ERRATA

Page	Line	Reads	Should Read
(Throughout the report)		formulism	formalism
12	Fig. 2	$\tau_c \ll \tau_N$	$\tau_c \gg \tau_N$
12	12	resolent time	resolving time
13	5	resolvent time	resolving time
13	12	$\dot{N}_1(k_1)$ becomes $\dot{N}_1(\underline{k}_1)$	$\dot{N}_1(\underline{k}_1)$ becomes $\dot{N}_1(k_1)$
13	17	$\dot{N}_c(\underline{k}_2\|k_1)$	$\dot{N}_c(\underline{k}_2\|\underline{k}_1)$
14	11	evalution	evolution
15	10	$\{\|\beta_i>\}$	$\{\|\beta>\}$
15	Eq. (2.10)	$\|\beta_i'>\underline{P}(\beta_i')<\beta_i'\|$	$\|\beta'>\underline{P}(\beta')<\beta'\|$
15	11	$\underline{P}(\beta_i')$	$\underline{P}(\beta')$
15	12	$\|\beta_i'>$	$\|\beta'>$
16	2	evalution	evolution
29	10	multiple	multipole
30	Eq. (3.31)	$I_{\nu_1}(j_i j j)II_{\nu_2}(j_f j j)$	$I_{\nu}(j_i j j)II_{\nu}(j_f j j)$
32	9	V_c^{NC}	V_τ^{NC}
35	8	$\underline{u}_{oci}$	$\underline{a}_{oci}$
39	2	$\hbar\omega_{KK}$	$\hbar\omega_{KK'}$
53	3	(12)	(11)
60	Eq. (5.10)	(right-hand side of Eq. (5.10) should be multiplied by)	$\alpha\omega'$
64	Fig. 7	D_γ	D_2
64	Fig. 7	γ_1 (along z-axis)	γ_2
65	Eq. (6.6)	$I_{\nu_2'}(j_i j j)$	$I_{\nu_1'}(j_i j j)$

ERRATA (Concluded)

Page	Line	Reads	Should Read
65	Eq. (6.7)	$II_{\nu_2}(j_f jj)$	$II_{\nu_2'}(j_f jj)$
84	13	the probability to choose the sources for experiments	the interpretability of experiments
71	7	$d_{\epsilon i} = 0$	$d_{ci} = 0$
75	Eq. (7.9)	$\sum_{KK'} G'_{KK'} T$	$\sum_{KK'} G'_{KK'}(0)T^2,$
76	Eq. (7.11)	$\sum_{K''' \neq K}$	$\sum_{K''' \neq K'}$
89	Eq. (I.15)	$V_{\beta_i \beta_f}$	$V_{\beta_i \beta_i}$
89	Eq. (I.16)	denominator	$i(E_{\beta_f} - E_{\beta_i})$
99	Eq. (IV.3)	$II_{\nu_2'}(j_f jj)$	$II_{\nu_2'}(j_f jj)$
103	5	$D^{(2)}_{\rho\text{-}\beta}(\Phi, \textcircled{H}, 0)$	$D^{(2)}_{\rho\text{-}\zeta}(\Phi, \textcircled{H}, 0).$
124	12	The University of Michigan, 1963,	1963, (To be published as an ANS Monograph)
124	15	Technical Report 04836-1-T, 1963.	University of Michigan Technical Report 04836-1-T 1963.

T H E U N I V E R S I T Y O F M I C H I G A N

COLLEGE OF ENGINEERING
Department of Nuclear Engineering

Technical Report

THEORY OF THE INFLUENCE OF THE ENVIRONMENT ON γ-γ ANGULAR CORRELATION AND ITS ROTATIONAL DEPENDENCE

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ABSTRACT

A theory of the influence of the environment on γ-γ angular correlation is presented. The theory is formulated by using the resolvent method, and the attenuation coefficient of the perturbed angular correlation function, which contains the effects of the changes of the states of the environment on the angular correlation, is obtained. In the solid environment, the effects of the lattice vibrations on the magnetic dipole-dipole coupling and the electric quadrupole coupling have been analyzed by using the normal mode expansion of the lattice displacements; and the perturbed angular correlation function is shown to be a function q the crystal temperature as experimentally observed.[9]

In order to determine the nuclear electric quadrupole moments in an excited state, which cannot be done by the usual microwave methods, a rotational technique is suggested and the theory has been developed for the case of asymmetric crystalline fields. For the special case of axially symmetric crystalline field, the present theory predicts the phase shifts in the rotational pattern of anisotropy, which agrees with the observations by the Zürich group.[8],[19],[20]

I. INTRODUCTION

It was first pointed out in 1940 by Dunworth[1] that the coincidence measurement of two radiations emitted by a radioactive nucleus shows a correlation in their relative propagation directions. In the same year the pioneer theoretical study of this angular correlation phenomena was made by Hamilton.[2] In this first theoretical paper Hamilton used the time-dependent perturbation theory and the result was restricted to the case when the intermediate state of the nucleus in the cascade is completely unperturbed. However, the experimentally observed angular correlations do not agree well with the theoretical predictions based on that simplifying assumption. Many observations are found to depend on the physical and chemical nature of the sources. Therefore one important aspect, to which attention was drawn, is the possible influence of extranuclear environments on the angular correlations.

In 1946 Goertzel[3] first made the theoretical investigations of the perturbations of the angular correlations by the hyperfine-structure interactions and externally applied magnetic fields. In his paper Goertzel followed closely Hamilton's approach and showed that only for an extremely short nuclear lifetime in the intermediate state would the correlation be unperturbed. The first efforts to look at perturbation were made in 1951 when Frauenfelder[4] suggested that the after-effects

of β-decay or K-capture could perturbe the angular correlation. The experiments showed indeed that there are perturbations, although the experiments were not conclusive whether all these perturbation effects were due to after-effects or due to static quadrupole interactions. K. Alder[5],[6] in his two papers reformulated Goertzel's results and showed that the effects of the extranuclear perturbation can be factored out as the so-called attenuation factor.

Abragam and Pound,[7] in 1953, reformulated the theory so that it can be used to describe the effect of the electric quadrupole interactions in liquids. In the same year Alber-Schönberg et al.[8] in Zürich performed the most conclusive experiments on the effect of the quadrupole interaction in a single crystal of indium. In their experiments a metallic indium single crystal of axial symmetry was used and the rotational patterns of the anisotropy with respects to the rotations of the crystal axis were observed. According to the theory one always predicts the maximum anisotropy at the zero degree of rotation. However, Alber-Schöberg's observation shows a phase shift in the rotational patterns.

Recently Ouseph and Canavan[9] have observed the temperature dependence of the anisotropy by using a hafnium single crystal. Saloman et al.[4] (1963) have also observed the temperature dependence of the anisotropy in case of Ta^{181} in a pure Hf-metal. They also found the same deviation from the Abragam and Pound's theory as found by Sommer-

feld and Schecter.[10]

According to Goertzel and Abragam's theories the explanation of the observations by the Zürich group and Ouseph et al. remains unclear. The after-effects of β-decay or K-capture will disturb the electronic shell and produce a magnetic shell which can cause a strong perturbation of the angular correlation. However, for long-lived isomer such as 48 min Cd^{111} the perturbations induced by the after-effects are unlikely. On the other hand a purely static electric interaction in the intermediate state is also unlikely. The thermal vibrations of the lattice points will change the electric field gradient at the position of the decaying nuclei and also produce the spin relaxation effect due to the interaction between the nuclear spin and the vibrating lattice system. That is, the decaying nuclei are influenced by different field gradients rather than a unique static field. Thus it would appear that the thermal vibration of the lattice is mainly the mechanism which causes the deviations from the theory. Therefore it is desirable to have a perturbed correlation function in which the effect of the lattice vibrations has been explicitely built in.

In this thesis the theory of the perturbed angular correlation is formulated by using the resolvent method. In this formulism the energy level displacements and the spin-lattice relaxations due to the thermal vibrations of the lattice system have been explicitely built

in the perturbed correlation function. As shown in the Section VIII, the difference between the present theory and the Abragam's formulism appears only in the last factors of the attenuation coefficients. In Abragam's formula the attenuation coefficient contains a factor

$$\langle jm|b\rangle\langle b|jm''\rangle\langle jm''|b'\rangle\langle b'|jm'\rangle\left[1-\left(\frac{i\tau_N}{\hbar}\right)(E_b-E_{b'})\right]^{-1}, \tag{1.1}$$

while in the present theory, instead of the expression (1.1), we have

$$\langle jm|K\rangle\langle K|jm'\rangle\langle jm''|K'\rangle\langle K'|jm''\rangle \times$$

$$\times\left\{\frac{\hbar}{2}\left[\langle\Gamma_K(V_\tau^{NC})\rangle_T+\langle\Gamma_{K'}(V_\tau^{NC})\rangle_T+2\tau_N^{-1}\right]-\right.$$

$$\left.-i\left[E_K-E_{K'}+\langle S_K(V_\tau^{NC})\rangle_T-\langle S_{K'}(V_\tau^{NC})\rangle_T\right]\right\}^{-1}, \tag{1.2}$$

In the expression (1.2), the effects of the dynamic part of the perturbing interaction on the angular correlation are described by the thermally averaged energy level displacement function $\langle S_K(V_\tau^{NC})\rangle_T$ and the relaxation function $\langle \Gamma_K(V_\tau^{NC})\rangle_T$.

In Abragam's formulism the coupled state is avoided and the effects of the changes of the environments on the angular correlation are ignored. Therefore, e.g. for the solid state environments, the expression (1.1) cannot describe the effects of the thermal vibrations of the lattice system on the correlation function and cannot predict

the temperature dependence of the anisotropy as experimentally observed. However, in the present theory these effects are systematically built in the attenuation coefficient. One also sees clearly that the expression (1.2) predicts the temperature dependence of the correlation function.

In fact the present theory is specially suitable for the solid state environments in which the perturbing interactions can be naturally decomposed into two parts. One part corresponds to the coupling Hamiltonian V_o^{NC} when the crystal is considered as a rigid lattice system and the other part represents the dynamic interactions V_τ^{NC} induced by the vibrations of the lattice points about their equilibrium positions. The effects of the lattice vibrations on the magnetic dipole-dipole coupling and the electric quadrupole coupling have been analyzed by using the normal mode expansion of the lattice displacements as shown in the Section IV.

It is a well-known fact that the mechanism of nuclear quadrupole coupling in angular correlation is specially suitable for the investigation of the electric moments of a nucleus in a short-lived excited states. This cannot be done by the usual microwave techniques. So a particular emphasis is given to the effects of the nuclear electric quadrupole couplings and the functions $\langle S_K(V_\tau^{NC})\rangle_T$ and $\langle \Gamma_K(V_\tau^{NC})\rangle_T$ have been calculated in detail for the solid state environments as shown in the Section IV. In order to determine the nuclear quadrupole moments

in an excited state a rotational technique is suggested and the theory has been developed for the case of asymmetric crystalline fields. As shown in the Section V, by introducing three sets of coordinate systems, the rotational dependence of the angular correlation has been investigated for the solid state environments with asymmetric crystalline fields and the functions $\langle S_K(V_\tau^{NC})\rangle_T$ and $\langle \Gamma_K(V_\tau^{NC})\rangle_T$ have been calculated as a function of the Euler angles between the three coordinate systems. This aspect of angular correlation studies has not been previously considered.

II. THE THEORETICAL INTERPRETATION OF THE ANGULAR CORRELATION MEASUREMENTS

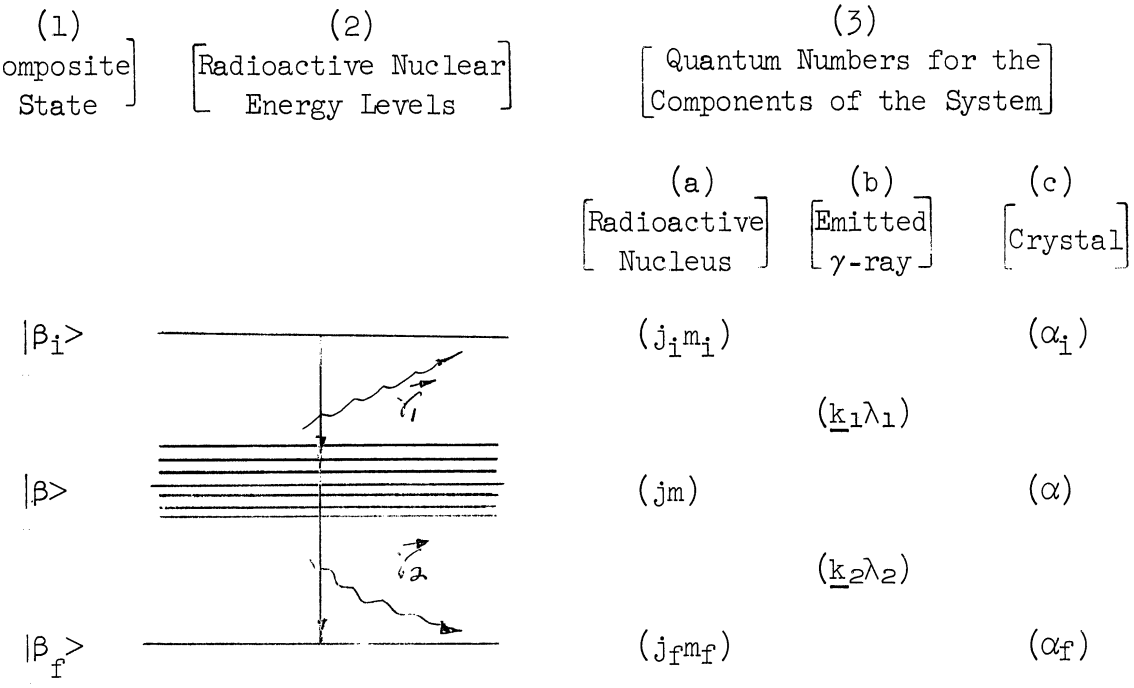
A. THE STATES OF THE PHYSICAL SYSTEM

The physical system which we are interested in here can be decomposed into two components. One is a radioactive nucleus and the other component is its environment which can be the electronic shells of an atom or ion, a molecule, a liquid or a lattice system of a solid. Now let us consider a radioactive nucleus, which is sitting at a crystal lattice site, undergoing a cascade γ-ray decay

$$ {}_{Z}X^{A**} \longrightarrow \vec{\gamma}_1 + {}_{Z}X^{A*} \longrightarrow \vec{\gamma}_1 + \vec{\gamma}_2 + {}_{Z}X^{A}. $$

We assume that the life time of ${}_{z}X^{A*}$ is long enough so that the interactions between the nucleus ${}_{z}X^{A*}$ and the crystalline fields measureably perturb the angular correlations of the cascade γ's.

In order to describe the events of the whole physical system, the quantum states of the system are designated according to the energy levels of the radioactive nucleus in the cascade decay.



The whole system is visualized as existing in any one of these three composite states $|\beta_i\rangle$, $|\beta\rangle$ and $|\beta_f\rangle$ corresponding to the intial state, the intermediate state and the final state of the radioactive nucleus in the cascade. The eigenkets $|\beta_i\rangle$, $|\beta\rangle$ and $|\beta_f\rangle$ belong to an orthonormal complete set of eigenkets $\{|\beta\rangle\}$ which satisfy the eigenvalue equation;

$$H_o|\beta\rangle = E_\beta|\beta\rangle .$$

The Hamiltonian H_o is a part of the total Hamiltonian $\mathcal{H}$ of the whole physical system, i.e.,

$$\mathcal{H} = H_o + V,$$

and

$$H_o = H^N + H^e + H^\gamma + V_o^{NC}$$
$$V = V^\gamma + V_e^{NC},$$

where H^N is the Hamiltonian of the radioactive nucleus and is taken to be a function of the internal degrees of freedom of the radioactive nucleus, H^c is the Hamiltonian of the lattice system and is taken to be a function of the external degrees of freedom of each lattice point including the radioactive nucleus, H^γ is the energy of the emitted photons, and V^γ is the radiation interaction Hamiltonian of the radioactive nucleus. The quantities V_o^{NC} and V_T^{NC} are respectively the static part and the dynamic part of the interaction Hamiltonian between the radioactive nucleus and the extranuclear fields.

Furthermore we define the following orthonormal complete sets of eigenkets $\{|\zeta_{jm}\rangle\}$, $\{|\alpha\rangle\}$, $\{|\eta_{\underline{k}_1\lambda_1}, \eta_{\underline{k}_2\lambda_2}\rangle\}$ and $\{|\zeta_{j\tau}\rangle\}$ such that

$$H^N|\zeta_{jm}\rangle = E^N_{\zeta j}|\zeta_{jm}\rangle,$$

$$H^c|\alpha\rangle = E_\alpha|\alpha\rangle,$$

$$H^\gamma|\eta_{\underline{k}_1\lambda_1}, \eta_{\underline{k}_2\lambda_2}\rangle = \left(\sum_{\underline{k}_1\lambda_1} \eta_{\underline{k}_1\lambda_1} \hbar c k_1 + \sum_{\underline{k}_2\lambda_2} \eta_{\underline{k}_2\lambda_2} \hbar c k_2\right) \times$$

$$\times|\eta_{\underline{k}_1\lambda_1}, \eta_{\underline{k}_2\lambda_2}\rangle \equiv$$

$$\equiv \left(E^\gamma_{\eta_{\underline{k}_1\lambda_1}} + E^\gamma_{\eta_{\underline{k}_2\lambda_2}}\right)|\eta_{\underline{k}_1\lambda_1}, \eta_{\underline{k}_2\lambda_2}\rangle,$$

and

$$(H^N + V_o^{NC})|\zeta_{j\tau}\rangle = E_{\zeta j\tau}|\zeta_{j\tau}\rangle,$$

can be solved. Then the eigenket $|\beta\rangle$ can be expressed as the product of eigenkets appropriate to each component of the physical system, i.e.,

$$|\beta\rangle = |\zeta j\tau\rangle |\alpha\rangle |\eta_{\underline{k}_1\lambda_1}, \eta_{\underline{k}_2\lambda_2}\rangle$$
$$\equiv |K\rangle |\alpha\rangle |\eta_{\underline{k}_1\lambda_1}, \eta_{\underline{k}_2\lambda_2}\rangle ,$$

where K stands for $(\zeta j\tau)$, then

$$H_0|\beta\rangle = (E_K + E_\alpha + E^{\gamma}_{\eta_{\underline{k}_1\lambda_1}} + E^{\gamma}_{\eta_{\underline{k}_2\lambda_2}})|\beta\rangle .$$

The α's are a set of quantum numbers which define a quantum state of the lattice system. The quantities j and m are respectively the angular momentum of the radioactive nucleus and its component on a quantization axis. The eigenket $|\eta_{\underline{k}_1\lambda_1}, \eta_{\underline{k}_2\lambda_2}\rangle$ defines the eigenstate of the cascade photons and $\eta_{\underline{k}\lambda}$ is the occupation number of photons of wave vector $\underline{k}$ and polarization λ.

In this representation the eigenstates corresponding to the initial state, the intermediate state and the final state of the radioactive nucleus can be expressed as;

$$|\beta_i\rangle = |K_i\rangle |\alpha_i\rangle |0_{\underline{k}_1\lambda_1}, 0_{\underline{k}_2\lambda_2}\rangle ,$$
$$|\beta\rangle = |K\rangle |\alpha\rangle |1_{\underline{k}_1\lambda_1}, 0_{\underline{k}_2\lambda_2}\rangle ,$$
$$|\beta_f\rangle = |K_f\rangle |\alpha_f\rangle |1_{\underline{k}_1\lambda_1}, 1_{\underline{k}_2\lambda_2}\rangle .$$

B. THE ANGULAR CORRELATION FUNCTION

An experimental arrangement for the measurements of the direction-direction angular correlation can be sketched as in Fig. 1.

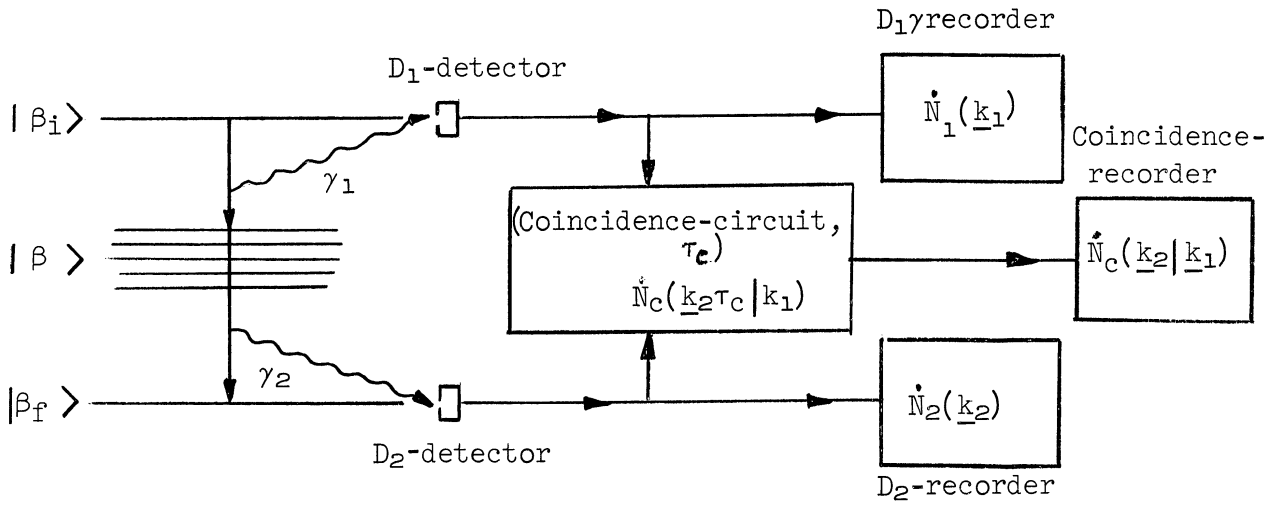


Fig. 1. Schematic representations of the measurements of γ-γ direction-direction correlation.

As shown in Fig. 1, the quantity $\dot{N}_1(\underline{k}_1)$ indicates the counting rate of the first emitted photon of wave vector $\underline{k}_1$ and $\dot{N}_c(\underline{k}_2|\underline{k}_1)$ indicates the coincidence counting rate of the cascade decay of $\gamma_1(\underline{k}_1)$ and $\gamma_2(\underline{k}_2)$. Since only the directional correlation is observed it is necessary to sum over all the possible polarizations of the emitted photons. Then $\dot{N}_1(\underline{k}_1)$ and $\dot{N}_c(\underline{k}_2|\underline{k}_1)$ can be written as

$$\dot{N}_1(\underline{k}_1) = \sum_{\lambda_1} \dot{N}_1(\underline{k}_1\lambda_1),$$

$$\dot{N}_c(\underline{k}_2,\underline{k}_1) = \sum_{\lambda_1,\lambda_2} \dot{N}_c(\underline{k}_2\lambda_2|\underline{k}_1\lambda_1),$$

where $\dot{N}_1(k_1\lambda_1)$ denotes the counting rate of the first emitted photon of wave vector $\underline{k}_1$ and polarization λ_1 and $\dot{N}_c(\underline{k}_2\lambda_2|\underline{k}_1\lambda_1)$ denotes the

coincidence counting rate of $\gamma_1(\underline{k}_1\lambda_1)$ and $\gamma_2(\underline{k}_2\lambda_2)$. The quantity τ_c is the resolution time of the coincidence circuit.

This is the characteristic time of the instruments and we suppose that $\tau_c \gg \tau_N$, τ_N being the mean life time of the radioactive nucleus in the intermediate state of the cascade decay.

It is important to consider here the time sequence of the coincidence measurement processes.

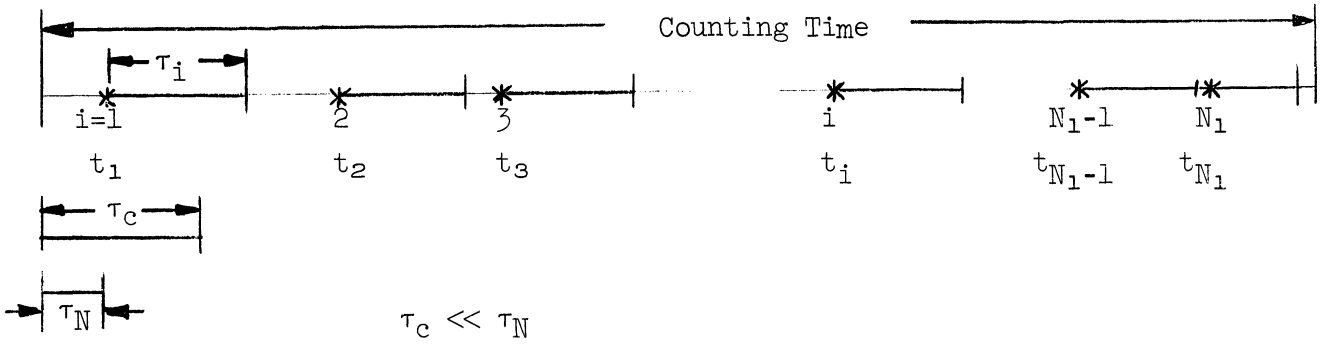


Fig. 2

As shown in Fig. 2, during a counting period the first detector D_1 is suppost to have detected γ, at the moments t_1, t_2,..., t_{N_1-1}, t_{N_1}. Once the first detector has detected a γ_1, it almost simultaneously opens the coincidence circuit for a small time interval of τ_c, the resolent time of the coincidence circuit. During this small time interval, if the second detector has detected a γ_2, a coincidence will be registered by the coincidence recorder. Therefore one sees that the coincidence counting rate $\dot{N}_c(\underline{k}_2|\underline{k}_1)$ is directly proportional to $\dot{N}_1(\underline{k}_1)$. For a direction-direction correlation measurements one can de-

fine a correlation function $W(\hat{k}_2|\hat{k}_1)$ by

$$W(\hat{k}_2/\hat{k}_1) = \frac{\dot{N}_c(\hat{k}_2/\hat{k}_1)}{\dot{N}_1(\hat{k}_1)} \tag{2.1}$$

Here $W(\hat{k}_2|\hat{k}_1)$ is actually the conditional probability to detect the second γ-ray of $\underline{k}_2$ following the detection of the first γ-ray of $\underline{k}_1$ within a certain small time interval. In general it will depend on the resolvent time τ_c and τ_N. But in the limit of $\tau_c \gg \tau_N$, $W(\hat{k}_2|\hat{k}_1)$ will depend only on the properties of the radioactive nucleus and its environment in the intermediate state.

If the temperature T of the source is such that the quantity kT is much larger than the nuclear energy level splitting due to the internal fields of the source or an external fields, then the nuclei in the source are not oriented and the counting rate of the first radiation will be isotropic and $\dot{N}_1(k_1)$ becomes $\dot{N}_1(\underline{k}_1)$, i.e., independent of directions of $\underline{k}_1$.

Furthermore, if the counting rate of the first γ-ray is a constant in time and the counter is not sensitive to the energy of the emitted photon, then $\dot{N}_1(k_1)$ is a constant $\dot{N}_1$.

Based on these assumptions the coincidence counting rate $\dot{N}_c(\underline{k}_2|k_1)$ also becomes a constant in time $\dot{N}_c(\hat{k}_2|\hat{k}_1)$ throughout the experiment. For a small time interval τ the counting rate $\dot{N}_c(\hat{k}_2|\hat{k}_1)$ can be written as;

$$\dot{N}_c(\hat{k}_2/\hat{k}_1) = \sum_{\lambda_1,\lambda_2} \frac{N_c(\hat{k}_2\lambda_2/\hat{k}_1\lambda_1;\tau) - N_c(\hat{k}_2\lambda_2/\hat{k}_1\lambda_1;0)}{\tau} \tag{2.2}$$

Then the direction-direction correlation function, defined by Eq. (2.1), becomes

$$W(\hat{k}_2|\hat{k}_1) = \sum_{\lambda_1,\lambda_2} \frac{N_c(\hat{k}_2\lambda_2|\hat{k}_1\lambda_1;\tau) - N_c(\hat{k}_2\lambda_2|\hat{k}_1\lambda_1)}{N,\tau} . \tag{2.3}$$

Any prediction about the behavior of a physical system is expressed as the mean value of a suitable operator. One must construct an operator $Q(\hat{k}_2\lambda_2|\hat{k}_1\lambda_1)$ such that the mean value of this operator corresponds to the correlation measurement of $N_c(\hat{k}_2\lambda_2|\hat{k}_1\lambda_1,t)$. Then one has

$$N_c(\hat{k}_2\lambda_2|\hat{k}_1\lambda_1;t) = Tr[Q(\hat{k}_2\lambda_2|\hat{k}_1\lambda_1)D(t)], \tag{2.4}$$

where D(t) is the density operator of the system at time t and satisfies

$$i\hbar\dot{D}(t) = [\mathcal{H}, D(t)] . \tag{2.5}$$

If $\mathcal{H}$ is not an explicit function of time, then the solution of Eq. (2.5) can be expressed in terms of the time evalution operator

$$D(t+\tau) = U(\tau)D(t)U^{\dagger}(\tau), \tag{2.6}$$

and

$$U(\tau) = e^{-i\mathcal{H}\tau/\hbar} , \qquad UU^{\dagger} = 1 .$$

Combining Eqs. (2.3), (2.4) and (2.6), the correlation function can be expressed as

$$W(\hat{k}_2|\hat{k}_1) = \frac{1}{N,\tau}\sum_{\lambda_1,\lambda_2} Tr\left[\frac{Q(\hat{k}_2\lambda_2|\hat{k}_1\lambda_1)}{\tau}\{U(\tau)D(0)U^{\dagger}(\tau) - D(0)\}\right] . \tag{2.7}$$

The operator $Q(\hat{k}_2\lambda_2|\hat{k}_1\lambda_1)$ can be chosen as

$$Q(\hat{k}_2\lambda_2|\hat{k}_1\lambda_1) = a^+(\hat{k}_2\lambda_2)a(\hat{k}_2\lambda_2)a^+(\hat{k}_1\lambda_1)a(\hat{k}_1\lambda_1), \tag{2.8}$$

where $a(\hat{k}\lambda)$ and $a^+(\hat{k}\lambda)$ are the conventional photon annihilation and creation operators. Using the representations defined in Section I and ignoring the off-diagonal elements of the density matrix, the trace of Eq. (2.7) can be expressed as;[11],[12]

$$W(\hat{k}_2|\hat{k}_1) = \sum_{\lambda_1\lambda_2}\sum_{\beta_i\beta_f} \langle\beta_f|U(\tau)|\beta_i\rangle\langle\beta_i|D(0)|\beta_i\rangle\langle\beta_i|U^+(\tau)|\beta_f\rangle\tau^{-1}. \tag{2.9}$$

In Eq. (2.9) we have dropped the factor $\dot{N}_1$ which is only a multiplicative constant in the correlation function.

The density operator D(o) describes the statistical mixture of the states at t=o. At this moment the system is represented by an orthonormal complete set of eigenkets $\{|\beta_i\rangle\}$ and D(o) can be written as

$$D(o) = \sum_{\beta'} |\beta_i'\rangle P(\beta_i')\langle\beta_i'|, \tag{2.10}$$

where $P(\beta_i')$ is the probability that the system is in an eigenstate $|\beta_i'\rangle$. The diagonal elements of D(o) become

$$\langle\beta_i|D(o)|\beta_i\rangle = P(\beta_i). \tag{2.11}$$

Substituting Eq. (2.11) into Eq. (2.9), the direction-direction correlation function can be expressed as

$$W(\hat{k}_2|\hat{k}_1) = \sum_{\lambda_1\lambda_2}\sum_{\beta_i\beta_f} P(\beta_i)\frac{|\langle\beta_i|U(\tau)|\beta_f\rangle|^2}{\tau}. \tag{2.12}$$

Essentially our problem has been reduced to the calculation of the absolute square of the off-diagonal matrix elements of the time evalution operator $U(\tau)$. By means of the resolvent method the off-diagonal matrix elements of $U(\tau)$ can be calculated as[11],[12] [Appendix I];

$$\frac{|\langle\beta_i|U(\tau)|\beta_f\rangle|^2}{\tau} = \left| \sum_{\beta\neq\beta_i,\beta_f} \frac{V^{\gamma}_{\beta_i\beta}\, V^{\gamma}_{\beta\beta_f}}{E_\beta - E_{\beta_i} + \frac{\hbar}{2}\gamma_\beta(-iE_{\beta_i})} \right|^2 \delta(E_{\beta_i} - E_{\beta_f}), \tag{2.13}$$

with

$$\frac{\hbar}{2}\gamma_\beta(-iE_{\beta_i}) = V_{\beta\beta} + \lim_{\lambda\to 0} \sum_{\beta_m\neq\beta}{}' \frac{|V_{\beta_m\beta}|^2}{E_{\beta_i} - E_{\beta_m} + i\lambda}, \tag{2.14}$$

and

$$V = V^{\gamma} + V^{NC}_{\tau}. \tag{2.15}$$

III. THEORY OF THE INFLUENCE OF EXTRANUCLEAR FIELDS ON ANGULAR CORRELATION

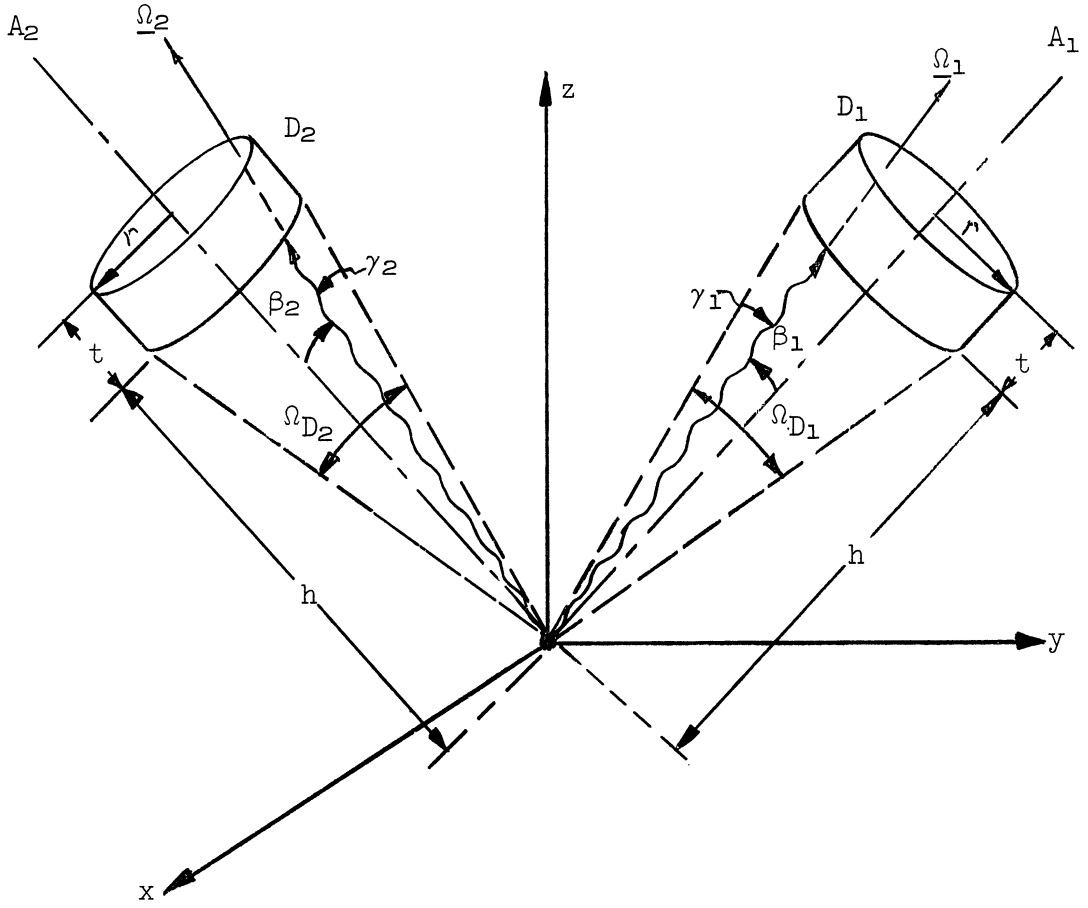


Fig. 3. Geometry for finite solid angle subtended by the detectors at the source.

By substituting Eq. (2.13) into Eq. (2.12) and taking into account the angular resolution and the efficiency of a detector for a γ-ray the direction-direction correlation function can be written as;

$$W(\vec{k}_2/\vec{k}_1) = \sum_{k_1 \epsilon \Delta k_1} \sum_{k_2 \epsilon \Delta k_2} \sum_{\lambda_1 \lambda_2} \sum_{\beta_i \beta_f} \mathcal{E}(x_1 \tau_1)\, \mathcal{E}(x_2 \tau_2)\, P(\beta_i) \times$$

$$\times \sum_{\substack{\beta\beta' \neq \\ \beta_i \beta_f}}{}' \frac{V^{\gamma}_{\beta_i\beta} V^{\gamma}_{\beta\beta_f} V^{\gamma *}_{\beta_i\beta'} V^{\gamma *}_{\beta'\beta_f}}{\left[E_\beta - E_{\beta_i} + \frac{\hbar}{2}\gamma_\beta(-iE_{\beta_i})\right]\left[E_{\beta'} - E_{\beta_i} + \frac{\hbar}{2}\gamma_{\beta'}(-iE_{\beta_i})\right]^*} \delta(E_{\beta_i} - E_{\beta_f}) \quad (3.1)$$

In Eq. (3.1) the $\sum_{\underline{k}_1 \epsilon \Delta \underline{k}_1}$ denotes the summation of directions of the first γ-ray over the finite solid angle Ω_{D_1} subtended by the detector D_1 at the source and the summation of all possible energies of the first γ-ray, $\mathcal{E}(x_1\tau_1)$ and $\mathcal{E}(x_2\tau_2)$ are respectively the efficiencies of the detector D_1 for the first γ-ray and of the detector D_2 for the second γ-ray. These efficiencies are functions of the distance traversed by the γ-ray inside the detector and can be expressed as

$$\mathcal{E}(x_1\tau_1) = 1 - e^{-\tau_1 x_1}$$

and

$$\mathcal{E}(x_2\tau_2) = 1 - e^{-\tau_2 x_2}$$

where τ_1 and τ_2 are the linear attenuation coefficients of γ_1 and γ_2 in the detectors D_1 and D_2 respectively, x_1 and x_2 are respectively the distances traversed by γ_1 and γ_2 inside the detectors D_1 and D_2 and can be calculated as a function of the thickness t and radius r of the detectors and the angle β's.[29]

The quantity $\frac{\hbar}{2}\gamma_\beta(-iE_{\beta i})$ is given by Eq. (2.14). It will be shown in thenext section that Eq. (2.14) can be expressed as

$$\frac{\hbar}{2}\gamma_\beta(-iE_{\beta_i}) = S_\beta(V_T^{NC}) + S_\beta(V^\gamma) - i\frac{\hbar}{2}\left\{\Gamma_\beta(V_T^{NC}) + \Gamma_\beta(V^\gamma)\right\}. \quad (3.2)$$

In Eq. (3.2) the quantities $S_\beta(V_T^{NC})$ and $S_\beta(V^\gamma)$ can be interpretated as the energy level displacements of the nuclear level corresponding to

state $|\beta>$ due to the dynamic part of the coupling between the radioactive nucleus and the crystal and due to the radiation interaction V^{γ}. The quantities $\frac{\hbar}{2}\Gamma_{\beta}(V_{\tau}^{NC})$ and $\frac{\hbar}{2}\Gamma_{\beta}(V^{\gamma})$ are respectively the level width due to the V_{τ}^{NC} and V^{γ}. For our purpose we will only note the existence of $S_{\beta}(V^{\gamma})$ and will not be concerned with its effects. Then the correlation function can be written as

$$W(\hat{k}_2|\hat{k}_1)=\sum_{\underline{k}_1\in\Delta\underline{k}_1}\sum_{\underline{k}_2\in\Delta\underline{k}_2}\sum_{\lambda_1\lambda_2}\sum_{\beta_i\beta_f}P(\beta_i)\mathcal{E}(\alpha_1\tau_1)\mathcal{E}(\alpha_2\tau_2)\sum_{\beta\beta'\neq\beta_i\beta_f}\times$$
$$\times V^{\gamma}_{\beta_i\beta}V^{\gamma}_{\beta\beta_f}V^{\gamma *}_{\beta_i\beta'}V^{\gamma *}_{\beta'\beta_f}\Big\{\big[E_{\beta}-E_{\beta_i}+S_{\beta}(V_{\tau}^{NC})-i\tfrac{\hbar}{2}\Gamma_{\beta}(V_{\tau}^{NC})-$$
$$-i\tfrac{\hbar}{2}\Gamma_{\beta}(V^{\gamma})\big]\big[E_{\beta'}-E_{\beta_i}+S_{\beta'}(V_{\tau}^{NC})+i\tfrac{\hbar}{2}\Gamma_{\beta'}(V_{\tau}^{NC})+$$
$$+i\tfrac{\hbar}{2}\Gamma_{\beta'}(V^{\gamma})\big]\Big\}^{-1}\delta(E_{\beta_i}-E_{\beta_f}). \qquad (3.3)$$

It has been shown that V^{γ} can be separated into two parts when the nuclear momentum is very small compared to the nucleonic momentum, one part depends only on the external degrees of freedom of the radioactive nucleus and the other part depends on the internal degrees of freedom of the radioactive nucleus, i.e.,

$$V^{\gamma}=e^{-i\underline{k}\cdot\underline{r}}V(\gamma). \qquad (3.4)$$

The factor $V(\gamma)$ in Eq. (3.4) depends only on the internal degrees of freedom of the radioactive nucleus and the photon field. The vector $\underline{r}$ is the position vector of the center of mass of the radioactive nucleus and $\underline{k}$ is the wave vector of the emitted photon.

Using Eq. (3.4) and making the expansion

$$|K\rangle = \sum_m |jm\rangle\langle jm|K\rangle , \tag{3.5}$$

the factor $V^{\gamma}_{\beta_i\beta} V^{\gamma}_{\beta\beta_f} V^{\gamma^*}_{\beta_i\beta_1} V^{\gamma^*}_{\beta_1\beta_f}$ in Eq. (3.3) can be expressed as

$$V^{\gamma}_{\beta_i\beta} V^{\gamma}_{\beta\beta_f} V^{\gamma^*}_{\beta_i\beta'} V^{\gamma^*}_{\beta'\beta_f} = \langle\alpha_i| e^{-i\vec{k}_1\cdot\vec{r}}|\alpha\rangle\langle\alpha'| e^{i\vec{k}_1\cdot\vec{r}}|\alpha_i\rangle \times$$

$$\times \langle\alpha| e^{-i\vec{k}_2\cdot\vec{r}}|\alpha_f\rangle\langle\alpha_f| e^{i\vec{k}_2\cdot\vec{r}}|\alpha'\rangle \times$$

$$\times \sum_{\substack{m\,m'\\ m''m'''}}{}' \langle jm|K\rangle\langle K|jm'\rangle\langle jm'''|K'\rangle\langle K'|jm''\rangle \times$$

$$\times \langle jm|V(\gamma)|j_i m_i\rangle \langle jm'''|V(\gamma)|j_i m_i\rangle^* \times$$

$$\times \langle j_f m_f|V(\gamma)|jm\rangle \langle j_f m_f|V(\gamma)|jm''\rangle^* . \tag{3.6}$$

The quantities E_β, $E_{\beta i}$ and $E_{\beta f}$ in Eq. (3.3) are the eigenvalues from the following equations;

$$H_0|\beta\rangle = E_\beta|\beta\rangle , \tag{3.7a}$$

$$H_0|\beta_i\rangle = E_{\beta_i}|\beta_i\rangle , \tag{3.7b}$$

and

$$H_0|\beta_f\rangle = E_{\beta_f}|\beta_f\rangle . \tag{3.7c}$$

Substituting

$$H_o = H^N + V_o^{NC} + H^C + H^\gamma$$

into Eqs. (3.7) one obtains;

$$E_{\beta_i} - E_{\beta_f} = E_{K_i} - E_{K_f} + E_{\alpha_i} - E_{\alpha_f} - \hbar c k_1 - \hbar c k_2 , \tag{3.8}$$

and

$$E_\beta - E_{\beta_i} = E_K - E_{K_i} + E_\alpha - E_{\alpha_i} + \hbar c k_1 \tag{3.9}$$

For our purpose we will neglect the interactions between the radioactive nucleus and the crystalline fields before the emission of the first photon and after the emission of the second photon. The probability $P(\beta i)$ in Eq. (3.3) can now be decomposed as $P(\beta i)=P(\alpha_i)P(m_i)$. Here $P(\alpha_i)$ and $P(m_i)$ are the probabilities for finding the crystal and the radioactive nucleus in states $|\alpha_i>$ and $|j_i^1 m_i>$ respectively. Then substituting Eqs. (3.6), (3.8) and (3.9) and neglecting the change of the crystal state due to the emission of the γ-rays, Eq. (3.3) can be expressed as;

$$W(\hat{k}_2/\hat{k}_1) = \sum_{\underline{k}_1 \in \Delta \underline{k}_1} \sum_{\underline{k}_2 \in \Delta \underline{k}_2} \sum_{K K'} \sum_{\substack{m m' \\ m'' m'''}} \mathcal{E}(x_1 \tau_1)\, \mathcal{E}(x_2 \tau_2) \times$$

$$\times \Lambda^{(1)}(m' m''')\, \Lambda^{(2)}(m m')\, <jm|K><K|jm'> \times$$

$$\times <jm'''|K'><K'|jm''> \{ E_K - E_{K_i} + \hbar c k_1 +$$

$$+\langle S_K(V_\tau^{NC})\rangle_T - i\frac{\hbar}{2}\{\langle \Gamma_K(V_\tau^{NC})\rangle_T + \langle \Gamma_K(V^\gamma)\rangle_T\}]\times$$

$$\times\{E_{K'} - E_{K_i} + \hbar c k_1 + \langle S_{K'}(V_\tau^{NC})\rangle_T +$$

$$+ i\frac{\hbar}{2}\{\langle \Gamma_{K'}(V_\tau^{NC})\rangle_T + \langle \Gamma_{K'}(V^\gamma)\rangle_T\}\}^{-1}\times$$

$$\times\delta(E_{K_i} - E_{K_f} - \hbar c k_1 - \hbar c k_2)\,. \tag{3.10}$$

where

$$\Lambda^{(1)}(m'm''') = \sum_{j_1}\sum_{m_i} P(m_i)\langle jm'|V(\gamma)|j_i m_i\rangle\times$$

$$\times\langle jm'''|V(\gamma)|j_i m_i\rangle^*, \tag{3.11}$$

$$\Lambda^{(2)}(mm'') = \sum_{j_2}\sum_{m_f}\langle j_f m_f|V(\gamma)|jm\rangle\times$$

$$\times\langle j_f m_f|V(\gamma)|jm''\rangle^*, \tag{3.12}$$

and

$$\langle S_K(V_\tau^{NC})\rangle_T = \sum_{\beta_i} P(\beta_i) S_K(V_\tau^{NC})\,, \tag{3.13}$$

is the averaged energy level displacement over the initial state of the system. In Eq. (3.10) we have assummed that the system is initially at thermal equilibrium and doing the thermal average we have followed

an approximation which is made by replacing the average of a ratio of two functions by the ratio of the averaged functions.[12]

The radiation interaction energy $V(\gamma)$ can be expressed as a contraction of two irreducible tensors of the same rank, i.e.;

$$V(\gamma) = \sum_{L} b_L \sum_{M=-L}^{L} (-1)^M A_{L,-M} T_{L,M}, \qquad (3.14)$$

where b_L is a constant and is a function of L, $T_{L,M}$ is an irreducible tensor operator of rank L and depends only on the internal degrees of freedom of the radioactive nucleus, and A_{L-M} is an irreducible tensor operator of rank L and depends only on the degrees of freedom of the emitted photon.

From Eq. (3.14) and using the equation for the rotational transformation of an irreducible tensor

$$T_{L,M} = \sum_{\mu} D^{L}_{\mu M}(R)\, T_{L,\mu},$$

and the Wigner-Eckart theorem one has;

$$\langle jm'|V(\gamma)|j_i m_i\rangle = \sum_{L_1 M_1} \sum_{\mu_1} (-1)^{M_1} b_{L_1} A_{L_1,-M_1} \langle j\|T_{L_1}\|j_i\rangle \times (jm'; L_1\mu_1|j_i m_i) D^{L_1}_{\mu_1 M_1}(R_1). \qquad (3.15)$$

Where $D^{L_1}(R_1)$ is a rotational matrix which transforms an irreducible tensor in the radiation axis into a chosen quantization axis through R_1 rotation which is a set of Enler angles. The quantity $(jm^1; L_1\mu_1|j_i m_i)$ is the clebsch—Gordan coefficient and $\langle j\|T_{L_1}\|j_L\rangle$ is the reduced matrix element of the nuclear tensor operator. By using Eq. (3.15), Eq. (3.11)

can be written as;

$$\Lambda^{(1)}(m'm''') = \sum_{j_1} \sum_{m_i} P(m_i) \sum_{L_1 L_1' M_1 M_1' \mu_1 \mu_1'} \sum (-1)^{M_1 + M_1'} \times$$

$$\times b_{L_1} b_{L_1'} A_{L_1 - M_1} A^{*}_{L_1' - M_1} \langle j \| T_{L_1} \| j_i \rangle \times$$

$$\times \langle j \| T_{L_1'} \| j \rangle^{*} (j m'; L_1 \mu_1 | j_i m_i) \times$$

$$\times (j m''; L_1' \mu_1' | j_i m_i) D^{L_1}_{\mu_1 M_1}(R_1) D^{L_1'*}_{\mu_1' M_1'}(R_1) . \qquad (3.16)$$

By using Racah's technique $\Lambda^{(1)}(m'm''')$ can be calculated as [Appendix II];

$$\Lambda^{(1)}(m'm''') = (2j_i + 1) \sum_{L_1 L_1' \tau_1 \nu_1} \sum (-1)^{m' + L_1 - j_i} \langle j \| T_{L_1} \| j_i \rangle \times$$

$$\times \langle j \| T_{L_1'} \| j \rangle^{*} C_{\nu_1 \tau_1}(L_1 L_1') (j m'; j - m''' \nu_1 | m' - m''') \times$$

$$\times W(j j L_1 L_1'; \nu_1 j_i) D^{\nu_1}_{m''' - m', -\tau_1}(R_1) , \qquad (3.17)$$

where

$$C_{\nu_1 \tau_1}(L_1 L_1') = \sum_{j_1} \sum_{M_1} (-1)^{L_1 - M_1} b_{L_1} b_{L_1'} A_{L_1, -M} A^{*}_{L_1', -M_1' - \tau_1} \times$$

$$\times (L_1 M_1; L_1' - M_1 - \tau_1 | \nu_1 - \tau_1) , \qquad (3.18)$$

is the first γ-ray radiation parameter which is a characteristic of the emitted radiation and is independent of the nuclear states involved in the tension. A detail discussion and calculations of the radiation parameters have been given by L.C. Biedenharn and M.E. Rose.[14]

Similarly $\Lambda^{(2)}(mm'')$ can be expressed as;

$$\Lambda^{(2)}(mm'') = (2j+1)\sum_{L_2 L_2'}\sum_{\nu_2 \tau_2}(-1)^{m''+L_2'-j_f}\langle j_f \| T_{L_2} \| j\rangle\langle j_f \| T_{L_2'} \| j\rangle^* \times$$
$$\times C_{\nu_2\tau_2}(L_2 L_2')(jm, j-m''|\nu_2 m-m'')W(jjL_2L_2';\nu_2 j_f)\times$$
$$\times D^{\nu_2}_{m-m'',-\tau_2}(R_2), \qquad (3.19)$$

and

$$C_{\nu_2\tau_2}(L_2L_2') = \sum_{\lambda_2}\sum_{M_2}(-1)^{L_2-M_2} b_{L_2} b_{L_2'} A_{L_2-M_2} A^*_{L_2'-M_2-\tau_2} \times$$
$$\times (L_2 M_2, L_2'-M_2-\tau_2|\nu_2-\tau_2). \qquad (3.20)$$

Combining Eqs. (3.17) and (3.19), the products of the radiation matrix elements in Eq. (3.10) can be written as;

$$\sum_{\lambda_1\lambda_2}\sum_{m_i m_f} P(m_i)\langle jm'|\bar{V}(\gamma)|j_i m_i\rangle\langle jm''|\bar{V}(\gamma)|j_i m_i\rangle^* \times$$

$$\times\langle j_f m_f|\bar{V}(\gamma)|jm\rangle\langle j_f m_f|\bar{V}(\gamma)|jm''\rangle^* =$$

$$= (2j+1)(2j_i+1)\sum_{L_1L_1'}\sum_{L_2L_2'}\sum_{\tau_1\tau_2}\sum_{\nu_1\nu_2}(-1)^{m'+m''+L_1+L_2'-j_i'-j_f}\times$$

$$\times \langle j \| T_{L_1} \| j_i \rangle \langle j \| T_{L_1'} \| j_i \rangle^* \langle j_f \| T_{L_2} \| j \rangle \langle j_f \| T_{L_2'} \| j \rangle^* \times$$

$$\times C_{\nu_1 \tau_1}(L_1 L_1') C_{\nu_2 \tau_2}(L_2 L_2') (jm; j-m''' | \nu_1 m'-m''') \times$$

$$\times (jm; j-m'' | \nu_2 m-m'') W(jjL_1L_1'; \nu_1 j_i) \times$$

$$\times W(jjL_2L_2'; \nu_2 j_f) D^{\nu_1}_{m'''-m', -\tau_1}(R_1) D^{\nu_2}_{m-m', -\tau_2}(R_2). \qquad (3.21)$$

For the direction-direction correlation and thepure multipole radiations with multipolarities of L_1 and L_2 Eq. (3.21) can be calculated as [Appendix III];

$$A^{(1)}_{(m'm''')} A^{(2)}_{(mm'')} = \sum_{\nu_1 \nu_2} I_{\nu_1}(j_i j j) \Pi_{\nu_2}(j_f j j) \times$$

$$\times (jm'; j-m''' | \nu_1 m'-m''') (jm; j-m'' | \nu_2 m-m'') \times$$

$$\times Y^*_{\nu_1, m'''-m'}(\theta_1 \varphi_1) Y_{\nu_2 m''-m}(\theta_2 \varphi_2). \qquad (3.22)$$

Combining Eqs. (3.22) and (3.10), the correlation function can be expressed as

$$W(\hat{k}_2/\hat{k}_1)=\sum_{\nu_1\nu_2}\sum_{\substack{mm'\\ m''m'''}}\sum_{KK'}(jm;j-m''/\nu_1 m'-m''')\times$$

$$\times(jm;j-m''/\nu_2 m-m'')\langle jm|K\rangle\langle K|jm'\rangle\langle jm'''|K'\rangle\times$$

$$\times\langle K'|jm''\rangle\,\widetilde{Y}^{*}_{\nu_1 m'''-m'}(\theta_1\varphi_1)\,\widetilde{Y}_{\nu_2 m''-m}(\theta_2\varphi_2)\times$$

$$\times\int k_1^2dk_1\int k_2^2dk_2\, I_{\nu_1}(j_i j j)\,II_{\nu_2}(j_f j j)\Big\{\big[E_K-E_{K_i}+$$

$$+\hbar c k_1+\langle S_K(V_\tau^{NC})\rangle_T-i\frac{\hbar}{2}\langle\Gamma_K(V_\tau^{NC})\rangle_T-i\frac{\hbar}{2}\langle\Gamma_K(V)\rangle_T\big]\times$$

$$\times\Big[E_{K'}-E_{K_i}+\hbar c k_1+\langle S_{K'}(V_\tau^{NC})\rangle_T+i\frac{\hbar}{2}\langle\Gamma_{K'}(V_\tau^{NC})\rangle_T+\frac{i\hbar}{2}\langle\Gamma_{K'}(V)\rangle_T\Big]^{-1}\Big\}\times$$

$$\times\delta(E_{K_i}-E_{K_f}-\hbar c k_1-\hbar c k_2)\,, \qquad (3.23)$$

where

$$\widetilde{Y}^{*}_{\nu_1 m'''-m'}(\theta_1\varphi_1)=\int_{\Omega_{D_1}} Y^{*}_{\nu_1 m'''-m'}(\theta_1\varphi_1)\,\mathcal{E}(x_1\tau_1)\,d\Omega_1\,, \qquad (3.24)$$

and

$$\widetilde{Y}_{\nu_2 m''-m}(\theta_2\varphi_2)=\int_{\Omega_{D_2}} Y_{\nu_2 m''-m}(\theta_2\varphi_2)\,\mathcal{E}(x_2\tau_2)\,d\Omega_2 \qquad (3.25)$$

In case of unperturbed correlation and the direction of the first γ-ray is chosen as the quantization axis, the quantities $\widetilde{Y}^{*}_{\nu_1 m''-m'}(\theta_1\phi_2)$ and $\widetilde{Y}_{\nu_2 m''-m}(\theta_2\phi_2)$ have been calculated by M.E. Rose.[29]

Carrying out the integrations of k_1 and k_2 in Eq. (3.23), the correlation function can be expressed approximately as (Appendix IV);

$$W(\vec{k}_2/\vec{k}_1) = \sum_{\nu_1\nu_2} \sum_{\substack{m m' \\ m''m'''}} I_{\nu_1}(j_i j_f) II_{\nu_2}(j_f j_i) III_{\nu_1\nu_2}(m m' m'' m''') \times$$
$$\times \tilde{Y}^*_{\nu_1 m''-m}(\theta_1 \varphi_1)\, \tilde{\tilde{Y}}_{\nu_2 m'-m}(\theta_2 \varphi_2), \qquad (3.26)$$

with

$$III_{\nu_1\nu_2}(m m' m'' m''') = \sum_{KK'} (j m'; j-m'''/\nu_1 m'-m''') \times$$

$$\times (j m; j-m''/\nu_2 m-m''') \langle j m/K\rangle\langle K/j m'\rangle \times$$

$$\times \langle j m''/K'\rangle\langle K'/j m''\rangle \Big\{ \frac{\hbar}{2}\Big[\langle \Gamma_K(\bar{V}_\tau^{NC})\rangle_T + \langle \Gamma_{K'}(\bar{V}_\tau^{NC})\rangle_T +$$

$$+ 2\Gamma_K(V^\gamma)\Big] - i\Big[E_K - E_{K'} + \langle S_K(\bar{V}_\tau^{NC})\rangle_T - \langle S_{K'}(\bar{V}_\tau^{NC})\rangle_T\Big]\Big\}^{-1}, \qquad (3.27)$$

and

$$E_K - E_{K'} = \sum_{m m'} \Big[\langle j m'/K\rangle\langle K/j m\rangle - \langle j m'/K\rangle\langle K/j m\rangle\Big] \times$$
$$\times \langle j m/V_0^{NC}/j m'\rangle, \qquad (3.28)$$

where V_0^{NC} is the static part of the interaction Hamiltonian between the radioactive nucleus and the extranuclear fields.

In Eq. (3.26) the factors $I_{\nu_1}(j_i jj)$ and $II_{\nu_2}(j_f jj)$ depend only on the degrees of freedom of the successive radiations. They are independent of the external perturbing fields. The influence of the external fields on the angular correlation is entirely contained in the third factor $III_{\nu_1\nu_2}(mm'm''m''')$. This perturbation factor is the so-called attenuation factor for the correlation function.

The selection rules of the clesch-Gordan coefficients, given in the Appendixes II and III, require that the indices ν_1 and ν_2 are even integers and are restricted by the following inequalities;

$$\left.\begin{aligned} &0 \leq \nu_1 \leq 2j, \quad 0 \leq \nu_1 \leq 2L_1; \\ &0 \leq \nu_2 \leq 2j, \quad 0 \leq \nu_2 \leq 2L_2; \end{aligned}\right\} \tag{3.29}$$

The integers L_1 and L_2 are the highest orders in the multiple expansions of the first and the second γ-rays respectively.

For vanishing perturbation, Eq. (3.27) reduces to

$$III_{\nu_1\nu_2}(mm'm''m''') = \left(\frac{\tau_N}{\hbar}\right)^2 (jm; j-m'''|\nu_1, m'-m''') \times$$

$$\times (jm; j-m''|\nu_2, m-m'') \delta_{mm'} \delta_{m'''m''}, \tag{3.30}$$

where τ_N is the mean life time in the intermediate state of the radioactive nucleus. Substituting Eq. (3.30) into Eq. (3.26) and making use of the orthogonal properties of the clebsch-Gordan coefficients and the addition theorem of the spherical harmonics and ignoring the angular

resolution corrections one can easily show that Eq. (3.26) reduces to the unperturbed angular correlation function

$$W(\hat{k}_2|\hat{k}_1) = \sum_{\nu} I_{\nu_1}(\gamma_i;jj)\, II_{\nu_2}(\gamma_f;jj)\, P_{\nu}(\cos\theta_{12}) . \qquad (3.31)$$

Equation (3.31) is the well-known unperturbed correlation function which can be expressed as the sum of even Legendre polynomials.

IV. THE ATTENUATION COEFFICIENTS

As shown in Eq. (3.27), the total influence of the extranuclear fields on the angular correlation can be factored out as a perturbation factor. This perturbation factor depends primarily on the strength of the interaction and on the length of time it can act on the nucleus, that is the mean life of the intermediate state. For a static perturbation, the angular correlation is measureably perturbed if $\omega\tau_N \gtrsim 1$. Here the magnitude of the perturbation is measured by a precession frequency ω. For a magnetic interaction ω is equal to the Larmor frequency. In case of the electric quadrupole interaction ω is proportional to the nuclear quadrupole moment in the intermediate state and the electric field gradient. For a time-dependent perturbation the magnitude of the perturbation is characterized by a relaxation time γ such that when $\gamma\tau_N \gtrsim 1$ the angular correlation is perturbed. This criterion of measurable perturbation depends on the sensitivety of the experimental arrangement. With the present experimental techniques the limits are given by $\omega\tau_N \gtrsim 0.01$ or $\gamma\tau_N \gtrsim 0.01$.[15] The precession frequencies for a large number of solids have been measured by microwave and nuclear resonance techniques.[16] For quadrupole interaction, values for $\omega/2\pi$ as large as 3000 MC/S have been observed. According to our criterion, cascade with a life time $\tau_N \gtrsim 10^{-12}$ sec can thus be measurably perturbed.

For a radioactive nucleus sitting in a lattice site the possible extranuclear perturbations will be the atomic hyperfine interaction, the nuclear magnetic dipole-dipole interaction with the surrounding nuclei (or the magnetic interaction with an extranally applied magnetic field) and the nuclear electric quadrupole interaction with the crystalline field. In our discussions we will focus our attention on the interactions between the radioactive nucleus and the crystalline fields.

By substituting $V=V^{\gamma}+V_c^{NC}$ into Eq. (2.14), the function $<\frac{\hbar}{2}\gamma_K(-iE_{\beta_i})>_T$ defined by

$$\left\langle \frac{\hbar}{2}\gamma_K(-iE_{\beta_i})\right\rangle_T = \sum_{\beta_i} P(\beta_i)\frac{\hbar}{2}\gamma_K(-iE_{\beta_i}), \tag{4.1}$$

can be expressed as;

$$\left\langle \frac{\hbar}{2}\gamma_K(-iE_{\beta_i})\right\rangle_T = \left\langle \frac{\hbar}{2}\gamma_K(V_c^{NC})\right\rangle_T + \frac{\hbar}{2}\gamma_K(V^{\gamma}), \tag{4.2}$$

with

$$\left\langle \frac{\hbar}{2}\gamma_K(V_c^{NC})\right\rangle_T = \lim_{\lambda\to 0}\sum_{\beta_i} P(\beta_i)\sum_{\beta'\neq\beta}\frac{|\langle\beta'|V_c^{NC}|\beta\rangle|^2}{E_{\beta_i}-E_{\beta'}+i\lambda}, \tag{4.3}$$

and

$$\left\langle \frac{\hbar}{2}\gamma_K(V^{\gamma})\right\rangle_T = \lim_{\lambda\to 0}\sum_{\beta_m\neq\beta}\frac{|\langle\beta_m|V^{\gamma}|\beta\rangle|^2}{E_{\beta_i}-E_{\beta_m}+i\lambda}. \tag{4.4}$$

In Eq. (4.2) we have neglected the changes of states of the lattice system due to the emissions of the cascade photons. The eigenkets $|\beta>$'s in Eqs. (4.3) and (4.4) should be replaced by the expansion

$$|\beta\rangle = \sum_m |jm\rangle|\alpha\rangle|\eta_{k_1\lambda_1}, \eta_{k_2\lambda_2}\rangle\langle jm|K\rangle . \tag{4.5}$$

In the following, we will first calculate the explicit expressions for the function $\langle \frac{\hbar}{2} \gamma_K (V_T^{NC}) \rangle_T$ for the nuclear electric quadrupole couplings and the magnetic dipole-dipole couplings and then consider in some details the effect due to the radiation interaction as given by Eq. (4.4).

A. THE ELECTRIC QUADRUPLE COUPLING

A nucleus with spin $j \geq 1$ may have an electric quadrupole moment. This electric quadrupole moment will interact with the crystalline field gradient and produce energy splitting. For a static quadrupole interaction one can show that the energy splittings are not uniform and the states are two-fold degenerate. Thus the influence of a quadrupole interaction on an angular correlation can no longer be described semiclassically by a single precession frequency.

The electrostatic interaction Hamiltonian can generally be expressed as the contraction of two tensors of the same rank,

$$V^{EQ} = \sum_{k=0}^{\infty} \sum_{\zeta=-k}^{k} (-1)^{\zeta} Q_{\zeta}^{k} V_{-\zeta}^{k} , \tag{4.6}$$

where Q_{ζ}^{k} and $V_{-\zeta}^{k}$ are respectively the nuclear electric moment tensor and the field tensor of rank k and can be written as;

$$Q_{\zeta}^{k} = \sum_{p} e_p r_p^{k} Y_{\zeta}^{-k}(\theta_p, \varphi_p) , \tag{4.7}$$

and

$$V_{-\zeta}^{k}=\sum_{i}\frac{e_i}{r_{ci}^{k+1}}\,Y_{-\zeta}^{k}(\theta_{ci},\phi_{ci}), \tag{4.8}$$

In Eq. (4.8) we have assumed that the electrostatic field is caused by the point charges in the lattice system and e_i is the point charge at the ith lattice point $(r_{ci},\theta_{ci}\phi_{ci})$. The charge e_p is the nucleonic charge in the nucleus at the point (r_p,θ_p,ϕ_p). By letting k=2 in Eq. (4.5), we have the electric quadrupole interaction Hamiltonian;

$$V^{EQ}=\sum_{\zeta=-2}^{2}(-1)^{\zeta}Q_{\zeta}^{(2)}V_{-\zeta}^{(2)}. \tag{4.9}$$

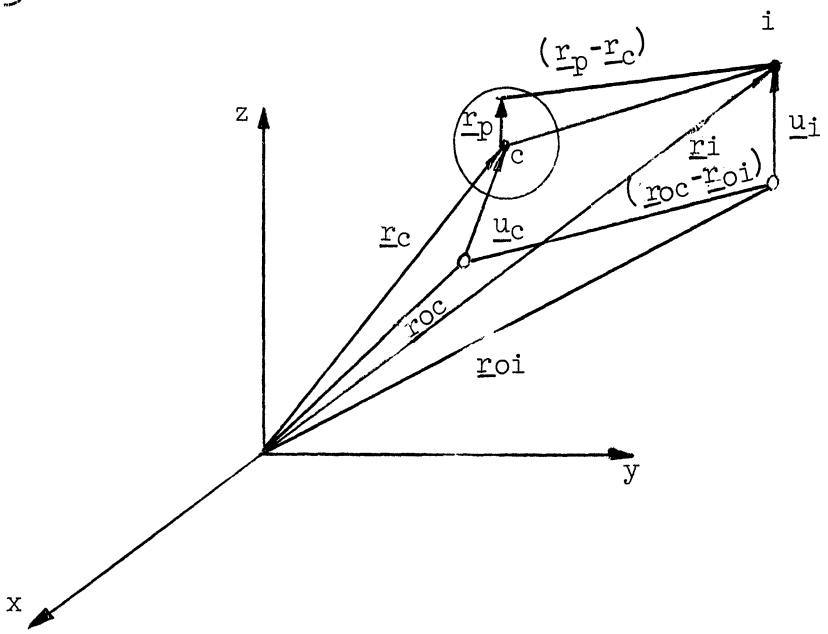


Fig. 4

From Eq. (4.8), one sees that the interaction depends on the length and orientation of the relative vector $\underline{r}_{ci}$ between the radioactive nucleus and the ith lattice point. For a rigid lattice the relative position vectors are fixed and the interaction becomes static.

Practically, there exist no ideal, rigid crystals and the nuclei in the crystal experience thermal vibrations. The effect of these thermal vibrations is to create at the radioactive nucleus the time-dependent electric field gradients on magnetic fields. As shown in Fig. 4 the vibration of a nucleus in a crystal is described by a vector $\underline{u}$ which indicates the displacement of the nucleus from its equilibrium position. Then the relative position vector between the radioactive nucleus and the ith lattice point becomes $\underline{r}_{ci}=\underline{u}_{oci}-\underline{u}_{ci}$. Here the vector $\underline{u}_{oci}$ is the relative equilibrium position vector and $\underline{u}_{ci}=\underline{u}_c - \underline{u}_i$ is the relative displacement vector between these two nuclei. When the temperature is far below the melting point of the crystal, one has $u \ll a_0$ then the field gradient tensors can be expanded at the equilibrium positions as;(27)

$$V_{-\zeta}^{(2)} = V_{-\zeta}^{(2)}(0) + \frac{1}{2!}\sum_{ij}\sum_{\mu\nu}\left(\frac{\partial^2 V_{-\zeta}^{(2)}}{\partial u_\mu(i)\partial u_\nu(j)}\right)u_\mu(i)u_\nu(j)+\cdots, \qquad (4.10)$$

where $V_{-\zeta}^{(2)}(0)$ is an irreducible component of the field gradient tensor evaluated at the equilibrium positions of the lattice points and $u_\mu(i)$ is the μth component of the relative displacement between the radioactive nucleus and the ith lattice point. If we assume the binary interaction potential as given by Eq. (4.8), then we have

$$\frac{\partial^2 V_{-\zeta}^{(2)}(r_{ci},\theta_{ci},\varphi_{ci})}{\partial u_\mu(i)\partial u_\nu(j)} = \frac{\partial^2 V_{-\zeta}^{(2)}(r_{ci},\theta_{ci},\varphi_{ci})}{\partial u_\mu(i)\partial u_\nu(i)}\delta_{ij}\,. \qquad (4.11)$$

From Eqs. (4.8) and (4.11), Eq. (4.10) can be calculated as

$$V_{-\zeta}^{(2)} = V_{-\zeta}^{(2)}(0) + \frac{15}{2}\sum_i \sum_{\mu\nu} \frac{\rho_i}{a_{oci}^5} Y_{-\zeta}^2(\theta_{oci}-\varphi_{oci}) \times$$

$$\times \eta_{i\mu}\eta_{i\nu} u_\mu(i) u_\nu(i) + \cdots , \qquad (4.12)$$

where ρ_i denotes the effective point charge at the ith lattice point and $\eta_{i\mu}$ and $\eta_{i\nu}$ are respectively the directional cosines of $\underline{a}_{oci}$ in the μth and νth directions. By using the normal mode expansion of the displacement, $\underline{u}$ can be expressed as a function of the phonon creaction and annihilation operators

$$\underline{u}_c(\underline{r}_{oc}) = \sum_{\underline{q}j} \left[\frac{\hbar}{2M_c\omega(\underline{q}j)}\right]^{\frac{1}{2}} \underline{e}(\underline{q}j)\left[a^+(\underline{q}j)e^{-i\underline{q}\cdot\underline{r}_{oc}} + a(\underline{q}j)e^{i\underline{q}\cdot\underline{r}_{oc}}\right], \qquad (4.13)$$

Then $\underline{u}(i)\equiv\underline{u}_c(r_{oc})-\underline{u}_i(\underline{r}_{oi})$ can be expressed as

$$\underline{u}(i) = \sum_{\underline{q}j} \left[\frac{\hbar}{2M_c\omega(\underline{q}j)}\right]^{\frac{1}{2}} \underline{e}(\underline{q}j)\left\{a^+(\underline{q}j)e^{-i\underline{q}\cdot\underline{r}_{oc}} \times (1-\epsilon_{ci}e^{-i\underline{q}\cdot\underline{a}_{oci}}) + a(\underline{q}j)e^{i\underline{q}\cdot\underline{r}_{oc}}(1-\epsilon_{ci}e^{i\underline{q}\cdot\underline{a}_{oci}})\right\}, \qquad (4.14)$$

where $\epsilon_{ci} = \dfrac{M_c}{M_i}$ is the ratio of the mass of the radioactive nucleus to the ith nucleus in the crystal. In case of single crystal ϵ_{ci} is the isotopic factor.

When the wavelength of phonon is appreciably larger than the interatomic spacing, we make the approximation

$$\exp(-i\underline{q}\cdot\underline{a}_{oci}) \simeq 1 - iqa_{oci}(\hat{q}\cdot\hat{a}_{oci}). \qquad (4.15)$$

This approximation will considerably simplify our calculations. Although it may break down at the upper end of the Debye spectrum, it will not affect the results appreciably.[18]

From Eqs. (4.15) and (4.14) and after some calculations one has;

$$u_\mu(i)u_\nu(i)=\sum_{\underline{q}\underline{q}'}\left(\frac{\hbar}{2M_c}\right)\left[\frac{1}{\omega(\underline{q}\mu)\omega(\underline{q}'\nu)}\right]^{\frac{1}{2}}a_{oci}^2\Big\{a^+(\underline{q}\mu)a^+(\underline{q}'\nu)\times$$

$$\times e^{-i(\underline{q}+\underline{q}')\cdot\underline{r}_{oc}}\left[d_{ci}+i\epsilon_{ci}q(\hat{q}\cdot\hat{a}_{oci})\right]\left[d_{ci}+\epsilon_{ci}q'(\hat{q}'\cdot\hat{a}_{oci})\right]+a(\underline{q}\mu)\times$$

$$\times a^+(\underline{q}'\nu)e^{i(\underline{q}-\underline{q}')\cdot\underline{r}_{oc}}\left[d_{ci}-i\epsilon_{ci}q(\hat{q}\cdot\hat{a}_{oci})\right]\left[d_{ci}+i\epsilon_{ci}q'(\hat{q}'\cdot\hat{a}_{oci})\right]+$$

$$+a^+(\underline{q}\mu)a(\underline{q}'\nu)e^{-i(\underline{q}-\underline{q}')\cdot\underline{r}_{oc}}\left[d_{ci}+i\epsilon_{ci}q(\hat{q}\cdot\hat{a}_{oci})\right]\left[d_{ci}-i\epsilon_{ci}q'\times\right.$$

$$\left.\times(\hat{q}'\cdot\hat{a}_{oci})\right]+a(\underline{q}\mu)a(\underline{q}'\nu)e^{i(\underline{q}+\underline{q}')\cdot\underline{r}_{oc}}\left[d_{ci}-i\epsilon_{ci}q(\hat{q}\cdot\hat{a}_{oci})\right]\times$$

$$\left.\times\left[d_{ci}-i\epsilon_{ci}q'(\hat{q}'\cdot\hat{a}_{oci})\right]\right\},\qquad(4.16)$$

where the quantity d_{ci} stands for $(1-\epsilon_{ci})/a_{oci}$.

Equation (4.16) describes all the possible two-phonon processes induced by the second term of Eq. (4.10). The first term of Eq. (4.16) describes the processes of creation of two phonons at the same time, while the last term describes the annihilations of two phonons at the same time. The second and the third term describe the processes of creating one phonon and annihilating another phonon at the same time. This process is the well-known Raman process and is overwhelmingly more

important than the creation or annihilation of two phonons simultaneously. Thus in our discussion we will only consider the Raman processes.

Then the quadrupole coupling Hamiltonian can be expressed as

$$V^{EQ} = V_o^{EQ}(0) + V_\tau^{EQ}, \tag{4.17}$$

with

$$V_o^{EQ}(0) = \sum_{\zeta=-2}^{2} (-1)^\zeta Q_\zeta^{(2)} V_{-\zeta}^{(2)}(0), \tag{4.18}$$

and

$$V_\tau^{EQ} = \left(\frac{15\hbar}{4M_a}\right) \sum_{\zeta} \sum_{i}^{(i\neq i')} \sum_{\mu\nu} \sum_{\underline{\xi}\underline{\xi}'} (-1)^\zeta \left(\frac{\rho_i}{a_{oi}^3}\right) \eta_{i\mu}\eta_{i\nu} \times$$
$$\times \left[\frac{1}{\omega(\underline{\xi}\mu)\omega(\underline{\xi}'\nu)}\right]^{\frac{1}{2}} \left[a_{oi} - i\epsilon_{oi}\underline{\xi}\cdot\underline{a}_{oi}\right] \times$$
$$\times \left[a_{oi} + i\epsilon_{oi}\underline{\xi}'\cdot\underline{a}_{oi}\right] Y_{-\zeta}^{(2)}(\theta_{oi}, \varphi_{oi})\, Q_\zeta^{(2)} \times$$
$$\times a(\underline{\xi}\mu)\, a^\dagger(\underline{\xi}'\nu)\, e^{i(\underline{\xi}-\underline{\xi}')\cdot\underline{r}_{oo}}. \tag{4.19}$$

Here $V_o^{EQ}(0)$ represents the static quadrupole coupling Hamiltonian when the crystal is considered as a rigid lattice system and V_τ^{EQ} describes the dynamic quadrupole coupling Hamiltonian induced by the Raman processes. Substituting Eq. (4.18) into (3.28) and Eq. (4.19) into (4.3), one obtains for the electric quadrupole coupling,

$$E_K - E_{K'} = \sum_{\zeta=-2}^{2} \sum_{mm'} \{ \langle jm'|K\rangle\langle K|jm\rangle - \langle jm'|K'\rangle\langle K'|jm\rangle \} \times$$

$$\times (jm; 2\zeta | jm') V^{(2)}_{-\zeta}(0) \langle j \| Q^{(2)} \| j \rangle , \qquad (4.20)$$

and

$$\left\langle \frac{\hbar}{2\gamma_K} (V_\tau^{EQ}) \right\rangle_T = \left(\frac{15\hbar}{4M_c}\right)^2 |\langle j \| Q^{(2)} \| j \rangle|^2 \lim_{\lambda \to 0} \sum_{\zeta=-2}^{2} \sum_{K' \neq K} \sum_{mm'} \sum_{\tau}^{(n,n)} \times$$

$$\times \sum_{\mu\nu} \sum_{\underline{q}\,\underline{q}'} (jm'; 2\zeta | jm)^2 |\langle jm|K\rangle\langle K'|jm'\rangle|^2 \times$$

$$\times \left(\frac{\rho_i}{a_{oci}^3}\right)^2 |Y^{-(2)}_{-\zeta}(\theta_{oci}, \varphi_{oci})|^2 \eta_{i\mu}^2 \eta_{i\nu}^2 \times$$

$$\times [d_{ci}^2 + \epsilon_{ci}^2 q^2 (\underline{q} \cdot \hat{a}_{oci})^2][d_{ci}^2 + \epsilon_{ci}^2 q'^2 (\underline{q}' \cdot \hat{a}_{oci})^2] \times$$

$$\times \frac{n_T(\underline{q}\mu)[n_T(\underline{q}'\nu) + 1]}{\omega(\underline{q}\mu)\omega(\underline{q}'\nu)[\hbar\omega(\underline{q}\mu) - \hbar\omega(\underline{q}'\nu) - \hbar\omega_{KK'} + i\lambda]} , \qquad (4.21)$$

where

$$n_T(\alpha) = \sum_{n(\alpha)} P(n(\alpha))\, n(\alpha) ,$$

is the thermally averaged phonon occupation number in the crystal and $\hbar\omega_{KK'}$ denotes $E_K - E_{K'}$.

For the summation of $\underline{q}$ in Eq. (4.21), one should sum over all the possible values of $\underline{q}$ in the first Brillouin zone of the reciprocal lattice space. However, for our purpose it is sufficient to take Debye's assumption that the sound propagation velocity $\underline{v}$ is indepen-

dent of the direction of propagation and of the polarization of the wave and the frequency function is taken to be

$$\omega(\underline{q}\mu) = v q . \tag{4.22}$$

Furthermore the total number of nuclei in a crystal is much greater than unity, one can replace the first Brillouin zone by a sphere, i.e., to replace $\sum_{\underline{q}}$ by

$$V\int d^3q = \frac{V}{v^3}\int \omega^2 d\omega \int d\Omega_{\hat{q}} , \tag{4.23}$$

where V is the volume of the crystal. By using the expressions (4.22) and (4.23) and the relation

$$\lim_{\lambda\to 0} \frac{1}{i\lambda + y} = P\left(\frac{1}{y}\right) - i\pi\delta(y) , \tag{4.24}$$

where $P(\frac{1}{y})$ is the principal value of $\frac{1}{y}$, then Eq. (4.21) can be expressed as;

$$\left\langle \frac{\hbar}{2}\Gamma_K(V_\tau^{EQ})\right\rangle_T = \left\langle S_K(V_\tau^{EQ})\right\rangle_T - i\left\langle \frac{\hbar}{2}\Gamma_K(V^{EQ})\right\rangle_T , \tag{4.25}$$

with

$$\left\langle S_K(V_\tau^{EQ})\right\rangle_T = \left(\frac{15\hbar V}{4M_c v^3}\right)^2 |\langle j\|Q^{(2)}\|j\rangle|^2 \sum_{\zeta=-2}^{2}\sum_{K'\neq K}\sum_{mm'} \times$$

$$\times \sum_{i}^{(n,n)}\sum_{\nu\mu}\left(\frac{\rho_i}{a_{oci}^3}\right)^2 \left| Y_{-\zeta}^{(2)}(\theta_{oci},\varphi_{oci})\right|^2 \times$$

$$\times (jm';2\zeta|jm)^2 |\langle jm|\kappa\rangle\langle\kappa'|jm'\rangle|^2 \eta_{i\mu}^2 \eta_{i\nu}^2 \times$$

$$\times \mathcal{P}\int_{\omega_{KK'}}^{\omega_M} d\omega \int_0^{\omega_M-\omega_{KK'}} d\omega' \int_{\Omega(\hat{\xi})} d\Omega_\xi \int_{\Omega(\hat{\xi}')} d\Omega_{\xi'}\, \omega\omega' \times$$

$$\times \left[d_{ci}^2 + \epsilon_{ci}^2 \frac{\omega^2}{v^2} (\hat{\xi}\cdot\hat{a}_{oci})^2\right]\left[d_{ci}^2 + \epsilon_{ci}^2 \frac{\omega'^2}{v^2} (\hat{\xi}'\cdot\hat{a}_{oci})^2\right] \times$$

$$\times \frac{n_T(\omega)[n_T(\omega')+1]}{\hbar\omega - \hbar\omega' - \hbar\omega_{KK'}}, \qquad (4.26)$$

and

$$\left\langle \frac{\hbar}{2}\Gamma_K(V_\tau^{EQ})\right\rangle_T = \pi\left(\frac{15\hbar V}{4M_c v^3}\right)^2 |\langle j\|Q^{(2)}\|j\rangle|^2 \sum_{\zeta=-2}^{2} \sum_{K'\neq K} \sum_{mm'} \times$$

$$\times \sum_{i}^{(n,n)} \sum_{\mu\nu} \left(\frac{\rho_i}{a_{oci}^3}\right)^2 |Y_{-\zeta}^{(2)}(\theta_{oci},\varphi_{oci})|^2 |(jm';2\zeta|jm)|^2 \times$$

$$\times |\langle jm|K\rangle\langle K'|jm'\rangle|^2 \eta_{i\mu}^2 \eta_{i\nu}^2 \int_{\omega_{KK'}}^{\omega_M} d\omega \int_0^{\omega_M-\omega_{KK'}} d\omega' \times$$

$$\times \int_{\Omega(\hat{\xi})} d\Omega_\xi \int_{\Omega(\hat{\xi}')} d\Omega_{\xi'}\, \omega\omega' \left[d_{ci}^2 + \epsilon_{ci}^2 \frac{\omega^2}{v^2} (\hat{\xi}\cdot\hat{a}_{oci})^2\right] \times$$

$$\times \left[d_{ci}^2 + \epsilon_{ci}^2 \frac{\omega'^2}{v^2} (\hat{\xi}'\cdot\hat{a}_{oci})^2\right] n_T(\omega)[n_T(\omega')+1] \times$$

$$\times \delta(\hbar\omega - \hbar\omega' - \hbar\omega_{KK'}). \qquad (4.27)$$

For a Raman process the frequencies of the two phonons ω and ω' satisfy $\omega-\omega' = \omega_{KK'}$ and ω can take all the values inside the frequency

spectrum from $\omega_{KK'}$ to ω_M. Here ω_M is the maximum frequency of the Debye spectrum.

Equation (4.26) describes the nuclear energy level displacement in the intermediate state due to the dynamic part of the nuclear electric quadrupole coupling. The quantity $\Gamma_K(V_\tau^{EQ})$ defined by Eq. (4.27) is actually the inversion of the nuclear spin-lattice relaxation time due to the electric quadrupole coupling.

The electric field gradient tensor appeared in Eqs. (4.26) and (4.27) is produced by the ith lattice point charge in the neighborhood of the radioactive nucleus in the absence of the nuclear quadrupole moment and is referred to as the direct field. The direct field will polarize the radioactive ions and this leads to an additional field at the nucleus oppositely directed to the direct field. This is the so-called shielding effect.[17] Furthermore the charge cloud of the ion can also be polarized by the nuclear quadrupole moment. The direct field can also interact with the quadrupole and higher moments of the charge cloud induced by the nuclear quadrupole moment. In some cases the induced quadrupole moment can be much larger than the nuclear quadrupole moment and therefore reinforces the nuclear quadrupole moment. This is called the antishielding effect.

In addition to the shielding and antishielding effects there still exists another effect called the co-valent effect which is usually much more important than the shielding effect. When the lattice

is deformed by the lattice vibrations some p- or d-like orbitals are mixed into the origional s-like wave function of the ions, then the electrons in the non-spherically symmetric orbitals give rise to an electric field gradient at the nucleus which in turn can interact with the nuclear quadrupole moment. Thus the resultant quadrupole spin-lattice coupling can be much larger than that due to the direct field alone. The dependence of the interaction energy on the displacements of the nuclei from their equilibrium positions will in general be much stronger than the direct field interaction. However, no detail understanding of the contributions of these effects to the spin-lattice coupling is available to us. In order to indicate the orders of magnitude of these effects one may introduce a multiplication parameter ξ such that the effective point charge of the lattice point is taken to be $\rho_i = \xi_i e_i$. Then the true potential becomes a parametic constant times the potential of the direct field.

B. THE MAGNETIC COUPLING

The magnetic coupling Hamiltonian of a lattice system of interacting spins in an external static magnetic field $\underline{H}_0$ can be written as

$$V^M = V^{H_0} + V^{DD}, \tag{4.28}$$

where

$$V^{H_0} = -\hbar H_0 \gamma_c I_z , \tag{4.29}$$

is the magnetic interaction energy between the external static field and the magnetic moment, $\underline{\mu}_c = \gamma_c \hbar \underline{I}_c$, of the radioactive nucleus, and V^{DD} represents the dipole-dipole interaction in the lattice system,

$$V^{DD} = \sum_{j<k} \frac{\hbar^2 \gamma_j \gamma_k}{r_{jk}^3} \left\{ \underline{I}_j \cdot \underline{I}_k - 3 \frac{(\underline{I}_j \cdot \underline{r}_{jk})(\underline{I}_k \cdot \underline{r}_{jk})}{r_{jk}^2} \right\}, \tag{4.30}$$

which is responsible for producing the spin-flip in the spin system. When we focus our attention to one radioactive nucleus, we need only to sum the indices j over the nearest neighbors of the radioactive nucleus. Then the dipole-dipole interaction Hamiltonian can be written as;

$$V^{DD} = \hbar \gamma_c \sum_{j}^{2(m,n)} \frac{b_{cj}}{r_{cj}^3} \left\{ \underline{I}_c \cdot \underline{I}_j - 3 \frac{(\underline{I}_c \cdot \underline{r}_{cj})(\underline{I}_j \cdot \underline{r}_{cj})}{r_{cj}^2} \right\}, \tag{4.31}$$

where γ_c is the gyromagnetic ratio of the radioactive nucleus and b_{cj} stands for γ_i/γ_c.

The vector $\underline{r}_{cj}$, appeared in Eq. (4.31), is the relative position vector between the radioactive nucleus and the jth nearest lattice point as shown in Fig. 5. By taking $(r_{cj}, \theta_{cj}, \phi_{cj})$ as the polar coordinates of

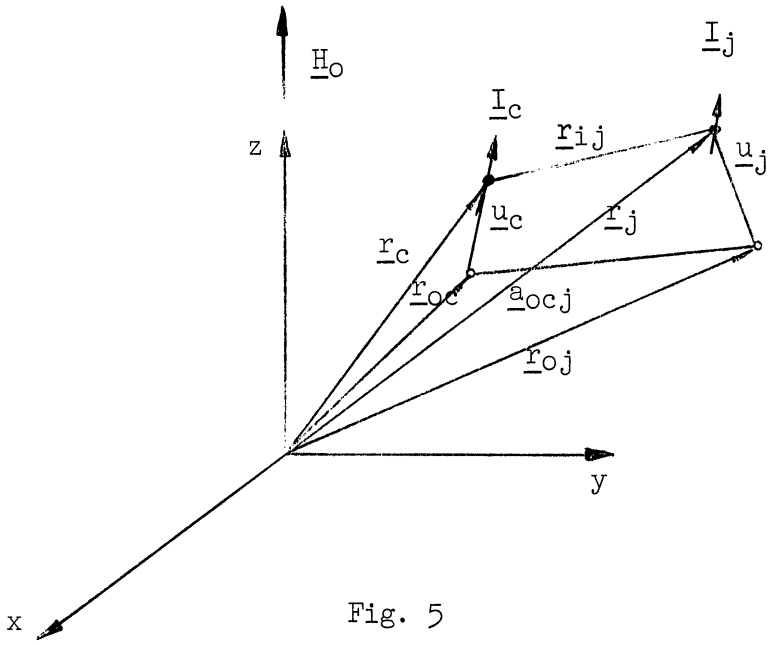


Fig. 5

the vector $\underline{r}_{cj}$, the dipole-dipole coupling Hamiltonian can be expressed as;

$$V^{DD} = \hbar^2 \gamma_c^2 \sum_j^{(n,n)} \frac{b_{cj}}{r_{cj}^3} \left\{ A_{cj} + B_{cj} + C_{cj} + D_{cj} + E_{cj} + F_{cj} + G_{cj} + H_{cj} \right\}, \quad (4.32)$$

where;

$$\left.\begin{aligned}
A_{cj} &= (1 - 3\cos^2\theta_{cj}) I_z^c I_z^j, \\
B_{cj} &= -\tfrac{1}{4}(1 - 3\cos^2\theta_{cj})(I_+^c I_-^j + I_-^c I_+^j), \\
C_{cj} &= -\tfrac{3}{2}\sin\theta_{cj}\cos\theta_{cj} e^{-i\varphi_{cj}} I_z^c I_+^j, \\
D_{cj} &= -\tfrac{3}{2}\sin\theta_{cj}\cos\theta_{cj} e^{-i\varphi_{cj}} I_+^c I_z^j, \\
E_{cj} &= -\tfrac{3}{2}\sin\theta_{cj}\cos\theta_{cj} e^{i\varphi_{cj}} I_z^c I_-^j,
\end{aligned}\right\} \quad (4.33)$$

$$\left.\begin{aligned} F_{cj} &= -\frac{3}{2}\sin\theta_{cj}\cos\theta_{cj}\,e^{i\varphi_{cj}} I_-^c I_z^j, \\ G_{cj} &= -\frac{3}{4}\sin^2\theta_{cj}\,e^{-2i\varphi_{cj}} I_+^c I_+^j, \\ H_{cj} &= -\frac{3}{4}\sin^2\theta_{cj}\,e^{2i\varphi_{cj}} I_-^c I_-^j. \end{aligned}\right\} \qquad (4.33)$$

One sees that the magnetic coupling between two spins depends on the orientations as well as the magnitudes of their magnetic moments and also on the length and orientation of the relative vector between them. The effects of this coupling depend strongly on whether this relative position vector is fixed in space or changes rapidly because of the relative motion between the nuclei due to, for example, the thermal vibrations. The effect of these thermal vibrations is to produce a time-dependent local magnetic field at the site of the radioactive nucleus. As shown previously one can expand the field gradient tensor at the equilibrium position as;

$$V^{DD} = V(0)^{DD} + \frac{1}{2!}\sum_j \sum_{\mu\nu} \left(\frac{\partial^2 V(r_{cj}, \theta_{cj}, \varphi_{cj})^{DD}}{\partial u_\mu(j)\,\partial u_\nu(j)}\right)_0 u_\mu(j) u_\nu(j) + \cdots, \qquad (4.34)$$

As shown in Fig. 5, the vectors $\underline{u}_c$ and $\underline{u}_j$ are the displacements from the equilibrium positions of the radioactive nucleus and the jth nearest neighboring nucleus respectively. Following the same procedures as in the case of electric quadrupole coupling, the magnetic

coupling Hamiltonian V^M can be written as

$$V^M = V_0^M(0) + V_\tau^{DD}, \tag{4.35}$$

with

$$V_0^M(0) = -\hbar H_0 \gamma_c I_z + \hbar^2 \gamma_c^2 \sum_{j'}^{(n,n)} \frac{b_{cj'}}{a_{ocj'}^3} f(\theta_{ocj'}, \phi_{ocj'}, I^c I^{j'}), \tag{4.36}$$

and

$$V_\tau^{DD} = \frac{15\hbar^3 \gamma_c^2}{4M_c} \sum_{j'}^{(n,n)} \sum_{\mu\nu} \sum_{\underline{\beta}\underline{\beta}'} \left(\frac{b_{cj'}}{a_{ocj'}^3}\right) f(\theta_{ocj'}, \phi_{ocj'}, I^c I^{j'}) \times$$

$$\times \eta_{j\mu}\eta_{j\nu}\left[\frac{1}{\omega(\underline{\beta}\mu)\omega(\underline{\beta}'\nu)}\right]^{\frac{1}{2}} \left[d_{cj} - i\epsilon_{cj}\beta(\underline{\beta}\cdot\hat{a}_{ocj})\right] \times$$

$$\times \left[d_{cj} + i\epsilon_{cj}\beta'(\underline{\beta}'\cdot\hat{a}_{ocj})\right] a(\underline{\beta}\mu) a^\dagger(\underline{\beta}'\nu) e^{-i(\underline{\beta}-\underline{\beta}')\cdot\underline{r}_{oc}}. \tag{4.37}$$

In Eqs. (4.36) and (4.37) we have defined, $f(\theta_{ocj}, \phi_{ocj}, I^C, I^j) = A_{cj} + B_{cj} + C_{cj} + D_{cj} + E_{cj} + F_{cj} + G_{cj} + H_{cj}$.

In Eq. (4.35) $V_O^M(0)$ is the static magnetic interaction Hamiltonian due to the external static field and the static dipole-dipole couplings in the lattice system and V_τ^{DD} is the dynamic part of the dipole-dipole interaction induced by the thermal vibrations of the lattice points.

The quantities A_{cj}, B_{cj}, C_{cj}, D_{cj}, E_{cj}, F_{cj}, G_{cj} and H_{cj} as defined in Eq. (4.33) are the operators which acting on the spin states $|I_c m_c\rangle$ and $|I_j m_j\rangle$ will produce the following spin-flips;

$$
\begin{aligned}
&A_{cj}: \ \Delta m_c = 0, \quad \Delta m_j = 0, \quad \Delta(m_c + m_j) = 0, \\
&B_{cj}: \ \Delta m_c = \pm 1, \quad \Delta m_j = \mp 1, \quad \Delta(m_c + m_j) = 0, \\
&C_{cj}: \ \Delta m_c = 0, \quad \Delta m_j = 1, \quad \Delta(m_c + m_j) = 1, \\
&D_{cj}: \ \Delta m_c = 1, \quad \Delta m_j = 0, \quad \Delta(m_c + m_j) = 1, \\
&E_{cj}: \ \Delta m_c = 0, \quad \Delta m_j = -1, \quad \Delta(m_c + m_j) = -1, \\
&F_{cj}: \ \Delta m_c = -1, \quad \Delta m_j = 0, \quad \Delta(m_c + m_j) = -1, \\
&G_{cj}: \ \Delta m_c = 1, \quad \Delta m_j = 1, \quad \Delta(m_c + m_j) = 2 \\
&H_{cj}: \ \Delta m_c = -1, \quad \Delta m_j = -1, \quad \Delta(m_c + m_j) = -2.
\end{aligned}
\tag{4.38}
$$

One sees that the operator A_{cj} causes no change in the spin states and describes the effect of the static local field and B_{cj} causes a simultaneous spin flips of two neighboring spins in opposite directions. Therefore the operators A_{cj} and B_{cj} cause no change in the interaction energy and can be referred to as the adiabatic operators of the interacting spin system. The operators C_{cj}, D_{cj}, E_{cj}, F_{cj}, G_{cj} and H_{cj} will cause the change in energy state of the coupling spins and allow to transfer energy from or to the spin system.

Substituting Eq. (4.37) into Eq. (4.3) one obtains

$$\left\langle \frac{\hbar}{2}\gamma_K(V_c^{DD})\right\rangle_T = \left\langle S_K(V_c^{DD})\right\rangle_T - i\left\langle \frac{\hbar}{2}\Gamma_K(V_c^{DD})\right\rangle_T , \tag{4.39}$$

with

$$\begin{aligned}\left\langle S_K(V_c^{DD})\right\rangle_T = &\left(\frac{15\hbar^3\gamma_c^2 V}{4M_c v^3}\right)^2 \sum_{K'\neq K}\sum_{\substack{m m'\\ m_j m_j'}}\sum_{j}^{(n,n)}\sum_{\mu\nu}\left(\frac{b_{cj}}{a_{ocj}^3}\right)^2 \times\\ &\times |f_{m'm_j';mm_j}|^2 \eta_{j\mu}^2\eta_{j\nu}^2 |\langle jm, j_j m_j|K\rangle|^2 \times\\ &\times |\langle K'|jm_j' j_j m_j'\rangle|^2 \mathcal{P}\int_{\omega_{KK'}}^{\omega_M} d\omega \int_0^{\omega_M-\omega_{KK'}} d\omega' \times\\ &\times \int_{\Omega(\hat{\xi})} d\Omega_{\xi} \int_{\Omega(\hat{\xi})} d\Omega_{\xi'}\, \omega\omega' \left[d_{cj}^2 + \epsilon_{cj}^2 \frac{\omega^2}{v^2}(\hat{\xi}\cdot\hat{a}_{ocj})^2\right] \times\\ &\times \left[d_{cj}^2 + \epsilon_{cj}^2 \frac{\omega'^2}{v^2}(\hat{\xi}'\cdot\hat{a}_{ocj})^2\right] n_T(\omega)\left[n_T(\omega')+1\right] \times\\ &\times \left[\hbar\omega - \hbar\omega' - \hbar\omega_{KK'}\right]^{-1} . \end{aligned} \tag{4.40}$$

and

$$\begin{aligned}\left\langle \frac{\hbar}{2}\Gamma_K(V_c^{DD})\right\rangle_T = &\left(\frac{15\hbar^3\gamma_c^2 V}{4M_c v^3}\right)^2 \sum_{K'\neq K}\sum_{\substack{m m'\\ m_j m_j'}}\sum_{j}^{(n,n)}\sum_{\mu\nu}\left(\frac{b_{cj}}{a_{ocj}^3}\right)^2 |f_{m'm_j';mm_j}|^2 \times\\ &\times \eta_{j\mu}^2\eta_{j\nu}^2 |\langle jm, j_j m_j|K\rangle\langle K'|jm_j' j_j m_j'\rangle|^2 \int_{\omega_{KK'}}^{\omega_M} d\omega \times\\ &\times \int_0^{\omega_M-\omega_{KK'}} d\omega' \int_{\Omega(\hat{\xi})} d\Omega_{\xi} \int_{\Omega(\hat{\xi}')} d\Omega_{\xi'}\, \omega\omega' \left[d_{cj}^2 + \epsilon_{cj}^2 \frac{\omega^2}{v^2}(\hat{\xi}\cdot\hat{a}_{ocj})^2\right] \times\\ &\times \left[d_{cj}^2 + \epsilon_{cj}^2 \frac{\omega'^2}{v^2}(\hat{\xi}'\cdot\hat{a}_{ocj})^2\right] \delta(\hbar\omega - \hbar\omega' - \hbar\omega_{KK'}) , \end{aligned} \tag{4.41}$$

where we have defined

$$f_{m'm_j';mm_j} = \langle Im' | \langle I_j m_j' | (B_j + D_j + F_j + G_j + H_j) | Im \rangle | I_j m_j \rangle, \quad (4.42)$$

and $|Im\rangle$ and $|I_j m_j\rangle$ are respectively the eigenkets of the spin operator I of the radioactive nucleus in the intermediate state and of the spin operator I_j of the jth neighboring nucleus.

Equation (4.40) and Eq. (4.41) describe respectively the nuclear energy level displacement and the spin-lattice relaxation effect due to the dynamic part of the magnetic dipole-dipole couplings. It has been shown experimentally that[18] this relaxation effect is inadequate to account for the nuclear relaxation times actually observed in most crystals and no single example is known where the observed relaxation could be assigned to this mechanism with certainty. On the other hand, the electric coupling of the lattice vibrations with the nuclear quadrupole moments is much more important and is known to be responsible for nuclear relaxation in many crystals.

In a strong external magnetic field the dipole-dipole couplings in the lattice system will be decoupled and the interaction Hamiltonian V^M of a radioactive nucleus is given by Eq. (4.29). If the external magnetic field has axial symmetry, one can choose the symmetric axis as the quantization axis oz and the projection of I_z of the nuclear spin on this axis commutes with V^M and they can be simultaneously diagonalized by the representation $\{|jm\rangle\}$. Then the perturbed cor-

relation functions of Eqs. (3.26) and (3.27) can be reduced to

$$W(\hat{k}_2/\hat{k}_1) = \sum_{\nu_1\nu_2} \sum_{mm'} I_{\nu_1}(j_i j j) II_{\nu_2}(j_f j j) III_{\nu_1\nu_2}(mm') \times \tilde{Y}^*_{\nu_1 m'-m}(\theta_1\varphi_1)\, \tilde{Y}_{\nu_2 m'-m}(\theta_2\varphi_2), \tag{4.43}$$

and

$$III_{\nu_1\nu_2}(mm') = \frac{(jm; j-m'|\nu_1 m-m')(jm; j-m|\nu_2 m-m')}{1 - ig^*\mu_N H_0 \tau_N (m-m')/\hbar}, \tag{4.44}$$

where g* is the g-factor of the radioactive nucleus in the intermediate state, μ_N is the nuclear magneton and H_o is thestrength of the external magnetic field.

Equations (4.43) and (4.44) are the same as Alder's formulas (Ref. (7) Eqs. (9) and (10)), except in the present formulism the angular resolution corrections for the detectors have been explicitly established in the correlation function. It has been pointed out by Abragam[7] that Alder's formulas are only valid if there exists an axis oz such that the projection I_z of the nuclear spin on this axis is a good quantum number. The present results, Eqs. (4.43) and (4.44), exactly meet this condition.

If the first detector is placed along the symmetric magnetic field, one has $\theta_1 = \phi_1 = 0$ and

$$Y_{\nu_1 m'-m}(\theta_1\varphi_1) = \left(\frac{2\nu_1+1}{4\pi}\right)^{\frac{1}{2}} \delta_{m',m},$$

then by using the orthonormal properties of the Clebsch-Gordan coef-

ficients Eq. (4.43) can be reduced to the unperturbed angular correlation function of Eq. (3.31). This is a well-known result, that the correlation is unperturbed by such a coupling if either one of the two radiations is emitted along the axially symmetric field.

C. THE RADIATION INTERACTION

For our purpose it is necessary to assume that the phenomenon of photon transport in the system does not exist. Therefore the physical system is taken to be optically thin so that the emitted photons escape the system without interacting with the particles in the system.

The effect due to the radiation interaction is described by Eq. (4.4). Where the summation over β_m is to be regarded as a summation over all possible sets of photon occupation numbers and over all states of the nuclei. According to the states defined in Section I this sum can be decomposed into the contributions arising from those states like the initial state $|\beta_i\rangle$ and those states like the final state $|\beta_f\rangle$. The contributions from the initial states correspond to the re-absorptions of the emitted photons by the radioactive nuclei. Since the system is assumed to be optically thin these contributions can be disregarded.

The contributions from the states like the final states can be calculated, by substituting Eq. (3.4) into Eq. (4.4), as

$$\frac{\hbar}{2}\gamma_K(V^\gamma) = S_K(V^\gamma) - i\frac{\hbar}{2}\Gamma_K(V^\gamma), \tag{4.45}$$

with

$$S_K(V^\gamma) = \sqrt{2}\left(\frac{Mc}{\hbar^2}\right)^{\frac{3}{2}} L^3 \sum_{j_2} \sum_{K' \neq K} \sum_{m m'} |\langle j'm'|K'\rangle\langle K|j'm\rangle|^2 \times$$

$$\times P\int \sqrt{E^N_{k_2}}\, dE^N_{k_2}\, d\Omega(\hat{k}_2)\, |\langle j'm'|V(\gamma)|j'm\rangle|^2 \times$$

$$\times \left[E^N_{k_1} + E^N_{k_2} + E_{K_i} - E_{K'} - E_{\gamma_1} - E_{\gamma_2}\right]^{-1}, \tag{4.46}$$

and

$$\frac{\hbar}{2}\Gamma_K(V^\gamma) = \pi\sqrt{2}\left(\frac{Mc}{\hbar^2}\right)^{\frac{3}{2}} L^3 \sum_{j_2} \sum_{m_f} \sum_{m m'} \sqrt{E^N_{K_i} - E^N_K - E_{\gamma_1} - E_{\gamma_2} + E^N_{k_1}} \times$$

$$\times \langle j'm|K\rangle\langle K|j'm'\rangle \int d\Omega(\hat{k}_2) \langle j_f m_f|V(\gamma)|j'm\rangle \times$$

$$\times \langle j_f m_f|V(\gamma)|j'm'\rangle^*, \tag{4.47}$$

where $V(\gamma)$ is the radiation interaction Hamiltonian for the emission of the photons, L is the quantization cell length of the photon in a coarse-grained configuration space,[12] E^N_k is the recoil energy of the radioactive nucleus due to the emission of a photon of wave vector $\underline{k}$, E_{γ_1}, and E_{γ_2} are respectively the first and second photon energy and E^N_{Ki} and E^N_{Kf} are respectively the total internal energy of the radioactive nucleus in the initial state and in the final state.

Equation (4.46) describes the energy level displacement caused by the radiation interaction of the photons. As we have mentioned in the previous section for our purpose we will only note the existance of $S_K(V^\gamma)$ and will not be concerned with its effects. The function $\Gamma_K(V^\gamma)$ defined by Eq. (4.47) is actually the inverse of the life-time of the radioactive nucleus in the intermediate state. By using Eq. (3.19), Eq. (4.47) can be calculated as

$$\frac{\hbar}{2}\Gamma_K(V^\gamma)=\left(\frac{\sqrt{2}\pi M_c L^2}{\hbar^2}\right)\sqrt{(2j+1)E_{K_1}^N}\sum_{L_2L_2'}(-1)^{j'+L_2'-j_f'}\times$$

$$\times\langle j_f'\|T_{L_2}\|j'\rangle\langle j_f'\|T_{L_2'}\|j'\rangle^* C_{00}(L_2L_2')\times$$

$$\times W(jj'L_2L_2';0j_f'). \tag{4.48}$$

One sees that the function $\Gamma_K(V^\gamma)$ is independent of the substates of the intermediate and the final states but depends on the angular momenta j and j_f and also on the types of transitions.

In our calculation we will let $\Gamma_K(V^\gamma) = (\tau_N)^{-1}$ and τ_N is taken to be the experimentally determined life-time of the radioactive nucleus in the intermediate state. Then Eq. (4.45) reduces to

$$\frac{\hbar}{2}\gamma_K(V^\gamma)=-i\frac{\hbar}{2\tau_N}. \tag{4.49}$$

Combining Eqs. (4.25), (4.46) and (4.2) one has

$$\left\langle\frac{\hbar}{2}\gamma_K(-iE_{\beta_i})\right\rangle_T=\left\langle S_K(V_c^{NC})\right\rangle_T-i\frac{\hbar}{2}\left[\left\langle\Gamma_K(V_c^{NC})\right\rangle_T+\tau_N^{-1}\right], \tag{4.50}$$

and the attenuation coefficient of Eq. (3.27) can be expressed as

$$\amalg_{\nu_1\nu_2}(mm'm''m''') = \sum_{KK'} \langle jm|K\rangle\langle K|jm'\rangle\langle jm'''|K'\rangle \times$$

$$\times \langle K'|jm''\rangle (jm', j-m'''|\nu_1, m'-m''') \times$$

$$\times (jm, j-m'''|\nu_2, m-m''') \times$$

$$\times \left\{ \frac{\hbar}{2}\left[\langle \Gamma_K(V_\tau^{NC})\rangle_T + \langle \Gamma_{K'}(V_\tau^{NC})\rangle_T + 2\tau_N^{-1} \right] - \right.$$

$$\left. - i\left[E_K - E_{K'} + \langle S_K(V_\tau^{NC})\rangle_T - \langle S_{K'}(V_\tau^{NC})\rangle_T \right] \right\}^{-1} . \qquad (4.51)$$

For electric quadrupole coupling the quantity $E_K - E_{K'}$ is given by Eq. (4.20). In Eq. (4.51) the energy level displacement $\langle S_K(V_T^{NC})\rangle_T$ is given by Eq. (4.26) for the electric quadrupole coupling and by Eq. (4.40) for the magnetic dipole-dipole coupling and the function $\langle \frac{\hbar}{2}\Gamma_K(V_T^{NC})\rangle_T$ is given by Eq. (4.27) for the electric quadrupole coupling and by Eq. (4.41) for the magnetic dipole-dipole coupling.

From the attenuation coefficient, given by Eq. (4.51), one sees clearly that in the present formulism the effects on the angular correlations due to the changes of the states of the environments have been explicitely and systematically built in the perturbed angular correlation function.

V. THE ROTATIONAL DEPENDENCE OF THE γ-γ ANGULAR CORRELATION

It has been proved experimentally by the Zürich group[8),(19),(20)] that the anisotropy of the γ-γ angular correlation is a function of the orientation of the symmetry axis of the crystalline field. Recently Paul and Brumner[21], Alder and Steffen[22] have calculated the anisotropy function based on the Abragram's formulism for the case of axially symmetric fields.

In this section we will investigate, by introducing a crystal coordinate system and a principal coordinate system of the crystalline field, the perturbing effect on the rotational pattern of the angular correlation for the case of asymmetric crystalline field based on the present formulism.

As shown in the Fig. 6, we let xyz be an arbitrarily chosen laboratory coordinate system, XYZ be a fixed coordinate system in the crystal and X*Y*Z* be the principal coordinates of the crystalline field. The Euler angles between XYZ and X*Y*Z* coordinate systems are taken to be $(\alpha\beta\gamma)$ which can be determined by EPR or NMR techniques. The Euler angles between the crystal coordinates XYZ and the laboratory coordinates xyz can be taken to the $(\Phi,(H),0)$. Here the Euler angles $(\alpha\beta\gamma)$ are defined in a right-handed coordinate system as;

$$xyz \xrightarrow{R(\alpha_z)} x'y'z' \xrightarrow{R(\beta_{y'})} x''y''z'' \xrightarrow{R(\gamma_{z''})} x'''y'''z'''.$$

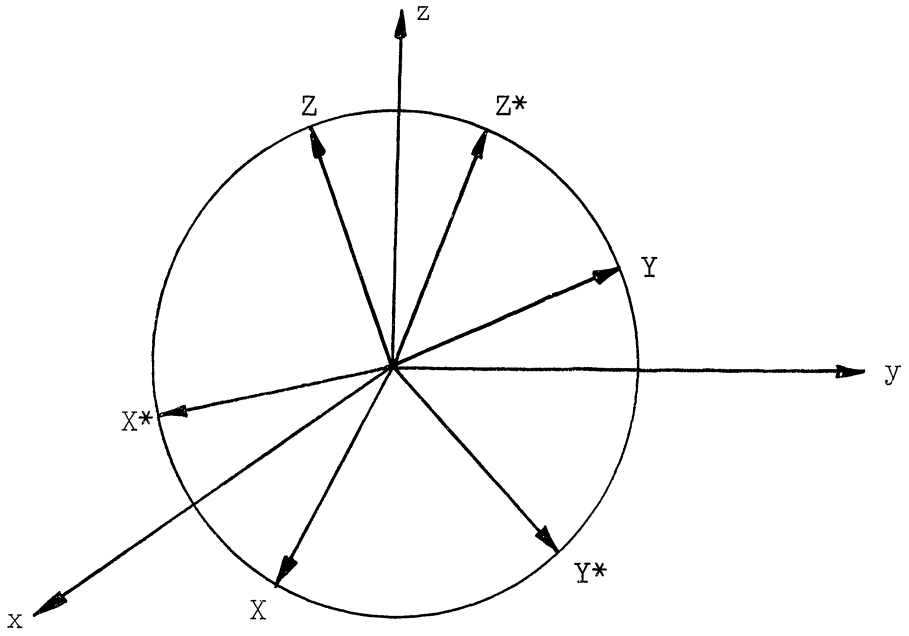


Fig. 6

Then the rotational relations between these three sets of coordinates can be expressed as;

$$\sum^{*}(X^{*}Y^{*}Z^{*}) \xrightarrow{R(\alpha\beta\gamma)} \sum^{(II)}(XYZ) \xrightarrow{R(\Phi\textcircled{H}0)} \sum^{(I)}(xyz).$$

Since an irreducible tensor operator of rank L will transform as $D^{(L)}$ rotational group of (2L + 1) dimensional representations under the rotation of the coordinate system, then an irreducible field gradient tensor of rank two ${}^{(II)}V^{(2)}$ in $\Sigma^{(II)}$ will transform as $D^{(2)}(\Phi, \textcircled{H}, 0)$ rotational group into the laboratory coordinate system $\Sigma^{(I)}$ under the rotation of $\Sigma^{(II)}$ and can be expressed as;

$${}^{(I)}V^{(2)}_{-\varsigma} = \sum_{\rho=-2}^{2} D^{(2)}_{\rho-\varsigma}(\Phi,\textcircled{H},0)\, {}^{(II)}V^{(2)}_{\rho} . \qquad (5.1)$$

Thus an irreducible tensor of rank two ${}^{*}V^{(2)}$ in the principal coordinate system Σ^{*} can be transformed into the laboratory coordinate

system as follows

$$ {}^{(I)}V_{-\zeta}^{(2)} = \sum_{\rho,r} D_{r\rho}^{(2)}(\alpha\beta\gamma)\, D_{\rho-\zeta}^{(2)}(\Phi\Theta O)\, {}^{*}V_{r}^{(2)}, \qquad (5.2) $$

where ${}^{(I)}V_{-\zeta}^{(2)}$ is the field gradient tensor in the $\Sigma^{(I)}$.

In a rectangular cartesion coordinates the field gradient tensors can be defined in their principal axes by two parameters, namely the field anisotropy η and the Z*—component of the field gradient. Furthermore the rotational transformation is an unitary transformation, then the transformation of Eq. (5.2) can generally be expressed as (Appendix V);

$$ {}^{(I)}V_{-\zeta}^{(2)} = {}^{*}V_{0}^{(2)}\left[F_{-\zeta}(\alpha\beta\gamma;\Phi\Theta O) + \eta G_{-\zeta}(\alpha\beta\gamma;\Phi\Theta O)\right], \qquad (5.3) $$

As shown in the Appendix V, the functions $F_{-\zeta}$ and $G_{-\zeta}$ can be calculated as a function of the Euler angles between these three sets of coordinate systems. With this field gradient tensor operators, the electric quadrupole interaction Hamiltonian can be expressed as

$$ V^{EQ} = \sum_{\zeta}(-1)^{\zeta}\left[F_{-\zeta}(\alpha\beta\gamma;\Phi\Theta O) + \eta G_{-\zeta}(\alpha\beta\gamma;\Phi\Theta O)\right] {}^{*}V_{0}^{(2)} Q_{\zeta}^{(2)}, \qquad (5.4) $$

where ${}^{*}V_{0}^{(2)}$ is an irreducible component of the field gradient tensor in its principal axes and can be expressed as

$$ {}^{*}V^{(2)} = \sum_{i}^{(n,n)} \left(\frac{\rho_i}{a_{ci}^{*3}}\right) Y_{0}^{(2)}(\theta_{ci}^{*}, \varphi_{ci}^{*}), \qquad (5.5) $$

the coordinates $(a_{ci}^{*}, \theta_{ci}^{*}, \phi_{ci})$ are the spherical coordinates of the ith nearest neighbor of the radioactive nucleus in the Σ^{*}.

If we assume that the thermal vibrations of the lattice points do not destroy the crystal axes but the principal axes of the field gradient will lose its identification due to the displacements of the lattice points from their equilibrium positions, then the electric quadrupole coupling Hamiltonian can be expressed as;

$$V^{EQ}(R_1 R_2) = V_o^{EQ}(R_1 R_2) + V_\tau^{EQ}(R_2), \tag{5.6}$$

with

$$V_o^{EQ}(R_1 R_2) = \sum_\zeta (-1)^\zeta \left[F_{-\zeta}(\alpha\beta\gamma; \Phi\Theta 0) + \eta G_{-\zeta}(\alpha\beta\gamma; \Phi\Theta 0) \right] V_o^{*(2)}(0) Q_\zeta^{(2)}, \tag{5.7}$$

and

$$V_\tau^{EQ}(R_2) = \left(\frac{15\hbar}{4M_c}\right) \sum_i^{(n,n)} \sum_\zeta \sum_\rho \sum_{\mu\nu} \sum_{\underline{\ell}\underline{\ell}'} (-i)^\zeta D^{(2)}_{\rho,-\zeta}(\Phi\Theta 0) \times$$

$$\times \left(\frac{\rho_i}{a_{oci}^3}\right) \eta_{i\mu} \eta_{i\nu} \left[\frac{1}{\omega(\underline{\ell}\mu)\,\omega(\underline{\ell}'\nu)}\right]^{\frac{1}{2}} \times$$

$$\times \left[d_{ci} - i\epsilon_{\beta\gamma}(\hat{\underline{\ell}}\cdot\hat{\underline{a}}_{oci})\right]\left[d_{ci} + i\epsilon_{ci}\beta\gamma(\hat{\underline{\ell}}'\cdot\hat{\underline{a}}_{oci})\right] \times$$

$$\times Y^{(2)}_{-\rho}(\theta_{oci}, \phi_{oci}) Q_\rho^{(2)} a(\underline{\ell}\mu) a^\dagger(\underline{\ell}'\nu) e^{i(\underline{\ell}-\underline{\ell}')\cdot\underline{r}_{oc}}, \tag{5.8}$$

where R_1 and R_2 represent the rotations of $R(\alpha\beta\gamma)$ and $R(\Phi\Theta 0)$ respectively. Equation (5.7) and Eq. (5.8) describe respectively the static part and the dynamic part of the electric quadrupole coupling under the rotations of the crystal coordinate system. The coordinates (a_{oci} θ_{oci} ϕ_{oci}) in Eq. (5.8) are the spherical coordinates of the relative

distance at equilibrium position between the radioactive nucleus and the ith lattice point in the crystal coordinate system. Since the thermal vibrations of the lattice points will destroy the identification of the principal axis, the dynamic part of the coupling Hamiltonian depends, as shown in Eq. (5.8), only on the rotations of the crystal axes and the field gradients are expressed in terms of the coordinates of the lattice points in the crystal coordinate system. With this coupling Hamiltonian the function $\langle \frac{\hbar}{2} \gamma_K(V_T^{NC}) \rangle_T$ as given by Eq. (4.3) can be calculated as;

$$\left\langle \frac{\hbar}{2}\gamma_K(\bar{V}_\tau^{EQ}(R_2))\right\rangle_T = \left\langle S_K(\bar{V}_\tau^{EQ}(R_2))\right\rangle_T - i\left\langle \frac{\hbar}{2}\Gamma_K(\bar{V}_\tau^{EQ}(R_2))\right\rangle_T , \tag{5.9}$$

with

$$\begin{aligned}\left\langle S_K(\bar{V}_\tau^{EQ}(R_2))\right\rangle_T = &\left(\frac{15\hbar\bar{V}}{4M_c v^3}\right)^2 |\langle j\|Q^{(2)}\|j'\rangle|^2 \sum_{\zeta=-2}^{2}\sum_{k'\neq K}^{1} \times \\ &\times \sum_{mm'}\sum_{i}^{(n,n)}\sum_{\mu\nu}\left(\frac{\rho_i}{a_{oci}^3}\right)^2 |F(\zeta, R_2, \theta_{oci}\varphi_{oci})|^2 \\ &\times (jm', 2\zeta | jm)^2 |\langle jm|K\rangle\langle K'|jm'\rangle|^2 \\ &\times \eta_{i\mu}^2\eta_{i\nu}^2 P\int_{\omega_{KK'}}^{\omega_M} d\omega \int_0^{\omega_M-\omega_{KK'}} d\omega' \int_{\Omega(\hat{\delta})} d\Omega \int_{\Omega(\hat{\delta}')} d\Omega \times \\ &\times \left[d_{ci}^2 + \epsilon_{ci}^2\frac{\omega^2}{v^2}(\hat{\delta}\cdot\hat{a}_{oci})^2\right]\left[d_{ci}^2 + \epsilon_{ci}^2\frac{\omega'^2}{v^2}(\hat{\delta}'\cdot\hat{a}_{oci})^2\right] \times \\ &\times n_T(\omega)[n_T(\omega')+1][\hbar\omega - \hbar\omega' - \hbar\omega_{KK'}]^{-1},\end{aligned} \tag{5.10}$$

and

$$\left\langle \frac{dP}{dt}\Big|_K (V_T^{EQ}(R_2)) \right\rangle_T = \pi \left(\frac{15\hbar V}{4M_c v^3}\right)^2 |\langle j\|Q^{(2)}\|j\rangle|^2 \sum_{\zeta=-2}^{2} \sum_{K\neq K} \times$$

$$\times \sum_{m m'} \sum_{i}^{(n,n)} \sum_{\mu\nu} \left(\frac{\rho_i}{a_{oci}^3}\right)^2 |F(\zeta, R_2 \theta_{oci} \varphi_{oci})|^2 \times$$

$$\times (jm'; 2\zeta | jm)^2 \langle jm|K\rangle\langle K|jm'\rangle|^2 \eta_{i\mu}^2 \eta_{i\nu}^2 \times$$

$$\times \int_{\omega_{KK'}}^{\omega_M} d\omega \int_0^{\omega_M - \omega_{KK'}} d\omega' \int_{\Omega(\hat{\xi})} d\Omega \int_{\Omega(\hat{\xi}')} d\Omega \, \omega\omega' \times$$

$$\times \left[d_{ci}^2 + \epsilon_{ci}^2 \frac{\omega^2}{v^2} (\hat{\xi}\cdot\hat{a}_{oci})^2\right]\left[d_{ci}^2 + \epsilon_{ci}^2 \frac{\omega'^2}{v^2} (\hat{\xi}'\cdot\hat{a}_{oci})\right] \times$$

$$\times n_T(\omega)[n_T(\omega')+1]\,\delta(\hbar\omega - \hbar\omega' - \hbar\omega_{KK'}), \qquad (5.11)$$

where

$$F(\zeta, R_2 \theta_{oci} \varphi_{oci}) = \sum_{\zeta} D^{(2)}_{\zeta,-\zeta}(\Phi,\Theta,0)\, Y^{(2)}_{-\zeta}(\theta_{oci}\varphi_{oci}). \qquad (5.12)$$

The quantity $E_K - E_{K'}$ in the denominator of Eq. (4.51) can now be calculated as;

$$E_K(R_1R_2) - E_{K'}(R_1R_2) \equiv E_{KK'}(R_1R_2) = \sum_{\zeta=-2}^{2} \sum_{mm'} {}^{*}V_0^{(2)}(0) \times$$

$$\times \langle j\|Q^{(2)}\|j\rangle [\langle jm|K\rangle\langle K|jm\rangle - \langle jm|K\rangle\langle K|jm\rangle] \times$$

$$\times (jm'; 2\zeta; jm)[F_{-\zeta}(\alpha\beta\gamma, \Phi,\Theta,0) + \eta G_{-\zeta}(\alpha\beta\gamma, \Phi,\Theta,0)]. \qquad (5.13)$$

The irreducible component of the field gradient tensor ${}^*V_0^{(2)}(0)$ appeared in Eqs. (5.7) and (5.13) is evaluated at the equilibrium positions of the lattice system in the principal axes.

From Eqs. (5.10), (5.11) and (5.13) one clearly sees that the attenuation coefficient $III_{\nu_1\nu_2}(mm'm''m''')$, as given by Eq. (4.51), is a function of the strength of interaction as well as a function of the rotations of the crystal axis and the orientations of the principal axes of the field gradients within the crystal.

VI. DETERMINATION OF THE NUCLEAR ELECTRIC QUADRUPOLE MOMENT IN EXCITED STATE

One of the immediate applications of the present theory is to investigate the nuclear quadrupole coupling by means of the rotational pattern of the γ-γ angular correlation. The nuclear electric quadrupole moment will interact with the non-spherical electric field. Even a nucleus, which occupies a site of cubic symmetry, will have quadrupole coupling with a fluctuating field gradients produced by the thermal vibrations of the neighboring nuclei. The quadrupole coupling also gives raise to an important nuclear spin-lattice relaxation and this relaxation is much more effective than that via the magnetic dipole-dipole coupling even at the room temperature.[18] As we have mentioned previously that for a cascade decay with a intermediate state of mean life $\tau_N \gtrsim 10^{-12}$sec the perturbing effects on the angular correlations still can be measured. This provides us a powerful tool to investigate the nuclear quadrupole moment in an excited states of considerably short life times. This cannot be done with the usual microwave techniques.

In an experimental arrangement for the measurement of the rotational pattern of the angular correlation, as shown in Fig. 7, we choose the direction of the first radiation as our z-axis. By placing the detector D_2, which detects the second cascade radiation, along the

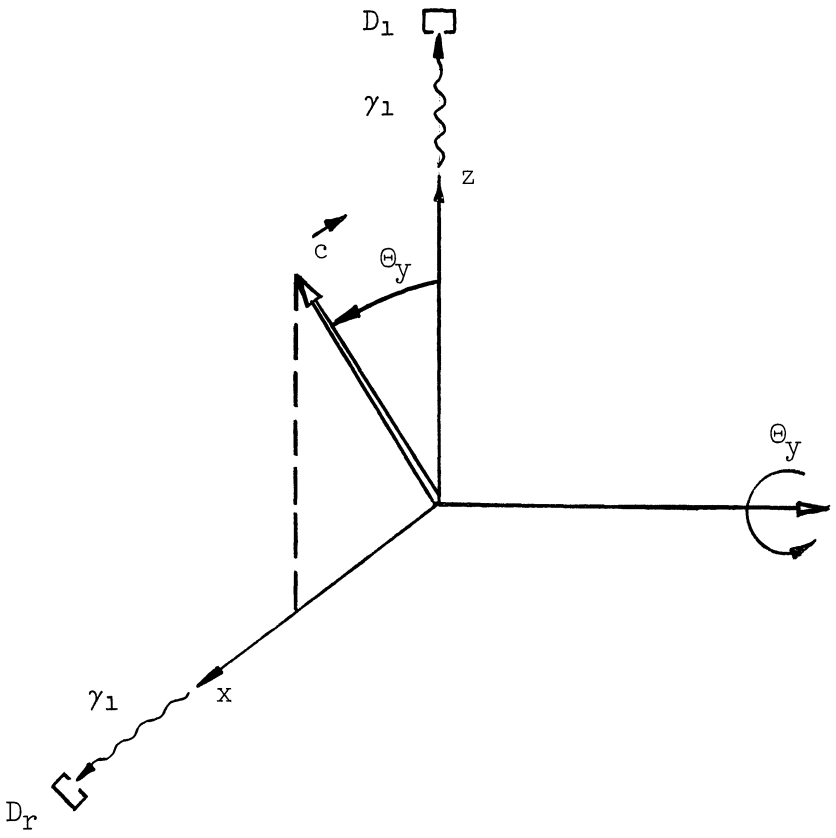


Fig. 7

x-axis we measure the 90° correlations as a function of the rotations, $\textcircled{H}_y$, of the crystal axis $\vec{C}$ which is in the plane of the detectors. Then placing D_2 along the negative z-axis we measure the 180° correlations as a function of $\textcircled{H}_y$.

Since the γ_1 is chosen as the z-axis, then one has $\Theta_1 = \phi_1 = 0$ and

$$Y_{\nu_2 m''-m'}(\theta_1, \varphi_1) = \left(\frac{2\nu_1+1}{4\pi}\right)^{\frac{1}{2}} \delta_{m''-m',0} \cdot \qquad (6.1)$$

For 90° and 180° correlations one has

$$Y_{\nu_2 m''-m}(\theta_2=\tfrac{\pi}{2},\varphi_2=0)=(-1)^{m''-m}\left[\frac{(2\nu_2+1)(\nu_2-m''+m)!}{4\pi(\nu_2+m''-m)!}\right]^{\frac{1}{2}}\times$$

$$\times P_{\nu_2 m''-m}(0), \tag{6.2}$$

and

$$Y_{\nu_2 m''-m}(\theta_2=\pi,\varphi_2=0)=(-1)^{\nu_2}\left[\frac{2\nu_2+1}{4\pi}\right]^{\frac{1}{2}}\delta_{m''-m,0}\,. \tag{6.3}$$

Using Eqs. (6.1), (6.2), (6.3), (4.51) and (3.26), the anisotropy function which is defined to be

$$A=\frac{W(\hat{k}_2/\hat{k}_1,\pi)-W(\hat{k}_2/\hat{k}_1,\frac{\pi}{2})}{W(\hat{k}_2/\hat{k}_1,\frac{\pi}{2})}, \tag{6.4}$$

can be calculated as

$$A=\frac{\sum_{\nu_1\nu_2}\sum_{mm'}A_{\nu_1\nu_2}\,\mathrm{III}_{\nu_1\nu_2}(mm'mm')}{\sum_{\nu_1\nu_2}\sum_{\substack{mm'\\m''}}B_{\nu_1\nu_2}(mm'')\,\mathrm{III}_{\nu_1\nu_2}(mm'm''m')}-1, \tag{6.5}$$

where

$$A_{\nu_1\nu_2}=(-1)^{\nu_2}\,\mathrm{I}'_{\nu_2}(j_i j_f j)\,\mathrm{II}'_{\nu_2}(j_f j_f j), \tag{6.6}$$

$$B_{\nu_1\nu_2}(mm'')=(-1)^{m''-m}\left[\frac{(\nu_2-m''+m)!}{(\nu_2+m''-m)!}\right]^{\frac{1}{2}}P_{\nu_2 m''-m}(0)\times$$

$$\times\,\mathrm{I}'_{\nu_1}(j_i j_f j)\,\mathrm{II}_{\nu_2}(j_f j_f j), \tag{6.7}$$

$$\Pi_{\nu_1\nu_2}(m m' m m') = \sum_{K K'} \langle j'm|K\rangle\langle K|j'm'\rangle\langle j'm'|K'\rangle\langle K'|j'm\rangle \times$$

$$\times (j'm'; j'-m'|\nu_1 0)(j'm; j'-m|\nu_2 0) \times$$

$$\times \left\{ \frac{\hbar}{2}\left[\langle \Gamma_{KK'}(\bar{V}_\tau^{EQ}(R_2))\rangle_T + 2\tau_N^{-1} \right] - \right.$$

$$\left. -i\left[E_{KK'}(R,R_2) + \langle \Delta_{KK'}(\bar{V}_\tau^{EQ}(R_2))\rangle_T \right] \right\}^{-1}, \qquad (6.8)$$

$$\Pi_{\nu_1\nu_2}(m m' m'' m') = \sum_{K K'} \langle j'm|K\rangle\langle K|j'm'\rangle\langle j'm'|K'\rangle\langle K'|j'm''\rangle \times$$

$$\times (j'm'; j'-m'|\nu_1 0)(j'm; j'-m''|\nu_2\, m-m'') \times$$

$$\times \left\{ \frac{\hbar}{2}\left[\langle \Gamma_{KK'}(\bar{V}_\tau^{EQ}(R_2))\rangle_T + 2\tau_N^{-1} \right] - \right.$$

$$\left. -i\left[E_{KK'}(R,R_2) + \langle \Delta_{KK'}(\bar{V}_\tau^{EQ}(R_2))\rangle_T \right] \right\}^{-1}, \qquad (6.9)$$

$$\langle \Delta_{KK'}(\bar{V}_\tau^{EQ}(R_2))\rangle_T \equiv \langle \Delta_K(\bar{V}_\tau^{EQ}(R_2))\rangle_T - \langle \Delta_{K'}(\bar{V}_\tau^{EQ}(R_2))\rangle_T , \qquad (6.10)$$

$$\langle \Gamma_{KK'}(\bar{V}_\tau^{EQ}(R_2))\rangle_T \equiv \langle \Gamma_K(\bar{V}_\tau^{EQ}(R_2))\rangle_T + \langle \Gamma_{K'}(\bar{V}_\tau^{EQ}(R_2))\rangle_T , \qquad (6.11)$$

and

$$\left.\begin{aligned} I'_{\nu_1}(j_i j' j') &= \left(\frac{2\nu_1+1}{4\pi}\right)^{\frac{1}{2}} I_{\nu_1}(j_i j' j'), \\ \Pi'_{\nu_2}(j_f j' j') &= \left(\frac{2\nu_2+1}{4\pi}\right)^{\frac{1}{2}} \Pi_{\nu_2}(j_f j' j'). \end{aligned}\right\} \qquad (6.12)$$

The functions $E_{KK'}(R_1R_2)$, $\langle S_K(V_\tau^{EQ}(R_2))\rangle_T$ and $\langle \frac{\hbar}{2}\Gamma_K(V_\tau^{EQ}(R_2))\rangle_T$ are given by Eqs. (5.13), (5.10) and (5.11) respectively. It will be shown in the next section that the functions $\langle S_K(V_\tau^{EQ}(R_2))\rangle_T$ and $\langle \frac{\hbar}{2}\Gamma_K(V_\tau^{EQ}(R_2))\rangle_T$ are functions of the crystal temperature. Therefore we have shown that the anisotropy of angular correlation is not only a function of the orientations of the crystal axis but also a function of the crystal temperature. This temperature dependence of the anisotropy has been experimentally observed by Ouseph and Canavan[9] by using hafnium single crystal and also observed by Sommerfeldt and Scheter,[10] by E. Matthias et al.[25] and by Hewitt and Taylar.[26]

In order to calculate the function $\langle S_K(V_\tau^{EQ}(R_2))\rangle_T$ one has to assume a model for the charge distribution in the crystal for the calculations of the crystalline field. The model we used is the so-called point-charge model. As mentioned before this model cannot take into account of the shielding, antishielding and co-valent effects. In order to compensate this point one introduces an adjustable multiplication parameter ξ such that the effective point charge of a lattice point is taken to be $\rho_i = \xi_i e_i$. This parameter ξ is determined by comparing the experimental results and the theoretical calculations in microwave techniques.[17],[24] The asymmetry of the crystalline, the Euler angles between Σ^* and $\Sigma^{(II)}$ and the Z^*-components of the field gradient can also be obtained from microwave experiments. Once these data are available, one can find the reduced

matrix element $\langle j\|Q^{(2)}\|j\rangle$ of the nuclear electric quadrupole moment in an excited state by comparing the experimentally measured anisotropy as a function of $\textcircled{H}_y$ with the theoretically calculated anisotropy from Eq. (6.5).

Once the reduced matrix element $\langle j\|Q^{(2)}\|j\rangle$ is known, the conventionally defined nuclear electric quadrupole moment Q, i.e.,

$$eQ = \langle jj|Q_0^{(2)}|jj\rangle = (jj;20|jj)\langle j\|Q^{(2)}\|j\rangle,$$

or

$$Q = \frac{1}{e}\sqrt{\frac{j(2j-1)}{(2j+3)(j+1)}}\,\langle j\|Q^{(2)}\|j\rangle,$$

can be calculated.

On the other hand, for a given nuclear model one can express the expectation values of the electric quadrupole moment operators in an excited state and in the ground state of a nucleus as follows

$$\sum_p e_p \int_{nucl.} \Psi^*_{\alpha jm} r_p^2 Y_0^{(2)}(\theta_p \varphi_p) \Psi_{\alpha jm}\, d\tau = (jm;20|jm)\langle \alpha j\|Q^{(2)}\|\alpha j\rangle,$$

and

$$\sum_p e_p \int_{nucl.} \Psi^*_{\beta JM} r_p^2 Y_0^{(2)}(\theta_p, \varphi_p) \Psi_{\beta JM}\, d\tau = (JM;20|JM)\langle \beta J\|q^{(2)}\|\beta J\rangle,$$

where $\Psi_{\alpha jm}$ and $\Psi_{\beta JM}$ represent respectively the eigenstate of the nucleus, according to the given nuclear model, in the excited state and in the ground state. The reduced maxtric element $\langle \beta J\|q^{(2)}\|\beta J\rangle$, on the left-hand side of the second equation, is the reduced maxtric element of the electric quadrupole moment in the ground state and can be found

by the conventional methods.[28] Once the reduced matrix elements $< \alpha_j \| Q^{(2)} \| \alpha_j >$ and $< \beta J \| q^{(2)} \| \beta J >$ are known, one expects, from these two expressions and for a given nuclear model, to investigate some changes, e.g. a change of charge distribution, within the nucleus due to a transition from an excited state to the ground state by emitting a photon. This subject is beyond the scope of this thesis. Here we only mention the possibility for a further investigation and will not intend to make further discussion.

VII. AN EXAMPLE FOR AXIALLY SYMMETRIC FIELD

As an example for the application of the general theory, let us consider a simple case of a lattice system of octahedron structure. In a regular octahedron structure a nucleus will experience a cubic symmetric field. However, if we assume that an axial symmetric field exists due to a vacancy at a nearest neighboring lattice site, as shown in Fig. 8,

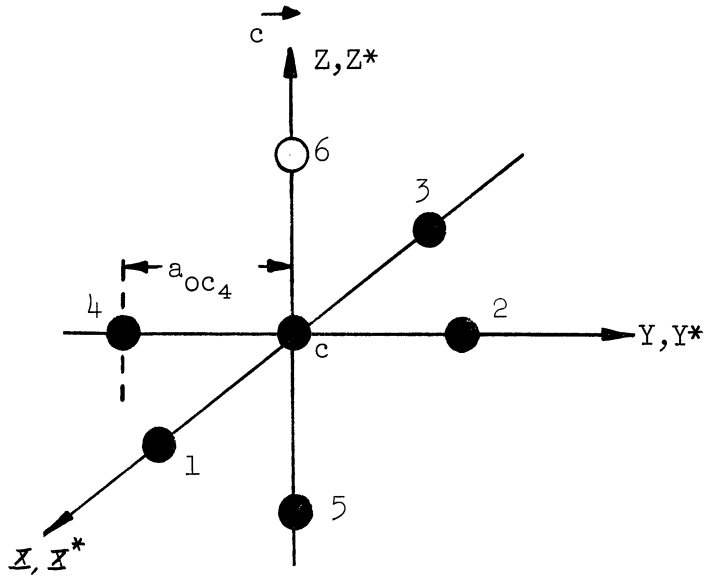


Fig. 8. An O_h point group.

one immediately has the following properties;

(1) $\eta = 0$ for axial symmetry field.

(2) The Euler angles between $\Sigma^{(II)}$ and Σ^* vanished. Since the field is axially symmetric one can choose X* along X and Y* along Y.

(3) $a^*_{oc_1} = a^*_{oc_2} = \cdots = a^*_{oc_6} = a$, which is the lattice constant.

(4) $\rho_1 = \rho_2 = \cdots = \rho_5 = \xi e$, $\rho_6 = 0$ due to a vacancy at the 6th neighbor.

(5) $\epsilon_{ci} = \epsilon_{c_2} = \cdots = \epsilon_{c_5} = 1$; $\epsilon_{c_6} = 0$. Here we have neglected the isotopic effect and assumed that all the nuclei have exactly the same mass, then

$$d_{ei} = 0, \text{ for } i = 1 \cdots 5, \text{ and } d_{c6} = \frac{1}{a}.$$

(6)

$$\theta^*_{oc1} = \frac{\pi}{2}, \quad \varphi^*_{oc1} = 0;$$

$$\theta^*_{oc2} = \frac{\pi}{2}, \quad \varphi^*_{oc2} = \frac{\pi}{2};$$

$$\theta^*_{oc3} = \frac{\pi}{2}, \quad \varphi^*_{oc3} = \pi;$$

$$\theta^*_{oc4} = \frac{\pi}{2}, \quad \varphi^*_{oc4} = \frac{3\pi}{2};$$

$$\theta^*_{oc5} = \pi, \quad \varphi^*_{oc5} = 0;$$

$$\theta^*_{oc6} = 0, \quad \varphi^*_{oc6} = 0.$$

(7)

$$\eta_{1X^*} = 1, \quad \eta_{1Y^*} = 0, \quad \eta_{1Z^*} = 0;$$

$$\eta_{2X^*} = 0, \quad \eta_{2Y^*} = 1, \quad \eta_{2Z^*} = 0;$$

$$\eta_{3X^*} = -1, \quad \eta_{3Y^*} = 0, \quad \eta_{3Z^*} = 0;$$

$$\eta_{4X^*} = 0, \quad \eta_{4Y^*} = -1, \quad \eta_{4Z^*} = 0;$$

$$\eta_{5x^*}=0, \qquad \eta_{5y^*}=0, \qquad \eta_{5z^*}=-1;$$

$$\eta_{6x^*}=0, \qquad \eta_{6y^*}=0, \qquad \eta_{6z^*}=1.$$

We then have

$$\eta_{i\nu^*}\eta_{i\mu^*}=\eta_{i\nu^*}^2\delta_{\nu^*\mu^*},$$

and

$$\eta_{i\nu^*}^2\eta_{i\mu^*}^2=\eta_{i\nu^*}^4\delta_{\nu^*\mu^*}.$$

Using these properties of the lattice system and making use of the fact that the phonons obey Bose statistics so that the number $n_T(\omega)$ of the phonons of energy $\hbar\omega$ presented in a crystal at temperature T is given by the Plank's law

$$n_T(\omega)=\left\{\exp\left(\frac{\hbar\omega}{kT}\right)-1\right\}^{-1},$$

and for high temperature limit by approximating

$$\exp\left(\frac{\hbar\omega}{kT}\right)\simeq 1+\frac{\hbar\omega}{kT},$$

and

$$n_T(\omega)\simeq\frac{kT}{\hbar\omega},$$

Equations (5.10) and (5.11) can be calculated as (Appendix VI);

$$\langle S_K(V_\tau^{EQ}(R_2))\rangle_T=G_K(R_2)T^2, \tag{7.1}$$

and

$$\left\langle\frac{\hbar}{2}\Gamma_K(V_\tau^{EQ}(R_2))\right\rangle_T=B_K(R_2)T^2, \tag{7.2}$$

with

$$G_K(R_2) = \left(\frac{15\hbar V \xi e \omega_M^2}{8\sqrt{\pi}\hbar M_c v^5 a^3}\right)^2 |\langle j\|Q^{(2)}\|j\rangle|^2 \sum_{\zeta=-2}^{2} \sum_{K'\neq K} \times$$
$$\times \sum_{m m'} \sum_{i=1}^{5} (jm'; 2\zeta | jm)^2 \omega_{KK'} |\langle jm|K\rangle| \times$$
$$\times |\langle K'|jm'\rangle|^2 |F(\zeta, \textcircled{H}_y \theta_{oci} \varphi_{oci})|^2 \times$$
$$\times \left[\left\{(\hat{\underline{\rho}} \cdot \hat{\underline{a}}_{oci})^2\right\}_{\Omega(\hat{\rho})}\right]^2 \qquad (7.3)$$

and

$$B_K(R_2) = \left(\frac{\pi \omega_M^5}{5\hbar}\right)\left(\frac{15\hbar V \xi e}{4 M_c v^5 a^3}\right)^2 |\langle j\|Q^{(2)}\|j\rangle|^2 \sum_{\zeta=-2}^{2} \sum_{K'\neq K} \times$$
$$\times \sum_{m m'} \sum_{i=1}^{5} (jm'; 2\zeta | jm)^2 |\langle jm|K\rangle\langle K'|jm'\rangle|^2 \times$$
$$\times |F(\zeta, \textcircled{H}_y \theta_{oci} \varphi_{oci})|^2 \times$$
$$\times \left[\left\{(\hat{\underline{\rho}} \cdot \hat{\underline{a}}_{oci})^2\right\}_{\Omega(\hat{\rho})}\right]^2 . \qquad (7.4)$$

The quantity ω_M in Eqs. (7.3) and (7.4) is the maximum frequency of the Debye spectrum in the crystal at temperature T. Substituting the expressions (7.1) and (7.2) into Eqs. (6.8) and (6.9) the attenuation coefficiencies for the 180° and 90° correlations can now be expressed as,

$$\text{III}_{\nu_1\nu_2}(m m' m m') = \sum_{K K'} \langle jm|K\rangle\langle K|jm'\rangle\langle jm'|K'\rangle\langle K'|jm\rangle \times$$
$$\times (j'm'; j'-m'|\nu_1 0)(j'm; j'-m|\nu_2 0) \times$$
$$\times \left\{\left[B_{KK'}(R_2) T^2 + \hbar \tau_N^{-1}\right] - \right.$$
$$\left. - i\left[E_{KK'}(R_1 R_2) + G_{KK'}(R_3) T^2\right]\right\}^{-1} \qquad (7.5)$$

and

$$\mathrm{III}_{\nu_1\nu_2}(mm'm''m') = \sum_{KK'} \langle jm|K\rangle\langle K|jm'\rangle\langle jm''|K'\rangle\langle K'|jm''\rangle \times$$

$$\times (jm;j-m'|2,0)(jm;j-m''|2,m-m'') \times$$

$$\times \{[B_{KK'}(R_2)T^2 + \hbar\tau_N^{-1}] -$$

$$-i[E_{KK'}(R_1R_2) + G_{KK'}(R_2)T^2]\}^{-1}, \qquad (7.6)$$

where

$$G_{KK'}(R_2) = \left(\frac{15\hbar V\sqrt{3}e\omega_M^2}{8\sqrt{\hbar}M_c v^5 a^3}\right)^2 |\langle j\|Q^{(2)}\|j\rangle|^2 \sum_{\zeta=-2}^{2}\sum_{mm'} \times$$

$$\times \sum_{i=1}^{5} (jm;2\zeta|jm)^2 \omega_{KK'} \times$$

$$\times |F(\zeta, \Theta_y, \theta_{occ}\varphi_{occ})|^2 \{[(\hat{\xi}\cdot\hat{a}_{occ})^2]_{\Omega(\hat{\xi})}\}^2 \times$$

$$\times \Big[\sum_{K''\neq K} |\langle jm|K\rangle\langle K''|jm'\rangle|^2 -$$

$$-\sum_{K'''\neq K'} |\langle jm|K'\rangle\langle K'''|jm'\rangle|^2\Big] \qquad (7.7)$$

and

$$B_{KK'}(R_2) = \left(\frac{\pi\omega_M^5}{5\hbar}\right)\left(\frac{15\hbar V\sqrt{3}e}{4M_c v^5 a^3}\right)^2 |\langle j\|Q^{(2)}\|j\rangle|^2 \times$$

$$\times \sum_{\zeta=-2}^{2}\sum_{mm'}\sum_{i=1}^{5} (jm;2\zeta|jm)^2 \{[(\hat{\xi}\cdot\hat{a}_{occ})^2]_{\Omega(\hat{\xi})}\}^2 \times$$

$$\times |F(\zeta, \Theta_y, \theta_{occ}\varphi_{occ})|^2 \Big[\sum_{K''\neq K} |\langle jm|K\rangle\langle K''|jm'\rangle|^2 +$$

$$+\sum_{K'''\neq K'} |\langle jm|K'\rangle\langle K'''|jm'\rangle|^2\Big]. \qquad (7.8)$$

From Eqs. (7.6), (7.5), (7.2), (7.1) and (6.5) one can calculate the rotational pattern of the anisotropy as a function of the rotations $\textcircled{H}_y$ of the crystal axes. One sees clearly that these functions are very complicated functions of $\textcircled{H}_y$ and no solution can be obtained without actually carrying out the tedious calculations of the summations and the rotational transformations. However, if we make some simplifications, for illustractive purpose, a considerable simple solution for the condition for $\frac{\partial A}{\partial \textcircled{H}_y} = 0$ can be obtained from Eq. (6.5) without carrying out the tedious numerical calculations. This solution is at least quanlitatively justified by the well-known experiments by the Zürich group.[8],[19],[20] As shown in the Appencix VII, the simplified solution for the condition for $\frac{\partial A}{\partial \textcircled{H}_y} = 0$ can be expressed as;

$$[\textcircled{H}_y]_{max.} = \frac{1}{2}\sin^{-1}\left[\frac{\sum_{KK'} G'_{KK'} T^2}{\sum_{KK'} A_{KK'}}\right], \tag{7.9}$$

where

$$A_{KK'} = \sum_{mm'} {}^{*}V_0^{(2)}(0)\langle j\|Q^{(2)}\|j\rangle\left[\langle jm'|K\rangle\langle K|jm\rangle - \langle jm'|K'\rangle\langle K'|jm\rangle\right]\left[\frac{\sqrt{6}}{2}(jm;22|jm) - \frac{3}{2}(jm;20|jm)\right], \tag{7.10}$$

$$G'_{KK'}(0) = 2\left(\frac{15kTV\xi e\omega_M^2}{8\sqrt{\pi}M_c v^5 a^3}\right)^2 |\langle j\|Q^{(2)}\|j\rangle|^2 \times$$

$$\times \sum_{\zeta=-2}^{2} \sum_{mm'} \sum_{i=1}^{5} (jm';2\zeta|jm)^2 \omega_{KK'} \times$$

$$\times F(\zeta,0,\theta_{oci}\varphi_{oci}) F'(\zeta,0,\theta_{oci}\varphi_{oci}) \times$$

$$\times \left[\left\{(\hat{\xi}\cdot\hat{a}_{oci})^2\right\}_{\Omega(\xi)}\right]^2 \left[\sum_{K'''\neq K} |\langle jm|K\rangle\langle K'''|jm'\rangle|^2 - \right.$$

$$\left. - \sum_{K''\neq K} |\langle jm|K\rangle\langle K''|jm'\rangle|^2\right], \qquad (7.11)$$

and

$$F(\zeta,\textcircled{H}_y,\theta_{oci}\varphi_{oci}) = \sum_{\rho} D^{(2)}_{\rho-\zeta}(0,\textcircled{H}_y,0)\, Y^{(2)}_{-\zeta}(\theta_{oci}\varphi_{oci}). \qquad (7.12)$$

The quantities $[\textcircled{H}_y]_{max}$, as given by Eq. (7.9), is the angle of rotations of the crystal axes for the maximum anisotropies in the rotational pattern. From Eq. (7.9) one sees that when the crystal can be considered as a rigid lattices one always has $[\textcircled{H}_y]_{max} = 0$ and Goertzel's and Abragam's theories can only predict this result. However, the present theory predicts for the rotational pattern of anisotropy the phase shift due to the thermal vibrations of the lattice points as given by Eq. (7.9) and this simplified solution also predicts the T^2 dependence of the phase shift.

The phase shift in the rotational pattern of anisotropy are experimentally observed by Albers—Schonberg's et al.[8] by using a

metallic indium single crystal containing the radioactive In^{111}. From the present theory, the phase shifts observed by Albers—Schönbergs can be explained as due to the thermal vibrations of the lattice points. The effect of these thermal vibrations is to produce at the site of the radioactive nucleus a fluctuating electric field gradients which are responsible to produce the phase shifts in the rotational pattern of anisotropy.

VIII. COMPARISON WITH OTHER PERTURBED CORRELATION THEORIES

The theory of the influence on angular correlations of perturbing interactions in the intermediate state was first investigated by Goertzel[3] in 1946 for the case of hyperfine-structure interactions. K. Alder[5],[6] reformulated Goertzel's results and extended that treatment in a way which very clearly displays that the effects of extranuclear perturbation can be factored out as a so-called attenuation factor. However Alder's results are only valid if the extranuclear fields are axially symmetric. In 1953 Abragam and Pound[7] extended Alder's formulations to more general perturbations and their result is written as[7]

$$W = I(k_1)\,II(k_2)\,III(k_1,k_2;\mu_1,\mu_2)\,Y_{k_1}^{\mu_1}(\Omega_1)\,Y_{k_2}^{\mu_2 *}(\Omega_2), \tag{8.1}$$

with

$$III(k_1,k_2;\mu_1,\mu_2) = (I k_1 m'\mu_1 / I k_1 I m)\times$$

$$\times (I k_2 m''\mu_2 / I k_2 I m'')\langle m|b\rangle\langle b|m''\rangle\times$$

$$\times \langle m''|b'\rangle\langle b'|m'\rangle\left[1 - i\left(\frac{\tau_N}{\hbar}\right)(E_b - E_{b'})\right]^{-1}, \tag{8.2}$$

where all the repeated indices should be summed over and E_b and $E_{b'}$ are two eigenvalues of the perturbing Hamiltonian $\mathcal{K}$ which is taken to

be static.

According to Abragam,[7] the procedure for deriving the above formula is as follows: one first starts with the well-known expression for the unperturbed angular correlation and then assumes the presence of an interaction of the nucleus in its intermediate state $|jm\rangle$ with some extranuclear fields. This interaction, which was described by the Hamiltonian $\mathcal{H}$, is assumed to act from the time the first radiation is emitted until the time at which the second radiation is emitted. During this time interval the state $|jm\rangle$ changes to different state $|jm'\rangle$ under the influence of the extranuclear perturbation. This change was represented by a time evolution operator which was simply taken as $U(t) = \exp(-i\mathcal{H}t/\hbar)$ for the static interaction. Finally, for the resolving time of the coincidence system is much larger then the life-time of the intermediate state, one performs the total-time integration for the delayed coincidences and obtains the perturbed correlation functions as given by Eqs. (8.1) and (8.2).

On the other hand, the results from the present systematical treatment can be summarized as

$$W(\hat{k}_2|\hat{k}_1) = \sum_{\nu_1\nu_2} \sum_{\substack{mm' \\ m''m'''}} I_{\nu_1}(jj') II_{\nu_2}(jj') \times$$
$$\times III_{\nu_1\nu_2}(mm'm''m''') \, \tilde{\bar{Y}}^{*}_{\nu_1 m'''-m'}(\theta_1\varphi_1) \times$$
$$\times \tilde{\bar{Y}}_{\nu_2 m''-m}(\theta_2\varphi_2), \qquad (8.3)$$

with

$$III_{\nu_1\nu_2}(mm'm''m''') = \sum_{KK'} (j'm'; j'-m'''/\nu_1, m'-m''') \times$$

$$\times (j'm; j'-m''/\nu_2 m-m'') \langle j'm/K\rangle\langle K/j'm'\rangle \times$$

$$\times \langle j'm'''/K'\rangle\langle K'/j'm''\rangle \times$$

$$\times \left\{ \frac{1}{2}\left[\langle \Gamma_K(V_\tau)^{NC}\rangle_T + \langle \Gamma_{K'}(V_\tau)^{NC}\rangle_T + 2\tau_N^{-1} \right] - \right.$$

$$\left. - i\left[E_K - E_{K'} + \langle S_K(V_\tau)^{NC}\rangle_T - \langle S_{K'}(V_\tau)^{NC}\rangle_T \right] \right\}^{-1} \quad (8.4)$$

First of all it should be pointed out that Eq. (8.1) from the Abragam's formulism and Eq. (8.3) from the present theory are equally valid for either axially symmetric or ansymmetric extranuclear fields. The general expressions for the perturbed angular correlation, as shown in Eqs. (8.1) and (8.3), have the same form and the factors $I(k_1)$ and $II(k_2)$ have exactly the same definitions as the factors $I_{\nu_1}(j_i jj)$ and $II_{\nu_2}(j_i jj)$. However the difference appears in the last factors of the attenuation coefficients, namely in Abragam's formulism the attenuation coefficient contains the factor

$$\langle j'm/b\rangle\langle b/j'm''\rangle\langle j'm'''/b'\rangle\langle b'/j'm'\rangle \left[1 - \left(\frac{i\tau_N}{\hbar} \right)(E_b - E_{b'}) \right]^{-1}, \quad (8.5)$$

but in the present theory we have

$$\langle jm|K\rangle\langle K|jm'\rangle\langle jm''|K'\rangle\langle K'|jm''\rangle \times$$

$$\times\left\{\frac{1}{2}\left[\langle\Gamma_K(V_\tau^{NC})\rangle_T+\langle\Gamma_{K'}(V_\tau^{NC})\rangle_T+2\tau_N^{-1}\right]-\right.$$

$$\left.-i\left[E_K-E_{K'}+\langle S_K(V_\tau^{NC})\rangle_T-\langle S_{K'}(V_\tau^{NC})\rangle_T\right]\right\}^{-1} \qquad (8.6)$$

The eigenket $|K\rangle$, in the expression (8.6), represents an eigenstate of the coupled system due to the static part of the coupling Hamiltonian between the radioactive nucleus and the environment. The effects of the dynamic part of the perturbing interaction on the correlation are described by the thermally averaged energy level displacement function $\langle S_K(V_\tau^{NC})\rangle_T$ and the relaxation function $\langle\Gamma_K(V_\tau^{NC})\rangle_T$. In Abragam's formulism the coupled state is avoided and the effects of the changes of the environments on the correlations are ignored. Therefore for the solid state environment Eq. (8.2) cannot describe the effects of the thermal vibrations of the lattice system on the correlation but in the present theory these effects are systematically built in the attenuation coefficient. In fact the present theory is specially suitable for the solid state environments in which the perturbing interactions can be so naturally decomposed into two parts as shown in the Section IV. One part corresponds to the interaction Hamiltonian V_O^{NC} when the crystal is considered as a rigid lattice system and the other part represents the dynamic interactions V_T^{NC} in-

duced by the vibrations of the lattice points about their equilibrium positions.

Furthermore in the present formulism the angular resolution corrections for the detectors, as shown in Eq. (8.3), are also systematically built in the correlation functions.

IX. CONCLUSION

The present theory of the perturbed angular correlation is developed based on the resolvent method. In this formulism one has a systematic treatment which contains a description of the effects of relaxation on correlations.

As shown in the Section VIII, the perturbed angular correlation functions obtained from the present theory and from the Abragam's formulism have exactly the same form and are equally valid for either axially symmetric or asymmetric extranuclear fields. However, the difference appears in the last factors of the attenuation coefficients as shown in Eqs. (8.5) and (8.6). In Abragam's formulism the coupled state is avoided and the effects of the changes of the environments on the angular correlations are ignored. Therefore, e.g. for the solid state environment, the expression (8.5) cannot describe the effects of the thermal vibrations of the lattice system on the correlation function, hence cannot predict the temperature dependence of the anisotropy as experimentally observed.[9] However, in the present theory these effects are systematically built in the attenuation coefficient. One also sees clearly that the expression (8.6) predicts the temperature dependence of the correlation function. In fact the present theory is developed specially suitable for the solid state environments in which the perturbing interactions can be

decomposed into the static interaction and the dynamic interactions as shown in Section IV.

The treatment of the rotational dependence of the angular correlation, as given in the Section V, has not been previously studied. The present result predicts the rotational dependence of the dynamic interaction. This prediction is justified at least quanlitatively, without carrying out the tedious numerical calculations, by the fact that for the special case of axially symmetric crystalline field the simplified solutions for the conditions for $\frac{\partial A}{\partial (H)_y} = 0$, as given by Eq. (7.9), predict the phase shifts in the rotational pattern of anisotropy, which agrees with the observations by the Zürich group.[8],[19],[20]

Furthermore the present theory has been developed for the cases of asymmetric crystalline fields. This greatly increases the probability to choose the sources for experiments and the probability to investigate the nuclear quadrupole moments in an excited state.

The problems of the recovery effects and the after-effect due to the K-capture and β-decay preceeding the γ-γ cascade have remained untouched. These effects are so complex that a detailed theoretical description becomes extremely complicated, and in fact has not yet been attempted. On the other hand the events happen within a very short time and it seems very difficult to follow the sequence of events experimentally.

Finally, one sees clearly from the present theory that the accuracy of the calculation of the nuclear electric quadrupole moment depends highly on the computations of the electrostatic field gradient at the nuclear site. The calculation of this crystalline field is extremely difficult and a model for the charge distribution in the lattice system is needed. Here we use the point-charge model and introduce an adjustable parameter ξ to take the shielding, antishielding and co-valent effects into account. For the determination of this parameter one has to rely entirely on other measurements. Therefore the progress in the determination of the electric quadrupole moments in an excited state depends highly on the future development of the theory of solid state physics and on a better understanding of the internal fields.

APPENDIX I. DERIVATION OF EXPRESSION (2.13)

It has been assumed that by a physical intuition one can separate the total Hamiltonian $\mathcal{H}$ of a whole physical system into the sum of two parts;

$$\mathcal{H} = H_0 + V,$$

and for an orthonormal complete set of eigenkets $\{|\beta\rangle\}$ the eigenvalue equation

$$H_0|\beta\rangle = E_\beta|\beta\rangle,$$

can be solved. In this representation we will calculate the off-diagonal matrix elements of the time evolution operator $U(\tau)$ by means of the resolvent method. In resolvent method one defines a resolvent function;

$$R(z) = (z + i\mathcal{H})^{-1}. \tag{I.1}$$

This resolvent function can be represented by the Laplase transformation of the time evolution operator $U(t) = e^{-i\mathcal{H}t/\hbar}$ as;

$$R(z) = \int_0^\infty d\frac{t}{\hbar} U(t) e^{-tz/\hbar}, \tag{I.2}$$

then

$$U(t) = \frac{1}{2\pi i}\int_{\lambda-i\infty}^{\lambda+i\infty} dz R(z) e^{zt/\hbar} \tag{I.3}$$

It should be noted that U(t) as represented by Eq. (I.3) satisfies the differential equation

$$i\hbar \frac{d}{dt} U(t) = \mathcal{H} U(t)$$

and the initial condition U(o) = 1.

From Eq. (I.1) we have

$$\sum_{\beta} (Z + i H_0 + i V)_{\beta_f \beta} R_{\beta \beta_i} = \delta_{\beta_f \beta_i} . \tag{I.4}$$

In the representations which diagonalize H_o in each nuclear state we introduce a non-diagonal matrix Q such that[11],[12],[13]

$$R_{\beta_f \beta_i} = R_{\beta_f \beta_f} Q_{\beta_f \beta_i} R_{\beta_i \beta_i} . \tag{I.5}$$

In order to calculate the diagonal matrix elements of the resolvent function let $|\beta_f\rangle = |\beta_i\rangle$ and from Eqs. (I.4) and I.5) one has;

$$\left[Z + i E_{\beta_i} + i V_{\beta_i \beta_i} + i \sum_{\beta \neq \beta_i} V_{\beta_i \beta} R_{\beta \beta} Q_{\beta \beta_i}\right] R_{\beta_i \beta_i} = 1 , \tag{I.6}$$

and

$$\left[Z + i E_{\beta_f} + i V_{\beta_f \beta_f} + i \sum_{\beta \neq \beta_f} V_{\beta_f \beta} R_{\beta \beta} Q_{\beta \beta_f}\right] R_{\beta_f \beta_f} = 1 . \tag{I.7}$$

We now define;

$$\left.\begin{aligned} \frac{i\hbar \gamma_{\beta_i}(Z)}{2} &= i V_{\beta_i \beta_i} + i \sum_{\beta \neq \beta_i} V_{\beta_i \beta} R_{\beta \beta} Q_{\beta \beta_i} , \\ \frac{i\hbar \gamma_{\beta_f}(Z)}{2} &= i V_{\beta_f \beta_f} + i \sum_{\beta \neq \beta_f} V_{\beta_f \beta} R_{\beta \beta} Q_{\beta \beta_f} . \end{aligned}\right\} \tag{I.8}$$

and the functions γ_{β_i} and γ_{β_f} have dimension of sec^{-1}. With this de-

finitions one has;

$$\left.\begin{aligned} R_{\beta_i\beta_i} &= \frac{1}{Z + iE_{\beta_i} + i\frac{\hbar\gamma_{\beta_i}}{2}}, \\ R_{\beta_f\beta_f} &= \frac{1}{Z + iE_{\beta_f} + i\hbar\gamma_{\beta_f}/2}, \end{aligned}\right\} \qquad \text{(I.9)}$$

provides that $Z + i E_{\beta_i} + \frac{i\hbar\gamma_{\beta_i}}{2} \neq 0$ and $Z + i E_{\beta_i} + \frac{i\hbar\gamma_{\beta_f}}{2} \neq 0$.

Letting $|\beta_i> \neq |\beta_f>$ and using Eqs. (I.4), (I.5) and (3.7) we get,

$$Q_{\beta_f\beta_i} = -iV_{\beta_f\beta_i} - i\sum_{\beta\neq\beta_i,\beta_f} V_{\beta_f\beta} R_{\beta\beta} Q_{\beta\beta_i} + i\sum_{\beta\neq\beta_i,\beta_f} V_{\beta_f\beta} R_{\beta\beta} Q_{\beta\beta_f} R_{\beta_f\beta_f} Q_{\beta_f\beta_i} . \qquad \text{(I.10)}$$

By iterating Eq. (I.10) and keeping terms only up to V^2 we get

$$Q_{\beta_f\beta_i} \simeq -iV_{\beta_f\beta_i} - \sum_{\beta\neq\beta_i,\beta_f} V_{\beta_f\beta} R_{\beta\beta} V_{\beta\beta_i} . \qquad \text{(I.11)}$$

Substituting Eq. (I.11) into (I.8) and keeping terms up to V^2 we have

$$\left.\begin{aligned} \frac{i\hbar\gamma_{\beta_i}(Z)}{2} &\simeq iV_{\beta_i\beta_i} + \sum_{\beta\neq\beta_i} V_{\beta_i\beta} R_{\beta\beta} V_{\beta\beta_i}, \\ \frac{i\hbar\gamma_{\beta}(Z)}{2} &\simeq iV_{\beta\beta} + \sum_{\beta'\neq\beta} V_{\beta\beta'} R_{\beta'\beta'} V_{\beta'\beta}, \\ \frac{i\hbar\gamma_{\beta_f}(Z)}{2} &\simeq iV_{\beta_f\beta_f} + \sum_{\beta\neq\beta_f} V_{\beta_f\beta} R_{\beta\beta} V_{\beta\beta_f}. \end{aligned}\right\} \qquad \text{(I.12)}$$

Since the radioactive nucleus ${}_zX^{A**}$ only undergoes the γ-ray cascade decay we therefore realize that $V_{\beta_f\beta_i} = 0$. Then from Eqs. (I.11), (I.5) and (I.9) we have

$$R_{\beta_f\beta_i}(Z) = \sum_{\beta\neq\beta_f,\beta_i} \frac{V_{\beta_f\beta} V_{\beta\beta_i}}{(Z + iE_{\beta_f} + \frac{i\hbar\gamma_{\beta_f}}{2})(Z + iE_{\beta} + \frac{i\hbar\gamma_{\beta}}{2})(Z + iE_{\beta_f} + \frac{i\hbar\gamma_{\beta_f}}{2})} . \qquad \text{(I.13)}$$

From Eqs. (I.13) and (I.3) we obtain the off-diagonal matrix element of U(t);

$$\langle \beta_f | U(t) | \beta_i \rangle = \frac{1}{2\pi i} \sum_{\beta \neq \beta_i, \beta_f} \int_{\lambda - i\infty}^{\lambda + i\infty} \frac{V_{\beta_f \beta} V_{\beta \beta_i} e^{tz/\hbar} dz}{(z + iE_{\beta_f} + \frac{i\hbar\gamma_{\beta_f}}{2})(z + iE_{\beta} + \frac{i\hbar\gamma_{\beta}}{2})(z + iE_{\beta_i} + \frac{i\hbar\gamma_{\beta_i}}{2})}, \tag{I.14}$$

where

$$\left.\begin{aligned} \frac{i\hbar\gamma_{\beta_i}(z)}{2} &\simeq i V_{\beta_i \beta_f} + \sum_{\beta \neq \beta_i}' \frac{V_{\beta_i \beta} V_{\beta \beta_i}}{z + iE_{\beta_i}}, \\ \frac{i\hbar\gamma_{\beta}(z)}{2} &\simeq i V_{\beta \beta} + \sum_{\beta' \neq \beta}' \frac{V_{\beta \beta'} V_{\beta' \beta}}{z + iE_{\beta}}, \\ \frac{i\hbar\gamma_{\beta_f}(z)}{2} &\simeq i V_{\beta_f \beta_f} + \sum_{\beta \neq \beta_f}' \frac{V_{\beta_f \beta} V_{\beta \beta_f}}{z + iE_{\beta_f}}, \end{aligned}\right\} \tag{I.15}$$

It will be shown that the function γ_i is the relaxation function and the energy level displacement function for the energy level corresponding to the ith eigenstate of the radioactive nucleus. Since our purpose is to investigate the effects on the angular correlation by the interactions between the nucleus in the intermediate state and the crystal, we will only consider the relaxation and energy displacement associated with the intermediate states.

By neglecting γ_{β_i} and γ_{β_f}, Eq. (I.14) can be written as;

$$\langle \beta_f | U(t) | \beta_i \rangle = \sum_{\beta \neq \beta_f, \beta_i}' V_{\beta_f \beta} V_{\beta \beta_i} \frac{1}{(E_{\beta_f} - E_{\beta_i})} \left[I_1(E_{\beta_i}, E_{\beta}) - I_2(E_{\beta_f}, E_{\beta}) \right], \tag{I.16}$$

where

$$\left.\begin{aligned} I_1(E_{\beta_i}, E_{\beta}) &= \lim_{\lambda \to 0+} \frac{1}{2\pi i} \int_{\lambda - i\infty}^{\lambda + i\infty} \frac{dz\, e^{zt/\hbar}}{(z + iE_{\beta_i})(z + iE_{\beta} + \frac{i\hbar\gamma_{\beta}(z)}{2})}, \\ I_2(E_{\beta_f}, E_{\beta}) &= \lim_{\lambda \to 0+} \frac{1}{2\pi i} \int_{\lambda - i\infty}^{\lambda + i\infty} \frac{dz\, e^{zt/\hbar}}{(z + iE_{\beta_f})(z + iE_{\beta} + \frac{i\hbar\gamma_{\beta}(z)}{2})}, \end{aligned}\right\} \tag{I.17}$$

In Eq. (I.17) we have made use of the fact that all the singularities of the function of Z lie on the imaginary axis. Since the inversion formula holds for all positive finite value of λ one can take the limit $\lambda \to 0+$ and still have the singularities lying to the left of the Bromwich path.

Making the transformation of $Z = Z - i E_{\beta_i}$ and using the convolution theorem, Eq. (I.17) can be expressed;

$$\left.\begin{aligned} I_1(E_{\beta_i}, E_\beta) &= \left[\frac{-i}{E_\beta - E_{\beta_i} + \hbar\gamma_\beta(-iE_{\beta_i})/2}\right] e^{-iE_{\beta_i}t/\hbar}, \\ I_2(E_{\beta_f}, E_\beta) &= \left[\frac{-i}{E_\beta - E_{\beta_f} + \hbar\gamma_\beta(-iE_{\beta_f})/2}\right] e^{-iE_{\beta_f}t/\hbar}, \end{aligned}\right\} \tag{I.18}$$

where

$$\frac{i\hbar\gamma_\beta(-iE_{\beta_i})}{2} = V_{\beta\beta} + \lim_{\lambda\to 0} \sum_{\beta_m \neq \beta} \frac{|V_{\beta_m\beta}|^2}{E_{\beta_i} - E_{\beta_m} + i\lambda}. \tag{I.19}$$

Substituting Eq. (I.18) into Eq. (I.16) and realizing that for a fixed E_{β_i} the most important contribution for Eq. (I.16) arises when $E_{\beta_i} \simeq E_{\beta_f}$ and making use of the fact that

$$\lim_{\tau/\hbar\to\infty} \left| e^{-iE_{\beta_i}t/\hbar} - e^{-iE_{\beta_f}t/\hbar} \right|^2 \left[(E_{\beta_i} - E_{\beta_f})^2 \tau \right]^{-1}$$

can be replaced by $\delta(E_{\beta_i} - E_{\beta_f})$, one obtains;

$$\frac{|\langle\beta_i|U(\tau)|\beta_f\rangle|^2}{\tau} = \left| \sum_{\beta\neq\beta_i\beta_f} \frac{V^{\gamma}_{\beta_i\beta} V^{\gamma}_{\beta\beta_f}}{E_\beta - E_{\beta_i} + \frac{\hbar}{2}\gamma_\beta(-iE_{\beta_i})} \right|^2 \delta(E_{\beta_i} - E_{\beta_f}). \tag{I.20}$$

APPENDIX II. DERIVATION OF THE EXPRESSION (3.17)

From Eq. (3.16), one has

$$\Lambda^{(i)}(m'm''') = \sum_{\lambda_1}\sum_{m_i} P(m_i) \sum_{L_1 L_1' M_1 M_1' \mu_1 \mu_1'} (-1)^{M_1+M_1'} b_{L_1} b_{L_1'} \times$$

$$\times A_{L_1,-M_1} A^{*}_{L_1',-M_1'} \langle j \| T_{L_1} \| j_i \rangle \langle j \| T_{L_1'} \| j_i \rangle^{*} \times$$

$$\times (jm'; L_1\mu_1 | j_i m_i)(jm''' L_1'\mu_1' | j_i m_i) \times$$

$$\times D^{L_1}_{\mu_1 M_1}(R_1)\, D^{L_1'*}_{\mu_1' M_1'}(R_1) . \qquad \text{(II.1)}$$

By the symmetry properties of the rotational matrices

$$D^{L_1'*}_{\mu_1' M_1'}(R_1) = (-1)^{\mu_1'-M_1'} D^{L_1'}_{-\mu_1' -M_1'}(R_1) , \qquad \text{(II.2)}$$

and the Clebsch-Gordan series

$$D^{L_1}_{\mu_1 M_1}(R_1)\, D^{L_1'}_{\mu_1' M_1'}(R_1) = \sum_{\nu_1} (L_1\mu_1; L_1'\mu_1' | \nu_1, \mu_1+\mu_1') \times$$

$$\times (L_1 M_1; L_1' M_1' | \nu_1, M_1+M_1')\, D^{\nu_1}_{\mu_1+\mu_1', M_1+M_1'}(R_1) ; \qquad \text{(II.3)}$$

the product of the two rotational matrices in Eq. (I.1) can be written as;

$$D^{L_1}_{\mu_1 M_1}(R_1)\, D^{L_1'*}_{\mu_1' M_1'}(R_1) = (-1)^{\mu_1'-M_1'} \sum_{\nu_1} (L_1\mu_1; L_1'-\mu_1' | \nu_1, \mu_1-\mu_1') \times$$

$$\times (L_1 M_1; L_1' -M_1' | \nu_1, M_1-M_1')\, D^{\nu_1}_{\mu_1-\mu_1', M_1-M_1'} . \qquad \text{(II.4)}$$

Combining Eqs. (II.1) and II.4), one has;

$$\Lambda^{(1)}(m'm''') = \sum_{\lambda_1} \sum_{m_i} P(m_i) \sum_{L_1 L_1' M_1 M_1'} (-1)^{M_1 - m''' + m_i} b_{L_1} b_{L_1'} \times$$

$$\times A_{L_1,-M_1} A^{*}_{L_1',-M_1'} \langle j \| T_{L_1} \| j_i \rangle \langle j \| T_{L_1'} \| j_i \rangle^{*} \times$$

$$\times (j m'; L_1 m_i - m' | j_i m_i)(j m'''; L_1' m_i - m''' | j_i m_i) \times$$

$$\times \sum_{\nu_1} (L_1 m_i - m'; L_1' m''' - m_i | \nu_1 m''' - m') \times$$

$$\times (L_1 M_1; L_1' - M_1' | \nu_1 M_1 - M_1') D^{\nu_1}_{m''' - m', M_1 - M_1'}(R_1) . \qquad \text{(II.5)}$$

In order to carry out the $\sum_{m_i}$, first let us consider the symmetry properties of the Clebsch-Gordan coefficiency, i.e.,

$$(j m'; L_1 m_i - m' | j_i m_i) = (-1)^{2j + j_i - L_1 - m'} \left(\frac{2j_i + 1}{2L_1 + 1}\right)^{\frac{1}{2}} \times$$

$$\times (j - m'; j_i m_i | L_1 m_i - m') , \qquad \text{(II.6)}$$

then the product of the two Clebsch-Gordan coefficients can be written as;

$$(j m'; L_1 m_i - m' | j_i m_i)(L_1 m_i - m'; L_1' m''' - m_i | \nu_1 m''' - m') =$$

$$= (-1)^{2j + j_i - L_1 - m'} \left(\frac{2j_i + 1}{2L_1 + 1}\right)^{\frac{1}{2}} (j - m'; j_i m_i | L_1 m_i - m') \times$$

$$\times (L_1 m_i - m'; L_1' m''' - m_i | \nu_1 m''' - m') . \qquad \text{(II.7)}$$

The right-hand side of Eq. (II.7) represents the coupling of three angular moments j, j_i and L_1' into ν_1 through the intermediate angular momentum L_1. One can choose another intermediate angular momentum state to couple these three angular momenta. The transformation between the possible different intermediate momentum states is an unitary transformation and is related by the Racah coefficients as

$$(j_1' m_1; j_2' m_2 | j'' m_1 + m_2)(j'' m_1 + m_2; j_3' m_3 | j' m_1 + m_2 + m_3) =$$

$$= \sum_{j''} [(2j'+1)(2j''+1)]^{\frac{1}{2}} W(j_1' j_2' j j_3'; j' j'') \times$$

$$\times (j_2' m_2; j_3' m_3 | j'' m_2 + m_3)(j_1' m_1; j'' m_2 + m_3 | j' m_1 + m_2 + m_3). \quad \text{(II.8)}$$

Using Eq. (II.8), Eq. (II.7) can be written as;

$$(j' m'; L_1 m_i - m' | j_i' m_i)(L_1 m_i - m'; L_1' m''' - m_i | 2_1 m'' - m) =$$

$$= (-1)^{2j' + j_i' - L_1 - m} (2j_i' + 1)^{\frac{1}{2}} \sum_t (2t+1)^{\frac{1}{2}} W(j j_i' 2_1 L_1'; L_1 t) \times$$

$$\times (j_i' m_i, L_1' m''' - m_i | t m''')(j - m'; t m''' | 2_1 m'' - m'). \quad \text{(II.9)}$$

The remaining Clebsch-Gordan coefficiency containing m' and m"' in (II.5) can be written

$$(j m''; L_1' m_i - m''' | j_i' m_i) = (-1)^{j_i' + L_1' - j' + L_1' + m_i - m'''} \times$$

$$\times \left(\frac{2j_i' + 1}{2j' + 1}\right)^{\frac{1}{2}} (j_i' m_i; L_1' m''' - m_i | j' m'''). \quad \text{(II.10)}$$

Substituting (II.10) and II.9) into (II.5) and using the orthogonal property of the Clebsch-Gordan coefficients;

$$\sum_{m_1} (j_1 m_1; j_2 m-m_1 | j m)(j_1 m_1; j_2' m-m_1 | j' m) = \delta_{jj'},$$

and the symmetric properties of the Racah coefficient;

$$W(jj_i' \nu_1 L_1'; L_1 j') = (-1)^{L_1+j'-j_i'-\nu_1} W(jj' L_1 L_1'; \nu_1 j_i')$$

one finally obtains

$$A^{(1)}(m'm''') = (2j_i'+1) \sum_{L_1 L_1'} \sum_{\tau_1 \nu_1} (-1)^{m'+L_1-j_i'} \langle j \| T_{L_1} \| j_i' \rangle \times$$

$$\times \langle j' \| T_{L_1'} \| j_i' \rangle^* C_{\nu_1 \tau_1}(L_1 L_1')(jm'; j-m''' | \nu_1 m'-m''') \times$$

$$\times W(jj' L_1 L_1'; \nu_1 j_i') D^{\nu_1}_{m'''-m', -\tau_1}(R_1), \qquad \text{(II.11)}$$

with

$$C_{\nu_1 \tau_1}(L_1 L_1') = \sum_{\lambda_1} \sum_{M_1} (-1)^{L_1-M_1} b_{L_1} b_{L_1'} A_{L_1,-M_1} A^{*}_{L_1',-M_1-\tau_1} \times$$

$$\times (L_1 M_1; L_1'-M_1-\tau_1 | \nu_1 - \tau_1). \qquad \text{(II.12)}$$

$C_{\nu_1 \tau_1}(L_1 L_1')$ is the so-called radiation parameter which is characteristic of the emitted radiation and is independent of the properties of the nuclear states involved in the transition. A detail discussion and calculations of this radiation parameter have been given by L.C. Biedenharn and M.E. Rose.[14]

APPENDIX III. DERIVATION OF THE EXPRESSION (3.22)

From Eq. (3.21), the products of the radiation matrix elements can be written as;

$$\sum_{\lambda_1\lambda_2}\sum_{m_i m_f} P(m_i)\langle j'm'|\bar{V}(\gamma)|j_i m_i\rangle\langle j'm''|\bar{V}(\gamma)|j_i m_i\rangle^* \times$$

$$\times\langle j_f m_f|\bar{V}(\delta)|j'm\rangle\langle j_f m_f|\bar{V}(\delta)|j'm''\rangle^* =$$

$$=(2j'+1)(2j_i+1)\sum_{L_1L_1'}\sum_{L_2L_2'}\sum_{\tau_1\tau_2}\sum_{\nu_1\nu_2}(-1)^{m'+m''+L_1'+L_2'-j_i-j_f}\times$$

$$\times\langle j'\|T_{L_1}\|j_i\rangle\langle j'\|T_{L_1'}\|j_i\rangle^*\langle j_f\|T_{L_2}\|j\rangle\langle j_f\|T_{L_2'}\|j\rangle^*\times$$

$$\times C_{\nu_1\tau_1}(L_1L_1')C_{\nu_2\tau_2}(L_2L_2')(j'm';j'-m''|\nu_1,m'-m'')\times$$

$$\times(jm;j'-m''|\nu_2,m-m'')W(jj'L_1L_1';\nu_1 j_i)\times$$

$$\times W(jjL_2L_2';\nu_2 j_f)D^{\nu_1}_{m''-m',-\tau_1}(R_1)D^{\nu_2}_{m-m'',-\tau_2}(R_2), \qquad \text{(III.1)}$$

where $\tau_1 = M_1-M_1'$ and $\tau_2 = M_2-M_2'$ are the polarization parameters of the emitted radiations. Since we are only interesting in the direction-direction correlation, the rotations of R_1 and R_2 for the γ_1 and the γ_2 need only two Euler angles to define their rotations for this unpolarized observations. The absence of any dependence of the third Euler angle will result $\tau = 0$. Thus for unpolarized γ-rays one has;[14]

$$\left.\begin{aligned} C_{\nu_1 0}(L_1 L_1') &= (-1)^{L_1+1}\left[(2L_1+1)(2L_1'+1)\right]^{\frac{1}{2}}(L_1 1; L_1' -1 | \nu_1 0), \\ C_{\nu_2 0}(L_2 L_2') &= (-1)^{L_2+1}\left[(2L_2+1)(2L_2'+1)\right]^{\frac{1}{2}}(L_2 1; L_2' -1 | \nu_2 0). \end{aligned}\right\} \quad \text{(III.2)}$$

Furthermore the reduced matrix element in Eq. (III.1) are in general neither real nor Hermitian but they satisfy;

$$\langle j \| T_{L'} \| j_i \rangle^* = (2j+1)^{\frac{1}{2}}(2j_i+1)^{-\frac{1}{2}}(-1)^{j_i - L' + j} \langle j_i \| T_{L'} \| j \rangle. \quad \text{(III.3)}$$

Using Eqs. (III.2) and (III.3), Eq. (III.1) can be written for a pure multipole radiation with multipolarity of L_1 and L_2 as;

$$\begin{aligned} &(2j'+1)\sum_{L_1 L_1' L_2 L_2' \nu_1 \nu_2}\sum\sum(-1)^{m'+m''+(L_2+L_2')-(L_1+L_1')-2j_f} \times \\ &\times f_{\nu_1}(L_1 L_1' j_i j') f_{\nu_2}(L_2 L_2' j_f j') \times \\ &\times (jm'; j'-m''' | \nu_1 m'-m''')(j'm; j'-m'' | \nu_2 m-m'') \times \\ &\times D^{\nu_1}_{m'''-m',0}(R_1) D^{\nu_2}_{m-m'',0}(R_2), \end{aligned} \quad \text{(III.4)}$$

where we have defined

$$\begin{aligned} f_\nu(LL' j_i j) &= (-1)^{j_i - j - 1}\left[(2j+1)(2L+1)(2L'+1)\right]^{\frac{1}{2}} \times \\ &\times (L1; L' -1 | \nu 0) W(jj' L L'; \nu j_i). \end{aligned} \quad \text{(III.5)}$$

The functions $f_\nu(LL' j_i j)$ have been calculated by Biendenharm and Rose for L = 1, 2, 3, 4, j_i = 0, 1, 2, 3, 4 and integer $j \leq 5$ and for all

the necessary ν values.

By making use of the expressions

$$D^{\nu}_{m_1-m_2,0}(\theta,\varphi,0) = \left(\frac{4\pi}{2\nu+1}\right)^{\frac{1}{2}} Y^{*}_{\nu\, m_1-m_2}(\theta,\varphi)$$

and

$$Y^{*}_{\nu\, m_1-m_2}(\theta,\varphi) = (-1)^{m_1-m_2}\, Y_{\nu\, m_2-m_1}(\theta,\varphi),$$

and defining;

$$\left.\begin{aligned} I_{\nu_1}(j_i j j) &= \left[\frac{4\pi(2j+1)}{2\nu_1+1}\right]^{\frac{1}{2}} \sum_{L_1 L_1'} (-1)^{L_1+L_1'} f_{\nu_1}(L_1 L_1' j_i j) \\ II_{\nu_2}(j_f j j) &= \left[\frac{4\pi(2j+1)}{2\nu_2+1}\right]^{\frac{1}{2}} \sum_{L_2 L_2'} (-1)^{L_2 L_2'} f_{\nu_2}(L_2 L_2' j_f j) \end{aligned}\right\} \quad \text{(III.6)}$$

Eq. (III.1) can be written as;

$$\begin{aligned} &\sum_{\lambda_1\lambda_2} \sum_{m_i m_f} P(m_i) \langle j m' | V(\gamma) | j_i m_i \rangle \langle j m''' | V(\gamma) | j_i m_i \rangle^{*} \times \\ &\quad \times \langle j_f m_f | V(\gamma) | j m \rangle \langle j_f m_f | V(\gamma) | j m'' \rangle^{*} = \\ &= \sum_{\nu_1\nu_2}{}' I_{\nu_1}(j_i j j)\, II_{\nu_2}(j_f j j)\, (j m; j-m'' | \nu_1 m'-m''') \times \\ &\quad \times (j m; j-m'' | \nu_2 m-m''')\, Y^{*}_{\nu_1\, m'''-m'}(\theta_1,\varphi_1) \times \\ &\quad \times Y_{\nu_2\, m''-m}(\theta_2,\varphi_2). \end{aligned} \quad \text{(III.7)}$$

The factors $I_{\nu_1}(j_i j j)$ and $II_{\nu_2}(j_f j j)$ depend only on the degrees of freedom of the successive radiations and are independent of the external perturbing fields.

The indices ν_1 and ν_2 are even integers and are restricted by the following inequalities;

$$\left.\begin{aligned} 0 \le \nu_1 \le 2j', \qquad & 0 \le \nu_1 \le 2L_1, \\ 0 \le \nu_2 \le 2j', \qquad & 0 \le \nu_2 \le 2L_2 \end{aligned}\right\} \tag{III.8}$$

The integers L_1 and L_2 are the highest orders in the multipole expansions of the first and the second γ-rays respectively.

APPENDIX IV. DERIVATION OF EXPRESSION (3.26) AND (3.27)

After integrating over k_2, the integration in Eq. (3.23) can be expressed as

$$I = \frac{1}{(\hbar c)^6} \frac{F(a a' \epsilon \epsilon')}{(a - a') - i(\epsilon + \epsilon')}, \tag{IV.1}$$

where

$$F(a a' \epsilon \epsilon') = \int_{E_{1m}}^{E_M} \frac{E_1^2 f(E_1) dE_1}{E_1 - a + i\epsilon} - \int_{E_{1m}}^{E_M} \frac{E_1^2 f(E_1) dE_1}{E_1 - a' - i\epsilon'} \tag{IV.2}$$

and

$$\left.\begin{aligned}
a &= E_{K_i} - E_K - \langle S_K(\bar{V}_\tau^{NC}) \rangle_T, \\
a' &= E_{K_i} - E_{K'} - \langle S_{K'}(\bar{V}_\tau^{NC}) \rangle_T, \\
\epsilon &= \frac{\hbar}{2} \left[\langle \Gamma_K(\bar{V}_\tau^{NC}) \rangle_T + \Gamma_K(\bar{V}^\gamma) \right], \\
\epsilon' &= \frac{\hbar}{2} \left[\langle \Gamma_{K'}(\bar{V}_\tau^{NC}) \rangle_T + \Gamma_{K'}(\bar{V}_\tau^{NC}) \right], \\
f(E_1) &= \left[E_{K_i} - E_{K_f} - E_1 \right]^2 I_{\nu_1}(j_i j j) \Pi_{\nu_2}'(j_f j j)
\end{aligned}\right\} \tag{IV.3}$$

The prime in $\Pi_{\nu_2}(j_f j j)$ denotes the replacement of E_2 in the function $\Pi_{\nu_2}(j_f j j)$ by $E_{ki} - E_{k_f} - E_1$, here E_1 is the first γ-ray energy. If the total width ϵ of the radioactive nucleus is much less than the nuclear level splitting, then by using the relations

and $$\lim_{\epsilon \to 0} \frac{1}{y + i\epsilon} = P\left(\frac{1}{y}\right) - i\pi\delta(y),$$

$$\lim_{\epsilon \to 0} \frac{1}{y - i\epsilon} = \mathcal{P}\left(\frac{1}{y}\right) + i\pi\delta(y),$$

Eq. (IV.2) can be approximated as;

$$F(aa'\epsilon\epsilon') \simeq \mathcal{P}\int_{E_{1m}} \frac{E_1^2 f(E_1)dE_1}{E_1 - a} - \mathcal{P}\int_{E_{1m}} \frac{E_1^2 f(E_1)dE_1}{E_1 - a'} - i\pi a^2 f(a) - i\pi a'^2 f(a'). \tag{IV.4}$$

Let E_{01} be the first γ-ray energy emitted by the nucleus without extranuclear perturbations, then the quantities a and a' can be written as;

$$\left.\begin{aligned} a &= E_{01} \pm \Delta E_K - \langle S_K(V_\tau^{NC})\rangle_T, \\ a' &= E_{01} \pm \Delta E_{K'} - \langle S_{K'}(V_\tau^{NC})\rangle_T. \end{aligned}\right\} \tag{IV.5}$$

where ΔE_K stands for the nuclear energy level splitting due to extranuclear perturbations. It is realized that the quantity E_{01}, which is in the order of Mev, is much greater than ΔE_K and $\langle S_K(V_\tau^{NC})\rangle_T$ which is in the order of ev. By neglecting ΔE_K and $\langle S_K(V_\tau^{NC})\rangle_T$ compared with E_{01}, one has $a \simeq E_{01}$ and $a' \simeq E_{01}$, and Eq. (IV.4) becomes

$$F(aa'\epsilon\epsilon') \simeq -2\pi i E_{01}^2 E_{02}^2 I_{\nu_1}(\gamma_i\gamma_f) \Pi_{\nu_2}(\gamma_f\gamma_f). \tag{IV.6}$$

Substituting Eq. (IV.6) into (IV.1) one has

$$I = \frac{2\pi E_{01}^2 E_{02}^2}{(\hbar c)^6} \frac{I_{\nu_1}(\gamma_i\gamma_f)\, \Pi_{\nu_2}(\gamma_f\gamma_f)}{(\epsilon + \epsilon') - i(a - a')}. \tag{IV.7}$$

Combining Eqs. (IV.7) and (3.26) one obtains Eqs. (3.26) and (3.27).

On the other hand, when the total width of the radioactive nucleus is much greater than the nuclear level splitting, one can write the integration in Eq. (3.23) as;

$$I = \frac{1}{(\hbar c)^6} \int_{E_{1m}}^{E_{1M}} \frac{E_1^2 f(E_1) dE_1}{|\zeta + E_1|^2 (1 + \frac{\delta_K}{\zeta + E_1})(1 + \frac{\delta_{K'}}{\zeta + E_1})^*}, \qquad \text{(IV.8)}$$

with

$$\left.\begin{aligned} E_1 &= \hbar c k_1 \\ \zeta &= -E_{01} + i \frac{\hbar}{2} \Gamma(V^\gamma), \\ \delta_K &= \Delta E_K + i \frac{\hbar}{2} \langle \Gamma_K(V_\tau^{NC}) \rangle_T, \end{aligned}\right\} \qquad \text{(IV.9)}$$

here we have neglected the nuclear level displacements due to the radiation interaction and extranuclear perturbations. The factor $|\zeta+E_1|^2$ in the denominator of Eq. (IV.8) can be expressed as

$$|\zeta + E_1|^2 = (E_1 - E_{01})^2 + \left[\frac{\hbar}{2} \Gamma(V^\gamma)\right]^2, \qquad \text{(IV.10)}$$

Since E_{01}, which is the first γ-ray energy, is much greater than $\frac{\hbar}{2}\Gamma(V^\gamma)$, one sees that the integrand of Eq. (IV.8) is a sharply peaked function at $E_1 = E_{01}$. Thus by evaluating the integrand at this point, one has

$$\begin{aligned} I = \frac{1}{(\hbar c)^6} & E_{01}^2 E_{02}^2 I_{\nu_1}(\gamma_i \gamma_f \gamma') II_{\nu_2}(\gamma_f \gamma_f \gamma') \times \\ & \times \left\{\left[\Delta E_K + i \frac{\hbar}{2}\left\{\langle \Gamma_K(V_\tau^{NC}) \rangle_T + \Gamma(V^\gamma)\right\}\right] \times \right. \\ & \left. \times \left[\Delta E_{K'} - i \frac{\hbar}{2}\left\{\langle \Gamma_{K'}(V_\tau^{NC}) \rangle_T + \Gamma(V^\gamma)\right\}\right]\right\}^{-1}. \end{aligned} \qquad \text{(IV.11)}$$

Using Eq. (IV.11), the attenuation coefficient for the perturbed angular correlation function can be expressed as;

$$\mathrm{III}_{\nu_1\nu_2}(m m' m'' m''') = \sum_{\kappa\kappa'} (j' m'; j'-m'' | \nu_1, m'-m''') \times$$

$$\times (j' m; j'-m'' | \nu_2 m-m'') \langle j' m | \kappa \rangle \langle \kappa | j' m' \rangle \times$$

$$\times \langle j' m'' | \kappa \rangle \times \kappa' | j' m'' \rangle \{ [\Delta E_\kappa + i \frac{\hbar}{2} \{ \langle \Gamma_\kappa (\bar{V}_\tau^{NC}) \rangle_T +$$

$$+ [(V^\gamma)\}] [\Delta E_{\kappa'} - i \frac{\hbar}{2} \{ \langle \Gamma_{\kappa'} (\bar{V}_\tau^{NC}) \rangle_T + [(V^\gamma)\}] \}^{-1} \qquad \text{(IV.12)}$$

Since the total width of the radioactive nucleus is much greater than the nuclear level splitting, then by neglecting $\frac{\hbar}{2} \langle \Gamma_k (V_\tau^{NC}) \rangle_T$ compared with $\frac{\hbar}{2} \Gamma(V^\gamma)$ and taking the limit $\frac{\Delta E_k}{\frac{\hbar}{2} \Gamma(V^\gamma)} \ll 1$, the attenuation coefficient can be reduced to

$$\mathrm{III}_{\nu_1\nu_2}(m m' m'' m''') = \left(\frac{2}{\hbar [(V^\gamma)} \right)^2 (j' m'; j'-m'' | \nu_1, m'-m''') \times$$

$$\times (j' m; j'-m'' | \nu_2 m-m'') \delta_{mm'} \delta_{m''m'''}. \qquad \text{(IV.13)}$$

Substituting Eq. (IV.13) into Eq. (3.26) and using the orthogonal properties of the clebsch-Gordan coefficients, one can easily show that the angular correlation is not perturbed in this case. This agrees with Goertzel's result for the unperturbed angular correlation.

APPENDIX V. DERIVATION OF EXPRESSION (5.3)

From Eq. (5.2), one has;

$$ {}^{(I)}V^{(2)}_{-\zeta} = \sum_{\rho,r} D^{(2)}_{r\rho}(\alpha\beta\gamma)\, D^{(2)}_{\rho-\zeta}(\Phi,\textcircled{H},0)\, {}^{*}V^{(2)}_{r} . \tag{V.1} $$

According to Wigner,[23] the rotational matrix $D^{(2)}_{r\rho}(\alpha\beta\gamma)$ can be written as;

$$ D^{(2)}_{r\rho}(\alpha\beta\gamma) = e^{-ir\alpha}\, e^{-i\rho\gamma} \sum_{\kappa} \frac{(-1)^{\kappa}\left[(2+r)!(2-r)!(2+\rho)!(2-\rho)!\right]^{\frac{1}{2}}}{\kappa!(2-\kappa-r)!(2+\rho-\kappa)!(r+\kappa-\rho)!} \times $$

$$ \times \left(\cos\frac{\beta}{2}\right)^{4+\rho-r-2\kappa} \left(-\sin\frac{\beta}{2}\right)^{r-\rho+2\kappa} . \tag{V.2} $$

In Σ^{*}, we need only to sum over $r = 0, \pm 2$. By Eq. (V.2), the non-vanishing matrix elements for $D^{(2)}_{r\rho}(\alpha\beta\gamma)$ and $D^{(2)}_{\rho-\beta}(\Phi, \textcircled{H}, 0)$ can be calculated as;

$$ \left.\begin{aligned} D^{(2)}_{02}(\alpha\beta\gamma) &= \frac{\sqrt{6}}{4} e^{-i2\gamma} \sin^2\beta , \\ D^{(2)}_{01}(\alpha\beta\gamma) &= \frac{\sqrt{6}}{2} e^{-i\gamma} \sin\beta\cos\beta , \\ D^{(2)}_{00}(\alpha\beta\gamma) &= 1 - \frac{3}{2}\sin^2\beta , \\ D^{(2)}_{0-1}(\alpha\beta\gamma) &= -\frac{\sqrt{6}}{2} e^{i\gamma} \sin\beta\cos\beta , \\ D^{(2)}_{0-2}(\alpha\beta\gamma) &= \frac{\sqrt{6}}{4} e^{i2\gamma} \sin^2\beta . \end{aligned}\right\} \tag{V.3} $$

$$
\left.
\begin{aligned}
D^{(2)}_{22}(\alpha\beta\gamma) &= \frac{1}{4} e^{-i2\alpha} e^{-i2\gamma} (1+\cos\beta)^2, \\
D^{(2)}_{21}(\alpha\beta\gamma) &= -\frac{1}{2} e^{-i2\alpha} e^{-i\gamma} \sin\beta (1+\cos\beta), \\
D^{(2)}_{20}(\alpha\beta\gamma) &= \frac{\sqrt{6}}{4} e^{-i2\alpha} \sin^2\beta, \\
D^{(2)}_{2-1}(\alpha\beta\gamma) &= -\frac{1}{2} e^{-i2\alpha} e^{i\gamma} \sin\beta (1-\cos\beta), \\
D^{(2)}_{2-2}(\alpha\beta\gamma) &= \frac{1}{4} e^{-i2\alpha} e^{i2\gamma} (1-\cos\beta).
\end{aligned}
\right\} \qquad (V.4)
$$

$$
\left.
\begin{aligned}
D^{(2)}_{-22}(\alpha\beta\gamma) &= \frac{1}{4} e^{i2\alpha} e^{-i2\gamma} (1-\cos\beta)^2, \\
D^{(2)}_{-21}(\alpha\beta\gamma) &= \frac{1}{2} e^{i2\alpha} e^{-i\gamma} \sin\beta (1-\cos\beta), \\
D^{(2)}_{-20}(\alpha\beta\gamma) &= \frac{\sqrt{6}}{4} e^{i2\alpha} \sin^2\beta, \\
D^{(2)}_{-2-1}(\alpha\beta\gamma) &= \frac{1}{2} e^{i2\alpha} e^{i\gamma} \sin\beta (1+\cos\beta), \\
D^{(2)}_{-2-2}(\alpha\beta\gamma) &= \frac{1}{4} e^{i2\alpha} e^{i2\gamma} (1+\cos\beta)^2.
\end{aligned}
\right\} \qquad (V.5)
$$

$$D^{(2)}_{22}(\Phi,\Theta,0)=\frac{1}{4}e^{-i2\Phi}(1+\cos\Theta)^2,$$

$$D^{(2)}_{12}(\Phi,\Theta,0)=\frac{1}{2}e^{-i\Phi}\sin\Theta(1+\cos\Theta),$$

$$D^{(2)}_{02}(\Phi,\Theta,0)=\frac{\sqrt{6}}{4}\sin^2\Theta,$$

$$D^{(2)}_{-22}(\Phi,\Theta,0)=\frac{1}{4}e^{i2\Phi}(1-\cos\Theta)^2.$$

$$D^{(2)}_{-12}(\Phi,\Theta,0)=\frac{1}{2}e^{i\Phi}\sin\Theta(1-\cos\Theta).$$

(V.6)

$$D^{(2)}_{21}(\Phi,\Theta,0)=-\frac{1}{2}e^{-i2\Phi}\sin\Theta(1+\cos\Theta),$$

$$D^{(2)}_{11}(\Phi,\Theta,0)=\frac{1}{2}e^{-i\Phi}(\cos2\Theta+\cos\Theta),$$

$$D^{(2)}_{01}(\Phi,\Theta,0)=\frac{\sqrt{6}}{2}\sin\Theta\cos\Theta,$$

$$D^{(2)}_{-11}(\Phi,\Theta,0)=\frac{1}{2}e^{i\Phi}(\cos\Theta-\cos^2\Theta),$$

$$D^{(2)}_{-21}(\Phi,\Theta,0)=\frac{1}{2}e^{i2\Phi}\sin\Theta(1-\cos\Theta).$$

(V.7)

$$D^{(2)}_{20}(\Phi,\Theta,0)=\frac{\sqrt{6}}{4}e^{-i2\Phi}\sin^2\Theta,$$

$$D^{(2)}_{10}(\Phi,\Theta,0)=-\frac{\sqrt{6}}{2}e^{-i\Phi}\sin\Theta\cos\Theta,$$

$$D^{(2)}_{00}(\Phi,\Theta,0)=1-\frac{3}{2}\sin^2\Theta,$$

$$D^{(2)}_{-10}(\Phi,\Theta,0)=\frac{\sqrt{6}}{2}e^{i\Phi}\sin\Theta\cos\Theta,$$

$$D^{(2)}_{-20}(\Phi,\Theta,0)=\frac{\sqrt{6}}{4}e^{i2\Phi}\sin^2\Theta. \qquad \text{(V.8)}$$

$$D^{(2)}_{2-1}(\Phi,\Theta,0)=-\frac{1}{2}e^{-i2\Phi}\sin\Theta(1-\cos\Theta),$$

$$D^{(2)}_{1-1}(\Phi,\Theta,0)=\frac{1}{2}e^{-i\Phi}(\cos\Theta-\cos2\Theta),$$

$$D^{(2)}_{0-1}(\Phi,\Theta,0)=-\frac{\sqrt{6}}{2}\sin\Theta\cos\Theta \qquad \text{(V.9)}$$

$$D^{(2)}_{-1-1}(\Phi,\Theta,0)=\frac{1}{2}e^{i\Phi}(\cos2\Theta+\cos\Theta),$$

$$D^{(2)}_{-2-1}(\Phi,\Theta,0)=\frac{1}{2}e^{i2\Phi}\sin\Theta(1+\cos\Theta),$$

$$
\left.
\begin{aligned}
D^{(2)}_{2-2}(\Phi,\Theta,0) &= \frac{1}{4}e^{-i2\Phi}(1-\cos\Theta)^2, \\
D^{(2)}_{1-2}(\Phi,\Theta,0) &= -\frac{1}{2}e^{-i\Phi}\sin\Theta(1-\cos\Theta), \\
D^{(2)}_{0-2}(\Phi\Theta 0) &= \frac{\sqrt{6}}{4}\sin^2\Theta, \\
D^{(2)}_{-1-2}(\Phi,\Theta,0) &= -\frac{1}{2}e^{i\Phi}\sin\Theta(1+\cos\Theta), \\
D^{(2)}_{-2-2}(\Phi,\Theta,0) &= \frac{1}{4}e^{i2\Phi}(1+\cos\Theta)^2.
\end{aligned}
\right\} \qquad (V.10)
$$

In the Σ^*, the irreducible field tensor operators have the following relations;

$$
\left.
\begin{aligned}
{}^*V^{(2)}_0 &= -\frac{1}{2}\left(\frac{\partial^2\phi}{\partial^2 z^*}\right), \\
{}^*V^{(2)}_{\pm 1} &= 0, \\
{}^*V^{(2)}_{\pm 2} &= \frac{\sqrt{6}}{6}\eta\,{}^*V^{(2)}_0,
\end{aligned}
\right\} \qquad (V.11)
$$

where η is the field gradient anisotropy and is defined;

$$
\eta = \left[\frac{\partial^2\phi}{\partial^2 x^*} - \frac{\partial^2\phi}{\partial^2 y^*}\right] \Big/ \frac{\partial^2\phi}{\partial^2 z^*}.
$$

By expanding Eq. (V.1), the irreducible components of the field gradient tensor operators can be expressed as a function of η and ${}^{*}V_{o}^{(2)}$ as follows;

$$
\begin{aligned}
{}^{(I)}V_{2}^{(2)} = {}^{*}V_{o}^{(2)}\Big\langle\Big\{ & \frac{\sqrt{6}}{16}\Big[(1+\cos\Theta)^{2} e^{-i2(\Phi+\gamma)} + (1-\cos\Theta)^{2} e^{i2(\Phi+\gamma)}\Big]\sin^{2}\beta + \\
& + \frac{\sqrt{6}}{8}\sin\Theta\Big[(1+\cos\Theta) e^{-i(\Phi+\gamma)} - (1-\cos\Theta) e^{i(\Phi+\gamma)}\Big]\sin 2\beta + \\
& + \frac{\sqrt{6}}{4}\sin^{2}\Theta\left(1-\frac{3}{2}\sin^{2}\beta\right)\Big\} + \\
& + \frac{\sqrt{6}}{6}\eta\Big\{\frac{1}{8} e^{-i2(\Phi+\gamma)}(1+\cos\Theta)^{2}\Big[\cos 2\alpha(1+\cos^{2}\beta) - 2i\cos\beta\sin 2\alpha\Big] + \\
& + \frac{1}{8} e^{i2(\Phi+\gamma)}(1-\cos\Theta)^{2}\Big[\cos 2\alpha(1+\cos^{2}\beta) + 2i\cos\beta\sin 2\alpha\Big] + \\
& + \frac{1}{2} e^{-i(\Phi+\gamma)}\sin\Theta(1+\cos\Theta)\sin\beta\Big[i\sin 2\alpha - \cos\beta\cos 2\alpha\Big] + \\
& + \frac{1}{2} e^{i(\Phi+\gamma)}\sin\Theta(1-\cos\Theta)\sin\beta\Big[i\sin 2\alpha + \cos\beta\cos 2\alpha\Big] + \\
& + \frac{3}{4}\sin^{2}\Theta\sin^{2}\beta\cos 2\alpha\Big\}\Big\rangle\Big\rangle .
\end{aligned}
\qquad \text{(V.12)}
$$

$$
{}^{(I)}V_1^{(2)} = {}^{*}V_0^{(2)}\Big[\Big\{\frac{\sqrt{6}}{8}\sin\Theta\{-(1+\cos\Theta)e^{-i2(\Phi+\gamma)} + (1-\cos\Theta)e^{i2(\Phi+\gamma)}\}\sin^2\beta +
$$

$$
+\frac{\sqrt{6}}{8}\{(\cos 2\Theta+\cos\Theta)e^{-i(\Phi+\gamma)} + (\cos 2\Theta-\cos\Theta)e^{i(\Phi+\gamma)}\}\sin 2\beta +
$$

$$
+\frac{\sqrt{6}}{4}\sin 2\Theta\left(1-\frac{3}{2}\sin^2\beta\right)\Big\} +
$$

$$
+\frac{\sqrt{6}}{6}\eta\Big\{-\frac{1}{4}e^{-i2(\Phi+\gamma)}\sin\Theta(1+\cos\Theta)\{\cos 2\alpha(1+\cos^2\beta) -
$$

$$
-2i\cos\beta\sin 2\alpha\} + \frac{1}{4}e^{i2(\Phi+\gamma)}\sin\Theta(1-\cos\Theta)\{\cos 2\alpha(1+\cos^2\beta) +
$$

$$
+2i\cos\beta\sin 2\alpha\} + \frac{1}{2}e^{-i(\Phi+\gamma)}(\cos 2\Theta+\cos\Theta)\sin\beta\{i\sin 2\alpha -
$$

$$
-\cos\beta\cos 2\alpha\} - \frac{1}{2}e^{i(\Phi+\gamma)}(\cos 2\Theta-\cos\Theta)\sin\beta\{i\sin 2\alpha+\cos\beta\cos 2\alpha\} +
$$

$$
+\frac{3}{4}\sin 2\Theta\sin^2\beta\cos 2\alpha\Big\}\Big]. \qquad \text{(V.13)}
$$

$$
{}^{(I)}V_0^{(2)} = {}^{*}V_0^{(2)}\Big[\Big\{\frac{3}{4}\sin^2\Theta\cos 2(\Phi+\gamma)\sin^2\beta - \frac{3}{4}\sin 2\Theta\cos(\Phi+\gamma)\sin 2\beta +
$$

$$
+\left(1-\frac{3}{2}\sin^2\Theta\right)\left(1-\frac{3}{2}\sin^2\beta\right)\Big\} +
$$

$$
+\frac{\sqrt{6}}{6}\eta\Big\{\frac{\sqrt{6}}{8}e^{-i2(\Phi+\gamma)}\sin^2\Theta\{\cos 2\alpha(1+\cos^2\beta) - 2i\cos\beta\sin 2\alpha\} +
$$

$$
+\frac{\sqrt{6}}{8}e^{i2(\Phi+\gamma)}\sin^2\Theta\{\cos 2\alpha(1+\cos^2\beta) + 2i\cos\beta\sin 2\alpha\} -
$$

$$-\frac{\sqrt{6}}{4}e^{-i(\Phi+\gamma)}\sin 2\Theta\sin\beta[i\sin 2\alpha-\cos\beta\cos 2\alpha]+$$

$$+\frac{\sqrt{6}}{4}e^{i(\Phi+\gamma)}\sin 2\Theta\sin\beta[i\sin 2\alpha+\cos\beta\cos 2\alpha]+$$

$$+\frac{\sqrt{6}}{2}(1-\frac{3}{2}\sin^2\Theta)\sin^2\beta\cos 2\alpha\}\Big]. \qquad (V.14)$$

$$^{(I)}T^{(2)}_{-1}=\ ^{*}T^{(2)}_{0}\Big[\Big\{\frac{\sqrt{6}}{8}\sin\Theta[-(1-\cos\Theta)e^{-i2(\Phi+\gamma)}+(1+\cos\Theta)e^{i2(\Phi+\gamma)}]\sin^2\beta-$$

$$-\frac{\sqrt{6}}{8}[(\cos 2\Theta-\cos\Theta)e^{-i(\Phi+\gamma)}+(\cos 2\Theta+\cos\Theta)e^{i(\Phi+\gamma)}]\sin 2\beta-$$

$$-\frac{\sqrt{6}}{4}\sin 2\Theta(1-\frac{3}{2}\sin^2\beta)\}+$$

$$+\frac{\sqrt{6}}{8}\eta\Big\{-\frac{1}{4}e^{-i2(\Phi+\gamma)}\sin\Theta(1-\cos\Theta)[\cos 2\alpha(1+\cos^2\beta)-2i\cos\beta\sin 2\alpha]+$$

$$+\frac{1}{4}e^{i2(\Phi+\gamma)}\sin\Theta(1+\cos\Theta)[\cos 2\alpha(1+\cos^2\beta)+2i\cos\beta\sin 2\alpha]-$$

$$-\frac{1}{2}e^{-i(\Phi+\gamma)}(\cos 2\Theta-\cos\Theta)\sin\beta[i\sin 2\alpha-\cos\beta\cos 2\alpha]+$$

$$+\frac{1}{2}e^{i(\Phi+\gamma)}(\cos 2\Theta+\cos\Theta)\sin\beta[i\sin 2\alpha+\cos\beta\cos 2\alpha]-$$

$$-\frac{3}{4}\sin 2\Theta\sin^2\beta\cos 2\alpha\}\Big]. \qquad (V.15)$$

$$
{}^{(I)}V_{-2}^{(2)} = {}^{*}V_{0}^{(2)}\Big\{\Big\{\frac{\sqrt{6}}{16}\Big[(1-\cos\Theta)^2 e^{-i2(\Phi+\gamma)} + (1+\cos\Theta)^2 e^{i2(\Phi+\gamma)}\Big]\sin^2\beta -
$$

$$
-\frac{\sqrt{6}}{8}\sin\Theta\Big[(1-\cos\Theta)e^{-i(\Phi+\gamma)} + (1+\cos\Theta)e^{i(\Phi+\gamma)}\Big]\sin 2\beta +
$$

$$
+\frac{\sqrt{6}}{4}\sin^2\Theta\Big(1-\frac{3}{2}\sin^2\beta\Big)\Big\} +
$$

$$
+\frac{\sqrt{6}}{6}\eta\Big\{\frac{1}{8}e^{-i2(\Phi+\gamma)}(1-\cos\Theta)^2\Big[\cos 2\alpha(1+\cos^2\beta) - 2i\cos\beta\sin 2\alpha\Big] +
$$

$$
+\frac{1}{8}e^{i2(\Phi+\gamma)}(1+\cos\Theta)^2\Big[\cos 2\alpha(1+\cos^2\beta) + 2i\cos\beta\sin 2\alpha\Big] -
$$

$$
-\frac{1}{2}e^{-i(\Phi+\gamma)}\sin\Theta(1-\cos\Theta)\sin\beta\Big[i\sin 2\alpha - \cos\beta\cos 2\alpha\Big] -
$$

$$
-\frac{1}{2}e^{i(\Phi+\gamma)}\sin\Theta(1+\cos\Theta)\sin\beta\Big[i\sin 2\alpha + \cos\beta\cos 2\alpha\Big] +
$$

$$
+\frac{3}{4}\sin^2\Theta\sin^2\beta\cos 2\alpha\Big\}\Big\}. \qquad (V.16)
$$

APPENDIX VI. DERIVATIONS OF THE EXPRESSIONS (7.1) AND (7.2)

Using the properties listed in Section VII the function $\langle S_K(V_\tau^{EQ}(R_2))\rangle_T$ as given by Eq. (5.10) can be written as

$$\langle S_K(V_\tau^{EQ}(R_2))\rangle_T = \frac{1}{\hbar}\left(\frac{15\hbar\sqrt{3}e}{4M_c v_s a^3}\right)^2 |\langle j\|Q^{(2)}\|j'\rangle|^2 \sum_{\zeta=-2}^{2} \sum_{K'\neq K} \sum_{mm'}' \times$$

$$\times \sum_{i}^{(n,n)} (j'm', 2\zeta | jm)^2 |\langle jm|K\rangle\langle K|j'm'\rangle|^2 \times$$

$$\times |F(\zeta, \Theta_y, \theta_{oci}, \varphi_{oci})|^2 \left[\{(\hat{\delta}\cdot\hat{a}_{oci})^2\}_{\Omega(\hat{\delta})}\right]^2 \times$$

$$\times P\int_{\omega_{KK'}}^{\omega_M} d\omega \int_0^{\omega_M-\omega_{KK'}} d\omega' \frac{\omega^3\omega'^3 n_T(\omega)[n_T(\omega')+1]}{\omega-\omega'-\omega_{KK'}}, \qquad \text{(VI.1)}$$

where

$$\{(\hat{\delta}\cdot\hat{a}_{oci})^2\}_{\Omega(\hat{\delta})} = \int_{\Omega(\hat{\delta})} (\hat{\delta}\cdot\hat{a}_{oci})^2 d\Omega \qquad \text{(VI.2)}$$

Since phonons obey Bose statistics, the number $n_T(\omega)$ of the phonons of energy $\hbar\omega$ presented in a crystal at temperature T is given by Plank's law

$$n_T(\omega) = \left\{\exp\left(\frac{\hbar\omega}{kT}\right) - 1\right\}^{-1}. \qquad \text{(VI.3)}$$

In the high temperature limit, i.e., $kT \gg \hbar\omega$, we approximate

$$\exp\left(\frac{\hbar\omega}{kT}\right) \simeq 1 + \frac{\hbar\omega}{kT}, \qquad \text{(VI.4)}$$

and

$$\eta_T(\omega) \simeq \frac{kT}{\hbar\omega}. \tag{VI.5}$$

Then Eq. (V.1) can be expressed as

$$\langle S_\tau(V_\tau^{EQ}(R_3))\rangle_T = \frac{1}{\tau}\left(\frac{15kV^3e}{4M_c v^5 a^3}\right)^2 T^2 |\langle j\|Q^{(2)}\|j\rangle|^2 \sum_{\zeta=-2}^{2}\times$$

$$\times\sum_{K'\neq K}\sum_{mm'}\sum_{i=1}^{5}\langle jm'|2\zeta|jm\rangle^2\times$$

$$\times|\langle jm|K\rangle\langle K'|jm'\rangle|^2|F(\zeta,\Theta_y,\varphi_{oci},\varphi_{oci})|^2\times$$

$$\times\left[\left\{(\vec{\zeta}\cdot\vec{a}_{oci})^2-\tfrac{1}{3}(\vec{\zeta})^2\right\}\right]^2 f(\omega_M\omega_{KK'}), \tag{VI.6}$$

where

$$f(\omega_M\omega_{KK'}) = \int_{\omega_{KK'}}^{\omega_M}\omega^2 d\omega\, P\int_0^{\omega_M-\omega_{KK'}}\frac{\omega'^2}{\omega-\omega'-\omega_{KK'}}d\omega'. \tag{VI.7}$$

Letting;

$$\frac{\omega'}{\omega_M} = x,$$

$$\frac{\omega-\omega_{KK'}}{\omega_M} = a,$$

and

$$\frac{\omega_{KK'}}{\omega_M} = b$$

Eq. (VI.7) can be written as

$$f(\omega_M\omega_{KK'}) = \omega_M^2\int_{\omega_{KK'}}^{\omega_M}\omega^2 d\omega \lim_{\epsilon\to 0}\left[\int_0^{a-\epsilon}\frac{x^2dx}{x-a}+\int_{a+\epsilon}^{1-b}\frac{x^2dx}{x-a}\right]. \tag{VI.8}$$

Carrying out the integration in x, Eq. (V.8) can be expressed as

$$f(\omega_M \omega_{KK'}) = I_1 - I_2 - I_3 - I_4 , \tag{VI.9}$$

with

$$I_1 = \omega_M^2 \int_{\omega_{KK'}}^{\omega_M} \omega^2 a^2 \ln a \, d\omega , \tag{VI.10}$$

$$I_2 = \omega_M^2 \int_{\omega_{KK'}}^{\omega_M} \omega^2 a^2 \ln(1-b-a) \, d\omega , \tag{VI.11}$$

$$I_3 = \omega_M^2 \int_{\omega_{KK'}}^{\omega_M} \omega^2 a^2 (1-b) \, d\omega , \tag{VI.12}$$

and

$$I_4 = \frac{\omega_M^2}{2} \int_{\omega_{KK'}}^{\omega_M} \omega^2 (1-b)^2 \, d\omega . \tag{VI.13}$$

The frequency $\omega_{KK'}$ lies in the radio-frequency region and is very small compared to the frequency ω_M of practically all of the lattice oscillators.[18] Then carrying out the integrations of I_1, I_2, I_3, and I_4 and neglecting the terms with higher than the first power in b one has,

$$I_1 = \omega_M^5 \left[\left(\frac{1}{5} - \frac{1}{2} b \right) \ln(1-b) - \frac{1}{25} + \frac{3}{40} b \right] , \tag{VI.14}$$

$$I_2 = \omega_M^5\left[\left(\frac{1}{5}-\frac{1}{2}b\right)\ln(1-b)-\frac{2}{3}+\frac{1}{4}-\frac{1}{25}+\frac{149}{120}b\right], \quad \text{(VI.15)}$$

$$I_3 = \omega_M^5\left(\frac{1}{4}-\frac{7}{12}b\right), \quad \text{(VI.16)}$$

and

$$I_4 = \omega_M^5\left(\frac{1}{6}-\frac{1}{3}b\right). \quad \text{(VI.17)}$$

Combining Eqs. (V.14), (V.15), (V.16), (V.17) and (V.9) one obtains

$$f(\omega_M \omega_{KK'}) = \frac{\omega_M^4 \omega_{KK'}}{4} \quad \text{(VI.18)}$$

Substituting Eq. (V.18) into Eq. (V.6), the function $\langle S_K(V_T^{EQ}(R_2))\rangle_T$ can be expressed as

$$\langle S_K(V_T^{EQ}(R_2))\rangle_T = G_K(R_2)\,T^2, \quad \text{(VI.19)}$$

where

$$G_K(R_2) = \left(\frac{15kV\xi e\omega_M^2}{8\sqrt{\pi}M_c v^5 a^3}\right)^2 |\langle j\|Q^{(2)}\|j\rangle|^2 \sum_{\zeta=-2}^{2} \times$$
$$\times \sum_{K'\neq K}\sum_{mm'}\sum_{i=1}^{5} (jm',2\zeta|jm)^2 \omega_{KK'} \times$$
$$\times |\langle jm|K\rangle\langle K'|jm'\rangle|^2 |F(\zeta\Theta_y\theta_{oci}\varphi_{oci})|^2 \times$$
$$\times \left[\left\{(\hat{\underline{\beta}}\cdot\hat{\underline{a}}_{oci})^2\right\}_{\Omega(\hat{\underline{\beta}})}\right]^2. \quad \text{(VI.20)}$$

Similarly, the function $\langle \frac{h}{2} \Gamma_K(V_T^{EQ}(R_2))\rangle_T$ as given by Eq. (5.11) can be expressed as

$$\left\langle \frac{\hbar}{2}\Gamma_K(V_T^{EQ}(R_2))\right\rangle_T = B_K(R_2)\, T^2 \tag{VI.21}$$

where

$$B_K(R_2) = \left(\frac{\pi \omega_M^5}{5\hbar}\right)\left(\frac{15\hbar V 3e}{4M_c v^5 a^3}\right)^2 |\langle j'\|Q^{(2)}\|j\rangle|^2 \sum_{\zeta=-2}^{2} \times$$

$$\times \sum_{K'\neq K}\sum_{mm'}\sum_{i=1}^{5} (jm';2\zeta|jm)^2 |\langle jm|K\rangle\langle K'|jm'\rangle|^2 \times$$

$$\times |F(\zeta \textcircled{H}_y \theta_{oci}\varphi_{oci})|^2 \left[\left\{(\hat{\underline{\delta}}\cdot\hat{\underline{a}}_{-oci})^2\right\}_{\Omega(\hat{\underline{\delta}})}\right]^2. \tag{VI.22}$$

APPENDIX VII. DERIVATIONS OF THE EXPRESSIONS (7.7) AND (7.8)

The anisotropy function of angular correlation as a function of the rotations of the crystal axes is given by Eq. (6.4);

$$A = \frac{W(\vec{k}_2 | \vec{k}_1; \pi)}{W(\vec{k}_2 | \vec{k}_1; \frac{\pi}{2})} - 1, \qquad \text{(VII.1)}$$

where

$$W(\vec{k}_2 | \vec{k}_1; \pi) = \sum_{\nu_1 \nu_2} \sum_{m m'} A_{\nu_1 \nu_2} \mathrm{III}_{\nu_1 \nu_2}(m m' m m'), \qquad \text{(VII.2)}$$

and

$$W(\vec{k}_2 | \vec{k}_1; \frac{\pi}{2}) = \sum_{\nu_1 \nu_2} \sum_{\substack{m m' \\ m''}} B_{\nu_1 \nu_2}(m m'') \mathrm{III}_{\nu_1 \nu_2}(m m' m'' m'), \qquad \text{(VII.3)}$$

and the functions $A_{\nu_1\nu_2}$, $B_{\nu_1\nu_2}(mm'')$, $\mathrm{III}_{\nu_1\nu_2}(mm'mm')$ and $\mathrm{III}_{\nu_1\nu_2}(mm'm''m')$ are given by Eqs. (6.6), (6.7), (6.8) and (6.9) respectively.

From Eq. (VI.1), one has the condition for $\frac{\partial A}{\partial (H)_y} = 0$ as

$$W(\vec{k}_2 | \vec{k}_1; \tfrac{\pi}{2}) W'(\vec{k}_2 | \vec{k}_1; \pi) - W'(\vec{k}_2 | \vec{k}_1; \tfrac{\pi}{2}) W(\vec{k}_2 | \vec{k}_1; \pi) = 0, \qquad \text{(VII.4)}$$

where the primes denote the first derivatives of the functions with respect to the rotations $(H)_y$ of the crystal axes. Substituting Eqs. (VI.2) and (VI.3) into (VI.4) one has

$$W(\hat{k}_2|\hat{k}_1;\tfrac{\pi}{2})W(\hat{k}_2|\hat{k}_1;\pi) - W(\hat{k}_2|\hat{k}_1;\tfrac{\pi}{2})W(\hat{k}_2|\hat{k}_1;\pi) =$$

$$= \sum_{\nu_1\nu_2}\sum_{\nu_1'\nu_2'}\sum_{mm'}\sum_{\substack{m_1m_1'\\ m_1''}}' A_{\nu_1\nu_2}B_{\nu_1'\nu_2'}(m_1m_1'') \times$$

$$\times\Big\{\mathrm{III}_{\nu_1\nu_2}(mm'mm')\,\mathrm{III}_{\nu_1'\nu_2'}(m_1m_1'm_1''m_1') -$$

$$-\mathrm{III}_{\nu_1'\nu_2'}(m_1m_1'm_1''m_1)\,\mathrm{III}_{\nu_1\nu_2}(mm'mm')\Big] = 0 \,. \qquad \text{(VII.5)}$$

Now one sees clearly that Eq. (VI.5), which gives the conditions for $\dfrac{\partial A}{\partial (H)_y} = 0$, is a very complicated function of $(H)_y$ and no solution can be obtained without actually carrying out the tedious calculations of the summations and the rotational transformations. However, if we make some simplications, for illustrative purposes, a considerable simple solutions for the conditions for $\dfrac{\partial A}{\partial (H)_y} = 0$ can be obtained and these solutions are at least quanlitatively, without carrying out the detail numerical calculations, justified by the well-known experiments by the Zürich group.[8),(9),(20)

In Eqs. (7.5) and (7.6) we let $B_{KK'}(R_2)T^2 = \tau^{EQ^{-1}}$ and τ^{EQ} has a dimension of time and is interpretated as the spin-lattice relaxation time through the electric quadrupole coupling. At room temperature τ^{EQ} is in the order of magnitude of 10^{-1} to 10^{-3} sec.[18),(24)] But the mean life τ_N in a intermediate state of a radioactive nucleus are mostly in the range of 10^{-7} to 10^{-12} sec. Therefore for most cases

we have $\tau^{EQ} >> \tau_N$ or $\frac{\hbar}{2\tau_N} >> \frac{\hbar}{2\tau^{EQ}}$. Then by neglecting the term $\frac{\hbar}{2\tau^{EQ}}$ compared with $\frac{\hbar}{2\tau_N}$ in the attenuation coefficiency and ignoring the rotational dependences of the unitary transformation coefficients the derivatives of $III_{\nu_1\nu_2}(mm'mm')$ and $III_{\nu_1\nu_2}(m_1m_1'm_1''m_1')$ with respect to $(H)_y$ can be calculated as;

$$III'_{\nu_1\nu_2}(mm'mm') = \sum_{KK'} f_{\nu_1\nu_2}(KK'mm') \left(\frac{i\tau_N}{\hbar}\right) \left\{E'_{KK'}(R_1R_2) + \right.$$

and

$$\left. + G'_{KK'}(R_2)T^2\right\} \left\{1 - \frac{i\tau_N}{\hbar}\left(E_{KK'}(R_1R_2) + G_{KK'}(R_2)T^2\right)\right\}^{-2}. \quad (VII.6)$$

$$III'_{\nu_1\nu_2'}(mm'm''m_1) = \sum_{KK'} f_{\nu_1\nu_2'}(KK'mm'm'') \left(\frac{i\tau_N}{\hbar}\right) \left\{E'_{KK'}(R_1R_2) + \right.$$

$$\left. + G_{KK'}(R_2)T^2\right\} \left\{1 - \frac{i\tau_N}{\hbar}\left(E_{KK'}(R_1R_2) + G_{KK'}(R_2)T^2\right)\right\}^{-2}, \quad (VII.7)$$

where

$$f_{\nu_1\nu_2}(KK'mm') = \langle j'm'|K\rangle\langle K|j'm\rangle\langle j'm'|K\rangle\langle K|j'm\rangle \times$$

$$\times (j'm';j'-m'|\nu_1 0)(j'm;j'-m|\nu_2 0), \quad (VII.8)$$

and

$$f_{\nu_1\nu_2'}(KK'm_1m'm'') = \langle j'm_1|K\rangle\langle K|j'm'\rangle\langle j'm''|K'\rangle\langle K'|j'm''\rangle \times$$

$$\times (j'm';j'-m'|\nu_1 0)(j'm'';j'-m''|\nu_2 m-m''), \quad (VII.9)$$

Combining Eqs. (VI.7), (VI.6) and (VI.5) one has;

$$W(\tfrac{\pi}{2})W'(\pi) - W(\pi)W'(\tfrac{\pi}{2}) =$$

$$= \sum_{\nu_1\nu_2} \sum_{\nu_1'\nu_2'} \sum_{mm'} \sum_{\substack{m_1 m_1' \\ m_1''}} \sum_{KK'} \sum_{K_1K_1'} A_{\nu_1\nu_2} \times$$

$$\times B_{\nu_1'\nu_2'}(m_1 m_1'') f_{\nu_1\nu_2}(KK'mm') f_{\nu_1'\nu_2'}(KK'm_1 m_1' m_1'') \times$$

$$\times \left[F_{KK'}(R_1R_2) F_{K_1K_1'}(R_1R_2)\right]^{-2} \left\{F_{K_1K_1'}(R_1R_2) - \right.$$

$$\left. - F_{KK'}(R_1R_2) \frac{F'_{K_1K_1'}(R_1R_2)}{F'_{KK'}(R_1R_2)}\right] F'_{KK'}(R_1R_2) = 0 , \qquad \text{(VII.10)}$$

where

$$F_{KK'}(R_1R_2) = 1 - \left(\frac{i\tau_N}{\hbar}\right)\left[E_{KK'}(R_1R_2) + G_{KK'}(R_2)T^2\right]. \qquad \text{(VII.11)}$$

The functions $F_{K_1K_1'}(R_1R_2)$ and $F_{KK'}(R_1R_2)$ are exactly the same function. By taking their first derivatives with respect to $(H)_y$ are approximately equal Eq. (VII.10) can be expressed as

$$W(\tfrac{\pi}{2})W'(\pi) - W(\pi)W'(\tfrac{\pi}{2}) =$$

$$= \left(\frac{i\tau_N}{\hbar}\right) \sum_{\nu_1\nu_2} \sum_{\nu_1'\nu_2'} \sum_{mm'} \sum_{\substack{m_1 m_1' \\ m_1''}} \sum_{KK'} \sum_{K_1K_1'} A_{\nu_1\nu_2} \times$$

$$\times B_{\nu_1'\nu_2'}(m_1 m_1'') f_{\nu_1\nu_2}(KK'mm') f_{\nu_1'\nu_2'}(K_1K_1'm_1 m_1' m_1'') \times$$

$$\times \left[F_{KK'}(R_1R_2) F_{K_1K_1'}(R_1R_2)\right]^{-2} \left[E_{KK'}(R_1R_2) - E_{K_1K_1'}(R_1R_2) + \right.$$

$$\left. + \left\{G_{KK'}(R_1R_2) - G_{K_1K_1'}(R_1R_2)\right\}T^2\right] F'_{KK'}(R_1R_2) = 0 . \qquad \text{(VII.12)}$$

Equation (VI.12) is still an extremely complicated function of $\textcircled{H}_y$. However, if one examines more carefully the expression one sees that for a given set of the indices the possible vanishing factor is the function $F'_{KK'}(R_1R_2)$. Therefore, by making a simple minded calculations, namely by taking $F'_{KK'}(R_1R_2)$ is zero for all possible indices K, one has the following condition for $\frac{\partial A}{\partial \textcircled{H}_y} = 0$;

$$\sum_{KK'} E_{KK'}(R_1R_2) + \sum_{KK'} G'_{KK'}(R_2) T^2 = 0 \,. \qquad \text{(VII.13)}$$

From Eq. (5.13) and the rotational transformation functions given in the Appendix IV and the symmetric properties of the Clesch-Gordan coefficients the quantity $E_{KK'}(R_1R_2)$ can be calculated as

$$E_{KK'}(R_1R_2) = \sum_{mm'} {}^{*}V_0^{(2)}(0) \langle j' \| Q^{(2)} \| j \rangle \{ \langle j'm'|K\rangle\langle K|j'm\rangle - \langle j'm'|K'\rangle\langle K'|j'm\rangle \} \left[\frac{\sqrt{6}}{2} (j'm'; 22|j'm) - \frac{3}{2}(j'm'; 20|j'm) \right] \sin^2 \textcircled{H}_y \,. \qquad \text{(VII.14)}$$

The function $G_{KK'}(R_2)$ is given by Eq. (7.7) which is still a very complicated function of $\textcircled{H}_y$. However, one realizes that the function $G_{KK'}(R_2)$ describes the effect of the thermal vibrations of the lattice points and these vibrations will in extreme case destroy the axially

symmetric field and becomes independence of $\textcircled{H}_y$. Hence for our simplified calculations we simply replace $G'_{KK'}(\textcircled{H}_y)$ by $G'_{KK'}(0)$.

Combining Eqs. (VI.13), (VI.14) and (7.7) one obtains the following condition for the maximum anisotropy function in the rotational pattern;

$$\left\langle \textcircled{H}_y \right\rangle_{max} = \frac{1}{2}\sin^{-1}\left[\frac{\sum' G'_{KK'}(0)\, T^2}{\sum' A_{KK'}} \right], \qquad \text{(VII.15)}$$

where

$$A_{KK'} = \sum_{mm'}{}' {}^{*}V_0^{(2)}(0) \langle j \| Q^{(2)} \| j \rangle \left[\langle jm'|K\rangle\langle K|jm\rangle - \right.$$

$$- \langle jm'|K\rangle\langle K|jm\rangle \left][\frac{\sqrt{6}}{2}(jm;22|jm) - \right.$$

$$\left. - \frac{3}{2}(jm;20|jm) \right], \qquad \text{(VII.16)}$$

$$G'_{KK'}(0) = 2\left(\frac{15 k \sqrt{3} e \omega_M^2}{8\sqrt{\pi} M_c v^5 a^3} \right)^2 |\langle j \| Q^{(2)} \| j \rangle|^2_x$$

$$\times \sum_{S=-2}^{2} \sum_{mm'} \sum_{i}^{5} (jm;2S|jm)^2 \omega_{KK'} \times$$

$$\times F(\xi, 0, \theta_{oci}, \varphi_{oci}) F'(\xi, 0, \theta_{oci}, \varphi_{oci}) \times$$

$$\times \left[\{ (\vec{\xi} \cdot \vec{a}_{-oci}) \}_{\Sigma(\vec{\xi})} \right] \left[\sum_{K'' \neq K} |\langle jm|K\rangle\langle K''|jm'\rangle|^2 - \right.$$

$$\left. - \sum_{K'' \neq K}{}' |\langle jm|K\rangle\langle K''|jm'\rangle|^2 \right], \qquad \text{(VII.17)}$$

and

$$F(\zeta, \textcircled{H}_y, \theta_{oci}, \varphi_{oci}) = \sum_{\rho}^{1} D^{(2)}_{\rho,-\zeta}(0, \textcircled{H}_y, 0)\, \bar{Y}^{-(2)}_{-\zeta}(\theta_{oci}, \varphi_{oci}). \quad \text{(VII.18)}$$

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