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Reorientation of Aligned Nuclei

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Where nuclei aligned at low temperatures decay by a two-stage cascade, the degree of alignment of the intermediate state may be influenced by magnetic coupling. If the second stage is γ radiation, the angular distribution will be influenced as a consequence. This effect is treated by assuming a static interaction and twofold ionic degeneracy. In particular for high temperatures very explicit results are given for Δ', the mean degree of alignment of the intermediate state. This is dependent primarily on the mean life and magnetic moment of the intermediate state. If is found that if the properties of the intermediate state are similar to those of the initial state. (1) Reorientation effects will be present if $\tau > 10^{-9}$ sec; (2) They may either increase or decrease Δ'; (3) A strong external field cancels the effect altogether; (4) Reorientation can introduce $\cos^4\theta$ terms into the angular distribution even at the highest temperatures; (5) Where τ is very long, Δ' tends to a finite limit. The calculations are applied to an experiment.

I. INTRODUCTION

THE phenomenon of nuclear alignment at low temperatures is experimentally well established,[1–3] and the elementary theory has been given.[4,5] The experimental method has been to substitute radioactive isotopes in salts whose low-temperature properties are known. Nuclear alignment is then measured by the anisotropy of emitted γ radiation. It has not been possible so far to compare theory and experiment exactly since the magnetic moment of the radioactive nucleus is an unknown. If the theory is accepted, a value for this parameter can be deduced. This is dependent, however, on the theoretical assumptions being correct. For example, if the ground-state spin of Co^{60} is 4, as has been recently proposed,[6] the experiments referred to will need reinterpretation.

The similarity between the problem of the angular distribution from aligned nuclei and the problem of angular correlations has been pointed out.[7] It is known that magnetic coupling in the intermediate state can affect the angular correlation of successive nuclear radiations[8] and that such effects may yield information about the magnetic moment and the lifetime of the intermediate state. Where nuclei aligned at low temperatures decay, for example by a $\beta-\gamma$ cascade, disturbances, such as hfs coupling and external fields, may be expected to influence the degree of alignment of the γ-emitting state and thus the angular distribution of the γ radiation. If the lifetime of the intermediate state is long enough the probability will grow that the nucleus is reoriented by such coupling.

This effect can most conveniently be incorporated into the population functions for the substates of the intermediate nucleus, that is, into the statistical matrix of the intermediate spin system.

In the following we treat in detail a situation of experimental interest in which, in a single crystal, the ionic degeneracy before emission is twofold, having anisotropic hfs. By assuming a static interaction in the intermediate state and restricting ourselves to terms of order $(1/kT)^2$, i.e., to "high" temperatures, very explicit results are obtained. These show that reorientation can have a pronounced effect and must be considered whenever the intermediate lifetime is greater than 10^{-10} sec.

The calculations are applied to the experiment of Ambler *et al.*[3] which disagrees with the simple theory. The result is inconclusive, however, in that the effect of reorientation in this case is not sufficient to wipe out the alignment altogether.

II. GENERAL

The first stage of this problem is similar to the problem of angular correlations and our starting point is the expression given by Biedenharn and Rose,[7] whose notation is adopted, for the probability amplitude of a two-stage transition as follows: The system ion plus nucleus goes over from a gross state A having substates α, emitting radiation ρ to a gross intermediate state B, substates β, and then emitting γ radiation σ to a final state C, substates γ. The nuclear spins involved are successively J, J_1, and J_2. The probability amplitude after emission is,

$$C_{\gamma\rho\sigma}=\sum_{\alpha\beta}\frac{P_\alpha(\alpha|H_\rho|\beta)^*(\beta|H_\sigma|\gamma)^*\exp[i(\omega_\rho+\omega_\sigma+\epsilon_\gamma)t]}{(\omega_\rho+\omega_\sigma-\epsilon_{\alpha\gamma}-i\gamma_A)(\omega_\sigma-\epsilon_{\beta\gamma}-i\gamma_B)}. \quad (1)$$

The fundamental difference between the present problem and that of angular correlations should here

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[1] Daniels, Grace, and Robinson, Nature **168**, 780 (1951).
[2] Gorter, Poppema, Steenland, and Beun, Physica **17**, 1050 (1951).
[3] Ambler, Grace, Halban, Kurti, Durand, Johnson, and Lemmer, Phil. Mag. **44**, 216 (1953).
[4] Simon, Rose, and Jauch, Phys. Rev. **84**, 1155 (1951); H. A. Tolhoek and J. A. M. Cox, Physica **19**, 101 (1953).
[5] N. R. Steenberg, Proc. Phys. Soc. (London) **A65**, 791 (1952); Proc. Phys. Soc. (London) **A66**, 399 (1953).
[6] G. L. Keister and F. H. Schmidt, Phys. Rev. **93**, 140 (1954).
[7] L. C. Biedenharn and M. E. Rose, Revs. Modern Phys. **25**, 729 (1953).
[8] Aeppli, Albers-Schönberg, Bishop, Frauenfelder, and Heer, Phys. Rev. **84**, 370 (1951).

be noted. For angular correlations $P_\alpha = e^{i\varphi_\alpha}$, that is, it can be assumed that both phases and spin orientations are distributed at random so that for an assembly of systems $\langle P_\alpha \bar{P}_{\alpha'} \rangle = \delta_{\alpha\alpha'}$. For the present problem the probability amplitude for the state α is $P_\alpha = |P_\alpha| e^{i\varphi_\alpha}$ and $\langle P_\alpha \bar{P}_{\alpha'} \rangle = |P_\alpha|^2 \delta_{\alpha\alpha'}$, that is, while we can assume that the phases are randomly distributed, the spin states α are occupied with a varying probability $|P_\alpha|^2 = w_\alpha$.

The angular distribution of the γ ray σ is given now by integrating $\langle |C_{\gamma\rho\sigma}|^2 \rangle$ over all radiation energies and summing over polarization and final states in the usual way and also integrating over all directions of the first radiation, $(\int d\Omega_\rho)$, i.e.,

$$I(\theta) = \mathfrak{S}_{\rho\sigma} \int d\Omega_\rho d\omega_\rho d\omega_\sigma \sum_\gamma \langle |C_{\gamma\rho\sigma}|^2 \rangle$$

$$= \mathfrak{S}_{\rho\sigma} \int d\Omega_\rho d\omega_\rho d\omega_\sigma \sum_{\alpha\beta\beta'\gamma} w_\alpha \times \frac{(\alpha|H_\rho|\beta)^*(\beta|H_\sigma|\gamma)^*(\alpha|H_\rho|\beta')(\beta'|H_\sigma|\gamma)}{(1+i\epsilon_{\beta\beta'}\tau)}, \quad (2)$$

where $\tau = 1/2\gamma_B$ is the mean life of the intermediate state B and $\epsilon_{\beta\beta'}$ is the energy difference between the substates β and β'. Because of the Hermitian property of the matrix elements we can replace $(1+i\epsilon_{\beta\beta'}\tau)$ at once by $(1+\epsilon_{\beta\beta'}^2\tau^2)$.

We must now consider the type of interaction which both is responsible for the initial nuclear alignment and may cause reorientation in the intermediate state. For the initial state ordinary hfs coupling with no external field cannot give rise to nuclear alignment since no direction in space is preferred. (It will be seen, however, that nuclear alignment can arise solely through the reorientation terms.) We assume our source to be a single crystal having an axis of magnetic symmetry along which an external field H is applied. The interaction Hamiltonian for the initial state is then,

$$\mathcal{H} = g\mu HS_z + g_n\mu_n HJ_z + AS_zJ_z + B(S_xJ_x + S_yJ_y), \quad (3)$$

where

$S_z = z$ component of ionic spin;

g, g_n = ionic and initial nuclear g factors;

μ, μ_n = Bohr and nuclear magnetons.

The nuclear interaction $g_n\mu_n$ is of the order of 10^{-3} times the ionic interaction, and since it is found to play no significant part by itself for magnetic ions, it is discarded in what follows.

In analogy with the theory of ordinary hfs, we label the zeroth-order states of the system with quantum numbers F and μ and express them as linear combinations of ionic and nuclear states as follows:

$$\chi_F{}^\mu = \sum_{\sigma M} (\sigma M|\mu F) \psi_J{}^M \varphi_S{}^\sigma. \quad (4)$$

The perturbation problem can now be solved for the energy levels, $E\mu^F$ and the transformation coefficients $(\sigma M|\mu F)$ of the system in the initial state. The $(\sigma M|\mu F)$ are real and unitary and vanish unless $\mu = \sigma + M$. If $H=0$ and $A=B$ (which would result, however, in no nuclear alignment at all) $(\sigma M|\mu F)$ becomes the regular vector addition coefficient, $(JSM\sigma|JSF\mu)$, in the notation of Condon and Shortley.[9] Note that while $\chi_F{}^\mu$ is an eigenfunction of $F_Z = J_Z + S_Z$ with eigenvalue μ, it is not (unless $H=0$, $A=B$) an eigenfunction of $\mathbf{F}^2 = \mathbf{J}^2 + \mathbf{S}^2 + 2\mathbf{J}\cdot\mathbf{S}$. F is, however, a convenient label.

For the interaction in the intermediate state we shall choose a static Hamiltonian $\mathcal{H}'$ of similar form and label the intermediate states with F_1 and μ_1. Equation (2) can now be rewritten replacing α by $F\mu$, β by $F_1\mu_1$, etc., $\epsilon_{\beta\beta'}$ by $\epsilon(F_1F_1'\mu_1\mu_1')$ and w_α by $w_\mu{}^F = \exp(-E_\mu{}^F/kT)$ and transforming the matrix elements as follows:

$$(\alpha|H_\rho|\beta) = \sum_{MM_1\sigma} (\sigma M|\mu F)(\sigma M_1|\mu_1 F_1)(M|H_\rho|M_1),$$

$$(\beta|H_\sigma|\gamma) = \sum_{M_1'M_2\sigma'} (\sigma' M_1'|\mu_1 F_1) \times (\sigma' M_2|\mu_2 F_2)(M_1'|H_\sigma|M_2).$$

We now assume for simplicity that the first radiation is emitted with one value only of total angular momentum j. It can then be shown that except for constants,

$$\mathfrak{S}_\rho \int d\Omega_\rho (M|H_\rho|M_1)^*(M'|H_\rho|M_1') = (J_1jM_1M-M_1|J_1jJM) \times (J_1jM_1'M'-M_1'|J_1jJM') \times \delta(M-M_1, M-M_1'). \quad (5)$$

It is further assumed that the γ ray is a pure multipole of order L, then

$$\mathfrak{S}_\sigma \sum_{M_2} |(M_1|H_\sigma|M_2)|^2 = I(M_1,\theta), \quad (6)$$

$$I(M_1,\theta) = 1 + \sum_k X_k(J_1) P_k(\cos\theta) \Pi_k(M_1J_1). \quad (7)$$

Here X_k is a factor[5] depending on the transition and the multipole order, $P_k(\cos\theta)$ is a Legendre polynomial, and k takes even values 2, 4, $\cdots$ up to $2L$ or $2J_1$, whichever is smaller. It will be found that under certain reasonable circumstances, only the term with $k=2$ will enter. $\Pi_k(M_1J_1)$ is related to $(kJ_10M_1|kJ_1J_1M_1)$, in particular $\Pi_2(M_1) = 3M_1^2 - J_1(J_1+1)$. The angular distribution (2) can now be written

$$I(\theta) = \sum_{M_1} W(M_1) I(M_1,\theta), \quad (8)$$

where by utilizing Eq. (5) and Eq. (6) and the orthogo-

[9] E. U. Condon and G. H. Shortley, *The Theory of Atomic Spectra* (Cambridge University Press, Cambridge, 1951).

nalities inherent in the transformation coefficients,

$$W_{M_1}=\sum_{FM\lambda\rho} w_{M+\rho}{}^F \times[(\rho M|\rho+MF)(\rho+\lambda M-\lambda|\rho+MF)H(\rho\lambda\sigma M_1) \times(J_1jM_1+\sigma-\rho\rho+M-M_1-\sigma|J_1jJM) \times(J_1jM_1+\sigma-\rho-\lambda M-M_1-\sigma+\rho|J_1jJM-\lambda)], \quad (9)$$

and

$$H(\rho\lambda\sigma M_1)=\delta_{\rho\sigma}\delta_{\lambda 0}-\tau^2K(\rho\lambda\sigma M_1),$$

$$K(\rho\lambda\sigma M_1)=\sum_{F_1F_1'}\frac{\epsilon^2(F_1F_1'M_1+\sigma M_1+\sigma)}{[1+\epsilon^2(F_1F_1'M_1+\sigma M_1+\sigma)\tau^2]} \times[(\rho M_1+\sigma-\rho|M_1+\sigma F_1)(\sigma M_1|M_1+\sigma F_1) \times(\rho+\lambda M_1+\sigma-\rho-\lambda|M_1+\sigma F_1')(\sigma M_1|M_1+\sigma F_1')]. \quad (10)$$

This may be compared with a similar expression given by Goertzel[10] in the theory of angular correlations. It should be noted that $H(\rho\lambda\sigma M_1)$ depends on properties of the intermediate state only. $W(M_1)$ can be interpreted as the mean relative population of the nuclear state M_1 at the emission of the second radiation.

III. STATISTICAL MATRIX INTERPRETATION

It can be seen from Eqs. (9) and (10) that when $\tau=0$, $W(M_1)$ reduces to

$$W(M_1)=\sum_M (J_1jM_1M-M_1|J_1jJM)^2W(M), \quad (11)$$

where

$$W(M)=\sum_{F\rho} w_{M+\rho}{}^F(\rho M|M+\rho F)^2.$$

It will be recalled that $W(M)$, the relative population of a given substate of the initial nucleus is the function to which special attention has been paid in previous treatments of this subject.[11] $W(M)$ [or more correctly, perhaps, $W(MM)$] is a diagonal element of the statistical matrix referring to the nuclear spin system alone, i.e.,

$$W(M)=(M|\mathrm{Tr}'\,\rho|M)(\mathrm{Tr}\,\rho)^{-1},$$

where Tr′ implies a sum over non-nuclear quantum numbers and ρ is the statistical operator $\rho=\exp\{-\mathcal{H}/kT\}$. From this point of view the mean value of any nuclear observable O is calculated from the formula

$$\langle O\rangle=\mathrm{Tr}[O\rho](\mathrm{Tr}\,\rho)^{-1},$$

which, if O is diagonal in M, becomes

$$\langle O\rangle=\sum_M W(M)(M|O|M).$$

However, $\langle O\rangle$ is independent of representation so that if it is more convenient (it generally is not, from the point of view of numerical computation) we can choose, for example, the representation in which the intermediate state is quantized, i.e.,

$$\langle O\rangle=\sum_{M_1} W(M_1)(M_1|O|M_1),$$

if O is diagonal in M_1, where

$$W(M_1)=(M_1|\mathrm{Tr}'\,\rho|M_1)(\mathrm{Tr}\,\rho)^{-1} =\sum_M(J_1jM_1M-M_1|J_1jJM)^2W(M).$$

(Tr′ ρ does not operate on coordinates of the emitted particle.) Thus Eq. (11) represents a change of representation of the statistical matrix from one in which M is a good quantum number to one in which M_1 is a good quantum number. Equation (11) gives $W(M_1)$ where the intermediate state is not perturbed and leads to an angular distribution which is identical to that previously given. Where it is disturbed it can be seen from Eqs. (9) and (10) that while our operator, the angular distribution from an intermediate state, is diagonal in M_1 $[I(M_1,\theta)]$ it is not diagonal in M. We have thus chosen the M_1 representation, and Eq. (9) represents the change of representation. The coefficient of τ^2 arises from nondiagonal elements of $[\rho]$ in the M representation. It may be observed here that the statistical matrix considered here develops in time, tending in fact as will be seen, to an equilibrium value.

From this point of view we can define the mean degree of nuclear alignment of the intermediate state at emission in analogy with a previous definition[4] as follows

$$\Delta'=\langle\Pi_2(J_1)\rangle/3J_1^2=\sum_{M_1} W(M_1)\Pi_2(M_1J_1)/3J_1^2. \quad (12)$$

It has been shown previously[5] that where τ is zero and $(A/kT)^2\ll 1$ the angular distribution depends on $\langle\Pi_2(J)\rangle$ alone, *viz.*,

$$I(\theta)=1+X_2(J)P_2(\cos\theta)\langle\Pi_2(J)\rangle.$$

In this approximation the anisotropy, $\epsilon=1-I(0)/I(\frac{1}{2}\pi)$, is given by

$$\epsilon=-\tfrac{3}{2}X_2(J)\langle\Pi_2(J)\rangle. \quad (13)$$

The same result is obtained if the J_1 representation is used in place of the J. Since $\Pi_k(M_1)=\Pi_k(-M_1)$ for k even, so far as $I(\theta)$ or Δ' are concerned, it is sufficient to consider $W^E(M_1)=\frac{1}{2}[W(M_1)+W(-M_1)]$. In what follows we shall deal exclusively with $W^E(M_1)$.

IV. TWOFOLD IONIC DEGENERACY

We now restrict our discussion to ionic systems which at low temperatures can be described by a fictitious "ionic spin," $S=\frac{1}{2}$, a situation of frequent occurrence and obtaining in the experiments already carried out on nuclear alignment. With this assumption the transformation coefficients and energy levels of the initial system become

$$E_\mu{}^{\pm}=\tfrac{1}{2}[2\lambda\mu-\tfrac{1}{2}A\pm\tfrac{1}{2}AQ(\mu)], \quad (14)$$

[10] G. Goertzel, Phys. Rev. **70**, 897 (1946).
[11] N. R. Steenberg, Phys. Rev. **93**, 678 (1954).

where

$$Q(\mu)=[(2\delta-2\lambda+2\mu)^2+4x^2(J+\tfrac{1}{2}+\mu)(J+\tfrac{1}{2}-\mu)]^{\frac{1}{2}},$$
$$\delta=g\mu H/A,\quad \lambda=g_n\mu_n H/A,\quad x=B/A,$$

and

$$(\tfrac{1}{2},\mu-\tfrac{1}{2}|\mu+)=(-\tfrac{1}{2},\mu+\tfrac{1}{2}|\mu-)=\left[\frac{2\delta-2\lambda+2\mu+Q(\mu)}{2Q(\mu)}\right]^{\frac{1}{2}},$$
$$(-\tfrac{1}{2},\mu+\tfrac{1}{2}|\mu+)=-(\tfrac{1}{2},\mu-\tfrac{1}{2}|\mu-)=\frac{2x(J+\tfrac{1}{2}-\mu)(J+\tfrac{1}{2}+\mu)}{[2Q(\mu)(2\delta-2\lambda+2\mu+Q(\mu))]^{\frac{1}{2}}}. \quad (15)$$

The $+$ and $-$ signs correspond to $F=J+\frac{1}{2}$ and $F=J-\frac{1}{2}$, respectively.

There now arises the difficult problem of the representation of the interaction in the intermediate state. While the choice made here of a static interaction, such as is assumed in the theory of the attenuation of correlations, is open to the objections discussed in VIII, it does have the merit of a straightforward, easily interpreted solution. The Hamiltonian for the intermediate state is then

$$\mathcal{H}'=g'\mu HS_z'+g_n'\mu_n HJ_{1z}+A'S_z'J_{1z}+B'(S_x'J_{1x}+S_y'J_{1y}),$$

where the constants have the same meaning as those given for Eq. (3) except that they refer to the intermediate state, and again we take $S'=\frac{1}{2}$.

The perturbation problem for the intermediate state is now identical to that for the initial state and the energy levels and transformation coefficients are given by expressions analogous to Eqs. (14) and (15). It then follows that the quantities $K(\rho\lambda\sigma M_1)$ Eq. (10) are given by

$$K(\tfrac{1}{2}0\tfrac{1}{2}M_1)=-K(-\tfrac{1}{2}0\tfrac{1}{2}M_1)=G(M_1);$$
$$K(\tfrac{1}{2}0-\tfrac{1}{2}M_1)=-K(-\tfrac{1}{2}0-\tfrac{1}{2}M_1)=-G(M_1-1);$$
$$K(\tfrac{1}{2}-1\tfrac{1}{2}M_1)=K(-\tfrac{1}{2}1\tfrac{1}{2}M_1)=-F(M_1);$$
$$K(\tfrac{1}{2}-1-\tfrac{1}{2}M_1)=K(-\tfrac{1}{2}1-\tfrac{1}{2}M_1)=F(M_1-1);$$
$$\tau^2G(M_1)=2x'^2\alpha^2(J_1+M_1+1)(J_1-M_1)[1+\alpha^2Q^2(M+\tfrac{1}{2})]^{-1};$$
$$\tau^2F(M_1)=\frac{x'\alpha^2[(J_1+M_1+1)(J_1-M_1)]^{\frac{1}{2}}(2\delta'-2\lambda'+2M_1+1)}{[1+\alpha^2Q^2(M+\tfrac{1}{2})]},$$

where $x'=A'/B'$ and $\alpha=\tau A'/2\hbar$. α may be thought of as the mean angle precessed through by the nucleus under the influence of the hfs field, analogous to the Larmor precession in an external field.

It should be observed here that $W(M_1)$ can be written in the form

$$W(M_1)=\textstyle\sum_M(J_1jM_1M-M_1|J_1jJM)^2W(M)-\tau^2[A(M_1)-A(M_1-1)], \quad (16)$$
$$A(M_1)=\sum_{FM\lambda\rho}w_{M+\rho}{}^F\times[(\rho M|\rho+MF)(\rho+\lambda M-\lambda|\rho+MF)K(\rho\lambda\tfrac{1}{2}M_1)\times(J_1jM_1-\rho+\tfrac{1}{2}\rho+M-M_1-\tfrac{1}{2}|J_1jJM)\times(J_1jM_1-\rho+\tfrac{1}{2}-\lambda\rho+M-M-\tfrac{1}{2}|J_1jJM-\lambda)]. \quad (17)$$

Thus, since

$$\sum_{MM_1}(J_1jM_1M-M_1|J_1jJM)^2W(M)=1,\quad \sum_{M_1}W(M_1)=1,$$

as should be expected on the interpretation of $W(M_1)$ as the probability of a nucleus to be in the state M_1.

Two important features can be seen here. The first is that for a sufficiently strong external field $K(\rho\lambda\sigma M_1)$ vanishes. Then, as is the case in angular correlations, the angular distribution is independent of interaction in the intermediate state. It is found that for typical values of the constants [$A=10^{-18}$ erg ($\simeq5\times10^{-3}$ cm^{-1}) and $g=3$] the effect of reorientation vanishes for $H\geq10^3$ gauss. The second feature is that the effect of reorientation vanishes altogether if $x'=A'/B'$ is zero. The intermediate interaction is then diagonal in M_1 and σ so that no transitions are possible which affect $W(M_1)$.

V. EXPANSION IN POWERS OF $1/kT$

For arbitrary temperatures no further reduction of the above expression, Eq. (16), for $W(M_1)$ is possible. However, it has been found that the leading terms in an expansion in powers of $1/kT$, i.e., for relatively high temperatures, contains explicitly nearly all the features which enter the exact expression and is often sufficient to interpret experimental results. Where this is not the case the series expansion does give an indication in direction and order of magnitude of the effect of introducing factors neglected in the simple theory.

Therefore, expanding the exponentials, $w_\mu{}^F=\exp(-E_\mu{}^F/kT)$ in powers of $\gamma=-A/2kT$, it is found that the sums indicated in Eq. (17) can be performed with the aid of the Racah sum formulas.[12] This expansion requires that $g\mu H$ be of the order of A or less. When $g\mu H\gg A$, the leading term in the nuclear alignment is[11] $(A/2kT)^2$ rather than $(A^2-B^2)(2kT)^2$.[13] For such a field the effect of reorientation is of course eliminated.

The first feature to emerge is the rather surprising

[12] See, for example, L. C. Biedenharn, Oak Ridge National Laboratory Report ORNL-1098 (unpublished).

[13] In reference 11, due to an error on the author's part, it is stated that as $H\rightarrow\infty$, $a_2\rightarrow(1-x^2)/8$. This should read instead $a_2\rightarrow\frac{1}{8}$, and hence Eq. (20) of reference 10 should read

$$W_M{}^E\simeq(2I+1)^{-1}\{1+\tfrac{1}{6}(A(2kT)^2\Pi_2(M)\}.$$

result that arising from the effect of reorientation there is in $W^E(M_1)$ a term of order γ. This is in contrast to previous results where the first term was in all cases of order γ^2. To order γ,

$$W^E(M_1)=(2J_1+1)^{-1}\{1-\gamma\alpha^2x'[2(\delta x'-x\delta' S_1)\Theta(M_1)+S_1(x'-x)\chi(M_1)]+\cdots\},\quad (18)$$

where $S_k=S_k(J_1jJ)$ is related to the Racah coefficient

$$S_k(J_1jJ)=\left[\frac{(2J_1-k)!(2J+k+1)!}{(2J_1+k+1)!(2J-k)!}\right]^{\frac{1}{2}}\times(2J_1+1)W(JjkJ_1|J_1J),$$

and

$$\Theta(M_1)=\tfrac{1}{2}[g(M_1)-g'(-M_1)-g'(M_1)+g(-M_1)];$$

$$\chi(M_1)=\tfrac{1}{2}[(g(M_1)+g'(-M_1))(2M_1+1)-(g'(M_1)+g(-M_1))(2M_1-1)];$$

$$g(M_1)=(J_1+M_1+1)(J_1-M_1)[1+\alpha^2Q^2(M_1+\tfrac{1}{2})]^{-1};$$

$$g'(M_1)=g(M_1-1).$$

It should be noted that where all variables (τ, H, x, and x') are arbitrary $\sum_{M_1}\Theta(M_1)\Pi_k(M_1J_1)$ and $\sum_{M_1}\chi(M_1)\Pi_k(M_1J_1)$ are both finite for all values of k except $k=0$ and $k>2J_1$. An important consequence of this fact is that even at the highest temperatures the effect of reorientation is to introduce $\cos^4\theta$, $\cos^6\theta$, etc., terms of order γ into the angular distribution [provided of course that they are present in $I_{M_1}(\theta)$]. In the absence of reorientation $\cos^4\theta$ terms only appear in terms of order γ^4. This $\cos^4\theta$ term may be large enough to provide a decisive test as to whether reorientation is taking place.

A further interesting point which is shown by Eq. (18) is that nuclear alignment in the intermediate state can arise through the reorientation terms alone where none would be present if the intermediate lifetime were zero. In the first place, where $H=0$ and $x=1$, isotropic hfs in the initial state, no alignment would normally be expected. However, $W^E(M_1)$ will still differ from its isotropic value $1/(2J_1+1)$ unless $x'=1$, that is unless the hfs in the intermediate state is isotropic also. Secondly, if the initial nucleus has spin $\frac{1}{2}$ no alignment under any circumstances would be expected on the simple theory. This is shown in the formula for $W^E(M_1)$ by the fact that $S_k(J_1j\frac{1}{2})=0$ for $k\geq 2$. However, for a $(\frac{1}{2}\xrightarrow{j}J_1\xrightarrow{L}J_2)$ transition where $J_1>\frac{1}{2}$, $S_1(J_1j\frac{1}{2})$ does not vanish. In particular if

$$J_1=j+\tfrac{1}{2},\quad S_1(J_1,J_1-\tfrac{1}{2},\tfrac{1}{2})=1/(2J_1).$$

It should also be noted that the sign of alignment to order γ will depend on the sign of magnetic moment of the initial nucleus through the coupling constant A.

While under some circumstances the term in $1/kT$ will be important, in general both this and the term in $(1/kT)^2$ will have to be considered together. To order γ^2,

$$\begin{aligned}W^E(M_1)=&(2J_1+1)^{-1}\{1-\gamma\alpha^2x'[2(\delta x'-\delta' xS_1)\Theta(M_1)+S_1(x'-x)\chi(M_1)]\}\\&-\tfrac{1}{2}\gamma^2\alpha^2(2J_1+1)^{-1}\{2S_1(xx'\delta'-x'^2\delta)\Theta(M_1)-[S_2x'^2(1-x^2)-S_1xx'(1-xx')]\chi(M_1)\}\\&+\tfrac{1}{6}\gamma^2(1-x^2)S_2\Pi_2(M_1)(2J_1+1)^{-1}+\cdots.\quad(19)\end{aligned}$$

The last term will be recognized as the only term responsible for nuclear alignment where the effect of reorientation is neglected.

We can now consider several reasonable situations for which the above formula simplifies considerably. If the first emission is a γ ray which does not appreciably disrupt the electronic system we can assume $x=x'$, i.e., that the intermediate state "sees" much the same field as does the initial state. Then in zero field the term in γ vanishes altogether and

$$W^E(M_1)=(2J_1+1)^{-1}[1+\tfrac{1}{2}\gamma^2\alpha^2\chi(M_1)x^2(1-x^2)(S_2-S_1)+\tfrac{1}{6}\gamma^2(1-x^2)S_2\Pi_2(M_1)+\cdots],\quad(20)$$

where

$$\chi(M_1)=g(M_1)(2M_1+1)-g'(M_1)(2M_1-1),$$

$$g(M_1)=g'(M_1+1)=\frac{(J_1+M_1+1)(J_1-M_1)}{1+\alpha^2[(2M_1+1)^2(1-x^2)+x^2(2J_1+1)^2]}.$$

Note in Eq. (20) how the alignment given by the simple theory may be either enhanced or diminished by the effect of reorientation through the factor (S_2-S_1). In particular for a $(J_1+j\xrightarrow{j}J_1\xrightarrow{L}J_2)$ transition $(S_2-S_1)=2(J_1+j+1)j/[(J_1+1)(2J_1+3)]$ which is always positive and thus causes a diminution of alignment; and for a $(J_1-j\xrightarrow{j}J_1\xrightarrow{L}J_2)$ transition $(S_2-S_1)=-2(J_1-j)j/[J_1(2J_1-1)]$ which is always negative causing enhancement, $\sum\chi(M_1)\Pi_2(M_1)$ being always negative.

It may be that, particularly where β emission initiates the cascade, the interaction in the intermediate state is better described by an isotropic hfs coupling $x'=1$. Then in zero field with $x'=1$, $Q^2(\mu)=(2J_1+1)^2$ and

$$\chi(M_1)=-2\Pi_2(M_1J_1)[1+\alpha^2(2J_1+1)^2]^{-1}.$$

Here, and indeed wherever the M_1 dependence of $W^E(M_1)$ is through $\Pi_2(M_1)$ alone, the angular distribution will contain only the $\cos^2\theta$ term, i.e., $P_2(\cos\theta)$.

$W^E(M_1)$, Eq. (19) is now

$$W^E(M_1)=(2J_1+1)^{-1}+\frac{\Pi_2(M_1J_1)}{(2J_1+1)}\left\{\frac{2\gamma\alpha^2S_1(1-x)}{1+\alpha^2(2J_1+1)^2}-\frac{\gamma^2\alpha^2[S_2(1-x^2)-S_1x(1-x)]}{1+\alpha^2(2J_1+1)^2}+\tfrac{1}{6}\gamma^2(1-x^2)S_2\right\}. \quad (21)$$

For this situation we can write the mean degree of nuclear alignment for the intermediate state Δ', defined in Eq. (12), to order $(1/kT)^2$ for an arbitrary lifetime and show that for very long lifetimes Δ' approaches a finite limit. Thus

$$\Delta'=\frac{(J_1+1)(2J_1+3)(2J_1-1)}{15J_1}\left\{\frac{\alpha^2(1-x)}{1+\alpha^2(2J_1+1)^2}\times[2\gamma S_1-\gamma^2S_2(1+x)+S_1\gamma^2x]+\tfrac{1}{6}\gamma^2(1-x^2)S_2\right\}, \quad (22)$$

which approaches an evident limit as $\alpha^2\to\infty$, i.e., as $\alpha^2/[1+\alpha^2(2J_1+1)^2]\to 1/(2J_1+1)^2$. When $A=10^{-18}$ erg, the requirement that $\alpha^2(2J_1+1)^2\gg 1$ implies a lifetime $\tau\geq 5\times 10^{-9}/(2J_1+1)$ sec. It may be borne in mind that for very long lifetimes the effects under discussion may overlap the effects of thermal relaxation.

From Eq. (22) an idea can be obtained of the minimum lifetime necessary in order that reorientation be appreciable. If we take as our criterion that the first term in the square bracket be at least $\frac{1}{4}$ of the second and put $x=0$, $\gamma=\frac{1}{4}$, $S_1=S_2=1$, we find that α must be at least 0.08 for $J_1=1$ or at least 0.15 for $J_1=5$. This implies, for $A=10^{-18}$ erg, lifetimes of 1.6×10^{-10} and 3×10^{-10} sec, respectively.

Another plausible assumption is that α is small enough that in an expansion in powers of α terms in α^4 may be neglected in comparison to terms in α^2. This approximation must be used with care, however, since the most stringent criterion would require that $\alpha^2 \ll 1/(2T_1+1)^2$, see Eq. (22), while α^2 must still be appreciable in order that reorientation have any effect at all. The two requirements are all but mutually exclusive. Nevertheless, if we discard terms in α^4, i.e., replace $\alpha^2/[1+\alpha^2Q^2(M_1+\frac{1}{2})]$ by α^2 we obtain

$$W^E(M_1)=(2J_1+1)^{-1}+\frac{\Pi_2(M_1)}{(2J_1+1)}\left\{2\gamma\alpha^2x'(x'-x)S_1-\gamma^2\alpha^2[S_2x'^2(1-x^2)-S_1xx'(1-xx')]+\tfrac{1}{6}\gamma^2(1-x^2)S_2+\cdots\right\}. \quad (23)$$

This expression is independent of the external field.

Where both the term in γ and that in γ^2 must be considered, the expression Eq. (13) for the anisotropy must be modified. When $I(\theta)=1+X_2(J_1)P_2(\cos\theta)\times\langle\Pi_2(J_1)\rangle$ and $\langle\Pi_2(J_1)\rangle=(C\gamma+D\gamma^2)$, ϵ is given by

$$\epsilon=-\tfrac{3}{2}X_2(J_1)[\langle\Pi_2(J_1)\rangle+\tfrac{1}{2}X_2(J_1)C^2\gamma^2]$$

in place of Eq. (13). When $W^E(M_1)$ is such that the angular distribution involves a $\cos^4\theta$ term, this will also affect ϵ.

VI. AN APPLICATION

To illustrate the ideas discussed above, they may be applied to the experiment of Ambler *et al.*[3] There Co^{60} was used in a Ce-Co salt and a high degree of alignment was obtained with an external field. The stable Co ions in this salt are found to be magnetically of two types,[14] X and Y in the ratio 2:1. The X-type ions have very nearly isotropic hfs at 20°K while the hfs of the Y ions is strongly anisotropic, $A=286.6\times 10^{-4}$ cm^{-1}, $B<1\times 10^{-4}$ cm^{-1},[15] i.e., $x\simeq 0$. When reorientation effects are neglected the theory predicts a substantial degree of alignment in zero field arising from the Y ions alone. In fact, for $1/T=46$ (deg)$^{-1}$, the anisotropy of γ radiation should be $\epsilon=0.11$,[16] whereas the experimental result in zero field at this temperature was $\epsilon=0.0\pm 0.03$. At an external field of 430 oersted, however, experiment and theory were in good agreement.

Let us now assume a finite lifetime for the first intermediate state in the decay of Co^{60}, i.e., the second excited state of Ni^{60}, and see for the Y ions what effect this has on the zero-field angular distribution of the first only of the two γ rays. It has been seen that if $x'=0$ there can be no reorientation at all. Since the first stage is a β emission it is reasonable that the coupling in the intermediate state (insofar as it can be described by a static interaction) should be different from that in the initial state. For simplicity then we take $x'=1$. The mean degree of nuclear alignment of the intermediate state is now given by Eq. (22) with $x=0$. The decay scheme of Co^{60} is here taken to be $(5\xrightarrow{1}4\xrightarrow{2+}2\xrightarrow{2+}0)$, whence $S_1=6/5$, $S_2=78/55$. A, to refer to Co^{60}, must be scaled according to the gyromagnetic ratios of Co^{60} and Co^{59}; thus for a nuclear magnetic moment of 3.5 nuclear magnetons for Co^{60},[17] $A=3\times 10^{-18}$ erg, larger in this case than that previously taken as typical. Taking $\alpha=0.3$ and $1/T=46$ deg^{-1} (i.e., $\gamma=-\frac{1}{2}$), it is found that the mean nuclear alignment and the anisotropy is reduced by 30 percent. If we assume that the alignment in the second intermediate state (first excited state of Ni^{60}) is disturbed by the same amount, the average reduction in the anisotropy is 38 percent, and ϵ is reduced from 0.11 to 0.068. For $\alpha=0.3$, $\alpha^2/(1+81\alpha^2)$ and thus the degree of reorientation is very nearly at its maximum value. Thus we must conclude that on this basis, while reorientation

[14] R. S. Trenam, Proc. Phys. Soc. (London) **A66**, 118 (1953).
[15] R. S. Trenam (unpublished).
[16] N. R. Steenberg, thesis, Oxford University, 1953 (unpublished).
[17] M. A. Grace and H. Halban, Physica **18**, 1227 (1952).

may be responsible for part of the disagreement, it cannot account entirely for it.

The value 0.3 for $\alpha = A'\tau/2\hbar$ implies if A' is taken equal to A, a mean life of $\tau = 2\times 10^{-10}$ sec for the second excited state of Ni^{60}. Engelder,[18] by delayed coincidence studies finds for this state $\tau < 10^{-9}$ sec. Aeppli *et al.*,[19] as a result of failure to restore an attenuated angular correlation, conclude that the mean life of the first excited state of Ni^{60} is of the order of 10^{-11} sec.

If the electronic g factor for the intermediate state is assumed to be the same as that for the initial state $g = 7.29$,[14] a field of 500 oersted is ample to restore the anisotropy to its unperturbed value.

These arguments are not materially affected if the original experiment[17] is reinterpreted as the basis of spin 4 for the ground state of Co^{60}.[6] This would require a magnetic moment of between 3.33 and 3.63 ± 0.5 nuclear magnetons depending on the relative contributions of Fermi and Gamow-Teller selection rules.

The considerations in this paragraph are put forward primarily to illustrate the arguments in the body of the paper. Before further speculation on this experiment is justified, certain additional experimental data are necessary. In particular, the hfs constants A and B should be remeasured at helium temperature and if possible it should be discovered if there is an antiferromagnetic transition between 4° and 0.01°. Furthermore it would be highly desirable to measure the spin and magnetic moment of Co^{60} by conventional methods in order to resolve the present ambiguity. As these experiments may serve as a pattern for future developments, they should be understood as fully as possible.

VII. DISCUSSION

This analysis, even if incomplete, at least shows clearly that the effect of finite lifetime and consequent nuclear reorientation must not be neglected in the theory of nuclear alignment at low temperatures. If the properties of the intermediate state are similar to those of the initial state, these effects may appear for lifetimes of the order of 10^{-10} sec and will certainly be present for lifetimes greater than 10^{-9} sec. A number of nuclei suitable for nuclear alignment fall into this class.

If the foregoing treatment is substantially correct, then we know further that the effect of reorientation reaches a maximum for $\tau \simeq 10^{-8}$ sec and that thereafter the alignment of the intermediate state is constant until the effect of thermal relaxation becomes appreciable. The application of an external magnetic field of the order of 10^3 oersted will eliminate the effect of reorientation. The effect on the anisotropy may be of the order of 40 percent, and it may either enhance or diminish the anisotropy. The effect may depend in part on the sign of the magnetic moment of the initial nucleus.

Abragam and Pound[20] have drawn attention to the importance of nonstatic interactions. A static perturbation for the intermediate state in a single crystal is most plausible when the first transition is a γ emission which does not disturb the electronic system. We would then expect the intermediate nucleus to see much the same field as the initial nucleus except for the change in nuclear spin and magnetic moment. In this case only the effect of nuclear recoil is neglected. For a nucleus of mass 60 the recoil energy from a 1-Mev γ ray is approximately 9 ev or 10^5 degrees. This may affect our assumption that the intermediate state can be described as an ionic doublet.

Where β emission initiates the cascade, the disruption of the atomic shell, autoionization and change of Z, are serious objections to a static perturbation. It would not be expected that the electronic structure would be settled into its final state as an impurity atom in the lattice of charge $Z\pm 1$ in times comparable to those we are considering, 10^{-7} sec. But how far the process would advance in this time, and what forces the atomic system would exert on the nucleus during the process are the questions involved.

The case of K capture is even more obscure. Here, until the K shell is filled, a time of $\sim 10^{-11}$ sec, the intermediate nucleus is in the field of the one odd K electron, of the order of 10^8 gauss. This field would be expected to produce pronounced effects even for very short-lived nuclear states. However, the results[21] for Co^{58} decaying 85 percent by K capture are very similar to those of Co^{60}. It may be that this field must be thought of as in fact oscillating so rapidly that it averages to zero.

VIII. ACKNOWLEDGMENT

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