ROLE OF NUMERICAL SIMULATIONS IN THE SEMICONDUCTOR HETEROSTRUCTURE TECHNOLOGY USING MOLECULAR BEAM EPITAXY

Jasprit Singh
Department of Electrical Engineering and Computer Science
University of Michigan, Ann Arbor MI 48109

K. K. Bajaj
Air Force Wright Aeronautical Laboratories
Avionics Laboratory, AFWAL/AADR
Wright-Patterson Air Force Base, OH 45433

(Received 26 March 1986)

The present and potential role of computer simulations in understanding the growth and fabrication of heterostructure interfaces grown by molecular beam epitaxy (MBE) is discussed. The important atomistic processes involved in MBE growth are identified and the ingredients of a computer simulation model are described. Key results on the effect of growth conditions on the quality of heterostructure interfaces are presented. Cation surface kinetics are shown to be critical in controlling the quality of the growth front and of the interfaces. Need for non-conventional growth approaches is identified and several different possibilities are explored.

1. INTRODUCTION

Advances in molecular beam epitaxy (MBE) have ushered in a new era of electronics technology based on thin metastable structures[1-4]. These structures are fabricated from two or more semiconductors and utilize the differences in bandedge line-ups to tailor the band structure of the composite structures. This has led to rapid advances in the areas of microwave and electro-optic devices for both fast and tailored response.

A number of theoretical and experimental studies of both optical and electronic properties of heterostructures (superlattices, quantum wells and heterojunctions) have been carried out. These studies have become a propelling force for the growth and fabrication of novel heterostructures. However, it must be recognized that even for the most studied semiconductor combination - GaAs/GaAlAs, the fabrication of high quality heterostructures is far from routine. The situation for more "difficult" heterostructures, such as InGaAs/InP, InAlAs/InGaAs, HgTe/CdTe, etc., is, of course, much worse. Since the success of any technology depends upon high yield, it is critical to understand, and if possible, overcome the growth/fabrication problems.

In heterostructures, as in conventional device structures, an important cause of poor performance is the presence of unwanted impurities, native defects, etc. However, an important additional reason for poor performance in heterostructures is the presence of interface roughness. Since quite often the active regions of heterostructure devices may be as narrow as 50 A (~20 atomic monolayers), interface perfection of up to one monolayer is required if the structure is to meet its expected potential.

Due to the stringent requirements of interface perfection in heterostructure technology, it is critical to understand the MBE growth process and the formation of interfaces. Also, since atomic scales (a few Angstroms) are critical, the atomistic nature of the growth/fabrication process must be understood. The problem of MBE growth is a many particle, far-from-equilibrium thermodynamics problem, representing an open system (number of particles not fixed). Due to the extremely complex nature of the problem, it is clear that microscopic details of the growth process can only be obtained by use of sophisticated computer simulation methods[5-11]. Considerable amount of analytical and Monte Carlo work has been carried out on the growth of a model cubic elemental system, within the framework of the so called "solid-on-solid" model, and also on elemental tetrahedral systems[5-7]. Although these studies have been pioneering in the use of computer simulations to study crystal growth, they were not carried out to study MBE growth and do not address the critical issues of this growth technique (discussed in Section IV). This paper focuses on Monte Carlo simulations carried out with the specific intent of understanding the MBE growth process. The potential role of computer simulations in

0749-6036/86/030185+11 $02.00/0
© 1986 Academic Press Inc. (London) Limited

MOLECULAR BEAM EPITAXY

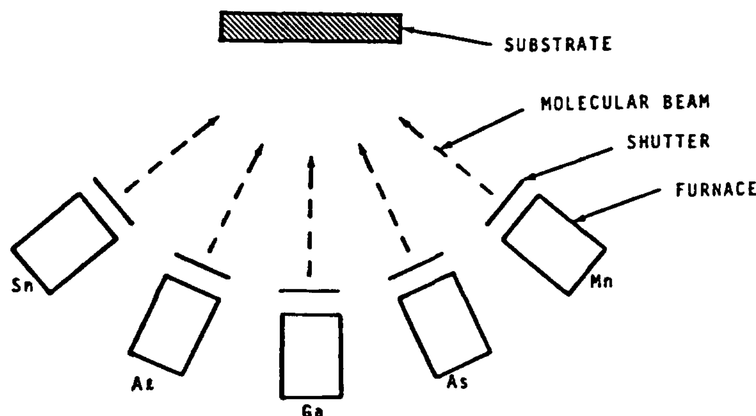

Fig. 1. A Schematic Diagram of the MBE Growth Setup.

heterostructure technology can be a very central one. Some of the key areas where simulations can make a significant impact are:

i) Identifying the key kinetic and energetic parameters controlling the growth process.

ii) Suggesting optimum growth conditions for fabrication of an interface from a given set of semiconductors.

iii) Providing insight towards the development of mathematical models for the microstructure of interfaces grown by MBE. These models could play a critical role in understanding the optical and transport properties in heterostructures as well as in identifying the effects of imperfect interfaces on device performance.

iv) Identifying semiconductor combinations which are likely to be "difficult" to grow with sharp interfaces using conventional MBE technology and suggesting novel approaches for their growth.

v) Suggesting key experimental studies [such as reflection high energy electron diffraction (RHEED) oscillations; masked growth, etc,] to understand the growth process and the determination of growth kinetics critical for the fabrication of high quality interfaces.

It is clear that information on these points will be extremely useful in the advancement of heterostructure technology. It also goes without saying that the theoretical/computational experience gained from these studies will be invaluable in developing a better understanding of other important processes in heterostructure technology such as doping, etching, contact formation, etc.

In Section 2 we will describe the conceptual picture of the MBE growth process and in Section 3 the details of the computer simulation are presented. Section 4 describes the key results obtained from growth simulations for the conventional MBE growth conditions, i.e., when the growth rate and growth temperature are kept fixed throughout the growth of the structure. In Section 5 we discuss the need for unconventional growth approaches and present results of computer simulations for several novel growth schemes. Finally, in Section 6 we discuss some important issues in MBE growth and the potential role of computer simulations.

2. Problem of Crystal Growth

A number of excellent articles[1-4] on the experimental aspects of MBE growth exist so we shall only briefly describe the growth process. Figure 1 shows a schematic picture of a MBE growth setup. Atomic cations (Ga, Al, etc.) and molecular anions (e.g., As_2) impinge on a heated substrate where they form chemical bonds and become part of the crystal. Interfaces are formed by opening/closing the relevant shutters.
Since the crystal growth rate is slow (~2.8 Å per sec.), the opening/closing of the shutters does not introduce any significant errors in the interface control. In order to understand the intrinsic growth mechanisms involved in the growth of a heterostructure, it is important to understand the energetics, the incorporation

process, and the surface kinetics of a semiconductor. Most of the discussion in this article will be devoted to III-V semiconductors.

A. Energetics of III-V Compounds

The thermodynamic aspects of the growing system are described by its energetics. For a simple lattice gas model, the energetics can be described by the following expression:

$$H = \frac{1}{2}\Big[\sum_{i_a, i_c} C_{ia} V_{AC} C_{ic} + \sum_{\substack{i_a, i_a' \\ 2^{nd} nn}} C_{ia} V_{AA} C_{ia}' + \sum_{\substack{i_c, i_c' \\ 2^{nd} nn}} C_{ic} V_{CC} C_{ic}'\Big] \qquad (1)$$

where C_{ia} and C_{ic} denote the occupation numbers at the anion and cation sublattices, respectively, and V_{AC}, V_{AA}, and V_{CC} are the bond energies for the nearest-neighbor, second-neighbor anion, and second-neighbor cation bonds, respectively. This simple description of the important energies involved can easily be extended to ternary and quaternary compounds, so that alloy growth can also be studied. It must be pointed out that in this simple model, the bond energies are additive (i.e., total energy for n bonds = n times the bond energy of a single bond). Relaxation of this condition is critical for studying the significance of surface reconstruction and growth of strained systems.

In addition to the bonding energies described by Eq. (1), one must realize that other energies describing the various possible defects (vacancies, antisites, etc.) can be important in determining the defect densities[10]. These energies play an important role in understanding high temperature growth. At high temperatures, due to extremely high atomic migration rates, the quality of the material grown is controlled by thermodynamic considerations. Defect densities in the system are then simply given by

$$N_D \propto \exp\left(-\frac{E_D}{kT_s}\right) \qquad (2)$$

where E_D is the activation barrier for defect formation and T_s is the substrate temperature. The value of E_D is determined using Eq. (1) and the corresponding equation when the defect is formed. For antisite defects this equation can be written as

$$H^D = \frac{1}{2}\Big[\sum_{i_a, i_a} C_{ia}' V_{AA}^{D1} C_{ia}' + \sum_{\substack{i_a, i_c' \\ 2^{nd} nn}} C_{ia} V_{AC}^{D2} C_{ic}' + \sum_{i_c, i_c'} C_{ic} V_{CC}^{D1} C_{ic}' \qquad (3)$$

Here V_{AA}^{D1} represents the bond energy of a nearest neighbor anion-anion bond, V_{CC}^{D1} that of a nearest neighbor cation-cation bond, and V_{AC}^{D2} that of a second neighbor anion-cation bond. There is little information available about these energies. They, however, can be quite crucial in determining the material quality. In this paper we will not concern ourselves further with the defect formation process.

B. Incorporation Process in MBE Growth

The process by which the atoms and molecules impinging from the vapor are incorporated into the growing crystal is a complex one and has been examined only for GaAs by Arthur[12], and be Foxon and Joyce[13-14]. These studies have been critical in developing an understanding of the incorporation process of atoms and molecules. Computer simulations using Monte Carlo methods have also provided a great deal of insight into the atomistic nature of MBE growth[8-11]. From the combination of experimental results and computer simulations a conceptual picture of the MBE growth process involving atomic Ga and dimer Arsenic (As_2), shown in Fig. 2, has emerged for the MBE growth process.

The first important ingredient in the growth of GaAs is the incorporation of anions. As shown in Fig. 2, the dissociative chemisorption is a key process in growth and a model proposed from experimental studies can be represented by the following equations:

$$As_2 \rightleftarrows As_2^* \qquad 4(a)$$

$$As_2^* + 2V(As) \rightleftarrows 2\,As \qquad 4(b)$$

where As_2^* is a physisorbed state of As_2 above the growing structure and V(As) represents an As vacancy on the surface. The microscopic details of the dissociative process are not provided by Eqs. 4(a) and 4(b), but have emerged instead from the Monte Carlo simulations and physical arguments. Three reaction pathways for dissociative pathways are shown in

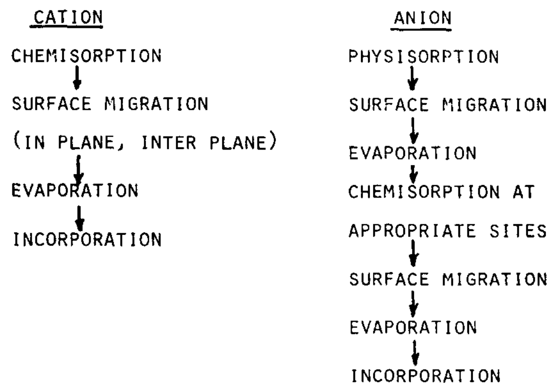

Fig. 2. Conceptual Picture of the MBE Growth Process.

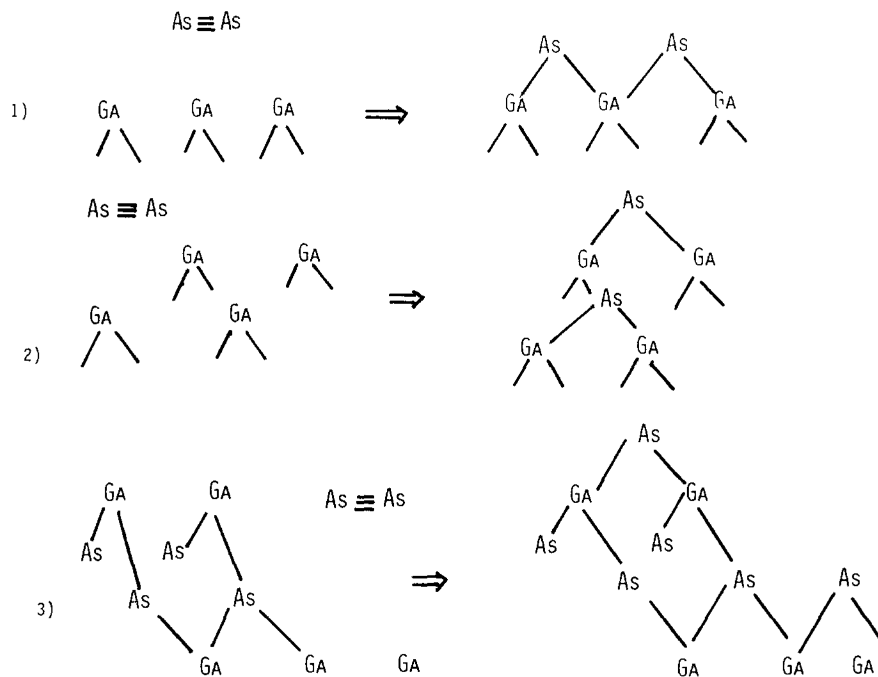

Fig. 3. Atomistic Model for the As_2 Incorporation During GaAs Growth.

Fig. 3 and are incorporated in the growth simulations. In addition to the pathways shown in Fig. 3, other pathways could be available, but are expected to have no significant contribution in the growth process.

The cation incorporation involves a simpler process as shown in Fig. 4. A cation is in atomic form when it impinges on the surface and attaches randomly to a surface site. It then undergoes several kinetically controlled steps before it is finally incorporated.

C. Kinetics of MBE Growth

Under typical MBE growth conditions used, the incorporation rate for cations (Ga, Al, etc.) is nearly 100%[15]. This means that the impingement flux for cations is much larger (usually ~ 10 times) than evaporation flux. Under these conditions, it has been shown that unless cation migration rate is high, growth will occur by a 3-dimensional island mode - a mode which is not capable of producing high quality interfaces[9,10].

In Fig. 4 we show the key steps involved in cation kinetics. The step shown in Fig. 4(a) is not expected to be controlled by any activation barrier since it involves formation of a single Ga-As bond. The intra- and inter-layer hops shown in Figs. 4(b) and 4(c) are controlled by activation barriers and have hopping rates given by the Arrhenious expressions as shown. Here R_o is a prefactor, E_{TOT} the cation site energy calculated according to Eq. (1) and E_{11} and E_{I} are activation barriers.

Under high temperature growth conditions, re-evaporation of cations can be significant and must be taken into account. The site evaporation is given by

$$R_e = R_{oe} \exp\left(-\frac{E_{TOT}}{kT_s}\right) \qquad (5)$$

It is clear from discussion in this section that a number of energetic and kinetic parameters are involved in crystal growth and these must be obtained carefully to fully utilize the outcome of the computer simulations, In the next section, we describe how these parameters are obtained.

3. Computer Simulation Model

The versatility and scope of any computer simulation is quite obvious. However, it is very important to realize the pitfalls of relying too heavily on the computer results and being carried away by the results of a fascinating but unphysical mathematical model. A great deal of care has been taken by us to evolve a computer simulation model that is quite realistic. One has to always compromise and make approximations in a simulation, since otherwise the time taken to run it would become

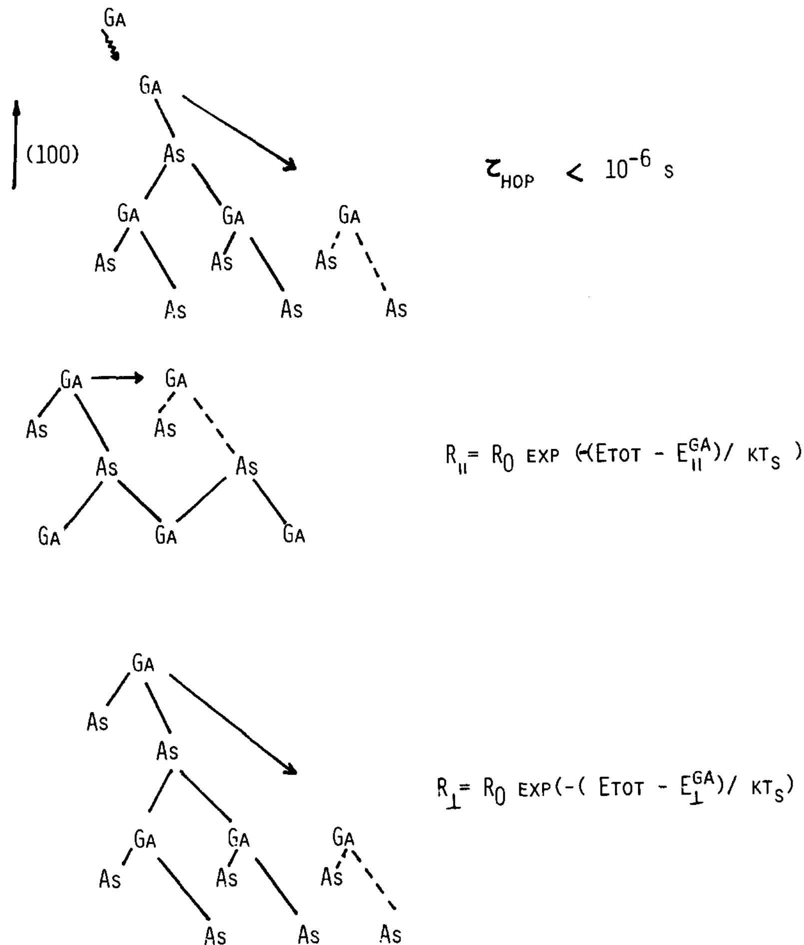

Fig. 4. Important Kinetic Steps in Ga Incorporation.

exorbitant. The following components have to be assembled to ensure a successful growth simulation:

a) An accurate concept of the physical processes involved.

b) An accurate set of parameters describing the physical processes.

c) An appropriate algorithm translating the physical ideas to the computer.

d) A proper determination of relevant physical properties which can then be compared with experiments.

We have already discussed component a) in Section 2. We must emphasize that the conceptual picture of MBE growth has emerged from GaAs studies. It is quite likely that a similar picture will emerge from experimental studies on other compound semiconductors and, therefore, we will use the same picture for the growth of other III-V and II-VI compounds.

Component b), i.e., adequate parameters to describe the crystal growth is perhaps the single most important ingredient of the simulations. As discussed in Section 2, the bond energies, activation barriers for hopping and prefactors for evaporation and hopping are the critical parameters for understanding growth. We have followed the following method to identify the parameters for GaAs and AlAs and propose to use a similar approach for other semiconductors. We examine the vapor pressure vs. 1/T data for Ga and As on GaAs. This gives the kink site bond energy (E_{KINK}) for the atoms when they are bonded on the GaAs surface. The kink site is defined as a site at a step edge where a Ga atom has two nearest and six second neighbor bonds satisfied. From Eq. (1) we then have for GaAs

$$E_{KINK} = 2V_{Ga-As} + 6V_{Ga-Ga} = E_{AC} \qquad (6)$$

where E_{AC} is the activation barrier for Ga obtained from vapor pressure vs. temperature data on GaAs. We have assumed that the nearest-neighbor bond is about five times as strong as the second-neighbor bond. Clearly this ratio is somewhat arbitrary, although on physical grounds one does expect it to be quite reasonable.

The determination of the kinetic parameters is much more difficult which, as will become clear in the next section, are of great significance in determining the growth quality. As noted earlier, the key parameters of significance are R_{od}, E_{11}, and E_1. These values have to be obtained from a careful comparison with available experimental results coupled with the physics and chemistry of surface bonds. We choose E_{11} and E_1 to be equal to $V_{A-C} + 2\,V_{A-A}$ corresponding to breaking half the surface bonds [on the (100) surface] during a hop.

This scheme which has only recently been developed by us, allows a simple scaling of activation barriers when we study growth of different semiconductors. The prefactor R_{od} is controlled by the phonon frequency and represents the attempt frequency for the hop. However, it is known that for adatoms the value of R_{od} can vary anywhere from $10^5\ s^{-1}$ to $10^{11}\ s^{-1}$. One therefore needs to have some information on hopping rates for the atoms on the growing surface before deciding on the value of R_{od}. There has been only one attempt to measure R_d for Ga atoms on GaAs using a masked growth technique[16]. This has suggested that Ga atoms can move ~ 2000 A in the absence of As overpressure at ~ 550°C. This has led us to suggest the following kinetic parameters for GaAs:

$$E_{11} = 1.14\ eV,\ E_1 = 1.14\ eV,\ \text{and}\ R_{od} = 3.76 \times 10^{10}\ s^{-1}$$

For other semiconductors we scale the activation barriers with the bond strengths in the manner described above.

The short discussion presented here highlights the need for more work to be done to obtain accurate values of the growth parameters. It is expected that the significance of these parameters in growth as brought out by our studies will encourage experimental efforts to measure them.

A software package has been put together to transmit the concepts discussed here into a computer system. The flowchart for this program is shown in Fig. 5. The program consists of simulating each of the steps described in Fig. 2 by a random process. The relative probabilities of the various kinetic events are used to decide which processes occur at any stage.

Component d) of the simulation effort is more important than one may initially think, since it involves a final comparison of the computer output with the measurements made in the laboratory. We have developed two different procedures to analyze the computer output which consists of the occupation numbers of the various sites on a zinc-blende lattice. In the first procedure, analysis is carried out during the crystal growth itself, while in the second, the analysis is done after the simulation run is finished.

The specific analysis done depends upon the structure being simulated, but in general, we calculate the following:

i) Partial layer coverages as a function of time.

ii) Surface strain energy during growth.

iii) Clustering effects in growth of alloys.

iv) Interface structure parameters when interfaces are formed between different semiconductors.

v) Surface hopping distances for growth under different conditions.

This information can then be compared with specific experiments, such as RHEED[17-19], photoluminescence in quantum wells[20,21], masked growth studies, etc., to authenticate the model.

We will now briefly discuss the computational requirements of carrying out the programs outlined here. We have found that a lattice size of 30 x 30 x 20 is required to obtain convergent results as far as global properties, such as differential layer coverages, are concerned. To obtain reliable information on microstructural details such as cluster sizes, etc., one needs to do several runs with different calls to the random numbers. The computer time requirement is the main problem associated with the runs. While for low temperature simulations only 15-20 minutes may be required (due to low migration rates), for simulation of high temperature growth (e.g., temperature where conventional MBE growth is done), several hours (2-3 hours) are required on the Cyber 205 System currently being used by us. Thus, it is our experience that to cover a reasonable growth-conditions parameter space, 15-20 hours of CPU time is required.

We must also emphasize that the results of the computer simulations can be fully appreciated if they are presented in a dynamic manner, e.g., as a movie. Much of the information is lost if the results are presented as graphs and figures. It is hoped that in the near future the current pace of developments in computer graphics will solve this important problem.

4. Results of Conventional MBE Growth

A considerable amount of theoretical work has been done to understand the growth of epitaxial crystals from the vapor phase. Some of the earliest results on growth modes were obtained by Burton, Cabrera, and Frank[22]. Later, several others used the Monte Carlo method to confirm these results and to identify new features in the growth process[5,6]. Much of

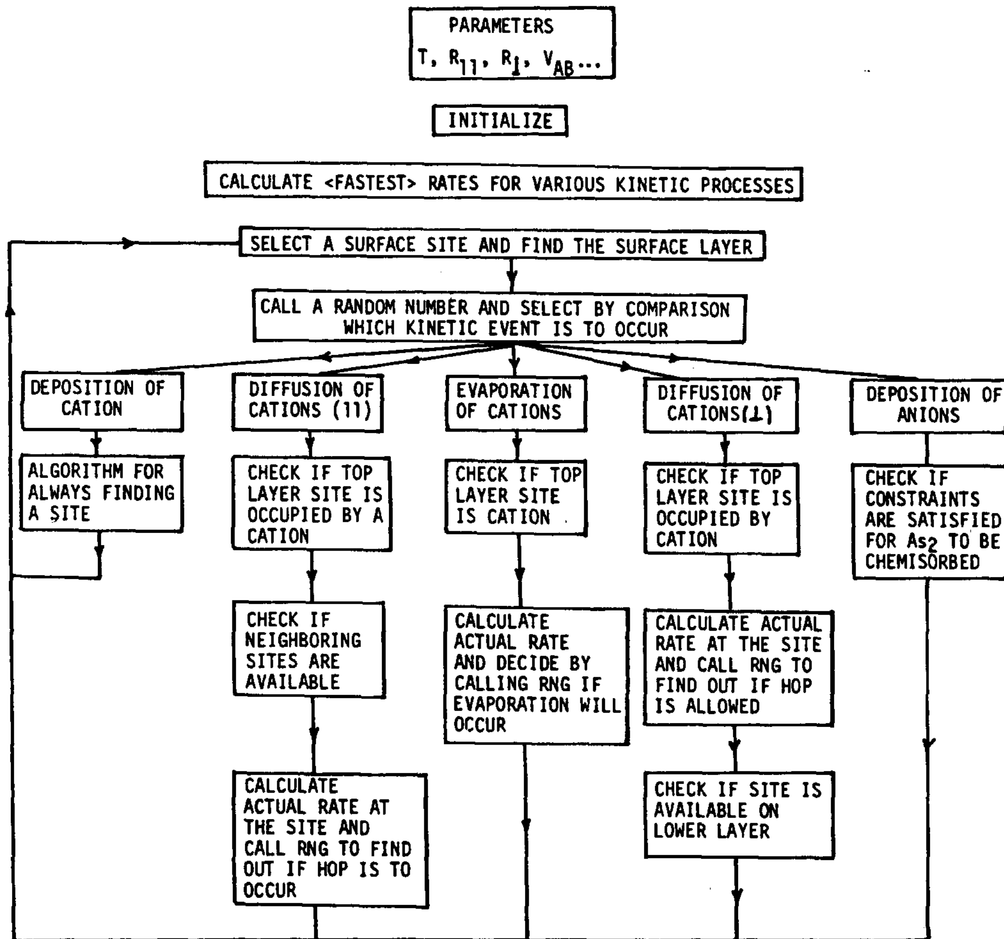

Fig. 5. A Flowchart for the Monte Carlo Process Used in the Simulations.

this work was done for the elemental cubic system (the so called "solid-on-solid" or SOS model), and the following important conclusions were obtained:

i) When the impinging flux from the vapor is close to the evaporating flux (small driving force case), growth occurs by the layer-by-layer mode. Two dimensional (2-D) nucleation is responsible for growth in this regime and the vapor-solid interface is atomically flat.

ii) In the case of larger driving force (impingement flux >> evaporation flux), the growth proceeds by the three dimensional (3-D) island growth mechanism in which the growth is continuous and the growth front extends over several monolayers.

iii) There exists a temperature, T_R, above which the growth front is always rough. This temperature, called the roughening transition temperature, represents a phase transition in the system.

Although these studies were pioneering in the area of crystal growth, it has become clear that the regimes and possibilities they cover do not include MBE growth of compound semiconductors. The principle differences between these studies and those by Singh and Madhukar[8], and Singh and Bajaj[9-11] lie in that the latter work recognizes the important role of cation surface migration and the presence of molecular anions.

Recently, Singh and Bajaj[9-11] have carried out a number of growth simulations for GaAs, AlGaAs as well as their interfaces. In order to understand the significance of these results it is important to realize that MBE growth takes place under an anion overpressure and the cation impingement flux is typically an order of magnitude higher than cation evaporation

rate (this may not be true for growth of semiconductors with very different bond-strengths). This puts the entire burden of kinetics on the cation and anion surface migration rates. Since the anions are initially in a highly mobile physisorbed state, the limiting kinetics controlling the growth is the cation surface kinetics.

Figure 6 shows the time evolution of the coverage of different monolayers as the growth is proceeding. Shown are plots of $d\theta_n/d\theta_{Tot}$ as a function of growth time when the cation hopping rate is a) 1.0 hop/sec and b) 3000 hops/sec. These correspond to GaAs growth at substrate temperature of 650 K and 810 K respectively when the growth rate is one monolayer per second. θ_n is the coverage of the nth layer and θ_{Tot} is the total coverage on the crystal so that $d\theta_n/d\theta_{Tot}$ determines what fraction of the impinging atoms are incorporated in the nth layer. It can be seen that for the low migration rate, several monolayers start growing simultaneously, while for the high migration rate, growth starts on a given monolayer only after the lower layer is almost filled. It is important to point out that the Monte Carlo method cannot be used to study growth of more than ~20 monolayers due to large computer expense involved. We have developed a scheme by which information from the Monte Carlo studies is utilized in a statistical model to predict surface quality for thicker films.[11] In Fig. 7 we show the growth-front roughness as a function of the thickness of the film grown for three different growth conditions identified by the value of n_c. Here n_c is the average number of sites visited by a cation between the time it impinges from the vapor and to the time it gets incorporated in the crystal. The values of n_c used are a) 25, b) 4, and c) 2. We note that for (100) growth where exchange reactions are not expected, the surface front is frozen in when an interface is formed. We finally summarize our results below:

i) Cation surface kinetics are critical in controlling the growth-front (and interface) quality.

ii) For a fixed growth rate, a given semiconductor can grow by the layer-by-layer mode above a certain temperature where the cation hopping rate is $\sim 10^4$ hops/time for monolayer growth.

iii) Since different semiconductors have different bond strengths and, hence, different activation barriers for cation hopping, the ideal temperatures for growth are different.

iv) Due to the observation noted in (iii), the quality of the normal and inverted interfaces can be quite different. The term "normal" interface is used for the interface produced by the sequence in which the lower melting temperature component is grown first. The term "inverted" interface is used for the reverse sequence.

v) The surface roughness of a growing structure gets worse as the film thickness increases due to statistical fluctuations of the impinging cation flux.

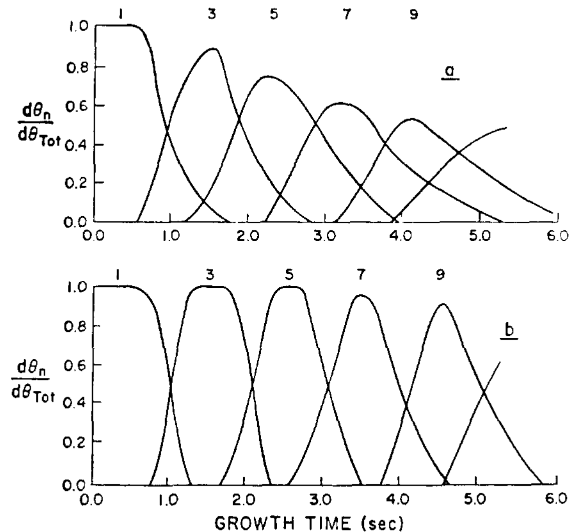

Fig. 6. Time Evolution of the Coverages on Different Monolayers During Growth (see Ref. 10).

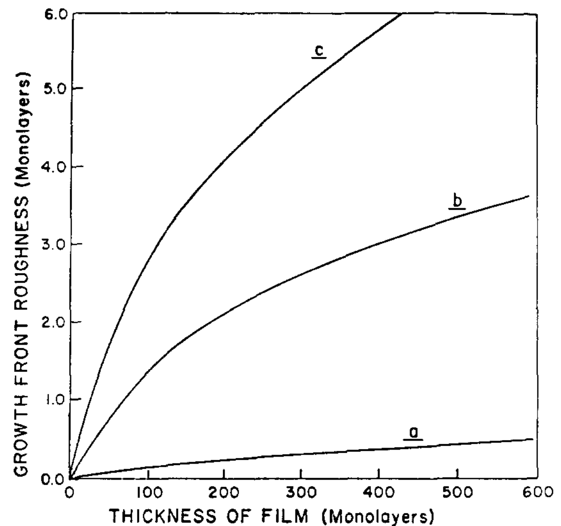

Fig. 7. Surface Roughness as a Function of Film Thickness for Different Cation Kinetics (see Ref. 11).

The results listed above have been very important in understanding a variety of observations in the MBE grown heterostructures. The continuing computer simulation work has now acquired some predictive power which is expected to be important in heterostructure technology.

FOR A FIXED GROWTH RATE :

AMORPHOUS HIGH INTRINSIC DEFECTS	FILM WITH ROUGH SURFACE/ INTERFACE	FILM WITH SHARP INTERFACE	BULK DIFFUSION HIGH DEFECT DENSITY

T_{c1} T_{c2} T_{c3}

FREE ENERGY = U- T.S

KINETICS ⟶ APPROACH TO FREE ENERGY MINIMUM

EXTREMELY KINETICS LIMITED	PARTIALLY KINETICS LIMITED	SUFFICIENT SURFACE KINETICS	ENTROPY CONTROLLED GROWTH

T_{c1} T_{c2} T_{c3}

Fig. 8. Role of Kinetics and Thermodynamics on the Surface/Interface Quality of MBE Structures Grown at Different Temperatures.

5. Novel Growth Approaches in MBE

The focus of the crystal growth simulations has been on lattice-matched systems, such as the GaAs/AlGaAs. A number of important results were obtained from these studies which have been discussed in Section 4. According to these studies certain generic conclusions can be drawn regarding heterostructures grown by the conventional MBE technique. In general, if one is interested in heterostructures grown from AC - BC (A, B cations; C anions), one finds that a) the ideal growth conditions (for layer-by-layer growth) are different for material AC than for material BC, and b) the ideal growth conditions for the normal and inverted structures may be quite different. The differences will increase as the energy difference between A-C and B-C bonds increases.

These ideas, coupled with experimental information, are illustrated in Fig. 8. Here we describe the structural properties of a film grown at different temperatures. Below a certain critical temperature Tc_1, the film is amorphous due to a complete lack of migration of the impinging atoms. Between temperatures Tc_1 and Tc_2 the film grows epitaxially, but with a poor surface due to lack of sufficient kinetics for the impinging atoms. Between Tc_2 and Tc_3, there exists a window of temperature over which the surface and, consequently, the corresponding interface is of high quality. At higher temperatures, the entropy controlled effects cause a poor quality of film due to high defect densities. The temperatures Tc_1, Tc_2, and Tc_3 are controlled by the bond strengths of the growing system. Thus, if the bond strengths of two materials AC and BC are quite different, there may be no temperature regime where the two materials grow with very smooth surfaces. However, as can be inferred from Fig. 8, modifications in the growth rate may allow an overlap of temperature regimes where both materials may grow well.

These problems are expected to be critical for semiconductor combinations such as HgTe-CdTe, InAlAs-InP, etc. Computer simulations are now being carried out to study both conventional and novel ways to improve interface quality in these and other systems. The results discussed in Section 4 apply to the conventional MBE growth approach where the growth rates and substrate temperatures are kept constant. However, from the above discussion it is clear that in the growth of certain heterostructures, conventional growth approaches may not be suited for high quality structures. Computer simulations are ideally suited for studying the effects of novel growth approaches. We will briefly describe four such approaches that are being explored by using computer simulations.

i) Use of resonant laser enhanced

cation surface kinetics for low temperature growth of heterostructures.

ii) Use of thin layers of a low melting temperature semiconductor for smoothening rough surfaces during MBE growth.

iii) Use of temperature pulses during growth for obtaining sharp interfaces at low average growth temperatures.

iv) Use of growth interruption before interface formation to improve interface quality.

All of these approaches are motivated by the realization that in any heterostructure, one of the components will almost always grow with high quality at a lower temperature than the other component. To alleviate this problem the approaches outlined above aim to grow heterostructures at a lower average temperature. We will briefly describe these approaches below.

i) Use of resonant laser enhanced cation kinetics: In conventional MBE growth, the cation kinetics is controlled primarily by the substrate temperature as shown in Fig. 4. If the energy required to move a surface atom from a given site to the adjacent surface site can be provided by photons from a laser source instead of the phonons, then semiconductors can be grown at lower substrate temperatures as long as the photon flux is present. This would allow the possibility of growing heterostructures at lower substrate temperatures as well as allow growth of high quality heterostructures from components that cannot be grown well at the same substrate temperature. Computer simulations carried out to establish the feasibility of this approach have been done assuming that the probability that a photon with energy $h\nu$ moves a surface atom from site i to the neighboring empty site i' is given as[23]

$$P(h\nu, C_i^{i'}) = P_m \exp\left[-\frac{1}{2}\left(\frac{E_i - E_{res}}{\sigma}\right)^2\right] + p^{NR} \quad (7)$$

where P_m is a coupling constant, E_{res} is the resonant energy, E_i is the energy of the surface atom at site i, σ is the width of the resonance and p^{NR} is the hopping probability due to non-resonant heating effects. Computer simulations show that the following conditions must be satisfied if this approach is to be feasible:

a) E_{res} must be equal to the non-kink site energy.

b) σ must be less than the second neighbor cation-cation bond energy. This energy is typically ~0.2 eV.

c) P_m must be larger than 10^{-4} per photon.

If the conditions mentioned above can be satisfied, high quality MBE growth would be feasible at temperatures lower than those employed in the conventional growth.

ii) Use of thin layers of a low melting temperature semiconductor for surface smoothening: This approach can be best understood by considering the AlAs system which grows with high quality surfaces at ~1000 K. If AlAs were to be grown at say 700 K, its surface quality would steadily worsen as growth continues making it useless for device applications. On the other hand InAs grows well at 700 K due to the much weaker In-As bond. Thus if a few monolayers ($\lesssim 5$) are periodically deposited during AlAs growth with a period of ~100 monolayers, AlAs with high quality surface can be grown at low temperatures[24]. Recently it has been shown that this approach is indeed feasible for growing high quality interfaces[25].

iii) Use of temperature pulses during MBE growth: This approach is again motivated by the need to grow heterostructures at lower average temperature. The concept is based on the recognition that in low temperature growth, the surface roughness increases with growth time, so that periodic or high temperature pulses could smoothen the growth front and thus allow growth of structures at lower temperatures. Our simulations have shown that high quality AlAs can be grown at 800 K if periodic pulses which raise the substrate temperature by ~200 K for 1-5 seconds are applied every minute or so[26].

iv) Use of growth interruption to grow high quality interfaces: we finally discuss this important technique for fabricating high quality interfaces at lower temperatures. In this approach the crystal growth is interrupted before the formation of the interface. During the interruption process the following kinetic events continue which are responsible for the smoothening of the surface and consequently are responsible for higher quality interfaces[27]:

a) Evaporation of anions as molecules from the surface,

b) Evaporation of cations from the surface,

c) Migration of cations on the surface.

Our computer simulations show that for low temperature growth the step a) is the rate limiting step to the smoothening process. During the interruption process background impurities can accumulate on the surface producing degradation of the film quality. The interruption times for producing smooth surfaces for GaAs being grown at 700 K are estimated to be as large as 10 minutes. Photoluminescence studies in InAlAs/InGaAs quantum well structures show that the interface quality steadily improves as the interruption time increases as inferred from the narrowing of the excitonic transition[28]. However, the emission intensity decreases dramatically suggesting impurity accumulation during interruption time. Our simulations suggest that the following additional steps must be taken to make this growth approach more beneficial:

a) Reduction of anion overpressure to allow the cation migration to occur with greater ease.

b) An increase in substrate temperature to shorten the interruption time to avoid impurity accumulation at the interface.

c) Since the interruption process is more useful for the inverted interface, it may be better to interrupt a few monolayers before the formation of the interface. This will keep the impurities accumulated from the background away from the interface and the active region of the device.

The brief discussion above on different unconventional approaches to MBE growth shows the potential of computer simulations in providing useful information on how high quality "difficult" heterostructures could be grown. As discussed earlier these approaches and modifications thereof are critical for the growth of heterostructures whose components have very different bondstrengths.

6. Conclusions

In this paper we have provided a motivation for using computer simulations to understand the growth of heterostructures by MBE. An atomistic model of MBE growth has been developed on the basis of experimental and theoretical work. The ingredients of a Monte Carlo simulation method have been described and results have been presented for the conventional MBE growth approach. The main conclusion of these studies is that it is difficult to grow high quality heterostructures from semiconductor components which have very different bondstrengths or melting temperatures. However, this difficulty can be overcome if unconventional growth approaches are used. The approaches may involve changing the growth rate, growth temperature as well as controlling surface kinetics of atoms through resonant laser coupling. Computer simulations provide an excellent mechanism to test the feasibility of these approaches. Such efforts are already paying rich dividends in terms of developing a better understanding of the growth process and, consequently, its control. The modeling effort is also acquiring a predictive power which when coupled with carefully controlled experiments may lead to the fabrication of high quality metastable structures.

References

1) A. Y. Cho and J. R. Arthur, in Progress in Solid State Chemistry, Eds. G. Somarjai and J. McCaldin (Pergamon, NY, 1975), Vol. 10, p. 175.
2) C. E. C. Wood, in Physics of Thin Films, Eds. G. Haas and M. H. Francombe (Academic, NY 1980) Vol. II, p. 35.
3) A. Y. Cho, Thin Solid Films, 100, 291 (1983).
4) L. Esaki and R. Tsu, IBM J. Res. Dev. 14, 61 (1970).
5) J. D. Weeks, G. H. Gilmer, and K. A. Jackson, J. Chem. Phys. 65, 712 (1976).
6) G. H. Gilmer and K. A. Jackson, in Crystal Growth and Materials, Eds. E. Kaldis and H. J. Scheel (North-Holland, Amsterdam, 1977).
7) W. J. P. van Enckewort and J. P. van der Eerden, J. of Cryst, Growth, 47, 501 (1979).
8) J. Singh and A. Madhukar, Phys. Rev. Lett. 51, 794 (1983).
9) J. Singh and K. K. Bajaj, J. of Vac. Sci. Technol. B2(2), 276 (1984).
10) J. Singh and K. K. Bajaj, J. Vac. Sci. Technol. B2(3), 576 (1984).
11) J. Singh and K. K. Bajaj, J. Vac. Sci. Technol. B3(2), 520 (1985).
12) J. R. Arthur, Surf. Sci. 43, 1999 (1974).
13) C. T. Foxon and B. A. Joyce, Surf. Sci. 64, 293 (1977).
14) B. A. Joyce and C. T. Foxon, J. Cryst. Growth, 31, 122 (1975).
15) J. R. Arthur, J. Phys. Chem. Solids, 28, 2257 (1967).
16) S. Nagata and T. Taneka, J. Appl. Phys. 48, 950 (1977).
17) J. M. van Hove, C. S. Lent, P. R. Pukite, and P. I. Cohen, J. Vac. Sci. Technol. B1(3), 741 (1983).
18) J. H. Neave and B. A. Joyce, Appl. Phys. A 34, 179 (1983).
19) A. Madhukar, T. C. Lee, M. Y. Yen, P. Chen, J. Y. Kim, S. V. Chaisas, and P. G. Newman, Appl. Phys. Lett. 46, 1148 (1985).
20) J. Singh, K. K. Bajaj, and C. Chaudhari, Appl. Phys. Lett. 44, 805 (1984).
21) J. Singh, K. K. Bajaj, D. C. Reynolds, C. W. Litton, P. W. Yu, W. T. Masselink, R. Fischer, and H. Morkoc, J. Vac. Sci. Technol. B3(4), 1061 (1985).
22) W. K. Burton, N. Cabrera, and F. C. Frank, Philos. Trans. Roy. Soc. London, Ser A 243. 299 (1951).
23) J. Singh and K. K. Bajaj, Appl. Phys. Lett, 46, 577 (1985).
24) J. Singh and K. K. Bajaj, Appl. Phys. Lett. 47, 594 (1985).
25) L. Goldstein, M. N. Charasse, A. M. Jean-Louis, G. LeRoux, M. Allovon and J. Y. Marzin, J. Vac. Sci. Technol. B 3, 947 (1985).
26) S. Dudley, K. K. Bajaj, and J. Singh, unpublished.
27) J. Singh, S. Dudley, and K. K. Bajaj, unpublished.
28) F. Y. Juang, P. K. Bhattacharya, and J. Singh, Appl. Phys Lett. (in press).