PHYSICAL REVIEW C VOLUME 17, NUMBER 1 JANUARY 1978

Nuclear orientation with combined electric and magnetic interactions

R. Haroutunian and M. Meyer
Institut de Physique Nucléaire and IN2P3, Université Lyon-1, 43 BD du 11 Novembre 1918-69621 Villeurbanne, France

R. Coussement
Instituut voor Kern-en-Stalingsfysika, Leuven University, 3030 Heverlee, Belgium
(Received 29 April 1977)

The combined interaction of a static electric field gradient and a static magnetic field with the electromagnetic moments of a nucleus is considered for the case of nuclear orientation at low temperature. The general expression of the angular directional distribution of radiation emitted by the oriented state is developed for polycrystalline samples, where the principal axes of the electric field gradients are randomly distributed with respect to a fixed magnetic field direction. Due to axial symmetry of the ensemble the effect of the quadrupole interaction is reduced to an attenuation factor on the usual B_K coefficients. Numerical calculations of these attenuation factors for $K = 1$, 2, and 4 have been performed in the case of the symmetric electric field gradient for a wide range of electric to magnetic interaction ratios and spin values $I = 1,3/2,... 8$. Typical attenuation curves for spin 5/2 and 9/2 are presented. Comparing the experimental anisotropies with the tabulated values, one can extract the quadrupole interaction value $\hbar\omega_Q$.

[RADIOACTIVITY Attenuation by random and symmetric electric quadrupole interaction of the low temperature nuclear orientation coefficient due to a polarizing magnetic field are computed for multipole order 1, 2, and 4 of the emitted radiation angular distribution.]

I. INTRODUCTION

The knowledge of electric quadrupole moments of nuclear states is quite important for understanding nuclear structure. Hyperfine interaction methods, especially the perturbed correlation method, have provided some quadrupole moment Q of short-lived excited states. For longer lifetimes, nuclear alignment by quadrupole interaction observed at low temperature can also be used, but in many cases single crystal growing time or preparation difficulties reduce considerably the experimental applicability.

In order to avoid monocrystal preparation, we consider the attenuation of the magnetic nuclear orientation by an electric quadrupole one in a polycrystalline sample, where the principal axes of the electric field gradient (EFG) are randomly distributed with respect to the magnetic field direction. We expect this attenuation to be very important if the quadrupole splitting is of the same order of magnitude as the magnetic one. This condition can be approached with the brute force orientation method; nevertheless, in many cases, quite low temperature is needed for measurable anisotropy and attenuation.

The recent technological developments on the ^{3}He-^{4}He dilution refrigerator [rapid sample loading facility ($\simeq\frac{1}{2}$ h) and much lower temperature ($\lesssim 10$ mK)] and the strong external field ($\simeq 10$ T) produced by the new available superconducting magnets, give experimenter the tools to fulfill the conditions required here.

In this paper we describe low temperature nuclear orientation with the combined interactions defined above. To allow evaluation of measurable cases, we have calculated and tabulated the attenuation coefficients for various spin values and several electric to magnetic interaction ratio values as a function of an orientation parameter defined by the magnetic interaction only and the temperature.

II. COMBINED ELECTRIC AND MAGNETIC INTERACTION

Eigenvalues E_N and eigenvectors $|N\rangle$ of the oriented state being needed to calculate the low temperature nuclear orientation coefficients (see Sec. III), we must consider the total Hamiltonian given by:

$$H = H_{\text{mag}} + H_{\text{elec}} = H_B + H_Q\,.$$

For our purpose it is convenient to take the magnetic field $\vec{B}$ direction as the z axis of the laboratory system (lab) (see Fig. 1). In this system, we use as basis, the eigenvectors $|Im\rangle = |m\rangle$ of I_z and I^2 operators, so we have:

$$H_B = -g\mu_n \vec{I}\cdot\vec{B} = -g\mu_n B I_z\,.$$

In each microcrystal we choose the XYZ coordinate axis along the principal axis system (PAS) of the EFG (see Fig. 1), and define by $|\tilde{\mu}\rangle$ the eigenvectors of I_Z and I^2. The electric quadrupole interaction is then given by the well-known expression[1]

$$H_Q = \frac{eQV_{ZZ}}{4I(2I-1)}[3I_Z^2 - I^2 + \tfrac{1}{2}\eta(I_+^2 + I_-^2)] .$$

A rotation $R(\alpha\beta\gamma)$, where $\alpha\beta\gamma$ are the usual Euler angles, connects the lab system to the PAS (see Fig. 1) and we have

$$RR^\dagger = 1 , \quad |\tilde{\mu}\rangle = R|\mu\rangle . \tag{1}$$

The total energy matrix elements in the lab are given by

$$\langle m|H|m'\rangle = -g\mu_n Bm\delta_{m,m'} + \langle m|H_Q|m'\rangle \tag{2}$$

and require knowledge of the electric part. Introducing projectors of the lab system, and using Eq. (1),

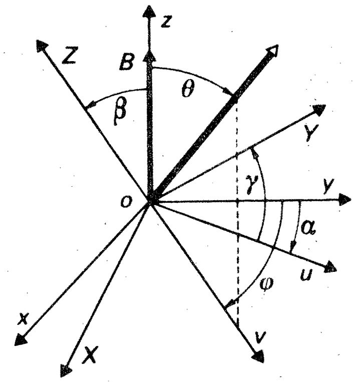

FIG. 1. The laboratory coordinate system (xyz) related to the principal axis system of EFG (XYZ) by the Eulerian angles α, β, and γ. The $\overrightarrow{0u}$ axis is the intersection of the two planes $\{0xy\}$ and $\{0XY\}$. The emitted radiation direction is shown as a double bar arrow, its projection on the $\{0xy\}$ plane is labeled $\overrightarrow{0v}$.

$$\begin{aligned}\langle m|H_Q|m'\rangle &= \sum_{\mu\mu'} \langle m|R|\mu\rangle\langle\mu|R^\dagger H_Q R|\mu'\rangle\langle\mu'|R^\dagger|m'\rangle \\ &= \sum_{\mu\mu'} \mathfrak{D}^I_{m,\mu}(\alpha\beta\gamma)\mathfrak{D}^{I*}_{m',\mu'}(\alpha\beta\gamma)\langle\tilde{\mu}|H_Q|\tilde{\mu}'\rangle \\ &= \sum_{\mu\mu'} e^{-i\alpha(m-m')}e^{-i\gamma(\mu-\mu')}d^I_{m,\mu}(\beta)d^{I*}_{m',\mu'}(\beta)\langle\mu|H_Q|\mu'\rangle ,\end{aligned}$$

where $\mathfrak{D}^I_{m,\mu}(\alpha\beta\gamma)$ are rotation matrix elements and $d^I_{m,\mu}(\beta) = d^{I*}_{m,\mu}(\beta)$ are reduced matrix elements.[2]

Writing Eq (2) as

$$\begin{aligned}\langle m|H|m'\rangle &= e^{-i\alpha(m-m')}\left\{-g\mu_n Bm\delta_{m,m'} + \sum_{\mu\mu'} e^{-i\gamma(\mu-\mu')}d^I_{m,\mu}(\beta)d^I_{m',\mu'}(\beta)\langle\tilde{\mu}|H_Q|\tilde{\mu}'\rangle\right\} \\ &= e^{-i\alpha(m-m')}\langle m|H(\alpha=0,\beta,\gamma)|m'\rangle ,\end{aligned}$$

the α dependence appears as a rotation $R(\alpha)$ around the z axis. Under this unitary transformation, the eigenvalues are invariant:

$$E_N(\alpha,\beta,\gamma) = E_N(\alpha=0,\beta,\gamma) . \tag{3}$$

The eigenvectors are transformed as

$$\begin{aligned}|N(\alpha,\beta,\gamma)\rangle &= R^\dagger_\alpha|N(\alpha=0,\beta,\gamma)\rangle \\ &= e^{i\alpha I_z}|N(\alpha=0,\beta,\gamma)\rangle\end{aligned}$$

and we get

$$\langle m|N(\alpha,\beta,\gamma)\rangle = e^{-im\alpha}\langle m|N(\alpha=0,\beta,\gamma)\rangle . \tag{4}$$

For axially symmetric EFG, $\eta = 0$ and the matrix element of H_Q in the PAS is diagonal:

$$\langle\tilde{\mu}|H_Q|\tilde{\mu}'\rangle = \frac{eQV_{ZZ}}{4I(2I-1)}[3\mu^2 - I(I+1)]\delta_{\mu\mu'} .$$

In this case, the total energy matrix element reduces to

$$\begin{aligned}\langle m|H(\alpha=0,\beta,\gamma)|m'\rangle &= -g\mu_n Bm\delta_{m,m'} + \frac{eQV_{ZZ}}{4I(2I-1)} \\ &\quad\times\sum_\mu d^I_{m,\mu}(\beta)d^I_{m',\mu}(\beta)[3\mu^2 - I(I+1)] .\end{aligned} \tag{5}$$

The γ dependence disappears, corresponding to the physical invariance of the interaction in the rotation of the microcrystal around the symmetry axis Z of PAS.

Our results have a different analytical form but are identical to those obtained by Matthias, Schneider, and Steffen,[3] just noting that they call the rotation around z a γ instead of α.

We now put

$$\hbar\omega_Q = \frac{eQV_{ZZ}}{4I(2I-1)},$$

$$\hbar\omega_B = g\mu_n B$$

and write Eq. (5) in the following way:

$$\langle m|H(\alpha=0,\beta,\gamma)|m'\rangle = \hbar\omega_B\left\{-m\delta_{m,m'} + \frac{\omega_Q}{\omega_B}\times\sum_\mu d^I_{m,\mu}(\beta)d^I_{m',\mu}(\beta)[3\mu^2 - I(I+1)]\right\}.$$

We remark that, taking $\hbar\omega_B$ just as a scale factor, the eigenvalues ϵ_N of the reduced Hamiltonian $H' = H/\hbar\omega_B$ depend only on β and ω_Q/ω_B and have the same eigenvectors as H:

$$H'|N(\beta,\hbar\omega_B,\hbar\omega_Q)\rangle = \epsilon_N\left(\beta,\frac{\omega_Q}{\omega_B}\right)|N(\beta,\hbar\omega_B,\hbar\omega_Q)\rangle,$$

$$|N(\beta,\hbar\omega_B,\hbar\omega_Q)\rangle = \left|N\left(\beta,\frac{\omega_Q}{\omega_B}\right)\right\rangle.$$

III. ANGULAR DISTRIBUTION

As the axial symmetry of the interaction is not conserved, we cannot use the well-known formula of directional distribution in each microcrystal:

$$W(\theta) = \sum_K (-)^K B_K A_K P_K(\cos\theta), \tag{6}$$

but we can use the general expression given by Steffen and Alder[4] in Eq. (12.294):

$$W(\theta,\varphi) = \sum_{K,n}(-)^K(2K+1)^{1/2}B^n_K A_K Y^n_K(\theta,\varphi=0)\cos n\varphi \tag{7}$$

with

$$B^n_K = \frac{1}{(2K+1)^{1/2}}\sum_m (-)^{I+m}\begin{pmatrix} I & I & K\\ -m & m' & n\end{pmatrix}\langle m|\rho|m'\rangle,$$

where θ and φ are the polar angles in lab of the propagation direction of the radiation (see Fig. 1). We introduce the normalized B^n_K coefficients in order to be consistent with the B_K notation of Eq. (6): $B^0_K = B_K$. They are related to the original statistical tensor ρ^n_K by the relation[4]

$$B^n_K = \frac{1}{2K+1}\rho^n_K.$$

The density matrix element $\langle m|\rho|m'\rangle$ has to be expressed in the eigenvectors $|N(\alpha\beta\gamma)\rangle$ of the I state:

$$\langle m|\rho|m'\rangle_{\alpha\beta\gamma} = \sum_{\substack{N(\alpha\beta\gamma)\\ N'(\alpha\beta\gamma)}}\langle m|N(\alpha\beta\gamma)\rangle\langle N(\alpha\beta\gamma)|\rho|N'(\alpha\beta\gamma)\rangle \times\langle N'(\alpha\beta\gamma)|m'\rangle.$$

As for low temperature nuclear orientation we have the diagonal Boltzmann distribution:

$$\langle N(\alpha\beta\gamma)|\rho|N'(\alpha,\beta,\gamma)\rangle = \frac{\exp(-E_{N(\alpha,\beta,\gamma)}/kT)}{\sum_{N''(\alpha\beta\gamma)}\exp(-E_{N''(\alpha,\beta,\gamma)}/kT)}\delta_{N,N'}.$$

The α dependence can be given explicitly using Eqs. (3) and (4):

$$B^n_K(\alpha,\beta,\gamma) = e^{in\alpha}B^n_K(\alpha=0,\beta,\gamma).$$

For polycrystalline samples, we have to integrate Eq. (7) over α, β, and γ:

$$W(\theta,\varphi) = \sum_{K,n}(-)^K(2K+1)^{1/2}A_K Y^n_K(\theta,\varphi=0)\cos n\varphi \times\int_0^{2\pi}e^{in\alpha}d\alpha \times\int_0^\pi\int_0^{2\pi}B^n_K(\alpha=0,\beta,\gamma)\sin\beta\, d\beta d\gamma.$$

Because the integration over α reduces to $2\pi\delta_{n,0}$, only B^0_K terms contribute to the angular distribution. This result indicates that, due to the random orientation of the PAS, the axial symmetry of the whole sample is conserved with the magnetic field direction as the symmetry axis. We get

$$W(\theta,\varphi) = \sum_K(-)^K(2K+1)^{1/2}A_K Y^0_K(\theta,\varphi=0)B_K$$

with

$$\bar{B}_K = 2\pi\int_0^\pi\int_0^{2\pi}B^0_K(\alpha=0,\beta,\gamma)\sin\beta\, d\beta d\gamma. \tag{8}$$

In the case of axially symmetric EFG as seen in the preceding section, neither eigenvalues nor eigenvectors in Eqs. (3) and (4) depends on γ, and Eq. (8) reduces to

$$\bar{B}_K = 4\pi^2\int_0^\pi B^0_K(\alpha=0,\beta,\gamma=0)\sin\beta\, d\beta. \tag{9}$$

These $\bar{B}_K$ appear as the actual nuclear orientation coefficients and depend on $\hbar\omega_B/kT$ and ω_Q/ω_B. In Eqs. (8) and (9) the 2π and $4\pi^2$ factors will disappear in the renormalization procedure of the $\bar{B}_K$ coefficients to $\bar{B}_0 = 1$.

The influence of the random oriented quadrupole interaction is to induce an attentuation factor G_K defined by the ratio

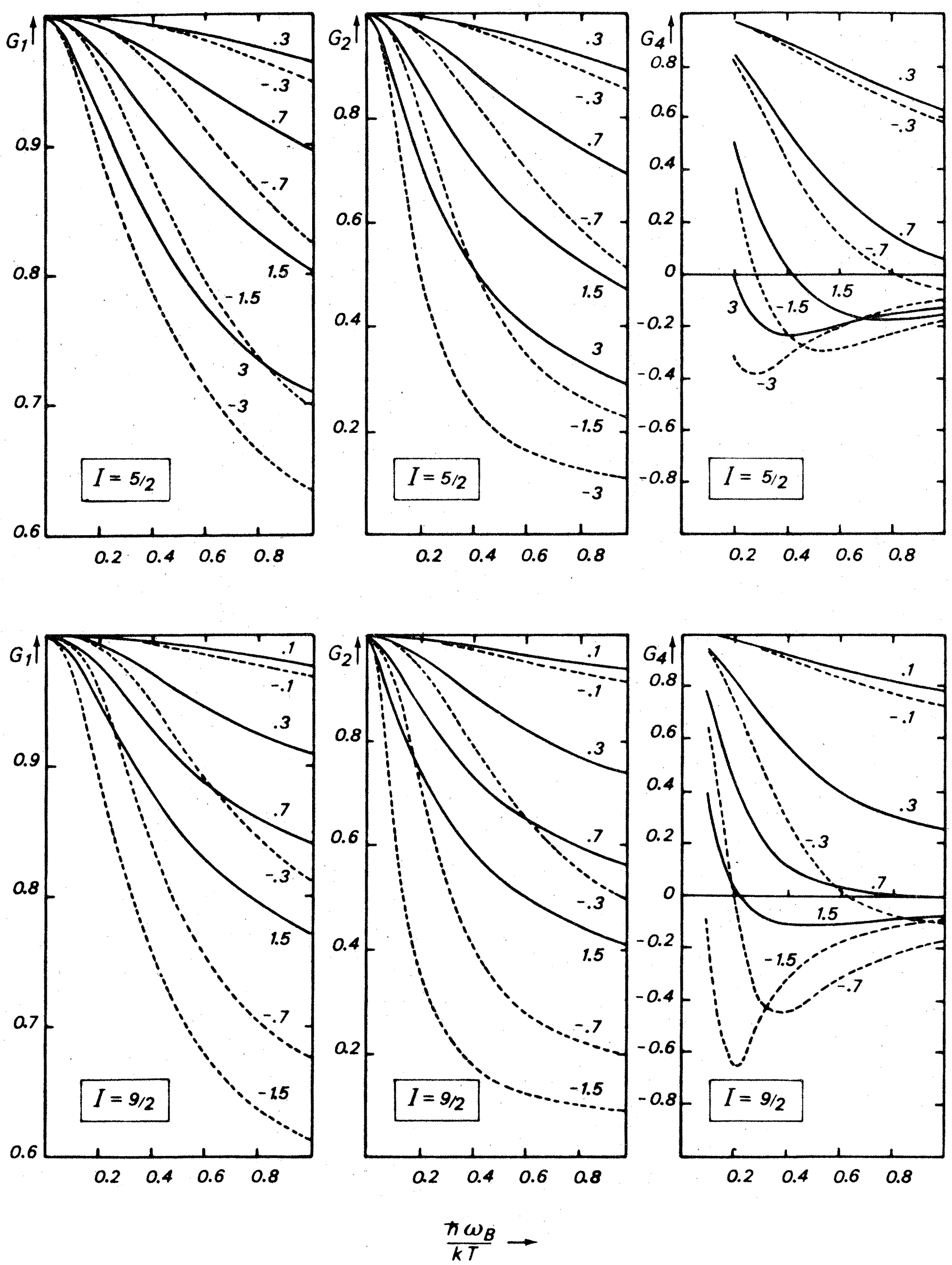

FIG. 2. Calculated attenuation coefficients, for spin $I=\frac{5}{2}$ and $\frac{9}{2}$, plotted for some values of the ratio $3\omega_Q/\omega_B$ used as a parameter on the curves. The positive and negative values of this parameter are, respectively, drawn in full and dashed lines. $G4$ curves are not drawn for $\hbar\omega_B/kT$ values giving $B4$ coefficients less than 10^{-4}. In the tabulation, the ratio $3\omega_Q/\omega_B$ is employed to be consistent with the $\Delta E/T$ notation introduced by Krane (Ref. 5).

$$G_K\left(\frac{\hbar\omega_B}{kT},\frac{\omega_Q}{\omega_B}\right)=\frac{\bar{B}_K[(\hbar\omega_B/kT),(\omega_Q/\omega_B)]}{B_K(\hbar\omega_B/kT)},$$

where B_K are the nuclear orientation coefficents for the pure magnetic interaction case, already tabulated by Krane.[5]

IV. NUMERICAL RESULTS

In order to estimate the remaining anisotropy we tabulate the values of the G_K coefficients as function of the orientation parameter $\hbar\omega_B/kT$ for a wide range of the parameter ω_Q/ω_B and several spin values, only for the case of the axially symmetric field gradient. These numerical calculations have been performed on a CDC6600 computer. The diagonalization procedure of the combined H' Hamiltonian has been done using the Givens-Householder method with Wilkinson inverse iterations, giving eigenvalues equal to those obtained by Matthias *et al.*[3] within 10^{-5}. For the integration over β, we apply the Gauss method and test the convergency of the results using various integration steps. For a 24 step procedure the precision is better than 10^{-5}. When the B_K coefficients are smaller than 10^{-4} we do not calculate the corresponding G_K because they do not contribute to the orientation and are not of any experimental interest. We have tabulated[6] the G_K factors for $K=1$, 2, 4 and spin values $I=1, \frac{3}{2}, \ldots, 8$. for the ratio $3\omega_Q/\omega_B$ varying from ± 0.05 to ± 10 and for orientation parameter $\hbar\omega_B/kT$ from 0.01 to 10. Typical attenuation curves for spin $\frac{5}{2}$ and $\frac{9}{2}$ are shown in Fig. 2.

Some general trends can be found from our results:

(1) The attenuation is larger on the coefficients of higher rank, so $G1>G2>G4$.
(2) The results are sensitive to the sign of the ratio ω_0/ω_B in contrast to the perturbed angular correlation method. This fact can be understood by the Boltzmann statistic.

Some of the brute force nuclear orientation experiments[7,8] reveal attenuation. If these are owing to a stable association of the radioactive atom with an impurity or a crystal defect, the calculation can be applied with an electric field gradient, assumed randomly oriented, for the case of polycrystalline samples. For example, we estimated the field gradient value needed to explain the attenuation in the Oxford experiment on ^{111}In in Cu[8] to be

$$V_{ZZ} \simeq +2.7\times 10^{19} \text{ or } -0.9\times 10^{19}\ \text{V/cm}^2 .$$

These values are higher than the usual ones found in noncubic metals. However, other measurements[9] indicate such large values. If this assumption is verified, these electric field gradients could be used to measure quadrupole moments of nuclei.

The authors thank Professor I. Berkes for helpful discussions and comments, and are very indebted to Dr. J. Meyer for his interest in this work and for enlightening advice concerning computational tests.

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