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GAMMA MAGNETIC RESONANCE

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Two-quantum resonance in the Mossbauer effect, which is a process consisting of simultaneous absorption of a Mossbauer quantum and a quantum of the external radio frequency field, is considered. The calculation is performed for Mossbauer nuclei imbedded in a ferromagnet. In this case the main contribution to the γ-ray absorption is from nuclei located in the Bloch walls. The analysis shows that transitions forbidden in one-quantum processes are made allowed by γ-magnetic resonance. Estimates show that an experimental detection of the lines should be feasible.

1. INTRODUCTION

THE Mossbauer effect is continuing to find more applications in solid-state physics [1]. Greatly contributing to this is the adoption by γ-resonance spectroscopy of all the methods which have become widely known in the radio and optical bands. We therefore deemed it of interest to realize two-quantum transitions in the Mossbauer effect [2]. In this paper we consider the case when the two-quantum transitions are produced by simultaneous action of γ radiation and a radio-frequency magnetic field on the absorber nuclei.

2. HAMILTONIAN OF THE QUANTUM SYSTEM

Let us consider Mossbauer nuclei placed in a ferromagnet. If we neglect the interaction between the nuclei and coherent effects [3], then the problem of the interaction of the nucleus with the radiation field can be solved for each nucleus separately. Then the Hamiltonian of the system in the single-particle approximation is written as follows:

$$\hat{\mathscr{H}} = \hat{\mathscr{H}}_0 + \hat{\mathscr{H}}_\gamma + \hat{\mathscr{H}}_{rf}, \qquad \hat{\mathscr{H}}_0 = \hat{\mathscr{H}}_a + \hat{\mathscr{H}}_b, \tag{1}$$

where $\hat{\mathscr{H}}_0$ is the Hamiltonian of the unperturbed nucleus, $\hat{\mathscr{H}}_\gamma$ is the Hamiltonian of the interaction with the γ radiation, and $\hat{\mathscr{H}}_{rf}$ is the Hamiltonian of interaction of the nucleus with the radio-frequency magnetic field, and $\hat{\mathscr{H}}_a$ and $\hat{\mathscr{H}}_b$ are the Hamiltonians of the spectrum of the ground and excited states [1)].

A. Spectrum of the System

For all real temperatures, the energy interval corresponding to the transitions $a \longleftrightarrow b$ will be such that $(E_b - E_a)/kT \gg 1$ always. This leads to population of only the ground-state levels. In addition, we assume that the ground and excited levels are determined by nuclear spins j_a and j_b. In a ferromagnet under the influence of the hyperfine magnetic field, these levels become split, forming Zeeman multiplets. However, in these splittings, in spite of the tremendous values of the hyperfine field (on the order of 10^5 G), are much smaller than the value of kT, down to infralow temperatures. Therefore the levels of one and the same multiplet are equally populated. If we neglect the quadrupole interaction of the nucleus with the electric field of the surrounding, we can assume furthermore that the levels of the ground and excited states are equidistant.

B. γ Radiation. Radiative Transitions

The Hamiltonian of the interaction of the γ radiation with the nucleus will be determined by assuming it to differ from zero only for transitions between ground and excited states. Describing it by the well-known formula

$$\hat{\mathscr{H}}_\gamma = \mathbf{A}\mathbf{j}, \tag{2}$$

where j is the current vector and A is the vector potential, we can readily obtain an expression for the matrix element of the operator $\hat{\mathscr{H}}_\gamma$. In the transition $a \rightarrow b$ with simultaneous absorption of the γ quantum we have

$$\langle b, n_\gamma - 1 | \hat{\mathscr{H}}_\gamma | n_\gamma, a \rangle = \left(\frac{2\pi\hbar c}{L^3} \frac{n_\gamma}{k_\gamma} \right)^{1/2} \exp[-i\omega_\gamma t]\, \mathbf{I}\mathbf{J}_{b,a}(-\mathbf{k}_\gamma), \tag{3}$$

1)The indices a and b will henceforth denote the ground and excited states, respectively.

$$\mathbf{J}_{b,a}(-k_\gamma) = \langle b|\exp\{-i\mathbf{k}_\gamma \mathbf{r}\}\mathbf{j}|a\rangle, \quad (3a)$$

where n_γ is the quantum number of the harmonic oscillator and determines the intensity of the incident flux of the γ quanta, ω_γ is the cyclic frequency of the γ quantum, k_γ is the wave vector of the γ quantum, directed from the source to the absorber, and $\mathbf{l}$ is the polarization vector of the incident wave.

The vector $\mathbf{J}_{b,a}(-\mathbf{k}_\gamma)$ can be represented in the following fashion [4] 2):

$$\mathbf{J}_{b,a}(-\mathbf{k}_\gamma) = \chi(M)(-)^{m_a - m_b}\begin{pmatrix} j_b & 1 & j_a \\ m_b & m_a - m_b & -m_a \end{pmatrix} \cdot$$

$$\times \sum_\nu \nu \xi_\nu D''_{\nu\, m_a - m_b}(\alpha', \beta', \gamma'); \quad \nu = \pm 1, \quad (4)$$

$$\xi_{\pm 1} = \mp 2^{-1/2}(\mathbf{e}_x \pm i\mathbf{e}_y), \quad \xi_0 = \mathbf{e}_z, \quad (4a)$$

where $\mathbf{e}_x$, $\mathbf{e}_y$, and $\mathbf{e}_z$ are the unit vectors of the laboratory frame, with $\mathbf{e}_z$ parallel to $\mathbf{k}_\gamma$; m_a and m_b are the magnetic quantum numbers of the nuclear moments, and $(:::)$ are Wigner 3j-symbols; $\chi(M)$ is a certain constant factor; $D^{(1)}_{m,\mu}(\alpha', \beta', \gamma') = \exp\{-im\alpha'\} d_{m\mu}(\beta')\exp\{-i\mu\gamma'\}$; finally, the Euler angles $(\alpha', \beta', \gamma')$ define a coordinate frame in which the Oz'' axis is the quantization axis relative to the laboratory coordinate frame.

We shall consider henceforth only isotropic g-factors of the ground and excited states. In this case, the direction of the quantization axis coincides with the direction of the constant magnetic field at the nucleus [5].

C. Interaction of Alternating Magnetic Field with the Nuclear Moment. Bloch Wall.

Let us consider the interaction between an alternating magnetic field and the nucleus in the absence of an external constant magnetic field. In this case, owing to the tremendous amplification of the alternating magnetic field at the nucleus, we can assume that the nuclei interacting with the perturbation are located only in the Bloch walls [6].

Let us now determine the operator $\mathscr{H}_{rf}$ for a nucleus situated in a 180-degree Bloch wall. It is easy to get

$$\mathscr{H}_{rf} = -\eta \sin\theta g \beta_N I_{x''} H_{z'}^1 \cos\omega t, \quad (5)$$

where η is a gain factor on the order of 10^3, g denotes the g-factor, β_N is the Bohr nuclear magneton, and $I_{x''}$ is the nuclear-spin component in the coordinate system x'', y'', z'' (see the figure) whose axes are rotated relative to the coordinate system x', y', z' by the Euler angles $(0, \theta, 0)$ (the Oz'' axis of this system coincides with the quantization axis), H_Z^1 is the amplitude of the projection of the alternating magnetic field on the easy magnetization direction, and ω is the frequency of the alternating magnetic field.

2)We assume here that the radiative transitions are purely magnetic-dipole.

The component $H_{Z'}^1$ can be expressed in terms of the corresponding components in the laboratory frame:

$$H_{z'}^1 = \sum_\mu D^{(1)}_{\mu,0}(\alpha, \beta, \gamma) H_\mu^1, \quad (6)$$

where the angles (α, β, γ) define the coordinate system $x'y'z'$ relative to the laboratory coordinate frame, $H^1_\mu = \mathbf{H}^1 \cdot \xi^*\mu$ are the components of the alternating magnetic field in the laboratory coordinate frame. With the aid of (6) we rewrite (5) in a final form:

$$\mathscr{H}_{rf} = -\eta \sin\theta g \beta_N I_{x''} \times \sum_\mu D^{(1)}_{\mu,0}(\alpha, \beta, \gamma) H_\mu^1 \cos(\omega t). \quad (7)$$

Let us also refine the expression (4). Using the group property of the D-functions, we can write

$$D^{(1)}_{\mu\mu'}(\alpha', \beta', \gamma') = \sum_{\mu''} D^{(1)}_{\mu\mu''}(\alpha, \beta, \gamma) D^{(1)}_{\mu'', \mu'}(0, \theta, 0). \quad (8)$$

This makes it possible for us to represent $\langle b, n_\gamma - 1|H_\gamma|n_\gamma, a\rangle$ in the following form

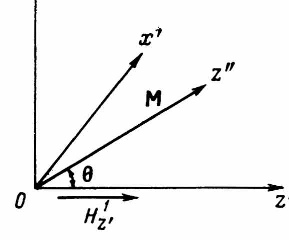
$$\langle b, n_\gamma - 1|\mathscr{H}_\gamma|n_\gamma, a\rangle = \left(\frac{2\pi\hbar c}{L^3}\frac{n_\gamma}{k_\gamma}\right)^{1/2} \chi(M)$$

$$\times (-)^{m_a - m_b}\begin{pmatrix} j_b & 1 & j_a \\ m_b & m_a - m_b & -m_a \end{pmatrix} e^{-i\omega_\gamma t} \sum_\nu \nu l_\nu^*$$

$$\times \sum_{m'} D^{(1)*}_{\nu, m'}(\alpha, \beta, \gamma) D^{(1)*}_{m', m_a - m_b}(0, \theta, 0). \quad (9)$$

3. PROBABILITY OF TWO-QUANTUM TRANSITION

The probability per unit time of the absorption of the γ quantum by a nucleus, if the latter goes



180-degree Bloch wall. The angle θ between the magnetization M and the easy-magnetization axis Oz' changes from 0 to π. The faces of the wall are parallel to the z'Ox' plane.

over in this case from the state a into the state b, can be written in the following manner:

$$W_{a\to b}=\lim_{t\to\infty}\frac{1}{t}\,|\langle b,n_\gamma-1|\hat{S}(t,0)|n_\gamma,a\rangle|^2, \tag{10}$$

where

$$\hat{S}(t,0)=T\exp\left[-\frac{i}{\hbar}\int_0^t\{\hat{\mathscr{H}}_\gamma'(t')+\hat{\mathscr{H}}_{rf}'(t')\}\,dt'\right],$$

$$\hat{\mathscr{H}}'(i)=\exp\left[\frac{it}{\hbar}\hat{\mathscr{H}}_0\right]\hat{\mathscr{H}}\operatorname{xep}\left[-\frac{it}{\hbar}\hat{\mathscr{H}}_0\right].$$

Considering in (10) terms of second order in the perturbation, we obtain the usual formula for the two-quantum transitions:

$$W_{a\to b}=W^+_{a\to b}+W^-_{a\to b}, \tag{11}$$

$$W^+_{a\to b}(\omega)=W^-_{a\to b}(-\omega), \tag{12}$$

$$W^+_{a\to b}(\omega)=\frac{\pi}{2\hbar^4}\left|\sum_s\left\{\frac{R_{bs}Q_{sa}}{\omega_{s,a}+\omega}+\frac{Q_{bs}R_{sa}}{\omega_{s,a}-\omega_\gamma}\right\}\right|^2$$
$$\times\,\delta(\omega_{b,a}-\omega_\gamma+\omega). \tag{13}$$

Here $R_{s,s'}$ and $Q_{s,s'}$ are determined by the following identities:

$$\langle s,n_\gamma-1|\hat{\mathscr{H}}_\gamma|n_\gamma,s\rangle=R_{s,s'}\exp\{-i\omega_\gamma t\}, \tag{14}$$

$$\langle s|\hat{\mathscr{H}}_{rf}|s'\rangle=Q_{ss'}\cos(\omega t). \tag{15}$$

From formulas (11)–(13) we see that the resonant frequency of the two-quantum transition a → b differs from the resonant frequency of the single-quantum transition a → b by $\pm\omega$ (provided, of course, that neither is forbidden).

4. AVERAGING. EFFECTIVE CROSS SECTION OF TWO-QUANTUM PROCESS

We shall henceforth carry out the calculations for a fine powder consisting of particles containing several domains. The ratio of the volume of the Bloch walls to the total volume of such particles is $\Lambda = 1/7$ [6]. We note also that a single-domain structure, say, in iron begins with particle dimensions $\leq 10^{-6}$ cm [7]. Averaging over all possible orientations of the powder particles will be carried out by taking the probability integral

$$\frac{1}{8\pi^2}\int d\Omega=\frac{1}{8\pi^2}\int_0^{2\pi}d\alpha\int_0^{\pi}\sin\beta\,d\beta\int_0^{2\pi}d\gamma.$$

In addition, we need to average over the nuclei contained in the Bloch wall. To this end we count the number of nuclei dN (see the figure) situated in an infinitesimally thin layer dy′ bounded by planes parallel to x′Oz′:

$$dN=An_0dy', \tag{16}$$

where A is the area of the face of the Bloch wall, and n_0 is the number of nuclei per unit volume. Substituting in (16) $dy'/d\theta = \delta/\sin\theta$ [8], where δ is the thickness of the Bloch wall, we obtain

$$dN/N=d\theta/\sin\theta. \tag{17}$$

We see that the relative content of the nuclei in the interval $d\theta$ becomes infinite at the points $\theta = 0$ and $\theta = \pi$. This is due to the imperfection of the theory of magnetization in the Bloch walls. However, there will be no divergences in the expression for the average probability, since the integrand is in final analysis proportional to $\sin\theta$.

Substituting (7) and (9) in (11)–(13) and averaging, we obtain the average probability. Using further the theorem of spectroscopic stability and comparing the probabilities of the two-quantum and single-quantum processes, we can obtain the normalized values of the effective absorption cross section in two-quantum transitions 3):

$$\sigma_{a\to b}=\sigma^+_{a\to b}+\sigma^-_{a\to b}; \tag{18}$$

$$\sigma^+_{a\to b}(\omega)=\sigma^-_{a\to b}(-\omega); \tag{19}$$

$$\sigma^+_{a\to b}=\frac{\sigma_0}{24}\left(\frac{\eta\beta_N}{\hbar}\right)^2\left\{(H^1)^2{\sum_{\rho=-2}^{2}}'\delta_{m_a-m_b,\,\rho}f^2_{\operatorname{sign}\rho}(\omega)\right.$$
$$\left.+\,\delta_{a-b}\,{}_0[(H^1)^2(f_1^2(\omega)+f_{-1}^2(\omega))+Rf_1(\omega)f_{-1}(\omega)]\right\}.$$
$$\times\frac{\Delta\omega_{\mathrm{rad}}}{\Delta\omega_n^2}\,\frac{(\Delta\omega_{n\,\mathrm{abs}}^{2})^2}{4(\omega_\gamma-\tilde{\omega}+m_b\omega_1-m_a\omega_0-\omega)^2+(\Delta\omega^{(2)}_{\mathrm{abs}})^2}, \tag{20}$$

where

$$f_\varepsilon=\frac{g_0[(j_a+\varepsilon m_a)(j_a-\varepsilon m_a+1)]^{1/2}}{(\omega_0\varepsilon+\omega)}$$
$$\times\begin{pmatrix}j_b & 1 & j_a\\ m_b & m_a-m_b-\varepsilon & -m_a+\varepsilon\end{pmatrix}$$
$$+\frac{g_1[(j_b-\varepsilon m_b)(j_b+\varepsilon m_b+1)]^{1/2}}{(\tilde{\omega}-\omega_1[m_b+\varepsilon]+\omega_0m_a-\omega_\gamma)}$$
$$\times\begin{pmatrix}j_b & 1 & j_a\\ m_b+\varepsilon & m_a-m_b-\varepsilon & -m_a\end{pmatrix}\quad\varepsilon=\pm1; \tag{21}$$

$$R={}^2/_5[(H_0^1)^2-|H_1^1|^2+3(l_1^*l_{-1}H_1^1H_{-1}^1+l_1l_{-1}^*H_1^{1*}H_{-1}^1],$$

$$(H^1)^2=\sum_\mu|H_\mu^1|^2; \tag{22}$$

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3)In the derivation we used the results of [9], concerning the value of the absorption cross section when the widths of the emission and absorption lines do not coincide.

The prime at the summation sign in (20) denotes that the value $\rho = 0$ does not take part in the summation, $\Delta\omega_{rad}$ is the width of the incident radiation, $\Delta\omega_{abs}^{(2)}$ is the width of the absorption line of the two-quantum process, $\tilde{\omega}$ is the frequency of the transition between the ground and excited levels if the hyperfine field at the nucleus in $H_N = 0$; g_0 and g_1 are the g-factors of the ground and excited states, respectively,

$$\omega_0 = -g_0\beta_N H_N/\hbar, \quad \omega_1 = -g_1\beta_N H_N/\hbar, \tag{23}$$

$$\sigma_0 = 2\pi\lambda\!\!\!^{-}{}^2 \frac{2j_b+1}{2j_a+1}\frac{f'}{1+\alpha}, \tag{24}$$

where f′ is the Mossbauer factor for the absorber and α is the conversion coefficient.

5. ESTIMATE OF THE RESULTS AND DISCUSSION

By way of an example let us consider an absorber consisting of nuclei in ferromagnetic iron. We have $f_a = 1/2$, $j_b = 3/2$, $g_0 = 0.18$, $g_1 = -0.103$, $H_N = -3.3 \times 10^5$ G, and $E = \hbar\omega = 14.4$ keV. The radiative transitions in Fe^{57} are magnetic dipoles (accurate to 0.01% [10]), so that the single-quantum transitions are determined by the selection rules $m_a - m_b = 0, \pm 1$. At the same time, two-quantum processes allow also the forbidden transitions $-1/2 \rightarrow 3/2$ and $1/2 \rightarrow -3/2$.

To find the most convenient conditions for the observation of two-quantum transitions, we specify the frequency of the alternating magnetic field to be such that the lines of the two-quantum transitions are to right and to the left of the spectrum of the lines of the single-quantum transitions. Satisfying this condition, we put $\tilde{\omega} = 4\omega_0/3$. Then at the frequencies $\tilde{\omega} + 1.7\omega_0$ and $\tilde{\omega} - 1.7\omega_0$ there will occur additional lines corresponding to the forbidden transitions. The choice of this value of ω is justified, first, by the convenience of observation (of the main spectrum), and second, in that the line width of the Mossbauer single-quantum transition, and all the more the width of the nuclear magnetic resonance, is smaller than $\omega_0/3$ [4,11]. Calculation shows that at frequencies $\tilde{\omega} + 1.7\omega_0$ and $\omega - 1.7\omega_0$ the cross sections will be determined, accurate to within 1 , by the following expressions:

$$\sigma_{res}^{(2)+}(-1/2 \rightarrow 3/2) \text{ and } \sigma_{res}^{(2)-}(1/2 \rightarrow -3/2), \quad \text{with}$$

$$\sigma_{res}^{(2)+}(-1/2 \rightarrow 3/2) = \sigma_{res}^{2}(1/2 \rightarrow -3/2) = \sigma'',$$
$$\sigma'' \approx \frac{\sigma_0}{24}\left(\frac{\eta\beta_N H^1}{\hbar}\right)^2 1{,}8\left(\frac{g_0}{\omega_0}\right)^2 \frac{\Delta\omega_{rad}}{\Delta\omega_{abs}^2}. \tag{25}$$

Putting further $\eta = 10^3$, $H_1 = 100$ Oe, $\sigma_0 = 1.48 \times 10^5$ b [4], and setting $\Delta\omega_{abs}^2$ equal to $\Delta\omega_{rad}$, we get $\sigma'' = 1.2 \times 10^3$ b.

We shall now determine the effective thickness of the absorber, $t = n\sigma''$, where n is the number of nuclei participating in the resonant absorption per unit target area. We shall use for the calculation the maximal absorber thickness $x_0 = 1/\mu_l$, where the linear coefficient of nonresonant absorption is $\mu_l \simeq 4.9 \times 10^2$ cm^{-1} [12]. The effective t thickness is determined in our case by the following formula:

$$t = N_A \rho M^{-1} x_0 c \Lambda \sigma'', \tag{26}$$

where N_A is Avogadro's number, c the concentration of Fe^{57}, M the molecular weight of the iron, ρ the density of the iron, Λ the relative volume of the Bloch walls. Substituting in (26) $N_A = 6 \times 10^{23}$, $c = 0.75$ (enriched iron), $\Lambda = 1/7$, and $M \approx 57$, we get $t \simeq 2 \times 10^{-2}$.

For $t \ll 1$, using the formulas for the dependence of the absorption on the thickness [9], we get the deviation from the nonresonant absorption in the following form:

$$\varepsilon = 1/2 ft, \tag{27}$$

where f is the Mossbauer factor for the emitter. Taking $f = 0.9$ [12], we get $\epsilon \simeq 0.9 \times 10^{-2}$. On the other hand, if we take into account the fact that the width of the two-quantum transition lines are much narrower than the width of the single-quantum transition lines, then the deviation from the nonresonant absorption can be increased by a factor of several times. Thus, ϵ will fluctuate in the range 10^{-2}—10^{-1}. In this interval of ϵ, the resonant absorption is perfectly observable with the aid of modern technology and with the isotopes used in the Mossbauer effect.

In conclusion let us discuss the experimental possibilities of γ-magnetic resonance.

First, the γ-magnetic resonance can yield greater information on the spectrum of the Mossbauer nucleus, than the Mossbauer effect and NMR. The point is that the Mossbauer effect causes only transitions between ground and excited levels. On the other hand, in view of the small number of nuclei and the short lifetime of the nuclei in the excited states, the NMR is used only for the investigation of the spin sublevels of the ground state. Besides the transitions investigated by both methods, γ-magnetic resonance makes it also possible to observe transitions between the spin sublevels of the excited state. In addition, the process under consideration makes also forbidden processes allowed, as we have indicated.

Second, it becomes possible to investigate the physics of the ferromagnetic Bloch walls with the aid of γ-resonance spectroscopy methods. This is due to the fact that a contribution to the resonance is made only by nuclei located in the Bloch walls.

Finally, inasmuch as the γ-magnetic resonance is a two-quantum process, it makes it possible to study the nonlinear phenomena in the γ-quantum range. By way of an example we can indicate that many-quantum processes have led, in spite of the small magnitude of the effect, to the creation of a new discipline in physics, namely nonlinear optics.

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