EFFECT OF MAGNETIC RESONANCE ON THE $\gamma\gamma$ ANGULAR CORRELATION

F. P. KHAĬMOVICH

Kazan' Physico-Technical Institute, Academy of Sciences, U.S.S.R.

Submitted July 14, 1967

Zh. Eksp. Teor. Fiz. 54, 292–297 (January, 1968)

Resonance phenomena due to an oscillating magnetic field arising in the cascade emission of two γ quanta by radioactive nuclei in a weak or strong static magnetic field are considered. An expression for the correlation attenuation factor is obtained for the case of a weak magnetic field. It is shown that the angular correlation changes abruptly as the magnetic resonance frequency is approached. "Allowed" and "forbidden" electronic paramagnetic resonance transitions leading to a resonance in the angular correlation are considered separately for the case of a strong field. These phenomena may be used for an exact determination of the g factor of excited nuclear states and also as a sensitive means of detecting the electron paramagnetic resonance in small concentrations of radioactive atoms.

1. INTRODUCTION

ABRAGAM and Pound[1] have proposed to detect the nuclear magnetic resonance through the change of the $\gamma\gamma$ angular correlation under the influence of a strong oscillating magnetic field. This idea is based on the fact that in the double γ decay of radioactive nuclei, the first radiation leads to a non-uniform population of the magnetic sublevels while the oscillating field with the resonance frequency tries to make the population the same in all sublevels. The experimental realization of this proposal would allow one to measure very accurately the magnetic moments of excited nuclear states. However, the correlation of emitted γ rays is destroyed in those cases where the lifetime of the intermediate state of the nucleus τ_n is comparable to or larger than the time necessary for a change of direction of the spin of the nucleus under the influence of the external field. Usually τ_n is small, so that this type of experiment cannot be carried out. The same authors have pointed out that this difficulty can be avoided by inducing resonance transitions between hyperfine sublevels without a static magnetic field in atoms with non-vanishing electronic moment.

On the basis of the ideas of [1] we propose in the present paper a theory of resonance effects in the double γ decay of nuclei in atoms with an unfilled electron shell. The problem is solved in two limiting cases: for a weak and for a strong magnetic field. The condition of a weak field has the form $g\beta H < A$, where g is the g factor of the electron shell of the paramagnetic atom containing the radioactive nucleus, β is the Bohr magneton, H is the constant magnetic field, and A is the constant of the hyperfine interaction. In this case the total atomic spin is $F = I + J$, where I and J are the nuclear and electron spins, respectively. Since the coupling of the atomic spin F to the radiofrequency field is a thousand times stronger than that of the nuclear spin, the decay probability during the time τ_n increases by a factor $\sim 10^6$. In the case of a strong field, we consider separately the "allowed" electronic paramagnetic resonance transitions which affect the angular correlation through the change in the population of the electronic sublevels, and the "forbidden" transitions which are accompanied by a reorientation of the electronic and nuclear spins. These cases are of particular interest, since they can be used for an exact determination of the g factor of excited levels with small τ_n. Besides the usual resonance effects there is a modulation of the angular correlation with respect to frequency, analogous to the light beats.[2,3]

The problem is solved by taking exact account of the influence of the oscillating field with frequency ω by going over to a rotating coordinate system. This method is often used in the theory of the electronic paramagnetic resonance and the radiooptical resonance.[3,4] We use the known formulas of the theory of angular correlations:[5,6]

$$W(\mathbf{k}_1, \mathbf{k}_2, t) = \sum_{k_1, k_2, N_1, N_2} A_{k_1}(1) A_{k_2}(2) G_{k_1 k_2}^{N_1 N_2}(t) \times [(2k_1+1)(2k_2+1)]^{-1/2} Y_{k_1}^{N_1}(\theta_1, \varphi_1) Y_{k_2}^{N_2}(\theta_2, \varphi_2). \quad (1)$$

Here $W(\mathbf{k}_1, \mathbf{k}_2, t)$ is the probability for emission of two γ quanta in the directions $\mathbf{k}_1$ and $\mathbf{k}_2$ during the time t: Y_k^N is the Nth spherical harmonic; $G_{k_1 k_2}^{N_1 N_2}(t)$ is the angular correlation attenuation factor taking account of the effect of the external fields:

$$G_{k_1 k_2}^{N_1 N_2}(t) = \sum_{m_a m_b} (-1)^{2I+m_a+m_b} [(2k_1+1)(2k_2+1)]^{1/2} \begin{pmatrix} I & I & k_1 \\ m_a' & -m_a & N_1 \end{pmatrix} \times \begin{pmatrix} I & I & k_2 \\ m_b' & -m_b & N_2 \end{pmatrix} \langle m_b | L(t) | m_a \rangle \langle m_b' | L(t) | m_a' \rangle^*, \quad (2)$$

where (:::) is the 3j symbol of Wigner; $|m_b\rangle$ is the wave function of the nucleus in the intermediate state with $I_z = m_b$; L(t) is the evolution operator which satisfies the Schrödinger equation.

Up to this time, the calculations of $W(\mathbf{k}_1, \mathbf{k}_2, t)$ and $G_{k_1 k_2}^{N_1 N_2}(t)$ in the presence of a time dependent perturbation have made use of approximate methods in which the interference terms entering in (1) and (2) were neglected. The advantage of the method proposed by us

lies primarily in the fact that it allows one to take the effect of these terms into account.

2. WEAK FIELD

Let us assume that the coupled system electronic shell + nucleus is in a crystal with a symmetry axis z along which a constant magnetic field H is directed and perpendicular to which a variable magnetic field with amplitude H_1 is applied. The Hamiltonian for the closed system has the form

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_1(t) = AJ_zI_z + B(J_+I_- + J_-I_+) + \omega_0F_z + {}^1/_2(F_+e^{-i\omega t} + F_-e^{i\omega t}). \quad (3)$$

Here $\omega_0 = g_F\beta H$, $\omega_1 = g_F\beta H_1$, where g_F is the atomic g factor and $F_\pm = 2^{1/2}(F_x \pm iF_y)$.

To determine (2), we must calculate the matrix elements of the evolution operator, which satisfies the Schrödinger equation

$$\frac{\partial}{\partial t}L(t) = -\frac{i}{\hbar}\mathcal{H}L(t). \quad (4)$$

Since the operator $F_Z = I_Z + J_Z$ commutes with the time independent part of the Hamiltonian $\mathcal{H}_0$, it is convenient to go over to a coordinate system which rotates about the z axis with an angular velocity ω for which the interaction Hamiltonian becomes independent of the time:

$$\mathcal{H}_1'(t) = \exp[-(i/\hbar)F_z\omega t]\mathcal{H}_1(t)\exp[(i/\hbar)F_z\omega t] = \omega_1F_x. \quad (5)$$

Solving (4) in the rotating coordinate system and then returning to the original system, we find

$$L(t) = \exp[-(i/\hbar)F_z\omega t]\exp[-(i/\hbar)(\mathcal{H}_0 + \mathcal{H}_1' - \omega F_z)t]. \quad (6)$$

The calculation of the matrix elements in (2) is performed in three steps. First we find the matrix elements $\langle m_F|L(t)|m_F'\rangle$ in the coordinate system whose z' axis coincides with the direction of the effective field H_{eff}^F:

$$H_{eff}^F = [(\omega_0 - \omega)^2 + \omega_1^2]^{1/2}/g_F\beta. \quad (7)$$

We then go over to the old coordinate system, taking into account that the state vectors in the old and new systems are related by a linear transformation:

$$|m_F\rangle = \sum_\mu |\mu_F\rangle\langle\mu_F|m_F\rangle = \sum_\mu D(0,\alpha,0)_{m\mu}|m_F\rangle, \quad (8)$$

where $D(0, \alpha, 0)_{m\mu}$ is Wigner's rotation matrix and $\alpha = \arctan(\omega_1/H_{eff}^F)$. Finally, using

$$|Fm_F\rangle = (-1)^{J-I+m}\sqrt{2F+1}\sum_{mM}\begin{pmatrix} I & J & F \\ m & M & m_F\end{pmatrix}|JM\rangle|Im\rangle, \quad (9)$$

we go over to the original matrix elements. In this way we find

$$G_{k_1k_2}^{N_1N}(t) = \sum_{m_am_b}\sum_{F,m_F,F',m_F'}(-1)^{2I+m_a+m_b}[(2k_1+1)(2k_2+1)]^{1/2} \times\begin{pmatrix} I & I & k_1 \\ m_a' & -m_a & N_1\end{pmatrix}\begin{pmatrix} I & I & k_2 \\ m_b' & -m_b & N_2\end{pmatrix}\begin{pmatrix} I & J & F \\ m_b & M & -m_F\end{pmatrix} \times\begin{pmatrix} I & J & F \\ m_a & M' & m_F'\end{pmatrix}\begin{pmatrix} I & J & F' \\ m_b' & M'' & -m_F'\end{pmatrix}\begin{pmatrix} I & J & F' \\ m_a' & M''' & -m_F'\end{pmatrix} \times\langle m_F|\mu_F\rangle\langle\mu_F|m_F'\rangle\langle m_F'|\mu_F'\rangle\langle\mu_F'|m_F'\rangle. \times\exp\{[(i/\hbar)(\lambda_\mu - \lambda_{\mu'}) - (m_a' - m_b')\omega]t - \Gamma t\} \quad (10)$$

Here $\lambda_\mu - \lambda_{\mu'} = (\mu - \mu')g_F\beta H_{eff}^F$, and $\Gamma = \hbar/\tau_n$. In deriving (10) we have neglected the spin-spin interactions, since the concentrations of radioactive atoms are usually very small. We have also assumed that the spin-lattice relaxation time $T_1 > \tau_n$. In some cases this condition is only fulfilled at helium temperatures.

Formula (10) takes the simplest form for vanishing magnetic field in the case of an isotropic hyperfine interaction. These conditions can be satisfied in gases, stabilized atoms,[7] or in crystals with a cubic lattice. In this case we have for the integral correlation attenuation factor $\overline{G_{kk}^{NN}(t)}$:

$$\overline{G_{kk}^{NN}}(t) = \frac{1}{\tau_n}\int_0^\infty e^{-t/\tau_n}G_{kk}^{NN}(t)\,dt = \sum_{FF'}[(2k_1+1)(2k_2+1)]^{1/2}(2F+1)(2F'+1) \times\begin{Bmatrix} F & F' & k_1 \\ I & I & J\end{Bmatrix}\begin{Bmatrix} F & F' & k_2 \\ I & I & J\end{Bmatrix}\sum_{m_Fm_F'}\begin{pmatrix} F & F' & k_1 \\ -m_F & m_F' & N\end{pmatrix}\begin{pmatrix} F & F' & k_2 \\ -m_F & m_F' & N\end{pmatrix} \times\frac{\langle m_F|\mu_F\rangle^2\langle m_F'|\mu_F'\rangle^2}{1+[(\lambda_\mu - \lambda_{\mu'})\tau_n/\hbar]^2}. \quad (11)$$

Here $\{:::\}$ is a Racah coefficient.

It follows from (10) and (11) that the dependence of the correlation attenuation coefficient on the magnetic field has resonance character. According to (7), the resonance occurs for $\omega_0 = \omega$. It is seen from (11) that the observation of this resonance requires $(\lambda_\mu - \lambda_{\mu'})\tau_n/\hbar \sim 1$. For the isotope Gd^{111} with $\tau_n = 8.5 \times 10^{-8}$ sec this requirement can be fulfilled with no particular difficulty. It also follows from (10) and (11) that $G_{k_1k_2}^{N_1N_2}(t)$ is modulated with the frequencies $[(\lambda_\mu - \lambda_{\mu'}) - (m_a' - m_b')\omega]$. This corresponds to the "beats" in the intensity in the double radio optical resonance.[3]

3. STRONG FIELD

Let us assume that $g\beta H \gg A$. We first consider the effect of an isotropic hyperfine interaction in the absence of the variable field. In the theory of angular correlations one usually restricts oneself to the approximation in which the wave functions of the nucleus and of the electron are independent and the total wave function can be written in the form $|M\rangle|m\rangle$. In the first approximation in the hyperfine interaction[8] we have

$$|M,m\rangle = q_0|M\rangle|m\rangle + q_\pm|M \pm 1\rangle|m \mp 1\rangle, \quad (12)$$

where

$$q_0^2 + q_+^2 + q_-^2 = 1, \quad q_\pm = \frac{A\langle M|J_\pm|M \pm 1\rangle\langle m \mp 1|I_\mp|m\rangle}{2g\beta H}. \quad (13)$$

The energy levels of the intermediate state of the nucleus E_m are equal to

$$E_m = AmM - g_I\beta_I mH - \frac{A^2}{2g\beta H}\{m^2M - m[J(J+1) - M^2]\}. \quad (14)$$

Taking account of the populations of the electronic Zeeman sublevels p_M, we obtain, using (12) and (14),

$$G_{k_1k_2}^{NN}(t)=\sum_M p_M \sum_m [(2k_1+1)(2k_2+1)]^{1/2}\begin{pmatrix} I & I & k_1 \\ m' & -m & N\end{pmatrix}$$
$$\times\begin{pmatrix} I & I & k_2 \\ m' & -m & N\end{pmatrix}\exp\left\{-\frac{i}{\hbar}(AM-g_I\beta_I H-[J(J+1)-M^2])Nt\right\},$$
$$\times\exp\left\{-\frac{i}{\hbar}\left[\frac{A^2}{2g\beta H}(m^2-m'^2)M\right]t\right\}. \qquad (15)$$

If the magnetic field is directed along the direction of propagation of one of the quanta, for example, $\mathbf{k}_1$, then $\theta_1 = 0$ in (1) and $Y_{k_1}^N(\theta_1, \varphi_1) = [(2k_1+1)/4\pi]^{1/2}\delta_{N0}$. For N = 0, formula (15) leads to the following criterion for a full restoration of the angular correlation up to its unperturbed value:

$$\frac{A^2}{2g\beta H}(m^2-m'^2)M\tau_R/\hbar \ll 1. \qquad (16)$$

For the isotope Gd^{154} formula (16) yields H = 2000 Oe. Experimentally,[9] a full restoration of the angular correlation was observed for H = 3300 Oe. We see that the criterion (16) gives good qualitative agreement with experiment, whereas theory up to now[6] yielded the result that full restoration of the angular correlation must occur for H ~ 100 Oe, and dubious assumptions had to be made to explain the experiments.

Let us now consider the effect of a radio-frequency field. An "allowed" electronic paramagnetic resonance transition does not affect the projections of the nuclear magnetic moment, but changes the population of the electronic Zeeman sublevels p_M in (15), which leads to a change of the correlation attenuation factor. Thus there is a possibility to detect, by a change in the angular correlation, an electronic paramagnetic resonance in a small number of atoms whose nuclei are in an excited state.

The "forbidden" transitions ($\Delta M = \pm 1$, $\Delta m = \mp 1$) affect the angular correlations directly. It follows from (14) that these transitions can be used for an exact determination of the g factors of excited nuclei. By calculations analogous to the ones leading to (10) and (11), we find for the correlation attenuation factor $G'^{NN}_{kk}(t)$ in the presence of "forbidden" transitions:

$$G_{kk}'^{NN}(t)=G_{kk}^{NN}(t)\left\{q_0^2+q_\pm^2\sum_{\mu\mu'}\langle M|\mu\rangle^2\right.$$
$$\left.\times\langle\mu'|M'\rangle^2\exp[-(i/\hbar)(\lambda_\mu-\lambda_{\mu'})t-\Gamma t]\right\}. \qquad (17)$$

It should be noted that the nuclei in the intermediate state have a rather large spin so that $q_\pm^2$ may attain ~0.1. The rotation matrices in (17) have a resonance character of the same type as in the case of a weak field.

In contrast to the isotropic case considered by us, additional resonance peaks appear in crystals in which the hyperfine interaction has axial symmetry; the position of these peaks is determined by

$$(\mu-\mu')g\beta H_{eff}-(m-m')\omega=0, \qquad (18)$$

where

$$H_{eff}=[(\omega-\omega_0)^2+\omega_1^2]^{1/2}/g\beta.$$

As an example we point out that in the isotopes Sm^{152}, Gd^{154}, Gd^{156}, Dy^{160}, and Er^{166} for which I = 2, the strongest resonance peaks for small values of ω_1 correspond to the frequencies $\omega = \omega_0/2$ and $\omega = 3\omega_0/2$. Additional peaks approach as ω_1 is increased. When $\omega_1 = \omega_0/2$, these coalesce with the main peak at the frequency ω_0.

[1] A. Abragam and R. V. Pound, Phys. Rev. **92**, 943 (1953).
[2] J. Brossel and F. Bitter, Phys. Rev. **86**, 308 (1952).
[3] J. N. Dodd and G. W. Series, Proc. Roy. Soc. **A263**, 353 (1961).
[4] V. G. Pokazan'ev and G. V. Skrotskiĭ, Zh. Eksp. Teor. Fiz. **49**, 163 (1965) [Sov. Phys.—JETP **22**, 117 (1966)].
[5] Collection Gamma-luchi (Gamma Rays), AN SSSR, 1961, p. 554.
[6] Int. Conf. on Perturbed Angular Correlations, ed. by E. Karlsson, E. Matthias, and K. Siegbahn, North-Holland, Amsterdam, 1964.
[7] R. A. Zhitnikov, N. V. Kolesnikov, and V. I. Kosyakov, Zh. Eksp. Teor. Fiz. **42**, 1186 (1962) [Sov. Phys.—JETP **16**, 839 (1962)].
[8] C. D. Jeffries, Dynamic Nuclear Orientation, Interscience, N. Y., 1963; Russ. Transl. Mir, 1965.
[9] R. Stiening and M. Deutsch, Phys. Rev. **121**, 1484 (1961).

Translated by R. Lipperheide
36