherically-shaped particles were of limited utility for self-assembly purposes as traditional spherical colloids are only capable of forming simple-symmetry phases such as face-centered cubic, hexagonally-closed-packed and body-centered cubic [2]. With the help of techniques such as surface templating, incorporation of bimodal size and charge distribution, and application of external fields, trigonal, tetragonal, simple cubic and ionic crystals can also be assembled by spherical building blocks [2, 42, 43]. In order to assemble more complicated structures, geometrical and/or interactional anisotropy of the building block is necessary.

Naturally occurring non-spherical colloids have been known for a long time. For example swelling clays (such as montmorillonite) are of discotic shape [2]. Accounts of the synthesis of artificial nonspherical nanoparticles can be found in an early work of Matijevic in 1976 where he reports the production of monodispersed colloidal dispersions of varying shapes including cubes, cube-octahedra, needles and ellipsoids [44]. By early 1990's, methods were in place for the synthesis of cubes, rods and discs from metals (such as gold and silver), and metal oxides (hematite, boehmite, and gibbsite) [2]. For instance, Ozaki *et al.* used forced hydrolysis of ferric chloride solutions at elevated temperatures to make spindle-type colloidal hematite particles [45]. Buining *et al.* made boehmite colloidal particle via hyrdrothermal treatment of an acidified aqueous alkoxide solution [46]. Keville *et al.* produced prolate micro-spheroids by embedding poly(methyl methacrylate) (PMMA) microspheres into a matrix of poly(dimethylsiloxane) (PDMS) and deforming the matrix under uniaxial extension [47]. The majority of these unusually-shaped particles were however of limited technological application due to low yields and insufficient shape and size selectivity [2].

The prospect of bottom-up assembly of complex structures from anisotropic building blocks spurred a resurgent interest in the field and a variety of physical, chemical and biosynthetic methods were developed in the last decade to allow for tighter control of particle size and geometry. In the selective crystallization and deposition methods, a class of solution-based bottom-up techniques that have become very popular recently, an initial crystallite, which is usually spherical, is formed through a chemical reaction. This crystallite is then preferentially grown along certain directions by the aid of templates, stabilizers or capping agents [48–51]. The arising anisotropic particles can be subsequently combined to make other anisotropic particles. For instance, nano-tetrapods can be obtained by growing rods in the [111] planes of zinc blend tetrahedron cadmium selenide seeds [48]. In top-down lithography-based methods such as selective deposition [52] and Particle Replication in Nonwetting Templates (PRINT) [53], a reactive substrate is selectively molded into the desired morphology using a beam of UV light or a nonwetting template respectively. A comprehensive discussion of different nanoparticle synthesis methods can be found in [54].

The emergence of these novel techniques has made possible the synthesis of nano- and colloidal particles of different shapes. The synthesized shapes include cubes [52, 55–61], rods [4, 56, 60, 62, 63], rings [52], dumbbells [58, 64, 65], decahedra [66–68], octahedra [57, 60, 61, 67, 69–72], tetrahedra [57, 67, 73–76], icosahedra [57, 67, 77–79], prisms [49, 80–85], disks [4, 86], triangular bipyramids [85, 87–89], stars [56, 90–92] and multipods [56, 92, 93]. These particles are made of a variety of materials ranging from noble metals [6] and metal oxides [5] to semiconductors [3] and polymers [4]. To this, one should add the hybrid nanoparticles that contain more than one chemical entity in their structure [7].

The availability of such a variety of nonspherical nano- and colloidal particles paves the way for assembling complex structures. In general, the individual nanoparticles can have opto-electrical, magnetic or catalytic properties that are different from the bulk material that they are made of, due to their large surface area [54, 94]. The structures assembled by these particles can in turn have interesting properties emanating from those of the individual building blocks. However, these structures can sometimes demonstrate collective physicochemical properties that are even different from those of the individual constituent nanoparticles [95]. Theoretical and experimental studies of self-assembly are further incentivized by these possibilities.

The phase behavior of these nano- and colloidal particles is not only determined by the range and directionality of interactions between them, but also by their shape. Hard core interactions are idealized models for studying the exclusive effect of the

shape of a building block on its phase behavior. We therefore follow by discussing the thermodynamics of hard particle systems and how it is related to the phase behavior of nano- and colloidal particles.

2.2   Thermodynamics of Hard Particles

For two arbitrary objects $\mathcal{R}_1$ and $\mathcal{R}_2$, the *hard potential* is defined by:

$$u_H(\mathcal{R}_1, \mathcal{R}_2) \quad = \quad \begin{cases} +\infty & \mathcal{R}_1 \cap \mathcal{R}_2 \neq \emptyset \\ 0 & \text{otherwise} \end{cases} \tag{2.1}$$

If $\mathcal{R}_1$ and $\mathcal{R}_2$ are congruent spheres of diameter σ, Eq. (2.1) takes the following form:

$$u_{HS}(r) \quad = \quad \begin{cases} +\infty & r < \sigma \\ 0 & r \geq \sigma \end{cases} \tag{2.2}$$

where r is the distance between the centroids of $\mathcal{R}_1$ and $\mathcal{R}_2$. Eq. (2.2) is called the *hard sphere potential*.

Hard core interactions are idealized representations of excluded volume effects that are prevalent in dense states of matter such as liquids and solids, and have been shown to approximate short-range repulsions [96]. In atomic and molecular systems, such repulsions arise when atoms and molecules get so close to one another that their electronic shells start to overlap and repel one another. That explains why hard particle systems, especially hard spheres, have for decades been such successful models of dense liquids and solids [11, 12, 97–99].

Likewise, hard core interactions can be used to explain the thermodynamic behavior of concentrated colloidal suspensions. What is remarkable however, is that colloidal particles can, under certain circumstances, behave like hard particles. For example, long-range electrostatic repulsions between charged nanoparticles can be screened by ionic solutions, effectively becoming short-range repulsions [100], which, as mentioned above, can be approximated with hard core interactions [96]. The qualitative phase behavior of such colloidal systems can therefore be predicted through theoretical and/or computational studies of the associated hard particle system.

For a long time, it was believed that hard particles cannot form ordered phases, as entropy was intuitively associated with 'lack of order'. A disorder-order transition was thus perceived to involve a decrease in entropy, which, due to lack of attractive interactions, could not be compensated by a decrease in a system's energy. This widely

accepted belief was first challenged by Onsager, who showed that, at sufficiently large number densities, an isotropic fluid of hard thin rods can undergo a weak first-order transition to a nematic liquid crystal [9]. Onsager made this remarkable- and rather counterintuitive- prediction by maximizing the virial expansion of the free energy functional:

$$
\begin{aligned}
\ln Q[f(\mathbf{a})] &= N_p \left[1 + \log \frac{V}{N_p} \right] - \int f(\mathbf{a}) \log[4\pi f(\mathbf{a})] d\Omega(\mathbf{a}) \\
&\quad + \frac{N_p}{2V} \int \beta_1 \left[\cos^{-1}(\mathbf{a}, \mathbf{a}') \right] f(\mathbf{a}) f(\mathbf{a}') d\Omega(\mathbf{a}) d\Omega(\mathbf{a}') \qquad (2.3)
\end{aligned}
$$

with respect to $f(\mathbf{a})$, the orientational distribution function, with the constraint that $\int f(\mathbf{a}) d\Omega = 1$. Here N_p is the number of particles in volume V and $\beta_1(\cdot)$ is the first cluster integral calculated for a fixed angle between the rods. It is argued that the higher-order cluster integrals can be neglected since $L \gg D$. Onsager showed that for sufficiently large $c = N_p/V$ there exists a non-uniform $f(\mathbf{a})$ that gives a higher value of $\ln Q$- and hence a lower value of the free energy- than $f_0 \equiv \frac{1}{4\pi}$, and such a non-uniform distribution function describes a nematic liquid crystal.

It took more than thirty years before Frenkel and Eppenga confirmed the existence of the isotropic-nematic (I-N) transition in hard particle systems in a computer simulation of hard thin platelets [13]. This phenomenon was experimentally observed in a colloidal suspension of plate-like particles in the late 1990's [101].

Perhaps the most well-known example of spontaneous ordering in hard particle systems is the freezing of hard spheres into an f.c.c. crystal. Kirkwood was the first to predict this, based on an approximate theoretical description of the hard sphere model [10]. This prediction was very controversial and met widespread skepticism. The issue was not settled until 1957 when Alder and Wainwright [11] and Wood and Jacobson [12] observed the freezing transition in their MC simulations of hard spheres. In recent decades, the transition has also been observed in experiments of colloidal hard spheres [102, 103].

These theoretical predictions were based on several approximations, but they could be qualitatively understood by making a distinction between the intuitive meaning of entropy i.e. lack of order, and its thermodynamic meaning i.e. the accessible volume of the phase space. In the case of hard rods for example, the nematic phase has more entropy than the isotropic phase at the same density since thin rods can have higher mobility by sliding past one another if they are on average aligned. In other words, the decrease in rotational entropy is compensated by yet a larger increase in

translational entropy, making the nematic phase more stable. A similar argument can be made for the freezing of hard spheres. Such transitions that involve an increase in entropy are generally referred to as *entropy-driven transitions* and are thought to occur in energetic systems as well [104].

The feasibility of disorder-order transitions can also be understood from a packing perspective as all permissible configurations of hard particle systems are packings of the corresponding shape(s). (The packing problem is discussed in detail in Section 2.3,). More particularly, the structure that is thermodynamically stable in the limit of infinite pressure should be among the packing(s) with the largest possible packing fraction. This can be seen by observing that the Gibbs free energy of a hard particle system is given by $G(P,T) = PV - ST$. As the entropy of any physical system is bounded from below, the Gibbs free energy is dominated by the PV term as $P \to \infty$ and the structure with the smallest specific volume i.e. the largest packing fraction will be eventually stable. Solving the packing problem is therefore akin to identifying the high-pressure equilibrium structure of the corresponding hard particle system. All known densest packings of two- and three-dimensional objects are ordered [105], which necessitates a disorder-order transition at some intermediate pressure/density.

2.2.1 Computational Studies of Hard Particles

Since Alder and Wainwright's pioneering work in 1957, numerous Monte Carlo (MC) and Molecular Dynamics (MD) studies of hard spheres have been performed. (A detailed description of hard particle simulation methods is given in Chapter III.) Different aspects of the hard sphere system have been considered including the equation of state [106], transport coefficients [97], nucleation rate [107], shear flow and hydrodynamics [108], glass transition [109] and binary mixtures [110, 111]. Simulation studies of non-spherical particles are however more recent and can only be dated back to the last three decades. Furthermore, the overwhelming majority of these studies are carried out using Monte Carlo due to computational difficulties of implementing an efficient, and yet accurate, event-driven Molecular Dynamics code for nonspherical hard particles. Here we go through the main results in this area. Although there have been several studies of the equations of state of hard particle fluids [112–115], we confine our attention only to studies in which non-isotropic portions of the phase diagram are also considered.

2.2.1.1 Infinitely Thin Disks

Hard infinitely thin disks were the first hard particle system for which an isotropic-to-nematic transition was observed by Frenkel and Eppenga [13, 116]. As circular planes with zero volume, they can only form orientationally ordered structures since translational order can only emerge for particles of nonzero volume [117]. In other words hard infinitely thin disks are excellent nematic formers due to the suppression of crystallization. This might have played a role in Frenkel and Eppenga's decision in using this particular system to demonstrate the possibility of an isotropic-nematic transition.

2.2.1.2 Cut Spheres

Hard cut spheres are obtained by symmetrically intersecting a sphere of radius D with two parallel planes distanced by L. Veerman and Frenkel have studied their phase diagram for $L/D < 0.4$. Prior to crystallization, the isotropic fluid transforms into a nematic and then a columnar phase for $L/D = 0.1$. However the nematic phase is no longer stable for $L/D = 0.2$, and is replaced by a cubatic liquid crystal; i.e. an orientationally ordered fluid with overall cubic symmetry. And finally for $L/D \geq 0.3$, only the isotropic phase and the solid phase are observed [118]. It has recently been suggested that the cubatic phase might indeed be metastable with respect to the columnar phase [119, 120].

In the limit of $L/D \to 0$, there is no distinction between a cut sphere and an infinitely thin cylinder of the same volume. Bates and Frenkel used this fact to study the phase behavior of thin cut spheres by simulating thin cylinders, They observed a nematic-to-columnar transition as had been observed for small– and yet nonzero– values of L/D in the earlier work of Veerman and Frenkel [118].

2.2.1.3 Spherocylinders

There have been several studies of hard spherocylinders since 1990s [15, 121, 122]. The most comprehensive study is by Bolhuis and Frenkel [122] who provided a complete picture of the phase diagram for all possible aspect ratios. For $L/D \to 0$, particles act as perfect spheres; a plastic phase is observed before crystallization to f.c.c. The phasic phase disappears for intermediate values of L/D and the isotropic fluid directly transforms into an f.c.c. crystal. At sufficiently large aspect ratios– i.e. $L/D > 3.0$– a nematic and a smectic phase are also observed in addition to the the solid. The f.c.c. phase is replaced by the AAA stacking for very large aspect ratios.

2.2.1.4 Ellipsoids

Hard ellipsoids can be obtained from hard spheres through an affine transformation, and they have been extensively studied in recent decades. Frenkel *et al.* [14, 123] studied the phase diagram of hard uniaxial ellipsoids– or as they call them 'ellipsoids of revolution'– of different aspect ratios and observed three distinct regions in the phase diagram. Sphere-like ellipsoids– i.e. ellipsoids with aspect ratios between 0.80 and 1.25– assemble into a rotator plastic phase before crystallizing into f.c.c, while prolate and oblate ellipsoids form a nematic liquid crystal instead before solidification. There is an intermediate region, however, where the isotropic fluid transforms directly into the crystal. Recently, Radu *et al.* demonstrated that the high-aspect-ratio stable solid phase is not an extended f.c.c.– as Frenkel and coworkers had assumed– but is a monoclinic phase with two ellipsoids of different orientation in a unit cell [124]. Hard biaxial ellipsoids however exhibit a richer phase behavior; two additional liquid crystalline phases- namely the discotic phase and the biaxial phase- are also observed in addition to the nematic liquid crystal [125].

2.2.1.5 Polyhedra

All previous simulations of hard polyhedra, in which the entire phase diagram has been mapped, have been for space-filling ones [17, 20, 126, 127], which, as discussed above, have *a priori* known stable solid phases. There are two approaches for simulating hard polyhedra; they are either represented perfectly with sharp edges and vertices [17, 20], or they are replaced with rigid clusters of hard spheres mimicking their shape [126, 127].

The simplest space-filling polyhedron is the hard cube. Hard cubes crystallize into a simple cubic lattice with $\phi = 1$ in the limit of infinite pressure. Simple cubic lattice has indeed been shown to be more stable with respect to other space-filling stacking variations [128]. Prior to that however, they form a liquid crystalline phase with cubatic order [126]. Depending on their aspect ratio $r = c/a$, hard tetragonal parallelepipeds tend to form various mesophases prior to crystallization [17, 127]. For $r \ll 1$, a nematic liquid crystal and a columnar phase forms prior to crystallization. At intermediate and high aspect ratios, a parquet phase and a smectic phase are observed instead.

Agarwal and Escobedo studied five other space-filling polyhedra in their recent work [20]. They observed hard triangular prisms and hard gyrobifastigia to directly transform into their space-filling crystal, while hard truncated octahedra, rhombic

dodecahedra and hexagonal prisms form rotator plastic phases before forming their corresponding space-filling solids. Using these examples, alongside cubes, Agarwal and Escobedo propose a general framework for predicting the type of mesophases that a certain building block can form by looking into its asphericity and rotational symmetry. They argue that high asphericity and low rotational symmetry leads to liquid crystalline phase i.e. phases with long-range rotational order but no translational order- while low asphericity and high rotational symmetry will lead to rotator phases– i.e. phases with long-range translational order but no rotational order. Systems of building blocks with Intermediate values of asphericity and intermediate rotational symmetry are thought to directly transform into their respective solid without going through any mesophases [20].

2.3 The Packing Problem

A family of objects $\{\mathcal{R}_i\}_{i=1}^{\infty} \subseteq \mathbb{R}^d$ is called a *packing* if their interiors are disjoint. For every packing, the *packing density* or the *packing fraction* is defined as:

$$\phi \ := \ \limsup_{r \to \infty} \frac{1}{\text{Vol}[B(r)]} \sum_{i=1}^{\infty} \text{Vol}[\mathcal{R}_i \cap B(r)] \tag{2.4}$$

with $B(r) = \{x \in \mathbb{R}^d : \|x\| \leq r\}$. For a periodic packing of congruent objects, Eq. (2.4) takes the form:

$$\phi \ = \ \frac{N V_{\mathcal{R}}}{V_B} \tag{2.5}$$

where N is the number of particles in the unit cell, and $V_{\mathcal{R}}$ and V_B are the volumes of the individual particle and the unit cell respectively. For congruent objects, the *packing problem* is to identify an arrangement with the largest possible packing fraction, and is posed as a part of the 18th of Hilbert's well-known *Mathematische Probleme*:

> "...How can one arrange most densely in space an infinite number of equal solids of a given form, e.g., spheres with given radii or regular tetrahedra with given edges (or in prescribed position), that is, how can one so fit them together that the ratio of the filled to the unfilled space may be as large as possible?" [26]

A packing with $\phi = 1$ is called a *tiling* of the d-dimensional space. The problem of identifying bodies that tile the space is also of interest to mathematicians, and has

received considerable attention in the literature [19, 23, 27, 129].

The packing problem is an optimization problem with infinite number of variables, which makes it very difficult to solve. For certain classes of packings describable by a finite number of parameters however, the local maxima of (2.4) can be easily calculated. For instance, the densest lattice packings[2] of three-dimensional compact convex sets can be obtained using the classical method of Minkowski [130], which has been used to obtain the densest lattice packings of a number of objects such as regular tetrahedra [37] and regular octahedra [130]. Betke and Henk have developed an efficient computer algorithm for analytic calculation of dense lattice packings of 3-polytopes, and have used it to obtain the densest lattice packings of all Platonic and Archimedean solids (Table. 2.1) [131]. Similar local maxima can be obtained for other classes of periodic packings with a few particles in the unit cell and have been used to obtain non-lattice dense packings of several three-dimensional objects [28, 30, 31, 105].

Table 2.1:
Densest lattice packings of Platonic and Archimedean solids compared to the best known packing of the corresponding polyhedron.

#	Body	Lattice Packing	All Packings
1	Tetrahedron [31, 37, 131]	36.7346938%	85.634768%
2	Cube	100%	100%
3	Octahedron [130, 131]	94.7368421%	94.7368421%
4	Dodecahedron [131]	90.4508497%	90.4508497%
5	Icosahedron [131]	83.6357445%	83.6357445%
6	Cubeoctahedron [131]	91.8367346%	91.8367346%
7	Icosidodecahedron [131]	86.4720371%	86.4720371%
8	Rhombic cubeoctahedron [131]	87.5805666%	87.5805666%
9	Rhombic icosidodecahedron [131]	80.4708487%	80.4708487%
10	Truncated cube [131, 132]	97.3747688%	97.3747688%
11	Truncated octahedron [131]	100%	100%
12	Truncated dodecahedron [131]	89.7787626%	89.7787626%
13	Truncated icosahedron [131]	78.49877759%	78.49877759%
14	Truncated cubeoctahedron [131]	84.9373252%	84.9373252%
15	Truncated icosidodecahedron [131]	82.7213595%	82.7213595%
16	Truncated tetrahedron [19, 131]	68.0921053%	99.5192308%
17	Snub cube [131]	78.769996%	78.769996%
18	Snub dodecahedron [131]	78.8640117%	78.8640117%

Although such local optima can be obtained, it is far more difficult to establish their global optimality. For instance, Lagrange (1773) and Gauss (1840) proved that

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[2]Lattice packings are periodic packings with one particle per unit cell.

the densest lattice packing of congruent circles in $\mathbb{R}^2$ and congruent spheres in $\mathbb{R}^3$ are the hexagonal lattice and the f.c.c. lattice respectively [22]. But the optimality of these solutions were not proven until 1892 for circles [25] and 2005 for spheres [24]. So far, the packing problem is only solved for a few non-space filling convex objects in $\mathbb{R}^3$ including the infinite cylinder [133], rhombic dodecahedron with a clipped corner [134] and spheres [24].

The difficulties of obtaining exact analytical solutions of packing problems on one hand, and the recent increase in the available computational power on the other hand has made numerical studies of packing more popular. The most commonly used numerical method is the Monte Carlo method in which the local maxima of the packing fraction are obtained through stochastic evolution of a periodic packing according to the laws of statistical mechanics [18]. (The technical details of the Monte Carlo approach is given in Chapter III.) Another method that has been recently used for obtaining dense packing of tetrahedra is the *periodic divide and concur (PDC) algorithm* which is based on the dynamics of the difference map on a constraint satisfaction iterative search algorithm and on the divide and concur constraint framework [135]. The main advantage of this method is its ability to evade local minima using a difference map; however since the number of constraints grows exponentially with the number of particles, its utility is confined to unit cells with a small number of particles only.

2.3.1 Sphere Packing

Perhaps no problem other than the packing of congruent spheres in three dimensions has exposed how little we know about solving packing problems. Studies of f.c.c. packings of spheres can be dated back to a Sanskrit work of 499 AD [22]. The first study of sphere packing in the modern era is due to T. Harriot who was interested in obtaining formulas for the number of cannonballs that can be stored in regularly stacked piles. Kepler's involvement in this problem came about through his correspondence with Harriot. In his 1611 essay *Strena seu de Nive sexangula (New Year's Gift Concerning Snowflakes)*, he asserts that the f.c.c. packing is the densest arrangement of spheres:

> "... the packing will be the tightest possible, so that in no other arrangement could more pellets be stuffed into the same container,"

This is widely known as *Kepler's conjecture.* In 1840 Gauss proved that the f.c.c. packing is the densest lattice packing of spheres. The two-dimensional analog of Kepler's conjecture was proven in 1892 [25]. Several upper bounds were obtained for

the sphere packing in $\mathbb{R}^3$ between 1919 and 1993 [136–142]. L Fejes-Tóth was the first to suggest that this infinite-variable optimization problem might be mappable onto a simpler finite-variable optimization problem. In particular, he attempted to obtain tight upper bounds for ϕ by minimizing the weighted volumes of thirteen neighboring Voronoi cells of a typical sphere packing. He also speculated that computers can be used in solving this optimization problem [143]. There were a series of unsuccessful attempts by Hsiang to solve the Kepler conjecture [144]. The final proof however came from Thomas Hales in the late 90's and early 2000's. He mapped the packing problem onto a number of finite-variable optimization problems by considering the weighted volumes of Voronoi cells, simplices and quarters. These problems were solved by the aid of a computer. A complete proof can be found in [24].

Not surprisingly, little success has been made in solving the sphere packing problem in higher dimensions. The densest lattice packings are, however, calculated for dimensions 4 and 5 [145, 146], $6-8$ [147] and 24 [148]. For dimensions beyond 8 no solution exists for the densest lattice packings except for $d = 24$ [148]. One remarkable fact about higher-dimensional spheres is that their apparent densest lattice packings might no longer be optimal; counterexamples can be found for dimensions $10, 11, 13, 18, 20$ and 22 [149]. It has indeed been conjectured that the densest packings are not lattice packings for sufficiently large dimensions [150]. There are several numerical results on the packing and thermodynamics of hyperspheres in the literature [135, 151–153].

2.3.2 Packing of Regular Tetrahedra

Platonic solids[3] were sources of fascination for ancient Greeks who were the first to identify and enumerate them. In his final book of *Elements*, Euclid gives a complete mathematical description of the five Platonic solids. They also play a central role in Plato's philosophy, and are associated with the five elements that mix together to form the universe[4].

The regular tetrahedron is the only self-dual Platonic solid, it is also the only Platonic solid that lacks central symmetry. Although Aristotle correctly identified the other three non-space-filling Platonic solids, he erroneously claimed that regular

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[3]In Platonic solids, all faces are congruent regular polygons which the same number of faces coming together at each vertex. All vertices are therefore equivalent in the sense that they can be mapped into one another via symmetry operations.

[4]In his dialogue *Timaeus* c. 360 BC, Plato associates air with octahedron, water with icosahedron, earth with hexahedron (cube), and fire with tetrahedron. Dodecahedron is saved for the *cosmos*.

tetrahedra tile Euclidean space. This led to an error that persisted for almost two millennia, and several thinkers after Aristotle tried to find the number of tetrahedra that can pack around a point. Only in the 15th century AD, Johannes Múller (1436-1476) managed to spot the flaw in Aristotle's claim in his lost manuscript "*On the five like-sided bodies, that are usually called regular, and which of them fill their natural place and which of them not, in contradiction to the commentator on Aristotle, Averroés*". The first existing written account of this problem can be found in the book of the Polish mathematician, J. Broscius (1591-1652) where he explicitly calculates the dihedral angle of a corner of a tetrahedron, showing that no integer multiple of it equals 4π [23].

The search for lattice packings of tetrahedra started by Minkowski, who used his method to find the densest lattice packing of tetrahedra [130]. He however made a mistake in identifying its difference polyhedra[5] and obtained an exceedingly low packing fraction of $\frac{9}{38} \approx 23.684211\%$ [31]. The densest lattice packing with $\phi = \frac{18}{49} \approx 36.734694\%$ was first proposed by Grömer in 1962 [36]; Hoylman proved its optimality in 1970 using Minkowski's method [37].

In 2006, Conway and Torquato proposed several dense packings and performed numerical compressions to obtain densities as high as 72% [27]. They obtained their 'reformed Scottish packing' from replacing each icosahedron with twenty tetrahedra in the densest lattice packing of icosahedra calculated by Betke and Henk [131]. The other dense arrangement– the Welsh bubbles– were based on polytetrahedral networks of clathrate hydrates. (The structural details of these packings are discussed in Chapter V.) Their failure to obtain packings denser than the sphere packing led them to question the validity of *Ulam's conjecture* which asserts that all convex bodies in $\mathbb{R}^3$ can pack better than spheres [154] (As will be seen in Chapter V, our numerical compression of the reformed Scottish and the Welsh packings leads to packings denser than spheres.)

In 2008, Chen debunked Conway and Torquato's doubts about Ulam's conjecture by proposing a packing with $\phi_{\max} = 77.86\%$ which packed significantly better than spheres [28]. The unit cell contained 18 tetrahedra that were arranged into two nonamers that were related to each other by inversion. Each nonamer contained nine tetrahedra forming two pentagonal bipyramids sharing a central tetrahedron. In 2009, Torquato and Jiao numerically compressed this packing to $\phi_{\max} = 78.20\%$ [29].

The last few months of 2009 witnessed a flurry of activity on this problem. These

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[5]For a convex set K, the difference set DK is defined as $DK = \left\{ \dfrac{x-y}{2}; x, y \in K \right\}$.

Table 2.2: Dense packings of regular tetrahedra.

Type	Reference	Date	Packing Fraction
Lattice Packing	Grömer [36], Hoylmann [37]	1970	$\frac{18}{49} \approx 36.73\%$
Reformed Scottish Bubble	Conway, Torquato [27]	2006	71.66%
Welsh Bubble	Conway, Torquato [27]	2006	71.74%
Nonamer Crystal	Chen [28]	2008	77.86%
Disordered	Haji-Akbari *et al.* [18]	2009	78.56%
Quasicrystal	Haji-Akbari *et al.* [18]	2009	83.24%
Approximant	Haji-Akbari *et al.* [18]	2009	85.03%
Dimer crystal, monoclinic	Kallus *et al.* [30]	2009	$\frac{100}{117} \approx 85.47\%$
Dimer crystal, triclinic	Chen *et al.* [31]	2010	$\frac{4000}{4671} \approx 85.63\%$

attempts will be fully discussed in Chapters V and VI of this thesis. In November 2009, we reported the spontaneous formation of a dodecagonal quasicrystal in a system of hard tetrahedraledra and compressed the quasicrystal and its first-order approximant to a maximum packing fractions of $\phi_{\max} = 83.24\%$ and $\phi_{\max} = 85.03\%$ respectively [18]. Shortly after, Kallus *et al* reported a double dimer lattice with four particles in a monoclinic unit cell with $\phi_{\max} = \frac{100}{117} \approx 85.470085\%$, slightly better than the approximant [30]. This current world record belongs to Chen, Engel and Glotzer [31], who optimized Kallus's packing to obtain a triclinic cell with $\phi = \frac{4000}{4671} \approx 85.634782\%$. Numerical simulations of the unit cells with 16 tetrahedra and less confirmed the optimality of Chen's structure. In 2010, Gravel *et al.* obtained a nontrivial upper bound for the packing density of regular tetrahedra [32]. Major milestones in solving the packing problem for tetrahedra are given in Table 2.2.

CHAPTER III

Methods

In this chapter, we discuss the technical details of various computational methods that will be used in the remainder of this thesis. This chapter is organized as follows. Section 3.1 provides a brief overview of the statistical mechanical principles used in this chapter and the rest of this dissertation. In Section 3.2, conventional methods used for sampling the configurational space of hard particles are discussed. This includes Monte Carlo and Molecular Dynamics simulations in different ensembles. Section 3.3 gives a general overview of free energy calculation methods, and is concluded by a detailed discussion of the Frenkel-Ladd method that we use for calculating the free energies of crystalline phases of hard tetrahedra and hard triangular bipyramids. Section 3.4 is dedicated to the discussion of the concept of free volume, and the methods with which it can be calculated. And finally, Section 3.5 gives a short description of the major analysis tools used in the thesis.

3.1 Statistical Mechanics Principles

All thermodynamic properties of a physical system are fully specified by its partition function $\mathfrak{F}$, which, in the classical treatment, is a multi-dimensional integral of a weight function $w(\Gamma)$ over all degrees of freedom of a system:

$$\mathfrak{F} = \int w(\Gamma) d\Gamma \tag{3.1}$$

The probability that the system visits a certain configuration is given by $w(\Gamma)d\Gamma/\mathfrak{F}$. This probability can be used to calculate the ensemble average of a given mechanical property X:

$$\langle X \rangle = \frac{1}{\mathfrak{F}} \int X(\Gamma) w(\Gamma) d\Gamma \tag{3.2}$$

Furthermore, the underlying thermodynamic potential that is maximized or minimized in a particular ensemble is proportional to the logarithm of $\mathcal{F}$. Therefore every thermodynamic property can be calculated with the knowledge of $\mathcal{F}$. The mathematical form of $w(\cdot)$ and $\mathfrak{F}$ depends on the thermodynamic ensemble; w and $\mathfrak{F}$ are given for several ensembles in Table. 3.1. In the thermodynamic limit, where all fluctuations become vanishingly small, all ensembles become equivalent.

The canonical (NVT) ensemble is arguably the most widely used ensemble in statistical mechanics, and will be used extensively in the remainder of this dissertation. We therefore introduce the notations that will be used for the NVT ensemble here. The weight function in the canonical ensemble is the famous *Boltzmann factor* which is $w = e^{-\beta H(\mathbf{x}^N, \mathbf{p}^N)}$ with $\beta = 1/k_B T$, $H(\mathbf{x}^N, \mathbf{p}^N)$ the Hamiltonian of the system; $\mathbf{x}^N \equiv (\mathbf{x}_1, \mathbf{x}_2, \cdots, \mathbf{x}_N)$ the configurational degrees of freedom and $\mathbf{p}^N \equiv (\mathbf{p}_1, \mathbf{p}_2, \cdots, \mathbf{p}_N)$ the conjugate momenta. The partition function $Q(N, V, T)$ is thus given by[1]:

$$Q(N, V, \beta) = \frac{1}{N!} \int e^{-\beta H(\mathbf{x}^N, \mathbf{p}^N)} d\mathbf{x}^N d\mathbf{p}^N \tag{3.3}$$

The thermodynamic potential that is minimized in equilibrium is the *Helmholtz free energy* and is given by $A = -k_B T \ln Q(N, V, T)$. Momentum contributions to the partition function– and the free energy– can be calculated analytically; they will therefore be included only if they are different for two different systems or state points. We will otherwise be dealing with the *configurational partition function Z*:

$$Z(N, V, T) = \int e^{-\beta U(\mathbf{x}^N)} d\mathbf{x}^N \tag{3.4}$$

with $A = -k_B T \ln Z$. The ensemble average of any mechanical observable that only depends on the positions (and orientations) of particles– and not their momenta– is therefore given by:

$$\langle X \rangle = \frac{1}{Z} \int X(\mathbf{x}^N) e^{-\beta U(\mathbf{x}^N)} d\mathbf{x}^N \tag{3.5}$$

In the quantum treatment of statistical mechanics, all integrals turn into sums over discrete microstates of the system, that are eigenfunctions of the Schrödinger equation. For instance $Q(N, V, T) = \sum_\nu e^{-\beta E_\nu}$. The classical and quantum treatments can be reconciled if the energy difference between the micorstates is $\ll k_B T$. Throughout

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[1]The $N!$ that appears in the denominator appears only if particles are indistinguishable. and is absent in systems where particles are distinguishable, like a crystal.

Table 3.1: w, Γ and $\mathfrak{F}$ for different ensembles.

Ensemble	Γ	w	$\ln \mathfrak{F}$
NVE	$(\mathbf{x}^N, \mathbf{p}^N)$	$\delta[H(\Gamma) - E]$	S/k_B
NVT	$(\mathbf{x}^N, \mathbf{p}^N)$	$e^{-\beta H(\Gamma)}$	$-A/k_B T$
NPT	$(\mathbf{x}^N, \mathbf{p}^N, V)$	$e^{-\beta PV - \beta H(\mathbf{q}^N, \mathbf{p}^N)}$	$-G/k_B T$
μVT	$(N; \mathbf{x}^N, \mathbf{p}^N)$	$e^{\beta \mu N - \beta H(\mathbf{q}^N, \mathbf{p}^N)}$	$PV/k_B T$

this dissertation, we will only use the classical treatment.

Unlike momentum contributions that can be calculated analytically, it is not possible to obtain analytical expressions for Z except for very few systems with simple potentials. Direct numerical integration of (3.4) or (3.5) is also not feasible as the amount of discretization needed for an accurate estimate of the integral is orders of magnitude beyond what can be handled with the existing computing power. These integrals can however be calculated indirectly. We will discuss the methods used for direct or indirect calculation of (3.4) in Section 3.3, but before that we discuss the numerical techniques used for indirect evaluation of (3.5).

3.2 Conventional Sampling Techniques

The success of statistical mechanics in explaining the macroscopic behavior of many-body systems by replacing time averages with ensemble averages lies in the fundamental assumption that most physical systems are *ergodic*; i.e. the likelihood that any particular configuration is visited by a typical sufficiently long trajectory in the configuration space is constant and independent of the trajectory. If a system is ergodic, the time average of *any* mechanical observable can be obtained from a stochastic trajectory provided that the stochastic process used for generating it has the same stationary (i.e. equilibrium) distribution as the physical system. The whole class of *Monte Carlo (MC)* methods employ this to study the equilibrium properties of many-body systems. Eq. (3.5) is therefore taken as its average over a sufficiently large stochastic trajectory i.e.:

$$\langle X \rangle = \lim_{n \to \infty} \frac{1}{n} \sum_{j=1}^{n} X_j$$

with $X_j := X(\mathbf{x}_j^N)$, the value of X in the jth snapshot of the stochastic trajectory.

Another alternative is to evolve the system according to its underlying Newtonian

Table 3.2: Acceptance probabilities of MC simulations in different ensembles.

Ensemble	$P_{\mathrm{acc}}(\Gamma^{(n+1)}|\Gamma^{(n)})$
NVT	$\min\left\{1, e^{-\beta(U_{n+1}-U_n)}\right\}$
NPT	$\min\left\{1, (V_{n+1}/V_n)^N e^{-\beta P(V_{n+1}-V_n)-\beta(U_{n+1}-U_n)}\right\}$
μVT	$\min\left\{1, \frac{N_n!}{N_{n+1}!}e^{\beta\mu(N_{n+1}-N_n)-\beta(U_{n+1}-U_n)}\right\}$

dynamics and replace (3.5) with the actual time average $\langle X\rangle_t = \lim_{t\to\infty}\frac{1}{t}\int_0^t X(\tau)d\tau$. This is what is done in *Molecular Dynamics (MD)* simulations. In the following sections, we provide a brief description of these two classes of methods, and outline their special application to hard particle systems. We also outline the technical details of the MC algorithm used in this dissertation

3.2.1 Monte Carlo

The conventional Monte Carlo algorithm that is widely used in numerical studies of condensed matter was first proposed in 1953 by Metropolis [155], who used it calculate the equation of state of two-dimensional hard disks. The success of his method can be attributed to his careful selection of the Markov process, which is still in use today. A particular *trial move* $\Gamma^{(n+1)}$ is generated from $\Gamma^{(n)}$ according to the distribution $\alpha(\Gamma^{(n)}|\Gamma^{(n+1)})$. The trial move is then accepted or rejected according to the following acceptance probability:

$$P_{\mathrm{acc}}\left(\Gamma^{(n+1)}|\Gamma^{(n)}\right) = \min\left\{1, \frac{\mathbb{P}(\Gamma^{(n+1)})}{\mathbb{P}(\Gamma^{(n)})}\right\} \tag{3.6}$$

i.e. $\Gamma^{(n+1)}$ is always accepted if it is more probable than $\Gamma^{(n)}$; otherwise it is accepted with the probability $\mathbb{P}(\Gamma^{(n+1)})/\mathbb{P}(\Gamma^{(n)})$. It can be easily shown that $\mathbb{P}(\cdot)$ is the stationary distribution of this Markov chain if $\alpha(\Lambda|\Gamma) = \alpha(\Gamma|\Lambda)$ for every Γ, Λ. The condition $\mathbb{P}(\Gamma)\alpha(\Lambda|\Gamma) = \mathbb{P}(\Lambda)\alpha(\Gamma|\Lambda)$, which is satisfied in the Metropolis algorithm, is called *detailed balance* and is a sufficient condition for the convergence of the Markov chain to the equilibrium distribution.

The Metropolis algorithm allows the system to evolve through the *important* regions of the configuration space without visiting the (high energy) low probability regions that contribute little to (3.4) and (3.5). Table. 3.2 summarizes the acceptance probabilities for MC simulations at different ensembles.

Monte Carlo simulations are very popular in studies of nonspherical hard particles.

This can be partly attributed to their relative ease of implementation, as the problem of calculating the internal potential is reduced to detecting collisions between rigid geometric objects. The acceptance criterion is usually very simple. For instance, in the canonical ensemble, all moves that do not generate an overlap are accepted. All the findings of this dissertation are obtained from MC simulations. We discuss the technical details of our implementation in the following sections.

3.2.1.1 Representation of Particles

We represent tetrahedra and triangular bipyramids in their full geometrical details, and do not adopt the convention of approximating them with rigid clusters of hard spheres. The orientation of each particle is described by a unit quaternion $\mathbf{q} = (q_t, q_x, q_y, q_z)$. The reference orientation, which corresponds to $\mathbf{q} = (1, 0, 0, 0)$, for each shape is given in the chapter dedicated to its study. Further details about quaternions and their relationship with rotation are given later in Section 4.3.3.2.

3.2.1.2 Units

MC simulations are performed in both NVT and NPT ensembles. Observe that $Z(N, V, T)$ is not a function of T and only depends on the number density N/V. The same assertion is true in the NPT ensemble where the configurational part of $\Delta(N, P, T)$ is a function of P/T only. We therefore choose the energy scale so that $T^* = k_B T/\epsilon$ is unity. A characteristic length of the system– e.g. the edge length of a tetrahedron or a triangular bipyramid– is also taken as the length scale σ. The dimensionless pressure therefore takes the form $P^* = P\sigma^3/k_B T$.

3.2.1.3 Thermodynamic Ensembles

Isochoric MC simulations are routinely used in studies of hard particles. Isobaric and the closely related isotensio MC simulations are also very common, much more so than in the soft particle systems where they are discouraged over NVT simulations due to their higher computational cost. This can be partly attributed to the difficulty of calculating pressure in isochoric simulations of nonspherical hard particles[2]; unlike soft particles where instantaneous pressure can be easily computed from the virial theorem [156]. In addition, it is easier for a periodic structure to find the appropriate

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[2]There are several method for calculating pressure in NVT simulations of hard particles; one such approach is presented in Section 3.5.1; however these methods are usually so computationally costly that the additional computational overhead for using NPT instead of NVT is justified.

box dimensions in an isobaric simulation. This is particularly important for hard particles where no attractive potential exists to guide the formation of the crystal with the right spacing, e.g. by arranging the particles to reside at the minima of the interparticle potential.

In conventional NPT MC, the simulation box is isotropically resized along all directions. However, there are other variants of NPT MC where both the size and the shape of the simulation box are altered. This can help a crystalline structure to find the optimal strain for a given external pressure by getting rid of its internal stresses. A similar method, known as the *Variable-Shape NVT MC*, exists where the shape of the simulation box is altered while its volume is kept constant [157].

We use all four ensembles for self-assembly simulations, while equations of state are calculated using isobaric and isotension simulations. Simulations that are used for calculating quantities such as free volume, mean-square displacement or autocorrelation functions are exclusively carried out in the canonical ensemble.

3.2.1.4 Trial Moves

The trial moves used in our simulations are local in nature, and do not involve any unphysical moves[3]. They can be classified into *particle sweeps* and *box sweeps*. Particle sweeps include trial translations and trial rotations, and are performed in all ensembles. Box sweeps are however performed in isobaric, isotension, and variable-shape isochoric simulations only. Each MC cycle includes N particle sweeps; each being a trial translation or a trial rotation with equal probability. An additional box sweep is carried out per cycle in isobaric, isotension and variable-shape isochoric simulations. Implementation details of each trial move are presented in Table 3.3. It is necessary to note that trial rotations are performed using the procedure given by Vesley [158].

$\Delta x_{\max}, \Delta q_{\max}$ and $\Delta b_{\max}$ are the maximum step sizes for different trial moves. They only affect the efficiency with which the configurational space is sampled without changing the asymptotic properties of the Markov chain (such as its stationary distribution). If these step sizes are too small, most trial moves will be accepted, but the system will hardly change since each trial move will only bring about a very small change to the configuration. On the other hand, too large step sizes will cause the majority of trial moves to be rejected, which will also lead to inefficient sampling. It is therefore customary to adjust the values of these parameters dynamically so that

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[3]The compression method used for obtaining dense packings is an exception, and involves unphysical compression moves that generate unphysical overlapping configurations.

Table 3.3:
Implementation details for different MC trial moves. $\mathbf{r}$ and $\mathbf{q}$ stand for the position and the quaternion of the particle randomly selected for translation or rotation. B is the box matrix having as its columns the vectors that span the simulation box. $S^3 = \{\mathbf{x} \in \mathbb{R}^4 : \|\mathbf{x}\| = 1\}$ is the unit sphere in $\mathbb{R}^4$, and $U(a, b)$ is a random variable uniformly distributed on $[a, b]$.

Trial Move	Ensemble	Implementation
Translation	All	$\mathbf{r}_{\text{new}} = \mathbf{r}_{\text{old}} + \Delta x_{\max} \mathbf{u},\, u_i \sim U\left(-\frac{1}{2}, \frac{1}{2}\right)$
Rotation	All	$\mathbf{q}_{\text{new}} = \dfrac{\mathbf{q}_{\text{old}} + \Delta q_{\max} \mathbf{q}_u}{\|\mathbf{q}_{\text{old}} + \Delta q_{\max} \mathbf{q}_u\|},\, \mathbf{q}_u \sim U(S^3)$
Box Sweep	NPT	$B_{\text{new}} = (1 + \Delta b_{\max} w) B_{\text{old}},\, w \sim U\left(-\frac{1}{2}, \frac{1}{2}\right)$
Box Sweep	NσT	$B_{\text{new}}^{ij} = B_{\text{old}}^{ij} + \Delta b_{\max} w_{ij},\, w_{ij} \sim U\left(-\frac{1}{2}, \frac{1}{2}\right)$
Box Sweep	VSNVT	$B_{\text{new}} = \left(\dfrac{\text{Vol}(B_{\text{old}})}{\text{Vol}(W)}\right)^{1/3} W,\, W^{ij} = B_{\text{old}}^{ij} + \Delta b_{\max} w_{ij},\, w_{ij} \sim U\left(-\frac{1}{2}, \frac{1}{2}\right)$

a certain fraction of moves are accepted. We use a target acceptance probability of 30% for the majority of our simulations. However we occasionally fix the maximum step sizes in simulations used for calculating dynamical properties such as mean-square displacements and autocorrelation functions. This is to assure the temporal equivalence of MC cycles in a simulation.

3.2.1.5 Collision Detection

The most time-consuming part of a hard particle MC simulation is the collision detection. For hard spheres, this task is as straightforward as checking their distances; there are more sophisticated formulae for other shapes including ellipsoids [159], spherocylinders [15], or thin disks [116]. In this section, we mention the three different algorithms that we used to detect overlaps between two convex polyhedra.

The simplest algorithm is based on the observation that two convex polyhedra overlap if an edge of one polyhedron is totally or partially located inside the other polyhedron. Since the interior of a convex polyhedron is *below* all its faces, this can be checked for each edge by intersecting it with the planes specifying those faces, and identifying the parts of an edge that lies below all those planes. The two polyhedra overlap if such a line segment is nonempty for at least one edge. This algorithm was used in some of our preliminary runs; it is also used to verify the densest packings obtained from simulations that use other collision detection algorithms.

For most of our production runs, we use the *separating axis theorem* [160], which

states that two convex sets do not intersect if and only if a plane can be found that completely separates them. In other words, the vertices of one convex polyhedron must lie on one side of the plane and the vertices of the other polyhedron on the other side. Taking two vertices of one polyhedron and one vertex of the other polyhedron defines a trial candidate for a separating plane. If none of these trial candidates is a separating plane, two polyhedra must intersect. For symmetric polyhedra such as tetrahedra, this algorithm can be further optimized by sorting the vertices of one polyhedron according to their distance to the other polyhedron and discarding the irrelevant trial candidates.

Another collision detection algorithm that is used for some production runs is the well-known Gilbert-Johnson-Keerthi (GJK) algorithm [161]. For a pair of convex sets C_1 and C_2, the *Minkowski difference* is defined as $C_1 - C_2 = \{x - y : x \in C_1, y \in C_2\}$. Observe that C_1 and C_2 intersect if $0 \in C_1 - C_2$. The GJK algorithm checks this condition using an iterative algorithm, which is well optimized for polytopes in $\mathbb{R}^3$. This method can also be used to determine the minimum distance of two nonintersecting convex polytopes.

3.2.1.6 Compression Algorithm[4]

The densest packing that can be attained from a given configuration is thermodynamically achieved at $P \to \infty$. This can be done by gradually increasing the pressure up to a sufficiently large value in the course of an isobaric MC simulation. Although this scheme works perfectly for small systems; i.e. systems with a few particles in a periodic box, it is very inefficient in compressing large systems. Note that a compression box sweep will be discarded even if it only generates one overlap. At a given density/pressure, the likelihood that a compression generates new overlaps is higher in larger systems. This in turn leads to a large number of volume moves to be rejected, and hence a smaller value of $\Delta b_{\max}$, and less efficient compression. In order to overcome this difficulty, we develop a modified compression Monte Carlo scheme which allows for a few minor overlaps during compression. It should be emphasized that our modified scheme is only used to obtain high density results; the conventional isochoric and isobaric Monte Carlo methods are used to produce all thermodynamic information, such as equations of state and phase transitions [18].

In our method, all expansion/compression moves are accepted even if they result in new overlaps and a different criterion is used to ensure that the number and amount

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[4]This section is partly based on the following manuscript: Amir Haji-Akbari, Michael Engel, *et al. Nature* **462**: 773–777 (**2009**) [18].

of overlaps remain small. We monitor p, the fraction of trial translations accepted since the previous volume move, and compare it to a target acceptance probability p_t. If $p < p_t$, the volume of the box is increased by a random factor f uniformly chosen from the interval $[1, 1 + 0.002\Delta x_{\max}]$ while for $p > p_t$, a compression is attempted by a random factor uniformly distributed in $[1 - 0.002\Delta x_{\max}, 1]$. Between volume moves, the system evolves through conventional trial translations and trial rotations and the moves that do not generate new overlaps are accepted.

$\Delta x_{\max}$, the maximum step size for a trial translation is the control parameter in this method and is inversely related to pressure in the conventional isobaric MC algorithm. In a compression run, $\Delta x_{\max}$ is exponentially decreased until the densest packing is obtained. A few hundred cycles of conventional NVT MC are carried out at the end to get rid of all minor overlaps generated by this method.

3.2.2 Molecular Dynamics

In a Molecular Dynamics simulation, the system evolves according to Newtonian dynamics. It therefore not only can be used for estimating (3.5), but also is the method of choice for studying dynamical phenomena such as diffusion [97] or granular flow [162]. Furthermore, it is more efficient in equilibrating systems of high densities [163].

A Molecular Dynamics simulation of soft particles– i.e. particles interacting through continuous potentials– is equivalent to solving a system of ordinary differential equations; a task that can be done by proper discretization of time. At each time step Δt, the force (and the torque) that is exerted on each particle is calculated, which is then used to update its position (and orientation). However, if there are discontinuities or singularities in the inter-particle potential, this scheme is no longer usable since the behavior of the system will heavily depend on collision events; i.e. events at which the relative positions (orientations) of a pair of particles coincides a discontinuity or singularity of the inter-particle potential. If such a system is evolved in constant time steps, some collisions are inevitably going to be missed, leading to a misrepresentation of the true physics of the system. An MD simulation of such systems should therefore involve a sequential processing of *all* collision events in addition to gradual evolution of the system according to continuous parts of their potential. This is in contrast to the inherently parallel scheme used for soft particles.

Hard particles can be considered on a *free flight mode* between successive collisions i.e. they will maintain their linear and angular momenta as no force (or torque) is exerted on them by other particles unless a collision occurs. Therefore an MD

simulation of hard particles only involves identifying and processing the collision events, and fast-forwarding the system between successive collisions. Although the problem of detecting collisions is mathematically well defined there is no unique recipé for processing collision events. Both the linear and angular momenta are conserved in a collision event. The total kinetic energy of the colliding particles is also conserved in *elastic collisions*. (Inelastic collisions are not very common and are mostly used in simulations of granular flow.) Additional assumptions are however needed for processing collisions. For instance, it is usually assumed that the change in the linear momentum of each colliding particle is perpendicular to the plane that is tangent to colliding particles in the point of contact [163, 164].

Determining collision times between spheres that move with constant velocities is as easy as solving a quadratic equation. It is therefore straightforward to use this *event-driven* approach for implementing efficient MD computer programs for hard spheres. For nonspherical particles however, collision times cannot be calculated analytically, and numerical solutions are only possible for very simple shapes. The only alternative is to fast-forward the system in small increments of time Δt until an overlap is detected, collision time can then be precisely calculated by an iterative process that involves going backwards and forward in time within the last timestep. Apart from the uncertainties in the estimates of the collision times, some *sliding collisions* might also be missed in this process. This could lead to an unphysical dynamics or an incorrect estimate of properties such as pressure.

Considering these limitations and the relative ease of implementing MC, MD is rarely used for nonspherical hard particles. Indeed all the examples mentioned in Section 2.2.1 are carried out using MC simulations. For the same reason, we did not use MD in any of our simulations, and this short discussion is included for completeness only.

3.3 Enhanced Sampling Techniques and Free Energy Calculation Methods

In this section, we present a brief overview of the free energy calculation methods suitable for studying phase equilibrium. In Section 3.3.1, the importance of free energy calculation methods is discussed. In Section 3.3.2, a short description will be given on conventional free energy calculation methods. And Section 3.3.4 is devoted to the adaptation of thermodynamic integration for calculating the free energy of crystals in hard particle systems.

3.3.1 Why Calculate Free Energy?

The 2nd law of thermodynamics states that the entropy of an isolated system that is not in equilibrium will tend to increase over time, approaching a maximum value in equilibrium i.e. $dS/dt \geq 0$ in the NVE ensemble. This inequality, along with the 1st law of thermodynamics, can be used to derive similar inequalities for other ensembles. In the canonical ensemble, for example, the Helmholtz free energy $A = E - ST$ is minimized in equilibrium. As mentioned in Section 3.1 we have:

$$S = k_B \ln \Omega(N, V, E) \tag{3.7}$$
$$A = -k_B T \ln Q(N, V, T) \tag{3.8}$$

The problem of identifying the most stable phase at a given state point is therefore reduced to calculating the types of integrals given in Eq. 3.3. In the following section, we go through several approaches used for estimating/comparing 3.3 for different structures.

3.3.1.1 Limitations of Conventional Sampling Techniques

Historically, methods such as Molecular Dynamics and Monte Carlo have been successfully used to predict the types of phases that a particular system can form under different conditions. Indeed if a physical system is ergodic, these sampling methods are guaranteed to reproduce the true equilibrium structure for sufficiently long times, or sufficiently large numbers of MC cycles. In the real world however, simulations are finite; the true equilibrium phase will therefore not be isolated if the simulation time is shorter than the decorrelation time of the system. This can typically arise if the pathway that connects the starting configuration to the true equilibrium structure travels through regions of configuration space that have a low probability of being visited in the corresponding thermodynamic ensemble.

Such kinetic traps can exist in all parts of the phase diagram especially at low temperatures (or high pressures); they however become particularly problematic in the vicinity of coexistence manifolds[5]. For instance, the driving force for a first-order phase transition is the difference between the chemical potential of two phases. This thermodynamic driving force can become vanishingly small in the vicinity of a coexistence manifold, which can, in turn, lead to diverging relaxation times. The coexistence manifolds of a phase diagram can therefore not be precisely determined with

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[5]Coexistence manifold is the generalization of a coexistence curve for systems with more than two intensive degrees of freedom, such as multicomponent systems.

conventional sampling methods such as MD or MC; instead the conditions of thermo-dynamic equilibrium i.e. the equality of chemical potentials should be explicitly or implicitly imposed. In general, the methods described in Section 3.3.2 can be used to determine the free energy potentials of the competing phases, which can in turn be used to equate their chemical potentials. Implicit methods explained in Section 3.3.3 however work by satisfying the conditions of phase equilibrium by exchanging work and particles between two phases.

3.3.2 Conventional Free Energy Calculation Methods

The outcome of every free energy calculation method is the free energy *difference* between two different systems, or two different structures of a given system. In *density-of-states methods*, this is done through equilibrium simulations while in the *work-based methods*, the work performed along an irreversible path is used to estimate the free energy difference. It is however noteworthy that the change in the free energy *within* a single phase can be easily calculated from mechanical observables and no specialized method is necessary. For instance:

$$A(V_2) - A(V_1) = -\int_{V_1}^{V_2} P dV \tag{3.9}$$

$$\frac{A(T_2)}{T_2} - \frac{A(T_1)}{T_1} = -\int_{T_1}^{T_2} \frac{E}{T^2} dT \tag{3.10}$$

3.3.2.1 Density-of-states (Equilibrium) Methods

The excess free energy difference between two systems with interparticle potentials $U_0(\cdot)$ and $U_1(\cdot)$ is given by:

$$\Delta A = -k_B T \ln Z_1/Z_0 \tag{3.11}$$

In the density-of-states methods [165], the ratio Z_1/Z_0 is calculated through equi-librium simulations of systems with potentials ranging from U_0 to U_1. There are three types of density-of-states methods: free energy perturbation (FEP) methods, the histogram methods and thermodynamic integration; they differ in the amount of discretization used for interpolating between $U_0(\cdot)$ and $U_1(\cdot)$.

Free Energy Perturbation (FEP) Methods One can rearrange Eq. (3.11) to obtain:

$$
\begin{aligned}
\Delta A &= -\beta^{-1} \ln \frac{\int \exp[-\beta U_1(\mathbf{x})] d\mathbf{x}}{\int \exp[-\beta U_0(\mathbf{x})] d\mathbf{x}} \\
&= -\beta^{-1} \ln \int \exp[-\beta(U_1(\mathbf{x}) - U_0(\mathbf{x}))] \mathcal{P}_0(\mathbf{x}) d\mathbf{x} \\
&= -\beta^{-1} \ln \langle \exp[-\beta(U_1(\mathbf{x}) - U_0(\mathbf{x})] \rangle_0 \qquad (3.12)
\end{aligned}
$$

where $\mathcal{P}_n(\mathbf{x}^N)$ is the probability density of configuration $\mathbf{x}^N$ for a system with the interparticle potential $U_n(\cdot)$. Eq. 3.12 is the basis for FEP methods [166, 167]. Sampling is performed in system 0, the reference or the unperturbed system, and $\exp[-\beta(U_1(\mathbf{x}) - U_0(\mathbf{x}))]$ values are used for estimating the free energy difference between system 1, the perturbed system, and system 0.

Many well-known methods fall into FEP category. Perhaps the most well known example is the *Widom particle insertion* [168]. In this method, a ghost particle is added to the system and the change in the potential energy of the system is used to calculate the chemical potential:

$$
\begin{aligned}
\mu = \left(\frac{\partial A}{\partial N} \right)_{T,V} &\approx -\beta^{-1} \ln \frac{Q_{N+1}}{Q_N} \\
&= -\beta^{-1} \ln \frac{\int \exp[-\beta U(\mathbf{x}^{N+1})] d\mathbf{x}^{N+1}}{\int \exp[-\beta U(\mathbf{x}^N)] d\mathbf{x}^N} \\
&= -\beta^{-1} \ln \int \exp[-\beta \Delta U] \mathcal{P}_N(\mathbf{x}) d\mathbf{x}_{N+1} \qquad (3.13)
\end{aligned}
$$

In the context of FEP methods, system 0 has $N + 1$ particles where the $(N + 1)$-th particle does not interact with the other N particles, while system 1 has $N + 1$ interacting particles. The actual simulation is done on system 0- i.e. the system with N interacting particles– and the ghost particle is removed after ΔU has been calculated.

Histogram Methods Suppose that system 0 can be transformed into system 1 by changing a parameter λ from λ_0 to λ_1. λ can be a macroscopic state variable, a parameter that transforms $\mathcal{H}_0$ to $\mathcal{H}_1$ or a generalized coordinate that allows different structural states to be distinguished. An extended ensemble can thus be defined with the partition function $\mathcal{N}$. The probability of observing the system in macrostate i is

therefore given by:

$$\mathcal{P}_i = \mathcal{P}(\lambda_i) \quad = \quad \frac{\int \exp[-\beta\mathcal{H}]\delta(\lambda(\mathbf{x},\mathbf{p})-\lambda_i)d\mathbf{x}d\mathbf{p}}{\mathcal{N}} \qquad (3.14)$$

Note that β and $\mathcal{H}$ can also depend on λ. In histogram methods [169], the interval $[\lambda_0, \lambda_1]$ is partitioned into a number of finite subintervals, and the number of times the system visits each macrostate is counted to estimate $\mathcal{P}_i$. The free energy difference is therefore calculated from $\Delta A = -k_B T \ln \frac{\mathcal{P}_1}{\mathcal{P}_0}$. In general, it is computationally inefficient to estimate $\mathcal{P}_i$'s in an unbiased simulations since $\mathcal{P}_i$'s might be orders of magnitude different for different λ_i's. To overcome this difficulty, it is customary to bias the system potential energy to allow low probability regions of the configuration space to be visited more frequently. More precisely, the probability of visiting the macrostate i is biased by introducing the bias factor η_i:

$$\mathbb{P}(\mathbf{x},\mathbf{p};\lambda_i) = \mathbb{P}_0(\mathbf{x},\mathbf{p})\exp[-\beta\eta_i] \qquad (3.15)$$

where $\mathbb{P}_0$ refers to the unbiased probability distribution and $\mathbb{P}$ refers to the biased distribution. Sampling is performed according to $\mathbb{P}$ while the averages of all observables are calculated in terms of $\mathbb{P}_0$.

There is no unique way of determining the value of η_i's. In the most widely used approach, known as *flat histogram methods*, η_i's are chosen so that $\mathbf{P}(\lambda_i) = \int \mathbb{P}(\mathbf{x},\mathbf{p};\lambda_i)d\mathbf{x}d\mathbf{p}$ is uniform in λ_i. There are different schemes for adjusting the η_i's to reach a flat distribution in λ_i, among them are themulticanonical method [170] and the Wang-Landau method [171, 172].

A good example of histogram methods applicable to hard particle systems is the *lattice switch Monte Carlo* method [173], which is used for calculating the free energy difference between two crystals. The position of a particle is decomposed into a *lattice component* $\mathbf{R}_i$, and a *displacement component* $\mathbf{u}_i$. In addition to conventional trial moves that change $\mathbf{u}_i$, the lattice type– and therefore the set of all $\mathbf{R}_i$'s– are also occasionally changed. For each lattice type, a bias factor is assigned, which is calculated in the course of simulation. The unbiased probability of visiting each crystal type is then calculated upon the convergence of bias factors. Histogram methods have also been used in free energy calculations of hard particles [17, 124].

Thermodynamic Integration In thermodynamic integration (TI) methods, the system is transformed from a macrostate 0 to a macrostate 1 through a reversible change in λ, a reaction coordinate or an order parameter. λ can be explicitly present

in the Hamiltonian, or be a generalized reaction coordinate, such as a torsional angle. In either case, it is easy to show that [174]:

$$\Delta A = \int_{\lambda_0}^{\lambda_1} \left\langle \frac{\partial \mathcal{H}}{\partial \lambda} \right\rangle_\lambda d\lambda \qquad (3.16)$$

Thermodynamic integration is relatively easy to implement if the Hamiltonian explicitly depends on λ; the integral in Eq. (3.16) can then be numerically estimated by choosing a sufficiently large number points in $[\lambda_0, \lambda_1]$ and estimating $\langle \partial \mathcal{H}/\partial \lambda \rangle_\lambda$ at each point in constant-λ simulations. It is however less straightforward to perform constant-λ simulations and estimate $dA/d\lambda$ if λ is a generalized coordinate of the system. More details on this latter case can be found in [174].

The most well-known example of thermodynamic integration is the method used for calculating the free energies of solids by reversibly transforming them to an Einstein crystal i.e. a group of independent Harmonic oscillators. The method was first used for continuous potentials by Broughton and Gilmer [175] and for discontinuous potentials by Frenkel and Ladd [176]. In the case of continuous potentials, the actual interparticle potential is gradually turned off and replaced by Harmonic springs that tether each particle to its average lattice position (and orientation). More precisely, the potential energy takes the form $U(\lambda) = (1 - \lambda)U_{\text{original}} + \lambda U_{\text{harmonic}}$; $\lambda = 0$ corresponds to the original system while at $\lambda = 1$, the system is fully composed of independent Harmonic springs. It is however not possible to turn off the hard core interaction gradually; the system potential is therefore changed along a path parameterized by $\lambda \in [0, \lambda_{\text{max}}]$ with $U(\lambda) = U_{\text{hard}} + \lambda U_{\text{harmonic}}$. For sufficiently large λ_{max}'s, the mobility of particles will be so restricted that the probability of an interparticle collision will be vanishingly small. The details of thermodynamic integration for nonspherical hard particles are presented at the end of this section.

It is noteworthy that these three categories differ in the level of discretization in interpolating between system 0 and system 1. In FEP methods, this transformation is instantaneous, and it is the equilibrium distribution of system 0 that is sampled, while in TI methods a reversible continuous path connects the two systems. Histogram methods are somewhere in between, with a few intermediate states also being sampled. It is however necessary for all these methods that the system that is sampled is always in *local* equilibrium.

3.3.2.2 Work-based (Non-equilibrium) Methods

In work-based methods, free energy differences are calculated from the work performed during an irreversible process. The Hamiltonian of system 0 is transformed in a finite time to the Hamiltonian of system 1 according to $\mathcal{H}[\lambda(t)]$ with $\mathcal{H}[\lambda(0)] = \mathcal{H}_0$ and $\mathcal{H}[\lambda(\tau)] = \mathcal{H}_1$. Jarzynski [177, 178] showed that the $\Delta A = A_1 - A_0$ is given by:

$$\exp[-\beta\Delta A] = \langle \exp[-\beta\mathcal{W}(\tau)] \rangle_0 \tag{3.17}$$

where $W(\tau)$ is the work performed in the irreversible process. Configurations sampled from the equilibrium distribution of system 0 are transformed into system 1 according to $\mathcal{H}[\lambda(t)]$ and $W(\tau)$ is calculated for each of them. Eq. (3.17) is then used to estimate ΔA. Although each configuration is initially in equilibrium, the irreversible process turns it into a *non-equilibrium* configuration of system 1. It can be shown that TI and FEP methods are indeed limiting cases of work-based methods when $\tau \to \infty$ and $\tau \to 0$ respectively. (See Appendix A.)

3.3.3 Implicit Methods

The methods discussed in this section are modified sampling methods optimized for studying first-order phase transitions and are not free energy calculation methods *per se*. Nevertheless they are still discussed due to their relevance to the problem of mapping phase diagrams. These methods are called *implicit* since the condition of the equality of chemical potentials between coexisting phases is implicitly imposed during a simulation.

Isolation of A Single Point on the Phase Boundary   In thermodynamic equilibrium, the temperature, the pressure and the chemical potential of every component in the system should be the same in all coexisting phases. The exchange of heat, work and matter that leads to these equalities occurs through the interfaces. However, there is a free energy penalty associated with every interface. Such penalties are vanishingly small in real systems, but in simulations where the number of particles are in the range of $10^2 - 10^6$, these interfaces are thermodynamically more expensive and hence difficult to form.

In order to study the coexistence problem in simulation, a class of methods have been developed in which the coexisting phases are allowed to exchange heat, work and particles without having a real interface. The most well-known of them is the Gibbs Ensemble Monte Carlo (GEMC) method developed by Panagiotopoulos [179]. Two

(or more) independent simulation boxes are simultaneously simulated. These boxes do not interact; i.e. every particle interacts with the particles within its own box. The conditions of thermodynamic equilibrium are enforced by allowing the boxes to exchange volume or particles with the constraint that the total volume of the system V and the total number of particles N are fixed. The acceptance probability of every trial move is determined from the following probability distribution:

$$\mathcal{P}(n_1, V_1, \mathbf{s}_1^{n_1}, \mathbf{s}_2^{N-n_1}) = \frac{1}{Q_G(N,V,T)V\Gamma^{3N}} \frac{V_1^{n_1}(V-V_1)^{N-n_1}}{n_1!(N-n_1)!} e^{-\beta[U(\mathbf{s}_1^{n_1})+U(\mathbf{s}_2^{N-n_1})]} \quad (3.18)$$

where Q_G, the Gibbs ensemble partition function, is given by:

$$
\begin{aligned}
Q_G(N,V,T) &= \sum_{n_1=0}^{N} \frac{1}{V\Gamma^{3N}n_1!(N-n_1)!} \int_0^V dV_1 V_1^{n_1}(V-V_1)^{N-n_1} \\
&\times \int d\mathbf{s}_1^{n_1} e^{-\beta U(\mathbf{s}_1^{n_1})} \int d\mathbf{s}_2^{N-n_1} e^{-\beta U(\mathbf{s}_1^{N-n_1})}
\end{aligned}
\quad (3.19)
$$

The coexistence densities are thus calculated from the densities of two boxes after equilibration. GEMC is a powerful tool for studying fluid/fluid coexistence problems. It is however less efficient when one of the coexisting phases is a crystal, a liquid crystal, or a dense fluid since it will take much longer to achieve chemical equilibrium due to relatively low probabilities of accepting particle exchange trial moves. Specific methods such as semigrand ensemble Monte Carlo [180] have been developed for dense systems.

Gibbs-Duhem Integration: Computing an Entire Coexistence Curve   If a single point on a coexistence curve is known from explicit free energy calculation or a GEMC simulation, the Gibbs-Duhem integration method proposed by Kofke [181] is an elegant way of calculating the rest of the coexistence curve through iterative integration of the Clasius-Clapyron equation (Eq. 3.21). For a pure substance the Gibbs-Duhem equation can be expressed as:

$$d(\beta\mu) = hd\beta + \beta v dP \quad (3.20)$$

For two coexisting phases of α and γ, the chemical potential is identical along the coexistence curve and $d\mu^\alpha = d\mu^\gamma$. Using Eq. 3.20 for both phases and after some

rearrangements one obtains:

$$\left(\frac{dP}{d\beta}\right)_\sigma = -\frac{h^\gamma - h^\alpha}{\beta(v^\gamma - v^\alpha)} \tag{3.21}$$

The subscript σ means that differentiation is performed along the saturation curve. Eq. 3.21 can be integrated from (β_0, P_0), an arbitrary point on the saturation line, to obtain the entire saturation curve. Since every quantity on the right hand side of Eq. 3.21 can be accurately obtained from simulation, two independent (NPT) simulations of two coexisting phases can be started from (β_0, P_0), and the values of P and β can be adjusted accordingly. Although the simulations are independent, the condition of chemical equilibrium is implicitly enforced by iterative integration of the Clasius-Clapyron equation.

3.3.4 Frenkel-Ladd Method for Anisotropic Hard Particles[6]

The free energy of a (quasi-)crystal is calculated using the Frenkel-Ladd method [176, 183] by transforming it reversibly into an Einstein crystal, which serves as a reference structure with known free energy. In the Einstein crystal, each particle is tethered to its average lattice position via harmonic springs. Although originally developed for spherical particles, this method can be extended to particles with rotational degrees of freedom, such as tetrahedra. Additional springs are needed to tether the orientations of the particles to their average orientations in the lattice. Alternative extensions of the Frenkel-Ladd method to systems of particles with rotational degrees of freedom can be found in the literature [184].

We describe the configuration of a tetrahedron by $(\mathbf{r}, \mathbf{q})$, with $\mathbf{r}$ being its center of mass position and $\mathbf{q}$ the unit quaternion describing its orientation. The potential energy of of the corresponding Einstein crystal can then be expressed as:

$$\frac{U(\mathbf{r}^N, \mathbf{q}^N)}{k_B T} = \sum_{i=1}^{N} \frac{||\mathbf{r}_i - \mathbf{r}_{i,0}||^2}{\sigma^2} + c\sum_{i=1}^{N} ||\mathbf{q}_i - \mathbf{q}_{i,0}||^2 \tag{3.22}$$

where $\mathbf{r}_{i,0}$ and $\mathbf{q}_{i,0}$ are the reference position and the reference orientation of the i-th particle in the crystal. The constant c allows us to adjust the relative strength of the rotational springs and does not affect the computed free energy differences. All the results in this study are obtained using a value of $c = 1/2$; we tested that using other

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[6]This section is partly based on the following manuscript: Amir Haji-Akbari, Michael Engel and Sharon C. Glotzer, *J. Chem. Phys.* **135**: 194101 (**2011**) [182].

values of c does not affect the outcome of the calculations.

Each system is transformed to the Einstein crystal along a reversible path parameterized by $\gamma \in [0, \gamma_{\text{max}}]$ using the isochoric-isothermal (NVT) ensemble and the Hamiltonian

$$\mathcal{H}(\mathbf{r}^N, \mathbf{q}^N; \gamma) = \mathcal{H}_{\text{hard}}(\mathbf{r}^N, \mathbf{q}^N) + \gamma U(\mathbf{r}^N, \mathbf{q}^N) \tag{3.23}$$

The hard particle system with Hamiltonian $\mathcal{H}_{\text{hard}}$ corresponds to $\gamma = 0$, while in the limit $\gamma \to \infty$ the Einstein crystal is obtained. In practice, we can stop at a sufficiently large value of γ_{max} when the springs are strong enough to suppress any particle collisions. The Helmholtz free energy difference $\Delta A = A_{\text{Ein}} - A_{\text{hard}}$ between the reference Einstein crystal and the hard particle system is given by:

$$\Delta A = \int\limits_0^{\gamma_{\text{max}}} \left\langle \frac{\partial \mathcal{H}(\gamma)}{\partial \gamma} \right\rangle_\gamma \mathrm{d}\gamma = \int\limits_0^{\gamma_{\text{max}}} \langle U \rangle_\gamma \mathrm{d}\gamma \tag{3.24}$$

Note that the Frenkel-Ladd method can only be used if there is no translational or rotational diffusion in the system; otherwise the ensemble average $\langle U \rangle_\gamma$ will not be well defined for small values of γ.

In our simulations of hard tetrahedra and hard triangular bipyramids, the system is held for 2×10^5 Monte Carlo cycles at each γ value during which $\langle U \rangle_\gamma$ is evaluated. The integral in equation (3.24) is then computed numerically. This allows us to determine the Gibbs free energy $G = A + PV$ of different crystals in a pressure range where no configurational rearrangements are observed in any of crystals. The free energy difference $\Delta G = G_X - G_Y$ is extrapolated to pressures outside this range using thermodynamic integration in addition to the Frenkel-Ladd method [185]:

$$\frac{\Delta G(P^*)}{Nk_BT} = \frac{\Delta G(P_0^*)}{Nk_BT} + \frac{V_p}{\sigma^3} \int\limits_{P_0^*}^{P^*} \left[\frac{1}{\phi_X(p)} - \frac{1}{\phi_Y(p)} \right] \mathrm{d}p \tag{3.25}$$

where X and Y are the crystal structures that are compared using this method, and V_p is the volume of an individual particle.

3.3.4.1 Fluid-solid Transition

We determine the melting pressure P_M^* by calculating the absolute free energies of the solid and fluid. For sufficiently large values of γ, the Helmholtz free energy of

the Einstein crystal is given by [183]:

$$\frac{A_{\mathrm{Ein}}}{Nk_BT} = -\frac{3}{2}\frac{N-1}{N}\ln\frac{\pi}{\gamma} - \frac{3}{2}\ln\frac{\pi}{c\gamma} - \ln N_{\mathrm{sym}} + 3\frac{N-1}{N}\ln\frac{\Lambda}{\sigma} \qquad (3.26)$$

where $\Lambda = h/(2\pi m k_B T)^{1/2}$ is the de Broglie wavelength. N_{sym} is the number of quaternions corresponding to orientations that are symmetry-equivalent, which is twice the order of the rotation group of the particle. The factor 2 arises from the fact that quaternions are inherently degenerate in describing the orientation; i.e. $\mathbf{q}$ and $-\mathbf{q}$ correspond to the same rotation matrix. For a non-symmetric particle, the rotation group will have one element (identity) only and $N_{\mathrm{sym}} = 2$. Here, for tetrahedra, the rotation group has twelve elements, so $N_{\mathrm{sym}} = 24$. For triangular bipyramids, the rotation group has six elements and $N_{\mathrm{sym}} = 12$. The first and the second terms are configurational contributions resulting from the translational and rotational springs. The last term corresponds to momentum contributions due to translational degrees of freedom. Momentum contributions due to rotational degrees of freedom are identical for the fluid and the solid and are therefore not included here.

The Gibbs free energy of an ideal gas, which approximates a real gas in the limit of infinite dilution, is

$$\frac{G_{\mathrm{id}}}{Nk_BT} = \ln\frac{P^*}{2\pi^2} + \frac{\ln(2\pi N)}{2N} + 3\ln\frac{\Lambda}{\sigma} \qquad (3.27)$$

The free energy of the fluid phase is then obtained from thermodynamic integration of the equation of state [186]:

$$\frac{G_{\mathrm{fluid}}(P^*)}{Nk_BT} = \frac{G_{\mathrm{id}}(P^*)}{Nk_BT} + \int_0^{P^*}\left[\frac{V_T/\sigma^3}{\phi(p)} - \frac{1}{p}\right]\mathrm{d}p \qquad (3.28)$$

3.4   Free Volume Calculation[7]

The free volume of a hard sphere is the volume of the region of space in which the sphere can be moved continuously without overlapping with its neighbors while keeping all the other particles fixed [187]. The definition generalizes to anisotropic particles with rotational degrees of freedom where free volume v_f is now the volume of the largest subset of configurational space connected to the origin that can be

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[7]This section is partly based on the following manuscript: Amir Haji-Akbari, Michael Engel and Sharon C. Glotzer, *J. Chem. Phys.* **135**: 194101 (**2011**) [182].

accessed by a given particle while fixing the positions and orientations of all other particles [188]:

$$v_f = \int I(\mathbf{r}, \mathbf{q}) \mathrm{d}^3\mathbf{r}\mathrm{d}^3\mathbf{q} \qquad (3.29)$$

Here, $I(\mathbf{r}, \mathbf{q})$ is the indicator function of motions $(\mathbf{r}, \mathbf{q})$ consisting of a translation by $\mathbf{r}$ and a rotation by $\mathbf{q}$ and connected to the origin. I is unity if the particle does not overlap with any other particle and zero otherwise. Due to the inherent periodicity of rotational motion, the free volume of an anisotropic particle has generally a more complicated topology compared to the free volume of a sphere. Here we calculate free volumes at high densities where the free volume is simply connected.

3.4.1 Shooting Method

We calculate the free volume of a particle using a new method we call the *shooting method* [182]. Let $(\mathbf{u}, \mathbf{v})$ correspond to a unit vector in the six dimensional configuration space and suppose that particle i is 'shot' in this direction until it hits another particle. The 'shooting distance' is the smallest value of α for which the particle first overlaps with its neighbors if translated by $\alpha\mathbf{u}$ and oriented according to the quaternion $(\mathbf{q}_i + \alpha\mathbf{v})/||\mathbf{q}_i + \alpha\mathbf{v}||$.

A lower bound for the free volume can be obtained by averaging over a sufficiently large number N_s of shots with shot distances α_j along randomly chosen directions:

$$v_f \gtrapprox \lim_{N_s \to \infty} \frac{1}{N_s} \sum_{j=1}^{N_s} \frac{\pi^3}{6} \alpha_j^6 \qquad (3.30)$$

Here $\pi^3/6$ is the volume of the six-dimensional unit sphere. Note that the periodic topology and the curvature of the six-dimensional configuration space are ignored, which is acceptable at high packing densities because $||\Delta\mathbf{q}|| \ll 1$.

Eq. (3.30) is a lower bound for concave free volumes, because shooting only allows access to the parts of the free volume connected to the origin by a straight line. Non-convex free volumes can arise from sliding collisions which, however, become increasingly rare at high packing densities. In fact, as we will show now for tetrahedra, the shooting method is accurate for high enough packing densities.

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3.4.2 Binning Method

To estimate the amount of error in the shooting method introduced by non-convexity, we using the alternative *binning method* which corresponds to a Monte Carlo integration of the free volume. The configuration space of a given particle is partitioned into N_{bins} small radial bins of volume V^{bin}. We perform N_t random ghost trial moves per bin to average out the orientational degrees of freedom and determine the number N^{NO} of trial moves not leading to an overlap. Free volume can then be estimated from:

$$v_f \gtrapprox \frac{1}{N_t} \sum_{j=1}^{N_{\text{bins}}} V_j^{\text{bin}} N_j^{\text{NO}} \tag{3.31}$$

Binning is much slower than shooting and might overestimate the free volume, if a trial move discovers an area of configuration space without overlap, but not connected to the original particle position. We find that the average of the logarithms of the free volumes calculated from the shooting method and the binning method agree within a relative error of 10^{-2} for all densities $\phi \geq 70\%$.

3.4.3 Mean-field Approximation

The distribution of free volumes is related to the entropy of a hard particle system in the mean-field approximation. If we assume that free volumes of neighboring particles are uncorrelated, then the partition function of the system is expressed as $Q_{\text{mf}} = \prod_{i=1}^{N} v_{f,i}$ and the Helmholtz free energy as $A_{\text{mf}}/Nk_BT = -\langle \ln v_f \rangle$. The thermodynamically relevant quantity is therefore the mean-log average of free volumes:

$$v_{f,\text{ML}} \quad := \quad \exp\langle \ln v_f \rangle \tag{3.32}$$

which will be used in the rest of this study instead of the simple average $\langle v_f \rangle$.

3.5 Analysis Tools

3.5.1 Pressure Estimation[8]

The acceptance probability of trial volume changes is an estimator of the pressure in Monte Carlo simulations [183]. Consider a trial expansion that increases the volume

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[8]This section is partly based on the following manuscript: Amir Haji-Akbari, Michael Engel and Sharon C. Glotzer, *J. Chem. Phys.* **135**: 194101 (**2011**) [182].

from V to $V + \Delta V$. To fulfill detailed balance, the acceptance probability of the volume change is given by the Boltzmann factor,

$$P_{\mathrm{B}} = \exp\left\{-\frac{P^*\Delta V}{\sigma^3} + N\ln\left(1 + \frac{\Delta V}{V}\right)\right\} \tag{3.33}$$

On the other hand, a trial compression that decreases the volume from V to $V - \Delta V$ is accepted if and only if no overlap is generated by the trial volume change. Let P_{NO} be the probability to generate an overlap in the trial compression. For small ΔV and in equilibrium the probabilities are equal, $P_{\mathrm{NO}} = P_{\mathrm{B}}$, and we can solve for the pressure:

$$P^* = \lim_{\Delta V \to 0}\left\langle \frac{N\sigma^3}{\Delta V}\left[\ln\left(1 + \frac{\Delta V}{V}\right) - \frac{1}{2}\ln p_{\mathrm{NO}}\right]\right\rangle \tag{3.34}$$

Here, $p_{\mathrm{NO}} = P_{\mathrm{NO}}^{2/N}$ is the probability of a single particle not having any overlap with any other particle after the trial compression that decreases the volume by ΔV.

3.5.2 Diffraction Patterns[9]

Diffraction images are powerful tools for studying periodic and quasiperiodic structures and will be extensively used in this dissertation. In the real world, diffraction images are obtained from small angle scattering experiments, where the transferred wave is perpendicular to the incoming and outgoing waves if the scattering angle is too small. For instance, if the incoming wave is in the z direction, the diffraction image on the xy plane is given by $|S(q_x, q_y)|^2$ with $S(q_x, q_y)$, the *structure factor*, given by:

$$S(q_x, q_y) \;=\; \int \rho(x, y, z)e^{-i(q_x x - q_y y)}dx dy dz \tag{3.35}$$

where $\rho(x, y, z)$ is the density function. For a set of n discrete point particles, Eq. (3.35) takes the form:

$$S(q_x, q_y) \;=\; \sum_{p=1}^{n} e^{-iq_x x_p - iq_y y_p} \tag{3.36}$$

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[9]Diffraction pattern and bond order diagram calculation algorithms are incorporated by Dr. Michael Engel to the visualization code `injavis`.

The diffraction images presented in this study are all calculated by assuming that scatterers are point particles. The positions of these scatterers are projected onto a two-dimensional plane perpendicular to the viewing direction $\mathbf{z}$. The plane is then sheared to obtain a square grid, which is then fast-Fourier-transformed to get the sheared structure factor. The shear is undone before calculating the diffraction image from the structure factor [189].

3.5.3 Bond Order Diagrams

Bond order diagrams will be extensively used in the remainder of this thesis. Here is the precise mathematical definition of a bond order diagram. Let $\mathcal{P} = \{\mathbf{r}_i\} \subset \mathbb{R}^3$ be a countable set of points in the three-dimensional Euclidean space, and let $f : \mathbb{Z}^2 \to \{0, 1\}$ be a given function with the property that $f(i, j) = 1$ if i and j are connected with a bond and zero otherwise. The bond order diagram is then obtained by calculating $\mathbf{u}_{ij} = (\mathbf{r}_j - \mathbf{r}_i) / \|\mathbf{r}_j - \mathbf{r}_i\|$ for each connected pair (i, j) and projecting it onto the surface of S^2, the three-dimensional unit sphere. The bond order diagram is the intensity function $\psi : S^2 \to \mathbb{R}^{\geq 0}$ which can be approximated by discretizing S^2 into bins and dividing the number of projections onto a certain bin by its surface area. A smooth function ψ can be obtained, if the surface area of each bin is sufficiently small, and the number of pairs in $\mathcal{P}$ is sufficiently large. For all bond order diagrams presented in this dissertation, $\mathcal{P}$ is the positions of centers of mass particles in a simulation. Usually, particles are defined neighbors if their distance lies within a certain range, different connectivity criteria will, however, be used occasionally.

CHAPTER IV

Quantification of Rotational Order For Systems of Symmetric Objects[1]

4.1 Introduction

Rotational order can in general arise in arrangements of nonspherical molecules or particles. It can be accompanied by translational order– as in crystals, quasicrystals and smectic liquid crystals– or it can be stand-alone– e.g. in nematic liquid crystals. Identifying and quantifying rotational order is of immense interest for condensed matter physicists. There are standard tools for detecting and quantifying translational order [190–192], while identifying rotational order is more challenging and the existing tools are far too complicated and confusing to be used in general [193, 194].

The orientation of an arbitrary object is fully determined by one polar angle in two dimensions and three Euler angles in three dimensions, This representation becomes degenerate for symmetric objects; i.e. objects with nontrivial rotation groups as the symmetry operations that keep the orientation of the object intact might change the values of these angles. (This is schematically depicted in an example in Fig. 4.1.)

Historically, the orientations of symmetric objects or symmetric phases have been described by the smallest non-vanishing symmetric traceless tensor that is invariant under the action of its rotation group [193]. A list of these tensors for some major symmetry groups in three dimensions is given in Table 1 of [194]. Such a tensor will not necessarily be a rotational coordinate, however. As an example, consider a square slab in three dimensions, which has a rotation group of D_4 (Fig. 4.2). The traceless symmetric tensor $S^{ij} = x^i x^j + y^i y^j - (2/3)\delta^{ij} = (1/3)\delta^{ij} - z^i z^j$ is invariant under D_4 but it is not a rotational coordinate since any rotation around $\mathbf{z}$ leaves S^{ij} invariant while changing the orientation of the slab.

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[1]The content of this chapter is based on a manuscript that is under preparation.
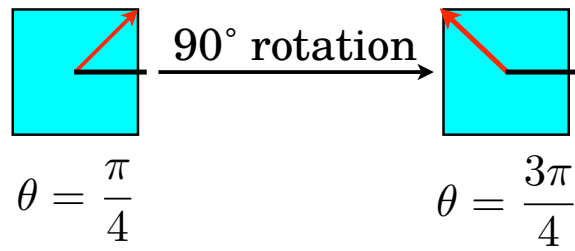
Figure 4.1: Degeneracy of polar angles for describing the orientation of a square in two dimensions.
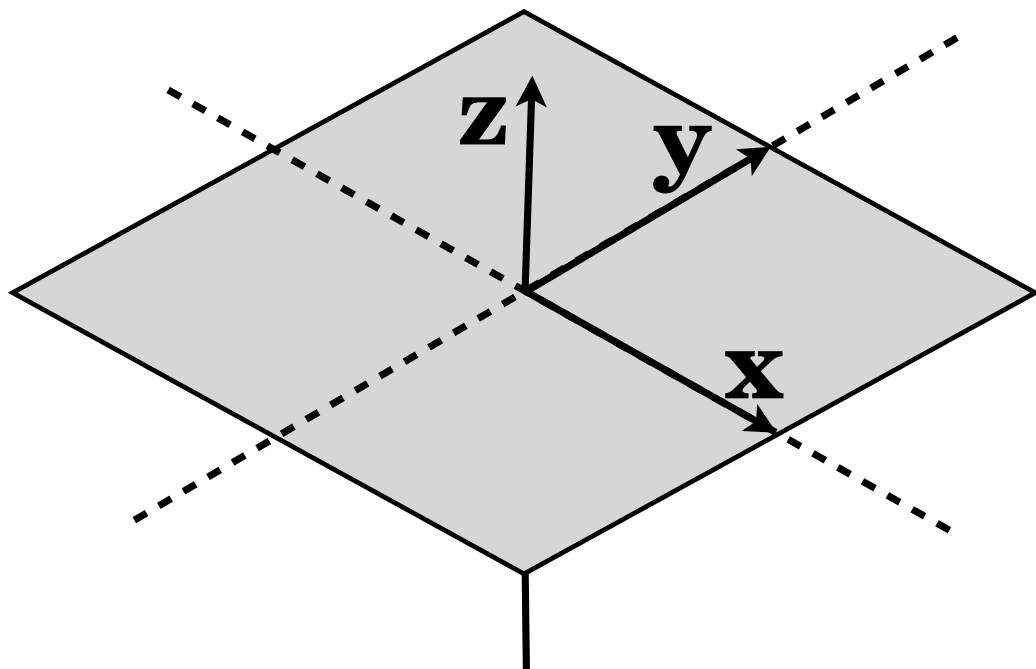
Figure 4.2: A square slab in three dimensions.

In this chapter, a general procedure is proposed for deriving rotational coordinates for symmetric objects. These coordinates, which are tensors with ranks depending on the type of symmetry, are bijective; i.e. each coordinate is associated with one and only one distinct rotation. Rotationally ordered phases of symmetric building blocks are then described using probability distributions of these rotational coordinates and order parameters are obtained as expected values of suitable moments of such coordinates. The extent and the geometry of order can be determined by solving a generalized optimization problem.

This chapter is organized as follows. We introduce the utilized notations and conventions in Section 4.2. We define the notion of strong orientational coordinate (SOC) In Section 4.3.1. In Section 4.3.2, we use the machinery of strong orientational coordinates and group theory to derive rotational coordinates for symmetric objects. We then derive the corresponding SOCs for all two- and three-dimensional rotation groups. In Section 4.3.3, we use distribution functions of SOCs to quantify different types of rotational order. We then use this framework to define orientational order parameters for several liquid crystalline phases. Finally in Section 4.4 we discuss further potential applications of SOCs other than the quantification of global orientational order.

Before closing the introduction, it is necessary to note that our approach is purely geometrical; i.e. we are not concerned about the physical realizability of the described structures; however our goal is to develop computational tools for quantifying such structures *in the event that they form.* Finally, since these tools will be used to quantify structures formed by systems of particles, we will use the terms 'object' and 'particle' interchangeably.

4.2 Notations And Definitions

Rotational coordinates derived in this chapter are contravariant tensors of different ranks. All tensors of rank 2 and higher will be denoted in capitalized italics (i.e. S, T) irrespective of their rank. Vectors, however, will be depicted as italicized small letters i.e. v, w etc. In all linear algebraic operations, vectors will always be considered 'column vectors'. The conventional Einstein notation will also be used for vectors and tensors frequently throughout this work. Finally, all scalars will be denoted by Greek letters (α, β, etc) respectively.

We denote the r-adic power of vector $v \in \mathbb{R}^d$ as v^r i.e. $(\mathbf{v}^r)^{i_1 i_2 \cdots i_r} = v^{i_1} v^{i_2} \cdots v^{i_r}$. Similarly, the r-adic product of r distinct vectors $v_1, v_2, \cdots, v_r$ will be denoted by

$v_1 v_2 \cdots v_r$. For every set of vectors $V = \{v_1, v_2, \cdots, v_n\}$, a rank r homogeneous tensor is defined by:

$$\mathcal{H}_r(V) \ := \ \sum_{p=1}^{n} v_p^r \tag{4.1}$$

For two arbitrary-rank tensors S and T, a generalized inner product $S \odot T$ is defined as the full contraction between them given by:

$$S \odot T \ = \ \overline{S^{i_1, i_2, \cdots, i_r}} T^{i_1, i_2, \cdots, i_r} \tag{4.2}$$

$\odot$ reduces to the Euclidean inner product for $r = 1$ i.e. when S and T are both vectors. Associated with this inner product, a generalized Frobenius norm of a rank r tensor is defined as:

$$\|S\|_F = (S \odot S)^{1/2} \tag{4.3}$$

Note that the Frobenius norm reduces into the Euclidean norm for $r = 1$; i.e. vectors and the matrix Frobenius norm for $r = 2$.

4.3 Tensor Order Parameters- General Construction

We derive proper orientational order parameters for arrangements of symmetric particles as follows. In Section 4.3.1, we develop the machinery of homogeneous tensors as tools of describing the orientation of an an arbitrary set of vectors in $\mathbb{R}^d$ and define the notion of *strong orientational coordinate* of a set. We then map the orientation of a symmetric object to a set of equivalent vectors in Section 4.3.2 and use the SOCs derived in Section 4.3.1 as rotational coordinates of symmetric objects. Finally in Section 4.3.3 we use distribution functions of these SOCs to identify and quantify order in orientationally ordered arrangements of symmetric objects.

4.3.1 Homogeneous Tensors

Let $V = \{v_1, v_2, \cdots, v_p\}$ be a finite set of unit vectors in $\mathbb{R}^d$ and let $W = \{Qv : v \in V\}$ with $Q \in O(d)$ an orthogonal transformation. A function $\mathcal{H}(V)$ is called a *strong orientational coordinate* of V if $\mathcal{H}(QV) = \mathcal{H}(V)$ implies $QV = V$. Since a set has no particular ordering, such a function should be invariant under permutations of elements of V, a property satisfied by homogeneous tensor forms defined in Eq. (4.1).

With the following chain of theorems, we establish that for any V, there exists an even number $2q \leq p$ and an odd number $2r - 1 \leq p$ so that $V = QV$ if and only if $\mathcal{H}_{2q}(V) = \mathcal{H}_{2q}(QV)$ and $\mathcal{H}_{2r-1}(V) = \mathcal{H}_{2r-1}(QV)$.

Lemma 1. *Let* $a_1, a_2, \cdots, a_n, b_1, b_2, \cdots, b_n \in \mathbb{R}$, *Then* $\{a_1, a_2, \cdots, a_n\} = \{b_1, b_2, \cdots, b_n\}$ *(up to multiplicities) if and only if* $\sum_{i=1}^{n} a_i^k = \sum_{i=1}^{n} b_i^k$ *for* $1 \leq k \leq n$.

Proof. Form the polynomials $p(z) = \prod_{i=1}^{n}(z - a_i)$ and $q(z) = \prod_{i=1}^{n}(z - b_i)$ and expand them to obtain $p(z) = z^n + \sum_{i=1}^{n}(-1)^i \alpha_i z^i$ and $q(z) = z^n + \sum_{i=1}^{n}(-1)^i \beta_i z^i$ where the coefficients are given by:

$$\phi_k = \sum_{i=1}^{k-1}(-1)^{i-1}\alpha_i \phi_{k-i} + (-1)^{k-1} k \alpha_k$$

$$\psi_k = \sum_{i=1}^{k-1}(-1)^{i-1}\beta_i \psi_{k-i} + (-1)^{k-1} k \beta_k$$

where $\phi_k = \sum_{i=1}^{n} a_i^k$ and $\psi_k = \sum_{i=1}^{n} b_i^k$. Note that $\phi_1 = \psi_1$ if and only if $\alpha_1 = \beta_1$ and by induction $\alpha_k = \beta_k$ for every $1 \leq k \leq n$. Thus $p(z)$ and $q(z)$ have the same coefficients, and thus the same roots. $\square$

Theorem 2. *Let* $V = \{v_1, v_2, \cdots, v_p\}$ *be an arrangement of* p *unit vectors in* $\mathbb{R}^d$ *and let* $W = \{Qv : v \in V\}$ *where* $Q \in O(d)$ *is an orthogonal transformation. Then* $V = W$ *if and only if* $\mathcal{H}_k(V) = \mathcal{H}_k(W)$ *for every* $k \leq p$.

Proof. If $V = W$, $\mathcal{H}_k(V) = \mathcal{H}_k(W)$ for every k including $k \leq p$. Now suppose $\mathcal{H}_k(V) = \mathcal{H}_k(W)$ for $1 \leq k \leq p$, which implies that $[\mathcal{H}_k(V) - \mathcal{H}_k(W)] \odot S = 0$ for any S. Set $S = v_i^k$ and obtain:

$$v_i^k \odot [\mathcal{H}_k(V) - \mathcal{H}_k(W)] = \sum_{j=1}^{p}\left[(v_i^T v_j)^k - (v_i^T w_j)^k\right]$$

$$= \sum_{j=1}^{p}\left[\mu_{ij}^k - \xi_{ij}^k\right]$$

where $\mu_{ij} = v_i^T v_j$ and $\xi_{ij} = v_i^T w_j$. Applying Lemma 1 yields $\{\xi_{i1}, \cdots, \xi_{ip}\} = \{\mu_{i1}, \cdots, \mu_{ip}\}$ for every i. We therefore have $\{\xi_{ij}\}_{i,j=1}^{p} = \{\mu_{ij}\}_{i,j=1}^{p}$ which is only possible if $V = W$. $\square$

This means that the $p-$tuple $(\mathcal{H}_1, \mathcal{H}_2, \cdots, \mathcal{H}_p)$ uniquely specifies the orientation of V; however we are interested in a single orientational coordinate. The following results refine the scope of our search.

Theorem 3. *Let V and W as defined in Theorem (2) and let $\mathcal{H}_r(V) \neq \mathcal{H}_r(W)$ for some integer r. Then $\mathcal{H}_{r+2}(V) \neq \mathcal{H}_{r+2}(W)$.*

Proof. From the properties of the norm it follows that:

$$||\mathcal{H}_r(V) - \mathcal{H}_r(W)||_F = \sum_{i,j=1}^{p} \left[\mu_{ij}^r - \xi_{ij}^r \right] > 0$$

Let $S^{i_1,i_2,\cdots,i_{r+2}} = \delta^{i_1 i_2}(\mathcal{H}_r(V) - \mathcal{H}_r(W))^{i_3,i_4,\cdots,i_{r+2}}$ and calculate $(\mathcal{H}_{r+2}(V) - \mathcal{H}_{r+2}(W) \odot S$ to get:

$$
\begin{aligned}
(\mathcal{H}_{r+2}(V) - \mathcal{H}_{r+2}(W)) \odot S &= \sum_{i=1}^{p} \left[(v_i^T I v_i) v_i^r \odot \mathcal{H}_r - (w_i^T I w_i) w_i^r \odot \mathcal{H}_r \right] \\
&= 2\mathcal{H}_r \odot \mathcal{H}_r > 0
\end{aligned}
$$

since $v_i^T I v_i = w_i^T I w_i = 1$. $\qquad\square$

Corollary 4. *Let V and W be as defined in Theorem (2) then there exists integers q, r with $2q \leq p$ and $2r - 1 \leq p$ so that $V = W$ if and only if $\mathcal{H}_{2q}(V) = \mathcal{H}_{2q}(W)$ and $\mathcal{H}_{2r-1}(V) = \mathcal{H}_{2r-1}(W)$.*

Proof. Let $Q \in O(d)$ so that $QV \neq V$. According to Theorem 2, there exists $s \leq p$ so that $H_s(V) \neq H_s(QV)$. Pick the smallest such integer and denote it s_Q. Define $E = \{Q; s_Q \text{ is even}\}$ and $O = \{Q; s_Q \text{ is odd}\}$. Taking $q = \frac{1}{2} \max_{Q \in E} s_Q$ and $r = \frac{1}{2}(1 + \max_{Q \in O} s_Q)$ completes the proof. Note that if either E or O is empty, one can pick any even or odd integer and the proof follows. $\qquad\square$

This allows us to refine the search for even arrangement of vectors for which every odd-rank homogeneous form vanishes.

Definition 1. A set V is **even** if for every $v \in V$, $-v \in V$ and **odd** if for every $v \in V, -v \notin V$.

For arrangements of vectors which are neither even nor odd, both an odd and even homogeneous form is needed for specifying their orientation since an even symmetric tensor form will be invariant under inversion which will change the orientation of V while an odd symmetric tensor form will be invariant under any orthogonal transformation which changes the orientation of the even subset while keeping the odd subset invariant. Intuitively, one expects the SOC of an even set to be even-ranked and the SOC of an odd set to be odd-ranked. This is proven in the following theorems.

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Corollary 5. *Let V and W be as defined in Theorem (2). If V and W are even, then there exists an integer $2q \leq p$ so that $V = QV$ if and only if $H_{2q}(V) = H_{2q}(QV)$.*

Proof. Applying Corollary (4) and note that $\mathcal{H}_{2r+1}(V) = 0$ for every r. $\qquad\square$

Lemma 6. *Let $x_1, \cdots, x_n, y_1, \cdots, y_n \in \mathbb{R}$ with the property that for every distinct $i, j \leq n$, if $x_i \neq 0$, then $x_i + x_j \neq 0$. Then $\{x_1, \cdots, x_n\} = \{y_1, \cdots, y_n\}$ (up to multiplicities) if and only if $\sum_{i=1}^n x_i^{2k-1} = \sum_{i=1}^n y_i^{2k-1}$ for every $k \leq n$.*

Proof. The proof follows from Proposition 4.1 in [195]. $\qquad\square$

Theorem 7. *Let V and W - as defined in Theorem (2)- be odd sets, then their exists $q \leq p$ so that $\mathcal{H}_{2q-1}(V) = \mathcal{H}_{2q-1}(W)$ if and only if $V = W$.*

Proof. If $V = W$, then $\mathcal{H}_{2q-1} = \mathcal{H}_{2q-1}(W)$ for every $q > 0$. Now let $\mathcal{H}_{2q-1}(V) = \mathcal{H}_{2q-1}(W)$ for every $q \leq p$, then for every $i, q \leq p$ we have $v_i^{2q-1} \odot [\mathcal{H}_{2q-1}(V) - \mathcal{H}_{2q-1}(W)] = \sum_{j=1}^p (\mu_{ij}^{2q-1} - \xi_{ij}^{2q-1}) = 0$; however μ_{ij}'s and ξ_{ij}'s satisfy the conditions specified in Lemma (6) since the odd-ness of V implies that for every $v_j \in V$, $-v_j \notin V$ and thus $v_i^T(-v_j)$ will not be among μ_{ij}'s and so is true for ξ_{ij}'s. Thus according to Lemma (6), $\{\xi_{ij}\} = \{\mu_{ij}\}$ (up to multiplicities) for every i, which implies that $V = W$. The proof is closed by applying Theorem (3). $\qquad\square$

Up to now, we have established upper bounds on the tensorial rank of strong orientational coordinates of even and odd sets. These upper bounds are generic since they only depend on the cardinality of the underlying set and not its structure. In general, tensor SOCs of smaller ranks might be possible for sets with certain symmetries. However, as it will be seen in Section 4.3.2, these bounds are still tight for certain types of symmetries.

4.3.2 Symmetric Objects

The strong orientational coordinates developed in Section 4.3.1 are 'permutation-invariant'; they can be used for describing the orientations of symmetric objects if a bijection is established between the orientation of a symmetric object and a set of 'equivalent' vectors. We will do this by generating orbits of suitable vectors under the action of the rotation group of the object. We are only using the rotation group since in a physical system, the orientation of an object can only change through rotations.

A rigid body $\mathcal{R} \subset \mathbb{R}^d$ is symmetric if there is a non-identity $Q \in SO(d)$ that $Q\mathcal{R} = \mathcal{R}$. The *rotation group* of a rigid body $\mathcal{R}$ is defined as:

$$\mathcal{G}_{\mathcal{R}} := \{Q \in SO(d) : Q\mathcal{R} = \mathcal{R}\} \tag{4.4}$$

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For every vector $v \in \mathcal{R}$, its *orbit* is defined as:

$$\mathcal{O}_v \ := \ \{Qv : Q \in \mathcal{G}_\mathcal{R}\} \tag{4.5}$$

In Lie groups orbits can be uncountably infinite sets.

Theorem 8. *$\mathcal{G}_\mathcal{R}$ has the following properties:*

1. *$\mathcal{G}_\mathcal{R}$ partitions $\mathcal{R}$ into equivalency classes i.e. $\mathcal{O}_v = \mathcal{O}_{Qv}$ for every $v \in \mathcal{R}$ and $Q \in \mathcal{G}_\mathcal{R}$.*

2. *$\mathcal{O}_v$ is either even or odd.*

Proof. (1) directly follows from group properties. If for every $u \in \mathcal{O}_v$, $-u \notin \mathcal{O}_v$, then $\mathcal{O}_v$ is odd an (2) follows. Suppose there is some $u \in \mathcal{O}_v$ with $-u \in \mathcal{O}_v$ and let $w \in \mathcal{O}_v$, then there exist $Q_+, Q_-, Q \in \mathcal{G}_\mathcal{R}$ with $u = Q_+v$, $-u = Q_-v$ and $w = Qv$. We thus have:

$$- w = -Qv = -QQ_+^* u = QQ_+^*(-u) = QQ_+^* Q_- v$$

but $QQ_+^* Q_- \in \mathcal{G}_\mathcal{R}$ and hence $-w \in \mathcal{O}_v$ and (2) follows. $\qquad\square$

All vectors in an orbit are 'equivalent' in the sense that they can be mapped onto one another by rotations that leave the orientation of $\mathcal{R}$ unchanged. Each orbit can therefore be uniquely described by a strong coordinate derived in Section 4.3.1 since it is either an even or an odd set. A single orbit is not necessarily sufficient for describing the orientation of $\mathcal{R}$. We might therefore need to map the orientation of a rigid body to a collection of distinct orbits $\mathcal{W}_\mathcal{R} = \cup_{i=1}^N \mathcal{O}_i$ instead. We call such a collection of orbits a *symmetric descriptor* of $\mathcal{R}$. An *irreducible symmetric descriptor* of $\mathcal{R}$ is defined as a symmetric descriptor that none of its subsets of constituent orbits is sufficient to describe the orientation of $\mathcal{R}$. Since each orbit in an irreducible symmetric descriptor is uniquely specified by $\mathcal{H}_i$, a strong coordinate derived in 4.3.1, the orientation of $\mathcal{R}$ can be uniquely specified by the N-tuple $(\mathcal{H}_1, \mathcal{H}_2, \cdots, \mathcal{H}_N)$.

There is no unique way of constructing an irreducible symmetric descriptor for a symmetric object as the size and the structure of a given orbit will depend on the vector that generate it. In general, one would prefer orbits with fewer vectors as their associated strong coordinates will be of smaller ranks, and hence easier to handle. The cardinality of an orbit is at most equal to the order of $\mathcal{G}_\mathcal{R}$, the rotation group of $\mathcal{R}$; however if the initial generating vector is chosen so that it is invariant under

certain group elements, a smaller orbit will be obtained. (Such vectors are equivalent to Wyckoff positions in a space group.) In general, the orientation of a rigid object in $\mathbb{R}^d$ can be uniquely specified by *at most* $d - 1$ linearly independent vectors. (This number will be smaller if the rotation group of $\mathcal{R}$ is a Lie group.) A symmetric descriptor should therefore have the following properties:

- It should have a sufficient number of linearly independent vectors.

- It is not invariant under any rotation that changes the orientation of $\mathcal{R}$.

Here we derive irreducible symmetric descriptors and the associated SRCs for all two- and three-dimensional rotation groups. The results are summarized in Table. 4.1.

4.3.2.1 Trivial Rotation Group

For a non-symmetric object, $\mathcal{G}_\mathcal{R} = \{I\}$ and $\mathcal{O}_v = \{v\}$ for every $v \in \mathcal{R}$. An irreducible symmetric descriptor is therefore given by $\cup_{i=1}^{d-1}\mathcal{O}_{v_i}$ with v_i's being linearly independent. A strong coordinate is therefore given by a $(d-1)$-tuple $(\mathcal{H}_1, \mathcal{H}_2, \cdots, \mathcal{H}_{d-1})$ $= (v_1, v_2, \cdots, v_{d-1})$. For a non-symmetric object in two and three dimensions, this will correspond, as expected, to one and two vectors.

4.3.2.2 C_n and D_n

C_n is the only non-trivial rotation group in two dimensions and corresponds to the symmetry of a regular n-gon. In three dimensions, C_n corresponds to the symmetry of a pyramid with a regular n-gonal basis. D_n however corresponds to the symmetry of a prism or a bipyramid with a regular n-gonal basis. A characteristic orbit of both C_n and D_n- denoted by $\mathcal{O}_n$- is generated by a arbitrary unit vector from within the plane perpendicular to the n-fold rotation axis, and contains n vectors. In two dimensions, $\mathcal{O}_n$ is an irreducible symmetric descriptor of a C_n object. In three dimensions however, it is only an irreducible symmetric descriptor of an object with $D_n (n \geq 3)$ rotation symmetry and it is not sufficient to describe the orientation of an object with the rotation group C_n; since a 180° rotation around one of those vectors will change the orientation of the object while leaving the orbit unchanged. An irreducible symmetric descriptor of a C_n object is therefore a union of the 'planar' orbit of n vectors and a single vector that is parallel to the axis of rotation. $\mathcal{O}_n$ is also not an irreducible symmetric descriptor of an object with D_2 symmetry as it only contains two collinear vectors. In that case, an irreducible symmetric descriptor can be constructed as the

union of $\{\pm z\}$ and $\mathcal{O}_n$ where z is the rotation axis. With the following chain of theorems, we show that $\mathcal{H}_n$ is the strong orientational coordinate for the planar orbit $\mathcal{O}_n$.

Lemma 9. *Let $v_1, v_2, \cdots, v_n \in \mathbb{R}^d$ be unit vectors so that $v_p^T v_q = \cos \dfrac{2\pi(p-q)}{n}$; then for $n \geq 3$, v_i's span a two-dimensional subspace of $\mathbb{R}^d$.*

Proof. Define unit vectors $x, y, z \in \mathbb{R}^d$ by $v_1 = x, v_2 = \cos \frac{2\pi}{n} x + \sin \frac{2\pi}{n} y$ and $v_k = \cos \frac{2\pi(k-1)}{n} x + \sin \frac{2\pi(k-1)}{n} z$ (for $k \geq 3$). Use $v_k^T v_2 = \cos \frac{2\pi(k-2)}{n}$ to deduce $z = y$. □

Lemma 10. *Let p, q be nonnegative integers and $\phi_k = \theta + \frac{2\pi k}{n}$. Then $I_{p,q}(\theta) = \sum_{k=0}^{n-1} \cos^p \phi_k \sin^q \phi_k$ is independent of θ for $p + q < n$, and is a function of θ if $p + q = n$.*

Proof. Note that:

$$\cos^p \phi_k = \left(\frac{1}{2}\right)^p \sum_{l=0}^{p} \binom{p}{l} \exp\left[i\phi_k(2l - p)\right] \tag{4.6}$$

$$\sin^q \phi_k = \left(\frac{i}{2}\right)^q \sum_{m=0}^{q} \binom{q}{m} (-1)^m \exp\left[i\phi_k(2m - q)\right] \tag{4.7}$$

$I_{p,q}(\theta)$ can therefore be written as:

$$I_{p,q}(\theta) = \frac{i^q}{2^{p+q}} \sum_{l=0}^{p} \sum_{m=0}^{q} \binom{p}{l} \binom{q}{m} (-1)^m e^{i\theta(2l+2m-p-q)} \sum_{k=0}^{n-1} \left[e^{2\pi i(2l+2m-p-q)/n}\right]^k$$

For $e^{2\pi(2l+2m-p-q)/n} \neq 1$ we have:

$$\sum_{k=0}^{n-1} \left[e^{2\pi i(2l+2m-p-q)/n}\right]^k = \frac{1 - e^{2\pi i(2l+2m-p-q)}}{1 - e^{2\pi i(2l+2m-p-q)/n}} = 0$$

Thus the sum over k survives only if $2l + 2m - p - q$ is a multiple of n. However $|2l + 2m - p - q| \leq p + q$. Thus if $p + q < n$, the only possibility is zero, which will take away the θ dependence of $I_{p,q}(\theta)$. For $p + q = n$, two θ-dependent terms will survive i.e. $e^{\pm i\theta n}$ and the proof follows. □

Theorem 11. *$\mathcal{H}_n$ is a strong orientational coordinate of $\mathcal{O}_n$.*

Proof. Let $V = \mathcal{O}_n = \{v_k\}_{k=1}^{n}$. Without loss of generality suppose $v_k = \cos\left(\theta + \frac{2\pi k}{n}\right) e_x + \sin\left(\theta + \frac{2\pi k}{n}\right) e_y$ with e_x and e_y being the unit vectors along the x and y directions.

The components of H_m are therefore either zero or $I_{p,m-p}$ which are always indepen-dent of θ for $m < n$ according to Lemma 10. For even n, the proof follows from Corollary 5. The proof for odd n is completed by noting that $\mathcal{H}_n(QV)$ will have some nonzero components that are zero for $\mathcal{H}_n(V)$ if QV and V are not in the same plane. $\qquad\square$

Note that for even n, $\mathcal{H}_n$, and for odd n, $\mathcal{H}_{2n-1}$ are guarranteed to be strong coordinates of $\mathcal{O}_n$. The bound given in Corollary 5 is therefore tight for even n.

4.3.2.3 C_∞ and D_∞

C_∞ is the symmetry of a cone with a circular base, while D_n corresponds to the symmetry of a cylinder. For both symmetries, there are two types of orbits. One is a finite set along the rotation axis which has one element for C_∞ and two elements for D_∞, and the other being an uncountably infinite set comprising one circle for C_∞ and one or two parallel circles for D_∞. The finite orbits $\mathcal{O}_f$ are irreducible symmetric descriptors. The corresponding SOC will therefore be $\mathcal{H}_1 = z$ for C_∞ and $\mathcal{H}_2 = zz$ for D_∞.

4.3.2.4 Tetrahedral Symmetry T

T corresponds to the rotation group of a regular tetrahedron and has twelve elements. A general orbit of T will therefore have twelve elements as well. The high symmetry vectors that connect the centroid of a regular tetrahedron to its vertices can however generate an orbit $\mathcal{O}_t = \{a_p\}_{p=1}^4$ that only has four elements. $\mathcal{O}_t$ is an irreducible symmetric descriptor of a regular tetrahedron, therefore its SOC will also be the SOC of a regular tetrahedron.

Since $\mathcal{O}_t$ is an odd set, $\mathcal{H}_1, \mathcal{H}_3, \mathcal{H}_5, \mathcal{H}_7$ are the candidate strong orientational coordinates given by Theorem 7, but if a_p's are unit vectors, we have:

$$\mathcal{H}_1^i = \sum_{p=1}^4 a_p^i = 0 \qquad (4.8)$$

$$\mathcal{H}_2^{ij} = \sum_{p=1}^4 a_p^i a_p^j = \frac{4}{3}\delta^{ij} \qquad (4.9)$$

We prove that $\mathcal{H}_3$ is the SOC for $\mathcal{O}_T$ as follows. Let $\mathcal{O}_T = \{a_p\}_{p=1}^4$ and $\mathcal{O}_{T'} = \{b_q\}_{q=1}^4$

be the corresponding orbits for two tetrahedra T and T' and observe that:

$$\|\mathcal{H}_3(\mathcal{O}_T) - \mathcal{H}_3(\mathcal{O}_{T'})\|_F^2 = 2\left[\frac{32}{9} - \sum_{p,q=1}^4 \xi_{pq}^3\right] \qquad (4.10)$$

with $\xi_{pq} = a_p^T b_q$. Contracting (4.8) and (4.9) with b_q^i and $b_q^i b_q^j$ yields:

$$\begin{array}{ll} \sum_{p=1}^4 \xi_{pq} = 0 & q = 1, \cdots, 4 \\ \sum_{p=1}^4 \xi_{pq}^2 = \frac{4}{3} & q = 1, \cdots, 4 \end{array} \qquad (4.11)$$

As shown in Appendix B.1, (4.10) can only be zero if ξ_{pq}'s are the permutations of $(1, -\frac{1}{3}, -\frac{1}{3}, -\frac{1}{3})$ for every q i.e. if $\mathcal{O}_T = \mathcal{O}_{T'}$.

4.3.2.5 Octahedral Symmetry O

O corresponds to rotational symmetry of a cube and an octahedron and has twenty-four elements. Orbits containing as few as six elements can however be obtained by choosing high-symmetry vectors, such as vectors connecting the center of a regular octahedron to its vertices- or equivalently the center of a cube to the centers of its faces. Such an orbit is an irreducible symmetric descriptor of a cube or an octahedron and has the form $\mathcal{O}_C = \{\pm x, \pm y, \pm z\}$ with x, y and z being mutually orthogonal.

Being an even set, Corollary 5 implies that $\mathcal{H}_2, \mathcal{H}_4$ and $\mathcal{H}_6$ are the candidate SOCs for $\mathcal{O}_C$. We however have:

$$\mathcal{H}_2^{ij} = 2(x^i x^j + y^i y^j + z^i z^j) = 2\delta^{ij} \qquad (4.12)$$

We prove that $\mathcal{H}_4$ is an SOC of $\mathcal{O}_C$ similar to what was done for regular tetrahedron. Let $\mathcal{O}_C = \{\pm a_p\}_{p=1}^3$ and $\mathcal{O}_{C'} = \{\pm b_q\}_{q=1}^3$ with $a_p^i a_q^i = b_p^i b_q^i = \delta_{pq}$ be the corresponding orbits for two cubes C and C' and note that:

$$\|\mathcal{H}_4(\mathcal{O}_C) - \mathcal{H}_4(\mathcal{O}_{C'})\|_F^2 = 8\left[3 - \sum_{p,q=1}^3 \xi_{pq}^4\right] \qquad (4.13)$$

with $\xi_{pq} = a_p^T b_q$. For every q we have $\sum_{p=1}^3 \xi_{pq}^2 = 1$. As shown in Appendix B.2, (4.13) is zero only if ξ_{pq}^2's are a permutation of $(1, 0, 0)$ for each q i.e. only if $\mathcal{O}_C = \mathcal{O}_{C'}$.

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4.3.2.6 Icosahedral Symmetry I

I corresponds to the rotational symmetry of a regular icosahedron and a regular dodecahedron, and has 60 elements. Orbits with as few elements as 12 can however be obtained by using vectors connecting the center of an icosahedron to its twelve vertices. The corresponding orbit denoted by $\mathcal{O}_I = \{\pm a_i\}_{i=1}^6$ is an irreducible symmetric descriptor of an icosahedron (dodecahedron). A prototypical set of such unit a_i's are given by $(\alpha, \pm\beta, 0), (0, \alpha, \pm\beta), (\pm\beta, 0, \alpha)$ with $\alpha = 1/\sqrt{1 + \phi^2}$, $\beta = \phi/\sqrt{1 + \phi^2}$ and $\phi = (\sqrt{5} + 1)/2$. The choice of a_i's are arbitrary in the sense that both $+a_i$ and $-a_i$ are valid choices. Also note that $a_i^T a_j = \pm 1/\sqrt{5}$ for $i \neq j$.

As an even set, the SOC of $\mathcal{O}_I$ will be amongst $\mathcal{H}_2, \cdots, \mathcal{H}_{12}$ according to Corollary 5. However one can show that:

$$\mathcal{H}_2^{ij} = 2\sum_{p=1}^6 a_p^i a_p^j = 4\delta^{ij} \tag{4.14}$$

$$\mathcal{H}_4^{ijkl} = 2\sum_{p=1}^6 a_p^i a_p^j a_p^k a_p^l = \frac{4}{5}\left(\delta^{ij}\delta^{kl} + \delta^{ik}\delta{jl} + \delta^{il}\delta^{jk}\right) \tag{4.15}$$

In an approach similar to what was done for the tetrahedral and octahedral symmetry, we prove that $\mathcal{H}_6$ is the SOC for $\mathcal{O}_I$. Let $\mathcal{O}_I = \{\pm a_p\}_{p=1}^6$ and $\mathcal{O}_{I'} = \{\pm b_q\}_{q=1}^6$ be two such orbits. We have:

$$\|\mathcal{H}_6(\mathcal{O}_I) - \mathcal{H}_6(\mathcal{O}_{I'})\|_F^2 = 8\sum_{p,q=1}^6 \left[\frac{156}{125} - \xi_{pq}^3\right] \tag{4.16}$$

with $\xi_{pq} = [a_p^T b_q]^2$. Contracting $\mathcal{H}_2(\mathcal{O}_I)$ and $\mathcal{H}_4(\mathcal{O}_I)$ with $b_q^i b_q^j$ and $b_q^i b_q^j b_q^k b_q^l$ yields:

$$\sum_{p=1}^6 \xi_{pq} = 2 \tag{4.17}$$

$$\sum_{p=1}^6 \xi_{pq}^2 = \frac{6}{5} \tag{4.18}$$

(4.16) can only be zero if ξ_{pq}'s are a permutation of $(1, \frac{1}{5}, \frac{1}{5}, \frac{1}{5}, \frac{1}{5}, \frac{1}{5})$ for each q i.e. if $\mathcal{O}_I = \mathcal{O}_{I'}$. (See Appendix B.3 for more details.)

Table 4.1: Strong orientational coordinates of two- and three-dimensional rotation groups. The 'optimal $\mathcal{H}$' corresponds to the smallest-rank strong rotational coordinate. 'Upper-bound SOC' however corresponds to the upper-bound rank predicted in Section 4.3.1. The vectors given in the column $\mathcal{W}$ correspond to a typical orbit. Also note that $\alpha = 1/\sqrt{\sqrt{1+\phi^2}}, \beta = \phi/\sqrt{\sqrt{1+\phi^2}}$ with $\phi = (\sqrt{5}+1)/2$.

d	g	Prototypes	$\mathcal{W}$	Optimal SOC	Upper bound SOC
2	C_n	regular n-gon	$\{v_p\}_{p=1}^n$ $v_p\left(\cos\frac{2\pi k}{n}, \sin\frac{2\pi k}{n}\right)$	$\mathcal{H}_n = \sum_{p=1}^n v_p^n$	$\mathcal{H}_n$, n even $\mathcal{H}_{2n-1}$, n odd
3	C_n	regular n-gonal pyramid	$\{v_p\}_{p=1}^n \cup \{z\}$ $v_p\left(\cos\frac{2\pi k}{n}, \sin\frac{2\pi k}{n}, 0\right)$ $z(0,0,1)$	$(\mathcal{H}_n, z)$	$(\mathcal{H}_n, z)$, n even $(\mathcal{H}_{2n-1}, z)$, n odd
3	$D_n, n > 2$	regular n-gonal pyramid regular n-gonal prism	$\{v_p\}_{p=1}^n$ $v_p\left(\cos\frac{2\pi k}{n}, \sin\frac{2\pi k}{n}, 0\right)$	$\mathcal{H}_n = \sum_{p=1}^n v_p^n$	$\mathcal{H}_n$, n even $\mathcal{H}_{2n-1}$, n odd
3	D_2	rectangular parallelepiped	$\{\pm z\} \cup \{\pm y\}$ $z(0,0,1)$ $y(0,1,0)$	(zz, yy)	(zz, yy)
3	C_∞	Cone, Hemisphere	$\{z\}, z(0,0,1)$	z	z
3	D_∞	Cyliner	$\{\pm z\}, z(0,0,1)$	zz	zz
3	T	Regular tetrahedron	$\{a_p\}_{p=1}^4$ $a_1\left(\frac{1}{\sqrt3},\frac{1}{\sqrt3},\frac{1}{\sqrt3}\right)$ $a_2\left(\frac{1}{\sqrt3},-\frac{1}{\sqrt3},-\frac{1}{\sqrt3}\right)$ $a_3\left(-\frac{1}{\sqrt3},-\frac{1}{\sqrt3},\frac{1}{\sqrt3}\right)$ $a_4\left(-\frac{1}{\sqrt3},\frac{1}{\sqrt3},-\frac{1}{\sqrt3}\right)$	$\mathcal{H}_3 = \sum_{p=1}^4 a_p a_p a_p$	$\mathcal{H}_7$
3	O	Octahedron Cube	$\{\pm a_p\}_{p=1}^3$ $a_1(1,0,0)$ $a_2(0,1,0)$ $a_3(0,0,1)$	$\mathcal{H}_4 = 2\sum_{p=1}^3 a_p a_p a_p a_p$	$\mathcal{H}_6$
3	I	Regular Icosahedron Regular Dodecahedron	$\{\pm a_p\}_{p=1}^6$ $a_{1,2}(\alpha,\pm\beta,0)$ $a_{3,4}(0,\alpha,\pm\beta)$ $a_{1,2}(\pm\beta,0,\alpha)$	$\mathcal{H}_4 = 2\sum_{p=1}^6 a_p a_p a_p a_p a_p$	$\mathcal{H}_{12}$

4.3.3 Ordered Arrangements and Order Parameters

To quantify any orientationally-ordered arrangement of symmetric particles, an interrelation should be established between the particle-level orientational information and the geometric description of order. To do this, we will characterize bulk order with a probability distribution specifying the probability that a single particle will take a certain orientation. The next step is to compare computationally-measured values of certain moments of particle-level orientational coordinates with their expected values calculated from the presumed rotational distribution function.

An arrangement of (symmetric) objects is called **isotropic** if each particle can take all permissible orientations with equal probability. This can be characterized by a uniform probability density function. However in an orientationally-ordered arrangement of (symmetric) objects, orientational symmetry is broken and each particle tends to preferentially take certain orientations more frequently. Such bias can be characterized by a non-uniform probability density function which can be viewed as the marginal probability density function for a single particle in such an arrangement. Such a connection can allow us define a structure- or a phase- as follows.

Definition 2. A **structure** or **phase** of an object $\mathcal{R}$ is characterized by a probability density function $p_0(\mathcal{H}_{m_1}, \cdots, \mathcal{H}_{m_N}; \Omega)$ where Ω stands for all geometric features needed for macroscopic characterization of the structure.

Example 1. Let $\mathcal{R}$ be a symmetric object with a normalized irreducible symmetric descriptor containing a single equivalence class ($N = 1$) and let $\mathcal{H}_m$ be a strong orientational coordinate constructed from $\mathcal{N}_\mathcal{R}$. The density function $p_0(\mathcal{H}_m; \hat{\mathcal{H}}_m) = \delta(\mathcal{H}_m - \hat{\mathcal{H}}_m)$ defines an arrangement of the object $\mathcal{R}$ where all objects have the same orientation with $\hat{\mathcal{R}}$ and $\Omega = \hat{\mathcal{H}}_m$.

More precisely, let $\mathcal{H}_\mathcal{R} :\equiv (\mathcal{H}_{m_1}, \cdots, \mathcal{H}_{m_N})$ be a strong orientational coordinate for $\mathcal{R}$ and $p_0(\mathcal{H}_\mathcal{R}; \Omega)$ be a phase. Also let $M(\mathcal{H}_\mathcal{R})$ be a tensor function of $\mathcal{H}_\mathcal{R}$ with the property that $\mathfrak{M}(\Omega) = \langle \mathcal{M}(\mathcal{H}_\mathcal{R}) \rangle_0$:

1. is a strong descriptor of Ω i.e. $\mathfrak{M}_1 = \mathfrak{M}_2$ if and only if $\Omega_1 = \Omega_2$.

2. is invariant under transformations which preserve Ω.

Property 1 is to assure that all distinct geometric instances of a phase are distinguishable by $\mathfrak{M}$ and is necessary for a *full* description of the geometry of order; however if one is not interested in all structural details, this condition can be relaxed so that certain *classes* of structures are resolved by a single $\mathfrak{M}$. The second condition is to

ensure that $\mathfrak{M}$ is unique for a certain instance of the phase and does not depend on structurally-irrelevant parameters.

Now let $\mathcal{R}_1, \mathcal{R}_2, \cdots, \mathcal{R}_n$ be an arrangement of (symmetric) particles and define the **experimental order estimator** as:

$$\overline{M} = \frac{1}{n} \sum_{i=1}^{n} M(\mathcal{H}_{\mathcal{R}_i}) \tag{4.19}$$

The more perfect the ordering of $\mathcal{R}_1, \mathcal{R}_2, \cdots, \mathcal{R}_n$ is, the closer will $\overline{M}$ be to $\mathfrak{M}$. (Indeed perfect ordering for $n \to +\infty$ implies that $\overline{M} \to \mathfrak{M}$ or equivalently $\left\|\overline{M} - \mathfrak{M}\right\|_F \to 0$). Letting $\mathfrak{M}_I = \langle M \rangle_{\text{isotropic}}$ it immediately follows that $\overline{M} \to \mathfrak{M}$ implies that $\left\|\overline{M} - \mathfrak{M}\right\|_F = \left\|(\overline{M} - \mathfrak{M}_I) - (\mathfrak{M} - \mathfrak{M}_I)\right\|_F \to 0$. For an isotropic structure however $\left\|\overline{M} - \mathfrak{M}\right\|_F = \left\|\mathfrak{M}_I - \mathfrak{M}\right\|_F$. Hence one can formulate the problem of finding the underlying geometric parameters of the phase as:

$$\min_{\Omega} \frac{\left\|\overline{M} - \mathfrak{M}_\Omega\right\|_F}{\left\|\mathfrak{M}_I - \mathfrak{M}_\Omega\right\|_F} \tag{4.20}$$

One can define **scalar order parameter** of the phase as:

$$\kappa_\Omega = 1 - \frac{\left\|\overline{M} - \mathfrak{M}_{\Omega^*}\right\|_F^2}{\left\|\mathfrak{M}_I - \mathfrak{M}_{\Omega^*}\right\|_F^2} \tag{4.21}$$

where Ω^* is the minimizer in (4.20). Note that $\kappa = 0$ for completely isotropic system while $\kappa = 1$ when matching is perfect. For a certain subclass of phases where $\|\mathfrak{M}_\Omega\|$ is constant, one can simplify (4.20) to:

$$\max_{\Omega} \overline{M} \odot \mathfrak{M}_\Omega \tag{4.22}$$

In general if the conditions outlined above are established for a given $\mathfrak{M}$, one can use (4.20) or (4.22) to obtain $\mathfrak{M}_{\Omega^*}$ and (4.21) to calculate the scalar order parameter. We will show the procedure of solving (4.20) or (4.22) in a few examples at the end of this section, but before doing so, we give explicit formulae that can be used to calculate $\mathfrak{M}_I$ for M's of the form introduced in this chapter.

Definition 3. Let $v_1, v_2, \cdots, v_m \in \mathbb{R}^d$ be arbitrary vectors and $a_1, a_2, \cdots, a_m$ nonnegative integers adding up to a, a *symmetrized tensor polynomial* $\mathfrak{S}^a_{a_1, a_2, \cdots, a_m}(v_1, v_2, \cdots, v_m)$ is defined as the sum of all possible direct products of the form $v_{i_1} v_{i_2} \cdots v_{i_a}$ where exactly a_1 of i_j's are 1, a_2 of i_j's two, etc. The number of distinct terms in such a

polynomial is $\left(\begin{smallmatrix} & a & \\ a_1,a_2,\cdots,a_m \end{smallmatrix}\right)$.

Proposition IV.1. *The symmetrized tensor polynomial defined in Definition (3) is given by:*

$$\mathfrak{S}^a_{a_1,a_2,\cdots,a_m}(v_1,v_2,\cdots,v_m) = \begin{cases} \mathfrak{S}^a_{a_1,a_2,\cdots,a_{k-1},0,a_{k+1},\cdots,a_m}(v_1,\cdots,v_{k-1},v_{k+1},\cdots,v_m) & a_k = 0 \\ \sum_{k=1}^m v_k \mathfrak{S}^{a-1}_{a_1,\cdots,a_{k-1},a_k-1,a_{k+1},\cdots,a_m}(v_1,\cdots,v_m) & a_i > 0 \text{ for all } i \end{cases}$$

with $\mathfrak{S}^0_{0,0,\cdots,0}(v_1,v_2,\cdots,v_m) = 1$.

Proof. First assertion follows from definition. For the second assertion look at each term of $\mathfrak{S}^a_{a_1,a_2,\cdots,a_m}(v_1,v_2,\cdots,v_m)$ and group them based on their initial multiplier. $\square$

Proposition IV.2. *Let* $\mathbf{t} \in \mathbb{R}^d, d = 2, 3$ *be a unit vector and* $k \in \mathbb{Z}^{\geq 0}$, *then :*

1. $\langle \mathbf{t}^{2k+1} \rangle_{isotropic} = 0$.

2. $\langle \mathbf{t}^{2k} \rangle_{isotropic} = \sum_{p=0}^k \dfrac{(2p)!(2k-2p)!}{4^k p! k! (k-p)!} \mathfrak{S}^{2k}_{2p,2k-2p}(\mathbf{x},\mathbf{y})$ *for* $d = 2$.

3. $\langle \mathbf{t}^{2k} \rangle_{isotropic} = \dfrac{1}{2k+1} \sum_{m=0}^k \sum_{n=0}^{k-m} \dfrac{\left(\begin{smallmatrix} & k & \\ m,n,k-m-n \end{smallmatrix}\right)}{\left(\begin{smallmatrix} & 2k & \\ 2m,2n,2k-2m-2n \end{smallmatrix}\right)} \mathfrak{S}^{2k}_{2m,2n,2k-2m-2n}(\mathbf{x},\mathbf{y},\mathbf{z})$.

where $(\mathbf{x},\mathbf{y})$ *and* $(\mathbf{x},\mathbf{y},\mathbf{z})$ *are orthogonal bases for* $\mathbb{R}^2$ *and* $\mathbb{R}^3$ *respectively.*

Proof. Observe that $\langle \mathbf{t}^n \rangle = (1/S_{d-1}) \int_{S^d} \mathbf{t}^n d\Omega$ where S_{d-1} is the surface of the d-sphere. Contributions from two hemispheres to the integral cancel out for odd n and (1) follows. For $n = 2k$ and $d = 2$ we have $\mathbf{t} = \cos\theta\, \mathbf{x} + \sin\theta\, \mathbf{y}$ and:

$$\langle \mathbf{t}^{2k} \rangle_I = \frac{1}{2\pi} \sum_{p=0}^k I_{2p,2k-2p} \mathfrak{S}^{2k}_{2p,2k-2p}(\mathbf{x},\mathbf{y})$$

where $I_{2p,2k-2p} = \int_0^{2\pi} \cos^{2p}\theta \sin^{2k-2p}\theta\, d\theta$. The odd terms vanish because $I_{2p,2q+1} = I_{2p+1,2q} = I_{2p+1,2q+1} = 0$ and (2) follows from Eq. (B.25). For $d = 3$, $\mathbf{t}(\theta,\phi) = \sin\theta\cos\phi\mathbf{x} + \sin\theta\sin\phi\mathbf{y} + \cos\theta\mathbf{z}$ and:

$$\langle \mathbf{t}^{2k} \rangle_I = \frac{1}{4\pi} \sum_{p=0}^k \sum_{q=0}^{k-p} I_{2p,2q} J_{2k-2p-2q,2p+2q+1} \mathfrak{S}^{2k}_{2p,2q,2k-2p-2q}(\mathbf{x},\mathbf{y},\mathbf{z})$$

$$\overset{(a)}{=} \sum_{p=0}^k \sum_{q=0}^{k-p} \frac{(2p)!(2q)!}{4^{p+q}p!q!(p+q)!} \frac{4^{p+q}(p+q)!(2k-2p-2q)!k!}{(k-p-q)!(2k+1)!} \mathfrak{S}^{2k}_{2p,2q,2k-2p-2q}(\mathbf{x},\mathbf{y},\mathbf{z})$$

$$(4.23)$$

(*a*) follows from (B.25) and B.28). Rearranging (4.23) completes the proof. $\qquad\square$

Using this proposition, one can thus calculate $\mathfrak{M}_I$ for any given tensor that is a sum of n-adic products of unit vectors, including moments of homogeneous tensors defined in this work. One can therefore always subtract $\mathfrak{M}_I$ in the definition of M so that $\|M\|_F$ on its own can be used as a measure of how anisotropic a certain arrangement of particles is. The rest of this section is devoted to some examples of how (4.20) and (4.22) can be formulated and solved. But before doing so, we outline the following useful result that can be used to calculate the expected value of a k-adic power of a vector that can uniformly rotate around a rotation axis.

Proposition IV.3. *Let $v, z, t \in \mathbb{R}^3$ be unit vectors and let $v = \alpha z + \beta t$ and t is uniformly distributed on the plane perpendicular to z. Then:*

$$
\langle v^k \rangle_z = \sum_{l=0}^{\lfloor k/2 \rfloor} \sum_{m=0}^{l} \alpha^{k-2l} \beta^{2l} \frac{(2m)!(2l-2m)!}{4^l l! m! (l-m)!}
$$
$$
\times \mathfrak{S}_{2m,2l-2m,k-2l}^{k}(x, y, z)
$$

with x and y being a pair of orthonormal vectors perpendicular to z.

Proof. Expand v^k in terms of t and z and use case 2 of Proposition IV.2 to complete the proof. $\qquad\square$

4.3.3.1 Unixial Nematics

Rodlike molecules or nanoparticles can assemble into a rotationally ordered phase known as the *uniaxial nematic* phase where the rotation axes of all particles are on average aligned to a common vector called a *director* [9, 116, 117]. As explained in Section 4.3.2.3, $\mathcal{H}_2(\{\pm z_i\}) = z_i^2$ is the proper SOC for rods and $\Omega = \{u\}$ i.e. the director, In a perfect nematic all particles will align along the same director. This perfectly fits into the type of structures described in Example 1. We therefore have:

$$
M^{ij} = H_2^{ij} - H_{2,I}^{ij} = z^i z^j - \frac{1}{3}\delta^{ij}
$$
$$
\mathfrak{M}_\Omega^{ij} = u^i u^j - \frac{1}{3}\delta^{ij}
$$
$$
\overline{M}^{ij} = \frac{1}{N}\sum_{p=1}^{N} z_p^i z_p^j - \frac{1}{3}\delta^{ij}
$$

60

Since $\|\mathfrak{M}_\Omega\|_F$ is constant, we can use the optimization problem (4.22) which takes the form:

$$\begin{aligned}
\max \quad & u^T \overline{M} u \\
\text{subject to} \quad & u^T u = 1
\end{aligned} \tag{4.24}$$

which can be solved by using Lagrange multipliers:

$$\begin{aligned}
\mathcal{L}(u, \lambda) &= u^T \overline{M} u - \lambda(u^T u - 1) \\
\nabla_u \mathcal{L} &= 2\overline{M} u - 2\lambda u = 0 \implies (\overline{M} - \lambda I) u = 0
\end{aligned}$$

i.e. u should be an eigenvector of $\overline{M}$. The largest eigenvalue of $\overline{M}$, λ_1 maximizes $u^T \overline{M} u$. The scalar order parameter is given by:

$$\kappa_{\text{nematic}} = 1 - \frac{3}{2} \left[\left(\lambda_1 - \frac{2}{3} \right)^2 + \left(\lambda_2 + \frac{1}{3} \right)^2 + \left(\lambda_3 + \frac{1}{3} \right)^2 \right] \tag{4.25}$$

with $\lambda_1 \geq \lambda_2 \geq \lambda_3$ being the eigenvalues of $\overline{M}$. This formula penalizes any divergence of λ_i's from their 'optimal' values of $(2/3, -1/3, -1/3)$ in a perfect nematic.

4.3.3.2 Cubatic Phase

At sufficiently large densities, hard cubes can assemble into the *cubatic phase*; a structure in which all particles have almost the same orientations while no long-range translational order exists [20]. The cubatic phase can therefore be described using the general framework of Example 1. As $\mathcal{H}_4$ is the proper SOC coordinate for cubes, we therefore have:

$$\begin{aligned}
M^{ijkl} &= 2\sum_{p=1}^{3} u_p^i u_p^j u_p^k u_p^l - \frac{2}{5} \left(\delta^{ij}\delta^{kl} + \delta^{ik}\delta^{jl} + \delta^{il}\delta^{jk} \right) \\
\mathfrak{M}_\Omega &= 2\sum_{p=1}^{3} v_p^i v_p^j v_p^k v_p^l - \frac{2}{5} \left(\delta^{ij}\delta^{kl} + \delta^{ik}\delta^{jl} + \delta^{il}\delta^{jk} \right) \\
\overline{M}^{ijkl} &= \frac{2}{N}\sum_{p=1}^{3}\sum_{q=1}^{N} u_{p,q}^i u_{p,q}^j u_{p,q}^k u_{p,q}^l \\
&\quad - \frac{2}{5} \left(\delta^{ij}\delta^{kl} + \delta^{ik}\delta^{jl} + \delta^{il}\delta^{jk} \right)
\end{aligned} \tag{4.26}$$

where v_p's are the orthogonal vectors specifying Ω i.e. the average cubic orientation that all particles align to, and $u_{p,q}$'s are the corresponding orthogonal vectors describing particle q. Since $\mathfrak{M}_\Omega$ is constant, the following optimization problem should be solved for quantifying cubatic order:

$$
\begin{array}{ll}
\text{max} & \overline{M}^{ijkl} \sum_{p=1}^{3} v_p^i v_p^j v_p^k v_p^l \\
\text{subject to} & v_p^i v_q^j = \delta_{pq} \qquad p, q = 1, 2, 3
\end{array}
\tag{4.27}
$$

The constraints of (4.27) can however be easily shown to be equivalent to $\sum_{p=1}^{3} v_p v_p^T = I$. (Multiply both sides by v_q and using the linear independence of v_p's to conclude that $v_p^T v_q = \delta_{pq}$.) This Lagrangian of the optimization problem is therefore given by:

$$
\mathcal{L} = \overline{M}^{ijkl} \sum_{p=1}^{3} v_p^i v_p^j v_p^k v_p^l - \sum_{i,j=1}^{3} \mu_{ij} \left[\sum_{p=1}^{3} v_p^i v_p^j - \delta^{ij} \right]
$$

with $\mu_{ij} = \mu_{ji}$. The derivative of the Lagrangian is given by:

$$
\begin{aligned}
\frac{\partial \mathcal{L}}{\partial v_q^s} &= \overline{M}^{sjkl} v_q^j v_q^k v_q^l + \overline{M}^{iskl} v_q^i v_q^k v_q^l + \overline{M}^{ijsl} v_q^i v_q^j v_q^l \\
&\quad + \overline{M}^{ijks} v_q^i v_q^j v_q^k - \mu_{sj} v_q^j - \mu_{is} v_q^i \\
&\overset{(a)}{=} 4\overline{M}^{sijk} v_q^i v_q^j v_q^k - 2\mu_{si} v_q^i \\
\nabla_q \mathcal{L} &= b_q - C v_q
\end{aligned}
\tag{4.28}
$$

with:

$$
b_q = 4 \begin{pmatrix} \overline{M}^{1ijk} v_q^i v_q^j v_q^k \\ \overline{M}^{2ijk} v_q^i v_q^j v_q^k \\ \overline{M}^{3ijk} v_q^i v_q^j v_q^k \end{pmatrix}, \quad C = 2 \begin{pmatrix} \mu_{11} & \mu_{12} & \mu_{13} \\ \mu_{21} & \mu_{22} & \mu_{23} \\ \mu_{31} & \mu_{32} & \mu_{33} \end{pmatrix}
\tag{4.29}
$$

Note that (a) follows from that $\overline{M}$ is invariant under index permutation. $\nabla_q \mathcal{L} = 0$ implies that $b_q = C v_q$. Multiplying both sides by v_q and summing over q yields:

$$
\sum_{q=1}^{3} b_q v_q^T = C \sum_{q=1}^{3} v_q v_q^T = C
\tag{4.30}
$$

Note that C is not symmetric for an arbitrary set of orthogonal v_q's and its symmetry is achieved if v_q's are amongst the Karush-Kuhn-Tucker solutions of (4.27). The global maximum of (4.27) can only be obtained if *all* solutions of $C = C^T$ are identified.

We achieve this by adopting a Newton-Raphson iterative scheme for solving $C = C^T$; and the global maximum will be achieved if a sufficient number of iterations using different initial guesses are obtained. In particular, we propose a quaternion-based, easy-to-implement Newton-Raphson scheme for this purpose. To do this, we define $\zeta_{st} := \mu_{st} - \mu_{ts}$ for $s \neq t$ and observe that:

$$\frac{\partial \zeta_{st}}{\partial v_p^m} = 12 \left(\overline{M}^{smij} v_p^i v_p^j v_p^t - \overline{M}^{tmij} v_p^i v_p^j v_p^s \right) \tag{4.31}$$

We can therefore approximate ζ_{st} as

$$\zeta_{st} \approx \zeta_{st,0} + \sum_{q=1}^{3} (\nabla_q \zeta_{st})_0^T (v_p - v_{p,0}) + \cdots \tag{4.32}$$

The Newton-Raphson iteration can therefore be carried out by simultaneously solving the three equations given by $\zeta_{st} = 0$ and the six equations ensuring the orthonormality of v_q's i.e. $\sum_q v_q^i v_q^j = \delta^{ij}$. We can however decrease the number of equations from nine to four by using quaternions, which are widely used in simulations of nonspherical particles. Described by the Irish mathematician William Rowan Hamilton in 1843 for the first time, quaternions form a non-commutative algebra extending complex numbers. Unit quaternions are used to describe rigid-body rotations in three dimensions. Rotating a rigid body $\mathcal{R}$ using a unit quaternion $\mathbf{q} = (q_1, q_2, q_3, q_4)$ maps every vector $v^\circ \in \mathbb{R}$ to $v = R(\mathbf{q})v^\circ$. The rotation matrix $R(\mathbf{q})$ is given by:

$$\begin{bmatrix} q_1^2 + q_2^2 - q_3^2 - q_4^2 & 2(q_2q_3 - q_1q_4) & 2(q_1q_3 + q_2q_4) \\ 2(q_1q_4 + q_2q_3) & q_1^2 - q_2^2 + q_3^2 - q_4^2 & 2(q_3q_4 - q_1q_2) \\ 2(q_2q_4 - q_1q_3) & 2(q_1q_2 + q_3q_4) & q_1^2 - q_2^2 - q_3^2 + q_4^2 \end{bmatrix}$$

One can thus consider v_p's- and ζ_{st}'s- as implicit functions $\mathbf{q}$. More specifically we have:

$$\frac{\partial \zeta_{st}}{\partial q_u} = \sum_{p,m=1}^{3} \frac{\partial \zeta_{st}}{\partial v_p^m} \frac{\partial v_p^m}{\partial q_u} = \sum_{p=1}^{3} (\nabla_p \zeta_{st})^T \frac{\partial v_p}{\partial q_u}$$

$$= \sum_{p=1}^{3} (\nabla_p \zeta_{st})^T \frac{\partial R(\mathbf{q})}{\partial q_u} v_p^\circ$$

where v_p° correspond to a set of orthonormal vectors corresponding to $\mathbf{q} = 1$. (In order to solve $C = C^T$ under the orthonormality constraint, on can solve the following four

equations denoted by $\mathbf{f(q)} = \mathbf{0}$):

$$\mathbf{f(q)} \quad = \quad \begin{pmatrix} \zeta_{12} \\ \zeta_{13} \\ \zeta_{23} \\ q_1^2 + q_2^2 + q_3^2 + q_4^2 - 1 \end{pmatrix} \tag{4.33}$$

The Newton-Raphson iteration can therefore be carried out using the following formula:

$$\mathbf{q}_{n+1} = \mathbf{q}_n - \Phi(\mathbf{q}_n)^{-1}\mathbf{f}(\mathbf{q}_n) \tag{4.34}$$

with:

$$\Phi(\mathbf{q}) \quad = \quad \begin{pmatrix} \partial\zeta_{12}/\partial q_1 & \partial\zeta_{12}/\partial q_2 & \partial\zeta_{12}/\partial q_3 & \partial\zeta_{12}/\partial q_4 \\ \partial\zeta_{13}/\partial q_1 & \partial\zeta_{13}/\partial q_2 & \partial\zeta_{13}/\partial q_3 & \partial\zeta_{13}/\partial q_4 \\ \partial\zeta_{23}/\partial q_1 & \partial\zeta_{23}/\partial q_2 & \partial\zeta_{23}/\partial q_3 & \partial\zeta_{23}/\partial q_4 \\ 2q_1 & 2q_2 & 2q_3 & 2q_4 \end{pmatrix}$$

Iteration (4.34) can be carried out for a number of randomly-selected unit quaternions as initial guesses. Once the global maximum is attained, Eq (4.21) can be used to calculate the scalar cubatic order parameter. Neither the accuracy nor the convergence rate depends on the selection of v_p° as they are benign parameters of $\mathbf{f(q)}$.

4.3.3.3 Tetrahedral Nematics

Regular tetrahedra can form a structure in which, on average, all particles have the same orientation. Although its existence has been predicted in some theoretical works of liquid crystals [194], it is yet to be observed in experiments or simulations. This phase is sometimes referred to as the *tetrahedral nematics*, and like axial nematics of rods and the cubatic phase, fall into the general category of structures outlined in

Example 1. As $\mathcal{H}_3$ is the proper SOC for tetrahedral symmetry, we have:

$$
\begin{aligned}
M^{ijk} &= \sum_{p=1}^{4} v_p^i v_p^j v_p^k \\
\mathfrak{M}_\Omega^{ijk} &= \sum_{p=1}^{4} u_p^i u_p^j u_p^k \\
\overline{M}^{ijk} &= \frac{1}{N} \sum_{l=1}^{N} \sum_{p=1}^{3} v_{p,l}^i v_{p,l}^j v_{p,l}^k \quad\quad (4.35)
\end{aligned}
$$

where u_p's are the four vectors specifying the high-symmetry orbit of the reference tetrahedron corresponding to the average orientation of the particles; while $v_{p,q}$'s are the elements of the corresponding orbit for particle q. The following optimization problem is therefore solved for quantifying the tetrahedral order:

$$
\begin{aligned}
&\text{max} && \overline{M}^{ijk} \sum_{p=1}^{4} u_p^i u_p^j u_p^k \\
&\text{subject to} && v_p^i v_q^i = \tfrac{4}{3}\delta_{pq} - \tfrac{1}{3} \quad p, q = 1, 2, 3, 4
\end{aligned} \quad\quad (4.36)
$$

As for the cubatic phase, we can replace the constraints of (4.36) with $\sum_{p=1}^{4} u_p^i u_p^j = (4/3)\delta^{ij}$ and $\sum_{p=1}^{4} u_p^i = 0$. (See Appendix B.5 for details.) The Lagrangian and its derivatives are thus given by:

$$
\begin{aligned}
\mathcal{L} &= \overline{M}^{ijk} \sum_{p=1}^{4} u_p^i u_p^j u_p^k - \sum_{p=1}^{4} \left[\lambda_i u_p^i - \mu_{ij} u_p^i u_p^j \right] \\
\nabla_q \mathcal{L} &= b_q - c - C u_q
\end{aligned}
$$

with:

$$
b_q = \begin{pmatrix} 3\overline{M}^{1jk} u_q^i u_q^j \\ 3\overline{M}^{2jk} u_q^i u_q^j \\ 3\overline{M}^{3jk} u_q^i u_q^j \end{pmatrix}, c = \begin{pmatrix} \lambda_1 \\ \lambda_2 \\ \lambda_3 \end{pmatrix}, C = \begin{pmatrix} \mu_{11} & \mu_{12} & \mu_{13} \\ \mu_{21} & \mu_{22} & \mu_{23} \\ \mu_{31} & \mu_{32} & \mu_{33} \end{pmatrix}
$$

with $C = C^T$. Multiplying $\nabla_q \mathcal{L}$ by u_q^T on the right and summing over q yields:

$$
C = \frac{3}{4} \sum_{q=1}^{4} b_q u_q^T \quad\quad (4.37)
$$

The symmetry of C can be enforced using a method similar to that explained for the cubatic phase and the scalar order parameter can be calculated accordingly.

4.3.3.4 Tetrahedral Axial Nematics

We call an arrangement of regular tetrahedra a *tetrahedral axial nematic* if a specific axis of each particle (modulo symmetry operations) aligns with a common director on average. Here we consider three plausible axes for such an alignment i.e. the normal to a face, an edge and the vector connecting the centers of two non-adjacent edges and the corresponding phases will be called the '*face nematics*', the '*edge nematics*' and '*z nematics*' respectively. Considering the inversion symmetry of these structures, the expected value of $\mathcal{H}_4$ will be used as a measure of order:

$$M^{ijkl}(\mathcal{H}_3) = \sum_{p=1}^{4} v_p^i v_p^j v_p^k v_p^l - \frac{4}{15}\left(\delta^{ij}\delta^{kl} + \delta^{ik}\delta^{jl} + \delta^{il}\delta^{jk}\right)$$

$$\overline{M}^{ijkl} = \frac{1}{N}\sum_{q=1}^{N}\sum_{p=1}^{4} v_p^i v_p^j v_p^k v_p^l - \frac{4}{15}\left(\delta^{ij}\delta^{kl} + \delta^{ik}\delta^{jl} + \delta^{il}\delta^{jk}\right)$$

For each structure, $\mathfrak{M}_\Omega$ is calculated by decomposing v_p's to vectors *along* and *perpendicular* to the director z and calculating the corresponding expected values using Proposition IV.3. More specifically, if $v_p = \alpha_p z + \beta_p t_p$, we have:

$$
\begin{aligned}
\mathfrak{M}_\Omega^{ijkl} = {}& \sum_{p=1}^{4}\alpha_p^4 z^i z^j z^k z^l + \frac{1}{8}\sum_{q=1}^{4}\beta_q^4\Big[3x^i x^j x^k x^l + 3y^i y^j y^k y^l + x^i x^j y^k y^l + y^i y^j x^k x^l \\
& + x^i y^j y^k x^l + y^i x^j x^k y^l + x^i y^j x^k y^l + y^i x^j y^k x^l\Big] + \\
& + \frac{1}{2}\sum_{p=1}^{4}\alpha_p^2\beta_p^2\Big[x^i x^j z^k z^l + y^i y^j z^k z^l + z^i z^j x^k x^l + z^i z^j y^k y^l \\
& + z^i x^j x^k z^l + z^i y^j y^k z^l + x^i z^j z^k x^l + y^i z^j z^k y^l + \\
& + z^i x^j z^k x^l + z^i y^j z^k y^l + x^i z^j x^k z^l + y^i z^j y^k z^l\Big] \\
& - \frac{4}{15}\left(\delta^{ij}\delta^{kl} + \delta^{ik}\delta^{jl} + \delta^{il}\delta^{jk}\right)
\end{aligned}
\tag{4.38}
$$

$\mathfrak{M}_\Omega$ clearly satisfies the properties outlined in Section 4.3.3. The extent of order can therefore be quantified by solving the following associated optimization problem:

$$
\begin{aligned}
\max \quad & \omega\overline{M}^{ijkl}z^i z^j z^k z^l \\
\text{subject to} \quad & z^i z^i = 1
\end{aligned}
\tag{4.39}
$$

where $\omega = \sum_{p=1}^{4}[\alpha_p^4 + \frac{3}{8}\beta_p^4 - 3\alpha_p^2\beta_p^2]$. (The derivation details can be found in Appendix B.6.) Note that for negative values of the coefficient, solving (4.39) reduces to *min-*

Table 4.2: Projection parameters and ω for face, edge and z uniaxial nematics.

Phase	α_1	β_1	α_2	β_2	α_3	β_3	α_4	β_4	$\sum_{i=1}^{4}\alpha_i^4$	$\sum_{i=1}^{4}\alpha_i^2\beta_i^2$	$\sum_{i=1}^{4}\beta_i^4$	ω
Face Nematics	1	0	$-\frac{1}{3}$	$\frac{\sqrt{8}}{3}$	$-\frac{1}{3}$	$\frac{\sqrt{8}}{3}$	$-\frac{1}{3}$	$\frac{\sqrt{8}}{3}$	$\frac{28}{27}$	$\frac{8}{27}$	$\frac{64}{27}$	$\frac{28}{27}$
Edge Nematics	$\frac{\sqrt{6}}{3}$	$\frac{\sqrt{3}}{3}$	$-\frac{\sqrt{6}}{3}$	$\frac{\sqrt{3}}{3}$	0	1	0	1	$\frac{8}{9}$	$\frac{4}{9}$	$\frac{20}{9}$	$\frac{7}{18}$
z nematics	$\frac{\sqrt{3}}{3}$	$\frac{\sqrt{6}}{3}$	$\frac{\sqrt{3}}{3}$	$\frac{\sqrt{6}}{3}$	$-\frac{\sqrt{3}}{3}$	$\frac{\sqrt{6}}{3}$	$-\frac{\sqrt{3}}{3}$	$\frac{\sqrt{6}}{3}$	$\frac{4}{9}$	$\frac{8}{9}$	$\frac{16}{9}$	$-\frac{14}{9}$

imizing $\overline{M}^{ijkl}z^i z^j z^k z^l$ under the same constraints. Table 4.2 gives the corresponding coefficients and OOPs for face, edge and z nematics. Unlike face and edge nematics, for z nematics, the corresponding prefactor is negative and the associated minimization problem is to be solved. The scalar order parameter can also be defined using (4.21).
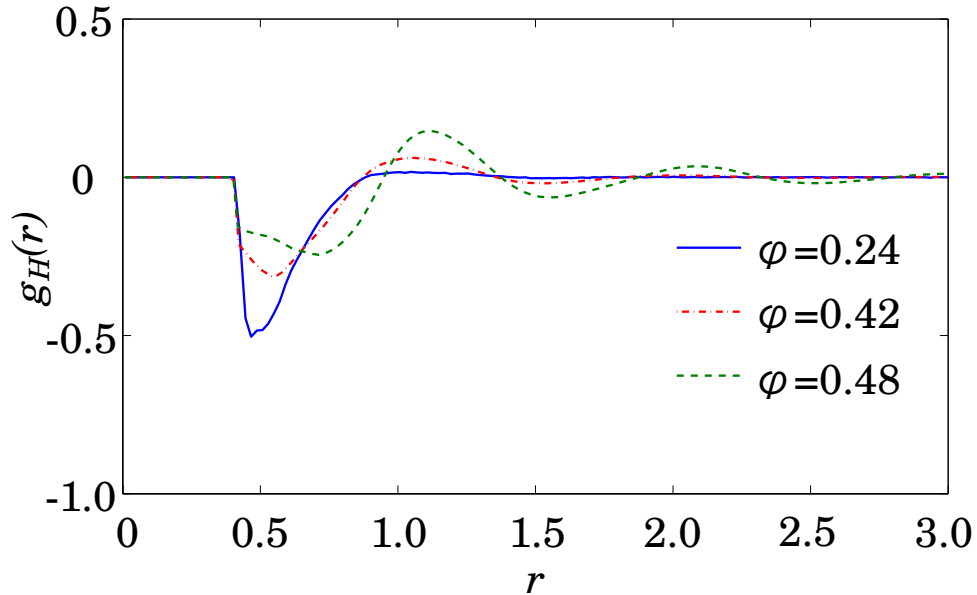
Figure 4.3: $g_H(r)$ calculated for a fluid of 4,096 hard tetrahedra at different packing fractions.

4.4 Other applications of Strong Rotational Coordinates

So far, we have only used SOCs to quantify global orientational order in systems of particles with non-trivial rotational symmetries. In this section, other potential

Table 4.3:
Expected values of $\mathcal{H}_{3,u}^{ijk}\mathcal{H}_{3,v}^{ijk}$ and $g_H(r)$ for different layers of nearest neighbors in the hard tetrahedron system

nth nearest neighbor	$\mathcal{H}_{3,u}^{ijk}\mathcal{H}_{3,v}^{ijk}$	$g_H(r)$
1	$-\frac{32}{81}$	$-\frac{1}{9}$
2	$-\frac{2464}{2187}$	$-\frac{77}{243}$
3	$\frac{79072}{59049}$	$\frac{2471}{6561}$

applications of SOCs in computational studies of soft condensed matter are discussed.

4.4.1 Time-averaged Orientations

The machinery of strong orientational coordinates can be used to calculate the average orientation of a particle in the course of a simulation. In general, this is done to get rid of thermal fluctuations, and can be considered the equivalent of determining inherent structures in energetic systems. More precisely, if $M_p(t)$ is the strong orientational coordinate of particle p at time t, one can maximize $\mathfrak{M}_p \odot \frac{1}{T}\int_0^T M_p(t)dt$ to find for the best average orientation that matches the trajectory of p. This problem also fits into the general class of problems outlined in Example 1.

4.4.2 Spatial Correlation of Local Rotational Order

Spatial correlation of orientations of particles in a simulation can also be quantified using SOCs. We explain this through an example; i.e. a fluid of regular hard tetrahedra. At low packing fractions, hard tetrahedra form a simple fluid where the orientations of different particles are not correlated. This however changes as the packing fraction increases, and a network structure emerges in which all neighboring particles are in a face-to-face configuration. At sufficiently high densities, this network transforms into a dodecagonal quasicrystal that is locally similar to the network fluid preceding it but has long-range quasiperiodicity [18].

It was shown in Section 4.3.2.4 that $\mathcal{H}_3(\mathcal{O}_t)$ is the strong orientational coordinate for a regular tetrahedron. Now consider an arrangement of N tetrahedra and let $N(r)$ be the number of pairs whose center-to-center distance lies in the interval $[r - dr/2, r + dr/2]$ and define the following correlation function:

$$g_H(r) = \frac{1}{N(r)\,\|\mathcal{H}_3\|_F^2} \sum_{<p,q>,|d(p,q)-r|<dr/2} \mathcal{H}_{3,p}^{ijk}\mathcal{H}_{3,q}^{ijk} \qquad (4.40)$$

We can quantify these differences using (4.40). We first analyze different scenarios for the first shell of nearest neighbors. Geometrically, the smallest distance between the centroids of two neighboring tetrahedra is achieved when their faces are touching one another. At low densities, the two neighboring tetrahedra can freely rotate around the axis perpendicular to this touching face, while at higher densities, rotation is restricted and two faces tend to match perfectly. The value of $g_H(r)$ can be calculated for these two idealized configurations. Let z be perpendicular to the common plane of the two touching faces. The characteristic vectors of each tetrahedron can thus be expressed as:

$$
\begin{aligned}
u_1 &= z \\
u_p &= -\frac{1}{3}z + \frac{\sqrt{8}}{3}s_p \\
v_1 &= -z \\
v_p &= \frac{1}{3}z + \frac{\sqrt{8}}{3}t_p
\end{aligned}
$$

with:

$$
\begin{aligned}
s_p &= \cos\frac{2\pi(p-2)}{3}x + \sin\frac{2\pi(p-2)}{3}y \\
t_p &= \cos\left(\frac{2\pi(p-2)}{3}+\theta\right)x + \sin\left(\frac{2\pi(p-2)}{3}+\theta\right)y
\end{aligned}
$$

and x and y two orthonormal vectors perpendicular to z. For a perfect non-rotating face-to-face contact, $\theta = 0$ and $\mathcal{H}_{3,u}^{ijk}\mathcal{H}_{3,v}^{ijk} = -\frac{32}{81}$; the corresponding value of $g_H(r)$ will be $-\frac{1}{9} \approx -0.11111$. For a freely-rotating face-to-face contact however, $\theta \sim U(0, 2\pi)$ and $\langle\mathcal{H}_{3,u}^{ijk}\mathcal{H}_{3,v}^{ijk}\rangle = -\frac{160}{81}$ which yields a $g_H(r)$ value of $-\frac{5}{9} \approx 0.5555$. Investigating the magnitude of the first valley of $g_H(r)$ will elucidate the type of the face-to-face contact prevalent in that particular phase. Fig. 4.3 shows $g_H(r)$ vs. r for three different packing fractions. At $\phi = 0.24$, the first valley of $g_H(r)$ has a value of -0.503 which is very close to the calculated value of $-\frac{5}{9}$ which is expected in a freely rotating face-to-face configuration. At a packing fraction of 0.48 however, the first valley value observed in simulation is around -0.15 which is also very close to the theoretically predicted value of $-\frac{1}{9}$ for the perfect face-to-face configuration. At intermediate densities (the red curve), the first valley is in between as expected. This analysis can be extended to second and third nearest neighbors. For perfect face-to-face configurations, the corresponding values of $\mathcal{H}_{3,u}^{ijk}\mathcal{H}_{3,v}^{ijk}$ and $g_H(r)$ are given in Table 4.3. The $g_H(r)$ values shown in Fig. 4.3 are consistent with these theoretically

predicted values, which confirms the existence of the network structure where all neighbors are in a perfect face-to-face contact.

CHAPTER V

Hard Tetrahedra: Self-assembly Simulations[1]

This chapter is dedicated to studying dense packings of regular tetrahedra using Monte Carlo simulations. In Section 5.1, some background information is provided as well as the technical specifications of the performed simulations. Section 5.2 discusses all dense packings of tetrahedra introduced prior to the completion of this project. Thermodynamics of the hard tetrahedron fluid is discussed in Section 5.3, while a discussion of the quasiperiodic phases formed by hard tetrahedra is given in Section 5.4. The effect of polydispersity on the formation of the quasicrystal is discussed in Section 5.5 and concluding remarks are reserved for Section 5.6.

5.1 Preliminaries

The regular tetrahedron is the Platonic solid P_5 and the uniform polyhedron[2] U_1. It is the only self-dual Platonic solid and lacks central symmetry. The vertices of a prototypical tetrahedron, which we also use as a reference in our simulations by assigning to it the quaternion $\mathbf{q} = (1, 0, 0, 0)$, are given by:

$$(+1, +1, +1) \tag{5.1a}$$

$$(+1, -1, -1) \tag{5.1b}$$

$$(-1, +1, -1) \tag{5.1c}$$

$$(-1, -1, +1) \tag{5.1d}$$

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[1]This chapter is partly based on the following manuscript: Amir Haji-Akbari, Michael Engel, *et al. Nature* **462**: 773–777 (**2009**) [18].

[2]Uniform polyhedra are the polyhedra with identical vertices. They include *Platonic solids* and *Kepler-Poinsot solids*

Table 5.1: Geometric features of a regular tetrahedron with edge length σ.

Property	Value
Volume	$\dfrac{\sqrt{2}}{12}\sigma^3$
Surface area	$\sqrt{3}\sigma^2$
Radius of Gyration	$\dfrac{\sigma}{2\sqrt{5}}$
Dihedral Angle	$\cos^{-1}\dfrac{1}{3} \approx 70.53°$
Solid Angle	$3\cos^{-1}\dfrac{1}{3} - \pi \approx 31.5863°$
Order of Rotation Group	12
Order of Symmetry Group	24

Indeed this is the smallest regular tetrahedron with all its vertices having integer coordinates, and has an edge length of $\sigma = \sqrt{8}$ and a volume of $V_T = 8/3$. Regular tetrahedron has seven axis of rotation (four C_3 axes and three C_2 axes) and six mirror planes. Its rotation group T and its symmetry group T_d have twelve and twenty-four elements respectively. Table 5.1 summarizes important geometric features of the regular tetrahedron.

As explained in Section 2.3.2, the problem of packing tetrahedra has been extensively studied. There have however been fewer studies of the thermodynamics of the hard tetrahedron system. In 1970, Gibbons studied the equation of state of a fluid of polydisperse hard regular tetrahedra– along with four other convex shapes– to assess the applicability of the scaled particle theory to systems of nonspherical hard particles [196]. In 1995, Kolafa and Nezbeda used Monte Carlo simulations to compute a low-density equation of state for the hard tetrahedron fluid, which they used as a model of liquid water [114]. The fact that many hard particle systems form liquid crystalline phases fueled the speculation that, at intermediate packing fractions, hard tetrahedra might also form liquid crystalline phases such as the *tetrahedratic phase* [194], which has been shown to have interesting hydrodynamic properties [197].

Studying the thermodynamics of the hard tetrahedron system is of more practical relevance today due to the recent synthesis of tetrahedrally-shaped nano- and colloidal particles [57, 66, 67, 73–76, 198]. The nanoparticles are made of a wide range of materials including platinum [73], gold [57, 66, 67], tin sulfide [74] and silicon [75, 76]. Micron-size colloidal tetrahedra made of colloidal spheres have also been reported [198]. As explained in Section 2.2, these particles can behave as hard particles under certain circumstances.

We use isobaric and isochoric Monte Carlo simulations with periodic boundary conditions to study systems of N regular tetrahedra, with N ranging from 512 to $21,952$. (General features of the applied Monte Carlo method is given in Section 3.2.1.) Simulations are initialized at low packing fraction in a random configuration and subsequently compressed to higher densities. The dodecagonal quasicrystals shown in Figs. 5.10, 5.11, 5.12 and 5.13 are obtained in isochoric simulations at packing fraction $\phi = 50\%$. Crystallization proceeds in three steps:

1. equilibration of the dense, metastable fluid. In a system of $N = 8,000$ particles, this step takes $< 12 \times 10^6$ MC cycles.

2. nucleation and growth, which takes between $12 - 23 \times 10^6$ MC cycles.

3. healing of defects, which starts after 23×10^6 Monte Carlo cycles.

The equation of state in Fig. 5.5a is calculated by increasing (or decreasing) the external pressure stepwise for compression (or expansion). Longer simulations facilitate equilibration in the transition region. For $P^* > 120$, compression with conventional Monte Carlo is inefficient. Therefore we apply the alternative method described in Section 3.2.1.6 to reach pressures as large as $P^* \leq 10^6$ and obtain maximum density packings.

5.2 Dense Packings of Tetrahedra

Regular tetrahedra do not tile the Euclidean space. However, if extra space is allowed between tetrahedra, or between groups of tetrahedra, dense ordered structures become possible. An *ex novo* approach for obtaining dense packings of tetrahedra is to construct locally dense clusters of tetrahedra and use them to build periodic dense superlattices. In this section, we introduce these locally dense clusters and the packings that can be constructed from them.

5.2.1 Locally Dense Clusters of Tetrahedra

Imagine building a dense cluster by adding one tetrahedron at a time. A *pnetagonal dipyramid (PD)* (Fig. 5.1a) is easily built from five tetrahedra that share a common edge. An internal gap of $2\pi - 5\cos^{-1}(1/3) \approx 7.36°$ is introduced in this process. Two pentagonal dipyramids can share a central tetrahedron to form a *nonamer* (Fig. 5.1b) in which the principal axes of the two interpenetrating PDs are along the opposite edges of the central tetrahedron, and are therefore perpendicular to one another.

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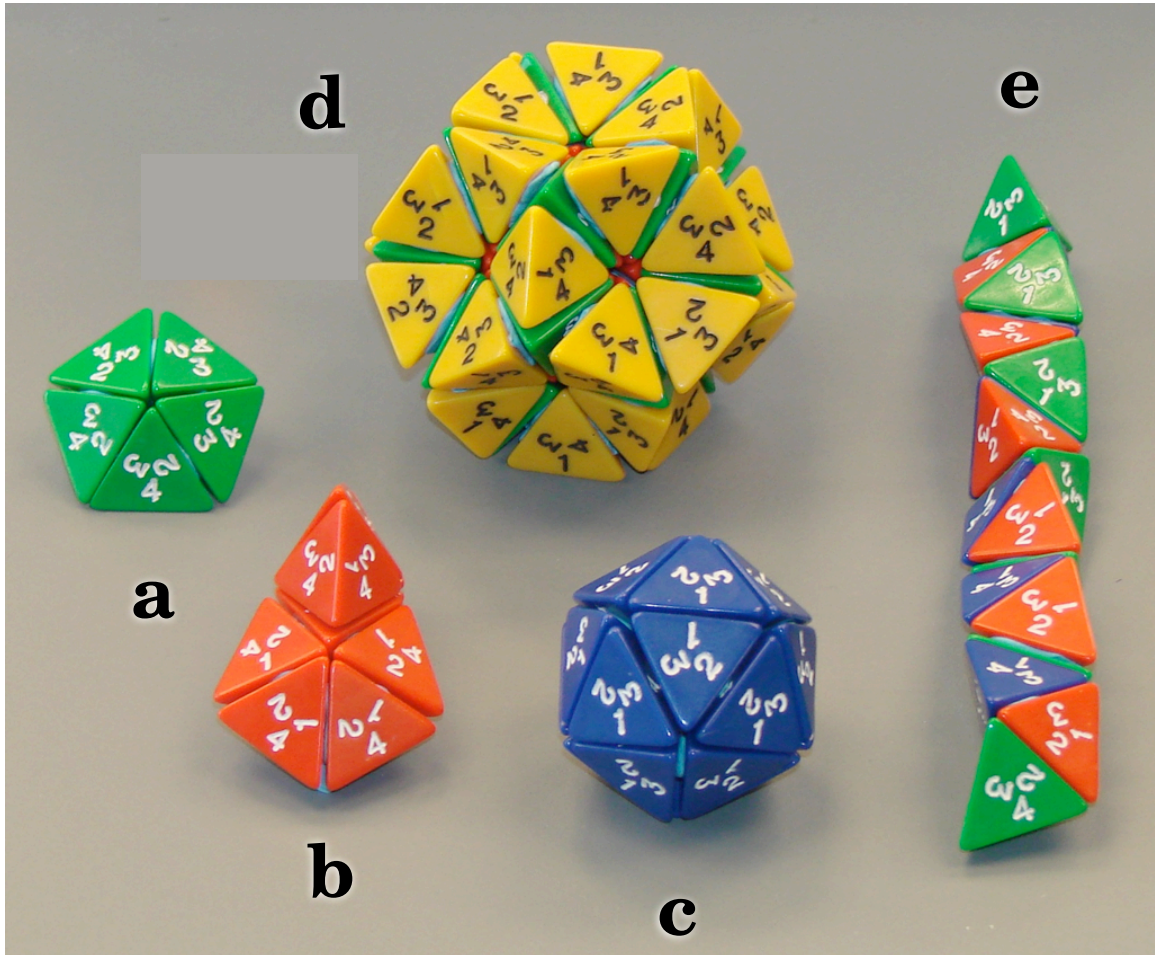
Figure 5.1:
Ideal local packing motifs built from tetrahedral dice stuck together with modeling putty. The pentagonal dipyramid (a), the nonamer (b) and the icosahedron (c) maximize local packing density. The icosahedron can be extended by adding a second shell (d), but then the large gaps between the outer tetrahedra lower the density. The tetrahelix (e) maximizes packing density in one dimension. (Images reproduced from [18])

Twelve interpenetrating pentagonal dipyramids define an *icosahedron* (Fig. 5.1c). It has twenty tetrahedra with a gap of $24\pi - 60\cos^{-1}(1/3) \approx 1.54$ steradians. A *hexecontahedron* (Fig. 5.1d), a motif with 70 tetrahedra, is obtained by adding an additional shell of tetrahedra to an icosahedron.

For later use, we introduce here a dense, one-dimensional packing given by a linear arrangement of tetrahedra with touching faces. It is called a *tetrahelix* or a *Bernal spiral*, and was first discovered by Hurley [199]. A tetrahelix has no internal gaps and therefore maximizes the packing fraction in one dimensions. It is also not periodic since the phase difference between two consecutive tetrahedra, $\theta_0 = \cos^{-1}(2/3)$ is not a rational multiple of 2π. Periodic tetrahelices can however be constructed by allowing minor gaps between consecutive tetrahedra.

In Fig. 5.1, tetrahedral dice are stuck together with modeling putty, which distributes the gap that would be present in each motif if most of the adjacent faces were touching. Pentagonal dipyramids and icosahedra are locally dense, but exhibit noncrystallographic symmetries. The problem of extending or arranging them into space-filling bulk structures is nontrivial. For example, adding a second shell to the icosahedron generates the hexecontahedron which has 70 tetrahedra, but decreases the packing fraction. However several relatively dense packings were obtained by arranging these motifs into dense structures that are explained in the following section.

5.2.2 Existing Dense Packings of Tetrahedra

The dense packings presented here are obtained from the numerical compression of geometrically constructed ordered arrangements of tetrahedra. They are either obtained by arranging the motifs shown in Fig. 5.1, or based on Frank-Kasper phases [200].

5.2.2.1 Nonamers

The nonamer crystal proposed by Chen [28] has a fundamental domain with eighteen tetrahedra, broken into two nonamers that are related to one another by inversion. These 'positive' and 'negative' nonamers form two alternating layers that are shown in 'blue' and 'red' in Fig. 5.2. The densest packing has a sheared tetragonal lattice and a maximum packing fraction of 77.86%. Torquato and Jiao numerically compress a unit cell to a packing fraction of 78.20% [29]. Using the compression algorithm described in Section 3.2.1.6 however, we obtain packing fractions as high as 78.38% (Appendix. C.2.1).
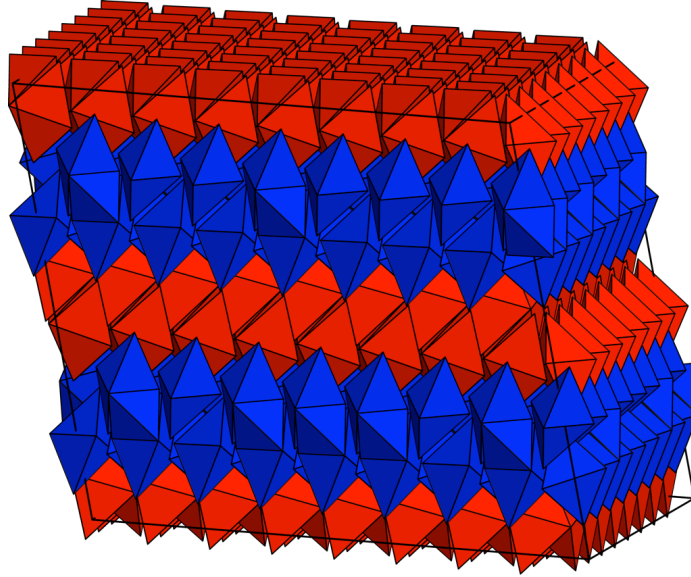
Figure 5.2: **The nonamer crystal.** $8 \times 8 \times 2$ unit cells of the nonamer crystal constructed by Chen [28]. Note the alternating layers of nonamers of opposite orientations that are depicted in red and blue, respectively.

5.2.2.2 Icosahedra

The densest cluster that can be obtained by packing tetrahedra around a single point is an icosahedron. There are two ways of arranging icosahedral clusters into dense packings of tetrahedra. One way is to arrange non-interpenetrating icosahedral clusters into the lattice packing of icosahedra proposed by Betke and Henk [131]. This structure was first proposed by Conway and Torquato [27], who called it the *reformed Scottish packing* (Fig. 5.3a). The original structure had a packing fraction of $\frac{45}{64} \approx 70.3125$. The authors achieved slightly higher packing fractions $> 71.655\%$ by moving the tetrahedra slightly; they speculate that further optimization of the same structure can give rise to packing fractions as high as 72%, but the arrangement cannot surpass $\phi_{\max} = \pi/\sqrt{18}$ of the densest packing of congruent spheres. We however compress this structure to a maximum packing fraction of 74.36%, which is denser than the densest sphere packing (Appendix C.2.2).

The maximum packing fraction can be increased by forming logs of interpenetrating icosahedral clusters and arranging them into a hexagonal lattice. Additional particles can be added in the void area between logs to increase the packing fraction even further. This is what we call the *fivefold log packing* (Fig. 5.3b), which can be numerically compressed to a packing fraction of 76.73% (Appendix C.2.3).
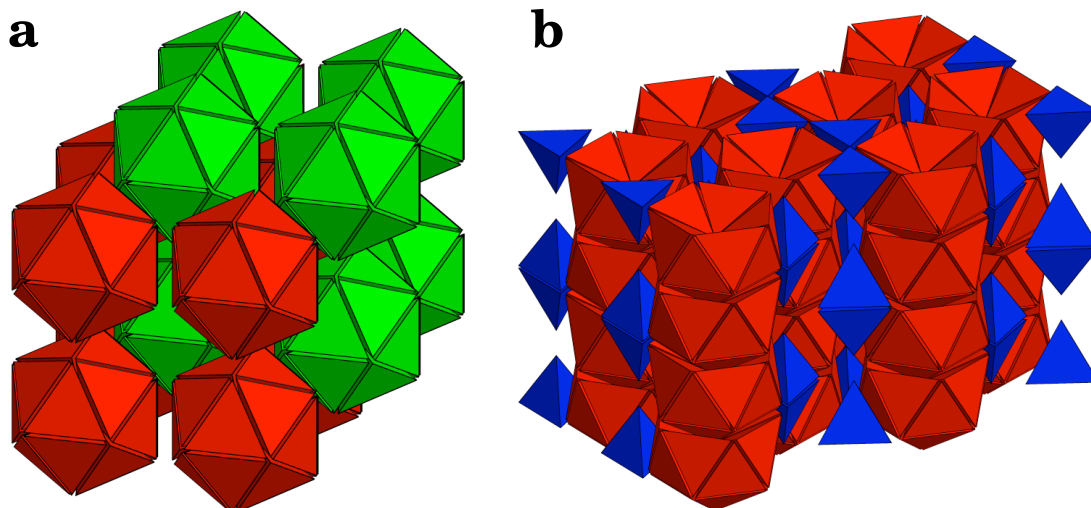
Figure 5.3: **Dense packings of regular tetrahedra with icosahedral motifs**: (a) a lattice packing of icosahedral motifs, (b) logs of interpenetrating icosahedra forming a hexagonal lattice, blue interstitials are added for increasing the packing fraction.

5.2.2.3 Frank-Kasper Phases

A different approach makes use of tetrahedrally close-packed structures, known as Frank-Kasper phases in atomic crystals [200]. These phases have tetrahedral interstices, which means that every vertex in their Voronoi decomposition is shared by four Voronoi cells only. In other words, the dual tessellation of the Voronoi decomposition is only made of irregular tetrahedra. A dense packing of regular tetrahedra can therefore be obtained from a Frank-Kasper phase by largest tetrahedra that can fit *inside* all its irregular interstices. Not all FK phases can be used for this purpose as the amount of irregularity in the tetrahedron network might be too high, which will make the corresponding packing very dilute, and practically uninteresting.

Conway and Torquato [27] utilized this approach to obtain dense packings of tetrahedra (Fig. 5.4). They obtain the simplest phase from the Voronoi tessellation of the body-centered cubic (BCC) lattice, and call it the *Scottish bubbles packing* (Fig. 5.4a). The Voronoi cells are truncated octahedra ($6^8 4^6$). The original structure is not particularly dense and has a packing fraction of $\frac{1}{2} = 50\%$ only. Using our compression algorithm, we can compress it to $\phi = 60.36\%$ (Appendix C.2.4).

They however obtain their densest *Welsh bubbles* packing (Fig. 5.4b) by decorating
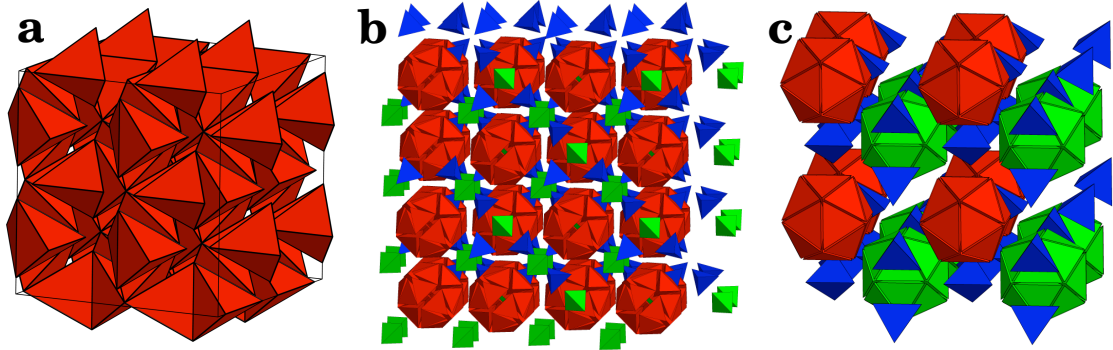
Figure 5.4:

Packings of tetrahedra obtained from Frank-Kasper phases: (a) Scottish bubbles packing obtained from the Voronoi decomposition of the BCC lattice, (b) Welsh bubbles packing based on Type II Clathrate structure, (c) Irish bubbles packing based on Type I Clathrate structure.

a Type II clathrate structure, which have two types of Voronoi cells- which they call bubbles- in its fundamental domain: sixteen small dodecahedra (5^{12}), and eight large hexadecahedra ($5^{12}6^4$); their centers form the C15 cubic laves structure, which is one of the most common intermetallic phases. The structure is very complicated and has 136 particles in the unit cell. It was the densest structure discovered by Conway and Torquato with $\phi_{\max} = 71.7455\%$. Using our compression algorithm, we were able to compress this structure to $\phi = 76.87\%$ (Appendix C.2.5).

They also applied the same procedure for the type I clathrate phase to obtain *Irish bubbles* packing (Fig. 5.4) which is not particularly dense. There are two types of Voronoi cells– bubbles– in the unit cell of the Irish bubbles packing. Smaller bubbles are dodecahedra (5^{12}). There are two of them in the fundamental domain and their centers lie on a BCC lattice. The bigger bubbles are tetradecahedra ($5^{12}6^4$). Together the centers of the bubbles form form the A15 (Cr_3Si) structure [201]. The structure has 46 tetrahedra per unit cell.

5.3   The Hard Tetrahedron Fluid

The packings mentioned in Section 5.2.2 are obtained either from geometric construction, or from numerical compression of geometrically constructed structures, and none of them has ever been observed in simulation or experiment. In order to obtain dense packings of tetrahedra, we follow a conceptually different approach i.e. we

use Monte Carlo simulations to let the system find dense packings itself by evolving it according to the rules of statistical mechanics. As mentioned in Section 2.2.1, a disorder-order transition is observed in Monte Carlo simulations of most hard particle systems, and most often, the ordered phase can be compressed into the densest known packing of that particular building block. In our Monte Carlo simulations of hard tetrahedra, we observe a disorder-order transition at sufficiently high packing fractions. A dodecagonal quasicrystal spontaneously assembles from the disordered fluid at $\phi \geq 50\%$, which can then be compressed to a packing fraction of 83.24%, the world record at the time of its discovery[3]. But before going through the properties of the ordered phase, we discuss the thermodynamics of the disordered fluid.

Fig. 5.5a shows the equation of state $\phi(P^*)$ obtained from simulations of a small system with 512 tetrahedra and a larger system with 4,096 tetrahedra. Here, $P^* = P\sigma^3/k_BT$ is the reduced pressure and σ the edge length of a tetrahedron. The large system undergoes a first-order transition on compression of the fluid phase and forms a quasicrystal. For the small system however, the equilibrium packing fraction exhibits an S-shaped transition at $P^* = 58$ and $\phi = 47\%$ from a simple fluid to a more complex fluid, discussed below. The same complex fluid forms in larger systems if the compression is too fast, i.e. if enough time is not given to the system for equilibration. The disordered fluid jams at sufficiently high pressures, and when compressed to nearly infinite pressure, attains packing fractions of $\geq 78\%$.

Fig. 5.6a shows a system of $N = 8,000$ tetrahedra rapidly compressed to a packing fraction of $\phi = 78.58\%$. This configuration is the densest known packing of disordered tetrahedra. We confirm the lack of global translational and orientational order in the system by calculating diffraction images, bond order diagrams and third rank tensor orientational order parameters derived in Chapter IV. The top row of Fig. 5.6b shows bond order diagrams obtained by projecting the vectors connecting the centers of nearest neighbor tetrahedra separated by $\leq 0.55\sigma$ on the surface of a sphere and the bottom row depicts diffraction patterns. As it can be seen bond order diagrams are uniform on S^2 and diffraction patterns are isotropic in all directions. Rotational order parameters calculated from third-rank tensor SOCs are also very small, and within the noise of a finite system.

In order to understand the structure of the complex fluid, we analyze the system for the presence of locally dense motifs introduced in Fig. 5.1. In particular, we count the PDs and the icosahedra that are visually observed in jammed configurations of the disordered fluid. We identify these motifs using a shape-matching algorithm described

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[3]Since then, we have obtained quasicrystals with packing fractions as high as 83.48%.
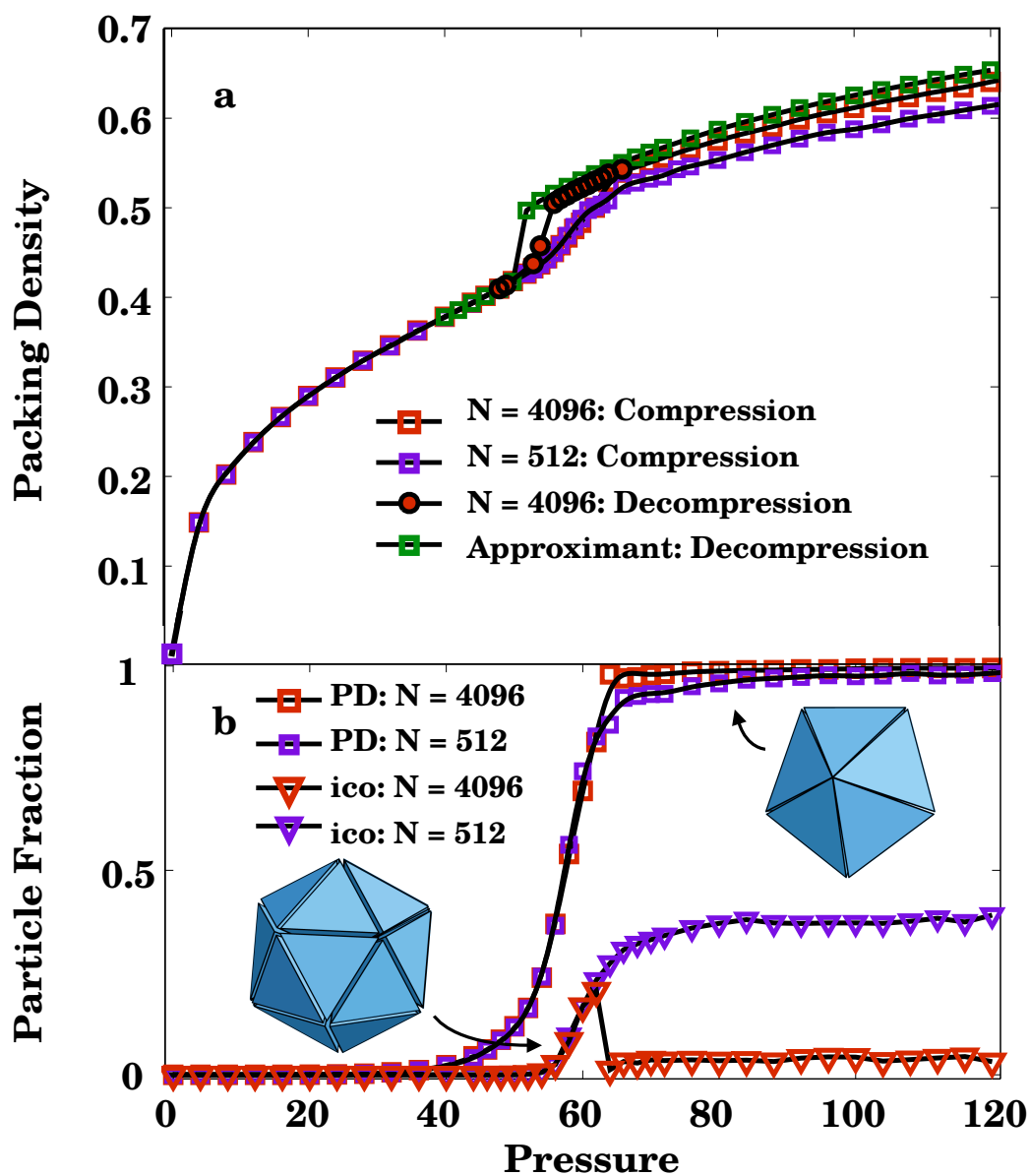
Figure 5.5:

Thermodynamics of the hard tetrahedron fluid. (a) Equation of state spanning the transition from the liquid to the solid state. Data are shown for various system sizes. Decompression of both the quasicrystal ($N = 4{,}096$) and the approximant (unit cell $N = 82$) shows a sharp melting transition. Hysteresis of the compression and decompression curves for the quasicrystal further indicates a first-order transition. For the system with $N = 512$, crystallization is inhibited in many runs, producing a jammed, disordered glass. (b) Fraction of tetrahedra participating in pentagonal dipyramids (PD, right inset) and icosahedra (Ico, left inset). (Images reproduced from [18])
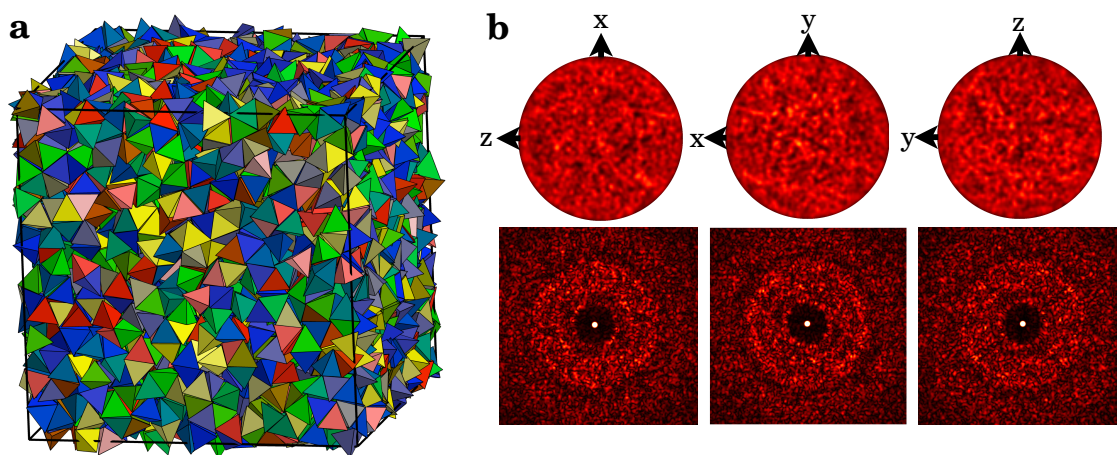
Figure 5.6:

The densest known disordered arrangement of tetrahedra (a) In short simulations or if the box is compressed too rapidly, crystallization does not occur. The image shows a disordered arrangement of $N = 8,000$ tetrahedra, compressed to $\phi = 78.58\%$, the densest known packing of disordered tetrahedra (Appendix C.2.7). Local ordering of the tetrahedra is visible. (b) Bond order diagrams (top) and diffractions patterns (bottom) of the disordered glass. (Images reproduced from [18])

by Keys *et al.* [191, 192]. The first step is to identify 'rough' candidates of each motif. For PDs, this is done by constructing the graph of nearest neighbors and isolating all pentagons on that graph. We define two particles as nearest neighbors if their distance is $\leq 0.65\sigma$. For icosahedra, we look for clusters of 20 tetrahedra packing around a single point.

After identifying these candidates, we compare them with perfect PDs and icosahedra using shape matching. For each motif, certain characteristic directions are projected onto the surface of a unit sphere. For a PD, these are the vectors connecting the center of the pentagon to its vertices, and for an icosahedron, the vectors connecting the center of the icosahedron to the centers of its faces. Each unit vector $\mathbf{u}$ is then mapped onto spherical angles (θ, ϕ), which are then used to calculate different spherical harmonic components $Y_{lm}(\theta, \phi)$. For a collection of characteristic vectors $\{\mathbf{u}_i\}$, $\sum_i Y_{lm}(\theta_i, \phi_i)$'s are calculated for a few values of l[4]. The corresponding Y_{lm} spectrum of the candidate motfi is then compared to that of the reference motif– i.e. the perfect motif– by measuring their scalar distance[5]. If the extent of matching is higher than a certain threshold, the candidate motif is declared as a positive match. The fact that each particle might be– and usually is– a member of several interpenetrating motifs is also considered in our implementation.

Fig. 5.5b depicts the outcome of the shape-matching algorithm. We see that the fraction of tetrahedra belonging to at least one pentagonal dipyramid increases well before jamming or crystallization. With increasing pressure, interpenetrating pentagonal dipyramids form icosahedra and finally merge into a percolating pentagonal dipyramid network as the fraction of tetrahedra in pentagonal dipyramids approaches unity. For the large system, the fraction of tetrahedra in icosahedra suddenly drops at $P^* = 62$, when crystallization occurs. Comparison with the glass shows that far fewer icosahedra remain in the quasicrystal.

The fact that a single tetrahedron can be a part of several interpenetrating PDs can be used to define clusters of interconnected PDs. Such clusters are small and isolated at lower pressures, but as the simple fluid transforms into the complex fluid, they become larger and larger, until they merge together to form a large cluster that spans the entire system. This notion of clustering is the basis of the percolation analysis

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[4]In order to detect a particular motif, it is necessary that Y_{lm}'s are calculated for select values of l only. For both PDs and icosahedra, we use $l = 5, 6, \cdots, 10$ since both these motifs have fivefold symmetries. It should however be emphasized that optimal choices of l might be different for other motifs.

[5]We used the Euclidean 2-norm in the Hilbert space of spherical harmonics as the measure of distance in this study.

presented in Fig. 5.7. The size of a cluster is defined as the number of particles it contains, and a spanning cluster is defined as a cluster that spans the simulation box in all three directions. Our findings suggest a percolation transition of the PD network in both the small and large systems at $P_p^* = 58 \pm 2$, before both the jamming and crystallization. Our free energy calculations suggest that the disorder-order transition should occur at the same range (See Section 6.4). The formation of the PD network, and the complex fluid, might therefore be a precursor to crystallization and not an isolated thermodynamic event.

Structural changes in the fluid are revealed by the unusual behavior of its radial distribution function $g(r)$, as shown in Fig. 5.8a. We find that the first peak near $r = 0.75\sigma$ disappears upon compression at low pressure, only to reappear for higher pressure, splitting into two peaks at $r = 0.55\sigma$ and $r = 0.80\sigma$. The positions of these peaks are characteristic of face-to-face and edge-to-edge arrangements, respectively, within a single pentagonal dipyramid. This initial loss of structure with increasing pressure or packing fraction is strikingly different from the well-known behavior of the hard sphere system depicted in Fig. 5.8b, and underscores the influence of shape in dense packings.

5.4 The Quasicrystal

Quasicrystals are ordered solids that lack periodicity. Therefore their structure *cannot* be described with repeating unit cells. Aperiodic geometric patterns have been known for a long time. For instance, quasiperiodic tilings of two-dimensional space with polygons can be found in the Islamic decorations of mosques and shrines in the medieval era [202] (Fig. 5.9). Rigorous construction of two-dimensional aperiodic tilings can be traced back to 1960's, the most notable being the infamous *Penrose tiling* which only has two types of congruent tiles in it [203]. It was however inconceivable that solids with aperiodic patterns could exist in nature. The first such solid was discovered by *Daniel Shechtman* in 1981 [204]. It is a binary alloy of Al–Mn formed from rapid cooling of the melt. Its diffraction pattern has sharp peaks like a crystal, but contains long-range fivefold symmetry which is incompatible with periodicity. In 2011, Schechtman was awarded the Nobel Prize in Chemistry for discovering this structure, but not surprisingly his discovery did not receive due enthusiasm back in 1980's. Some famous scientists, such as the Nobel laureate Linus Pauling, questioned the possibility of quasiperiodic phases and tried to explain Schechtman's findings with theories such as *multiple twinning*. Indeed the issue was not fully settled until high-
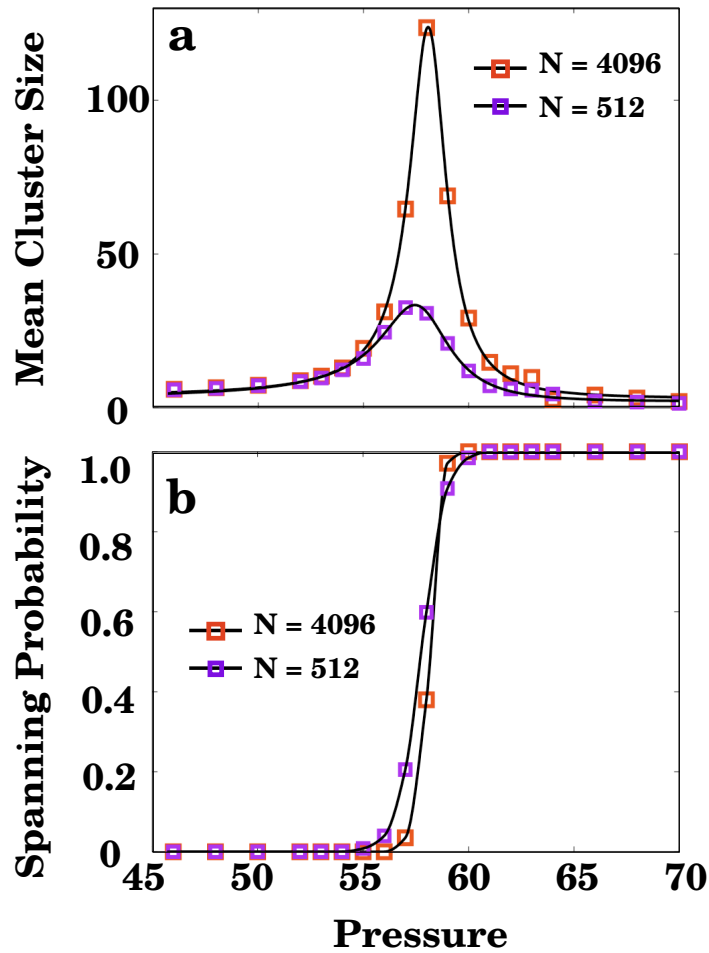
Percolation analysis of the PD network. (a) Mean cluster size of interpenetrating pentagonal dipyramids. (b) Spanning probability of the largest cluster of interpenetrating pentagonal dipyramids. (Images reproduced from [18])
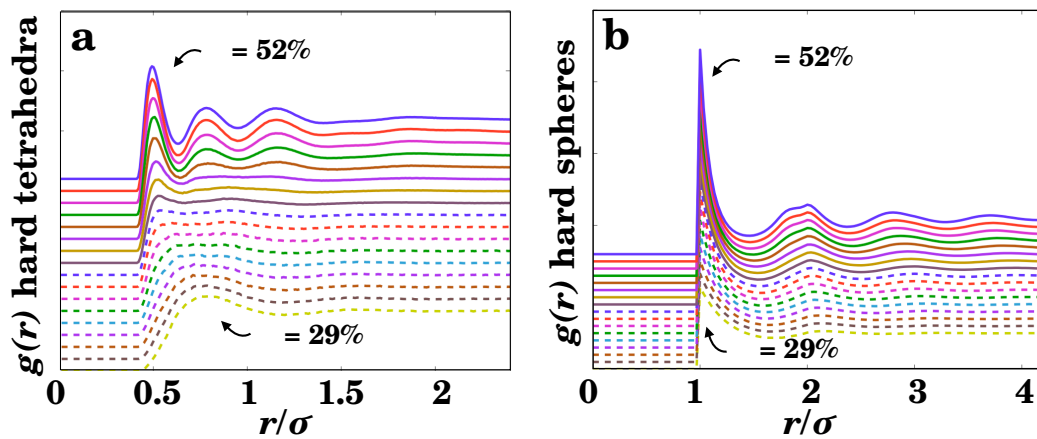
Figure 5.8:
Comparison of radial distribution functions of the hard tetra-hedron and the hard sphere fluids. (a) Radial distribution function $g(r)$ of regular tetrahedra for packing fractions ranging from $\phi = 29\%$ to $\phi = 52\%$. Curves are vertically offset for clarity. (b) Radial distribution function for the same densities as in (a) for a hard sphere system. (Images reproduced from [18])

Figure 5.9:
Islamic tilings with aperiodic patterns: (a) Darb-i Imam shrine, Isfahan, Iran (1453 C.E.) (b) Gunbad-i Kabud tomb tower in Maragha, Iran (1197 C.E.). (Images reproduced from [202])

resolution electron microscopy images of an Al–Mn–Pd alloy provided visual evidence for quasiperiodic arrangements of atoms in an alloy.

Shortly after Schechtman's groundbreaking discovery, quasicrystals with dodecagonal [205], decagonal [206] and eightfold [207] symmetries were also discovered. Since then, hundreds of quasicrystals have been observed, most of which being metastable states of binary and ternary alloys formed as a result of rapid quenching., there are however stable quasicrystals as well [208]. The overwhelming majority of identified quasicrystals are in atomic systems, and only a few quasicrystals have been recently observed in micellar systems [209–212].

The spontaneous formation of a quasicrystal from the hard tetrahedron fluid is remarkable since all previously observed crystalline structures of hard particles have unit cells consisting of only a few particles. Furthermore most simulation studies of quasicrystals employ exotic interparticle potentials with several minima [185, 213, 214], it is thus not expected *a priori* that interactions as simple as hard potentials can lead to quasiperiodic order. The quasicrystal formation is indeed robust and occurs for all systems with as few as 3,000 particles.

Fig. 5.10a depicts the quasicrystal that forms in a system of $N = 13,824$ tetrahedra. From Fig. 5.10b it can be seen that the quasicrystal consists of a periodic stack of corrugated layers with the spacing 0.93σ. The view along the direction of the stacking
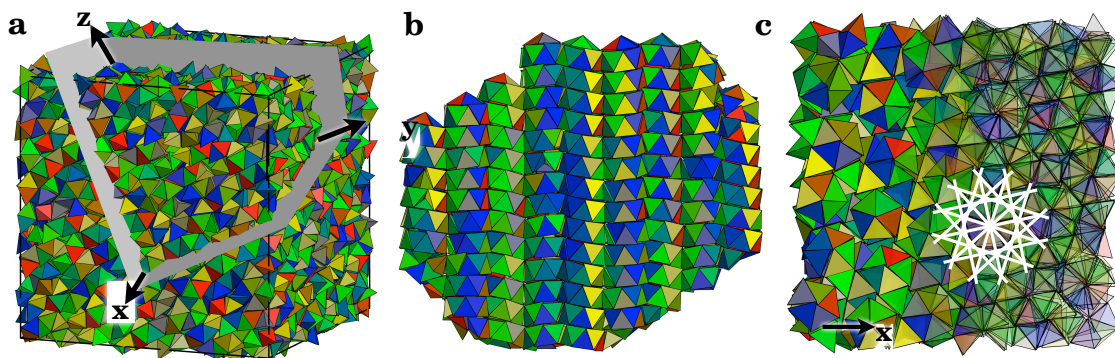
Figure 5.10: **The dodecagonal quasicrystal of hard tetrahedra**. A quasicrystal with packing fraction $\phi = 83.24\%$ obtained by first equilibrating an initially disordered fluid of 13,824 hard tetrahedra using Monte Carlo simulation and subsequent numerical compression. The images show an opaque view of the system (a) and opaque and translucent views of two rotated narrow slices (b) and (c). The white overlay in (c) shows the distinctive twlvefold symmetry of the dodecagonal quasicrystal. Corrugated layers with normals along the **z** axis are apparent in (b). The coloring of the tetrahedra is based on their orientation. (Images reproduced from [18])

vector (Fig. 5.10c) reveals details of the structure within the layers. Twelvefold symmetric rings formed by interpenetrating tetrahelices exist throughout the structure. The helix chirality is switched by 30° rotations, lowering the symmetry and resulting in a generalized point group of D_{6d} [215].

The structure of the quasicrystal can be understood more easily by examining the dual representation constructed by connecting the centers of mass of neighboring tetrahedra; i.e. the tetrahedra distanced within the first peak of the radial distribution function. In the dual representation, pentagonal dipyramids are represented by pentagons. The mapping is applied to a layer of an 8,000-particle quasicrystal in Fig. 5.11a. Recurring motifs are rings of twelve tetrahedra that appear as dodecagons in the dual representation. Each ring is capped with a pentagonal dipyramid. These PD-capped rings are stacked periodically to form logs (Fig. 5.11b), similar to the hexagonal antiprismatic clusters in the tantalum–tellurium system [212]. As indicated in Fig. 5.11a, the symmetry axes of the logs arrange into a non-repeating pattern of squares and triangles (tile edge length 1.83σ)), an observation that we confirm in systems with 13,824 and 21,952 particles. Note that additional interstitial

particles are also needed to fill in the void that exists at the center of each triangle and square. The diffraction pattern obtained by positioning scatterers at the centers of tetrahedra shows rings of Bragg peaks, indicating the presence of long-range order with twelvefold symmetry not compatible with periodicity. The effect of choosing different scatterers e.g. vertices of tetrahedra rather than their centers is explained in Appendix D.

Perfect quasicrystals are aperiodic while extending to infinity; they therefore cannot be realized in experiments or simulations, which are, by necessity, finite. The observed tilings and diffraction patterns with twelvefold symmetry are sufficient in practice for the identification of our self-assembled structures as dodecagonal quasicrystals. Such an identification is in agreement with previous theoretical analysis of random square-triangle tilings [216] and findings of dodecagonal quasicrystals in recent experiments [209–212] and simulations [185, 213, 214].

Further evidence for the quasiperiodicity of the assembled structure is obtained by analyzing the tiling obtained in a system of 13,824 particles. There are three types of tiles in a quasicrystal, most tiles are either squares S or triangles T, but a few rhombi R are also present in the structure. A perfect– mathematical– quasicrystal is self-similar i.e. the same tiling is obtained in different length scales; a tiling should in principle be mappable to a tiling at a larger length scale by inflation rules. This constrains the concentration of different types of tiles in a mathematically perfect quasicrystal. For the dodecagonal quasicrystal for instance, the ideal ratio is $\frac{n_T}{n_S + n_R/2} = \frac{4}{\sqrt{3}} \approx 2.31$. Fig. 5.12 depicts the full tiling for the quasicrystal forming in the 13,824-particle system with the $\frac{n_T}{n_S + n_R/2} = 2.33$, which is very close to the ideal ration in the perfect quasicrystal.

This inflation symmetry can be viewed more vividly in the quasicrystals formed at bigger systems. Fig. 5.13 shows the transparent top view of the quasicrystal formed in a system of $N = 28^3 = 21,952$ particles. Bigger rings R_2 are related to smaller rings R_1 by inflation symmetry. The ratio between the sizes of two rings is $\frac{r_2}{r_1} = \frac{\sqrt{3}+1}{\sqrt{2}}$. Note that R_1 corresponds to twelvefold rings shown in Fig. 5.10b.

Quasicrystal approximants are periodic crystals with local tiling structures identical to that in the quasicrystal [215]. Because they are closely related, and are often observed in experiments, we consider them as candidates for dense packings. The dodecagonal approximant with the smallest unit cell (space group $P\bar{4}n2$) has 82 tetrahedra (Fig. 5.14b) and corresponds to one of the Archimedean tilings [217]. At each vertex we see the logs of twelvefold rings (shown in red) capped by single pentagonal
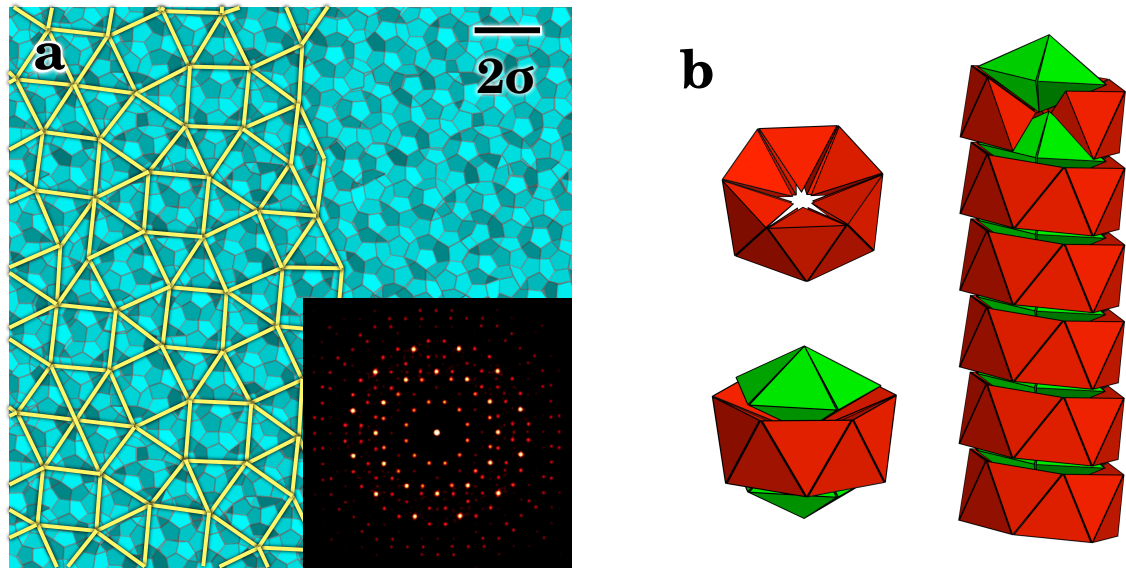
Figure 5.11:

Structural details of the quasicrystal. (a)Network of connected tetrahedra in a slice through a quasicrystal with $8,000$ tetrahedra, viewed along the direction of the twelvefold axis. Lines connecting the centre of mass of nearest neighbor tetrahedra form turquoise pentagons, which correspond to the pentagonal dipyramid network. The tiling structure is highlighted in yellow. A diffraction pattern indicating twelvefold symmetry is shown in the inset. (b) The vertices of the tiling are formed by logs comprised of rings of twelve tetrahedra, with neighboring rings enclosing a pentagonal dipyramid. The packing fraction within the logs can be increased by a tilt of the rings with respect to the log axis. This allows neighboring pentagonal dipyramids to avoid each other, as indicated in the figure, where two tetrahedra have been removed from the top ring to expose pentagonal dipyramids. (Images reproduced from [18])
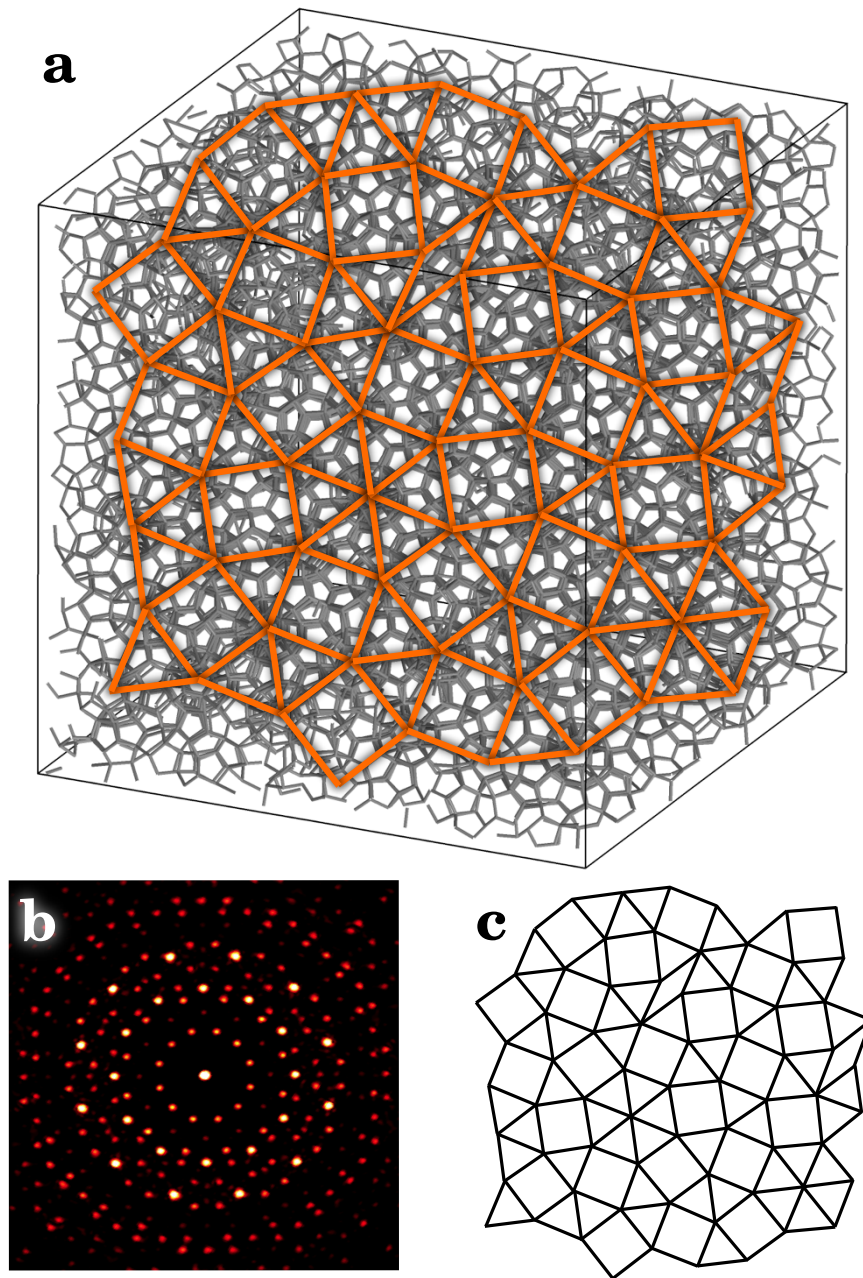
Figure 5.12:
Analysis of the tiling substructure in the quasicrystal with $N = 24^3 = 13,824$ particles shown in Fig. 5.10. (a) The gray lines connect tetrahedra that are nearest neighbors. Pentagons in the neighbor network correspond to PDs. Centers of logs are connected with orange lines. (b) The diffraction pattern shows twelvefold symmetry. (c) The square-triangle tiling contains $n_S = 23$ squares, $n_T = 56$ triangles and $n_R = 2$ thin rhombi. Their ratio $n_T/(n_S + n_R/2) \approx 2.33$ is close to the ideal value for a mathematically perfect quasicrystal, $4/\sqrt{3} \approx 2.31$. For the first-order approximant the ratio equals 2. (Images reproduced from [18])
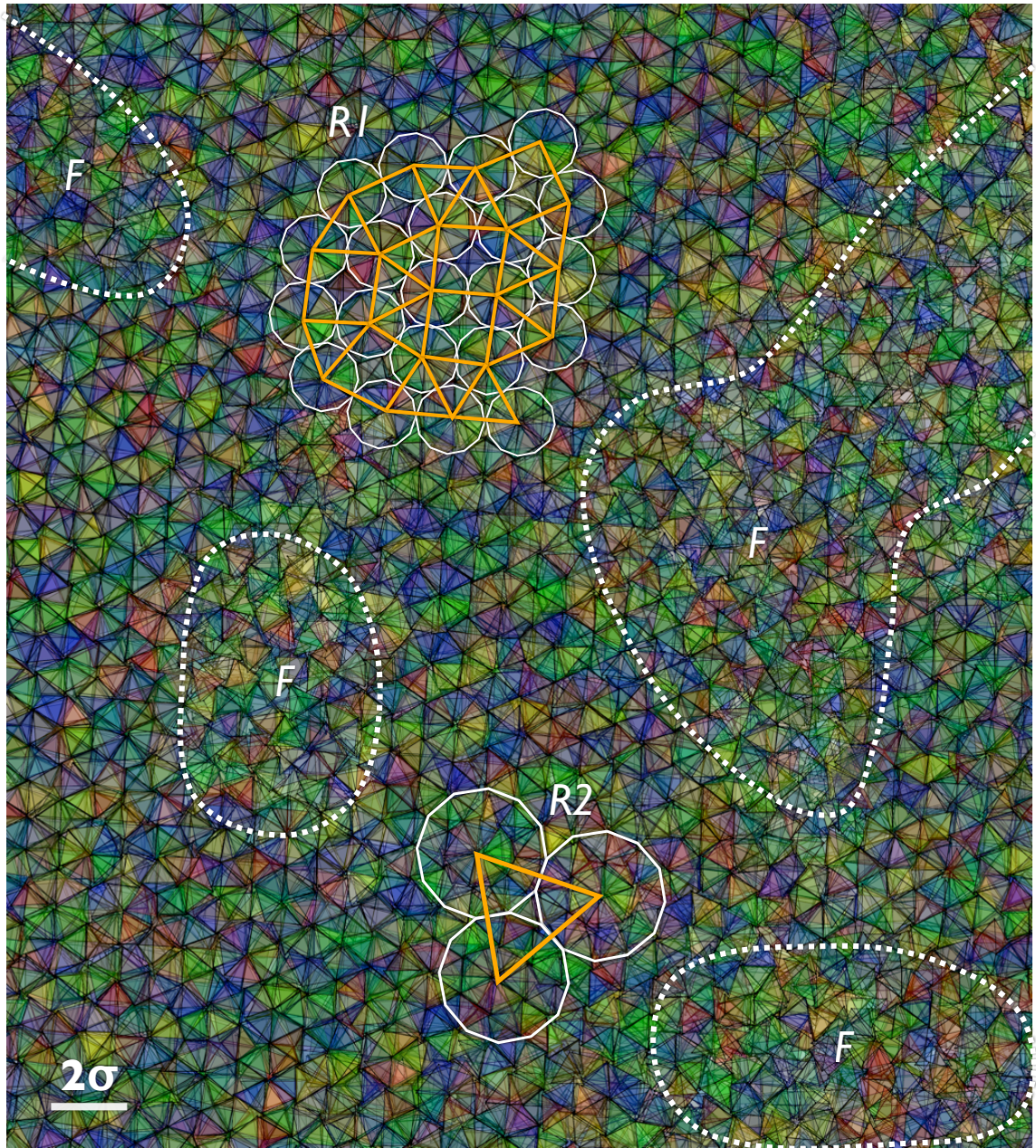
Figure 5.13: **Inflation symmetry of the quasicrystal**. Translucent view showing a cut through a system with $N = 28^3 = 21{,}952$ tetrahedra. The viewing direction is along the dodecagonal axis. Parts of the sample are still in the fluid (F) state. Tetrahedra arrange into small rings $(R1)$ and large rings $(R2)$. Their diameters have an irrational ratio of $r_2/r_1 = (\sqrt{3}+1)/\sqrt{2}$. (Images reproduced from [18])

dipyramids (green). The logs pack well into squares and triangles with additional, intermediary tetrahedra (blue). The vertex configuration of the tiling is $(3.4.3^2.4)$, as shown in Fig. 5.14a. Interpenetrating tetrahelices can also be seen in the approximant (Fig. 5.14c). Building and numerically compressing a unit cell of this ideal structure achieves a packing fraction of $\phi = 84.79\%$. If we compress a $2 \times 2 \times 2$ unit cell, the packing fraction marginally increases to $\phi = 85.03\%$ (Appendix C.2.6), the densest packing of tetrahedra at the time of its discovery (Fig. 5.14d-e). Compressing approximants with more complex unit cells, more faithful to an ideal quasicrystal, does not further improve the packing (Fig. 5.15).

Why should square-triangle tilings be preferred for dense packings of tetrahedra? First, we compare the packing fraction of the square tile with 22 tetrahedra (Fig. 5.16a) to that of the triangle tile with 9.5 tetrahedra (Fig. 5.16b). Their ratio $\frac{\phi_{\text{triangle}}}{\phi_{\text{square}}} = \frac{19}{11\sqrt{3}} \approx 0.9972$ is nearly unity, which suggests that tetrahedra pack equally well in both tiles. Second, we note that rings comprising the logs are tilted (Fig. 5.11b) and the layers of the structure are corrugated (Fig. 5.10a). This is a direct consequence of the face-to-face packing of tetrahedra where neighboring logs kiss. As a result, the square tile has a negative Gaussian curvature whereas the triangle tile has a positive one. Alternating the two tiles produces a net zero curvature in the layers, as observed in the quasicrystal and its approximant.

As shown in Fig. 5.17, the local structures of the $(3.4.3^2.4)$ approximant, the dodecagonal quasicrystal and the disordered glass (as characterized by their radial distribution functions) are very similar. The peak positions are identical: only the peak heights differ. This implies that the local structure of the glass and quasicrystal are only subtly different, and more sensitive measures of local order, as in Fig. 5.5b, are required. The quasicrystal and the approximant can be also be distinguished by visual inspection of the tiling, as well as comparison of diffraction patters (Fig. 5.18). The crucial step during crystallization is the transformation of the percolating pentagonal dipyramid network into layers, and the elimination of icosahedra. This intriguing process will be investigated in subsequent studies.

5.5   The Effect of Polydispersity[6]

In Section 2.2, we discussed the predictive power of hard particle simulations in forecasting the phase behavior of experimental systems of nano- and colloidal particles. This was indeed one of our main motivations of studying the self-assembly of

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[6]This section will be part of a manuscript that is currently under preparation.
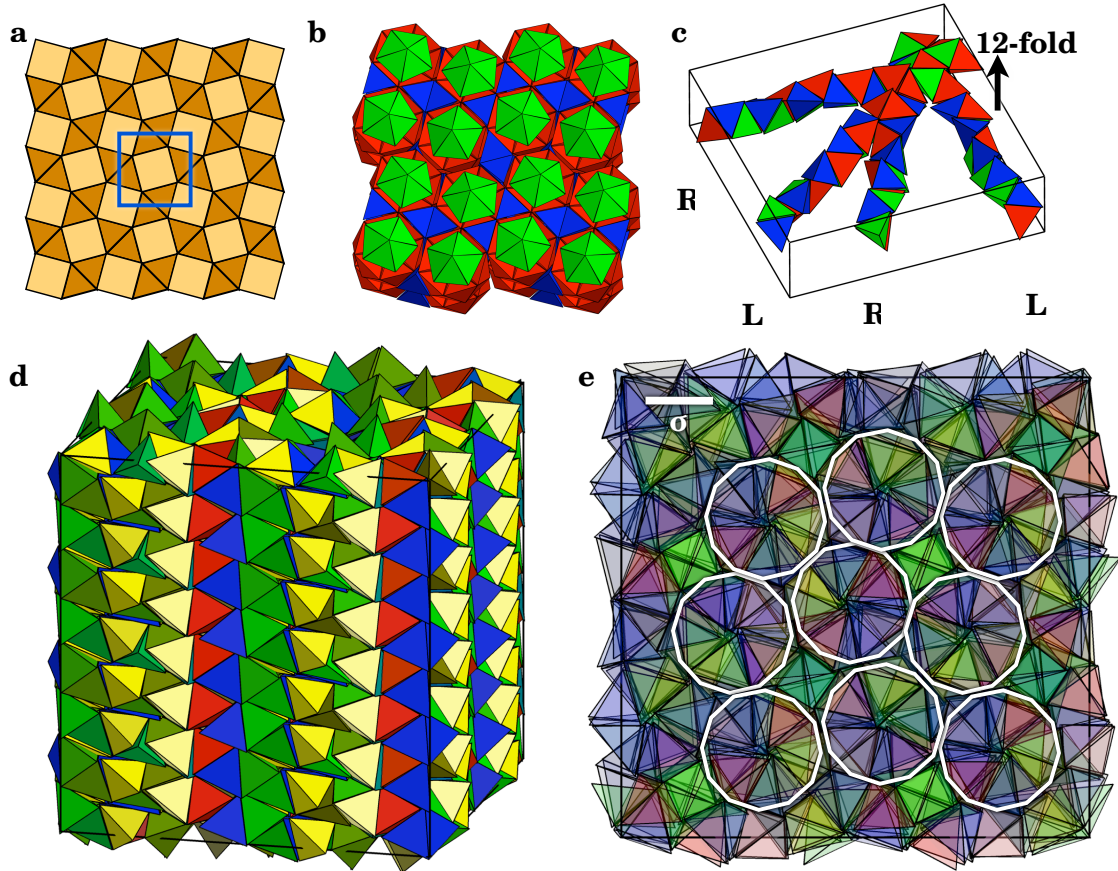
Figure 5.14:

The $(3.4.3^2.4)$ **approximant**. The tiling (a) and the structure (b) of the $(3.4.3^2.4)$ approximant. Colors are described in the text. (c) Similar to the quasicrystal, interpenetrating tetrahelices are present throughout the approximant. Their chirality alternates between left (L) and right (R) by 30° rotations. (d-e) Details of the high-density $\phi = 85.03\%$ packing of hard tetrahedra. This density was obtained by compressing a $2 \times 2 \times 2$ cell of the $(3.4.3^2.4)$ approximant with 656 tetrahedra. (d) For ease of viewing, the $2 \times 2 \times 2$ cell has been periodically continued into a $2 \times 2 \times 8$ cell. (e) In the translucent image, the twelvefold logs can be identified. (Images reproduced from [18])
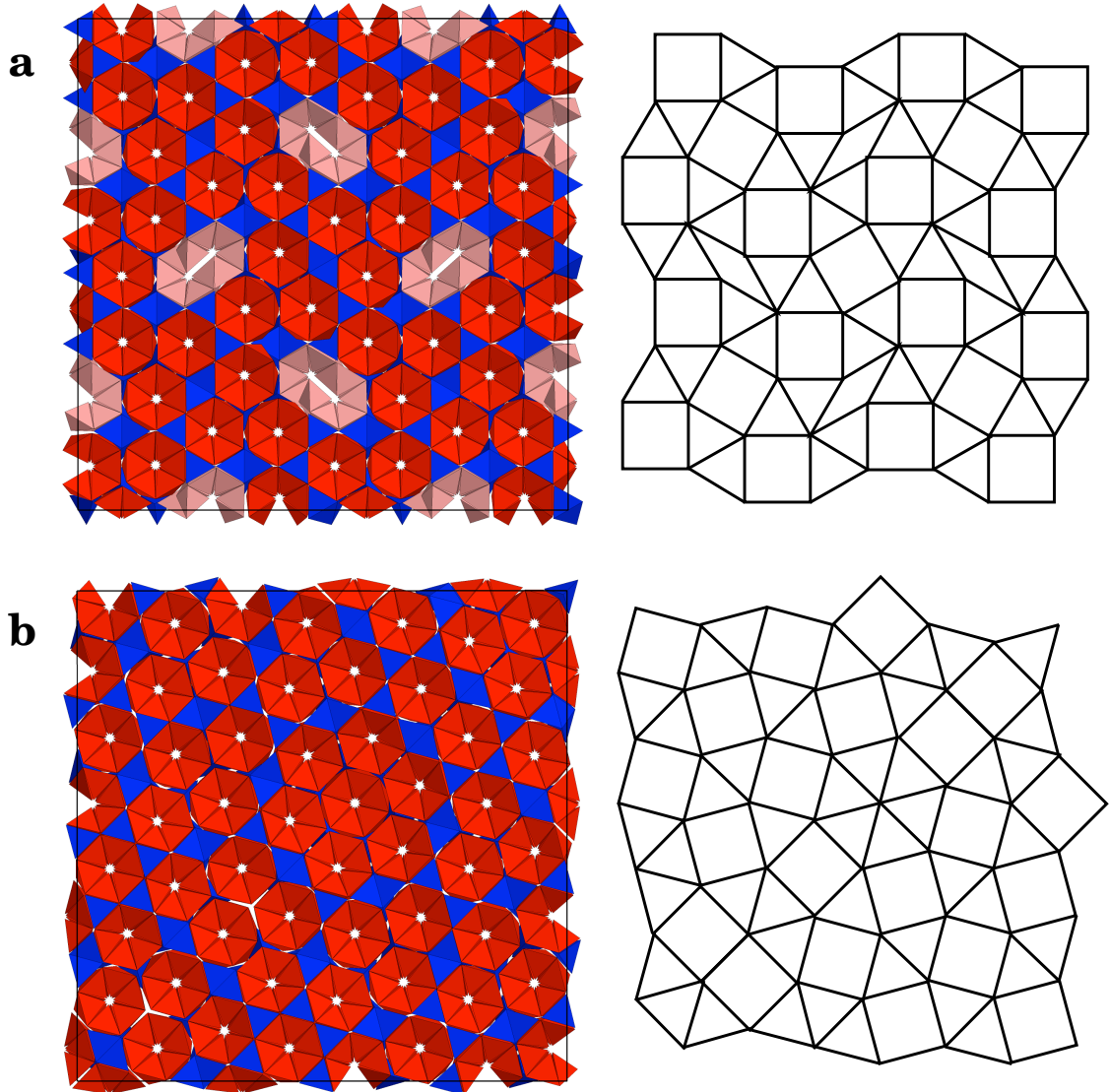
Figure 5.15: **Two higher approximants of the dodecagonal quasicrystal obtained via geometric construction**. Arrangements of tetrahedra (left) and tilings (right) are shown. PDs capping the logs are not drawn for ease of viewing. (a) The second approximant has a body-centered tetragonal unit cell with 306 tetrahedra. Four unit cells form an orthorhombic box. (b) The third approximant has a primitive tetragonal unit cell with 1,142 tetrahedra. As expected, the ratio of the number of tetrahedra in successive approximants converges to the self-similarity scaling factor of the square-triangle tiling, $\sqrt{3} + 2 \approx 3.732051 : \frac{306}{82} \approx 3.731707$ (2nd vs. 1st), $\frac{1142}{306} \approx 3.732026$ (3rd vs. 2nd). The approximants compress to $\phi = 82.84\%$ (a) and $\phi = 83.52\%$ (b). (Images reproduced from [18])

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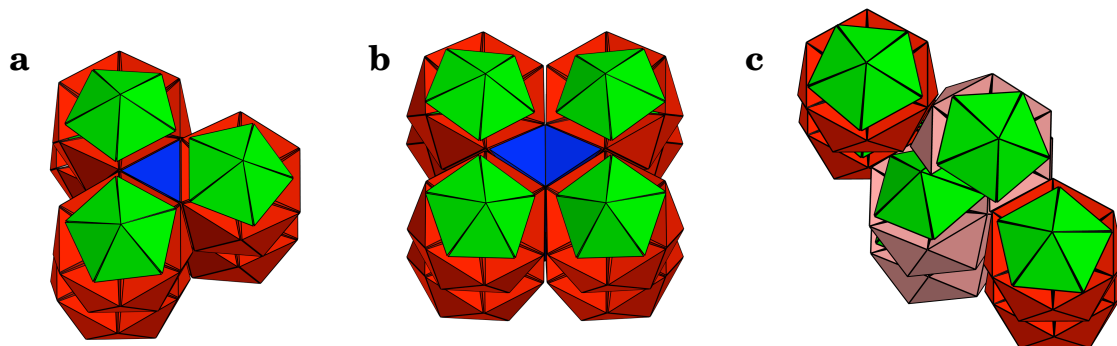
Figure 5.16:

Decoration of individual tiles in the quasicrystal and the approximants: (a) a triangle with 9.5 tetrahedra, (b) a square with 22 tetrahedra. The ratio of the packing densities of the tiles is $\phi_{\text{triangle}}/\phi_{\text{square}} = 19/11\sqrt{3} \approx 0.9972$. (c) Thin rhombi are frequently observed in connection with zipper motion, a dynamical mechanism to rearrange squares and triangles [216]. A rhomb is a structural defect. It consists of 11 tetrahedra and has half the volume of a square. As can be seen in the image, tetrahedra in the middle form a spiral of two interpenetrating logs (light red). (Images reproduced from [18])
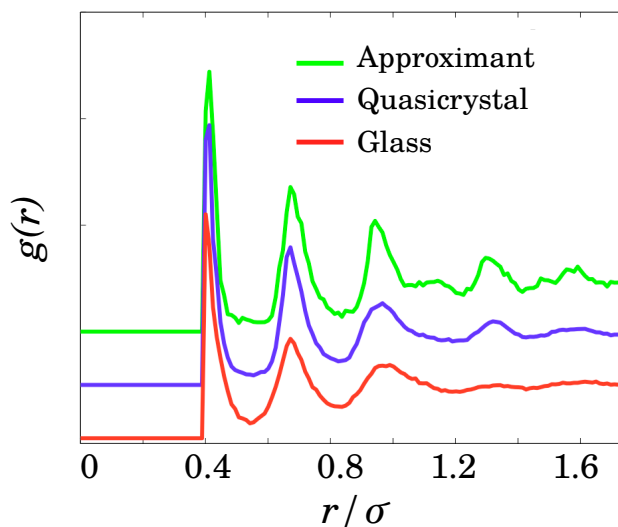
Figure 5.17:

Radial distribution functions for the approximant ($N = 82$), quasicrystal ($N = 8{,}000$), and glass ($N = 8{,}000$). Curves are vertically offset for clarity. (Images reproduced from [18])

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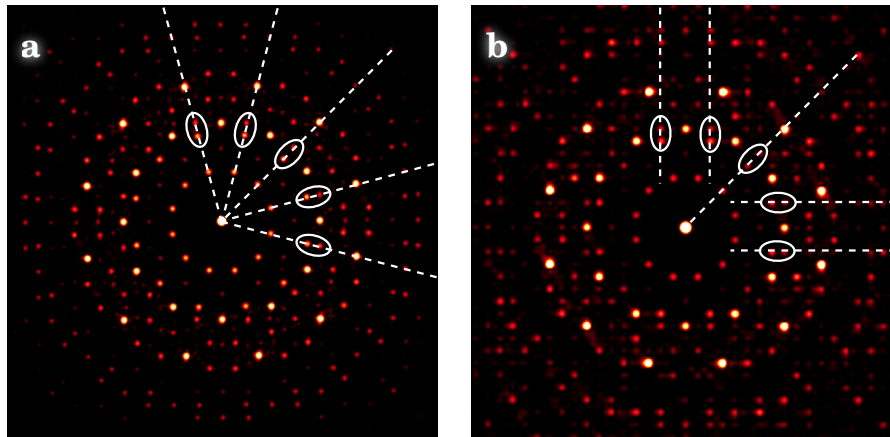
Figure 5.18:

Comparison of the diffraction patterns of the compressed quasicrystal (Fig. 5.10) and the compressed $(3.4.3^2.4)$ **approximant (Fig. 5.14d-e).** While the Bragg peaks have perfect twelvefold symmetry in the dodecagonal quasicrystal (a), the symmetry is broken to fourfold symmetry in the approximant (b). As indicated by white dashed lines and ellipsoids, weak Bragg peaks of the approximant are shifted slightly from their positions in the quasicrystal. Such behavior can be understood within the theory of quasicrystals [215]. (Images reproduced from [18])

hard tetrahedra, to be able to predict the types of phases that could emerge in systems of colloidal and nanotetrahedra. It should however be noted that real colloidal particles are not perfect, and the fact that the quasicrystal forms in a system of perfect hard tetrahedra does not necessarily imply that it will also form in real colloidal systems where the particles are polydispersed. It is therefore of great practical relevance to investigate the robustness of the dodecagonal quasicrystal to polydispersity of the building blocks.

In order to do this, we carry out simulations of polydisperse tetrahedra at pressures/densities that perfect tetrahedra assemble into a quasicrystal. However, the formation of the quasicrystal is a very slow process, and can take up to 3×10^7 MC cycles even for perfect tetrahedra. In order to accelerate this, we focus our attention on systems of $N = 328$ particles, which is the magic number for forming four unit cells of the $(3.4.3^2.4)$ approximant in a cubic box. In the case of monodisperse tetrahedra, a system of 328 particles crystallizes an order of magnitude faster i.e. in $2 - 5 \times 10^6$ MC cycles at 50% packing fraction. It is therefore reasonable to assume that the approximant will form in much shorter times in polydisperse systems of $N = 328$ particles, if the system has the propensity to form the quasicrystal at all.

In order to study the effect of size polydispersity, we first generate a dilute random configuration of perfect tetrahedra, we then set the edge length of each particle to a number randomly selected from the distribution $\sqrt{8}\mathcal{N}(1, \nu^2)$ with ν being the percentage of polydispersity. We then run isobaric simulations of the resulting configuration at $P^* = 64$, a pressure at which the approximant is routinely observed to form in systems of perfect tetrahedra. We run simulations for polydispersities in the range $\nu = 1 - 19\%$, for each value of ν, we run ten simulations, each randomly generated using the procedure described above. Fig. 5.19 shows the diffraction images calculated for the densest packing obtained for each value of polydispersity. Diffraction patterns are of superb quality for polydispersities as high as 8%, and of acceptable quality for polydispersities as high as 12%. One can however observe traces of long-range order for polydispersities as high as 15%. This is good news from an experimental perspective, since the reported polydispersity of colloidal and nanotetrahedra are in the range of 10% [66], which is less than the 12% threshold found in this study. This keeps the hope of obtaining a dodecagonal quasicrystal of colloidal tetrahedra alive, and well within rich in current experimental systems. We also consider calculate the maximum density of the densest packing observed for polydispersities below 15% and observe that $\phi_{\max}(\nu)$ is monotonically decreasing function of ν (Table 5.2).

It is also noteworthy that the formation of the quasicrystal in a system of poly-

0%   1%   2%   3%   4%

5%   6%   7%   8%   9%

10%   11%   12%   13%   14%

15%   16%   17%   18%   19%
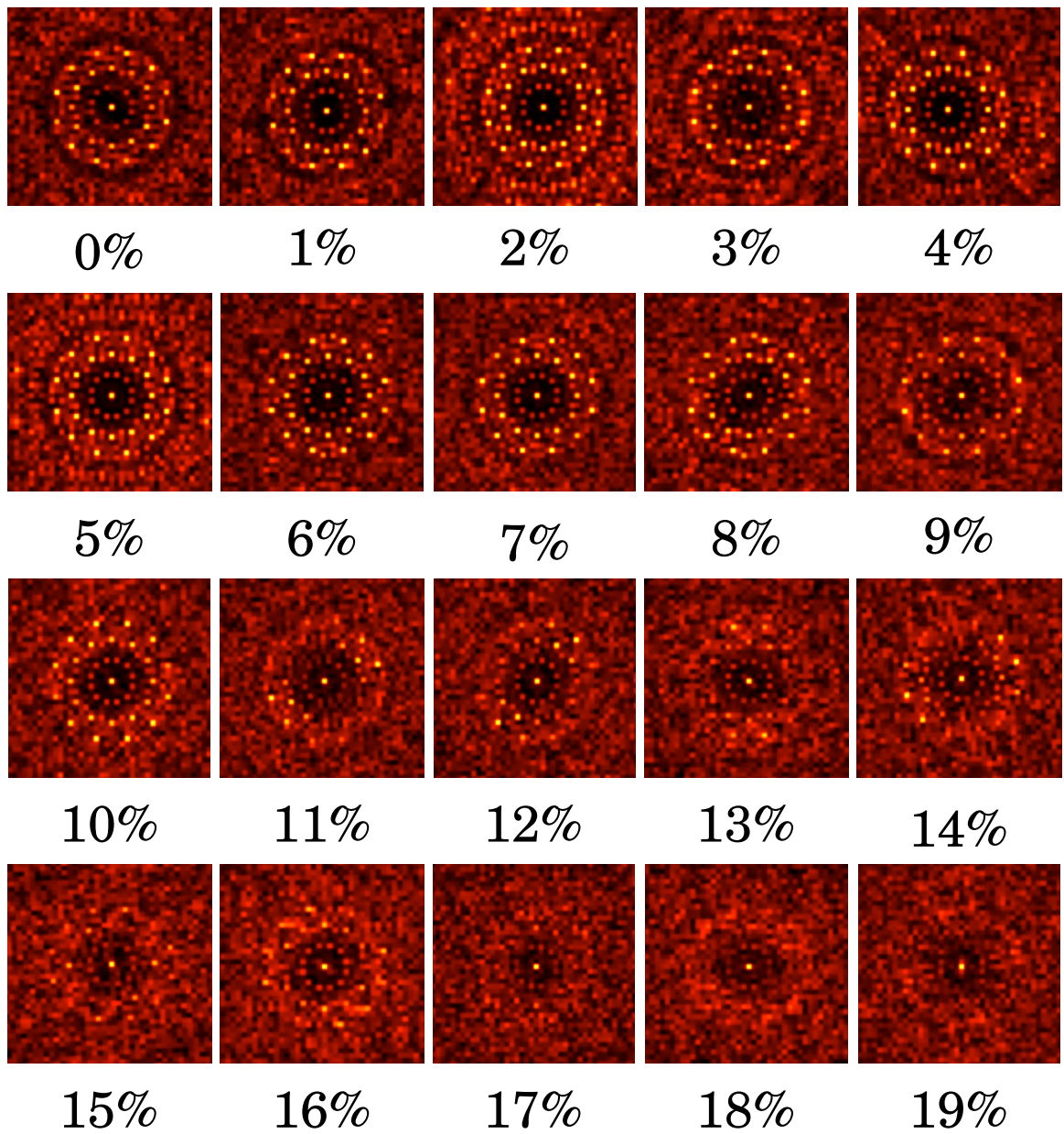
Figure 5.19: **Diffraction images calculated for the polydisperse systems.** Each system is simulated for 10^8 MC cycles at $P^* = 64$, and then quickly compressed to its maximum packing fraction.

Table 5.2: Maximum densities of the approximant for different polydispersities

Polydispersity (%)	Maximum Packing Fraction
1	0.84697582
2	0.84544326
3	0.84339035
4	0.83890998
5	0.83553847
6	0.83003463
7	0.82358400
8	0.81634127
9	0.80990100
10	0.80330100
11	0.79151252
12	0.78462746
13	0.77818900
14	0.77407546

disperse tetrahedra is more robust than the formation of the fcc crystal in a system of polydisperse hard spheres, where the solid phase cannot exist for polydispersities beyond 5.7% [218]. This might be explained by the fact that the quasicrystal is more similar to the hard tetrahedron fluid than is the fcc to the hard sphere fluid.

5.6 Conclusions

In conclusion, we report the spontaneous formation of a dodecagonal quasicrystal by hard tetrahedra, which is the first quasicrystal to be formed by hard particles, as well as the first quasicrystal formed by nonspherical building blocks. The quasicrystal and its approximant can pack very efficiently, and the $(3.4.3^2.4)$ approximant was the world record at the time of its discovery. This study clearly shows that shape alone can produce remarkable structural complexity through solely entropic interactions. We also show that the quasicrystal formation is very robust and occurs for size polydispersities as high as 12 per cent. This last finding is important since it encompassed the ranges of polydispersities reported for the synthesis of colloidal and nanotetrahedra [66].

CHAPTER VI

Hard Tetrahedra: Phase Diagram[1]

Shortly after our reporting of the quasicrystalline phase and the then-record maximum packing density of 85.03% [18], Kallus *et al.* introduced a simple periodic structure with $\phi = \dfrac{100}{117} \approx 85.47\%$ [30]. The unit cell is monoclinic and has four tetrahedra grouped into pairs of triangular bipyramids (TBPs). This structure was slightly optimized by Torquato and Jiao to $\phi = \dfrac{12250}{14319} \approx 85.55\%$ by breaking its monoclinic symmetry [219]. A few days after Toquato and jiao, Chen *et al.* found the real local optimum for the triclinic unit cell (Fig. 6.1), a structure with a packing density $\phi = \frac{4000}{4671} \approx 85.63\%$ [31, 220]. They also isolated the same structure in NPT simulations of small systems with 16 or fewer particles. Throughout this chapter, we will call the densest known packing of tetrahedra the *dimer crystal*.

Since the dimer phase is denser than the $(3.4.3^2.4)$ approximant, it should become thermodynamically stable in the limit of infinite pressure, however it is never observed to form in our self-assembly simulations. In this chapter, we carry out a detailed investigation of the phase behavior of hard tetrahedra from the fluid up to the densest packing. In contrast to previously studied systems of hard particles, the phase diagram of tetrahedra entails a non-symmetry-breaking solid-solid transition. We confirm its existence by Monte Carlo simulation and free energy calculations and discuss the origin of the transition. The present study complements previous works on hard tetrahedra which studied some aspects of the equation of state [18, 114, 196] as well as dense packings [30, 31], and extends those to provide a complete picture of the phase diagram. By comparing the results of self-assembly simulations to those obtained from free energy calculations, we assess the likelihood of various candidate phases to be observed both in simulations and in experiments of hard tetrahedra.

The chapter is organized as follows. A detailed account of performed simulations is given in Section 6.1. In Section 6.2, the thermodynamics of the dimer phase is reported. The thermodynamics of the quasicrystal and its approximant follows in Section 6.3. The results of free energy calculations are presented in Section 6.4. A computer experiment in which the dimer crystal spontaneously transforms into the quasicrystal at $\phi = 50\%$ is reported in Section 6.5. The origin of the stability of the approximant over the dimer crystal at experimentally realizable densities is discussed in Section 6.5.1 and discussions and concluding remarks are provided in Section 6.6.

6.1   Technical Specifications of Performed Simulations

Equations of state, $\phi(P^*)$, are calculated with isobaric Monte Carlo simulations. (Further details of Monte Carlo simulations can be found in Section 3.2.1.) Changes in the Gibbs free energy within a single phase are obtained via thermodynamic integration:

$$\frac{G_2 - G_1}{Nk_BT} = \frac{V_T}{\sigma^3} \int_{P_1^*}^{P_2^*} \frac{\mathrm{d}p}{\phi(p)}, \qquad (6.1)$$

where $V_T = \sigma^3\sqrt{2}/12$ is the volume of a tetrahedron.

Simulations are carried out in the pressure range $50 \leq P^* \leq 4000$ for the dimer crystal ($4\times6\times6\times6 = 864$ tetrahedra), quasicrystal (8,000 tetrahedra) assembled from the fluid and compressed to a packing density up to 83.36%, and the approximant ($82 \times 2 \times 2 \times 3 = 984$ tetrahedra).

We use a modified version of the Frenkel-Ladd method for free energy calculation. Implementation details are given in Section 3.3.4. We use a maximum string constant of $\gamma_{\max} = 4\times10^6$. The interval $[0, \gamma_{\max}]$ is discretized and the system is held for 2×10^5 MC cycles at each γ value. During this period, the value of $\langle U \rangle_\gamma$ is evaluated. Explicit free energy calculation is only done in the pressure range $250 \leq P^* \leq 600$ where no configurational rearrangements are observed. The Gibbs free energy of each phase is then extrapolated for higher/lower pressures from equation of state using Eq. (3.25).
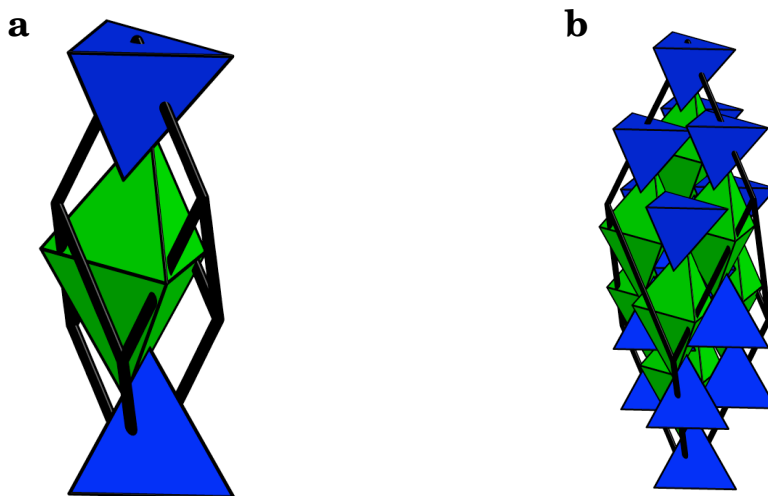
Figure 6.1:
The dimer crystal. The densest known tetrahedron packing [31] is a crystal with four particles per unit cell forming two triangular bipyramids (or 'dimers') shown in green and blue. The unit cell breaks the threefold symmetry of the dimers. (Images reproduced from [182])

6.2 Symmetrization of the Dimer Packing on Decreasing Pressure

We construct the dimer crystal analytically [31] and slowly expand it by reducing the pressure. The crystal remains stable during the simulation for pressures $P^* \geq 60$ while at lower pressures it melts abruptly. No hysteresis is observed in the equation of state (Fig. 6.2a), if the decompression is stopped before melting and the system is re-compressed. This suggests that the system remains at least in metastable equilibrium over this range of pressures and densities.

The compressibility $\kappa = (1/\phi)(\partial\phi/\partial P^*)$ (Fig. 6.2b) reveals a complicated phase behavior, with an anomalous peak indicative of a second-order phase transition appearing at around $P^* = 90$. We verify that this is a displacive phase transition; i.e. it only involves a lattice distortion and the particles in the lower density phase still remain in dimers. Analyzing the lengths of the vectors spanning the simulation box and the angles between them (Figs. 6.2c,d) indicates that the transition takes place in two stages. While in the lower density phase D_I ($P^* < 90$) all lengths and angles are equivalent, they are completely split only in the phase D_{III} ($P^* > 220$). There is also an intermediate phase D_{II} ($90 < P^* < 220$) in which only two of the lengths and angles are still degenerate. The symmetrization of the lattice therefore follows

the sequence: triclinic (D_{III}) → monoclinic (D_{II}) → rhombohedral (D_I).

It is known that the threefold symmetry of the dimers must be broken to achieve optimal bulk packing [30, 31], and we observe this in the sequence of transitions. We note that D_{II} was initially reported by Kallus *et al.* as a candidate for the densest packing of tetrahedra [30]. Its maximum packing density is only 0.2% lower than the maximum packing density of D_{III}, the structure predicted by Chen *et al.* [31]. Note also that the integrated area under the peak is a measure of the difference in packing densities. This explains the missing peak in the compressibility for the transition D_{III} → D_{II}. In contrast, the difference in maximum packing densities for the transition D_{II} → D_I is much larger, and of the order of a few percent.

6.3 Comparison of the Quasicrystal and its $(3.4.3^2.4)$ Approximant

The equations of state of the quasicrystal, the approximant, and the dimer crystal are presented in Fig. 6.3a. We observe that the approximant is not only denser than the quasicrystal at all pressures above the melting transition, it also melts at lower pressure. These observations together with Eq. (6.1) suggest that the quasicrystal is generally less stable than the approximant.

Further evidence for the stability of the approximant over the quasicrystal is obtained through constructing higher order approximants; i.e. approximants that have larger unit cells than the $(3.4.3^2.4)$ approximant, and comparing their equations of state with the quasicrystal and the $(3.4.3^2.4)$ approximant. Fig. 5.15 depicts two such approximants that we constructed using inflation operation [221]. The approximant depicted in Fig. 5.15a is not particularly dense due to the presence of rhombi in its tiling, and is therefore not considered here. The other approximant (Fig. 5.15b) however has no rhombi in its tiling and is relatively dense. We therefore calculate its equation of state in the transition region; we call the approximant with a 'rhomb-free' tiling the *second-order approximant* in the rest of this chapter.

As observed in Fig. 6.4, the second approximant is denser than the densest quasicrystal that formed in our simulations but less dense than the first approximant. Neither structure is expected to have a significant entropic advantage over others since tetrahedra experience similar local environments in all these structures. It is therefore safe to conclude that the first approximant is more stable than the quasicrystal and the second approximant because of its higher density. Higher-order approximants can be constructed similarly using inflation symmetry; however, such approximants will
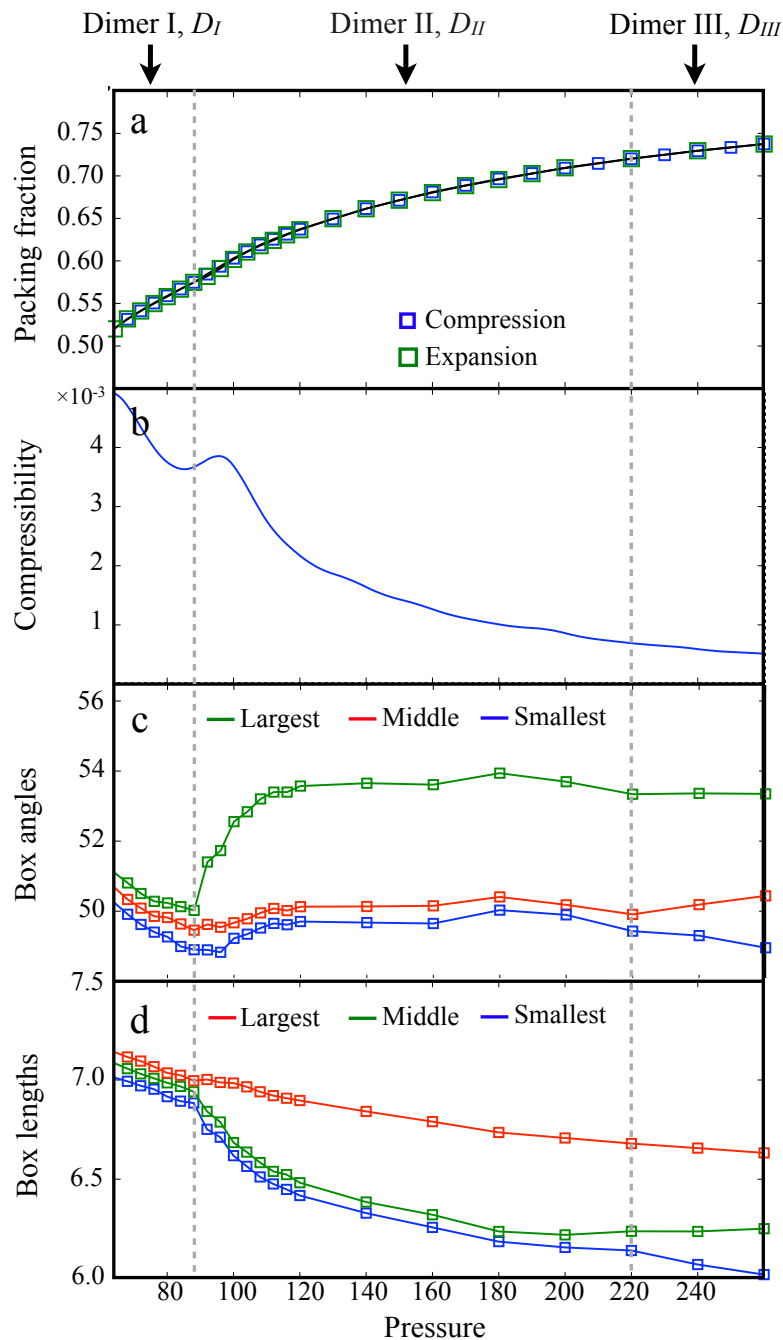
Figure 6.2: **Symmetrization of the dimer crystal.** (a) The equation of state shows no hysteresis between compression and expansion. (b) A peak is observed in the compressibility near pressure $P^* = 90$ at a second order displacive phase transition. The (c) three box angles and (d) box lengths, obtained by sorting the angles and lengths and then averaging the sorted values, are plotted as a function of pressure. We observe two transitions, from triclinic (D_{III}) to monoclinic (D_{II}) to rhombohedral (D_I). The phase D_{III} is thermodynamically stable, D_{II} and D_I are metastable. (Images reproduced from [182])
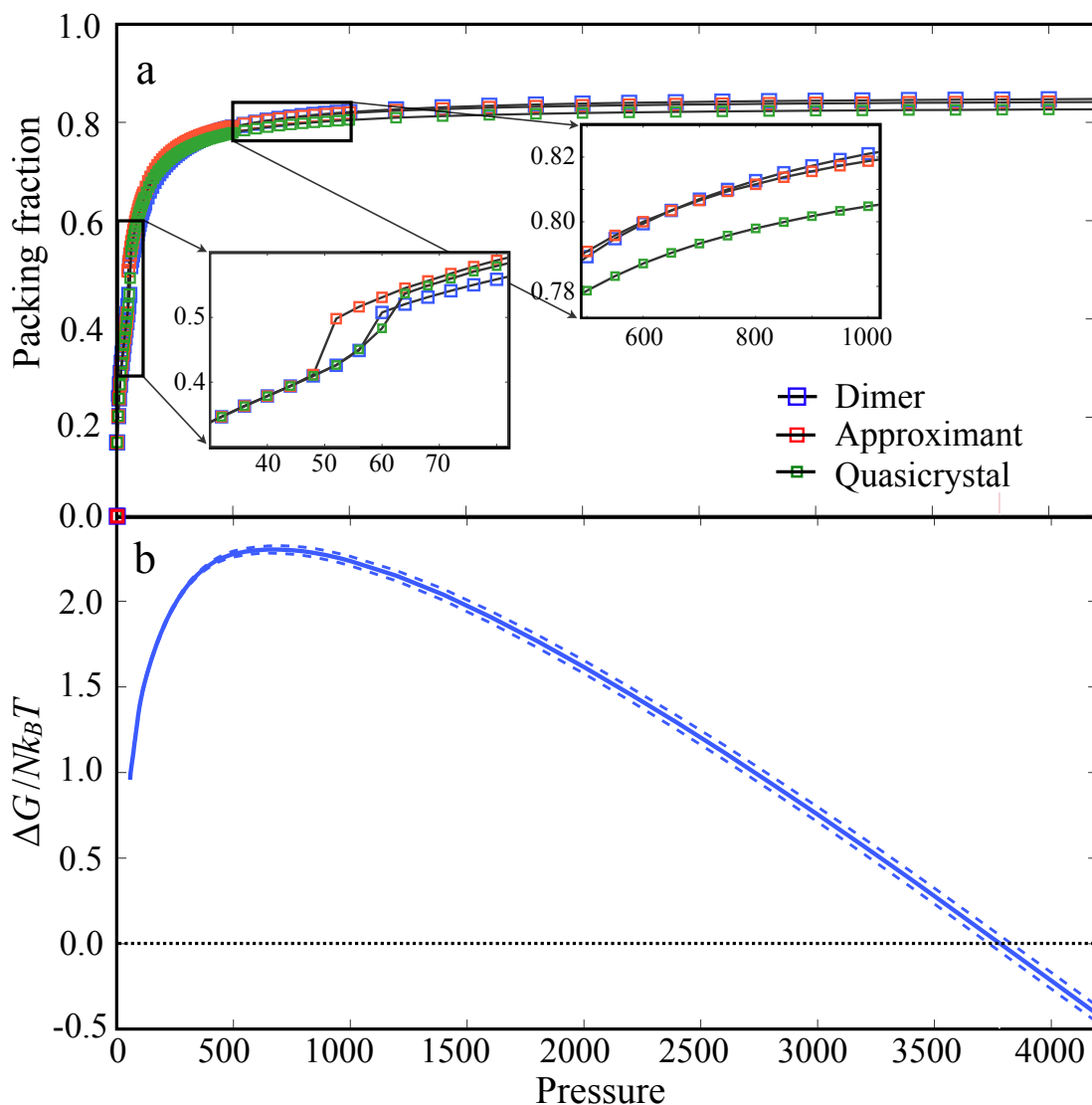
Figure 6.3: **Thermodynamic stability of the dimer crystal.** (a) The equation of state for the dimer crystal, the approximant, and the quasicrystal shows that the dimer crystal is the densest packing for $P^* > 700$. The approximant is always denser than the quasicrystal. Error bars are smaller than the size of the symbols. Insets show the equations of state in the melting region as well as near $P^* = 700$ where the dimer crystal first becomes denser than the approximant. (b) The Gibbs free energy difference between the dimer crystal and the approximant $\Delta G/Nk_BT = (G_D - G_A)/Nk_BT$ calculated using thermodynamic integration and the Frenkel-Ladd method. The dimer crystal is stable only at very high pressures.(Images reproduced from [182])
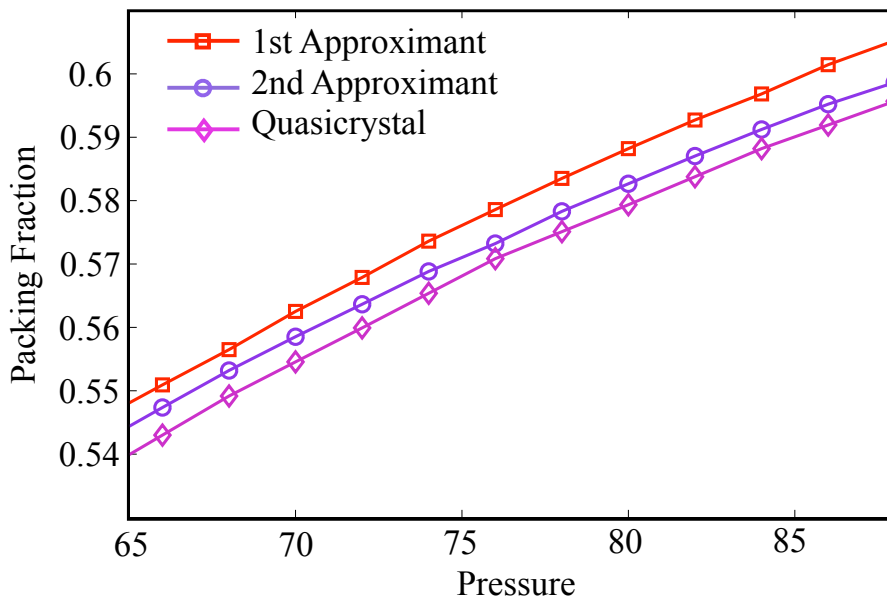
Figure 6.4: **Equations of state of the quasicrystal and the first and second approximants computed from NPT simulations.** The second approximant (with a unit cell of 1,142 particles) is less dense than the first approximant (with a 82-particle unit cell). (Images reproduced from [182])

have very large unit cells with tens of thousands of particles. Based on the observed trend, we expect higher order approximants to become successively less dense but still denser than the quasicrystal.

The question of comparing the relative thermodynamic stability of quasicrystals and their approximants plagues nearly all reports of new quasicrystals in the literature. The difficulty in obtaining perfect quasicrystals in experiments and simulations, along with the slow kinetics that would be involved in the transformation of even an imperfect quasicrystal to any of its approximants, confounds attempts to address quasicrystal stability. In this spirit, we remark that the quasicrystal configuration used in this study is obtained in simulation and an ideal, perfect quasicrystal might be slightly denser. The structure of such an ideal quasicrystal, however, is unknown. A denser quasicrystal would shift the curve in Fig. 6.3a slightly upwards, and hence make the quasicrystal thermodynamically more stable than the approximant in a narrow region close to melting. Based on all evidence however, we use the $(3.4.3^2.4)$ approximant as the most stable quasicrystal-like structure for free energy and free volume calculation purposes.

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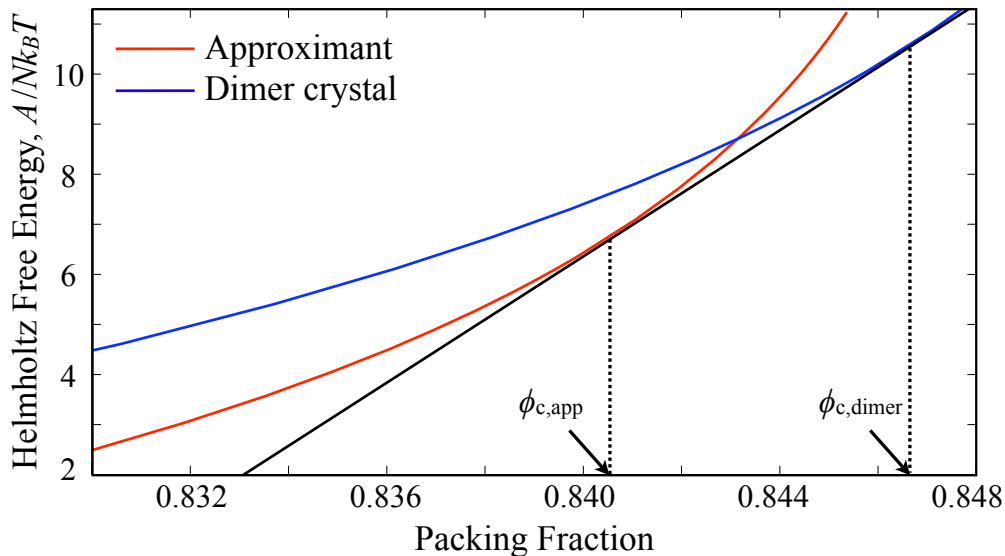
Figure 6.5:
The common tangent construction for the approximant-dimer transition. The critical packing fractions for the approximant to dimer transition can be calculated via the common tangent construction from the Helmholtz free energies of the approximant (red) and the dimer crystal (blue). (Images reproduced from [182])

6.4 Relative Thermodynamic Stability

The Gibbs free energy difference between the dimer and the approximant is calculated using the method described in Sec. 3.3.4. We find that the dimer crystal is stable only for pressures above $P_c^* = 3780 \pm 60$ (Fig. 6.3b), while the approximant is favored below P_c^*. At the critical pressure, the approximant and the dimer crystal have packing densities of $(84.0 \pm 0.1)\%$ and $(84.6 \pm 0.1)\%$ respectively. The transition densities can be alternatively calculated from the Helmholtz energy using the common tangent construction (Fig. 6.5). P_c^* is significantly higher than the melting pressure for the approximant, $P_M^* = 55 \pm 1$ (Fig. 6.6), which is determined using the approach described in Section 3.3.4.1.

It is noteworthy that the above calculations are based on the assumption that the dimer crystal of [31, 220] is the densest possible arrangement of hard tetrahedra. Although we cannot rule out the possibility that an even denser arrangement of tetrahedra that is different from the approximant and the dimer crystal might exist, our observation that the dimer crystal is the densest structure that forms in simulations of 16 tetrahedra and fewer [31, 220] substantiates this assumption. The quasicrystal
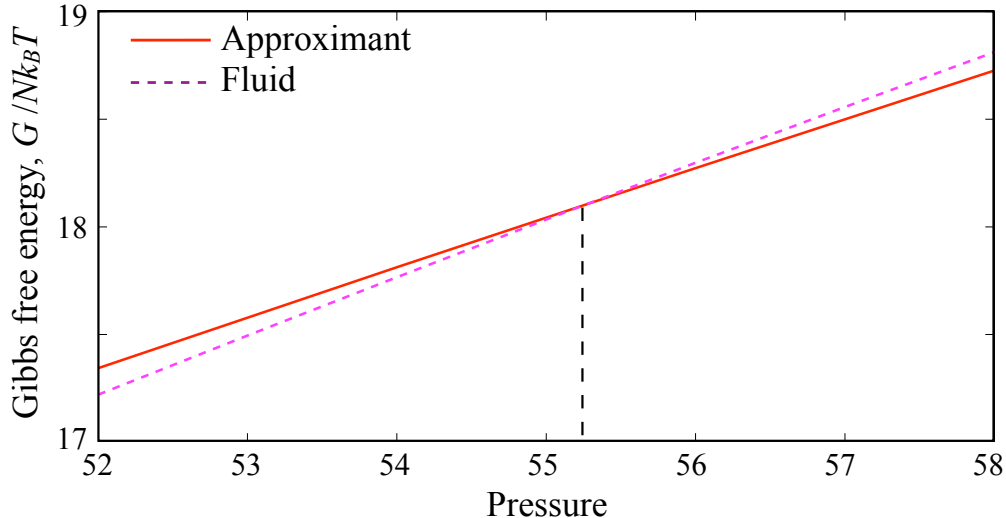
Figure 6.6:
Gibbs free energies of the approximant and the fluid close to the melting transition. The transition occurs at $P_M^* = 55$. (Images reproduced from [182])

that we are using for comparison with the approximant has been assembled in simulations from the disordered fluid and therefore contains imperfections. We cannot rule out that a perfect quasicrystal might be thermodynamically more stable than the approximant at all pressures. If this were the case, then the transition between the approximant and the dimer crystal reported above would be substituted by a transition between the quasicrystal and the dimer crystal in the phase diagram. Therefore, while such a discovery could alter certain details of the phase transition, it will not eliminate the existence of a solid-solid phase transition reported in this work.

6.5 Dimer-Quasicrystal Transformation

To compare the relative thermodynamic stability of the dimer crystal and the quasicrystal in simulation, we set up a Monte Carlo simulation of a large dimer crystal with 2,916($= 4 \times 9 \times 9 \times 9$) tetrahedra in the isochoric ensemble. To facilitate the transformation, the box dimensions are occasionally distorted in a random direction with the constraint that the total volume remains unchanged (variable-shape ensemble, [157]). This distortion allows the system to adjust to arbitrary lattice symmetries by relaxing shear stresses.

We choose a constant packing density of $\phi = 50\%$, because at this density the

quasicrystal is routinely observed to form spontaneously from the fluid. Structural changes are detected by counting the number of particles that form PDs and icosahedra using a shape-matching algorithm [191]; icosahdral motifs vanish when the quasicrystal forms [18]. Additionally, the pressure is determined from the acceptance probability of trial volume changes [116, 222].

The pressure shows a sharp spike after 4 million MC cycles accompanying the melting of the dimer crystal (Fig. 6.7a). The spike quickly decreases to a plateau, which, after $15 - 20$ million MC cycles, relaxes to its equilibrium value. PDs and icosahedra form as the preferred local configurations in the melt (Fig. 6.7b). On the other hand, in the final solid structure, most particles are members of PDs and virtually no icosahedra remain. Diffraction images in Figs. 6.7c-f show that the final solid structure is the dodecagonal quasicrystal. The fact that the quasicrystal forms in the simulation with the melt as an intermediate state confirms that both the quasicrystal and the melt are thermodynamically favored over the dimer crystal at the packing density $\phi = 50\%$.

6.5.1 Origin of Stability of the Approximant

To investigate the superior stability of the quasicrystal approximant compared to the dimer crystal over such a wide range of densities, we investigate the significance of collective particle motions by comparing the free energy estimates obtained from the mean-field approximation introduced in Section 3.4.3 with the exact free energy differences. We also analyze the dynamics in the approximant by calculating the van Hove correlation function [223] and visually inspecting the high-mobility particles [224] in our simulations.

6.5.1.1 Free volumes

We calculate the mean-log average of the free volumes $v_{f,\mathrm{ML}}$ of tetrahedra (Eq. 3.32) in the approximant, the dimer crystal, and the quasicrystal using the shooting method described in Section 3.4.1. The results are presented in Fig. 6.8a. Whereas particles in the quasicrystal generally have a smaller $v_{f,\mathrm{ML}}$ than in the approximant, the curves are shifted along the abscissa relative to one another by a fixed amount as indicated with arrows in Fig. 6.8a. This implies an identical thermodynamics for the quasicrystal and the approximant except for their different maximum packing densities. Indeed, tetrahedra experience similar local environments in the quasicrystal and its approximant.
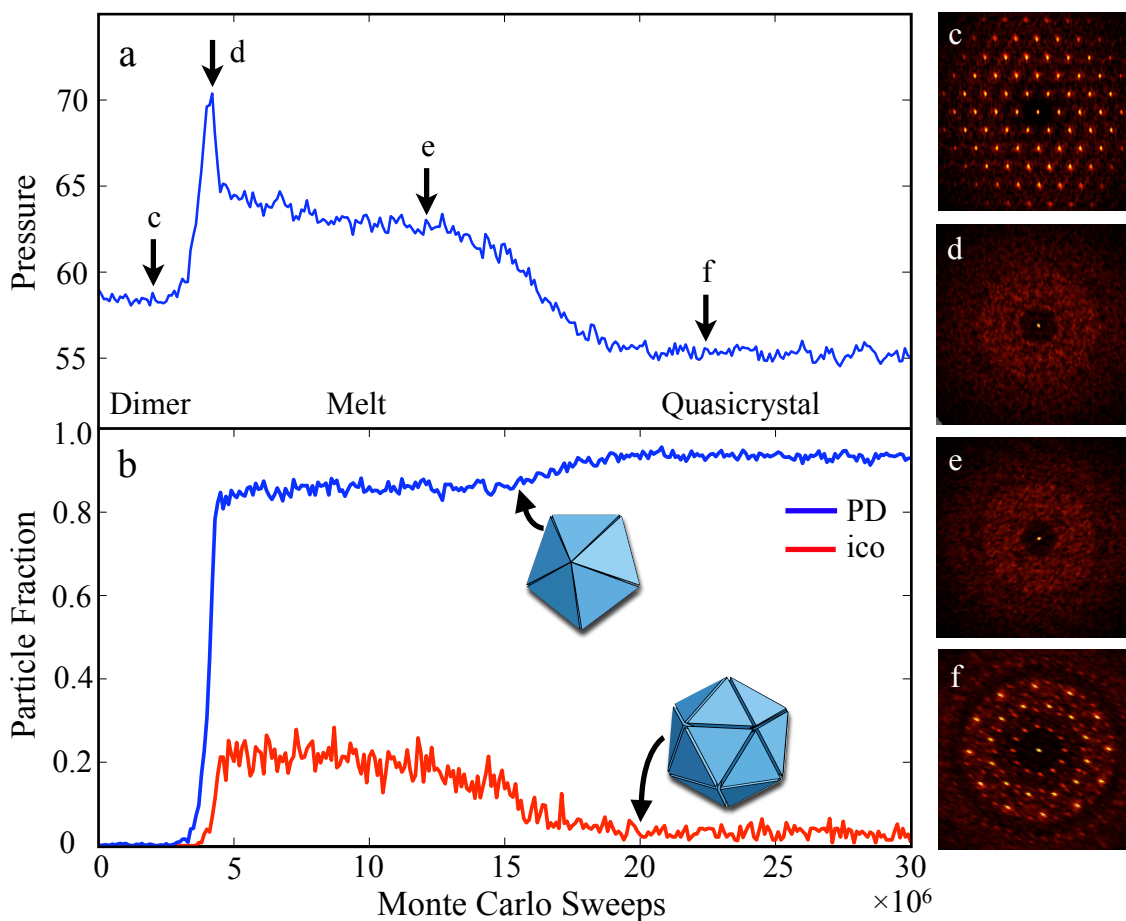
Figure 6.7: **Transformation of the dimer crystal to the dodecagonal quasicrystal in an isochoric simulation.** (a) The pressure first spikes after 4 million MC cycles and then relaxes during the melting of the dimer crystal. Between 15 and 20 million MC cycles, the quasicrystal forms from the melt. (b) The number of particles arranged in pentagonal dipyramids (PDs) or icosahedra (ico) increases rapidly during melting. In the quasicrystal essentially all particles form PDs while icosahedra disappear. Diffraction patterns confirm the transformation from the dimer crystal (c) to the melt (d,e) and then to the quasicrystal (f). (Images reproduced from [182])

In contrast, the mean log free volume of the dimer crystal decays much more slowly with packing density and intersects the two other free volume curves. This finding suggests that the approximant relaxes more efficiently during expansion, creating free volume for the particles more readily. Note that the packing density where the two curves cross is considerably below 84%, the density where the approximant becomes thermodynamically unstable, which underscores the significance of collective motions of particles in stabilizing the approximant even at very high densities.

The importance of collective motions may be further inferred by comparing the free energy difference estimated from a mean-field approximation with the exact value. As shown in Fig. 6.8b, the mean-field approximation underestimates the stability of the approximant, which indicates that entropic contributions from collective motions are significant. We suspect that slight rearrangements of particles in the approximant during expansion also increase its stability at lower packing densities. This is confirmed by estimating ΔG from a cell model approximation. The cell model is similar to the mean-field approximation except that free volumes are calculated for a non-equilibrated structure obtained by isotropically expanding the densest packing to a given packing density [188]. As shown in Fig. 6.8b, the cell-model approximation underestimates the stability of the approximant even more than the mean-field approximation, which suggests the significance of small local rearrangements that occur while the structure is equilibrated after expansion.

6.5.1.2   Dynamics in the Approximant

Correlated motions of tetrahedra are observed in long simulations of both the approximant and the quasicrystal at all densities. These motions are most apparent at packing densities below 65% where they give rise to local structural rearrangements, but they are still present at higher densities in the form of correlated vibrations of clusters of tetrahedra. The fundamental mechanism through which these rearrangements proceed is the rotation of single PDs around their principal axes by multiples of 72°. The particular lower asphericity of PDs allows an easy rotation even in relatively dense configurations.

van Hove correlation functions are powerful computational tools for characterizing such dynamical heterogeneities. The full van Hove correlation function is given by [223]:

$$G(\mathbf{r}, t) = \frac{1}{N} \left\langle \sum_{i=1}^{N} \sum_{j=1}^{N} \delta\left[\mathbf{r} + \mathbf{r}_j(t) - \mathbf{r}_i(0)\right] \right\rangle \tag{6.2}$$
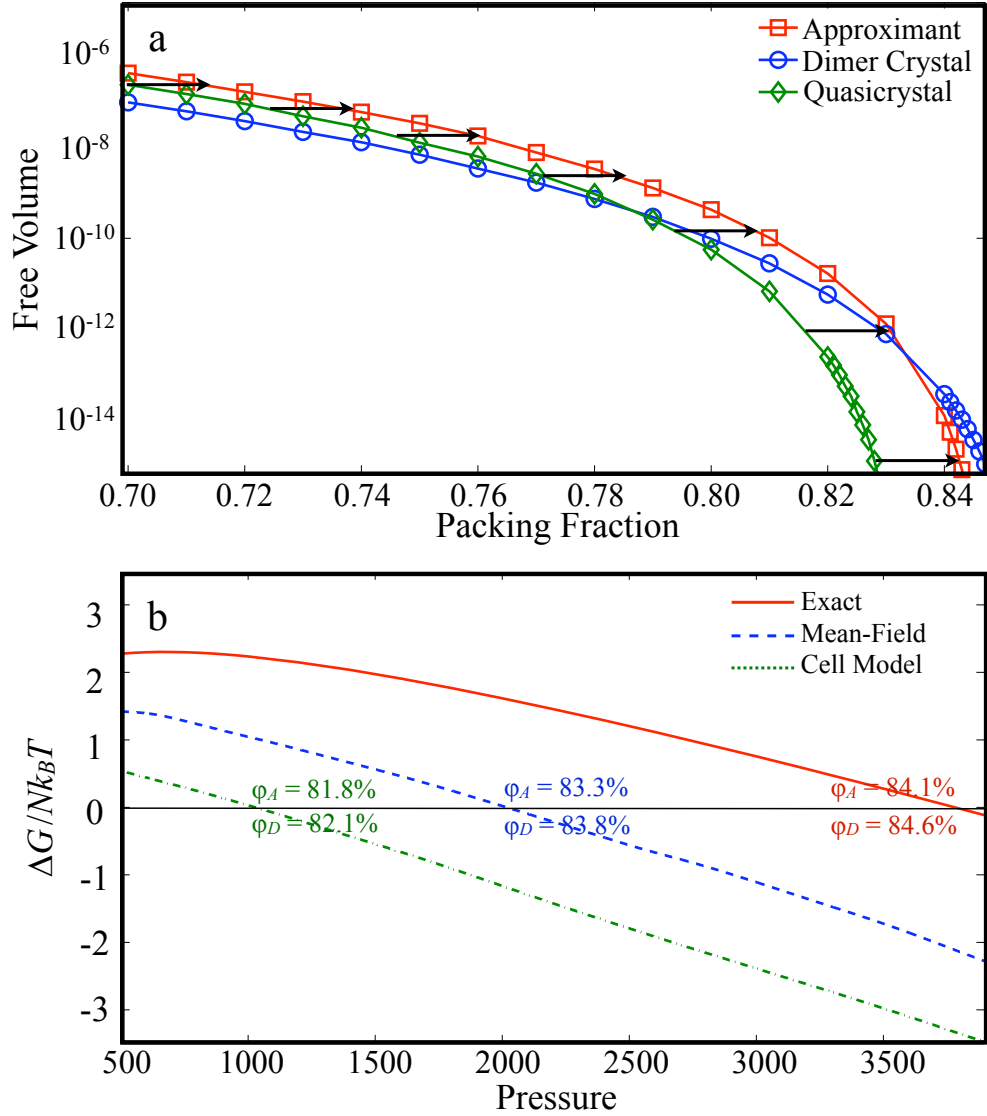
Figure 6.8: **Relative stability of the dimer crystal, quasicrystal, and quasicrystal approximant.** (a) Up to packing density 83% the dimer crystal has lower average free volume per particle. This helps to stabilize the approximant entropically. At high packing densities the dimer crystal should eventually have the highest average free volume, because its maximally achievable density is the highest of the three candidate structures. (b) Comparison of the Gibbs free energy differences between the dimer crystal and the approximant using the exact Frenkel-Ladd method, the mean-field approximation, and the cell-model approximations. The transition is predicted with all three methods even though the critical densities ϕ_A (approximant) and ϕ_D (dimer crystal) vary slightly. (Images reproduced from [182])

with N the number of particles in the system. $G(\mathbf{r}, t)$ can be decomposed into the *self part* $G_s(\mathbf{r}, t)$ and the *different part* $G_d(\mathbf{r}, t)$:

$$G_s(\mathbf{r}, t) = \frac{1}{N} \left\langle \sum_{i=1}^{N} \delta \left[\mathbf{r} + \mathbf{r}_i(t) - \mathbf{r}_i(0) \right] \right\rangle \tag{6.3}$$

$$G_d(\mathbf{r}, t) = \frac{1}{N} \left\langle \sum_{i=1}^{N} \sum_{j=1, j \neq i}^{N} \delta \left[\mathbf{r} + \mathbf{r}_j(t) - \mathbf{r}_i(0) \right] \right\rangle \tag{6.4}$$

The function $G_d(\mathbf{r}, t)$ measures any spatiotemporal correlations between *distinct* particles, and for $t = 0$, it simply is proportional to the pair distribution function i.e. $G_d(\mathbf{r}, 0) = \rho g(\mathbf{r})$. $G_s(\mathbf{r}, t)$ however measures the extent to which a single particle is correlated to its own original position at a given time i.e. $G_s(\mathbf{r}, t) d^3 \mathbf{r}$ is the infinitesimal probability that an arbitrary particle has moved by a vector falling into a $\Delta \mathbf{r}$-sided box centered at $\mathbf{r}$. It is of particular interest to calculate its radially-averaged version $G_s(r, t)$ and its associated probability density $4\pi r^2 G_s(r, t)$. We calculate these quantities in isochoric Monte Carlo simulations wherein average step sizes are fixed throughout the simulation and not adjusted. This is done to make sure that each MC cycle is equivalent to the same period of time if the real dynamics of the system is considered.

The rotation of PDs is confirmed by observing several peaks in $G_s(r, t)$, the self-part of the van Hove correlation function [223], which implies that the tetrahedra indeed move between discrete sites separated by geometric barriers (Fig. 6.9a,b). As reported in our earlier work [18], each tetrahedron in the quasicrystal and the approximant is part of a spanning network of interpenetrating PDs (that is, PDs that share a tetrahedron). The locations of the peaks in $G_s(r, t)$ correspond to the characteristic distances of the nearest neighbor distances in the spanning network.

We observe that not all PDs are equally likely to rotate. At high densities, the PDs capping the twelvefold rings in the center of logs (shown in green in Fig. 5.14b)[18] rotate more frequently as they are spatially separated from the rest of the structure. This can be seen in the trajectories of the high-mobility particles in the approximant at $\phi = 65\%$ (Fig. 6.9c). Close to melting, however, rotations involve the full network of neighboring PDs, which allows the particles to diffuse over arbitrary distances (Fig. 6.9d). The underlying dynamics is identical in the quasicrystal. However the presence of defects leads to higher mobility in the quasicrystals that form in simulation as compared with "perfect" quasicrystals. Both the quasicrystal and the approximant exhibit some 'liquid-like' behavior since unlike simple crystals, diffusion can take place
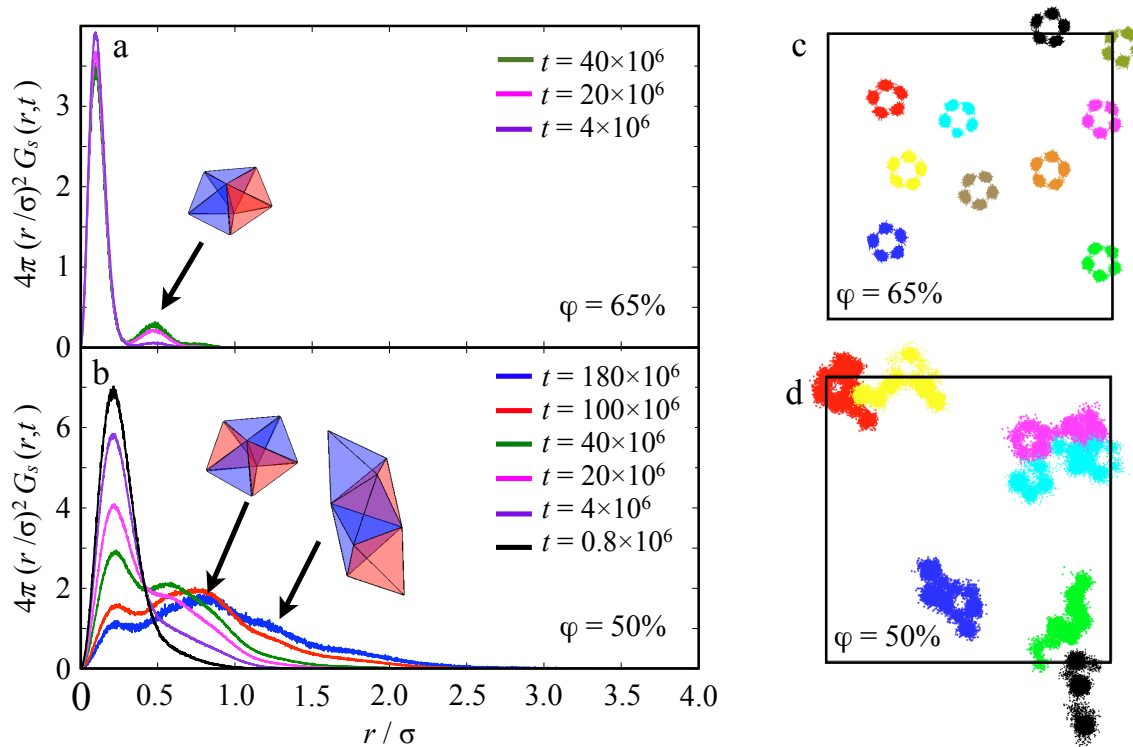
Figure 6.9:
Particle dynamics in the quasicrystal approximant. (a,b) The self-parts of the van Hove correlation functions at $\phi = 65\%$ (a) and $\phi = 50\%$ (b) show various peaks, which indicates that the particles do not move continuously but have to overcome (geometric) barriers. The peak positions correspond to different levels of nearest neighbor distances in the underlying PD network. (c,d) The trajectories of particles with the highest mobility are plotted. At high density, $\phi = 65\%$ (c), tetrahedra move along the edges of pentagons. This motion corresponds to rotations of the PDs in log centers. At intermediate densities, $\phi = 50\%$ (d), neighboring PDs start to rotate and the tetrahedra are more mobile. In the infinite time limit the tetrahedra can diffuse through successive PD rotations. (Images reproduced from [182])

a

b

φ = 75%

φ = 80%
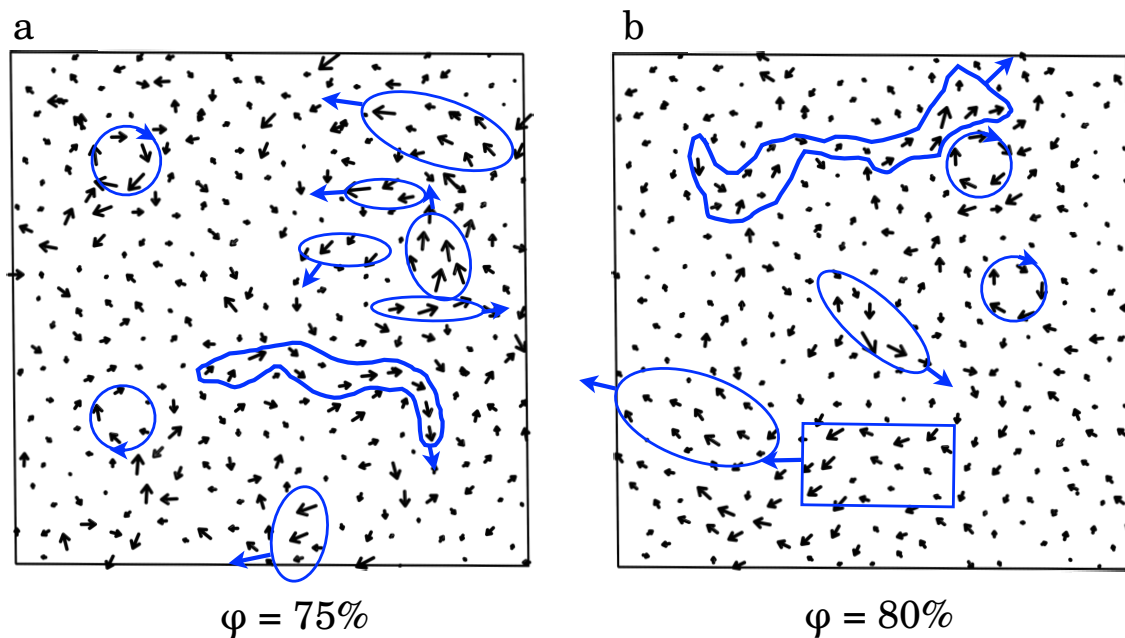
Figure 6.10: **Correlated motion of clusters in a slab of the approximant at (a)** $\phi = 75\%$ **and (b)** $\phi = 80\%$**.** Dark arrows correspond to the direction towards which each particle has moved after $t = 5 \times 10^7$ MC cycles; the length of each arrow is twice the distance the corresponding particle hard travelled. There are several clusters of neighboring tetrahedra moving collectively. A few of these clusters are highlighted in blue. Not surprisingly, the mobility is higher at $\phi = 75\%$ as evidenced by longer arrows. (Images reproduced from [182])

in these systems even in the absence of defects.

At packing densities beyond 65%, PD rotations become extremely unlikely, but clusters of tetrahedra, including PDs, can still vibrate collectively. Figs. 6.10a-b show such correlated motions occurring in a time period of 50 million MC cycles in a layer of the approximant at $\phi = 75\%$ and $\phi = 80\%$ respectively. The vibrations are extremely slow, but their existence adds additional entropy to the system making the mean-field approximation and the cell model inaccurate. No dynamics is observed in the dimer crystal.

In general, thermodynamically equivalent local rearrangements are a characteristic feature of quasicrystals and their approximants. The transformation among these takes place via phason modes [225–227]. Elementary excitations are phason flips, which previously have been observed with high-resolution transmission electron microscopy [228] and in simulations of two dimensional model systems [229].

6.6 Discussion and Conclusion

In general one might expect a 'simple' structure like the dimer crystal to form more easily than 'complex' structures like the quasicrystal or its approximant. The observation that tetrahedra defy this expectation suggests that structural complexity is not always a good indicator of thermodynamic stability. Indeed, although it has been argued in the literature [230] that the dimer crystal first reported by [31, 220] and studied here might be the stable phase even at densities where the quasicrystal is reproducibly observed (down to densities of 50%), our free energy calculations demonstrate that the dimer crystal is in fact preferred thermodynamically only at very high densities (above 84%). On the other hand, insofar as structural complexity increases a system's entropy, structurally complex arrangements of hard particles may be thermodynamically preferred over simpler ones.

Indeed, we have shown that the structural features of the quasicrystal and the approximant allow for more complex dynamics than the dimer crystal at moderate and high densities as manifested in the behavior of the free volume as a function of packing density and the collective motions in the form of PD rotations. The existence of the PD network facilitates collective particle motions at low densities. Although rearrangements become vanishingly unlikely at higher densities, they appear to contribute additional entropy to the system and stabilize it over the dimer crystal, in which each particle can only 'rattle' independently in its own cage. Rearrangements are impossible in the dimer crystal because no rearrangeable network exists there.

The superior stability of the quasicrystal and its approximant relative to the dimer crystal may also be attributed to the presence of almost-perfect face-to-face contacts between tetrahedra. There is a natural tendency for hard polyhedra to optimize face-to-face contacts at high densities in order to maximize configurational entropy. For instance, there are uncountably infinite cubic arrangements of hard cubes with packing fraction one, but among them the simple cubic lattice, where all cubes are perfectly face-to-face, has the highest entropy and is thermodynamically stable [128].

Within the approximant, we observe that face-to-face contacts between neighboring tetrahedra are nearly perfect in the sense that the touching faces are not significantly shifted with respect to one another. This is not true in the dimer crystal where inter-dimer face-to-face contacts are shifted and therefore not close to being perfect. Abundance of strong face-to-face contacts makes the PD network more rearrangeable and collective motions of particles more feasible, which in turn leads to a higher entropy and superior stability.

We summarize our findings in a schematic phase diagram in Fig. 6.11. We note that hard tetrahedra are one of the few examples of hard particles with two distinct solid phases not mutually related by symmetry breaking. Our results show that entropic effects alone are sufficient for inducing highly nontrivial solid-solid phase transitions.

Not all phase transformations are accessible in simulations on finite time scales. The observation that simulations only form the quasicrystal but never the approximant suggests that the quasicrystal is kinetically more easily accessible than the approximant– independent of whether it is thermodynamically preferred or not. This can be attributed to the fact that the transformation of a dodecagonal quasicrystal to one of its approximants proceeds through a process called zipper motion [216], which is extremely slow even in experiment [211]. Furthermore, transformation to the dimer crystal at packing densities greater than 84% is not observable in simulations, and may be unobservable in experiments, due to the extremely slow kinetics at such high densities.

In conclusion, we have shown that the quasicrystal and its approximant are thermodynamically favored over the dimer crystal at all experimentally realizable packing densities. We also observe a very rich dynamical behavior in the quasicrystal and its approximant induced by rotations of pentagonal dipyramids within an interconnected network. We have shown the significance of collective motions in stabilizing the approximant for a wide range of packing densities.
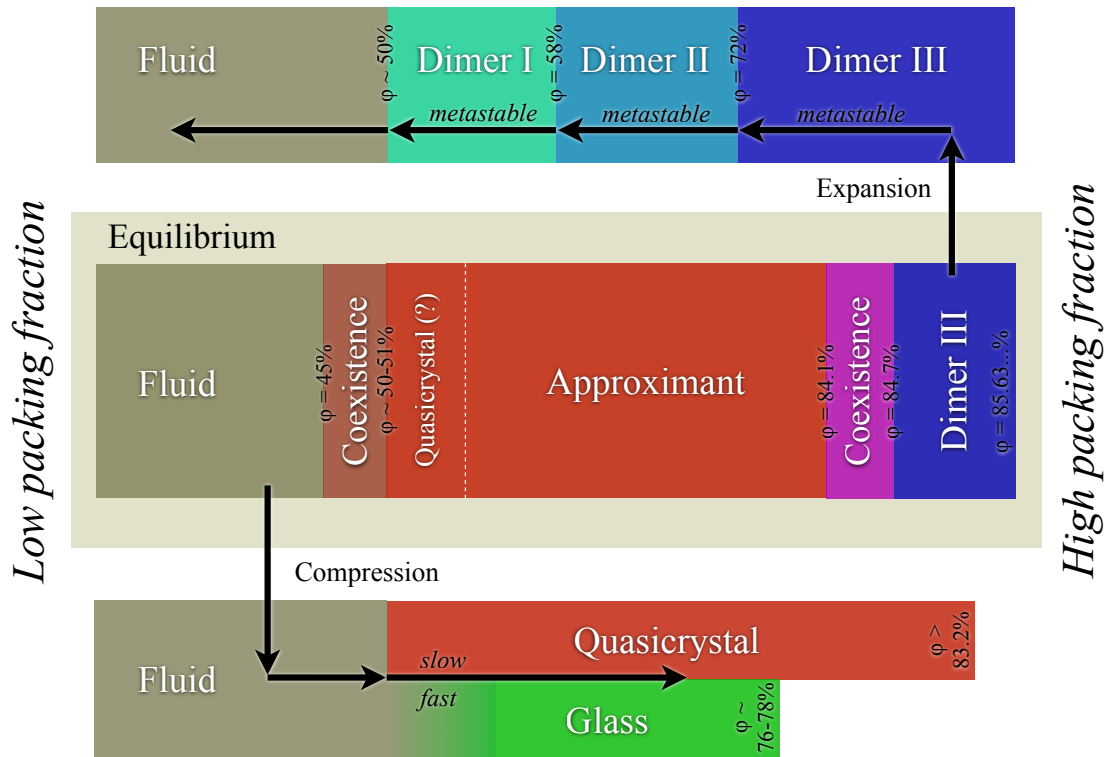
Figure 6.11: **Schematic phase diagram of hard tetrahedra** summarizing our findings. In thermodynamic equilibrium the Dimer III crystal and the approximant are stable (Middle panel). In compression simulations the approximant is never observed, and only the quasicrystal forms. If crystallization is suppressed, then a jammed packing with local tetrahedral order forms [18, 231] (Lower panel). The transformation of the approximant or quasicrystal directly to and from the Dimer III crystal is not observed in simulation. Instead, during expansion the Dimer III crystal transforms into the Dimer II crystal, and then the Dimer I phase prior to melting to the fluid (Upper Panel). (Images reproduced from [182])

CHAPTER VII

Hard Triangular Bipyramids[1]

In the densest known packing of regular tetrahedra, particles are paired into perfect triangular bipyramids. Despite being the densest– and hence the thermodynamically stable phase in the limit of infinite pressure–, this structure does not form in simulation. As explained in Chapter VI, quasicrystal-like phases are more efficient in opening up rattling space for individual particles when expanded. This can be partly attributed to the collective motions of tetrahedra in the quasicrystal and its approximants helping the system explore more configurations than it would have otherwise accessed. In a system of hard triangular bipyramids– i.e. dimers of hard tetrahedra– the dynamics will be far more restricted due to rigid connections between tetrahedra that are part of a single dimer. It is therefore reasonable to expect that the quasicrystal-like phases be destabilized in that system, maybe to the extent that the dense dimer crystal could become thermodynamically superior over them at intermediate densities as well. The dimer crystal will not be as adversely affected due to the natural pairing of tetrahedra into dimers in the tetrahedron-based dimer crystal.

Motivated by this consideration, we studied the system of hard triangular bipyramids (TBPs) using Monte Carlo simulation. To our surprise, the system still forms a dodecagonal quasicrystal, which is identical to the quasicrystal formed by hard tetrahedra in the monomer level. However it is characterized by an additional level of randomness in the way individual tetrahedra are paired into TBPs on the network of nearest neighbors. This pairing degeneracy leads to a nondeterministic decoration of individual tiles, something that is observed for the first time for any quasicrystal. Similar pairing degeneracy is observed in *degenerate crystals* formed by hard sphere dimers. We therefore call this new phase a *degenerate quasicrystal*.

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[1]This chapter is partly based on the following manuscript: Amir Haji-Akbari, Michael Engel, Sharon C. Glotzer, *Phys. Rev. Lett.* **107**: 215702 (**2011**) [21].

This chapter is organized as follows. In Section 7.1, we mention the technical specifications of the simulations performed in this study. Section 7.2 is dedicated to the notion of degenerate phases. In Section 7.3, we report the formation of the degenerate quasicrystal. In Section 7.4 different approximant phases are discussed. The thermodynamic stability of different phases are discussed in Section 7.5. A brief study of truncated TBP system is given in Section 7.6. And concluding remarks are given in Section 7.7.

7.1 Technical Specifications of Performed Simulations

Isochoric and isobaric Monte Carlo simulations are used for self-assembly simulations, while the isobaric Monte Carlo is used in calculating the equations of state of different phases. For crystalline phases, the isotension MC is used wherein the shape of the simulation box is also altered. Free energies of different phases are calculated using the Frenkel-Ladd-based method described in Section 3.3.4. Table 7.1 gives the technical specifications of all simulations performed in this study. The vertices of a prototypical triangular bipyramid that corresponds to the quaternion $\mathbf{q} = (1, 0, 0, 0)$ are given by:

$$
\begin{aligned}
\mathbf{v}_{1,2} &= \left(0, 0, \pm\frac{4\sqrt{3}}{3}\right) \\
\mathbf{v}_{3,4} &= \left(-\frac{\sqrt{6}}{3}, \pm\sqrt{2}, 0\right) \\
\mathbf{v}_5 &= \left(\frac{2\sqrt{6}}{3}, 0, 0\right)
\end{aligned}
$$

The dimensionless pressure is defined as $P^* = P\sigma^3/k_B T$ with σ being the edge length of a single TBP. All simulations are carried out in boxes with periodic boundary conditions.

7.2 Degenerate Phases

Hard disks and spheres order into hexagonal and face-centered cubic crystals, respectively, above a certain packing fraction. A more complex phase behavior is observed if the disks or spheres are rigidly bonded into dimers (dumbbells) [16, 184, 233, 234]. A solid phase, disordered in the orientation of dimers while ordered on the

Table 7.1: **Simulation details for the calculation of the equation of state (EOS), quasicrystal (QC) assembly, and thermodynamic integration.** Several independent runs were performed for each state point to assure accurate statistics. The smallest system used for quasicrystal formation has 1,458 particles. Quasicrystal formation is robust and is routinely observed at densities above 54% and system sizes larger than 1,400 particles. We do not observe a quantitative difference in the structure of the quasicrystal for such large system sizes. The small, 432-particle system is only used for the mathematically constructed TBP crystal (with 216 two-particle unit cells), which is used for estimating the equation of state and free energy calculations only.

Phase	Objective	Ensemble	System size	MC sweeps	Parameters
Fluid	EOS calculation	isobaric	2,624	10^7	$0.01 \leq P^* \leq 60$
Fluid	QC assembly	isobaric	$1{,}458 - 2{,}624$	10^8	$40 \leq P^* \leq 60$
Fluid	QC assembly	isochoric	8,000	10^8	$0.5 \leq \phi \leq 0.6$
TBP crystal	EOS calculation	isobaric	432	10^7	$32 \leq P^* \leq 10{,}000$
Quasicrystal	EOS calculation	isobaric	2,624	10^7	$36 \leq P^* \leq 10{,}000$
Approximant	EOS calculation	isobaric	656	10^8	$35 \leq P^* \leq 10{,}000$
Approximant	Dynamics	isochoric	656	3×10^8	$0.57 \leq \phi \leq 0.80$
TBP crystal	Therm. Integration	isochoric	432	2×10^5 per γ	$0 \leq \gamma \leq 4 \times 10^6, c = 1/2$ $0.70 \leq \phi \leq 0.80$
Approximant	Therm. Integration	isochoric	656	2×10^5 per γ	$0 \leq \gamma \leq 4 \times 10^6, c = 1/2$ $0.60 \leq \phi \leq 0.80$
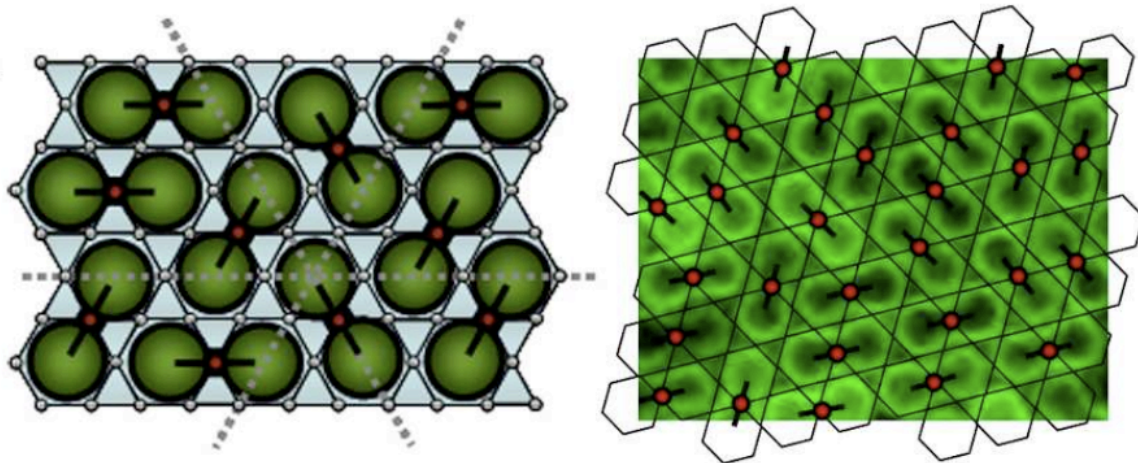
Figure 7.1:
Degenerate crystals of hard sphere dimers. The structure is a hexagonal lattice in the monomer level, but the orientations of dimers on the lattice is random. (Images reproduced from [232])

monomer level, forms if the distance between monomers within a dimer is roughly the diameter of a monomer (Fig. 7.1). This equilibrium solid phase can be alternatively understood as a random pairing of neighboring monomers within the native monomer crystal. The resulting thermodynamic ensemble of ground states is degenerate and the structure is therefore called a *degenerate crystal*. As shown by Wojciechowski *et al.* [16] for hard disks, every configuration of the degenerate crystal that is obtained from a certain pairing pattern has less entropy than a regular arrangement of dimers, but the degenerate crystal is stabilized due to the entropy associated with different ways of pairing hard spheres into dimers. Other consequences of the pairing of monomers into dimers include topological defects [235], a restricted, glassy dislocation motion [236, 237], and unusual elastic properties [238]. Similar degenerate phases have also been observed for freely-joined chains of hard spheres [239, 240].

Although degenerate crystals can potentially assemble from dimers of hard shapes other than disks and spheres, few examples have been reported. One reason is the competition between degenerate crystals and the liquid crystalline phases frequently observed for particles with large aspect ratios. For example, elongated tetragonal parallelepipeds, which for an aspect ratio of 2:1 can be viewed as dimers of face-sharing cubes, form a degenerate *parquet* phase at intermediate densities before transforming into a smectic liquid crystal that eventually crystallizes [17]. In the parquet phase, the centers of monomers form a simple cubic lattice, but their pairing into dimers is random.

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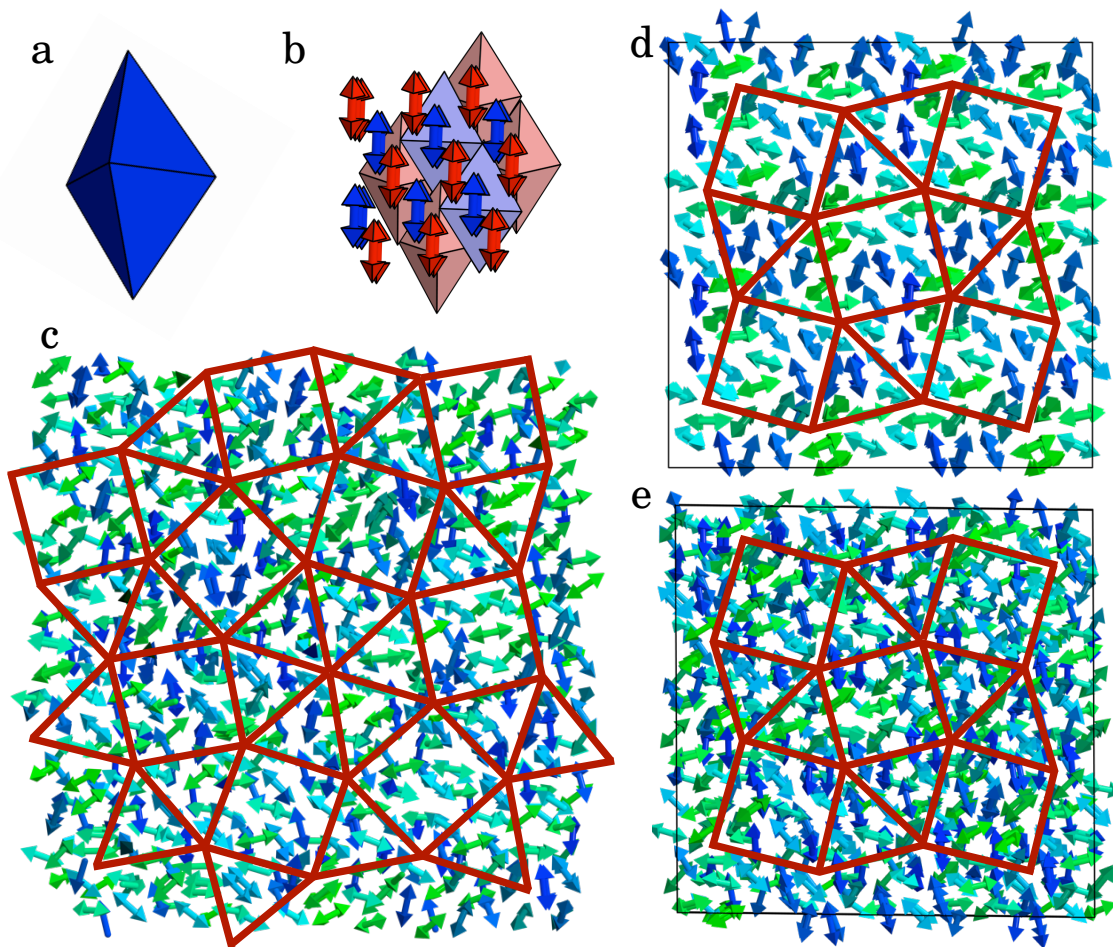
Figure 7.2: **Phases formed by (a) triangular bipyramids (TBPs)**: (b) TBP crystal, (c) degenerate quasicrystal, (d) regular quasicrystal approximant, (e) degenerate quasicrystal approximant. For visualization purposes, we show member tetrahedra of most TBPs at 30% actual size and connect their centers with bonds. In (c-e), tetrahedra and bonds are colored according to their orientation projected on the plane. (Images reproduced from [21])

7.2.1 Feasibility of Degenerate Phases in the Hard Triangular Bipyramid System

Another simple dimer is the triangular bipyramid (TBP), which consists of two face-sharing, regular tetrahedra (Fig. 7.2a). The TBP is the simplest face-transitive bipyramid and the twelfth of the 92 Johnson solids[2] [241]. The lack of inversion symmetry of the TBP, however, makes lattice packings non-optimal [29], and thus it is potentially more interesting as a dimer than dimers of spheres and cubes. Moreover, the recent synthesis of TBP-shaped nanoparticles and colloids [85, 87–89] makes the investigation of this building block of practical relevance.

In both of the known ordered phases of hard, regular tetrahedra, each tetrahedron is in almost-perfect face-to-face contact with at least one other tetrahedron. The densest known packing of tetrahedra ($\phi = \frac{4000}{4671} \approx 85.63\%$) i.e. the dimer crystal is a parallel arrangement of two dimers (four tetrahedra) – that is, two TBPs – in a triclinic unit cell. We refer to this phase in the present chapter as the *TBP crystal* (Fig. 7.2b). At lower packing fractions, hard tetrahedra assemble into a dodecagonal quasicrystal [18], in which the tetrahedra form a decorated square-triangle tiling [216]. Degenerate phases are impossible in the TBP crystal because the contacts between neighboring tetrahedra in different TBPs are highly imperfect [182], but are possible in the quasicrystal due to the almost perfect face-to-face contacts between all neighboring tetrahedra.

7.3 Degenerate Quasicrystal

The dodecagonal quasicrystal of TBPs forms spontaneously from the equilibrium fluid phase at packing fractions above 54%. Fig. 7.3a depicts a side view of the quasicrystal formed in an isobaric simulation of 2,624 TBPs at reduced pressure $P^* = P\sigma^3/k_BT = 46$ and subsequently compressed to a packing fraction of 81.34%. TBPs arrange into layers (white lines), which stack on top of each other perpendicular to the twelvefold symmetry axis (dark arrow). We confirmed that the formation of the quasicrystal occurs reproducibly in systems with at least a few thousand particles and does not depend on the shape of the simulation box. It even forms in the boxes commensurate to the unit cell of a TBP crystal.

The quasicrystal structure can be best understood by replacing each bipyramid

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[2] A Johnson solid is a convex polyhedron whose faces are all regular polygons, but is not a Platonic or Archimedean, prism or antiprism. They are called Johnson solids in the honor of Norman W. Johnson who enumerated them and showed that there are only 92 polyhedra with this property [241].
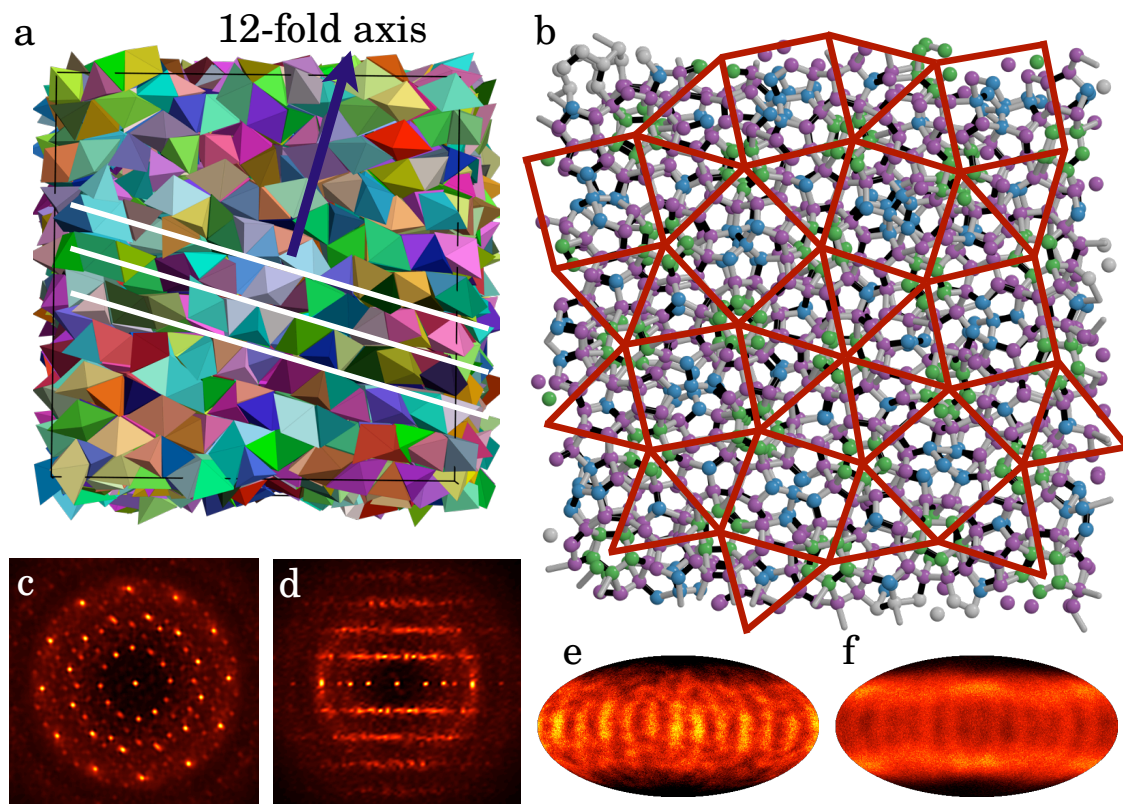
Figure 7.3: **Degenerate quasicrystal of hard TBPs**: (a) TBPs assemble into a dodecagonal quasicrystal in isobaric and isochoric Monte Carlo simulations. (b) The square-triangle tiling obtained by connecting the centers of twelvefold rings of member tetrahedra. Intra- and inter-TBP bonds are depicted in black and gray respectively. (d,e) Diffraction patterns with centers of member tetrahedra as scatterers calculated (c) perpendicular to and (d) across the layers. (e) Intra-TBP and (f) total bond order diagrams. (Images reproduced from [21])

by its two member tetrahedra. Fig. 7.3b depicts the centroids of tetrahedra within a few layers of Fig. 7.3a. Neighboring tetrahedra are connected with bonds. Two tetrahedra are defined as neighbors if their distance lies within the first peak of $g_t(r)$, the radial distribution function based on the centroids of the tetrahedra. Dodecagons that are depicted in purple in Fig. 7.3b correspond to rings of twelve member tetrahedra (Fig. 5.11b), a structural motif characteristic of the tetrahedron-based quasicrystal [18]. These rings are further capped with pentagonal dipyramids (PDs), five tetrahedra sharing an edge (Fig. 5.1a), visible in the figure as pentagons (green) within dodecagons. Additional member tetrahedra, referred to as interstitials, fill the space between the rings and are depicted in dark blue. Together, dodecagons and PDs form motifs whose centers are the vertices of square and triangle tiles. Their mixing gives the square-triangle tiling its overall twelvefold symmetry as observed in the diffraction pattern depicted in Fig. 7.3c. Layering along the twelvefold axis can be seen in Fig. 7.3d. Overall, the arrangement of the member tetrahedra is identical to that reported in the hard tetrahedron system [18].

In order to prove that the self-assembled structure is indeed quasiperiodic and not a periodic arrangement of the six fold supercluster shown in Fig. 7.3b, we carry out self-assembly simulations in larger systems. Fig. 7.4 shows the tiling for the quasicrystal that we assembled in an NVT simulation of 8,000 hard TBPs– i.e. 16,000 tetrahedra– at $\phi = 54\%$. The tiling is different from what is observed in the smaller system and is in line with what is observed in large systems of hard tetrahedra. For instance, it contains a few rhombi that are the defects of the quasicrystal formation and are routinely observed in dodecagonal quasicrystals formed in experiments or simulations.

To elucidate how the bipyramids are arranged within the quasicrystal, we compare statistical distributions of intra-TBP bonds (bonds that connect member tetrahedra within TBPs) and the set of all bonds in the quasicrystal by projecting both sets onto the surface of a unit sphere[3]. The resulting diagrams are referred to as intra-TBP and total bond order diagrams, respectively, and are visualized using the Mollweide projection with the twelvefold axis pointing in the vertical direction. Comparing these bond order diagrams (Fig. 7.3e and Fig. 7.3f), we observe no significance difference in the distribution of bond directions within the twelvefold layers. This suggests that pairing of tetrahedra in the quasicrystal does not follow a predefined set of rules and is instead random. However tetrahedra tend to pair more strongly within layers

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[3]All bond order diagrams presented in this chapter are prepared using a Molleweide projection, which maps the surface of S^2 onto a plane.
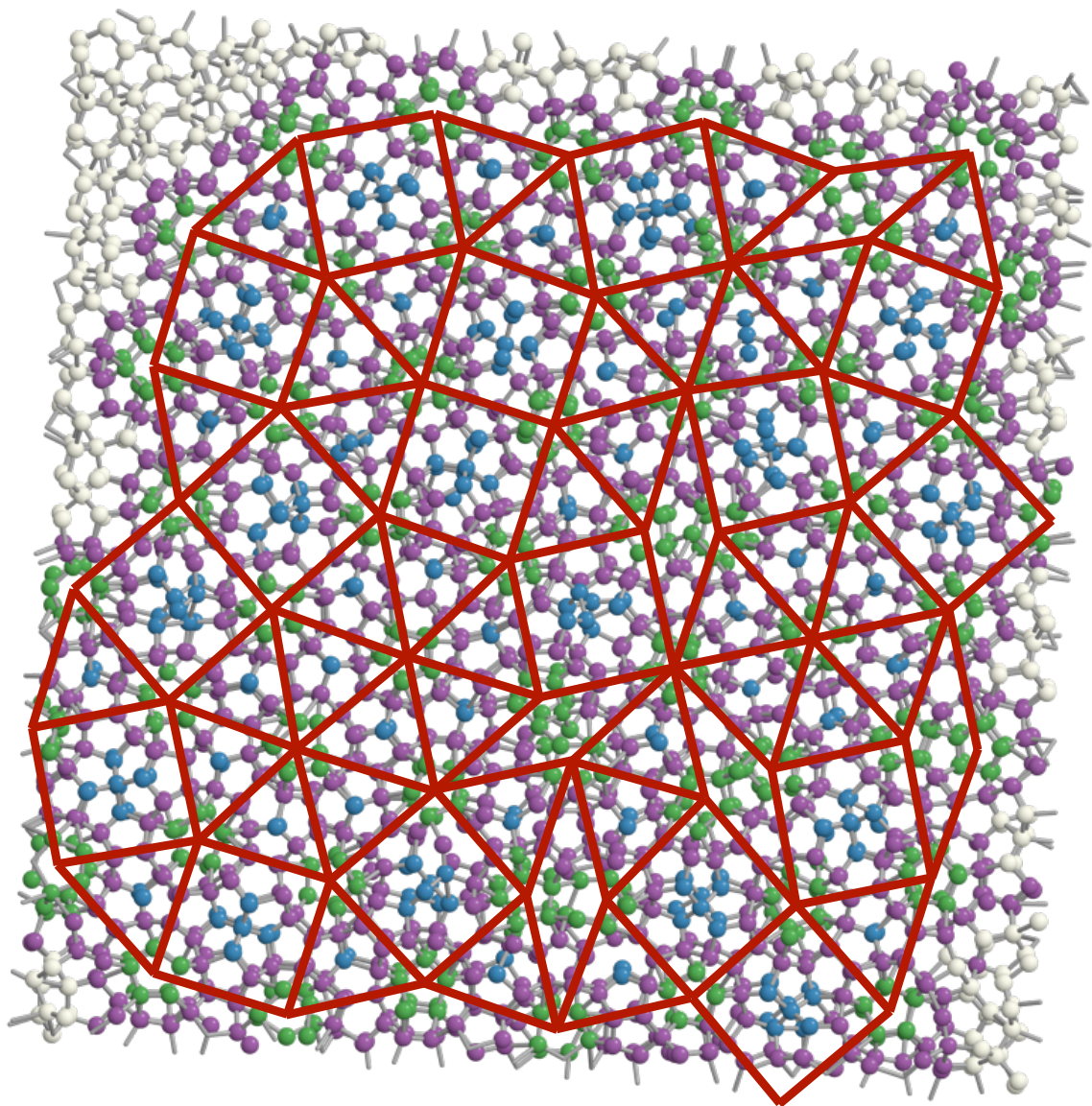
Figure 7.4: **Tiling obtained for a slab of the degenerate quasicrystal formed in an NVT simulation of** $8,000$ **particles at** $\phi = 54\%$**.** Like Fig. 7.3b, the centers of member tetrahedra are depicted and they are colored according to their environment: twelvefold ring (purple), capping PDs (green) and interstitials (blue). Compared to the quasicrystal shown in Fig. 7.3b, the assembled quasicrystal has more defects and is yet to fully crystallize in the layers above and below the ones shown in the figure. This explains the abundance of rhombs and zippers that have shown to be important in the process of crystallization. (Images reproduced from [21])

than between neighboring layers, a fact that can be explained by noting that face-to-face contacts are more perfect within layers. Motivated by studies of hard sphere dimers [16], we refer to the TBP quasicrystal as a degenerate quasicrystal (DQC). The randomness can be seen clearly in Fig. 7.2c. The notion of degeneracy discussed here should not be confused with the extensively studied degeneracy associated with random tiling quasicrystals [242, 243] where tiles with unique decoration patterns mix to form random tilings. We instead report a new type of randomness in the level of decorating individual tiles, in addition to the degeneracy of the random tiling.

It is surprising that the structural quality of the DQC is not compromised despite the additional geometrical constraints imposed on the system by pairing tetrahedra into TBPs. For instance, we find that the maximum packing fraction achieved by replacing the bipyramids with individual member tetrahedra and then compressing is statistically identical to that obtained in simulations of hard tetrahedra.

The thermodynamics of hard TBP fluid is also very similar to its hard tetrahedron counterpart. The simple fluid gradually transforms into a complex fluid prior to crystallization. A kink is observed at $P^* = 46$ in the equation of state of the fluid (Fig. 7.5a), and so is an increase in the fraction of particles that are part of a PD or an icosahedron (Fig. 7.5b). Fig. 7.6 shows $g_t(r)$, the radial distribution function based on centers of tetrahedra, for different densities. In order to show the structural changes in the fluid more clearly, we do not show the trivial Dirac peak at $r = \sigma/\sqrt{6} \approx 0.41\sigma$ which corresponds to intra-TBP bonds. There is a change in structure between $50\% \leq \phi \leq 56\%$ wherein the second peak disappears and then reappears splitting into two peaks, the same behavior observed in the hard tetrahedron fluid (Fig. 5.8a). As in the hard tetrahedron system, the locations of the first three peaks in $g_t(r)$ correspond to characteristic distances in a PD network. It is noteworthy that this PD network exists in both the disordered fluid and the quasicrystal in both these systems.

7.4 Quasicrystal Approximants

Due to the additional level of randomness in pairing tetrahedra into TBPs, constructing an approximant of the TBP quasicrystal involves not only choosing a periodic tiling and decorating it with tetrahedra, but also pairing the tetrahedra into bipyramids. We choose the $(3.4.3^2.4)$ Archimedean tiling which, in the case of hard tetrahedra, gives rise to the densest approximant (Fig. 5.14). There is no unique way of pairing tetrahedra into TBPs even within a single unit cell of the approximant due
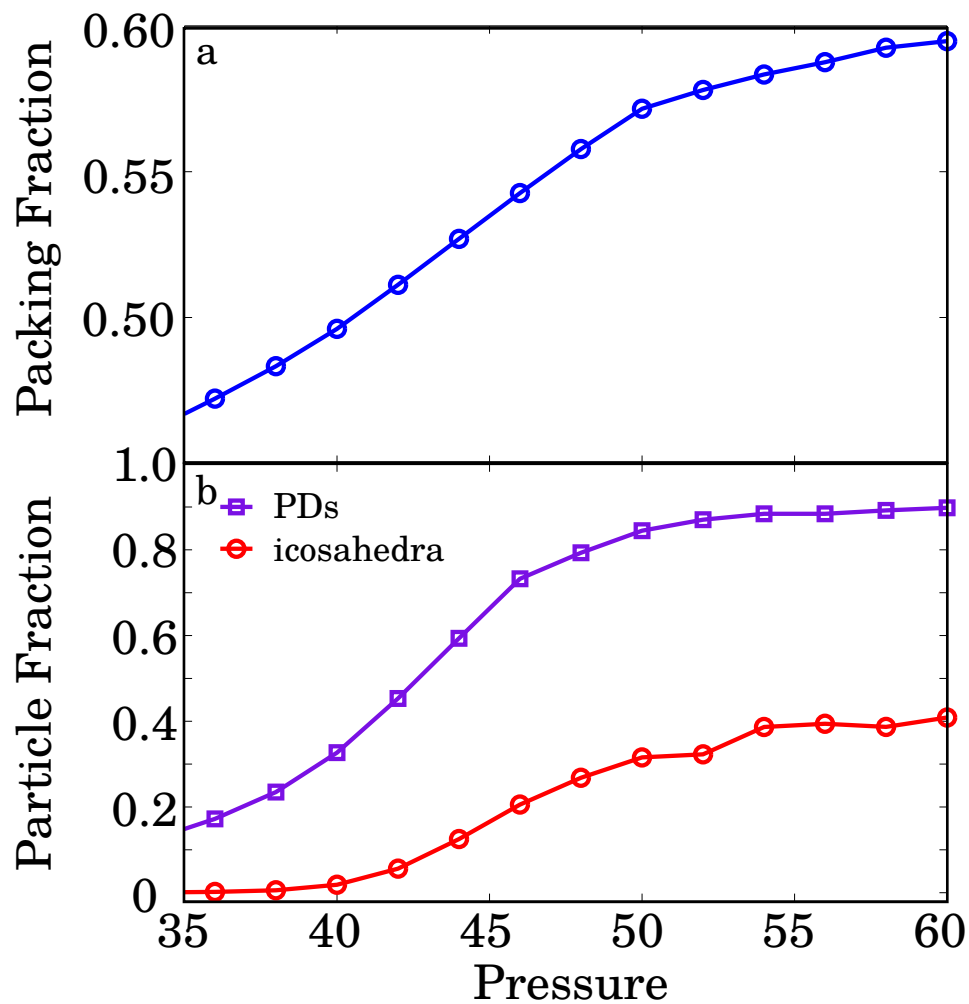
Figure 7.5: **Thermodynamics of the hard TBP fluid.** (a) Equation of state, (b) Fractions of constituent tetrahedra in PDs and icosahedra calculated in isobaric simulations of a system of 2,624 particles.
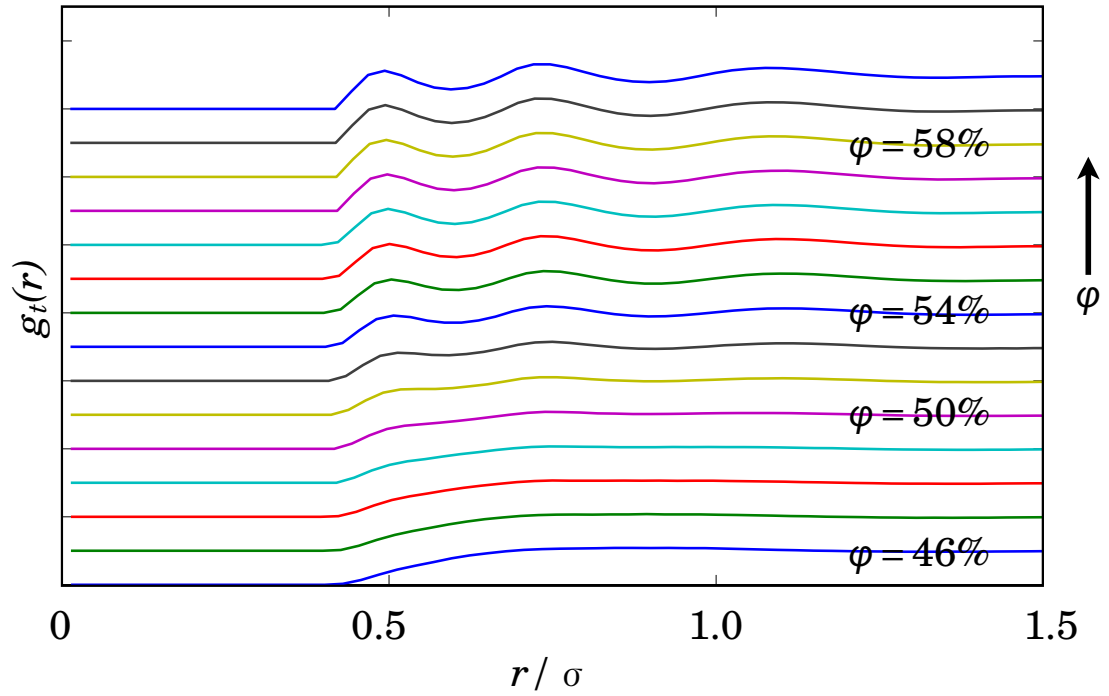
Figure 7.6:
Radial distribution function of the hard TBP fluid. $g_t(r)$, the radial distribution function based on centers of tetrahedra, calculated in isobaric simulations of $N = 2,624$ TBPs in a cubic box. the trivial Dirac peak at $r/\sigma = 1/\sqrt{6}$ is removed for clarity.

to degeneracies associated with rotations of the capping PDs. In particular, it is not possible to avoid breaking the fourfold symmetry of the approximant unit cell in the pairing process. We constructed a *regular approximant* by retaining as much of the symmetry as possible. Top and bottom views of the constructed approximant are depicted in Figs. 7.7a,b. There are six types of particles in the structure:

- **Intra-PD Particles**: Member tetrahedra are both part of one of the four PDs that cap twelvefold rings. There are eight such pairs in the unit cell and are all depicted in light green.

- **PD-ring Particles**: They connect a capping PD to a dodecagonal ring. There are four such pairs in the unit cell, and they are depicted in dark green.

- **Intra-ring Particles:** The tetrahedra that are paired are part of the same twelvefold ring. There are twelve such pairs in the unit cell, that are shown in purple.

- **Inter-ring Particles:** They connect tetrahedra within neighboring rings. There are five such pairs in the system and are depicted in red.

- **Ring-Interstitial Particles:** They connect twelvefold rings to one of the interstitial tetrahedra. There are ten such pairs in the system colored in cyan.

- **Interstitial-Interstitial Particles:** The connect the two central tetrahedra in the center of the two square tiles to one of its neighbors. Obviously there are only two such pairs in the unit cell that are colored in blue.

A schematic of the unit cell is shown in Fig. 7.7c where the ring-ring and ring-interstitial connections are highlighted in red and blue respectively. We find that the regular approximant can be compressed to a maximum packing fraction of 83.39% (Appendix C.3.1), a bit less than the maximum packing fraction of 85.03% achieved for the quasicrystal approximant constructed of individual tetrahedra [18]. Compressing a $2 \times 2 \times 2$ unit cell of the approximant marginally improves the packing to $\phi_{\max} = 83.43\%$. The distinctive difference between the intra-TBP (Fig. 7.7d) and the total bond order diagrams (Fig. 7.7e) is a result of this deterministic pairing (Fig. 7.2d).

By expanding the regular approximant, we find that it melts at $P^* \leq 35$ and packing fractions $\phi < 54\%$. But before melting, the crystal slowly transforms into a more loosely packed structure in which tetrahedra are paired at random into TBPs, just as
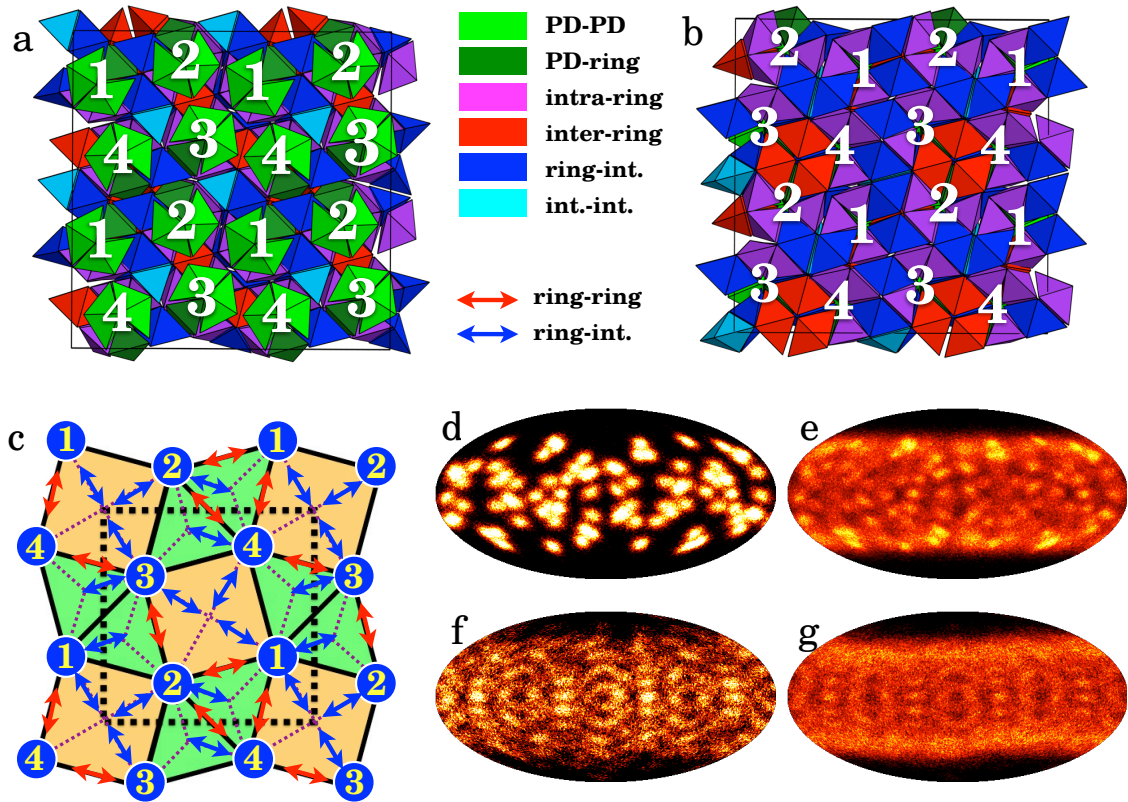
Figure 7.7: **Quasicrystal approximants**: (a) Top and (b) bottom views of the regular approximant. The unit cell has 41 triangular bipyramids. Particles are colored according to their environment: PD-PD (light green), PD-ring (dark green), intra-ring (purple), ring-ring (red), ring-interstitial (blue), interstitial-interstitial (cyan). (c) Schematics of the unit cell with connections between neighboring rings and between rings and central interstitials shown with red and blue double-arrows, respectively. (d,f) Intra-TBP and (e,g) total bond order diagrams for (d,e) the regular approximant and (f,g) the degenerate approximant. In the legends, 'int.' stands for 'interstitial'. (Images reproduced from [21])

in the DQC, although their positions and orientations are unchanged (Fig. 7.2e). The resulting structure is therefore degenerate to the tetrahedron-based approximant and we refer to it as a *degenerate approximant (DA)*. The angular distribution of intra-TBP bonds around the fourfold axis (Fig. 7.7f) is more similar to that of all bonds (Fig. 7.7g) in the degenerate approximant than in the case of the regular approximant (Figs. 7.7d,e), which again suggests random pairing. We find that the transformation from regular to degenerate approximant is irreversible on the time scale of our simulations ($\approx 10^8$ MC cycles). Since the DA can only be recompressed to a density of 82.88%, which is lower than the maximum density of the regular approximant, the DA has to be stabilized by its pairing disorder close to melting. We refer to the entropy associated with random pairing on a network as the *pairing entropy*.

To understand how the regular approximant transforms into the DA, we note (Fig. 7.3b) that the arrangement of the member tetrahedra can be alternatively understood as a spanning network of interpenetrating PDs [18]. In the hard tetrahedron system, PDs can easily rotate around their principal axes [182]. Such rotations are also essential in understanding the local rearrangements of bipyramids at densities below 60%. As shown in Fig. 7.8b, TBPs move very little at $\phi = 60\%$. Even after 250 million MC cycles only a small fraction of TBPs have moved as much as σ. Much faster dynamics occurs at $\phi = 57\%$. Particles at or near that density move over discrete distances that are characteristic of a PD network (Fig. 7.8a). These rearrangements change neither the tiling nor its decoration. Instead, they reshuffle the pairing pattern by a sequence of PD rotations. After a sufficiently large number of reshuffling moves the DA emerges from the regular approximant.

7.5 Thermodynamics

Next we study the relative thermodynamic stability of various phases. We first compare the DQC obtained in simulation and its constructed approximants. As observed in Fig. 7.10a, both the regular and the degenerate approximant are slightly denser than the DQC at all pressures. The relation $G(P_2^*) - G(P_1^*) \propto \int_{P_1^*}^{P_2^*} \phi^{-1} dP^*$ between the free energy and the equation of state then suggests that the approximants are thermodynamically preferred over the DQC at sufficiently high pressures because their Gibbs free energies increase more slowly with pressure. Furthermore, the approximants melt at lower pressures than the quasicrystal, which indicates that they might even be more stable than the quasicrystal at all pressures. Nevertheless, the DQC remains the only ordered phase that forms in our simulations. It is also
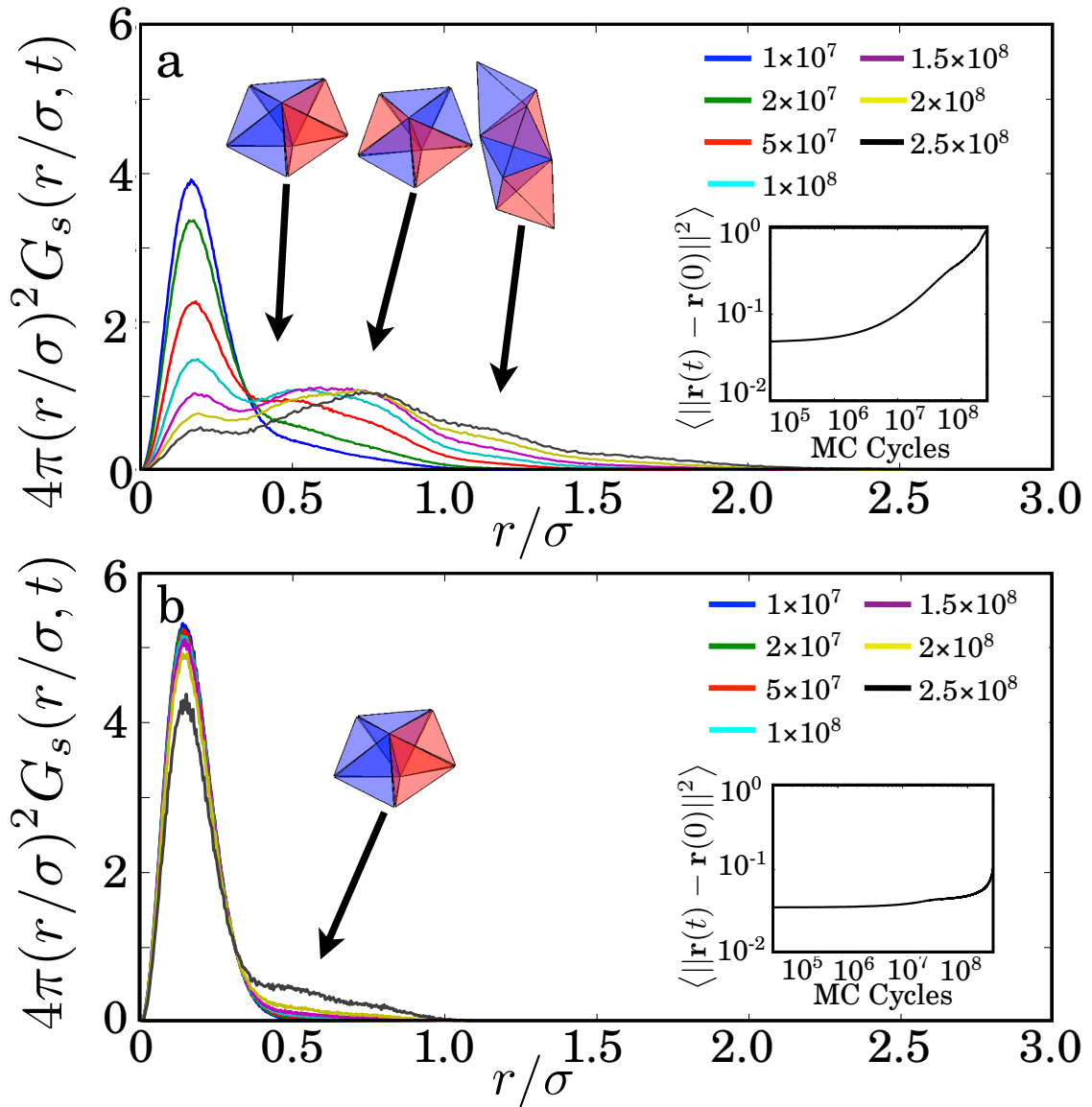
Figure 7.8: **Dynamics of the hard TBP system**. The self part of the van Hove correlation function $G_s(r,t)$ measures the particle motion in the approximant. The separation distance $r(t)$ is calculated between centers of mass of member tetrahedra. (a) Large rearrangements occur at $\phi = 57\%$. (b) There is little motion present at $\phi = 60\%$. The observed dynamics is similar to that observed in the hard tetrahedron system [182]. (Images reproduced from [21])

the only structure we expect to be observed in experiments of hard nanocolloidal TBPs since the kinetic process of transforming from the DQC into the approximant is extremely slow. Considering the local structural similarity of the DQC and the fluid in terms of the PD network, the formation of the less stable DQC and not the approximant in simulation may be another example of Ostwald's rule [244].

The problem of comparing the relative thermodynamic stability of the regular approximant and the degenerate approximant is however less conclusive. What we know for certain is that the regular approximant will be more stable in the limit of infinite pressure thanks to its higher packing fraction while the degenerate approximant is more stable at pressures close to melting since it spontaneously emerges in simulations of the regular approximant. This transition is irreversible so its location cannot be exactly determined from conventional simulations. Instead, the pairing entropy must be determined and used, alongside thermodynamic integration of each phase, to determine the pressure at which the degenerate approximant becomes less stable. In principle, the number of different ways that a nearest-neighbor graph can be partitioned into dimers can be determined from Wang-Landau type simulations, but determining the pairing entropy is more tricky since different degenerate approximants that can be obtained from different coverings of the network do not have identical densities; each of them therefore has a different vibrational entropy. In short, this question cannot be addressed with the tools that we currently have, and can be the subject of future research.

Next we calculate the equation of state of the analytically constructed TBP crystal using isotension Monte Carlo simulation (Fig. 7.9). The TBP crystal melts at $P^* = 32$ which is *lower* than the melting pressure for the quasicrystal $P^* = 36$ and the approximant $P^* = 35$. This is in contrast with the hard tetrahedron system where the dimer crystal melts at higher pressures than the quasicrystal and its approximants. We also monitor the equation of state carefully and observe the same anomalous behavior explained in Section 6.2. A peak is observed in the compressibility curve at $P^* \approx 60$, which is indicative of a second-order transition (Fig. 7.9b). This is further confirmed by re-compressing the TBP crystal relaxed right before melting to observe that there is no hysteresis in the equation of state(Fig. 7.9a). As we did for the dimer crystal in the hard tetrahedron system, we sort the box dimensions and box angles at each snapshot, and average these sorted values (Fig. 7.9c–d). This reveals that the transition at $P^* \approx 60$ is a displacive transition from a high-density monoclinic phase to a low-density rhombohedral phase. This analysis also reveals another displacive transition at $P^* \approx 150$ from the monoclinic phase to a triclinic phase. Inspired by

our notations in Chapter VI, we denote these three phases as T-I, T-II and T-III respectively.

Next, we compare the approximant with the TBP crystal by calculating the free energy difference between them. As shown in Fig. 7.10b, the approximant has a lower free energy than the TBP crystal for packing fractions below 79%. A phase transition occurs at $P_c^* = 356 \pm 50$, corresponding to coexistence packing fractions of $\phi_{c,\mathrm{app}} = (79.7 \pm 0.8)\%$ and $\phi_{c,\mathrm{TBP}} = (80.7 \pm 0.7)\%$. The thermodynamic stability of the approximant at lower densities can be attributed to the additional configurational entropy associated with collective motions of particles. Such motions are not present in the TBP crystal. Their role in stabilizing the quasicrystal approximant has been shown for the structurally and dynamically similar system of hard tetrahedra (Section 6.5.1). The phase diagram of the hard TBP system is depicted in Fig. 7.10c.

It is necessary to mention that the dotted curves in Fig. 7.10b are the free energy differences calculated using the regular and the degenerate approximant in Frenkel-Ladd-type thermodynamic integration, and the solid curve is their average. We do this simply because we are not aware of the precise pressure at which the degenerate approximant becomes unstable. It is noteworthy that the lower dotted curve is only obtained for one instance of the degenerate approximant, and no pairing entropy is included in its calculation. Including the pairing entropy will stabilize the degenerate approximant with respect to the TBP crystal, and will shift the dotted curve to the right. This approach therefore gives us very conservative error-bars for the transition pressure and coexistence densities. In the case of a disordered fluid however, this approach cannot be used since inclusion of the pairing entropy will shift the error-bars in the wrong direction. We therefore do no free energy calculation for determining the melting pressure- as we did in Chapter VI and the reported coexistence densities are from melting simulations only.

7.6 Hard Truncated Triangular Bipyramids[4]

Motivated by a recent study of hard truncated tetrahedra by Damasceno *et al.* [19], we study the self-assembly and dense packings of hard truncated triangular bipyramids especially to explore the possibility of new non-quasicrystalline phases that are degenerate to their corresponding truncated tetrahedra counterparts. We also want to know how robust the quasicrystal is to the truncation of the building blocks. Below we present the technical specifications of our simulations as well as our preliminary

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[4]The content of this section is based on a manuscript that is under preparation.
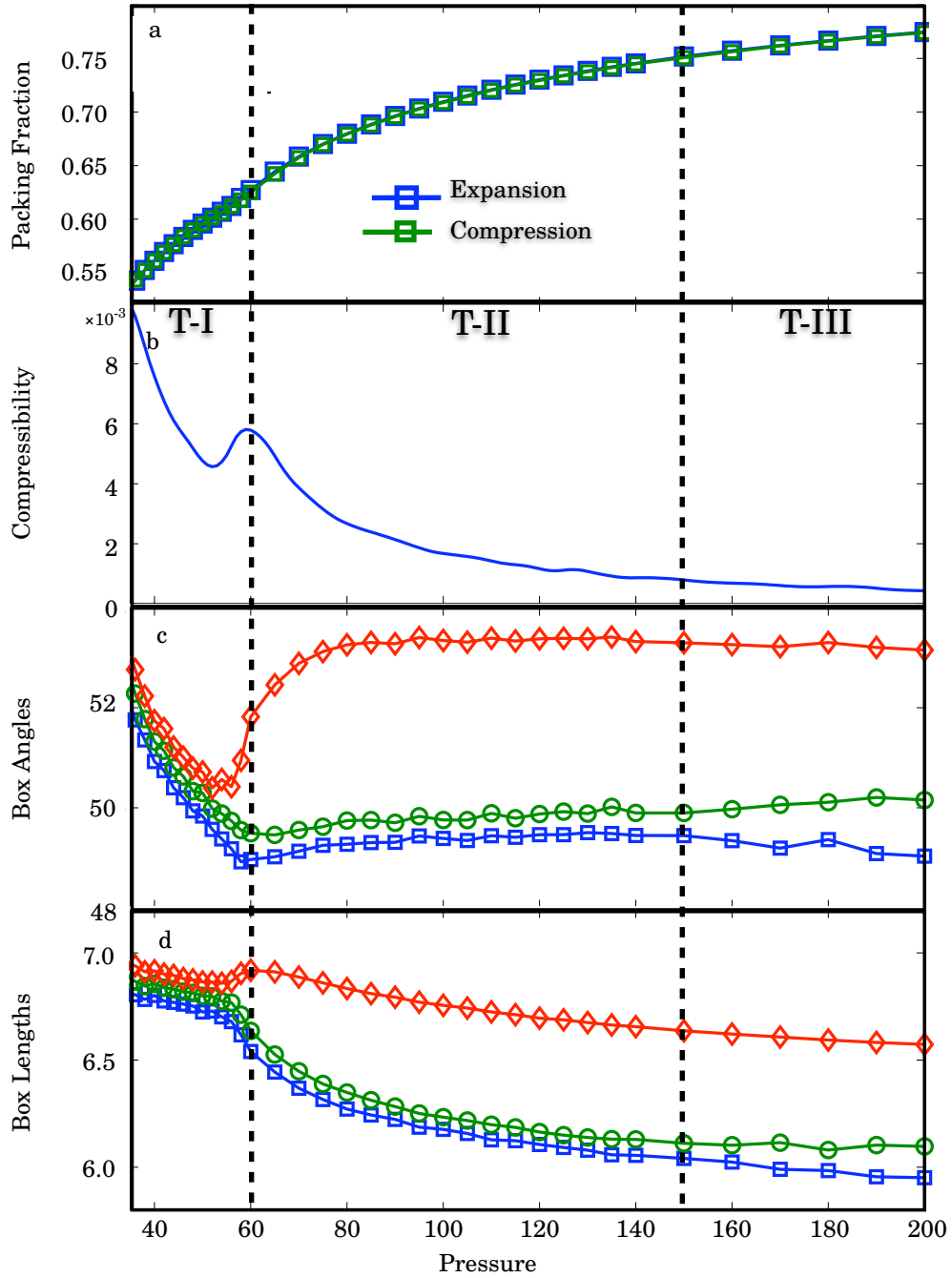
Figure 7.9: **Thermodynamics of the TBP crystal.** (a) Equation of state obtained by expanding the analytically constructed dense crystal, and recompressing the low-density crystal after proper relaxation. Compressibility (b), sorted box angles (c) and dimensions (d) vs. pressure.
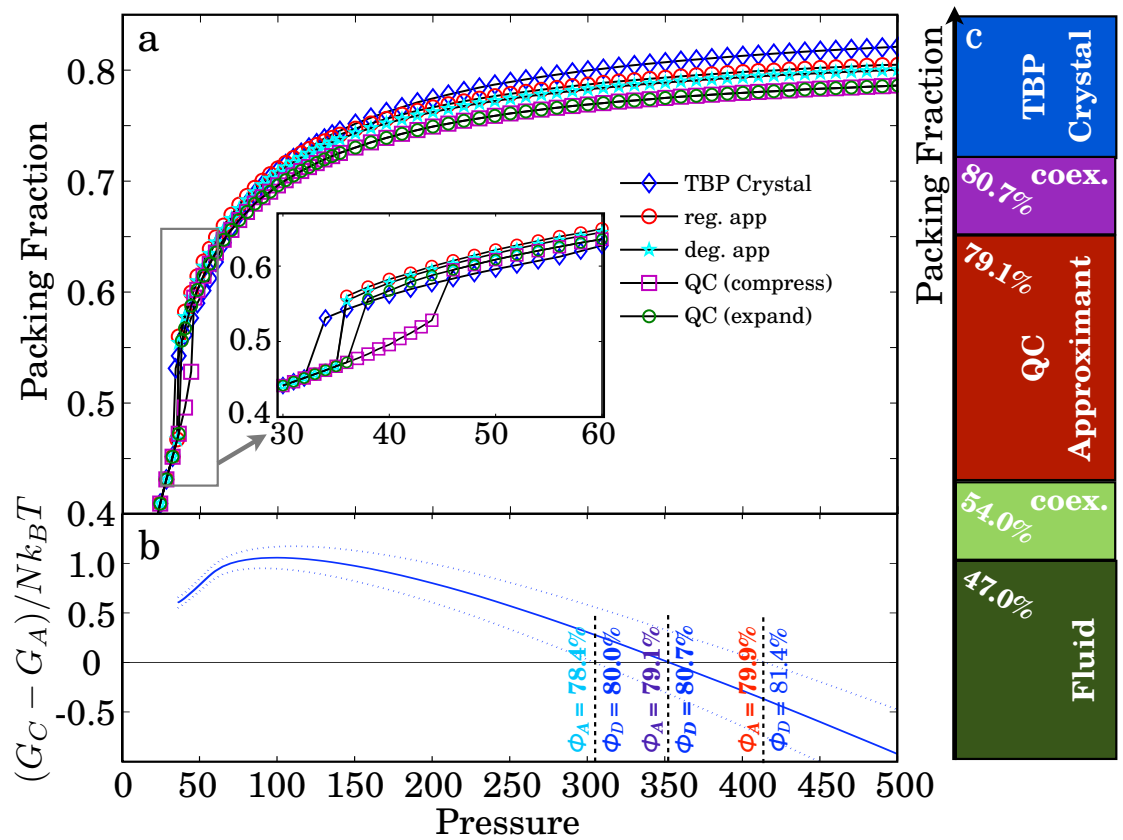
Figure 7.10: **Thermodynamics and the phase diagram of the hard TBP system**. (a) Equation of state for the TBP crystal, the degenerate quasicrystal, the regular and the degenerate approximants. (b) The free energy difference between the TBP crystal and the approximant. (c) Equilibrium phases of hard TBPs. (Images reproduced from [21])

$t = 1/4$ $t = 1/2$ $t = 2/3$ $t = 1$
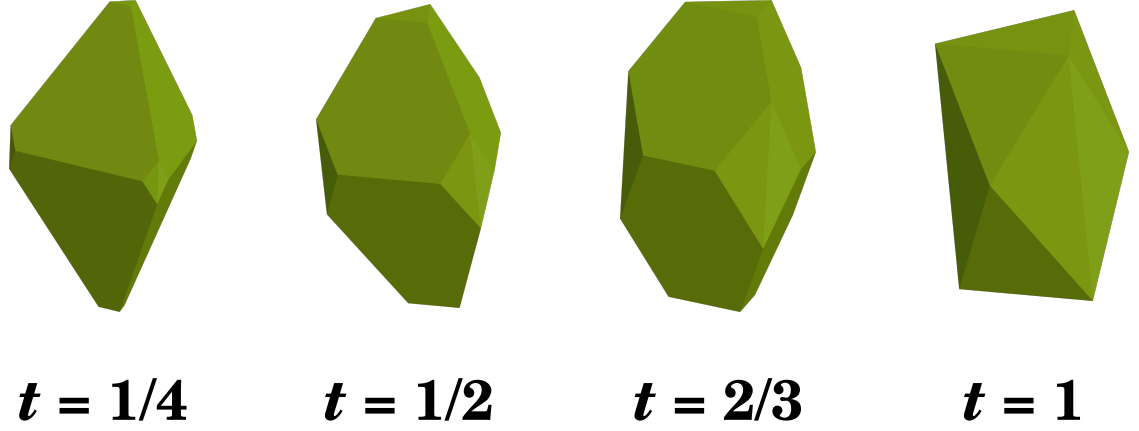
Figure 7.11: **Truncated triangular bipyramids for different values of t.**

results.

Let $\mathcal{P}$ be a perfect (non-truncated) triangular bipyramid centered at the origin and having the vertices:

$$\mathbf{v}_1 = \left(\frac{1}{\sqrt{3}}, 0, 0\right) \tag{7.1a}$$

$$\mathbf{v}_{2,3} = \left(-\frac{1}{2\sqrt{3}}, \pm\frac{1}{2}, 0\right) \tag{7.1b}$$

$$\mathbf{v}_{4,5} = \left(0, 0, \pm\frac{\sqrt{6}}{3}\right) \tag{7.1c}$$

A truncated TBP with the truncation t is then defined as the convex hull of $\{\mathbf{w}_{ij}\}_{i,j=1}^{5}$ with $\mathbf{w}_{ij}$'s given by:

$$\mathbf{w}_{ij}(t) \;=\; \mathbf{v}_i + \frac{1}{2}\left(\mathbf{v}_j - \mathbf{v}_i\right)t \tag{7.2}$$

with $1 \leq i, j \leq 5$. The volume of this truncated TBP (tTBP) is given by:

$$V(t) \;=\; V_p\left[1 - \frac{3t^3}{8}\right] \tag{7.3}$$

where $V_p = \sqrt{2}/6$ is the volume of a perfect non-truncated TBP. Fig. 7.11 depicts truncated TBPs for different values of t.

In this study, we carry out two types of simulations. The densest packings of hard tTBPs are obtained from isotension simulations of small unit cells i.e. boxes
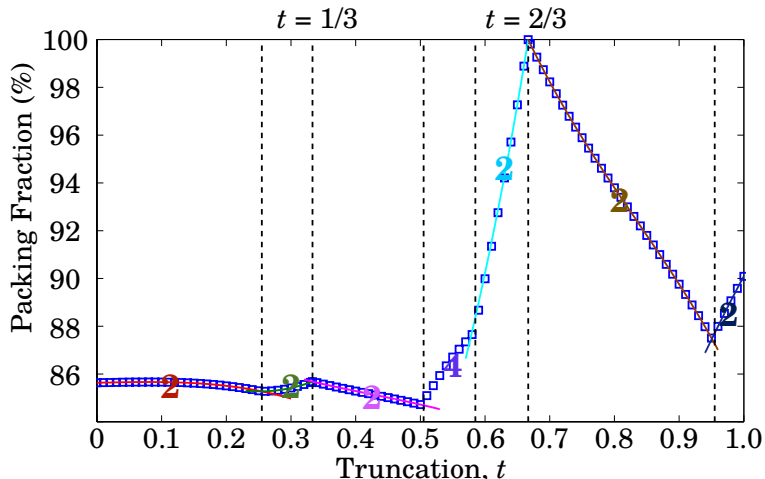
Figure 7.12: $\phi_{\max}(t)$ **vs.** t **for dense packings of truncated TBPs.** The symbols are the maximum packing densities obtained from Monte Carlo simulations while the curves are calculated from the analytical expressions given in Appendix E.

with as many as four particles. Each of these simulations start at a very low pressure of $P^* = PV(t)/k_B T = 1$. The external pressure is then exponentially increased to $P^* = 10^7$. Up to a hundred replica are performed for each truncation and the densest packing is recorded. Self-assembly simulations are however all performed in the isochoric ensemble for systems of $N = 2{,}048$ particles quickly compressed to a packing fraction of 70%. These simulations are performed at $\phi = 55 - 69\%$.

7.6.1 Dense Packings

We perform isotension compressions of small unit cells for $0 \le t \le 1$ and observe that $\phi_{\max}(t)$ changes continuously with t, however there are six kinks in the $\phi_{\max}(t)$ vs. t curve that separate eight distinct families of packings (Fig. 7.12). All these packings have $\phi_{\max}$'s that exceed 84%, which shows that truncated TBPs are efficient packers. In particular, the tTBP corresponding to $t = \dfrac{2}{3}$ is a space-filling polyhedron, previously indexed as 14-III by Goldberg [245].

Six of these eight packings have two particles per unit cell, and can be constructed using the analytical formulae given in Appendix E[5]. Each is comprised of alternate lay-

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[5]These analytical formulae are calculated by Dr. Elizabeth R. Chen, using the approach given in [31].

ers of particles that are related to one another by inversion. These packings however differ on the types of contacts between neighboring particles. For small truncations ($0 \leq t \leq 0.25$), the packing is structurally similar to the TBP crystal (Fig. 7.2b), slightly sheared to optimize the packing of slightly truncated TBPs. In this packing the triangular tips of tTBPs are not adjacent to one other, instead they touch the hexagonal faces of the particles in the alternate layer (Fig. 7.13b). In the next two families of packings, that are observed for $0.25 \leq t \leq \frac{1}{3}$ and $\frac{1}{3} \leq t \leq 0.51$ respectively, triangular tips of particles in opposite layers touch each other, creating a star-like pattern (Fig. 7.13d), while in the two families of packings observed for $0.59 \leq t \leq \frac{2}{3}$ and $\frac{2}{3} \leq t \leq 0.95$, the triangular tips of particles in identical layer touch one another (Fig. 7.13f). For truncations beyond 0.95, a structure is observed wherein the identical layers do not touch one another at all, and triangular tips of particles touch the associated tips on the alternate layers (Fig. 7.13h).

The two families of densest packing for $0.52 \leq t \leq 0.58$ are however completely different (Fig. 7.14b), and have four particles in the unit cell. The basic building element of the packing is a dimer of two tTBPs with inverted orientations, and partially sharing a hexagonal face (Fig. 7.14a). Each unit cell contains two such dimers, rotated with respect to one another. No analytical expression could be obtained for these packings although intersection equations can be solved numerically for arbitrary precision. The numerically-compressed dense packings are given in C.4.

7.6.2 A Space-filling Packing of Truncated TBPs for $t = \frac{2}{3}$

The unit cell of the space-filling packing shown in Fig. 7.13e is spanned by the following vectors:

$$\mathbf{b}_1 = \left(\frac{1}{\sqrt{3}}, -\frac{1}{3}, 0 \right) \tag{7.4a}$$

$$\mathbf{b}_2 = \left(\frac{1}{\sqrt{3}}, \frac{1}{3}, 0 \right) \tag{7.4b}$$

$$\mathbf{b}_3 = \left(0, 0, \frac{4\sqrt{6}}{9} \right) \tag{7.4c}$$

The corresponding particles are related to one another by inversion symmetry. The particle $\mathcal{I}_+$ is obtained from truncating the TBP described by Eq. (7.1) and $\mathcal{I}_-$ is
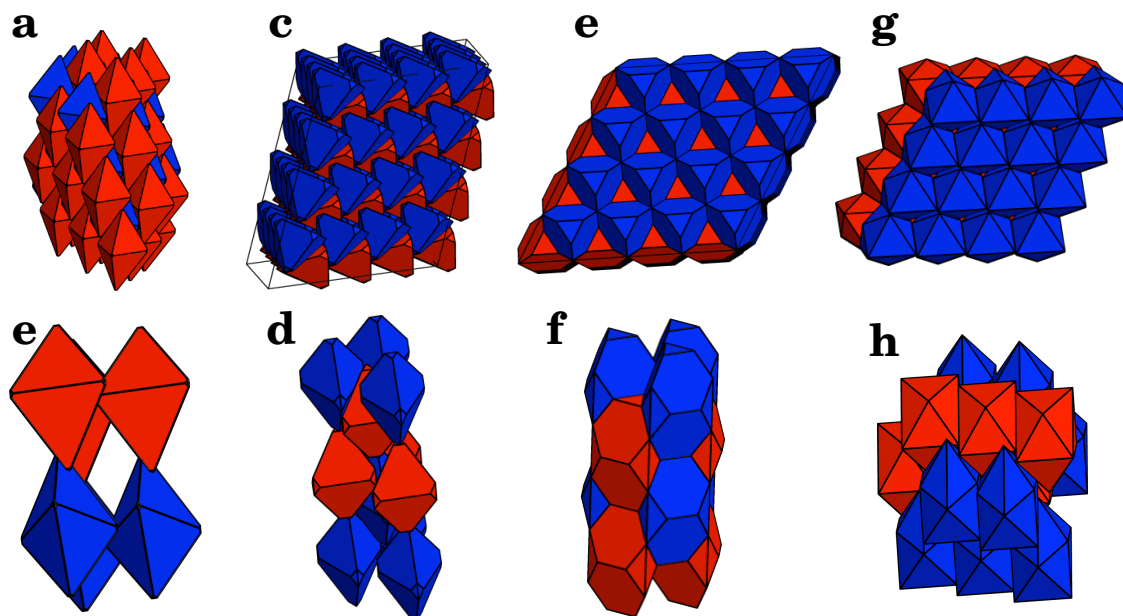
Figure 7.13: **Dense packings of tTBPs with two particles per unit cell.** Densest packings of truncated TBPs for (a) $t = 1/10$, (b) $t = 1/3$, (c) $t = 2/3$ and (d) $t = 1$. All these packings have two particles per unit cell. For $t = 1/10$, the triangular tips of tTBPs touch the hexagonal faces of the particles in the alternate layer. For $t = 1/3$ however, triangular tips of particles in alternate layers kiss one another to create star-like pattens (d). The tTBP with $t = 2/3$ is a space-filling polyhedron. In this packing, tips of particles in identical layers touch one another (f). For $t = 1$ however, layers with identical orientations do not touch at all, and triangular tips partially match the corresponding tips in the alternate layer (h).

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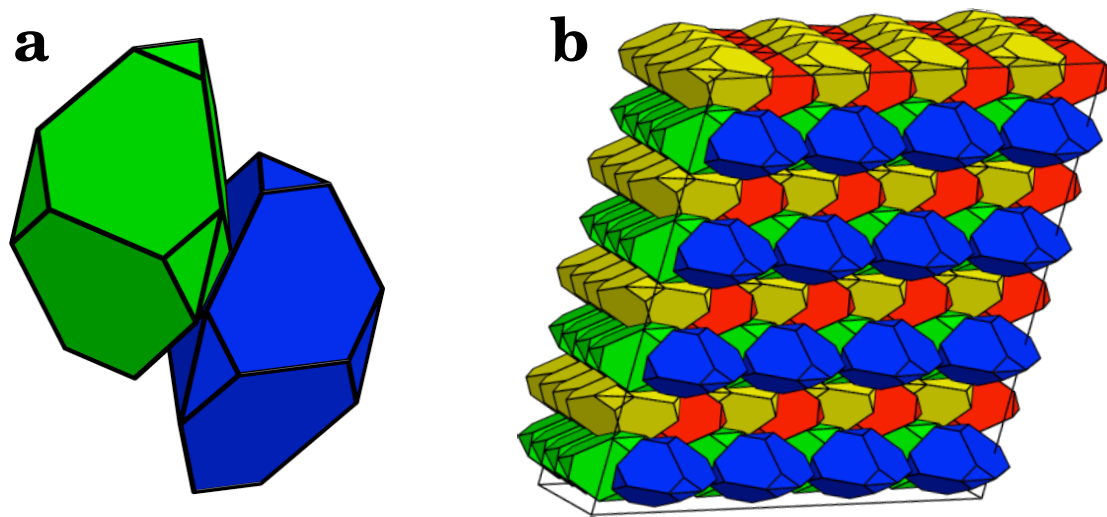
Figure 7.14: **The only dense packing of tTBPs with four particles in the unit cell.** (a) A dimer of tTBPs with $t = 0.55$, which is the basic building blocks in the densest packing given in (b). The blue and green tTBPs belong to the same dimer while the red and yellow particles belong to the other dimer in the unit cell.

Table 7.2:
The properties of the Degenerate Quasicrystal of hard truncated TBPs. $\phi_{\min}$ corresponds to the smallest packing fraction at which the quasicrystal forms from the disordered fluid, $\phi_{\max}$ corresponds to the maximum packing fraction obtained from fast compression of the quasicrystal. (Reproduced from [21])

Truncation	$\phi_{\min}$	MC Cycles	$\phi_{\max}$
0.1	57%	4×10^7	81.21%
0.2	57%	6×10^7	80.93%
0.3	58%	9×10^7	79.74%
0.4	60%	15×10^7	81.31%

obtained by truncating its inverse. The positions of $\mathcal{I}_+$ and $\mathcal{I}_-$ are given by:

$$\mathbf{r}_\pm = \left(\pm\frac{\sqrt{3}}{9}, 0, \pm\frac{\sqrt{6}}{9} \right) \tag{7.5}$$

As far as the centers of individual particles is concerned, this packing can be described as an elongated hexagonally close-packed structure.

7.6.3 Self-Assembly Simulations

The hard tTBP system assembles far fewer ordered structures than the hard truncated tetrahedron system. In particular we observe no ordering for $0.4 < t < 0.7$ and $t > 0.8$. The two ordered phase that form in our self-assembly simulations are the degenerate quasicrystal formed for small truncations ($t \le 0.4$) and the elongated hcp packing for $0.7 \le t \le 0.8$. Except for the degenerate quasicrystal, we do not observe any other ordered phase that is degenerate to the phases formed in the hard truncated tetrahedron system.

Table. 7.2 shows the minimum density and the number of MC cycles needed to form the quasicrystal . As expected, it is far more difficult for the truncated system to form the degenerate quasicrystal than the perfect hard TBP system, and high densities and longer simulation times are needed. Looking at diffraction patterns (Fig. 7.15) and $\phi_{\max}$'s obtained from fast compressions (Table. 7.2) reveals that the degenerate quasicrystal is of very superb structural quality.

The other ordered phase assembles for $t = 0.7$ and $t = 0.8$ at packing fractions beyond 61%. It is structurally similar to the space-filling packing shown in Fig. 7.13d-e, and is not degenerate. Fig. 7.16a shows a snapshot of an NVT simulation of
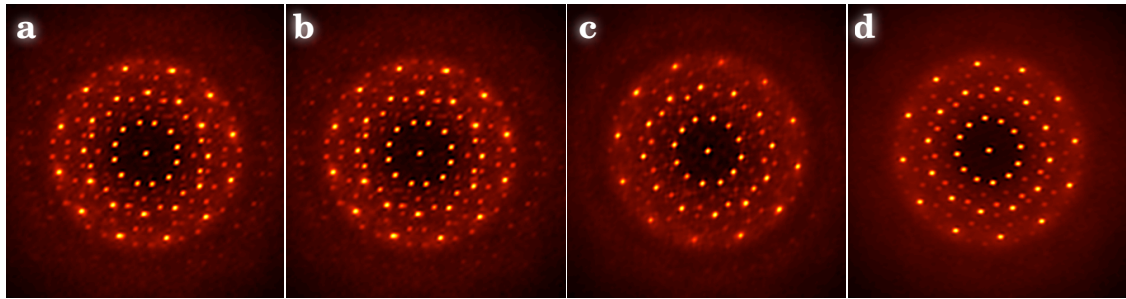
Figure 7.15: **Diffraction images of the degenerate quasicrystals of hard tTBPs**: (a) $t = 0.1$, (b) $t = 0.2$, (c) $t = 0.3$ and (d) $t = 0.4$. Scatters are placed at the centers of constituent truncated tetrahedra within each tTBP.

2,048 truncated TBPs ($t = 0.7$) performed at $\phi = 61\%$, the layers with alternate orientations are shown with yellow and cyan lines respectively. The diffraction image calculated from above the layers, reveals sixfold symmetry which is consistent with hexagonal arrangement of particles in each layer.

7.7 Conclusion

Remarkably, hard TBPs not only prefer a complex quasicrystal over the simpler TBP crystal at intermediate packing fractions, but also form it on timescales comparable to that previously observed in the hard tetrahedron system. This is surprising because, in comparison to tetrahedra, the motion of the highly anisotropic bipyramids is considerably more constrained. Nevertheless, the degeneracy of the quasicrystal helps it form easily in simulation. Random pairing allows TBPs to join existing seeds of the DQC without forming configurations that are kinetically trapped due to incorrect pairing. Particle rearrangements needed for the formation and growth of the seed are also feasible due to the local similarity of the fluid and the quasicrystal [18, 214]. Finally, the degeneracy and the existence of ring-ring and ring-interstitial 'cross-links' adds rigidity to the TBP structures. This means that the TBP system might be superior over the tetrahedron system in terms of its mechanical properties, just as for crystals of hard sphere dimers compared to crystals of their monomers [238].

Despite similarities to the hard tetrahedron system in the types and order of equilibrium phases appearing in the phase diagram, the hard TBP system is different in several ways: All ordered phases melt at higher packing fractions in the hard TBP

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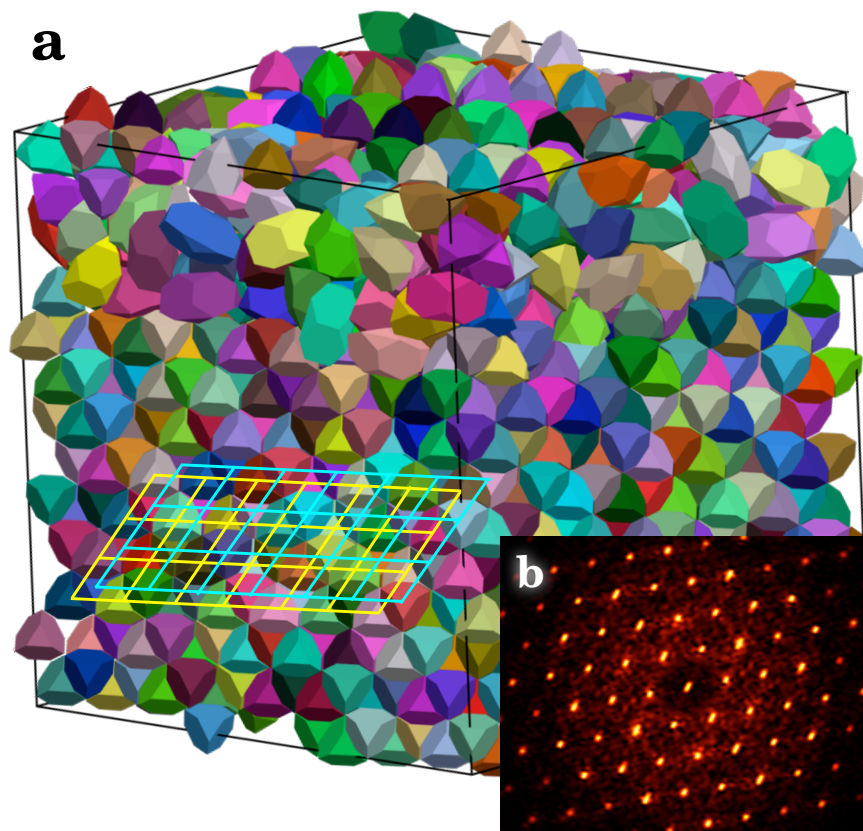
Figure 7.16: **Simulation snapshot of the elongated hexagonally close-packed structure formed in an isochoric simulation of** 2,048 **tTBP** ($t =$ 0.7) **at** $\phi = 61\%$**.** Layers with alternate orientations are shown with yellow and cyan lines respectively. (b) Diffraction image calculated from above the layers.

system. The sequence at which these phases melt upon expansion is also different. Unlike the hard tetrahedron system where the TBP crystal melts earlier, the QC and the approximant tend to melt at higher pressures in the hard TBP system. The approximant is also stable within a narrower range of packing fractions. All these differences can be attributed to reduced stability of ordered phases in the hard TBP system due to a decrease in the degrees of freedom accessible to the system. As expected, the stability of the quasicrystal and its approximants is more adversely affected since tetrahedra are naturally paired into dimers in the dimer crystal in the hard tetrahedron system and pairing them into TBPs does not have any significant effect on the packing and the entropy of the system. But this destabilization is not big enough to discourage the formation of the quasicrystal.

The behavior of the hard truncated TBP system is however completely different from the corresponding hard truncated tetrahedron system both in terms of the densest packings, and the structures that form in self-assembly simulations. It is also noteworthy that the degenerate quasicrystal still forms for truncations up to $t = 0.4$.

In conclusion we have shown that hard triangular bipyramids form a degenerate dodecagonal quasicrystal. Our finding is only the second quasicrystal formed with hard particles, the first reported degenerate quasicrystal, and one of only a few quasicrystals formed in nonatomistic systems. Our results suggest that degenerate phases are not restricted to simple close-packed crystals and might be common in dimer systems.

CHAPTER VIII

Conclusion and Outlook

8.1   Summary of Key Findings

In this dissertation we studied the thermodynamics of hard tetrahedra and hard triangular bipyramids– dimers of hard tetrahedra. Both these building blocks are simple polyhedra, and have the same densest packing which is a simple crystal with four (two) particles in the unit cell and $\phi = \dfrac{4000}{4671} \approx 85.63\%$. Yet, they exhibit a nontrivial phase behavior by preferring complex quasiperiodic arrangements over this densest packing at intermediate densities. This shows that shape and entropy alone is sufficient for promoting complex phases and unusual phase behaviors.

The dodecagonal quasicrystal formed by hard tetrahedra is the first quasicrystal observed in hard particle systems. It forms at $\phi \approx 50\%$ and its basic structural motifs are twelvefold rings capped with pentagonal dipyramids that sit at the vertices of a planar tiling of square, triangles and rhombs. Additional interstitial tetrahedra fill the voids at the centers of squares and triangles. The quasicrystal can also be understood as a network of interpenetrating PDs, a fact that is quantitatively verified. We also observe this network in the disordered fluid prior to crystallization, a finding that suggests the possibility of a second transition from a simple fluid in which particles move independently, to a complex fluid where particles move collectively within the network.

The quasicrystal and its approximants pack very efficiently and can be compressed to very high packing fractions. In particular, the $(4.3^2.4.3)$ approximant can be compressed to $\phi = 85.03\%$, which was the world record at the time of its discovery, and we believe that it is the most stable form of the quasicrystal-like phases observed in our simulations. The disordered complex fluid is also relatively dense, and can be packed to a packing fraction of $\phi = 78.56\%$, which to date is still the densest

known disordered arrangement of tetrahedra. It also exceeds all dense packings of tetrahedra– ordered or otherwise– reported prior to the start of this project.

Using free energy calculation, we show that the $(3.4.3^2.4)$ approximant of the quasicrystal formed in our simulations is thermodynamically more stable than the simple dimer crystal for $\phi \leq 84\%$. The thermodynamic transition from the approximant to the dimer crystal does not involve any symmetry breaking. This is in contrast to most hard particle systems where the densest known packing can be obtained by a series of symmetry-breaking first- and second-order transitions.

In order to understand why the approximant is stable for such a wide range of densities, we performed free volume calculations, and observed that the mean-log average free volume grows more quickly in the approximant than in the dimer crystal. Furthermore, tetrahedra were observed to move collectively in both the quasicrystal and the approximant. Such correlated motions occurred in the form of PD rotations at moderately high densities and led to particle rearrangements, but were still present at dense packings and manifested themselves in correlated vibrations of groups of particles, including PDs.

The thermodynamic behavior of hard triangular bipyramids– dimers of tetrahedra– is qualitatively similar. They also form the same quasicrystal– in the level of monomers – and the quasicrystal approximant is still more stable than the dimer crystal at intermediate packings. The way that tetrahedra are paired into dimers is however random, which adds some randomness to the way any particular tile (e.g. square, triangle) can be decorated with triangular bipyramids. We are the first to observe such degeneracy in any quasicrystal, and therefore call the structure a degenerate quasicrystal. This degeneracy should however not be confused with the well-known notion of degeneracy associated with random tilings and phason flips.

8.2   Future Work

Like any other endeavor in science, the effort presented in this thesis poses more questions than it manages to answer. In this section, we pose some of these impending questions that might one day be addressed by future generations of scientists.

1. **How does the quasicrystal form?** The fluid and the quasicrystal have very similar local configurations as particles are a part of a PD network in both structures. It is conceivable that the formation of the quasicrystal can proceed through some subtle changes in the PD network. This speculation is in

line with recent simulation studies of model systems revealing that a quasiperiodic nucleus grows by incorporating into itself kinetically trapped particles by minimal rearrangements [214]. The exact mechanism is however unknown for the quasicrystal formed by hard tetrahedra and can be determined by enhanced sampling techniques such as transition path sampling [246] or forward flux sampling [247].

2. **How robust is the quasicrystal formation?** Our initial reporting of the quasicrystal was in a system of perfect monodisperse tetrahedra and TBPs. However real experimental systems are not perfect. For instance colloidal tetrahedra might not be completely regular, they might also be polydisperse, or with rough edges and faces. It is therefore of practical relevance to understand the effect of these imperfections of the quasicrystal formation. We have studied some imperfections e.g. polydispersity and truncation in this dissertation. The remainder is being studied by another member of the Glotzer Group and will be presented in a separate publication.

3. **Physical properties of the quasicrystal:** Quasicrystals have been shown to exhibit distinct, and potentially interesting, mechanical [248], electrical and optical properties [249]. It is therefore interesting to study the physical properties of the quasicrystal formed by hard tetrahedra. Particularly, physical properties of the degenerate quasicrystal can in principle deviate from those of the original tetrahedron-based quasicrystal. Identifying those differences can help us understand the practical importance of pairing degeneracy in dimer systems.

4. **Estimation of pairing entropy in the degenerate quasicrystal:** If different ways of pairing monomers into dimers have the same maximum packing fraction, the problem of identifying the pairing entropy is essentially a combinatorics problem, which is easy to solve for square or triangular lattices. In the case of a complex network such as a quasicrystal, this enumeration is less trivial. Furthermore, since not all configurations corresponding to different pairings have the same $\phi_{\max}$ in the hard tetrahedron system. It is therefore interesting to identify the types of networks for which an analytical– or an asymptotically analytical– solution exists for the pairing entropy. Developing efficient numerical techniques for calculating pairing entropy in nontrivial networks is also useful.

5. **Possibility of degenerate phases in other systems:** As mentioned in Chap-

ter VII, the notion of degeneracy might be more commonplace than originally thought. It will therefore be interesting to study other systems of dimers of soft and hard monomers to explore the possibility of a new class of degenerate phases. Such structures might, for instance, be optically identical to the original monomer-based ordered structure, but yet mechanically stronger. It will also be informative to study how various physical properties are affected by introducing pairing degeneracy.

APPENDICES

APPENDIX A

Limiting Behavior of Jarzynski's Equality for $\tau \to 0$ and $\tau \to \infty$

A.1 Limiting Behavior of Jarzynski's Equality for $\tau \to 0$ and $\tau \to \infty$

Let the Hamiltonian of the system evolve according to $\mathcal{H}(t)$ so that $\mathcal{H}(0) = \mathcal{H}_0$ and $\mathcal{H}(\tau) = \mathcal{H}_1$. According to Jarzynski's equality 3.17:

$$\exp\left[-\beta\Delta A\right] = \langle\exp\left[-\beta\mathcal{W}\left(\tau\right)\right]\rangle_0 \;\; = \;\; \left\langle\exp\left[-\beta\int\limits_0^\tau \frac{d\mathcal{H}(t)}{dt}dt\right]\right\rangle_0 \qquad \text{(A.1)}$$

For a finite τ, Eq. A.1 takes the form:

$$\Delta A = -\beta^{-1}\ln\left\langle\exp\left\{-\beta\left[\mathcal{H}_1\left(\mathbf{x}^{(1)}(\tau),\mathbf{p}^{(1)}(\tau)\right) - \mathcal{H}_0\left(\mathbf{x}^{(0)}(0),\mathbf{p}^{(0)}(0)\right)\right]\right\}\right\rangle \qquad \text{(A.2)}$$

If $\tau \to \infty$, the transformation will be so slow that the process will essentially be *reversible* and the system will be in equilibrium for every finite t. The only difference with thermodynamic integration will be the fact that here, all samples are taken from system 0, but one should remember that the initial equilibrium configuration taken from system 0 always remains in local equilibrium for $\tau \to \infty$ and the equivalence with thermodynamic integration is established.

For $\tau \to 0$, the transformation is almost instantaneous, so the initial configuration taken from system 1 does not have enough time to evolve, and thus $\mathbf{x}^{(1)}(\tau) = \mathbf{x}^{(0)}(0)$

and $\mathbf{p}^{(1)}(\tau) = \mathbf{p}^{(0)}(0)$. This is exactly equivalent to Free Energy Perturbation (FEP) methods performed using Eq. 3.12.

APPENDIX B

Derivation Details of Theorems and Lemmas of Chapter IV

B.1 Derivation Details of Tetrahedral SOC

We need to solve the following optimization problem:

$$
\begin{aligned}
\text{maximize} \quad & \sum_{l,m=1}^{4} \xi_{lm}^3 \\
\text{subject to} \quad & \sum_{m=1}^{4} \xi_{lm}^2 = \tfrac{4}{3} \quad l = 1, 2, 3, 4 \\
& \sum_{m=1}^{4} \xi_{lm} = 0 \quad l = 1, 2, 3, 4
\end{aligned}
\tag{B.1}
$$

The form of (B.1) suggests that we can solve it by breaking into four independent problems of the form:

$$
\begin{aligned}
\text{maximize} \quad & \sum_{i=1}^{4} x_i^3 \\
\text{subject to} \quad & \sum_{i=1}^{4} x_i = 0 \\
& \sum_{i=1}^{4} x_i^2 = \tfrac{4}{3}
\end{aligned}
\tag{B.2}
$$

We solve this problem by enumerating all solutions of Karush-Kuhn-Tucker criteria [250, 251] and show that global maximum is attained if x is a permutation of $(1, -\frac{1}{3}, -\frac{1}{3}, -\frac{1}{3})$. The Lagrangian for (B.2) is:

$$
\mathcal{L}(x, \nu, \mu) = \sum_{i=1}^{4} x_i^3 + \nu \sum_{i=1}^{4} x_i + \mu \sum_{i=1}^{4} \left(x_i^2 - \frac{4}{3} \right)
\tag{B.3}
$$

$$
\frac{\partial \mathcal{L}}{\partial x_j} = 3x_j^2 + \nu + 2\mu x_j
\tag{B.4}
$$

KKT conditions requires that $\nabla_x \mathcal{L} = 0$. Eliminating μ one easily obtains:

$$\nu = -\frac{3}{4}\sum_{i=1}^{4} x_i^2 = -1 \tag{B.5}$$

Using this, one can obtain x_i as:

$$x_i = \frac{-\mu \pm \sqrt{\mu^2 + 3}}{3} \tag{B.6}$$

Denote the roots of this equation with ξ_+ and ξ_- and let $n_+(n_-)$ be the number of x_i's equalling $\xi_+(\xi_-)$. Using $n_+\xi_+ + n_-\xi_- = 0$ one gets:

$$\mu = \frac{3m}{\sqrt{3(16 - m^2)}} \tag{B.7}$$

$$\xi_\pm = -\frac{m \mp 4}{\sqrt{3(16 - m^2)}} \tag{B.8}$$

where $m = n_+ - n_-$. The global maximum is thus obtained for $m = -2, \xi_+ = 1, \xi_- = -\frac{1}{3}$, for which $\sum_{i=1}^{4} x_i^3 = \frac{8}{9}$. The global maximum of (B.1) is therefore $\frac{32}{9}$.

B.2 Derivation Details of Octahedral SOC

Letting $\zeta_{pq} = \xi_{pq}^2$, we need to solve the following optimization problem:

$$\begin{aligned}
\text{max} \quad & \textstyle\sum_{p,q=1}^{3} \zeta_{pq}^2 \\
\text{subject to} \quad & \textstyle\sum_{p=1}^{3} \zeta_{pq} = 1 \quad q = 1, 2, 3 \\
& \zeta_{pq} \geq 0 \qquad p, q = 1, 2, 3
\end{aligned} \tag{B.9}$$

Observe that (B.9) can be broken into three independent and yet identical optimization problems of the form:

$$\begin{aligned}
\text{max} \quad & \textstyle\sum_{i=1}^{3} x_i^2 \\
\text{subject to} \quad & \textstyle\sum_{i=1}^{3} x_i = 1 \\
& x_i \geq 0, i = 1, 2, 3
\end{aligned} \tag{B.10}$$

which is solved by identifying x's that satisfy Karush-Kuhn-Tucker solutions criteria. The Lagrangian is given by:

$$\mathcal{L} = \sum_{i=1}^{3} x_i^2 - \sum_{i=1}^{3} \nu_i x_i - \lambda \sum_{i=1}^{3} x_i \tag{B.11}$$

$$\frac{\partial \mathcal{L}}{\partial x_j} = 2x_j - \nu_j x_j - \lambda x_j = 0 \tag{B.12}$$

which yields $x_j = (\nu_j + \lambda)/2$. Let $n \leq 3$ be the number of nonzero x_j's. For each such x_j, $\nu_j = 0$ due to complementary slackness. We thus have $\lambda = \frac{2}{n}$ and $x_j = \frac{1}{n}$ for nonzero x_j's. Choosing $n = 1$ maximizes the objective function with $\sum_{i=1}^{3} x_i^2 = 1$. The global maximum for (B.9) is therefore 3 with ξ_{pq}'s being a permutation of $(\pm 1, 0, 0)$ for each q.

B.3 Derivation Details of Icosahedral SOC

We need to solve the following optimization problem:

$$\begin{array}{ll}
\text{max} & \sum_{p,q=1}^{6} \xi_{p,q}^3 \\
\text{subject to} & \sum_{p=1}^{6} \xi_{pq} = 2 \quad q = 1, \cdots, 6 \\
& \sum_{p=1}^{6} \xi_{pq}^2 = \frac{6}{5} \quad q = 1, \cdots, 6 \\
& \xi_{pq} \geq 0 \qquad p, q = 1, \cdots, 6
\end{array} \tag{B.13}$$

which can be broken into six independent and yet identical optimization problems of the form:

$$\begin{array}{ll}
\text{max} & \sum_{i=1}^{6} x_i^3 \\
\text{subject to} & \sum_{i=1}^{6} x_i^2 = \frac{6}{5} \\
& \sum_{i=1}^{6} x_i = 2 \\
& x_j \geq 0 \qquad j = 1, 2, \cdots, 6
\end{array} \tag{B.14}$$

In order to solve (B.14), we first solve the following optimization problem:

$$\begin{array}{ll}
\text{max} & \sum_{i=1}^{n} x_i^3 \\
\text{subject to} & \sum_{i=1}^{n} x_i^2 = \frac{6}{5} \\
& \sum_{i=1}^{n} x_i = 2
\end{array} \tag{B.15}$$

using Lagrange multipliers. The Lagrangian is given by:

$$\mathcal{L}(x_1, \cdots, x_n; \lambda, \mu) = \sum_{i=1}^{n} x_i^3 - \lambda \left[\sum_{j=1}^{n} x_j^2 - \frac{6}{5} \right]$$
$$- \mu \left[\sum_{i=1}^{n} x_i - 2 \right]$$

which can be differentiated to get:

$$\frac{\partial \mathcal{L}}{\partial x_j} = 3x_j^2 - 2\lambda x_j - \mu = 0 \tag{B.16}$$

Summing over j yields $\mu = \frac{1}{n} \left[\frac{18}{5} - 4\lambda \right]$. The roots of the quadratic equation (that we denote by ξ_+ and ξ_-) are thus given by:

$$\xi_{\pm} = \frac{1}{3} \left[\lambda \pm \sqrt{\lambda^2 + \frac{3}{n} \left(\frac{18}{5} - 4\lambda \right)} \right] \tag{B.17}$$

Let $n_+(n_-)$ be the number of x_j's being equal to $\xi_+(\xi_-)$ and let $m = n_+ - n_-$. From $n_+ \xi_+ + n_- \xi_- = 2$ we have:

$$\lambda = \frac{6}{n} \left[1 - m \sqrt{\frac{3n - 10}{10(n^2 - m^2)}} \right] \tag{B.18}$$

$$\xi_{\pm} = \frac{2}{n} \left[1 - (m \mp n) \sqrt{\frac{3n - 10}{10(n^2 - m^2)}} \right] \tag{B.19}$$

which suggests that the problem is not feasible for $n < 4$. The value of the objective function is given by:

$$n_+ \xi_+^3 + n_- \xi_-^3 = \frac{36}{5n} - \frac{16}{n^2} - \frac{8m(3n - 10)}{5n^2} \sqrt{\frac{3n - 10}{10(n^2 - m^2)}} \tag{B.20}$$

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Since $d[m/\sqrt{n^2 - m^2}]/dm = n^2/\sqrt{(n^2 - m^2)^3} > 0$, the function is maximized for the *smallest* possible m i.e. $m = 2 - n$ and the global maximum is given by:

$$f(n) = \frac{36}{5n} - \frac{16}{n^2} + \frac{4(n-2)(3n-10)}{5n^2}\sqrt{\frac{3n-10}{10(n-1)}}$$

(B.21)

Now we solve (B.14) by identifying x's that satisfy the Karush-Kuhn-Tucker criteria. Its Lagrangian is of the form:

$$\mathcal{L} = \sum_{i=1}^{6} x_i^3 - \nu_i x_i - \lambda \left[\sum_{i=1}^{n} x_i^2 - \frac{6}{5}\right] - \mu \left[\sum_{i=1}^{n} x_i - 2\right]$$

$$\frac{\partial \mathcal{L}}{\partial x_j} = 3x_j^2 - \nu_j - 2\lambda x_j - \mu$$

It can be easily shown that for every $x_j = 0$, $\nu_j = -\mu$ and is independent of j. For $x_j \neq 0$, complementary slackness implies that $\nu_j = 0$ and $\partial \mathcal{L}/\partial x_j = 0$ implies $\mu \neq 0$. Note that non-zero x_j's are amongst the Karush-Kuhn-Tucker solutions of (B.15) for $n = 6 - n_0$ with n_0 being the number of x_j's that are zero. Since $f(4) < f(5) < f(6)$ from Eq. (B.21), $n_0 = 0$ gives the global maximum with $\sum_{i=1}^{6} x_i^3 = \frac{26}{25}, \xi_+ = 1, \xi_- = \frac{1}{5}$. The value of the objective function of (B.13) is thus $\frac{156}{25}$ which can be *only* achieved for ξ_{pq}'s being permutations of $(1, \frac{1}{5}, \frac{1}{5}, \frac{1}{5}, \frac{1}{5}, \frac{1}{5})$.

B.4   Some Useful Trigonometric Integrals

We are interested in integrals of the form:

$$I_{m,n} = \int_0^{2\pi} \cos^m\theta \sin^n\theta \, d\theta$$

(B.22)

$$J_{m,n} = \int_0^{\pi} \cos^m\theta \sin^n\theta \, d\theta$$

(B.23)

Note that $I_{2m-1,2n} = I_{2m,2n-1} = 0$ since the integrand is odd around $\theta = \pi$. $I_{2m-1,2n-1} = 0$ since:

$$I_{2m-1,2n-1} = 2\int_0^{\pi} \cos^{2m-1}\theta \sin^{2n-1}\theta \, d\theta = 0$$

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since the integrand is odd around $\theta = \frac{\pi}{2}$. $I_{2m,2n}(m, n > 0)$ can be calculated by considering the identity:

$$I_{2m,0} = \frac{2\pi(2m)!}{4^m(m!)^2} \tag{B.24}$$

and integration by part:

$$\int_0^{2\pi} \cos^{2m}\theta \sin^{2n}\theta d\theta = \left[\frac{\cos^{2m-1}\theta \sin^{2n+1}\theta}{2n+1}\right]_0^{2\pi}$$

$$+ \frac{2m-1}{2n+1} \int_0^{2\pi} \cos^{2m-2}\theta \sin^{2n+2}\theta d\theta$$

$$= \frac{2m-1}{2n+1} I_{2m-2,2n+2}$$

$$= \frac{2\pi(2m)!(2n)!}{4^{m+n}m!n!(m+n)!} \tag{B.25}$$

(B.24) can be proven as follows:

$$I_{2m,0} = \int_0^{2\pi} \cos^{2m}xdx = \int_0^{2\pi} \cos^{2m-2}x(1-\sin^2 x)dx$$

$$\stackrel{(a)}{=} I_{2m-2,0} - \left\{\left[-\frac{\cos^{2m-1}x \sin x}{2m-1}\right]_0^{2\pi}\right.$$

$$\left. + \frac{1}{2m-1} \int_0^{2\pi} \cos^{2m}xdx\right\}$$

$$I_{2m,0} = \frac{2m-1}{2m} I_{2m-2,0} = I_{0,0} \prod_{k=1}^{m} \frac{2k-1}{2k} \stackrel{(b)}{=} \frac{(2m)!}{2^{2m}(m!)^2} 2\pi$$

(a) follows from integration by part by setting $u = \sin x, dv = \cos^{2m-1}x \sin xdx$ and (b) follows from $I_{0,0} = \int_0^{2\pi} dx = 2\pi$. Note that $J_{2m+1,n} = 0$ since the integrand is odd around $\pi/2$. For $m = 2p, n = 2q$ both even we have:

$$J_{2p,2q} = \int_0^{\pi} \cos^{2p}x \sin^{2q}xdx = \frac{1}{2}I_{2p,2q} \tag{B.26}$$

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We also have:

$$J_{2m,1} = \int_0^\pi \cos^{2m} x \sin x dx = \left. \frac{-\cos^{2m+1} x}{2m+1} \right|_0^\pi = \frac{2}{2m+1}$$

(B.27)

Similar to what was done for $I_{2m,2n}$, using integration by part we have:

$$
\begin{aligned}
J_{2m,2n+1} &= \frac{2n}{2m+1} J_{2m+2,2n-1} \\
&= \frac{2n}{2m+1} \frac{2n-2}{2m+3} \cdots \frac{2}{2m+2n-1} \frac{2}{2m+2n+1} \\
J_{2m,2n+1} &= \frac{2^{2n+1} n! (2m)! (m+n)!}{m! (2m+2n+1)!}
\end{aligned}
$$

(B.28)

B.5   Equivalency of Rigidity Constraints for the Regular Tetrahedron

We need to show that the following set of constraints are equivalent:

$$\sum_{p=1}^4 u_p^i u_p^j = \frac{4}{3} \delta^{ij}$$

(B.29a)

$$\sum_{p=1}^4 u_p^i = 0$$

(B.29b)

$$u_p^i u_q^i = \frac{4}{3} \delta_{pq} - \frac{1}{3}, \, p,q = 1,2,3,4$$

(B.30)

To prove that (B.29) implies (B.30), multiply (B.29a) by u_q and observe that:

$$\sum_{p=1}^4 (u_p^T u_q) a_p = \frac{4}{3} u_q$$

which, after some rearrangement takes the form:

$$\sum_{p \neq q} \left[\frac{4}{3} + \xi_{pq} - \xi_{pp} \right] u_p = 0$$

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with $\xi_{pq} = a_p^T a_q$. However since $\sum_{p=1}^4 u_p u_p^T$ is a full-rank matrix, any three of four u_p's are linearly independent and $\xi_{qq} - \xi_{pq} = \frac{4}{3}$. We also have $\left\|\sum_{p=1}^4 u_p u_p^T\right\|_F^2 = 16/3$ which yields $\xi_{qq} = 1, \xi_{pq} = -\frac{1}{3}(p \neq q)$ and (B.30) follows.

To prove (B.29) from (B.30) take an arbitrary set of vectors satisfying (B.30) and show that (B.29) holds; however since 0 and δ^{ij}'s are isotropic tensors, they are invariant under any unitary transformation and the proof follows.

B.6 Derivation Details of Uniaxial Nematics Order Parameter

The objective function of the associated optimization problem- i.e. $\mathfrak{M}_\Omega^{ijkl}\overline{M}^{ijkl}$- can be simplified by observing that $\overline{M}$ is both invariant under index permutation and traceless. More specifically:

$$
\begin{aligned}
\overline{M}^{ijkl}(z^i x^j x^k z^l + z^i y^j y^k z^l) &= \overline{M}^{ilkj} z^i z^l (x^k x^j + y^k y^j) \\
&= \overline{M}^{ijkl} z^i z^j (\delta^{kl} - z^k z^l)
\end{aligned}
$$

The same thing can be done for the six terms containing $\mathbf{z}$, $\mathbf{x}$ and $\mathbf{y}$. With a similar argument one can verify that:

$$
\overline{M}^{ijkl}(x^i x^j x^k x^l + x^i y^j x^k y^l) = \overline{M}^{ijkl} x^i x^j (\delta kl - z^k z^l)
$$

The objective function can therefore be written as:

$$
\begin{aligned}
\mathfrak{M}_\Omega^{ijkl}\overline{M}^{ijkl} &= \sum_{q=1}^4 \left[\alpha_q^4 z^i z^j z^k z^l + 3\alpha_q^2\beta_q^2 z^i z^j(\delta^{kl} - z^k z^l) + \frac{3\beta_q^4}{8}(\delta^{ij} - z^i z^j)(\delta^{kl} - z^k z^l)\right]\overline{M}^{ijkl} \\
&= \omega z^i z^j z^k z^l \overline{M}^{ijkl}
\end{aligned}
$$

APPENDIX C

Numerically-Compressed Dense Packings

C.1 Data Format

The numerically compressed dense packings introduced in this thesis are presented here. The data format used in this appendix is the `.pos` file format used in `injavis`, the visualization software developed by Dr. Michael Engel in Glotzer's Group and used for visualization purposes in this thesis. The vectors spanning the simulation box are given by the identifier `boxMatrix`. The line:

```
boxMatrix b11 b12 b13 b21 b22 b23 b31 b32 b33
```

corresponds to the box spanned by the vectors $\mathbf{b}_1 = (b_{11}, b_{21}, b_{31}), \mathbf{b}_2 = (b_{12}, b_{22}, b_{32})$ and $\mathbf{b}_3 = (b_{13}, b_{23}, b_{33})$. A convex polyhedron is defined by the identifier `def`:

```
def TAG "poly3d N v_1x v_1y v_1z ⋯ v_Nx v_Ny v_Nz "
```

where N is the number of vertices of the convex polyhedron, and $\mathbf{v}_1 = (v_{1x}, v_{1y}, v_{1z})$, $\cdots$, $\mathbf{v}_N = (v_{Nx}, v_{Ny}, v_{Nz})$ are its vertices. For instance, a tetrahedron can be defined as:

```
def tetra "poly3d 4 1 1 1 1 -1 -1 -1 1 -1 -1 -1 1"
```

A particle of the type `TAG` with the position $\mathbf{r} = (x, y, z)$ and the quaternion $\mathbf{q} = (q_t, q_x, q_y, q_z)$ is specified by:

```
TAG color x y z qt qx qy qz
```

with the quaternion $\mathbf{q} = (1, 0, 0, 0)$ corresponding to the orientation specified in the `def` command.

C.2 Dense Packings of Hard Tetrahedra

C.2.1 Nonamer Packing

The packing presented below is a numerical compression of the unit cell introduced by Chen [28] and has a density of $\phi = 78.38\%$. (Fig. 5.2).

```
boxMatrix 2.560259039 -0.447328826 -1.143405974 0.383685733 2.575659831 -1.154175128 0.207358905 0.154123516 8.887307999
def S0 "poly3d 4 1.689459 -0.310541 -0.222021 -0.310541 1.689459 -0.222021 -0.467438 -0.467438 1.600938 -0.911479 -0.911479 -1.156897"
def S1 "poly3d 4 1.377833 -0.622167 0.8452716 -0.622167 1.377833 0.8452716 -1.223105 -1.223105 -0.089605 0.4674383 0.4674383 -1.600938"
def S2 "poly3d 4 0.5624301 -1.43757 0.7855349 -1.43757 0.5624301 0.7855349 -0.347965 -0.347965 -1.660675 1.223105 1.223105 0.08960497"
def S3 "poly3d 4 0.3304534 -1.669547 -0.321582 -1.669547 0.3304534 -0.321582 0.9911282 0.9911282 -1.017512 0.347965 0.347965 1.660675"
def S4 "poly3d 4 -1 -1 -1 1 1 -1 -1 1 1 1 -1 1"
def S5 "poly3d 4 -1.689459 0.3105412 0.2220205 0.3105412 -1.689459 0.2220205 0.4674383 0.4674383 -1.600938 0.9114793 0.9114793 1.156897"
def S6 "poly3d 4 -1.377833 0.6221667 -0.845272 0.6221667 -1.377833 -0.845272 1.223105 1.223105 0.08960497 -0.467438 -0.467438 1.600938"
def S7 "poly3d 4 -0.56243 1.43757 -0.785535 1.43757 -0.56243 -0.785535 0.347965 0.347965 1.660675 -1.223105 -1.223105 -0.089605"
def S8 "poly3d 4 -0.330453 1.669547 0.3215816 1.669547 -0.330453 0.3215816 -0.991128 -0.991128 1.017512 -0.347965 -0.347965 -1.660675"
def S9 "poly3d 4 1 1 1 -1 -1 1 1 -1 -1 -1 1 -1"
def S10 "poly3d 4 1.676411 0.3235889 0.2914379 -0.323589 -1.676411 0.2914379 -0.384973 0.3849733 -1.64426 -0.967849 0.967849 1.061384"
def S11 "poly3d 4 1.419762 0.5802377 -0.804736 -0.580238 -1.419762 -0.804736 -1.224498 1.224498 -0.034789 0.3849733 -0.384973 1.64426"
def S12 "poly3d 4 0.6034304 1.39657 -0.827928 -1.39657 -0.60343 -0.827928 -0.431359 0.4313586 1.621067 1.224498 -1.224498 0.03478902"
def S13 "poly3d 4 0.315858 1.684142 0.2527834 -1.684142 -0.315858 0.2527834 0.9369254 -0.936925 1.115501 0.4313586 -0.431359 -1.621067"
def S14 "poly3d 4 -1.676411 -0.323589 -0.291438 0.3235889 1.676411 -0.291438 0.3849733 -0.384973 1.64426 0.967849 -0.967849 -1.061384"
def S15 "poly3d 4 -1.419762 -0.580238 0.8047355 0.5802377 1.419762 0.8047355 1.224498 -1.224498 0.03478902 -0.384973 0.3849733 -1.64426"
def S16 "poly3d 4 -0.60343 -1.39657 0.8279282 1.39657 0.6034304 0.8279282 0.4313586 -0.431359 -1.621067 -1.224498 1.224498 -0.034789"
def S17 "poly3d 4 -0.315858 -1.684142 -0.252783 1.684142 0.315858 -0.252783 -0.936925 0.9369254 -1.115501 -0.431359 0.4313586 1.621067"
S0 ff0000ff -0.154432292 -0.585884752 -0.150972715 0.998509448 -0.048051485 0.011763417 -0.023055547
S1 ff0000ff 0.155654298 -0.325552043 -1.234618145 0.998531184 -0.053601022 0.007163775 -0.003329496
S2 ff0000ff 0.9680784 0.495084921 -1.254949121 0.99852946 -0.05363929 0.007117843 -0.003328674
S3 ff0000ff 1.210360141 0.797214207 -0.165168564 0.998443267 -0.054686688 0.005043225 -0.009745485
S4 ffff0000 0.361358044 1.372644779 -3.891885513 0.999584851 -0.019730187 -0.011201971 -0.017758418
S5 ffff0000 1.483483516 -0.526823379 -3.337142229 0.998517425 -0.048233172 0.011607006 -0.022400701
S6 ffff0000 1.173870174 -0.785030919 -2.252996635 0.998531274 -0.053599533 0.007161901 -0.003330563
S7 ffff0000 -0.085884264 0.970030642 -2.078173636 0.998529355 -0.053640724 0.007120608 -0.003331138
S8 ffff0000 -0.328488245 0.66716888 -3.166775325 0.998428091 -0.054977153 0.005502218 -0.00941203
S9 ff0000ff 0.524099139 0.08964577 0.55774948 0.999571471 -0.020275866 -0.010299563 -0.018430503
S10 ff0000ff -0.188457808 0.877382911 1.231385837 0.999526732 0.00486902 0.027593165 -0.012697329
S11 ff0000ff 0.113039901 0.607270023 2.312865412 0.999579335 0.005704506 0.026809908 -0.009478432
S12 ff0000ff 0.945381121 -0.196246853 2.353739171 0.999573192 0.005423836 0.027096043 -0.009477373
S13 ff0000ff 1.217026204 -0.515412828 1.261118397 0.999306659 0.003265643 0.0258853 -0.026561013
S14 ffff0000 -0.072101962 -0.573225617 4.317570339 0.999495888 0.006306834 0.028937872 -0.011436472
S15 ffff0000 -0.379159483 -0.302055009 3.237874425 0.999541604 0.007136676 0.028208827 -0.008361274
S16 ffff0000 -1.210407278 0.502635202 3.201111353 0.999547665 0.007424136 0.027917833 -0.0083632
S17 ffff0000 -1.034305108 -1.753123088 4.142917717 0.999267825 0.002366557 0.026484825 -0.027509394
eof
```

C.2.2 Reformed Scottish Packing

Reformed Scottish packing(Fig. 5.3a) was first proposed by Conway and Torquato [27]. We compressed a unit cell of it to a packing fraction of $\phi = 74.36\%$.

```
boxMatrix 5.23463941 0 0 0 5.23463941 0 0 0 5.23463941
def S0 "poly3d 4 1 1 1 1 -1 -1 -1 1 1 -1 -1 1"
S0 ffff0000 -0.756773627 -2.244651662 2.351203256 0.881507077 0.356625021 -0.075548311 -0.300093853
S0 ffff0000 -1.38616614 -1.794055571 1.492490701 0.632311173 0.447059355 0.449753654 -0.445019285
S0 ffff0000 -2.505547105 -1.120288756 1.714331948 0.194686512 0.330281073 0.832603477 -0.399728688
S0 ffff0000 -2.494197661 -1.074269164 -2.36204157 -0.345220353 0.095919121 0.893793129 -0.269733708
S0 ffff0000 -1.549595202 -1.771373859 -1.869402904 -0.75076058 -0.106440813 0.651940491 -0.001581712
S0 ffff0000 -2.449340267 0.815945885 1.885520934 0.825648921 0.433528646 0.179626059 -0.313195229
S0 ffff0000 1.820612841 1.537858171 1.408340401 0.458583575 0.459235211 0.606462927 -0.459354814
S0 ffff0000 1.080040105 1.966996561 2.434120301 -0.15175185 0.264350981 0.878093718 -0.368837847
S0 ffff0000 1.70386055 1.549563794 -1.913435336 -0.627713221 -0.003571332 0.766364597 -0.136560107
S0 ffff0000 -2.439709329 0.865146589 -2.192453799 -0.875419003 -0.299763739 0.367139091 0.094826988
S0 ffff0000 -0.872783113 1.835472156 2.403769874 0.876916322 0.375857347 0.12990942 0.269949183
S0 ffff0000 -1.786389732 2.53366036 0.831206324 0.526366801 0.71522839 0.453504414 0.075631261
S0 ffff0000 1.824742505 -1.830369152 1.241746415 -0.001161779 0.763318405 0.632103956 -0.133372606
```

```
S0 ffff0000 1.812000521 -1.672038814 -2.024324151 -0.538466435 0.552345158 0.549292113 -0.321320556
S0 ffff0000 -1.969202273 2.426280746 -1.120583489 -0.882459862 0.076358996 0.296804491 -0.356848693
S0 ffff0000 1.158926063 -2.111221415 2.338099543 -0.286277679 -0.100703744 0.35256993 0.885210874
S0 ffff0000 2.100789945 2.431212236 -1.281777653 -0.375562125 0.261197228 -0.150221409 0.876448873
S0 ffff0000 -1.537298044 1.552793478 -1.741334528 -0.353316339 0.539977644 -0.541361337 0.538998712
S0 ffff0000 -1.535395049 1.417867619 1.516386661 -0.153467569 0.617047447 -0.771785184 0.006912523
S0 ffff0000 2.295524447 2.544741591 0.625824883 0.03776461 0.462778259 -0.678807081 -0.56888581
S0 ffff0000 1.725170307 0.536707036 -0.196596314 0.885160058 0.354445845 -0.115669304 -0.278353061
S0 ffff0000 1.06236591 0.957434449 -1.116115394 0.638022319 0.452111375 0.441155987 -0.44034557
S0 ffff0000 0.172790525 1.575205611 -0.706930897 0.15956397 0.374443044 0.807117668 -0.427659698
S0 ffff0000 0.168910136 1.471710739 0.44708544 -0.370514632 0.140165764 0.878803312 -0.266039855
S0 ffff0000 1.062483637 0.838380289 0.85132658 -0.766901979 -0.139918353 0.626313803 -0.003902476
S0 ffff0000 0.152098419 -1.727713462 -0.918383104 0.814967788 0.432725033 0.159213874 -0.351037736
S0 ffff0000 -0.745400266 -1.081980423 -1.338554406 0.45223413 0.439611605 0.639334033 -0.439861254
S0 ffff0000 -1.445656706 -0.778542445 -0.276552754 -0.11382948 0.285672736 0.872418244 -0.379895175
S0 ffff0000 -0.77974491 -1.212424498 0.633112104 -0.618566785 0.001969295 0.774189471 -0.134171225
S0 ffff0000 0.115341186 -1.826834366 0.23558989 -0.870679867 -0.267350375 0.384974921 0.149112899
S0 ffff0000 1.852792709 -0.620694149 -0.319718146 0.878216286 0.366796203 0.157801165 0.263240369
S0 ffff0000 0.797921994 -0.180509573 -1.845851274 0.563641964 0.683844677 0.461354794 0.042613945
S0 ffff0000 -0.835302294 0.791452553 -1.190465105 0.000407138 0.750236478 0.652659608 -0.105737875
S0 ffff0000 -0.922437943 0.792117136 0.721370847 -0.541644895 0.538776864 0.533813416 -0.362468668
S0 ffff0000 0.668333653 -0.054153852 1.469206773 -0.878209443 0.145067736 0.258368936 -0.375431776
S0 ffff0000 -1.558186469 0.382430009 -0.15116573 -0.293928483 -0.066753789 0.366761831 0.880133932
S0 ffff0000 -0.488162293 -0.094362574 1.381687591 -0.354421824 0.298798543 -0.091887615 0.881283875
S0 ffff0000 1.124290929 -1.036339097 0.72290609 -0.384864219 0.537574494 -0.530087901 0.530942571
S0 ffff0000 1.22783391 -1.053127767 -1.189566185 -0.127812368 0.634041705 -0.762663163 0.000123396
S0 ffff0000 -0.357595172 -0.186232874 -1.95560007 0.081194334 0.453074398 -0.723208505 -0.51487914
eof
```

C.2.3 Fivefold Log Packing

This is a packing of fivefold logs into a hexagonal lattice (Fig. 5.3b), which we numerically compress to a maximum packing fraction of 76.73%.

```
boxMatrix 8.813219058 0 0 0 5.269759849 0 0 0 5.088314394
def S0 "poly3d 4 1 1 1 1 -1 -1 -1 1 -1 -1 -1 1"
S0 ffff0000 -3.743751649 -0.385888457 1.179645074 0.19949478 0.566525651 -0.404683143 0.689552082
S0 ffff0000 -3.115868405 -0.399852273 2.272177419 0.140031107 0.009375453 -0.368683634 0.918899215
S0 ffff0000 -3.524929661 -0.272970677 -1.719877012 0.084690004 0.536078706 0.241642843 -0.804397887
S0 ffff0000 -4.109726368 -1.732947738 0.52806809 0.086392005 0.188724988 0.073225974 0.975477963
S0 ffff0000 -2.660602853 -1.530007469 2.152376678 0.222937292 -0.309576585 0.116259129 0.917030597
S0 ffff0000 -3.641515805 -1.412674537 -1.269972197 -0.258491075 0.805713639 -0.047636209 -0.530790624
S0 ffff0000 -3.253685556 -2.215774124 1.404435973 -0.072109974 -0.114821571 0.592616556 0.793991043
S0 ffff0000 -2.852512372 -2.10285253 -1.951807671 0.260944718 -0.536155275 0.560311117 0.574888535
S0 ffff0000 -3.529968803 2.033978083 -1.817156332 0.104071188 0.264813045 0.378972196 -0.880581236
S0 ffff0000 -3.637835559 2.093513478 2.105421002 0.124763473 0.306420629 -0.374135628 0.866350394
S0 ffff0000 -2.109520962 0.922973258 -1.549029797 -0.349418561 0.644727946 -0.100139873 -0.672461561
S0 ffff0000 -2.028389797 0.973897008 1.028897235 0.345525196 -0.140099489 0.221582453 0.901046996
S0 ffff0000 -3.062612406 0.51348861 -0.94050432 0.505306738 -0.634614652 0.514928224 0.277088918
S0 ffff0000 -3.545102488 0.745978517 0.73402765 -0.262842077 0.107770712 0.463854931 0.839129382
S0 ffff0000 -2.271847188 0.464357811 2.120637417 0.233030174 -0.304160524 0.694074694 0.609461756
S0 ffff0000 -0.393776859 1.058959425 -1.101720149 0.353476767 0.867241648 -0.100263803 0.335995936
S0 ffff0000 -0.471833917 1.143221259 0.050246355 0.379694313 0.888833838 0.113592092 -0.230007549
S0 ffff0000 -0.959742441 -0.066044028 -1.191308598 0.034790483 0.710294539 0.228796215 0.664773331
S0 ffff0000 -1.116649249 0.265334271 0.566775314 -0.105703821 0.969422782 0.161590032 0.151442511
S0 ffff0000 -0.006718623 -0.611362884 -1.866587292 0.398042811 -0.365841521 -0.452984125 -0.708891588
S0 ffff0000 -0.269314151 -0.261237191 1.37333134 0.579641755 -0.774587503 0.253030262 0.002307089
S0 ffff0000 -1.363748449 -0.5878672 -0.242619642 0.531626756 -0.691672448 -0.136438621 -0.469411035
S0 ffff0000 -0.4429617 -1.512584769 0.955068156 0.287062196 0.792863649 0.095056185 -0.529081139
S0 ffff0000 -0.891345768 -1.648286775 -0.128328314 0.404796327 0.906896891 -0.116045556 0.014539337
S0 ffff0000 0.057489534 -1.555766182 -0.927544628 0.383307976 0.679870858 -0.240321994 0.577144654
S0 ffff0000 -0.91089816 -2.333208897 1.620228896 -0.126288769 0.981182886 -0.019054843 -0.144804021
S0 ffff0000 -1.759933315 -2.471471155 -0.014968832 0.007196851 0.907360539 0.069473161 0.414509996
S0 ffff0000 -0.98340612 -2.626341591 -1.780845661 0.190631326 0.538509379 0.126976116 0.810891123
S0 ffff0000 -1.790096818 2.283434209 1.047356853 0.531447384 -0.778926887 0.133756317 -0.304870187
S0 ffff0000 -1.908872257 2.242441902 -1.19338963 0.403671885 -0.567793426 -0.194816223 -0.690439189
S0 ffff0000 -2.524522915 -0.727342981 -0.209453305 0.885324384 -0.148335762 0.309233688 0.313961404
S0 ffff0000 -2.803328074 -1.847414698 -0.246439521 0.534073439 -0.35630253 0.184859047 0.744070697
S0 ffff0000 -1.303585806 -0.156478161 2.223471827 0.78065659 0.157997204 0.278441466 -0.536733195
S0 ffff0000 -1.481061134 -1.310119867 2.321580439 0.341958503 0.28639269 0.097454275 -0.889688863
```

```
S0 ffff0000 3.832496067 -0.704584434 1.584847866 0.331128443 0.889782727 -0.203882347 0.238898811
S0 ffff0000 3.817670047 -0.849824237 -2.337754567 0.281166802 0.904110256 0.063471749 -0.315438125
S0 ffff0000 2.045199376 -1.664677069 1.666721697 -0.027680336 0.67366733 0.360812527 0.644376014
S0 ffff0000 3.054154319 -1.622938833 -1.168785676 -0.121354814 0.969974337 0.171850492 0.12202542
S0 ffff0000 3.101253258 -2.153112034 1.295977159 0.407075997 -0.272921765 -0.559618802 -0.668303553
S0 ffff0000 3.841510249 -2.270997395 -0.361683356 0.632650094 -0.745658011 0.203301761 0.049156731
S0 ffff0000 2.869412023 -2.377933833 -2.11620821 0.610792338 -0.673958893 -0.148100663 -0.388301847
S0 ffff0000 4.148377126 2.098924909 -1.068233191 0.265024951 0.788933143 0.231155895 -0.503898028
S0 ffff0000 3.540415893 1.951285423 -2.139300567 0.361647194 0.932275997 -0.007144168 0.00466193
S0 ffff0000 4.03332317 2.126896458 1.867446463 0.288031006 0.763352854 -0.238423209 0.526768387
S0 ffff0000 4.27794816 0.838614335 -0.847532229 -0.203636014 0.956670709 0.08630591 -0.189380087
S0 ffff0000 3.068688641 0.867488462 -2.243556506 -0.098763635 0.913639393 0.138768786 0.369123323
S0 ffff0000 4.160690494 1.078088275 1.195067139 0.024405303 0.545416462 0.145996127 0.824991149
S0 ffff0000 3.572421505 0.090014486 -1.488256381 0.609211714 -0.760569406 0.095525071 -0.203150749
S0 ffff0000 3.300978358 0.425268371 1.794570785 0.513342676 -0.55602924 -0.225530838 -0.613552461
S0 ffff0000 0.870884232 0.971958271 -0.992439471 0.235318146 0.498663368 -0.207433065 0.808041917
S0 ffff0000 1.350194787 0.964238428 0.181793571 -0.080357104 0.032900922 -0.341212238 0.935967133
S0 ffff0000 0.736920868 0.82622409 1.232637487 0.315906067 0.467707796 0.362503843 -0.741649337
S0 ffff0000 1.420861565 -0.128431969 -1.433407431 0.202949566 0.14143218 0.208229501 0.946281611
S0 ffff0000 2.176950166 0.161354413 0.255282056 0.059614814 -0.386568457 0.072445361 0.917476197
S0 ffff0000 1.216852837 -0.005374371 1.874276931 -0.080773506 0.814517491 0.124392526 -0.560859516
S0 ffff0000 2.067612021 -0.720726886 -0.679918166 0.022023259 -0.205166044 0.627911812 0.750432293
S0 ffff0000 2.125435444 -0.423216246 1.283262843 0.174505994 -0.681075428 0.457110767 0.544732656
S0 ffff0000 0.709646851 -1.708436822 0.825569469 0.058326429 0.379982451 0.27670301 -0.880708129
S0 ffff0000 1.110080582 -1.583765112 -0.306053652 0.143356025 0.190727002 -0.335530789 0.911312981
S0 ffff0000 0.960435881 2.483107415 1.290306239 -0.231561459 0.706106375 -0.101405207 -0.661445434
S0 ffff0000 1.488417038 -2.628979802 -0.800025076 0.159816114 -0.129883605 0.189133362 0.960113342
S0 ffff0000 -0.105729572 2.134166226 1.832979101 0.447682354 -0.741516948 0.472847892 0.161703423
S0 ffff0000 0.561551944 2.318058479 -1.719213716 -0.206507418 0.011604228 0.614183825 0.761576166
S0 ffff0000 1.706431935 2.119209571 0.208549356 0.104371842 -0.440667156 0.605801747 0.654158406
S0 ffff0000 1.797647364 2.198550823 -2.362409543 0.813363579 -0.165607581 0.32066916 0.456273061
S0 ffff0000 1.909269948 1.04957602 -2.416330771 0.405865795 -0.319654474 0.152254407 0.842563095
S0 ffff0000 2.88074176 2.041318507 0.22721976 0.869946221 0.1238089 0.313719173 -0.359784949
S0 ffff0000 3.132464914 0.836519777 0.204851582 0.480259928 0.311212065 0.165960248 -0.803090685
```

C.2.4 Scottish Bubbles Packing

This packing is obtained from numerical compression of the Scottish bubbles packing proposed in [27] (Fig. 5.4a) and has a packing fraction of 60.36%:

```
boxMatrix 3.75659289 0 0 0 3.756591615 0 0 0 3.756591138
def S0 "poly3d 4 1 1 1 1 -1 -1 -1 1 1 -1 -1 -1 1"
S0 ffff0000 -1.674083833 -1.100295886 -0.124996452 0.94016149 -0.042599614 0.336251447 0.034879925
S0 ffff0000 -0.875913473 0.486606822 1.486858222 0.930230982 0.363977172 -0.022512482 0.041038097
S0 ffff0000 -0.464823235 -1.5165244 -1.113317263 0.925560286 -0.027139011 0.001426762 0.377623615
S0 ffff0000 1.417857763 0.70866869 0.393789566 0.924591991 0.031836882 -0.379610991 0.003399923
S0 ffff0000 0.993205856 -0.417557107 -1.317239266 0.929479616 -0.366155169 -0.038651369 0.022452331
S0 ffff0000 0.198600908 1.68138479 0.809160667 0.939255326 0.04605815 -0.042754144 -0.337416897
S0 ffff0000 -1.078839116 1.338409215 0.197359991 0.910956442 -0.41050441 -0.038125796 -0.013817143
S0 ffff0000 -1.333117292 0.099607279 -0.905918956 0.930110955 -0.011594415 -0.001194094 -0.367093659
S0 ffff0000 -0.323981778 -0.90254482 1.400864563 0.902459478 0.010229579 -0.427345702 0.053271916
S0 ffff0000 0.812756438 -1.715809995 -0.501078152 0.910386889 0.412112219 0.011321049 0.035086542
S0 ffff0000 1.526778256 -0.501033493 1.044560772 0.902392232 -0.011437949 -0.046502259 0.428246393
S0 ffff0000 0.532944231 0.932648804 -1.726306652 0.93018232 0.013843979 0.366804118 -0.004892335
```

C.2.5 Welsh Bubbles Packing

This packing is obtained from numerical compression of the Welsh Bubbles packing proposed in [27] (Fig. 5.4b) and has the packing fraction 76.87%.

```
boxMatrix 7.784688508 0 0 0 7.784688508 0 0 0 7.784688508
def S0 "poly3d 4 1 1 1 1 -1 -1 -1 1 1 -1 -1 -1 1"
S0 ffff0000 -3.656425137 -2.7170994 -2.882909613 -0.06660635 0.778642176 -0.41928268 -0.462041113
S0 ffff0000 -2.919910754 -2.333637748 3.838857225 0.241908183 0.68889824 -0.68011529 -0.065900213
S0 ffff0000 -2.926722025 -2.187143099 -2.154142264 0.994670274 0.084680525 0.044884482 -0.038021546
```

```
S0 ffff0000 -2.100500334 -1.432385882 -2.556781262 0.365757001 0.443425251 0.817488379 0.036174766
S0 ffff0000 -2.084932928 -1.55227131 -3.743213641 -0.048854607 0.682131892 0.634703806 0.359806043
S0 ffff0000 -2.257348845 -2.994504728 -1.667309791 0.409246669 -0.407923576 0.103766885 -0.809535641
S0 ffff0000 -3.30967244 -1.698578145 -1.1387313 0.846043545 0.070788406 -0.409938699 0.333391037
S0 ffff0000 -0.14570978 -0.937537453 -0.119136944 -0.027029742 -0.066757898 0.691765648 -0.718521443
S0 ffff0000 -2.116680328 -0.038743212 -3.792927383 0.452800609 0.733102249 0.407692498 0.30219121
S0 ffff0000 -2.754653515 -2.272583752 -0.174656844 0.658207801 -0.257816251 -0.055270665 0.705151349
S0 ffff0000 -2.298315176 -0.344266621 -1.743140782 0.650952477 0.063885178 0.723348149 -0.221239717
S0 ffff0000 -0.802224851 -0.074164592 -1.929777615 -0.214246191 0.941954729 -0.076846733 -0.246808503
S0 ffff0000 -3.052756749 -0.521039786 -0.852425943 0.665791138 0.373690997 -0.639843166 -0.087623757
S0 ffff0000 -1.006295647 -0.29444137 -0.577859199 0.549444512 -0.548811847 0.008910767 0.629949905
S0 ffff0000 -0.199188989 -3.640017128 3.623358402 0.070441907 0.002541169 0.732066121 -0.677577062
S0 ffff0000 -0.988648394 -1.706693432 -2.408856221 0.494473767 -0.120959983 0.83369087 0.214065199
S0 ffff0000 -0.202853223 -1.889086807 -3.57042934 0.679840739 -0.226211854 0.455973514 0.527951628
S0 ffff0000 -1.016611975 -2.715869514 -1.818061602 0.1375085 0.581812904 -0.196680525 0.777111271
S0 ffff0000 -3.649266571 -2.658903962 2.470923267 0.079229274 0.512932062 -0.188962078 0.833616672
S0 ffff0000 -3.830501037 -1.456870074 3.585316973 0.286144823 0.646426332 -0.514418542 0.485414978
S0 ffff0000 -2.080490543 -3.2032115 2.769338768 0.42463512 0.314696991 -0.696733021 0.484988574
S0 ffff0000 -3.314120126 -3.850289269 1.69696918 0.743108554 0.317080103 -0.390910883 -0.440951887
S0 ffff0000 -1.095283567 -1.792391665 3.19973254 0.938508384 -0.186407229 0.037682053 0.288156937
S0 ffff0000 -0.220145257 -1.014166572 1.69500867 0.967303967 0.202218756 -0.144839845 0.049517978
S0 ffff0000 -1.802419848 -3.047383367 -0.346099704 0.690799993 0.024017985 0.395288867 -0.604950591
S0 ffff0000 -0.679126044 -2.733642232 0.153888898 0.199192258 0.943911669 -0.249914224 0.083042676
S0 ffff0000 -0.208270393 -3.630942459 1.836197004 -0.203860692 0.960510511 0.075792159 -0.173539407
S0 ffff0000 -3.761966178 -1.7789207 1.556663774 0.5045116 -0.192976486 0.802597779 0.253110503
S0 ffff0000 -3.572402441 -3.01740914 0.228969535 0.662470439 -0.249637638 0.51034651 0.488221678
S0 ffff0000 -1.870976476 -1.421926218 0.839553724 0.481336387 -0.185253723 -0.484192151 0.706791555
S0 ffff0000 -3.077954505 -0.774596403 1.987272754 -0.27281331 0.70966606 0.486182539 -0.430782452
S0 ffff0000 -3.113145001 -0.405380825 3.362457215 0.604739175 0.049273969 0.53085995 -0.591650505
S0 ffff0000 -0.803669034 -3.028408058 2.84362096 0.570732544 -0.583701468 -0.064490288 0.573932019
S0 ffff0000 -0.666378361 -1.604718536 0.675010993 0.627456597 0.580026085 -0.51947716 -0.003382196
S0 ffff0000 -2.014579026 -0.47759982 0.038671319 0.490093065 0.297885507 -0.680593094 0.455923296
S0 ffff0000 -0.929065753 -0.143474949 2.77548868 -0.006658204 0.694096615 0.320226712 0.6447018
S0 ffff0000 -1.800704702 -0.185659477 2.019313151 -0.285097966 0.654410728 -0.014729053 -0.700177694
S0 ffff0000 -3.470227136 3.688039943 -2.99037146 -0.277441964 0.570389274 -0.04189176 -0.771963155
S0 ffff0000 -2.362761162 3.677865568 -2.26274455 -0.014154193 0.716725711 0.272740634 0.641651355
S0 ffff0000 -3.158729347 2.207802509 -3.517363246 0.674267292 -0.223091291 0.533759425 0.459015001
S0 ffff0000 -1.996399301 3.714675216 3.527598501 0.432357575 -0.225630362 -0.524504791 0.697891533
S0 ffff0000 -0.356053895 2.315485253 -3.683801899 0.183167665 0.959394209 -0.214504223 0.000543861
S0 ffff0000 -2.13994739 0.655840555 -2.851435057 0.665080766 0.555772685 0.442291015 -0.230570934
S0 ffff0000 -0.67508576 0.944528378 -0.502542752 0.954407038 -0.185588948 0.032322164 0.231558255
S0 ffff0000 -2.152527678 2.690157319 -1.356387446 0.37795614 0.710258452 0.541099188 0.244732011
S0 ffff0000 -0.88203605 3.523870374 -2.423940549 0.943063136 0.244917241 -0.22328179 0.028155094
S0 ffff0000 -3.602568327 1.121892809 -0.524115796 0.277759394 0.72161673 -0.446297245 0.450486163
S0 ffff0000 3.665673141 0.361280635 -2.090922645 0.729360146 0.314951286 -0.466146891 -0.38928979
S0 ffff0000 -3.368488324 1.029176818 -3.089144055 0.036110722 0.546847628 0.569465473 0.612668558
S0 ffff0000 -3.517684428 2.223607599 -0.917302037 0.516764881 -0.05807308 0.619722156 -0.587814618
S0 ffff0000 -0.97472195 3.427355656 -3.674117624 0.52432607 0.626384423 -0.5766376 0.014622073
S0 ffff0000 -0.087132565 -3.427102159 -2.328812987 0.692125428 0.304014295 -0.4587327 -0.467013929
S0 ffff0000 -3.839020342 0.296982728 -3.862651338 0.03546639 0.064350629 0.700098924 -0.710255327
S0 ffff0000 -0.713299771 1.544009876 -2.256488305 0.093085014 0.815251787 0.235325856 -0.520885251
S0 ffff0000 -0.724031925 2.19717653 -1.302062083 0.855061259 -0.058415589 -0.4900872 -0.158972947
S0 ffff0000 -1.89556548 3.475628575 1.607329444 0.673336095 0.318631317 -0.66661025 0.026895398
S0 ffff0000 -0.901498942 3.524960399 0.884073091 0.704678645 0.029859995 0.6586006 -0.262262536
S0 ffff0000 -2.173327799 3.344817225 -0.394201976 0.638739691 0.485050863 0.539902776 -0.255425643
S0 ffff0000 -3.175206349 3.589840189 0.233905412 -0.040892434 0.632648476 0.579207252 0.512447728
S0 ffff0000 -2.178757573 2.304156944 3.526714605 0.62114143 -0.347927792 -0.011669097 0.702134893
S0 ffff0000 -1.1765487 1.575428853 3.670991233 0.69670756 -0.060252631 0.309969236 -0.644117434
S0 ffff0000 -2.236246762 2.488178261 2.374239226 0.8068635 -0.010420904 -0.397860269 0.436543129
S0 ffff0000 -3.396258598 2.123111978 2.111338256 0.82618006 -0.127138041 -0.536912359 -0.113962916
S0 ffff0000 -0.763663114 1.12167322 2.659304016 0.414852775 -0.43249361 -0.04435412 -0.799299171
S0 ffff0000 -0.63476218 2.169467062 1.229987985 0.388323476 0.415054077 0.822410706 -0.02399211
S0 ffff0000 -1.324417388 1.79214979 0.158624699 0.042953192 0.708695997 0.617955603 0.337691991
S0 ffff0000 -2.110846996 0.822175708 1.458300035 0.050128269 0.82664914 -0.391688823 -0.400896773
S0 ffff0000 -2.278323799 1.022660244 0.118248435 0.307734746 0.616106852 -0.725040004 -0.005354031
S0 ffff0000 -3.287962025 1.116365861 1.491945345 0.179617703 0.819716411 0.190416873 -0.509454512
S0 ffff0000 -1.480366271 1.674743443 1.942369241 0.998603402 -0.040358608 0.005985032 0.033565095
S0 ffff0000 -3.794582551 0.135440771 1.981488177 -0.245339409 0.953791031 -0.004142765 -0.173418801
S0 ffff0000 3.044428656 -2.629183942 -2.327229202 0.174970842 0.80042437 0.158120978 -0.551093267
S0 ffff0000 3.125997902 -1.719407049 -1.508814949 0.850780474 -0.190037485 -0.469369693 -0.140536225
S0 ffff0000 1.889846376 -1.524674074 3.728203168 0.680718286 -0.036092404 0.32729091 -0.654325216
S0 ffff0000 2.985712031 -1.331603744 -3.558530939 0.221414463 0.93597406 -0.273291975 0.015481974
S0 ffff0000 1.618492332 -2.448063715 -3.145649728 0.727706079 0.387816858 0.477405133 -0.303523122
S0 ffff0000 2.988470418 3.779089358 -1.917257867 -0.199304292 0.946989825 -0.038154952 -0.249062783
```

```
S0 ffff0000 0.644507553 -1.141566328 -3.848929526 0.655499289 -0.234871412 -0.122252254 0.707255604
S0 ffff0000 -0.093873899 -1.003344037 -1.97158382 -0.299536684 0.70418059 0.407994752 -0.497943526
S0 ffff0000 0.326153563 -2.730753371 -0.529961853 0.200867753 0.655229808 -0.496231541 0.532991841
S0 ffff0000 3.269574571 -3.042649844 -0.442984814 0.948554465 -0.212889346 0.044329048 0.23012494
S0 ffff0000 1.663575768 -1.039782934 -1.08223132 0.431170992 0.698302681 0.491760735 0.290923221
S0 ffff0000 2.859536337 -0.515202795 -2.134753003 0.941187887 0.236479804 -0.21608397 0.107472703
S0 ffff0000 0.230414964 -2.943023224 -3.338926398 -0.041297363 0.516959961 0.582137204 0.626229352
S0 ffff0000 0.431552321 -1.514902675 -0.946434413 0.553920733 -0.014323291 0.603760152 -0.573097151
S0 ffff0000 2.915094721 3.607439779 -0.599205774 0.57278985 -0.529616253 0.037595905 0.624503772
S0 ffff0000 2.869740848 -0.167383356 -3.507449773 0.539259608 0.561354713 -0.627757286 0.000866874
S0 ffff0000 1.632059126 -0.300242955 -0.195899751 0.671602684 0.488173607 0.510557114 -0.223534778
S0 ffff0000 1.87995982 0.374200862 3.826639386 0.484777782 -0.253803952 -0.523018505 0.653472034
S0 ffff0000 2.714269602 -2.447693862 2.812521963 0.397142884 -0.426971087 -0.074560433 -0.808958567
S0 ffff0000 2.807582982 -3.749817968 2.757727911 0.041038831 0.659806542 0.323384572 0.677047679
S0 ffff0000 1.577123595 -1.179627667 2.328230985 0.788343147 -0.042425471 -0.402640337 0.463244991
S0 ffff0000 0.589922748 -1.817022981 2.054679118 0.809286425 -0.197077456 -0.544155127 -0.100554247
S0 ffff0000 1.924499961 -2.93795881 1.273954843 0.050729249 0.82544227 -0.426708903 -0.366047968
S0 ffff0000 1.570023809 -2.960107423 -0.018262555 0.367888077 0.621572706 -0.690025782 0.046584916
S0 ffff0000 1.970918044 3.825346473 1.969169809 -0.281629582 0.700095037 -0.019517718 -0.655874056
S0 ffff0000 0.745494284 -2.76183852 1.392580589 0.219456566 0.821206872 0.123634484 -0.512027931
S0 ffff0000 2.326938294 -1.887613361 1.807260106 0.997175685 -0.059382178 -0.024234847 0.039077905
S0 ffff0000 2.855606413 -1.426992978 0.759130921 0.414665875 0.365886936 0.830525187 -0.066384306
S0 ffff0000 2.442302568 -2.200099955 -0.169059294 0.065766803 0.692379215 0.677651721 0.238901432
S0 ffff0000 2.655024199 -0.128418988 0.749211159 0.655021494 -0.075989654 0.698850782 -0.277092042
S0 ffff0000 0.603006999 -0.129575813 0.357242005 -0.03501938 0.588632734 0.593188819 0.548098688
S0 ffff0000 2.942227869 2.123229291 -2.353518626 0.504778188 -0.156632962 0.820770025 0.216798666
S0 ffff0000 3.763507482 2.91806823 -1.9470923 -0.277840392 0.688401568 0.436508258 -0.508299654
S0 ffff0000 3.036501603 1.220231621 -1.639625038 0.090635432 0.550005048 -0.165666844 0.813531906
S0 ffff0000 1.625050363 3.681980816 -2.08598629 0.684918132 0.041385392 0.689256031 -0.232595198
S0 ffff0000 0.615577488 3.781932075 -1.255255294 0.61516452 0.350397624 -0.705442601 0.033835702
S0 ffff0000 1.831048148 2.643493464 -2.570144047 0.357613558 0.402826802 0.837185266 0.094678089
S0 ffff0000 0.445088517 1.440600259 -2.620910238 -0.054910624 0.822016463 -0.330318026 -0.460612375
S0 ffff0000 0.644471534 0.226748362 -2.988754206 -0.326142489 0.624124478 0.084097484 -0.70500165
S0 ffff0000 1.083605339 2.023550681 -1.853568733 0.994037175 0.09840466 0.017910254 0.04342626
S0 ffff0000 0.367481012 2.527642456 -1.02442029 0.783545923 0.102630234 -0.437960803 0.428617728
S0 ffff0000 1.794159406 1.099342438 -1.541404365 0.470447827 -0.383374684 0.084873998 -0.790252553
S0 ffff0000 1.518180325 0.166486856 -2.227480973 -0.100583539 0.67385205 0.279706577 0.676439648
S0 ffff0000 1.030414554 1.84164268 -0.152344276 0.649413134 -0.277440301 -0.068880723 0.704659426
S0 ffff0000 1.937118159 1.128517011 -0.204081893 0.716736945 0.033686754 0.326269213 -0.615387483
S0 ffff0000 1.949821395 3.41715498 0.163598335 0.504710977 0.267832439 -0.668422605 0.476176265
S0 ffff0000 3.115356328 2.786206686 3.6226102 0.135858771 0.709460473 0.639027097 0.264296426
S0 ffff0000 1.815556854 2.36127738 3.848544492 0.427872359 0.583783213 -0.686978432 -0.064676411
S0 ffff0000 3.742867726 2.920756021 -0.159604024 -0.07089322 -0.060710205 0.693420084 -0.714462742
S0 ffff0000 3.770559611 1.879807305 3.149408289 0.966178764 -0.212080742 -0.029399311 0.143722079
S0 ffff0000 1.618086542 -3.403702453 -3.793879725 0.521684751 0.635129122 0.53069162 0.206935794
S0 ffff0000 3.777158048 3.008945738 1.692805519 0.946386141 0.278754487 -0.145976965 0.073074855
S0 ffff0000 1.932300366 -0.180383877 1.648271311 0.714754367 0.326472021 -0.616921869 0.044154522
S0 ffff0000 0.413652072 1.093618266 2.352144611 0.171351815 0.517878397 -0.161142055 0.822480249
S0 ffff0000 0.612782143 2.336620869 3.365951618 0.270046488 0.705603612 -0.45482067 0.471525817
S0 ffff0000 2.131092409 1.286347929 2.895613269 0.454836465 0.183270788 -0.774439287 0.399724154
S0 ffff0000 0.485817352 -0.110160598 1.56224039 0.750885399 0.244457475 -0.43060606 -0.437024121
S0 ffff0000 0.4981774 1.921396798 1.428913126 0.489331273 -0.146425134 0.844778116 0.159576067
S0 ffff0000 0.259547599 0.967844873 0.181428358 0.672823333 -0.234578352 0.496803673 0.495447142
S0 ffff0000 1.998277415 2.624589566 1.00239716 0.490385589 -0.24864581 -0.449367601 0.704106522
S0 ffff0000 0.703328833 3.450842236 1.927031569 -0.20635697 0.682464331 0.537236468 -0.450595402
S0 ffff0000 3.46964324 1.146202101 0.461603158 0.260664122 0.943839921 -0.199777811 -0.036183483
S0 ffff0000 0.735424583 3.599613939 3.161018246 0.633827183 -0.003442385 0.56637748 -0.526752126
S0 ffff0000 3.335354493 0.845380235 2.867666749 0.64194019 -0.599383181 -0.003954412 0.478159971
S0 ffff0000 3.261376405 2.25589417 0.711405855 0.626117929 0.581648308 -0.519163327 -0.011446597
```

C.2.6   The Quasicrystal Approximant

The packing is the densest arrangement of a single unit cell of the $(3.4.3^2.4)$ approximant in the hard tetrahedron system. It has a packing fraction of 84.78%:

```
boxMatrix 10.208845351446810  0   0   0 2.571686440054523  0   0   0 9.823376081309215
def tetra "poly3d 4 1 1 1 1 -1 -1 -1 -1 1 -1 1 -1"
tetra FFFF0000   0.538327947382685 -1.081636567075107 -0.034601148989334 0.817337586591655 0.327880695725213 0.296792389611936 0.369280105588854
tetra FFFF0000  -1.038510164943297 -1.113760600594468 -1.257832842360639 0.572409504226847 0.555069682225026 0.586810519323514 0.141061765750514
tetra FFFF0000  -2.936248068239788 -0.760953042828404 -0.857718710157713 0.181230846549929 0.627571732250111 0.744225012130181 -0.139421061749015
```

```
tetra FFFF0000  -3.189060515609178 -0.610367833064769 1.083708754383395 -0.262808429185872 0.555597125444297 0.696642276980510 -0.370044729283231
tetra FFFF0000  -1.700792884507799 -0.596037624237901 2.343482422988033 -0.606076412917511 0.313456853804025 0.510476549992585 -0.523287563788776
tetra FFFF0000  0.093321350868941 -0.778493967504492 1.780561049880858 -0.831871087530107 -0.002668094829472 0.097034161527852 -0.546413530668893
tetra FFFF0000  -2.767938113199055 -1.232301986627499 1.982530638007453 -0.511466780056498 0.110877850877013 0.167797356284971 0.835435145482465
tetra FFFF0000  -0.761345399388574 1.256504453932962 2.393360544933835 -0.487044537614804 0.482367246946034 -0.142012005764080 0.714102266956315
tetra FFFF0000  0.549414258868341 0.992553257462912 1.084636990857025 -0.303386855923978 0.746141048841573 -0.457236868165683 0.377046943067038
tetra FFFF0000  -0.125152393012702 0.823723080366044 -0.775141581446323 -0.056835804113628 0.752769535285890 -0.655696177477151 -0.013047641858777
tetra FFFF0000  -2.194642199677001 0.987952038557198 -1.226087546632044 0.159579359045945 0.623480493426552 -0.642537327176357 -0.415875324668502
tetra FFFF0000  -3.381243877588915 1.229767581486165 0.073893654179913 0.393124942344290 0.255341799443682 -0.453167851680405 -0.758216488456317
tetra FFFF0000  -0.708113781697524 -0.021495439484946 -0.196829692232588 0.877925426273603 0.307669977993508 0.300202508678201 -0.210866271193619
tetra FFFF0000  -1.857907264675644 0.041304477416064 -0.356556276456923 0.506204629628837 0.215785933070325 0.731901659349072 -0.401887129768268
tetra FFFF0000  -2.435572428894385 0.233651581666976 0.682098169607358 -0.077285676855159 0.027685802901592 0.890770431506546 -0.446977022702931
tetra FFFF0000  -1.477550748677476 0.339731983158568 1.419936214180924 -0.632164311257724 -0.165735287509000 0.689639949026538 -0.312115425059444
tetra FFFF0000  -0.452581552857670 0.154795581620399 0.921348760887522 -0.925126149367389 -0.289148777663002 0.237492658937657 -0.064279305249676
tetra FFFF0000  -4.696066630116136 -0.884464942233475 -1.346366031479770 0.752798795508367 0.294740657666394 0.342476451388553 0.478677133821784
tetra FFFF0000  4.134873454476181 -1.755657332350324 -2.558587215359589 0.510749237041532 0.484786698026172 0.687858834606214 0.175975276480974
tetra FFFF0000  2.031764476656213 -1.243319524637724 -2.137790930785434 0.204314433293040 0.515845672497673 0.824581764224713 -0.110561153311766
tetra FFFF0000  1.658507621859929 -1.137723342775472 -0.313403753042214 -0.237414150952348 0.395554503426156 0.768344744968793 -0.443641193562661
tetra FFFF0000  3.271541703109135 -0.876038338172507 0.965908625133347 -0.541999631237362 0.250655568049121 0.494577015532645 -0.631507530953795
tetra FFFF0000  -4.989165291905720 -0.593659657065584 0.517834054501861 -0.756664421751636 -0.019744211393859 0.101846515268841 -0.645520260177239
tetra FFFF0000  2.360360741610543 0.956477746914198 0.513260555626028 -0.642953883823895 0.136119708720473 0.156883192088804 0.737203765734854
tetra FFFF0000  4.377127913593054 1.285047852548412 1.082363925029749 -0.562284737816316 0.528052666184054 -0.071990696462948 0.632308148751484
tetra FFFF0000  -4.522007830461938 1.177264584894022 -0.227473951806370 -0.350439854078513 0.804758552082334 -0.356105856313270 0.320537362288966
tetra FFFF0000  5.042810954123738 0.878814607332197 -2.052047428809768 -0.042946284143429 0.836754778718335 -0.542672105156007 -0.059194959716595
tetra FFFF0000  2.967736292179869 0.721734491222510 -2.523174127599491 0.215380422872353 0.707681157304578 -0.548393453731362 -0.389952911187965
tetra FFFF0000  1.706919969633839 0.634717871709929 -1.195703636912878 0.495220229784038 0.339443008716617 -0.417846454807487 -0.681864875214195
tetra FFFF0000  4.458147895925859 0.038082237703646 -1.360190323111128 0.909700775650608 0.271464815741076 0.308484459239547 -0.059905987939942
tetra FFFF0000  3.377947850623143 -0.190438203561325 -1.699150163680346 0.589151094465234 0.124784175111518 0.741999472511364 -0.294561844652378
tetra FFFF0000  2.679837668157425 -0.210713352231313 -0.773160162333578 0.050065553221748 -0.070476349661202 0.902188261894857 -0.422590658460607
tetra FFFF0000  3.299641709982919 -0.016235173743299 0.188802285702022 -0.502797492167502 -0.254502471747885 0.726292166284481 -0.393602417342784
tetra FFFF0000  4.434606170769150 0.209092014499054 -0.182723363338539 -0.886139019530960 -0.322768069970230 0.249038169297686 -0.220359708897201
tetra FFFF0000  1.838753749622308 -1.146358338570484 -4.799537185590947 0.761545429760626 0.438995628617801 0.203153385841501 0.431346842212042
tetra FFFF0000  0.565515277303737 -0.882811702074357 3.569803940521781 0.506505569765903 0.628751849091636 0.544904832457930 0.226278464801665
tetra FFFF0000  -1.419074054456052 -0.716688317680052 4.147667296626637 0.106431735532988 0.664146106765873 0.739981379454344 0.003129313026134
tetra FFFF0000  -2.023364552388024 -0.888020884264969 -3.841453326569261 -0.322264907562681 0.498243385207910 0.757353775892369 -0.272606156555113
tetra FFFF0000  -0.721714298812761 -1.145568117266694 -2.394982752119308 -0.654942751494002 0.179376699958866 0.565729883002002 -0.467785945979434
tetra FFFF0000  1.256918477712772 -1.238965549276295 -2.998815047856791 -0.803951198898402 -0.123352733140934 0.203300359724991 -0.545083054911976
tetra FFFF0000  -1.582509759020920 0.899223946443282 -2.979946333368044 -0.527536145069068 -0.000555452719667 0.088149375860735 0.844946740719974
tetra FFFF0000  0.318513131849556 0.750726066698343 -2.651095164365957 -0.579651042901480 0.380051042245615 -0.230173035892449 0.683071187578061
tetra FFFF0000  1.662757405244697 0.644999535532183 -3.962811714543805 -0.454695423162165 0.656030783788427 -0.481561645846869 0.361903390598343
tetra FFFF0000  1.229334960140172 0.885161672843021 4.072607722726285 -0.221182532901262 0.744793716808440 -0.625999884307172 -0.066968286445587
tetra FFFF0000  -0.623356423578924 1.220872866225897 3.589547386680102 0.046514475467346 0.647494662849288 -0.585750939942802 -0.485265805007738
tetra FFFF0000  -2.028474652833631 1.208524566926964 4.896481551915810 0.346491312945109 0.362205189894005 -0.371621997027021 -0.781439864476513
tetra FFFF0000  0.732813381118706 -0.230974583943343 -4.214301749883523 0.897224798578043 0.418766209101065 -0.137456251145785 -0.026987070101444
tetra FFFF0000  0.281543473048466 -0.056667333023830 4.533576555041718 0.771082601838942 0.406538614154196 0.438950057901022 -0.217900947706894
tetra FFFF0000  -0.840433642170010 0.161138764428531 4.764206456256443 0.358257593893781 0.250043854487951 0.843857719460435 -0.311502354015632
tetra FFFF0000  -1.135898930333364 0.035889438478583 -3.945568679994772 -0.178262850759858 -0.020910242912084 0.936364211899445 -0.301673963832243
tetra FFFF0000  -0.064071054956771 -0.149241850813768 -3.375331641098099 -0.687372703780503 -0.274723809635332 0.649616753364577 -0.173331094339700
tetra FFFF0000  -3.184861293590484 -0.564957882024360 3.615344114948864 0.714078251182203 0.372789724051026 0.243475541273074 0.540221929980920
tetra FFFF0000  -4.534508497980070 -0.560452127293761 2.446196612448177 0.452670246466278 0.534694067769814 0.647464186851024 0.299970046176399
tetra FFFF0000  3.794489596766005 -0.772837126442072 2.807060839105459 0.074428277124671 0.534678333083905 0.841554806561626 -0.019105476908318
tetra FFFF0000  2.964789259049481 -1.060923931933388 4.755308046086619 -0.252676323194415 0.367286688430913 0.835113079691828 -0.322244175007146
tetra FFFF0000  4.257131873414839 -1.135933039257589 -3.772954964360948 -0.587155648381717 0.092678300591149 0.562569841704335 -0.574607823108720
tetra FFFF0000  -3.910613890829325 -0.862450485822128 -4.455605701246658 -0.711213609369052 -0.120302657336473 0.187075282677951 -0.666862287955312
tetra FFFF0000  3.514489393930678 0.739954569704744 -4.351893531318328 -0.661020108993362 0.011701276333246 0.082461870300173 0.745731543911877
tetra FFFF0000  -4.887865006924959 0.977117512384285 -4.023385963234607 -0.659470748831491 0.468730927255962 -0.201957159871623 0.551908466003182
tetra FFFF0000  -3.146079227204990 1.223813587855193 4.573634386166327 -0.491682853916059 0.742885359592871 -0.348114220371918 0.291866070728309
tetra FFFF0000  -3.623158449674954 -1.241200547539019 2.760307516692577 -0.173707614333799 0.832664245227928 -0.513411619556687 -0.113597659947926
tetra FFFF0000  4.699546920854318 -1.228102796296048 2.246263973630346 0.171204252361910 0.707302047004369 -0.475735993446091 -0.494052813792706
tetra FFFF0000  3.208756964352296 1.077471296984867 3.691819310677610 0.451868902494213 0.437674909129520 -0.327803331666591 -0.704840510062684
tetra FFFF0000  -4.204608300488951 0.293313547434629 4.171787185734140 0.900478930630327 0.423294357587475 -0.011500550335313 0.099132838555449
tetra FFFF0000  -4.898168953737978 0.316694155565076 3.211272283637288 0.773320592173426 0.305368810232691 0.541868512284486 -0.122896976578802
tetra FFFF0000  4.204403489853616 0.145311207934812 3.503722216171458 0.353881981974805 0.087892521068740 0.889397179112492 -0.275708370131388
tetra FFFF0000  4.106949301365407 -0.054522550694620 4.638078384083317 -0.182543735350168 -0.186672710819419 0.904692261194453 -0.336694217732655
tetra FFFF0000  -4.916059109246240 0.071168558842361 -4.768939963391872 -0.689581805270762 -0.380781632199243 0.547226850057171 -0.282887003935676
tetra FFFF0000  1.974581091485459 -1.210720077155658 2.416637031661228 0.021668269501219 0.935253196799797 0.016834137978934 0.352914374560017
tetra FFFF0000  2.110363173395732 0.657973996980618 3.345775278271824 -0.505310953002522 0.783627204765564 -0.222545809037109 0.284714958523451
tetra FFFF0000  1.621649214702879 0.656642344341665 1.370776900917460 0.485272052035142 0.803199407761802 0.268369036762872 0.217623084693838
tetra FFFF0000  2.950700977580040 -0.452266134078486 2.079034761298958 -0.252476329339501 0.808735181283946 0.483088980661470 0.220970917634733
tetra FFFF0000  0.940323938642802 -0.504288306036050 2.537035254114325 0.244167892763267 0.797411910598554 -0.488517549180879 0.256645454121862
tetra FFFF0000  0.671289283234656 0.235839005664591 -1.517486858207975 0.280815575269599 0.215389303570009 0.854292724461511 0.380701985187573
tetra FFFF0000  2.683382633179328 0.232772979971984 -3.584102992500505 0.314051482143453 -0.193490460074717 0.841894384783555 -0.393886980355399
tetra FFFF0000  -2.130671228628549 -0.129803775215756 3.354746342821389 0.311116539493081 -0.395696289638968 0.836076266530751 0.218191250429080
tetra FFFF0000  -4.219931604871536 -0.109882500349770 1.259410522196842 0.295727109456136 0.413921403937317 0.827822470220452 -0.236483627114834
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tetra FFFF0000  -3.394388192655966 1.231891135755789 -2.732609794016115 0.031812382980703 0.981340904812329 0.002619109379136 -0.189607861380449
tetra FFFF0000  -2.492950056583079 0.622251353451831 -2.319830595473770 -0.301730998869111 0.817827531747219 -0.469943878130378 -0.138813846735404
tetra FFFF0000  -4.484404754184109 0.572007745972042 -3.000374860468890 0.252466480528742 0.807304941584386 0.487527247277332 -0.216417630208467
tetra FFFF0000  -3.112641842535461 -0.527872771756743 -3.561978275061939 -0.471096066244149 0.818736048052575 0.287307751275629 -0.158726292861267
tetra FFFF0000  -3.650459631519871 -0.571845157892545 -1.745648036092591 0.491642781260037 0.806959135021187 -0.282548794443044 -0.165137847871210
```

The compression of eight unit cells, with 656 particles. It has a packing fraction of 85.03%:

```
\boxMatrix 20.124027715878380 0 0 0 5.192920842530376 0 0 0 19.687498870777841
def tetra "poly3d 4 1 1 1 1 -1 -1 -1 -1 1 1 -1 1 -1"
tetra FFFF0000 2.967194957690658 2.525475006938243 1.820984433765668 -0.006043679186931 0.978852119705851 -0.018126493561838 0.203674819063132
tetra FFFF0000 8.083070662718706 2.456598480470830 -2.943126690206037 0.020471739625235 0.944363062680458 -0.011277099212200 -0.328073377089425
tetra FFFF0000 2.836738975044177 2.543630128702523 -0.814550485074518 -0.290974565891018 0.381009221557803 0.740119087106881 -0.471581925006173
tetra FFFF0000 3.135588506123002 -2.571739524296127 -5.282546353406340 0.747127277665386 0.492260789020921 0.257935234820228 0.364622491351047
tetra FFFF0000 1.743432222624314 -2.578910324981241 -0.435280431076952 0.804467062009140 0.352214321079964 0.343734010785478 0.332602988558270
tetra FFFF0000 -7.382131571031975 2.490596377417793 6.377316239030486 -0.820920282632604 -0.183116292719497 0.131206296135632 -0.524731570193220
tetra FFFF0000 0.702323484126362 2.536865086001133 -2.815232356051019 -0.705761093444262 0.135502573887805 0.488114250880235 -0.495262364346178
tetra FFFF0000 -7.461147862160111 2.504271077861492 -3.473727160720102 -0.814800154844888 -0.199520428246462 0.108113487982419 -0.533482689591115
tetra FFFF0000 5.271094482923667 2.540974979975772 -2.954577478869326 0.521065417175715 0.466942148864871 0.686546398806556 0.197762238363911
tetra FFFF0000 -6.580413638711274 2.477407182044689 7.232789608448060 0.121806413992915 0.514095321382885 0.835967861122167 -0.148414733814551
tetra FFFF0000 3.457653596752102 2.491611100004213 7.304998929526876 0.117903492300278 0.521170931609876 0.832318758490541 -0.147394405636906
tetra FFFF0000 -7.146981808554102 2.526421414278008 -8.080344834376364 -0.002748189485390 0.978695142394874 -0.035088812911946 0.202279610726491
tetra FFFF0000 8.417798091377312 2.491312182688579 1.524733317944170 -0.534858492524898 0.196286264660411 0.100727151067544 0.815629901559201
tetra FFFF0000 -6.600412136174926 2.482952514369005 -2.589787543710112 0.108272012389513 0.519234217584184 0.834159254020317 -0.151166588752717
tetra FFFF0000 5.191432051561147 2.528033422186501 6.806466985269205 0.520922894371677 0.460910120227276 0.697645195159533 0.171733458773557
tetra FFFF0000 8.430271675936103 2.481441151412164 -8.285699051392756 -0.541719030338291 0.199865806102611 0.119395490001136 0.807674977133552
tetra FFFF0000 0.689477644565347 2.565105300534294 7.072629349506821 -0.696923792980905 0.141535808849536 0.499661995546652 -0.494573282534301
tetra FFFF0000 2.637226212620744 2.512258637154960 6.406808926504192 -0.820181236044125 -0.184239211050716 0.122591062332396 -0.527569980750191
tetra FFFF0000 -7.174884187400078 2.449709592607006 1.923636019096525 -0.027485287289556 0.976433751639474 0.003259283148399 0.214035195038644
tetra FFFF0000 2.752251427683678 2.522093501503124 -3.483532911730250 -0.822302452147397 -0.176908095140797 0.141729796586445 -0.521952936408635
tetra FFFF0000 -4.868881883924582 2.540907449296828 -3.095120072610091 0.515220479986719 0.467313986622787 0.695069168174476 0.181781039611437
tetra FFFF0000 3.491196747285091 2.476542489255813 -2.576486099564288 0.122163507336598 0.512724659640270 0.837796067821320 -0.142433316380420
tetra FFFF0000 7.681319098055285 2.445423775301966 2.414176671062616 -0.133909149038431 0.809000228907282 -0.529580369665491 -0.217098137939343
tetra FFFF0000 7.654610557403570 2.444307374782363 -7.426224240004402 -0.133125941394519 0.809001863545675 -0.532244973104299 -0.210970986423850
tetra FFFF0000 -2.444971659913953 2.467234557381597 -7.368264285733147 -0.124643195323015 0.804383357566828 -0.539650959567185 -0.214961228521336
tetra FFFF0000 -4.813421198930431 2.514275826442203 6.861835405579372 0.524851907545194 0.463335992255484 0.688402348843920 0.189611285352351
tetra FFFF0000 2.853430188863830 2.457420654727494 -7.850599568137761 -0.023056208556438 0.979290073865177 0.004844083255961 0.201086790549622
tetra FFFF0000 7.803246588194591 2.411438089106807 9.589071561621216 0.432359860640060 0.203436086519546 -0.425602737295751 -0.768466667862233
tetra FFFF0000 -2.099270862669440 2.391264618785062 -2.843793457729366 0.001466715796809 0.946015275588607 0.018859014706699 -0.323569597864317
tetra FFFF0000 -2.369614785502925 2.423157410402030 2.383081937891202 -0.139021299159943 0.810428556477273 -0.532570860694564 -0.200616329258352
tetra FFFF0000 7.843901449744857 2.432189681952493 -0.265007164523572 0.419037797024265 0.231895089425307 -0.437701688938800 -0.760952839317656
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tetra FFFF0000 -3.441338198893986 -2.361112741932193 8.209916772627624 0.744314786178763 0.290527903870296 0.377303796308377 0.468220975010391
tetra FFFF0000 -1.625553903328091 -2.290222290619773 8.801937262960410 0.147398574496449 0.631177577930373 0.743062255050997 -0.166574339178906
tetra FFFF0000 4.341362217363210 -2.337163073361825 0.614701789863041 -0.581076897133441 0.207949172948135 0.446060195438258 -0.648179823145971
tetra FFFF0000 9.236986751475749 -2.528085758492844 5.710832433009003 -0.390829499440638 0.461090623197558 0.723877550478846 -0.332639491760997
tetra FFFF0000 -0.767935454072553 -2.479611813687495 -4.146168528390935 -0.397587845906165 0.464206886792605 0.720675925154728 -0.327203425936207
tetra FFFF0000 4.238589336360546 -2.371679109492621 -9.133254620417667 -0.613981839669195 0.184501419377730 0.419912642767618 -0.642385319919400
tetra FFFF0000 6.565661218763045 -2.373439859329669 8.266720754508460 0.731362827099567 0.302603708375997 0.409082841918940 0.454082194389460
tetra FFFF0000 1.670396127146395 -2.366138743688324 -6.527644587705836 0.453947860332053 0.658960654090252 0.579008902751991 0.156367793982119
tetra FFFF0000 -0.669342436248618 -2.454190266341454 5.550569047202194 -0.412921422485053 0.470216297885706 0.699558464771591 -0.344978965187729
tetra FFFF0000 6.671745487403873 -2.354750248832981 -1.657348238511313 0.745822014381656 0.294604004314494 0.378855123648126 0.461981382880302
tetra FFFF0000 -7.242643878651593 2.544087190722114 8.970654219184473 -0.293393157494236 0.378843707442506 0.738553062768432 -0.474275525345919
tetra FFFF0000 -9.642265605024276 -2.578495627823026 -1.679035179530498 0.523068391855574 0.551730583896774 0.645188929444077 0.075657554529300
tetra FFFF0000 -8.347421824593154 -2.586001075831008 9.368842104420994 0.803163528855695 0.349318417841759 0.342761492677921 0.339734525784179
tetra FFFF0000 -9.735857698717165 -2.565182228759846 8.117745362087296 0.520594002402777 0.548238093610625 0.651136310172726 0.066621189955538
tetra FFFF0000 -7.170599890405131 2.549980948710925 -0.690317753965651 -0.277071105068735 0.393799824863923 0.749582897660943 -0.454179237974787
tetra FFFF0000 -5.822357261423109 -2.582698509201728 4.338025140350798 -0.335242632921199 0.356125813629201 0.784517268754828 -0.381207865821873
tetra FFFF0000 1.746094216002231 -2.556611902442719 9.459301219463585 0.818140359874613 0.341168152571276 0.318897966257505 0.335491773866616
tetra FFFF0000 -8.295192912764747 -2.584395390901583 -0.421402386460342 0.812832225409504 0.347388728865717 0.322865739486484 0.338204850853376
tetra FFFF0000 -4.327624866446451 2.575095665651418 5.697884130217448 -0.601332215821527 0.104630740364578 0.542956362908406 -0.576965902791581
tetra FFFF0000 0.389494089502717 -2.544597262824960 -1.694940889844645 0.511690977416034 0.551449329134836 0.655366105728915 0.067611008641193
tetra FFFF0000 -5.911075786952734 -2.533485500484303 -5.498541933232143 -0.311974160607164 0.364810071894808 0.790158842055856 -0.381096757895564
tetra FFFF0000 -9.351884738643188 2.513299999440539 -2.798343098380527 -0.689916801452872 0.138314652654103 0.500239722599085 -0.504622714379964
tetra FFFF0000 4.219338555171120 -2.553596287807105 4.337452853813563 -0.339902934969168 0.357596910851959 0.780608073146856 -0.384817158408294
tetra FFFF0000 2.989505169995194 2.593334893554811 4.565404208838261 0.740304687292038 0.482035533915351 0.264517199580486 0.386809210260663
tetra FFFF0000 -4.311158932773361 -2.585119540777645 -4.266361495818183 -0.590826815894939 0.117413827838316 0.548744198732871 -0.579670139828525
tetra FFFF0000 0.394123063587221 -2.545552326163515 8.210418547561909 0.525914391032781 0.556355817832193 0.637826365982804 0.084022521540718
tetra FFFF0000 4.315451440930419 -2.562383840965269 -5.496684388149269 -0.342014521512394 0.360832973463682 0.779768731416523 -0.380508025987722
tetra FFFF0000 -9.414084858033110 2.506689760898897 7.002551552794424 -0.698037590157069 0.141747599800504 0.490142864343879 -0.502405327610372
tetra FFFF0000 -7.174539406526942 -2.590754066768112 -5.268656910790956 0.712540312631642 0.502334058772559 0.296510564678952 0.389908042114624
```

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```
tetra FFFF0000 5.782548437289897 2.591075910447661 -4.126624836186291 -0.604433528294719 0.099881280342371 0.542715390415985 -0.574581451768147
tetra FFFF0000 5.691700999047301 2.582022574718054 5.679570468489615 -0.602436343682148 0.101283583814408 0.539412449187966 -0.579522473353394
tetra FFFF0000 -7.007999279329397 -2.595313237078896 4.547587155944033 0.743628274185108 0.490615057669189 0.253671184355630 0.376782145607178
tetra FFFF0000 2.871120689451793 2.563199085058656 9.198140148343240 -0.273347744650279 0.400812628284607 0.748346788332464 -0.452335419677341
```

C.2.7   The Disordered Glass

The disordered glass obtained in a system of 8,000 tetrahedra and compressed to a packing fraction of 78.58%, the densest known packing of disordered tetrahedra (Fig. 5.6):

```
boxMatrix 29.809106661 0 0 0 30.354809922 0 0 0 30.001907762
def S0 "poly3d 4 1 1 1 1 -1 -1 -1 1 1 -1 -1 -1 1"
S0 ffff0000 -12.280534804 -13.776151837 -12.636977685 0.561308761 -0.31718098 0.342345194 -0.683467972
S0 ffff0000 -11.53457426 -13.32490531 -14.477024052 0.499857058 0.102774551 -0.095963683 -0.854617625
S0 ffff0000 -14.684439874 -14.191529521 -13.41926109 -0.531044913 -0.777467484 0.334331825 0.041926643
S0 ffff0000 -13.455723235 -11.973899655 -14.223451258 -0.622549806 -0.383930436 -0.407672564 0.546655504
S0 ffff0000 -13.484130313 -12.716632333 -12.690103475 -0.052771742 -0.353591547 -0.881530667 -0.308369654
S0 ffff0000 -14.205284333 -14.060496848 -12.370286953 -0.122514561 -0.726353011 0.597729604 -0.31641872
S0 ffff0000 -14.082518658 -13.007230784 -14.462132333 0.319939361 -0.464718268 -0.81001249 0.159860888
S0 ffff0000 -11.586720375 -11.627607548 -12.718897843 -0.216754167 0.719765085 -0.373201171 -0.543761657
S0 ffff0000 -12.121135523 -14.445429577 -11.635097452 -0.127583998 -0.916595904 -0.3149875 0.210611363
S0 ffff0000 -13.863672726 -14.894511018 -14.05172989 -0.392331377 -0.455944785 -0.712945095 -0.360416057
S0 ffff0000 -11.463801207 -12.847694569 -12.877794526 -0.387219417 -0.25527529 -0.352002627 0.813012792
S0 ffff0000 -12.567125055 -14.441883354 -13.590589859 -0.685478259 -0.311840202 0.650765733 -0.096846301
S0 ffff0000 -11.483323475 -12.066308748 -9.612842997 0.331343314 0.87017851 0.063832707 0.359063163
S0 ffff0000 -14.895288824 -12.663927262 -8.995008632 0.993941801 0.036408612 0.082592098 0.062710878
S0 ffff0000 -11.837875378 -11.638988356 -8.248576242 0.130736456 -0.447093031 -0.310944559 0.828449927
S0 ffff0000 -12.285613086 -12.694601924 -7.78897287 -0.005658152 -0.297670958 0.934608448 0.19459454
S0 ffff0000 -14.660058755 -14.340853114 -9.055798335 0.726758055 0.087733007 -0.627541843 0.265173309
S0 ffff0000 -14.097462191 -11.583226601 -9.495632303 0.196267754 -0.787394787 -0.568061837 0.137091821
S0 ffff0000 -14.199999699 -15.013074386 -7.777254356 0.51730816 0.126780367 -0.103479616 -0.840006533
S0 ffff0000 -11.759127149 -14.205081172 -8.81328894 -0.683951084 -0.316454013 0.60136812 0.2653755
S0 ffff0000 -13.525708437 -14.542694702 -9.918222387 -0.220373729 0.954365146 -0.132298939 0.152051232
S0 ffff0000 -14.050343989 -13.287273799 -8.514106963 0.859947172 -0.354491772 -0.360353981 0.070508532
S0 ffff0000 -14.361813611 -12.237068543 -10.631030867 0.665835538 -0.684757087 -0.295578717 -0.020099518
S0 ffff0000 -12.857957916 -11.818915092 -10.937260725 0.380955208 -0.001028226 0.771193148 0.510032548
S0 ffff0000 -14.209504893 -14.731267693 -10.936036338 0.903639616 -0.286391074 -0.165401073 0.272136147
S0 ffff0000 -12.578515183 -13.400272584 -8.667182352 0.102266805 0.438746539 -0.046261182 0.89157326
S0 ffff0000 -12.104570554 -13.129912743 -10.075251029 0.541423621 -0.775054448 -0.22938388 0.231374373
S0 ffff0000 -13.008238218 -13.15098694 -10.952354454 -0.14868363 0.549999782 -0.783190427 -0.249010389
S0 ffff0000 -13.684783593 -15.132005016 -4.112569862 -0.501431567 0.076639675 -0.259199518 0.821893152
S0 ffff0000 -11.582677564 -15.011426682 -6.474412953 -0.411607168 -0.892186525 0.1856815 -0.01025304
S0 ffff0000 -14.200265073 -13.770522025 -5.330949387 -0.37969556 0.263849577 0.791703988 0.399273687
S0 ffff0000 -12.454406714 -12.652935894 -4.9364481 -0.161920551 -0.440827995 0.830621052 0.299200738
S0 ffff0000 -12.976600431 -13.146382019 -3.859921472 -0.018139014 -0.963008114 0.0318139 0.266972328
S0 ffff0000 -14.04332278 -13.162813707 -7.239552377 -0.866790209 -0.195893714 0.453465659 0.068332153
S0 ffff0000 -13.885631839 -13.808053586 -4.141624642 -0.662990994 0.109156195 -0.609137081 0.421283616
S0 ffff0000 -14.479156751 -11.577932054 -6.355479101 -0.365420639 0.137285468 -0.030409194 0.920160713
S0 ffff0000 -13.411160628 -11.523882667 -4.787723584 -0.318605854 0.055233833 -0.890473284 -0.320151314
S0 ffff0000 -11.538397923 -13.634140354 -6.274193762 0.353864606 0.841773498 0.406999293 -0.023426365
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S0 ffff0000 -14.233778187 -14.879057837 -5.862046092 0.10545583 -0.216632078 -0.961436934 0.132622145
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S0 ffff0000 -13.378922537 -11.466729769 -5.959955375 -0.420635652 0.340471816 -0.816095162 0.202813404
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S0 ffff0000 -12.305823366 -14.610550659 -1.684422783 -0.3360416 -0.687524506 -0.554283701 -0.327346416
S0 ffff0000 -14.413965136 -12.666976537 -2.641472596 -0.765243275 -0.272966974 -0.356504827 -0.461298244
S0 ffff0000 -13.327281043 -12.386760304 -2.930546669 -0.206117647 0.413315371 -0.882292196 -0.090809698
S0 ffff0000 -13.493428015 -14.089755905 -0.360998447 -0.373146557 0.334300441 -0.403415141 0.765676881
S0 ffff0000 -14.166598471 -14.671032578 -1.098021814 -0.596692222 -0.794402523 -0.113450769 -0.003456254
S0 ffff0000 -14.854791023 -13.114219823 -1.269151123 -0.467391098 -0.609310166 0.273738631 -0.57909744
S0 ffff0000 -14.023474899 -13.062347934 -0.245280391 -0.136307583 -0.654875733 -0.60572022 0.430884013
S0 ffff0000 -12.136867124 -12.587198105 -2.097515618 -0.531582392 0.051181204 -0.790085658 0.300940688
S0 ffff0000 -11.925216258 -13.673103904 -2.437554685 0.329706377 -0.1562388 0.503008807 -0.783495554
```

```
S0 ffff0000 -14.704745593 -13.515368963 -3.381783982 -0.834579389 -0.009591281 -0.054576571 0.548093649
S0 ffff0000 -12.532678108 -12.703085378 -1.010037232 0.869412183 -0.457085439 0.036798589 0.183959836
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S0 ffff0000 -12.389742178 -13.961385986 -0.732179977 0.293666719 0.188520713 0.135724616 -0.927253271
S0 ffff0000 -11.656725606 -13.91141107 2.628842087 -0.885506828 -0.061871144 -0.412578958 -0.204519492
S0 ffff0000 -11.375233013 -13.979539061 0.171686044 0.616497211 -0.0646127 0.772647645 -0.137010965
S0 ffff0000 -14.082476138 -12.143421546 1.758493133 -0.797931814 -0.133649439 0.122002139 0.574941845
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S0 ffff0000 -14.373525001 -14.090166772 1.936332948 0.161691981 0.956946601 -0.134925574 0.199759844
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S0 ffff0000 -14.001352102 -12.766089851 6.873761757 -0.144876507 0.609383958 -0.172193449 0.760270613
S0 ffff0000 -14.463899886 -11.537395621 5.072617528 0.5184096 -0.356538445 0.277774845 0.725929032
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S0 ffff0000 -13.524284414 -12.933939899 4.238335516 -0.079453898 0.913995111 -0.397350106 -0.020320143
S0 ffff0000 -11.717098355 -13.733304498 3.78750853 -0.799287941 -0.03604912 0.520014025 0.299039564
S0 ffff0000 -11.454563314 -12.058184049 4.526344967 -0.978174655 -0.041505955 -0.18026613 -0.094634674
S0 ffff0000 -12.11135739 -13.267380125 6.541538588 -0.93300323 -0.245614997 -0.182827594 0.189082831
S0 ffff0000 -12.603794726 -14.996924522 5.80563352 0.151605792 -0.790375946 0.030963833 -0.592758626
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S0 ffff0000 -13.198564582 -13.139208973 6.132082721 0.201207564 -0.609917735 0.440948412 0.62696122
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S0 ffff0000 -12.202924089 -14.641060759 8.698013216 -0.236130392 0.950809959 -0.16651658 -0.111691935
S0 ffff0000 -13.756696946 -14.137608375 9.609186725 0.491291281 0.081180804 -0.791474898 0.354415068
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S0 ffff0000 -14.176291103 -14.237915786 8.530711798 0.682401863 0.128236448 -0.302194389 0.653116883
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S0 ffff0000 14.67314587 13.578540464 4.240897396 0.622679189 -0.633193395 -0.050009697 -0.456985538
S0 ffff0000 11.19364482 14.701100342 10.493105836 0.963135236 0.089374391 0.193372362 0.164285923
S0 ffff0000 12.488766154 13.46218209 10.327746807 0.420730291 0.071853818 -0.612746866 -0.665104751
S0 ffff0000 13.238905351 13.316383158 9.386501216 -0.628736781 0.257416785 0.594629534 -0.429932992
```

```
S0 ffff0000 14.103618491 12.555126856 9.50192805 0.007509965 -0.329902824 0.146660028 0.932522688
S0 ffff0000 11.206675895 12.620122619 8.842904166 0.327159096 0.141564901 0.861984985 -0.360427787
S0 ffff0000 12.760867316 11.684680395 8.026466902 -0.600305039 -0.25756919 0.514697002 0.555318799
S0 ffff0000 12.556981781 13.543609437 7.815954352 -0.047422406 -0.745614685 0.623438638 0.230508396
S0 ffff0000 11.42937266 14.109652289 7.576649421 -0.529324907 0.788320084 0.301047933 0.087958682
S0 ffff0000 14.793064029 14.398766739 8.939333008 0.09963611 0.44147247 0.425382424 -0.783724759
S0 ffff0000 12.769229412 12.732498183 11.205805525 -0.172626459 -0.068308571 -0.678955619 -0.710319162
S0 ffff0000 12.319139511 12.660146752 8.525358429 -0.136056755 0.395943718 0.683970761 -0.597412027
S0 ffff0000 13.250082782 14.888423561 7.854319017 -0.975399349 0.18641576 0.111210687 0.03843772
S0 ffff0000 13.269977579 11.882581796 10.589855637 -0.264920529 -0.116946474 0.927131498 -0.237839905
S0 ffff0000 12.819489483 14.467040985 10.790607413 0.137056945 -0.101333691 0.964971532 -0.19944127
S0 ffff0000 13.274790291 15.130652922 9.677680925 -0.061984655 0.786364612 0.609702326 -0.077792502
S0 ffff0000 13.612799007 14.320732623 8.927252981 -0.210675636 0.850630573 -0.418442397 0.238640661
S0 ffff0000 12.440565946 14.945087711 12.513546059 0.248325125 0.788507097 -0.540334074 -0.156940366
S0 ffff0000 12.964266623 11.489531714 14.220877952 -0.457476157 -0.77688884 0.398404082 -0.168622309
S0 ffff0000 14.303511293 12.742716452 12.159464073 0.684247176 0.206293048 0.405297534 0.570072706
S0 ffff0000 14.419342247 12.070075866 13.206519923 0.57511842 -0.425838373 -0.697451312 -0.038238066
S0 ffff0000 14.283181355 11.838026541 11.431128039 -0.076898681 0.394063225 0.915609777 -0.021436028
S0 ffff0000 11.585799353 12.253667554 11.485087709 -0.298373716 -0.11467388 0.24140145 -0.91626872
S0 ffff0000 12.65535455 13.062248909 13.165366768 0.136541215 -0.085383602 -0.936914378 0.310254066
S0 ffff0000 13.364620598 13.430453623 12.062372292 -0.480945931 -0.586073494 0.240785831 0.60599592
S0 ffff0000 13.688501669 14.47098425 11.680386532 -0.640411417 -0.635213504 -0.054879784 -0.428211665
S0 ffff0000 12.497164447 14.244585009 13.471313401 0.233673311 0.538482346 -0.091620788 0.804387455
S0 ffff0000 13.383217264 12.75446229 14.055604859 0.120089512 0.269707795 -0.657283478 -0.693407992
S0 ffff0000 13.624386062 14.860809821 14.022380048 -0.387848082 0.361341921 -0.354386062 -0.770335252
S0 ffff0000 11.639617298 12.270943712 13.013167492 0.291465977 0.013435887 0.926994673 0.2356861
S0 ffff0000 14.750734474 14.319955288 14.331182692 0.309650065 -0.198794502 0.927073982 -0.071633898
```

C.3 Dense Packings of Hard TBPs

C.3.1 Regular Approximant

The unit cell of the regular approximant is numerically compressed to a maximum packing fraction of 83.39%. This packing is presented below:

```
boxMatrix 9.96864731 0 0 0 10.12720286 0 0 0 2.597424124
def S0 "poly3d 5 0 0 2.30940108 0 0 -2.30940108 1.63299316 0 0 -0.81649658 1.41421356 0 -0.81649658 -1.41421356 0"
S0 ffff0000 -3.086054086 -2.363988414 0.413277757 0.724311194 0.366868384 0.073525829 0.579115564
S0 ffff0000 -4.11840487 -0.892236112 0.415845679 0.432135294 -0.510211566 0.631248159 0.393025455
S0 ffff0000 -3.924682848 -3.466017697 -0.350811567 -0.318217496 0.434494644 0.725846175 0.427901111
S0 ffff0000 -0.462122468 -4.240376441 -0.782025752 0.852598155 0.38776541 -0.303556385 0.174836762
S0 ffff0000 -0.327948406 -1.568864202 -1.104708664 0.131985547 -0.297203708 -0.551333205 0.768297773
S0 ffff0000 -2.672857633 0.001417971 -0.337285874 -0.555926418 -0.431797813 0.706493586 -0.073234406
S0 ffff0000 -2.069478657 -1.471298448 -0.432573444 0.177650291 0.552837588 0.096194039 0.808429145
S0 ffff0000 -0.826248624 -0.277415522 -0.173475293 0.812731912 0.302601899 -0.36038252 -0.343545295
S0 ffff0000 -2.318916062 -4.112336247 0.659756236 -0.410542691 -0.392643954 0.520886557 0.637151959
S0 ffff0000 -1.344851368 -2.859967477 0.633186749 0.636896655 0.725358601 0.219148044 0.142097453
S0 ffff0000 -4.696641186 1.276225643 0.602240973 0.360108299 0.252500774 0.269483792 0.856705234
S0 ffff0000 -3.747047319 5.05528104 -1.103638744 0.780999587 -0.449388327 0.430045356 -0.056131712
S0 ffff0000 -0.05899754 4.025246097 -0.779658046 -0.218095268 0.293099666 0.406142214 0.837601063
S0 ffff0000 -0.019924758 1.968363142 -0.681742157 -0.575391179 0.092652143 0.548061557 0.59997425
S0 ffff0000 -2.64948323 4.205046998 -0.356921625 0.50778325 -0.424324513 0.582993354 0.471406013
S0 ffff0000 -1.457204318 4.260893847 1.190217106 0.729944614 -0.577104503 0.065356154 0.360360689
S0 ffff0000 -1.778009861 2.641765034 -0.715619163 0.666191766 0.458836083 -0.016867546 0.587684836
S0 ffff0000 -4.267156145 3.386854105 -0.471378412 -0.000224002 0.782668146 0.478172787 0.398473723
S0 ffff0000 -1.197762201 1.002543156 -1.100736732 0.473332539 -0.577185196 -0.123466505 0.653888048
S0 ffff0000 -2.94412608 1.001577617 0.918738785 0.691606294 0.561167807 -0.315127681 -0.327820027
S0 ffff0000 -3.431835202 2.531003145 0.76732624 0.489058954 0.502023864 0.569884742 0.428981072
S0 ffff0000 4.166061318 -1.755893743 0.614482584 0.857339372 0.268730116 -0.248620481 -0.361857958
S0 ffff0000 4.707576086 -0.23855974 -0.536923289 0.079504631 -0.419075935 -0.566678392 0.704932603
S0 ffff0000 4.456114027 -2.99420821 -0.34284377 0.780466896 0.401090725 -0.425640796 0.220969608
S0 ffff0000 4.827932349 -4.702316768 -0.159021811 -0.126511507 0.202433383 0.41472186 0.87807821
S0 ffff0000 3.567745446 -4.59466625 -1.082778182 0.754021412 -0.607585046 -0.001432754 0.249579784
S0 ffff0000 1.938503081 -3.80573308 -0.665797086 -0.348135133 -0.057177952 0.878347065 0.322550839
S0 ffff0000 0.916420109 -4.715357725 1.157640853 -0.342223016 -0.075509837 -0.566097787 0.746133344
S0 ffff0000 0.595676354 -2.632461476 -0.391055241 0.686531636 -0.073835559 0.706109295 0.156946761
S0 ffff0000 1.361780627 -1.041508444 -0.497237432 -0.218226841 0.281976797 0.279418344 0.891510808
S0 ffff0000 2.522729926 -1.36697548 0.913207732 0.340971281 0.338907933 0.555567249 0.678398872
```

```
S0 ffff0000 2.66630994 -2.95070587 0.792567704 0.396438787 0.527197369 -0.529906306 -0.533008939
S0 ffff0000 2.576140986 2.039647502 0.467914212 -0.571761301 -0.070597781 0.373290369 0.727158351
S0 ffff0000 3.524756753 0.477954765 0.412277663 0.3506445 0.670036628 0.463160891 -0.462148614
S0 ffff0000 3.992316141 3.709275417 0.053283182 0.183126889 0.7896533 0.085915702 -0.579250119
S0 ffff0000 4.19922719 2.276875385 -0.60051038 -0.359557729 0.681364585 0.265991831 -0.579403908
S0 ffff0000 2.385703449 4.617199613 -0.648455546 0.236634396 -0.425417036 0.711443254 -0.506826404
S0 ffff0000 0.932212413 0.55991917 0.041230498 0.541559035 -0.498432529 0.611824825 0.2897399
S0 ffff0000 1.870351885 3.740201639 0.686805299 0.635673515 0.523964733 0.374951541 0.425195818
S0 ffff0000 0.852464474 2.539550949 0.649899985 -0.138338952 -0.219309086 0.717949504 0.645998737
S0 ffff0000 2.376730451 0.544997559 -0.642579527 -0.670448515 0.706351286 -0.038435571 -0.223806515
```

C.4 Dense Packings of Hard Truncated TBPs

No analytical expression could be obtained for tTBP dense packings obtained for $0.52 \leq t \leq 0.58$. We therefore present the numerically compressed packings of tTBPs for $t = 0.52, 0.55$ and 0.58.

C.4.1 $t = 0.52$

The following periodic structure has a packing fraction of 85.5156%.

```
boxMatrix 0.929783738 0.42643432 0.397564256 0.006695808 0.376996507 -0.668619644 -0.784291171 1.083915404 0.076143193
def S0 "poly3d 18 0.352183664 0.13 0 -0.06350853 0.37 0 0.352183664 -0.13 0 -0.06350853 -0.37 0 0.427239199 0 0.212289111 0.15011107
0 0.60420747 0.427239199 0 -0.212289111 0.15011107 0 -0.60420747 -0.288675135 0.24 0 -0.288675135 -0.24 0 -0.2136196 0.37 0.212289111
-0.075055535 0.13 0.60420747 -0.2136196 0.37 -0.212289111 -0.075055535 0.13 -0.60420747 -0.2136196 -0.37 0.212289111 -0.075055535
-0.13 0.60420747 -0.2136196 -0.37 -0.212289111 -0.075055535 -0.13 -0.60420747"
S0 82a917 0.384617993 0.10249313 -0.313383768 -0.27034337 -0.395456186 0.185847616 0.857898322
S0 58d3e8 -0.021024708 0.407605411 0.472241693 0.372187069 -0.65574116 0.648512879 0.10445747
S0 17732c 0.06959559 -0.291883464 -0.011523909 0.194825019 -0.435398734 -0.036779481 0.878133489
S0 5d1f48 0.077673933 0.071623816 -0.873292339 0.104457336 -0.648512835 -0.655741124 -0.372187246
```

C.4.2 $t = 0.55$

The following periodic structure has a packing fraction of 86.7102%.

```
boxMatrix -0.274357602 0.933044336 -0.652226465 0.962757852 -0.48917934 -0.351893014 -0.668788071 -0.557484339 -0.239005248
def S0 "poly3d 18 0.339193283 0.1375 0 -0.050518149 0.3625 0 0.339193283 -0.1375 0 -0.050518149 -0.3625 0 0.418578945 0 0.22453656
0.158771324 0 0.591960021 0.418578945 0 -0.22453656 0.158771324 0 -0.591960021 -0.288675135 0.225 0 -0.288675135 -0.225 0 -0.209289473
0.3625 0.22453656 -0.079385662 0.1375 0.591960021 -0.209289473 0.3625 -0.22453656 -0.079385662 0.1375 -0.591960021 -0.209289473
-0.3625 0.22453656 -0.079385662 -0.1375 0.591960021 -0.209289473 -0.3625 -0.22453656 -0.079385662 -0.1375 -0.591960021"
S0 82a917 0.414124063 -0.274653502 0.206132948 0.225121897 0.622770188 -0.667487621 0.340496257
S0 58d3e8 -0.219950272 -0.061363168 -0.310346482 0.477825091 -0.104685974 0.830322583 -0.266998945
S0 17732c -0.320123873 0.240942531 0.182223665 -0.324500609 0.547308128 0.007684822 -0.771423432
S0 5d1f48 -0.027614183 -0.156631908 0.573537654 -0.340496203 -0.667487646 -0.622770236 0.225121771
```

C.4.3 $t = 0.58$

The following periodic structure has a packing fraction of 87.6521%.

```
boxMatrix 1.103113851 -0.318715935 -0.131822416 -0.346049334 -0.415583696 -0.926908294 -0.164412273 0.574742671 -0.743072049
def S0 "poly3d 18 0.326202902 0.145 0 -0.037527768 0.355 0 0.326202902 -0.145 0 -0.037527768 -0.355 0 0.409918691 -0 0.236784009 0.167431578
-0 0.579712573 0.409918691 -0 -0.236784009 0.167431578 -0 -0.579712573 -0.288675135 0.21 0 -0.288675135 -0.21 0 -0.204959346 0.355
0.236784009 -0.083715789 0.145 0.579712573 -0.204959346 0.355 -0.236784009 -0.083715789 0.145 -0.579712573 -0.204959346 -0.355
0.236784009 -0.083715789 -0.145 0.579712573 -0.204959346 -0.355 -0.236784009 -0.083715789 -0.145 -0.579712573"
S0 82a917 0.062907082 -0.208212244 -0.580032881 0.25458918 0.768594616 0.154611703 0.566164188
S0 58d3e8 0.468530137 -0.106102771 0.244497674 -0.474227861 0.314824403 0.783272267 0.24995617
S0 17732c -0.113183194 -0.060585028 0.228112568 0.28571525 0.118990172 -0.83574588 -0.453582362
S0 5d1f48 0.479333701 -0.469800435 -0.265171466 -0.154611873 -0.56616429 0.254589255 0.768594481
```

APPENDIX D

Different ways of Calculating Diffraction Patterns

In all diffraction images presented in this thesis, scatters are placed at centroids of tetrahedra. Another alternative, which might be of greater experimental relevance, is to put scatters at the vertices of tetrahedra rather than at their centroids. We used both these approaches in the beginning of this study, we however obtained no further information from the latter approach. For instance, Fig. D.1 shows the diffraction patterns calculated for the quasicrystal formed in a system of $N = 8{,}000$ tetrahedra using these different choices of scatterer positions. The diffraction patterns are very similar, but there are minor differences between them, mostly in the intensity of peaks. We therefore adopted the convention of using centroids as scatterers noting that general features of the diffraction pattern will not be affected by the particular choice of the scatterer positions. We therefore expect similar diffraction images to be observed in experimental systems.
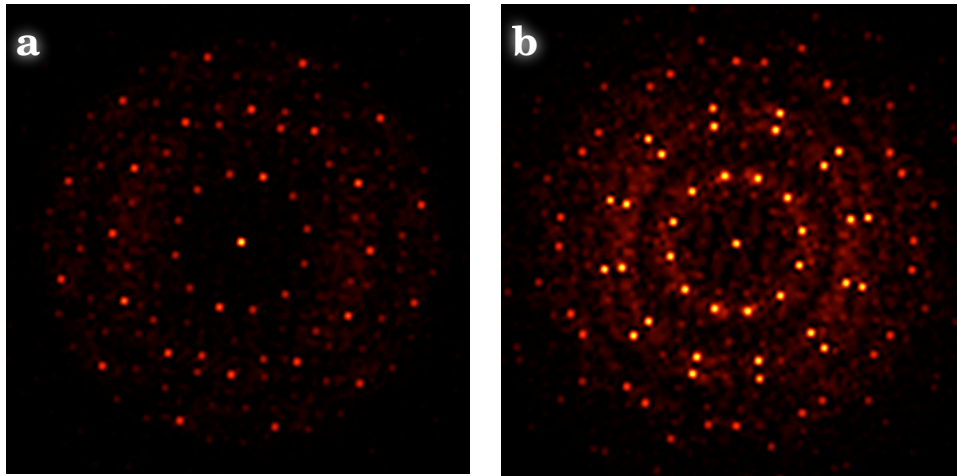
Figure D.1:
**Effect of scatterer selection on diffraction patterns of hard tetra-
hedra.** Diffraction patterns of the dodecagonal quasicrystal formed in a
system of $N = 8{,}000$ hard tetrahedra calculated by putting scatterers at
(a) the centroids and (b) the vertices of tetrahedra.

APPENDIX E

Dense Packings of Truncated Triangular Bipyramids with Two Particles Per Unit Cell

In this appendix we present the analytically constructed dense packings of truncated TBPs with two particles per unit cell. The vertices of a non-truncated (perfect) TBP are given by:

$$(+2, +2, +2) \tag{E.1a}$$

$$(+2, -1, -1) \tag{E.1b}$$

$$(-1, +2, -1) \tag{E.1c}$$

$$(-1, -1, +2) \tag{E.1d}$$

$$(-2, -2, -2) \tag{E.1e}$$

Each unit cell contains two truncated TBPs with opposite orientations $\mathcal{T}$ and $-\mathcal{T}$. The box vectors of the unit cell are given by $\mathbf{u} + \mathbf{v}, \mathbf{v} + \mathbf{w}$ and $\mathbf{w} + \mathbf{u}$ respectively with $\mathcal{T}$ residing at the center of the unit cell and $-\mathcal{T}$ residing at $\mathbf{t}$. Here we present $\mathbf{u}, \mathbf{v}, \mathbf{w}, \mathbf{t}$ and $\phi_{\max}$ for different values of truncation.

E.1 The Densest Packing for $0 \le t \le 0.2544$

-	x	y	z
u	$\dfrac{t+87}{32}$	$\dfrac{321-25t}{320}$	$\dfrac{3(15t-7)}{320}$
v	$-\dfrac{5t+51}{160}$	$\dfrac{831+25t}{320}$	$-\dfrac{9(t-9)}{64}$
w	$\dfrac{3(25t+47)}{160}$	$\dfrac{145t-249}{320}$	$\dfrac{35t+741}{320}$
t	$\dfrac{5t+19}{160}$	$\dfrac{7t+1}{64}$	$-\dfrac{3t+5}{64}$
$\phi_{\max}(t) = \dfrac{4000(1-3t^3/8)}{4671-30t+25t^2}$			

E.2 The Densest Packing for $0.2544 \le t \le \dfrac{1}{3}$

-	x	y	z
u	$\dfrac{19t+88}{36}$	$\dfrac{67}{36}-\dfrac{23t}{9}$	$\dfrac{1-23t}{36}$
v	$\dfrac{16-101t}{36}$	$\dfrac{103-32t}{36}$	$\dfrac{t+37}{36}$
w	$\dfrac{19t+40}{36}$	$\dfrac{4t}{9}-\dfrac{17}{36}$	$-\dfrac{97-23t}{36}$
t	$\dfrac{5(7t-2)}{36}$	$\dfrac{5(1-2t)}{36}$	$\dfrac{5(1-5t)}{36}$
$\phi_{\max}(t) = \dfrac{216(1-3t^3/8)}{238+131t-319t^2+64t^3}$			

E.3 The Densest Packing for $\dfrac{1}{3} \leq t \leq 0.5010$

-	x	y	z
u	$\dfrac{97 - 14t}{36}$	$\dfrac{2(5 - t)}{9}$	$-\dfrac{2t + 17}{36}$
v	$-\dfrac{1 - 38t}{36}$	$\dfrac{2(t + 22)}{18}$	$\dfrac{67 - 62t}{36}$
w	$\dfrac{37 - 2t}{36}$	$\dfrac{4 - 17t}{9}$	$\dfrac{103 - 26t}{36}$
t	$\dfrac{5(1 - 2t)}{36}$	$\dfrac{5(2t - 1)}{18}$	$\dfrac{5(1 - 2t)}{36}$
$\phi_{\max}(t) =$	$\dfrac{108(1 - 3t^3/8)}{(1 + t)(119 - 80t + 8t^2)}$		

E.4 The Densest Packing for $0.5776 \leq t \leq \dfrac{2}{3}$

-	x	y	z
u	$3 - \dfrac{5t}{2}$	$6 - \dfrac{11t}{2}$	$\dfrac{6 - 7t}{4}$
v	$-t$	$6 - \dfrac{11t}{2}$	$\dfrac{18 - 13t}{4}$
w	$2t$	$\dfrac{t}{2}$	$\dfrac{5t + 6}{4}$
t	$\dfrac{7t - 2}{4}$	$7t - 5$	$\dfrac{5t}{2} - 2$
$\phi_{\max}(t) =$	$\dfrac{16(1 - 3t^3/8)}{3(t - 2)(7t^2 - 4t - 4)}$		

E.5 The Densest Packing for $\dfrac{2}{3} \le t \le 3 - \dfrac{\sqrt{105}}{5}$

-	x	y	z
u	$\dfrac{t+130}{56}$	$\dfrac{22-5t}{14}$	$\dfrac{118-149t}{56}$
v	$\dfrac{94-113t}{56}$	$\dfrac{34-23t}{14}$	$\dfrac{106+37t}{56}$
w	$\dfrac{250-179t}{56}$	$\dfrac{5t-22}{28}$	$\dfrac{t+130}{56}$
t	$\dfrac{29t-38}{56}$	$\dfrac{50-47t}{28}$	$\dfrac{65t-62}{56}$
$\phi_{\max}(t) = \dfrac{224(1-3t^3/8)}{3(t-2)(65t^2-124t+4)}$			

E.6 The Densest Packing for $3 - \dfrac{\sqrt{105}}{5} \le t \le 1$

-	x	y	z
u	$\dfrac{11}{5} - \dfrac{t}{2}$	$\dfrac{118-65t}{40}$	$\dfrac{29}{20} + \dfrac{5t}{8}$
v	$\dfrac{5t}{2} - \dfrac{13}{5}$	$\dfrac{46-5t}{40}$	$\dfrac{53}{20} + \dfrac{5t}{8}$
w	$\dfrac{5t}{2} - \dfrac{7}{5}$	$\dfrac{23t}{8} - \dfrac{73}{20}$	$\dfrac{17(5t+2)}{40}$
t	$-\dfrac{13(5t-6)}{20}$	$\dfrac{18-11t}{8}$	$\dfrac{t-6}{8}$
$\phi_{\max}(t) = \dfrac{160(1-3t^3/8)}{3(t-2)(35t^2-20t-52)}$			

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BIBLIOGRAPHY

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