Angular Correlation of Nuclear Radiation

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ABSTRACT

A method is developed for the evaluation of the perturbed angular correlation of two particles successively emitted by nuclei embedded in sources where the interaction between the nuclei and the environment is isotropic on the average. The method can be applied with any value of the correlation time τ_c . It is shown that in the limit of small τ_c , if the source is also homogeneous and the perturbing process is stationary during the nuclear lifetime, the attenuation coefficients $G_k(t)$ are single exponentials, a result which is not restricted to those cases where perturbation theory is applicable. The case of a time dependent hyperfine interaction associated with randomly distributed static quadrupole interactions is studied.

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I. INTRODUCTION

The problem of the angular distribution of particles successively emitted by atomic nuclei has been studied since 1940.¹ Measurements of these distributions have been used not only to obtain nuclear properties but also as a tool for measuring the fields which interact with the radioactive nuclei in solids, liquids and gases. Although the theory of angular correlations is quite well established, there are still many problems to be solved. One of these problems is related to the correlation time of randomly varying time dependent interactions. By correlation time we mean a characteristic time interval τ_c such that the directions of the fields which interact with the nucleus at a time t_1 may be considered to be uncorrelated with the directions at a time t_2 , whenever $|t_2 - t_1| > \tau_c$. A mathematical definition of τ_c is given in first order perturbation theory by Abragam and Pound.² The time dependence of the angular correlation $W(\theta, t)$ of two radiated particles is well known in the limits of $\tau_c \ll t$ and $\tau_c \rightarrow \infty$ (static interaction) but no general theory exists for the cases where τ_c is comparable with the time interval t between the two emissions. Some models have been developed,^{3,4,5} but all of them have very limited ranges of applicability. Even in the limit of $\tau_c \ll t$ there are still some questions to be answered. For example, Abragam and Pound, using first order perturbation theory for isotropic sources, have shown that the attenuation coefficients $G_k(t)$ are given by single exponentials, namely

$$G_k(t) = e^{-\lambda_k t} \quad (1)$$

where λ_k are parameters depending on the characteristics of the interactions. In 1962 Dillenburg and Maris⁶ pointed out that in certain cases some of the individual interactions may be so strong that perturbation theory is not applicable. They found that in these cases the attenuation coefficients

are given by a sum of exponentials. Nevertheless, no experimental confirmation of this effect has so far been found. In Sec. IV-A we give an answer to this dilemma.

In the present paper we develop a method for calculating the attenuation coefficients $G_k(t)$, applicable for sources where the interaction between the nuclei and their environments are isotropic on the average. No restriction on the value of τ_c and on the strength of the interactions is made, except in some of the applications in Sec. IV.

II. DEFINITIONS AND BASIC RELATIONS

Since we will be concerned throughout this paper with sources which are isotropic on the average, the general attenuation coefficients $G_{k_1 k_2}^{N_1 N_2}(t)$ of the angular correlation will always have $N_1 = N_2 = 0$ (see Ref. 2). Its general form is given by⁷

$$G_{k_1 k_2}^{00}(t) = \sum_{mm'} (-)^{2I+m+m'} [(2k_1 + 1)(2k_2 + 1)]^{1/2} \times \begin{pmatrix} I & I & k_1 \\ m & -m & 0 \end{pmatrix} \begin{pmatrix} I & I & k_2 \\ m' & -m' & 0 \end{pmatrix} P_{mm'}(t, 0) \quad (2)$$

where I is the spin of the intermediate nuclear state and $P_{mm'}(t, 0)$ is the conditional probability for finding the nucleus in the sub-state m at time t if it was in the sub-state m' at $t = 0$ (immediately after the first emission). In the notation of Ref. 7 we would have $|\langle m | \Lambda(t) | m' \rangle|^2$ instead of $P_{mm'}(t, 0)$, but we prefer the latter notation because we intend to extend our treatment to cases where the quantum mechanical system is not the nucleus by itself, but the nucleus plus the environment. In these cases an evolution operator $\Lambda(t)$ for the nuclear states may not exist. Even the quantities $P_{mm'}(t, 0)$ only exist in some cases. We will show in

the following section that they exist when the nuclear environment is isotropic on the average. The expressions for $P_{mm'}(t,0)$ will be shown to have the form

$$P_{mm'}(t,0) = \sum_k g_k(t) (-)^{m-m'} (2k+1) \begin{pmatrix} I & I & k \\ m & -m & 0 \end{pmatrix} \begin{pmatrix} I & I & k \\ m' & -m' & 0 \end{pmatrix} \quad (3)$$

Substituting Eq. (3) into Eq. (2) and performing the sums over m and m' one gets

$$G_{k_1 k_2}^{00}(t) = \delta_{k_1 k_2} g_k(t) \quad (4)$$

which shows that the angular correlation can be written in the form

$$W(\theta, t) = \sum_k A_k(1) A_k(2) G_k(t) P_k(\cos \theta) \quad (5)$$

where

$$G_k(t) = g_k(t) \quad (6)$$

and $A_k(1)A_k(2)$ are the usual expansion coefficients of the unperturbed angular correlation $W(\theta)$ in Legendre polynomials.

To calculate the conditional probabilities $P_{mm'}(t,0)$ we will study the time behavior of the density matrix $\rho(t)$ of the entire quantum mechanical system, which consists of "nucleus + environment". The density matrix $\rho^N(t)$ of the ensemble of nuclei is given in terms of $\rho(t)$ by⁸

$$\rho^N(t) = \text{Tr } \rho(t) \quad (7)$$

where the trace is taken over all the variables of the environment. This matrix will be shown to be diagonal in the nuclear magnetic quantum numbers m , i.e.,

$$\rho_{mm'}^N(t) = \delta_{mm'} \rho_{mm}^N(t). \quad (8)$$

Therefore, the quantity

$$P_m(t) = \rho_{mm}^N(t) \quad (9)$$

may be interpreted as the probability for finding the nucleus in the sub-state m at time t . The probability distributions $P_m(t)$ and $P_m(t_0)$, $t_0 < t$, are clearly related by

$$P_m(t) = \sum_{m'} P_{mm'}(t, t_0) P_{m'}(t_0) \quad (10)$$

which extends the concept of the conditional probability, introduced in Eq. (2), to non-zero values of the second argument t_0 .

Some properties of $P_{mm'}(t, t_0)$ are:

- a) $P_{mm'}(t_0, t_0) = \delta_{mm'}$, from its definition.
- b) $\sum_m P_{mm'}(t, t_0) = 1$, from probability conservation.
- c) $P_{-m, -m'}(t, t_0) = P_{m, m'}(t, t_0)$, from parity conservation.
- d) $P_{mm'}(t, t_0) = P_{m'm}(t, t_0)$, from time-reversal invariance.
- e) If τ_c is finite, then $P_{mm'}(t_0 + t', t_0) \xrightarrow{t' \rightarrow \infty} \frac{1}{2I + 1}$, since the isotropy of the source causes equi-population of the sub-states m after a sufficiently long time.
- f) $P_{mm'}(t, t_0) = \sum_{m''} P_{mm''}(t, t_1) P_{m''m'}(t_1, t_0)$, $t_0 \leq t_1 \leq t$, from conservation of probability.

III. THE SMALL NEIGHBORHOOD METHOD

In this section we will study the evolution of the nuclear spins during the time interval between the two radiations, i.e., from $t = 0$ to the time t when the second radiation occurs. Since it is not always possible to decouple the nucleus from its surroundings and consider it as a quantum mechanical system by itself, moving under the influence of external potentials, we define a larger system in the following way: Select a small amount of matter around the nucleus and call it the "the neighborhood". The nucleus plus the neighborhood will be considered a quantum mechanical "system". The influence of the rest of the source on the "system" will be taken as randomly varying potentials. In doing so we are neglecting the

"feed-back" effects on the system resulting from the influence of its movement on the rest of the source. Thus the neighborhood has to be chosen in such a way that this effect is negligible. This will be a restriction only when we intend to apply the method developed here in obtaining numbers for the attenuation coefficients $G_k(t)$, because then the neighborhood has to be chosen so small that it is possible to take into account explicitly all the relevant quantum numbers. In the present section it will be left unspecified. In particular, it can be the whole source and the properties derived here are then completely general for an isotropic source.

In what follows two different sets of basis functions will be used as representations for the density matrix $\rho(t)$ of the ensemble of systems. One of them is the set of state-vectors $|AIJFM\rangle$ (the coupled representation), where I is the nuclear spin, J is the angular momentum of the neighborhood, F is the total angular momentum of the system, M is its z -component and A stands for all other quantum numbers of the system. The other representation is the set of state-vectors $|AI m J n\rangle$ (the uncoupled representation), where m and n are I_z and J_z respectively.

It is well known that at $t = 0$ there exists a probability distribution $P_m(0)$ for the nuclear sub-states m , i.e., $\rho(0)$ is diagonal in m , as long as the z -axis is chosen in the direction of the first radiation and its transversal polarization is not measured. On the other hand, $\rho(0)$ is also diagonal in n because of the isotropy of the source, and its diagonal elements are independent of n . Thus we can write

$$\langle AI m J n | \rho(0) | A' I m' J' n' \rangle = \delta_{mm'} \delta_{nn'} P_m(0) f_{AJ, A'J'}(0) \quad (11)$$

where $f_{AJ, A'J'}(0)$ is independent of m and n and is clearly the trace over m of $\rho(0)$, representing, therefore, a density matrix element for the ensemble of

neighborhoods at $t = 0$. Since any density matrix can be expanded as sums of irreducible spherical tensors, the Wigner-Eckart theorem can be used to show that, being diagonal in n with diagonal elements independent of n , $\rho(0)$ is also diagonal in J . Thus Eq. (11) can be written as

$$\langle A'ImJn | \rho(0) | A'Im'J'n' \rangle = \delta_{mm'} \delta_{nn'} \delta_{JJ'} P_m(0) f_{AJ,A'J}(0) \quad (12)$$

A basis transformation of Eq. (12) to the coupled representation leads to

$$\langle A'IJFM | \rho(0) | A'IJ'F'M' \rangle = \sum_{mn} \langle ImJn | FM \rangle \langle Im'J'n' | F'M' \rangle P_m(0) f_{AJ,A'J}(0) \delta_{JJ'} \quad (13)$$

where the symbols in brackets are Clebsch-Gordan coefficients.

Our next task is to see how the matrix elements of $\rho(t)$ are related to those of $\rho(0)$. For this purpose we divide the ensemble of systems into sub-ensembles, which we label by σ , in the following way: two systems are in the same sub-ensemble σ if the sequence of interactions between each of them and the rest of the source is equal, except for an overall rotation. In other words, the evolution operator of two elements in the same sub-ensemble σ can be identified through a convenient rotation. We will denote by $U^\sigma(t,0;\alpha\beta\gamma)$ the evolution operator from 0 to t of the element for which a rotation through the Euler angles α , β and γ has to be made to identify it with a "reference" element whose evolution operator is denoted by $U^\sigma(t,0;000)$. In other words,

$$U^\sigma(t,0;\alpha\beta\gamma) = R^\dagger(\alpha\beta\gamma) U^\sigma(t,0;000) R(\alpha\beta\gamma) \quad (14)$$

Due to the isotropy of the source, any set of Euler angles $\alpha\beta\gamma$ occurs the same number of times as any other set, and we can write

$$\rho^\sigma(t) = \frac{1}{8\pi^2} \int_0^{2\pi} d\gamma \int_0^{2\pi} d\alpha \int_0^\pi \sin \beta d\beta \{ U^\sigma(t,0;\alpha\beta\gamma) \rho(0) U^{\sigma\dagger}(t,0;\alpha\beta\gamma) \} \quad (15)$$

for the density matrix of the sub-ensemble σ at time t . Inserting identity

operators between U^σ , ρ and $U^{\sigma\dagger}$ in Eq. (15) and taking the matrix elements of $\rho(t)$ between states of the coupled representation one obtains

$$\begin{aligned} \langle A_1 I J_1 F_1 M_1 | \rho^\sigma(t) | A_2 I J_2 F_2 M_2 \rangle &= \Sigma \frac{1}{8\pi^2} \int d\Omega \{ \langle A_1 I J_1 F_1 M_1 | U^\sigma(t, 0; \alpha\beta\gamma) | A I J F M \rangle \\ &\times \langle A I J F M | \rho(0) | A' I J' F' M' \rangle \langle A' I J' F' M' | U^{\sigma\dagger}(t, 0; \alpha\beta\gamma) | A_2 I J_2 F_2 M_2 \rangle \} \quad (16) \end{aligned}$$

where $\Omega \equiv \alpha\beta\gamma$ and the sum is over $AA'JJ'FF'MM'$. Using Eqs. (13) and (14), Eq. (16) becomes

$$\langle A_1 I J_1 F_1 M_1 | \rho^\sigma(t) | A_2 I J_2 F_2 M_2 \rangle = \Sigma \langle I m J n | F M \rangle \langle I m J n | F' M' \rangle P_m(0) f_{AJ, A'J}(0) I^\sigma \quad (17)$$

where

$$\begin{aligned} I^\sigma &= \frac{1}{8\pi^2} \int d\Omega \langle A_1 I J_1 F_1 M_1 | R^\dagger(\alpha\beta\gamma) U^\sigma(t, 0) R(\alpha\beta\gamma) | A I J F M \rangle \\ &\times \langle A' I J' F' M' | R^\dagger(\alpha\beta\gamma) U^{\sigma\dagger}(t, 0) R(\alpha\beta\gamma) | A_2 I J_2 F_2 M_2 \rangle \quad (18) \end{aligned}$$

and

$$U^\sigma(t, 0) \equiv U^\sigma(t, 0; 000)$$

The "vectors" $R(\alpha\beta\gamma) | A I J F M \rangle$ can be written as linear combinations of the basis vectors $| A I J F M \rangle$ in the form

$$R(\alpha\beta\gamma) | A I J F M \rangle = \Sigma_\mu D_{\mu M}^F(\alpha\beta\gamma) | A I J F \mu \rangle. \quad (19)$$

The integral (18) can be performed by using the well known integration formulas for the D matrices. Substituting the result into Eq. (17) one obtains, after some algebra,

$$\begin{aligned} \langle A_1 I J_1 F_1 M_1 | \rho^\sigma(t) | A_2 I J_2 F_2 M_2 \rangle &= \delta_{M_1 M_2} \Sigma \{ (-)^{J+m-M_2+\mu-\mu_1} P_m(0) f_{AJ, A'J}(0) \\ &\times (2k+1)((2F+1)(2F'+1))^{1/2} \begin{Bmatrix} I & I & k \\ F & F' & J \end{Bmatrix} \begin{Bmatrix} I & I & k \\ m & -m & 0 \end{Bmatrix} \begin{Bmatrix} F_1 & F_2 & k \\ M_2 & -M_2 & 0 \end{Bmatrix} \\ &\times \begin{Bmatrix} F & F' & k \\ -\mu & \mu' & \mu-\mu' \end{Bmatrix} \begin{Bmatrix} F_1 & F_2 & k \\ \mu_1 & -\mu_2 & \mu_2-\mu_1 \end{Bmatrix} \cdot \langle A_1 I J_1 F_1 \mu_1 | U^\sigma(t, 0) | A I J F \mu \rangle \\ &\times \langle A_2 I J_2 F_2 \mu_2 | U^\sigma(t, 0) | A' I J' F' \mu' \rangle^* \} \quad (20) \end{aligned}$$

where the sum is over all the variables which do not appear in the L.H.S.

Transforming Eq. (20) back to the uncoupled representation and taking the trace over the variables of the neighborhood (see Eq. (7)) one obtains:

$$\rho_{m_1 m_2}^{N\sigma}(t) = \delta_{m_1 m_2} \sum_{km} g_k^\sigma(t) P_m(0) \alpha_{mm_1}^k \quad (21)$$

with

$$\alpha_{mm_1}^k = (-)^{m-m_1} (2k+1) \begin{pmatrix} I & I & k \\ m & -m & 0 \end{pmatrix} \begin{pmatrix} I & I & k \\ m_1 & -m_1 & 0 \end{pmatrix} \quad (22)$$

and

$$\begin{aligned} g_k^\sigma(t) = & \sum (-)^{J-J_1+\mu-\mu_1} f_{AJ,A',J}(0) [\hat{F}\hat{F}'\hat{F}_1\hat{F}_2]^{1/2} \begin{Bmatrix} I & I & k \\ F & F' & J \end{Bmatrix} \begin{Bmatrix} I & I & k \\ F_1 & F_2 & J_1 \end{Bmatrix} \\ & \times \begin{pmatrix} F & F' & k \\ -\mu & \mu' & \mu-\mu' \end{pmatrix} \begin{pmatrix} F_1 & F_2 & k \\ -\mu_1 & \mu_2 & \mu_1-\mu_2 \end{pmatrix} \langle A_1 I J_1 F_1 \mu_1 | U^\sigma(t,0) | A I J F \mu \rangle \\ & \times \langle A_1 I J_1 F_2 \mu_2 | U^\sigma(t,0) | A' I J F' \mu' \rangle^* \end{aligned} \quad (23)$$

where the sum in Eq. (23) is over all the variables except I and k and where $\hat{F}$ means $(2F+1)$.

Since $\rho^{N\sigma}(t)$ is diagonal in m for any sub-ensemble σ , its ensemble average

$$\rho^N(t) = \langle \rho^{N\sigma}(t) \rangle_\sigma \quad (24)$$

also has this property, as was stated in the preceding section. Thus we can write

$$P_{m_1}^N(t) = \rho_{m_1 m_1}^{N\sigma}(t) = \langle \rho_{m_1 m_1}^{N\sigma}(t) \rangle_\sigma = \sum_{km} \langle g_k^\sigma(t) \rangle_\sigma \alpha_{mm_1}^k P_m(0) \quad (25)$$

From Eqs. (10) and (25) one obtains

$$P_{mm_1}(t,0) = \sum_k \langle g_k^\sigma(t) \rangle_\sigma \alpha_{mm_1}^k \quad (26)$$

which, according to Eqs. (3), (6) and (22) leads to

$$G_k(t) = \langle g_k^\sigma(t) \rangle_\sigma \quad (27)$$

In conclusion, we may say that Eq. (23) provides us with a means for calculating the conditional probabilities $P_{mm_1}(t,0)$ and the attenuation factors $G_k(t)$ in isotropic sources. To perform such a calculation we need

to know the density matrix for the neighborhood $f_{AJ,A'J}(0)$ at time $t = 0$ and the evolution operators $U^\sigma(t,0)$ for the arbitrarily chosen "reference" elements. The contribution of the other elements was already taken into account in the derivation of Eq. (23), namely, in performing the average given by Eq. (15). Applications to actual physical situations requires the construction of suitable models. Some of these will be discussed in the next section.

IV. APPLICATIONS

A) The Limit $\tau_c \ll t$

In this section we will consider sources which besides being isotropic on the average, are also homogeneous and stationary, and where the correlation time τ_c is much smaller than the time interval t between the two emissions. By homogeneous we mean that all the nuclei are subjected during their lifetimes to the same sequence of interactions, except for statistical deviations. We exclude, for example, sources where identical intermediate nuclei are contained in different chemical compounds. By stationary we mean that in a time interval $\Delta t \gg \tau_c$, the average sequence of interactions undergone by the nucleus is the same, regardless of whether Δt is chosen at the beginning or at the end of the lifetime. We exclude, for example, the case of atoms recoiling into a gas for which the average time interval between collisions is increasing as the atom is slowing down.

Since $t \gg \tau_c$ we can define a time interval Δt such that

$$\tau_c \ll \Delta t < t \quad (28)$$

From the stationarity of the source and Eq. (26) we have

$$P_{mm'}(t, t_0) = \sum_k \langle g_k^\sigma(t - t_0) \rangle_\sigma \alpha_{mm'}^k, \quad (29)$$

for any t_0 between 0 and t . Using Eqs. (28) and (29) and property (f) of the conditional probabilities (Sec. II) one sees that:

$$P_{mm'}(2\Delta t, 0) = \sum_{m''} P_{mm''}(2\Delta t, \Delta t) P_{m''m'}(\Delta t, 0) = \sum_{kk'm''} \langle g_k^\sigma(\Delta t) \rangle_\sigma \langle g_{k'}^\sigma(\Delta t) \rangle_\sigma \alpha_{mm''}^k \alpha_{m''m'}^{k'}$$

The sum over m'' can be performed by use of the definition of $\alpha_{mm''}^k$, Eq. (22), leading to

$$P_{mm'}(2\Delta t, 0) = \sum_k (\langle g_k^\sigma(\Delta t) \rangle_\sigma)^2 \alpha_{mm'}^k \quad (30)$$

By the same procedure we obtain

$$P_{mm'}(s\Delta t, 0) = \sum_k (\langle g_k^\sigma(\Delta t) \rangle_\sigma)^s \alpha_{mm'}^k \quad (31)$$

Taking $s\Delta t = t$ and comparing the above equation with Eqs. (3) and (6) one sees that

$$G_k(t) = (\langle g_k^\sigma(\Delta t) \rangle_\sigma)^{t/\Delta t} \quad (32)$$

Defining

$$\lambda_k = -\frac{1}{\Delta t} \ln(\langle g_k^\sigma(\Delta t) \rangle_\sigma), \quad (33)$$

Eq. (32) becomes identical to Eq. (1), which is the classical result obtained by Abragam and Pound² by the use of first order perturbation theory. We understand that the above demonstration answers the question of whether the attenuation coefficients $G_k(t)$ are always single exponentials in the limit $\tau_c \ll t$ or whether they may be a sum of exponentials. Our answer is: they are single exponentials whenever the source is isotropic, homogeneous and stationary. We point out that, since we did not need to calculate explicitly the functions $g_k^\sigma(\Delta t)$ to obtain this result, no use was made of the smallness of the neighborhood. Thus our quantum mechanical system may be the whole source, which renders the demonstration completely general. This result does not imply, of course, that what was done in Ref. 6 is wrong, but rather, it indicates that the symmetry properties of the source have not been explored far enough to reduce the sum of exponentials to a single term. We also point out that the

existence of a hard core in Blume's model³ is not in contradiction with the present result, since in that model the direction of the perturbing field at a given nuclear site is constant, the fluctuations being only in its sign. Thus, recalling our definition of τ_c (see Introduction), the present demonstration is not applicable to it.

B) A Gas Model

Recently we have published a paper⁵ where the functions $G_k(t)$ are calculated for a monoatomic gas model, which is valid for any value of the correlation time τ_c . We will show here how the same results are derivable from the equations obtained in Sec. III.

Two hypotheses define the model:

- 1) The only relevant interactions between the atoms in the gas are the collisions, i.e. there are no interatomic correlations such as long distance magnetic or electric interactions.
- 2) Each collision is an event of very small duration compared to the average time interval between collisions and its only effect on the colliding atom is that of rendering the direction of the electronic angular momentum $\vec{J}$ (whose magnitude is supposed constant) completely random.

The second hypothesis allows us to say that the correlation time τ_c corresponds to the average time interval between collisions. The first hypothesis allows us to treat the atom as a free system during the time interval between collisions, having a constant angular momentum $\vec{F}$. Denoting by $\sigma = 0$ the sub-ensemble of atoms which don't undergo any collision between 0 and t , its evolution operator is

$$U^0(t,0) = e^{-i/\hbar H t} \quad (34)$$

where H is the Hamiltonian, having eigenvectors $|IJFM\rangle$ and eigenvalues

E_F independent of M . Using Eq. (34) and

$$f_{AJ,A'J}(0) = \frac{1}{2J+1} \quad (35)$$

in Eq. (23) one obtains

$$g_k^0(t) = \sum_{FF'} \frac{(2F+1)(2F'+1)}{2J+1} \left\{ \frac{I}{F} \frac{I}{F'} \frac{k}{J} \right\}^2 e^{-i\omega_{FF'}t} \quad (36)$$

where

$$\omega_{FF'} = 1/\hbar (E_F - E_{F'}).$$

Eq. (36) agrees with the known result for free atoms.⁷

The conditional probabilities for this sub-ensemble is given, according to Eq. (26), by

$$P_{mm'}^0(t,0) = \sum_k g_k^0(t) \alpha_{mm'}^k \quad (37)$$

Let $\sigma = (1, t_1)$ be the sub-ensemble in which one collision occurs between 0 and t , at time t_1 . The atom is free during the open intervals $(0, t_1)$ and (t_1, t) . Thus we can write, by hypothesis (2),

$$P_{mm'}^1(t,0;t_1) = \sum_{m''} P_{mm''}^0(t,t_1) P_{m''m'}^0(t_1,0) = \sum_k g_k^0(t-t_1) g_k^0(t_1) \alpha_{mm'}^k \quad (38)$$

Similarly, for the sub-ensemble $\sigma = (s; t_1, t_2 \dots t_s)$ for which s collisions occur at times $t_1, t_2 \dots t_s$, one obtains

$$P_{mm'}^s(t,0;t_1, t_2 \dots t_s) = \sum_k g_k^0(t-t_s) g_k^0(t_s-t_{s-1}) \dots g_k^0(t_1) \alpha_{mm'}^k \quad (39)$$

The conditional probabilities for the whole ensemble is the average of Eq. (39) over the number s of collisions and over the times $t_1, t_2 \dots t_s$. Equivalently,

$$\begin{aligned} G_k(t) &= \langle g_k(t) \rangle_\sigma = \langle g_k^s(t; t_1 \dots t_s) \rangle_{s; t_1 \dots t_s} = \\ &= \langle g_k^0(t-t_s) g_k^0(t_s-t_{s-1}) \dots g_k^0(t_1) \rangle_{s; t_1 \dots t_s} \quad (40) \end{aligned}$$

To perform this average we suppose that the probability per unit

time for the occurrence of a collision is $\frac{1}{\tau_c}$, independent of time. From this it follows that the probability for the j^{th} collision to occur at time t_j in dt_j if the $(j-1)^{\text{th}}$ collision had occurred at time t_{j-1} is

$$\frac{1}{\tau_c} \exp(-(t_j - t_{j-1})/\tau_c) \theta(t_j - t_{j-1}) dt_j$$

where the step function $\theta(t_j - t_{j-1})$ is introduced to ensure that the j^{th} collision does not occur before the $(j-1)^{\text{th}}$.

Clearly, the probability for the first collision to occur at time t_1 in dt_1 , the second at time t_2 in $dt_2 \dots$, the s^{th} at time t_s in dt_s and none between t_s and t ($t \geq t_s$) is

$$\begin{aligned} W^s(t, 0; t_1 \dots t_s) &= \frac{1}{\tau_c} e^{-t_1/\tau_c} \theta(t_1) \cdot \frac{1}{\tau_c} e^{-(t_2 - t_1)/\tau_c} \theta(t_2 - t_1) \dots \frac{1}{\tau_c} \\ &\cdot e^{-(t_s - t_{s-1})/\tau_c} \theta(t_s - t_{s-1}) e^{-(t - t_s)/\tau_c} dt_1 dt_2 \dots dt_s \\ &= \frac{1}{(\tau_c)^s} e^{-t/\tau_c} \theta(t_1) \theta(t_2 - t_1) \dots \theta(t - t_s) dt_1 \dots dt_s \end{aligned} \quad (41)$$

Eq. (40) becomes

$$\begin{aligned} G_k(t) &= \sum_{s=0}^{\infty} \int_0^t dt_1 \int_0^t dt_2 \dots \int_0^t dt_s W^s(t, 0; t_1 \dots t_s) g_k^s(t; t_1, t_2 \dots t_s) = \\ &= \sum_{s=0}^{\infty} \frac{e^{-t/\tau_c}}{(\tau_c)^s} \int_0^t dt_1 \int_{t_1}^t dt_2 \dots \int_{t_{s-1}}^t dt_s \{g_k^0(t_1) g_k^0(t_2 - t_1) \dots g_k^0(t_s - t_{s-1}) g_k^0(t - t_s)\} \end{aligned} \quad (42)$$

which, except for a trivial transformation of the integration variables, is identical to Eq. (4) of Ref. (9). Explicit calculations of $G_k(t)$ were done⁵ and the integrations in Eq. (42) have been performed⁹ by the use of Laplace transforms. The numerical results show that the plots of $G_k(t)$ versus t for various values of τ_c change smoothly from the limit $\tau_c \ll t$

to the limit $\tau_c \rightarrow \infty$, agreeing in these limits with the known results.^{2,7}

In particular, for an interaction Hamiltonian of the form

$$H = \hbar \omega_J \vec{I} \cdot \vec{J} \quad (43)$$

the parameters λ_k (see Eq. (1)) corresponding to the limit $\tau_c \ll t$ can be obtained from Eq. (42). They are

$$\lambda_k = \frac{1}{3} \omega_J^2 \tau_c J(J+1) k(k+1) \quad (44)$$

The same result was obtained by Abragam and Pound² for paramagnetic ions in crystals. In this case the relaxation of the electronic spin may be due to the lattice vibrations or to the scattering of the conduction electrons by the ion. It is interesting to note that in this limit it makes no difference whether the relaxation of the electronic spin is done discontinuously by collisions (or by electronic scattering), or smoothly by lattice vibrations, as long as we define correctly the correlation time τ_c .

C) Time Dependent Hyperfine and Static Quadrupole Interactions in Solids

Angular correlation experiments¹⁰ done by implantation of rare earth ions into nonmagnetic metals have given values of $G_k(t)$ which can be roughly approximated by single exponentials. Attempts to interpret this result as due to a pure time dependent hyperfine interaction $\vec{I} \cdot \vec{J}$ are not completely satisfactory in that λ_4/λ_2 is 10/3 for this interaction while the experiments give λ_4/λ_2 close to 2. The influence of the associated time dependent quadrupole interaction is far too small to account for this difference. It has been suggested¹¹ that a very small static electric field gradient could be present, which would account for the difference. In the absence of a theory to take this randomly distributed static perturbation into account some authors^{12,13} have simply multiplied the coefficients $G_k(t)$ corresponding to pure random static quadrupole interactions (we call them

$G_k^S(t)$) by the ones corresponding to pure time dependent hyperfine interactions (we call them $G_k^H(t)$), i.e.,

$$G_k(t) = G_k^H(t) G_k^S(t) \quad (45)$$

The results obtained in this way give reasonably good fits^{10,12} to the experimental data, but there is no good theoretical support for assumption (45). We will derive an expression for $G_k(t)$ using the "small neighborhood method" introduced in Sec. III.

We consider the ion containing the intermediate nucleus to be the quantum mechanical system. The electronic shell is then the "neighborhood", whose only relevant quantum numbers are supposed to be the angular momentum J and its projection n on the z -axis. In most actual solids the correlation time τ_c for the relaxation of the electronic angular momentum direction is a very small quantity ($\approx 10^{-12}$ sec) compared to the resolution time of the measuring apparatus. Thus we will take for granted the condition $\tau_c \ll t$. On the other hand, we saw in Sec. IV-B that in this limit it makes no difference whether the relaxation of $\vec{J}$ is done continuously or discontinuously. Thus, for simplicity, we consider it to be done by the scattering of the conduction electrons which renders the direction of $\vec{J}$ completely random. The magnitude J of $\vec{J}$ will be taken as constant. The influence of the small static field gradient on the electronic wave functions is neglected, i.e., we suppose the static quadrupole term K in the Hamiltonian to operate only on the nuclear variables. The total Hamiltonian will be written in the form

$$H(t) = H(t) + K \quad (46)$$

where $H(t)$ is the time dependent hyperfine interaction, which may contain a magnetic term $\vec{I} \cdot \vec{J}$ and a time dependent electric quadrupole term.

To evaluate Eq. (23) we proceed in a way similar to that for the gas model. First, consider the sub-ensemble for which the first scattering

occurs at time t_1 and for which the static field gradient will be labeled by v , or, equivalently, $K = K^v$. Remember that in the derivation of Eq. (23) we defined the evolution operator $U^\sigma(t,0;\alpha\beta\gamma)$ in terms of a "reference element" in the sub-ensemble σ , having evolution operator $U^\sigma(t,0;000)$. In the present case we take the "reference element" to be one for which the static electric field gradient is in the z -direction. We also suppose axial symmetry for the field gradient, i.e., the vectors $|ImJn\rangle$ are eigenvectors of K^v , with eigenvalues⁷

$$E_m^v = (3m^2 - I(I+1)) \omega_Q^v, \quad (47)$$

where the quadrupole frequency ω_Q^v is given by

$$\omega_Q^v = \frac{eQV_{zz}^v}{4I(2I-1)\hbar} \quad (48)$$

Since we will restrict our treatment to very small values of the quadrupole frequencies ω_Q^v , this seems to be a reasonable approximation even for the cases where the field gradient is not axially symmetric.

For times $t < t_1$, the evolution operator of the reference element is

$$U^{0,v}(t,0) = e^{-i/\hbar (H+K^v)t} \quad (49)$$

Since the operators H and K^v do not commute, we will approximate Eq. (49) by

$$U^{0,v}(t,0) = 1/2 (e^{-i/\hbar Ht} e^{-i/\hbar K^v t} + e^{-i/\hbar K^v t} e^{-i/\hbar Ht}) \quad (50)$$

Eq. (50) is equal to Eq. (49) to second order in $\frac{1}{\hbar} (H + K^v)t$. A discussion of this approximation will be made below Eq. (60).

For the matrix elements in Eq. (23) one obtains

$$\begin{aligned} \langle IJF\mu | U^{0,v}(t,0) | IJF_1\mu_1 \rangle &= \frac{1}{2} \{ (e^{-i/\hbar E_F t} + e^{-i/\hbar E_{F_1} t}) \\ &\quad \cdot \sum_{mn} \langle ImJn | F\mu \rangle \langle ImJn | F_1\mu_1 \rangle e^{-i/\hbar E_m^v t} \} \quad (51) \end{aligned}$$

where E_F and E_m^v are the eigenvalues of H and K^v with eigenvectors $|IJFM\rangle$ and $|ImJn\rangle$ respectively. Substituting Eq. (51) into Eq. (23) and using

$f_{AJ,A,J}(0) = \frac{1}{2J+1}$ one obtains

$$g_k^{o,v}(t) = \sum \left\{ \frac{1}{2J+1} \hat{F}\hat{F}'\hat{F}_1\hat{F}_2 \begin{pmatrix} I & I & k \\ F & F' & J \end{pmatrix} \begin{pmatrix} I & I & k \\ F_1 & F_2 & J \end{pmatrix} \begin{pmatrix} F & F' & k \\ -\mu & \mu' & \mu_k \end{pmatrix} \begin{pmatrix} F_1 & F_2 & k \\ -\mu & \mu' & \mu_k \end{pmatrix} \right. \\
\cdot \begin{pmatrix} I & J & F \\ m & n & -\mu \end{pmatrix} \begin{pmatrix} I & J & F_1 \\ m & n & -\mu \end{pmatrix} \begin{pmatrix} I & J & F' \\ m' & n' & -\mu' \end{pmatrix} \begin{pmatrix} I & J & F_2 \\ m' & n' & -\mu' \end{pmatrix} \\
\cdot \left. \frac{1}{4} (e^{-i\omega_{FF'}t} + e^{-i\omega_{F_1F_2}t} + e^{-i\omega_{F_1F'}t} + e^{-i\omega_{FF_2}t}) e^{-i\omega_{mm'}^v t} \right\} \quad (52)$$

where $\omega_{FF'} = (1/\hbar)(E_F - E_{F'})$ and $\omega_{mm'}^v = (1/\hbar)(E_m^v - E_{m'}^v)$.

The first and second exponentials in Eq. (52) give the same contribution because all the F's are summed over and their coefficients are symmetric under the interchange $F \leftrightarrow F_1$ and $F' \leftrightarrow F_2$. By the same reason the third and fourth exponentials also give equal contributions, but their contributions are not equal to those of the first two. Thus we write

$$g_k^{o,v}(t) = \frac{1}{2} g_{1,k}^{o,v}(t) + \frac{1}{2} g_{2,k}^{o,v}(t) \quad (53)$$

where $g_{1,k}^{o,v}$ and $g_{2,k}^{o,v}$ are defined by Eq. (52) by dropping the factor $1/4$ and taking into account only $\exp(-i\omega_{FF'}t)$ and $\exp(-i\omega_{F_1F'}t)$, respectively. If we had forgotten the non-commutativity of H and K^v we would have obtained

$$g_k^{o,v}(t) = g_{1,k}^{o,v}(t)$$

which, as we shall see, would result in the approximation (45).

After some algebra one obtains

$$g_{1,k}^{o,v}(t) = g_k^o(t) G_k^v(t) \quad (54)$$

and

$$g_{2,k}^{o,v}(t) = \sum_{k',k''} C(k,k',k'') g_k^o(t) G_{k'}^v(t) G_{k''}^v(t) \quad (55)$$

where $g_k^o(t)$ is the same as in the gas model, Eq. (36), $G_k^v(t)$ are the attenuation coefficients corresponding to pure random static quadrupole, namely

$$G_k^v(t) = \sum_{mm'} \left(\begin{pmatrix} I & I & k \\ m' & -m & m-m' \end{pmatrix} \right)^2 e^{-i\omega_{mm'}^v t} \quad (56)$$

and

$$C(k, k', k'') = (2k' + 1)(2k'' + 1) \begin{Bmatrix} I & I & k' \\ I & I & k'' \end{Bmatrix} \begin{Bmatrix} I & I & k' \\ I & I & k'' \end{Bmatrix} \quad (57)$$

Now consider the sub-ensemble for which s scatterings occur, the first at t_1 , the second at $t_2, \dots$, and the s^{th} at t_s . Since these scatterings do not change the static field gradient we suppose that the only modification we have to make in Eqs. (53), (54) and (55) is to substitute $g_k^0(t)$ by $g_k^s(t; t_1, t_2, \dots, t_s)$, given by Eq. (40). Then we have

$$G_k(t) = \frac{1}{2} G_{1,k}(t) + \frac{1}{2} G_{2,k}(t) \quad (58)$$

with

$$G_{1,k}(t) = G_k^H(t) G_k^S(t) \equiv \langle g_k^s(t; t_1, t_2, \dots, t_s) \rangle_{\text{av}} \langle G_k^V(t) \rangle_{\text{av}} \quad (59)$$

and

$$G_{2,k}(t) = \sum_{k', k''} C(k, k', k'') G_{k'}^H(t) G_{k''}^S(t) \quad (60)$$

and it was possible to take the averages separately for each factor because they are taken over different variables.

Let us now examine the approximation we have made in writing Eq. (50). As we have mentioned, it is exact only to second order in $(1/\hbar)(H + K^V)t$. For $t \gg \tau_c$, the first term will not remain in the form $(1/\hbar)Ht$, but rather, in the form $(1/\hbar)H(t_i - t_{i-1})$. Since $t_i - t_{i-1}$ is of the order of τ_c , its eigenvalues will be very small quantities and this term will not cause any problem for our approximation. On the other hand K^V is a constant operator and $(1/\hbar)K^Vt$ may have large eigenvalues. Nevertheless, in many physical situations K^V is a very small perturbation and $(1/\hbar)K^Vt$ is still a small quantity. To be explicit, we will say that the approximation is good as long as $\omega_{\text{mm}}^V t \lesssim 1$, since $\cos(1)$ is still reasonably close to its second order Taylor expansion. (This does not exclude the possibility that it may be good for larger values of $\omega_{\text{mm}}^V t$. For example, in the limit $\tau_c \rightarrow 0$ it is exact for any value of K^Vt . Thus we may expect that the condition $(1/\hbar)H\tau_c \ll 1$ is sufficient to ensure the appropriateness of the approximation).

The functions $G_k^H(t)$ and $G_k^S(t)$ in Eqs. (59) and (60) are given respectively by (see gas model)

$$G_k^H(t) = e^{-\lambda_k t} \quad (61)$$

and by the average over v of Eq. (56).

Taking the average of Eq. (56) is very much simplified by the restriction of second order approximation for the static interaction. Indeed, if $f(v)$ is the distribution function for the strength V_{zz}^v of the static field gradient, then

$$\begin{aligned} G_k^S(t) &= \sum_{mm'} \left(\frac{I}{m}, \frac{I}{-m} \frac{k}{m-m'} \right)^2 \int_0^\infty f(v) \cos(\omega_{mm'}^v t) dv \\ &\approx \sum_{mm'} \left(\frac{I}{m}, \frac{I}{-m} \frac{k}{m-m'} \right)^2 \int_0^\infty f(v) \left(1 - \frac{1}{2} (\omega_{mm'}^v t)^2 \right) dv \\ &\approx \sum_{mm'} \left(\frac{I}{m}, \frac{I}{-m} \frac{k}{m-m'} \right)^2 \cos(\bar{\omega}_{mm'} t) \end{aligned} \quad (62)$$

where

$$\bar{\omega}_{mm'} = [\langle (\omega_{mm'}^v)^2 \rangle_{av}]^{1/2} \quad (63)$$

Thus, to second order, we may approximate the integral over the distribution function $f(v)$ by using a mean value for $(\omega_Q^v)^2$. We find

$$\bar{\omega}_{mm'} = 3(m^2 - m'^2) [\langle (\omega_Q^v)^2 \rangle_{av}]^{1/2} \equiv \omega_0 (m^2 - m'^2) \quad (64)$$

In Fig. (1) we compare the coefficient $G_2(t)$ for $I = 2$ obtained from Eqs. (58) through (60) (curve a) with the corresponding one obtained from Eq. (59) only (curve b), which corresponds to the approximation made in Ref. 12. The parameters λ_2 and ω_0 were arbitrarily chosen. From the similarity of the results we see that Eq. (59) by itself provides a quite reasonable approximation, confirming the assumption of Ref. 12. Calculations made with other values of the parameters also give very similar values for the two curves. In the absence of a static perturbation both Eqs. (58) and (59) reduce to Abragam and Pound's result (curve c in Fig. (1)).

It is also of interest to have the time integrated coefficients

$$\overline{G_k(\infty)} = \frac{1}{\tau} \int_0^{\infty} e^{-t/\tau} G_k(t) dt \quad (65)$$

Writing

$$\overline{G_k(\infty)} = \frac{1}{2} \overline{G_{1,k}(\infty)} + \frac{1}{2} \overline{G_{2,k}(\infty)} \quad (66)$$

one obtains

$$\overline{G_{1,k}(\infty)} = \sum_{mm'} \left(\begin{matrix} I & I & k \\ m & -m & m-m' \end{matrix} \right)^2 \frac{1}{1 + \lambda_k \tau + \bar{\omega}_{mm'}^2 \tau^2 / (1 + \lambda_k \tau)} \quad (67)$$

and

$$\overline{G_{2,k}(\infty)} = \sum_{k'k''} C(k, k', k'') \sum_{mm'} \left(\begin{matrix} I & I & k'' \\ m & -m & m-m' \end{matrix} \right)^2 \frac{1}{1 + \lambda_k \tau + \bar{\omega}_{mm'}^2 \tau^2 / (1 + \lambda_k \tau)} \quad (68)$$

To take into account the time dependent quadrupole interaction, λ_k in Eq. (61) has to be written in the form

$$\lambda_k = \lambda_k^M + \lambda_k^E \quad (69)$$

where λ_k^M is the magnetic hyperfine contribution, given by Eq. (44) and λ_k^E is the electric contribution, given by⁷

$$\lambda_k^E = \frac{3}{80} \frac{(eQ)^2}{\hbar^2} \langle V'_{zz} \rangle_{av} \tau_c \frac{k(k+1)[4I(I+1) - k(k+1) - 1]}{I^2 (2I-1)^2} \quad (70)$$

Here V'_{zz} is the time dependent electric field gradient at the nuclear site due to the non-spherical configuration of its electronic shell.

The experiments mentioned at the beginning of this section have been done at various temperatures.

If we suppose that the only effect of varying the temperature is a change in τ_c , then we can obtain the ratio λ_k^E/λ_k^M as a function of

$$R = \frac{9(eQ)^2 \langle V'_{zz} \rangle_{av}}{80 \hbar^2 \omega_J^2 J(J+1) I^2 (2I-1)^2} \quad (71)$$

namely

$$\frac{\lambda_k^E}{\lambda_k^M} = R [4I(I+1) - k(k+1) - 1] \quad (72)$$

Unfortunately there are only a few cases where the quantities involved in the definition of R are known. Nevertheless, λ_k^E is in general much smaller than λ_k^M ($R \approx 10^{-3} - 10^{-2}$ for rare earths^{14,15}) such that no appreciable error is made if we put $R = 0$. In any case, we have

$$\lambda_k = \lambda_k^M \{1 + R[4 I(I + 1) - k(k + 1) - 1]\} \quad (73)$$

In Fig. (2) we compare the curve $\overline{G_4(\infty)}$ versus $\overline{G_2(\infty)}$ for $I = 2$ obtained from Eqs. (66) through (68) with the corresponding one obtained from Eq. (67) only, which again corresponds to the approximation made in Ref. 12. The various points in each curve correspond to different values of the temperature, i.e., different values of $\omega_J^2 \tau_c$. The chosen value for $\omega_0 \tau$ is 0.4 and R was set equal to zero. Again, the curves are very similar. Curve c in Fig. (2) corresponds to pure time dependent magnetic interaction. The experimental results obtained by the Wisconsin group¹⁰ are quite well approximated by curves a and b.

D) Comments on Further Application

The method developed in Sec. III may be applied in a variety of physical situations, as long as we have a good model for calculating the matrix elements involved in Eq. (23). Some experiments have been done for ions recoiling into gas or vacuum.¹⁶ In the first case the correlation time τ_c is varying while the ion is slowing down. In the second case it is the photon emission from the excited ion which causes the relaxation of the electronic spin. Here we have an extra complication due to the variation of the magnitude of J as the photons are emitted and also due to the dipole selection rule which inhibits transitions with $\Delta n > 1$. In actual calculations these effects have to be taken into account together with the temporal variation of τ_c . Simplifying assumptions may be necessary.

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FIGURE CAPTIONS

Fig. 1: $G_2(t)$ versus time for $I = 2$, $\lambda_2 = 0.3/\text{ns}$ and $\omega_0 = 0.1/\text{ns}$.

- a) Calculated from the results of Sec. IV-C, Eq. (58).
- b) Calculated as in Ref. 12.
- c) Assuming pure time dependent interaction ($\omega_0 = 0$).

Fig. 2: $\overline{G_4(\infty)}$ versus $\overline{G_2(\infty)}$ as a function of $\omega_J^2 \tau_c$, for $I = 2$, $R = 0$ and $\omega_0 = 0.4$.

- a) Calculated from the results of Sec. IV-C, Eq. (66).
- b) Calculated as in Ref. 12.
- c) Assuming pure time dependent interaction ($\omega_0 = 0$).

Fig. 1

Fig. 2