ACOUSTIC MODULATION OF γ RADIATION IN FERROMAGNETIC MATERIALS

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A theory is presented of the acoustic modulation of γ radiation emitted by nuclei of a ferromagnetic material in which ultrasound with frequency Ω is excited in the crystal or a radio-frequency field is applied. It is shown that, in addition to excitation of satellites, resonance absorption occurs of ultrasound (acoustic resonance) or the radio-frequency field if Ω coincides with the spin precession frequency of the excited nuclei. The calculations are made on the assumption that the nuclear spins interact with lattice vibrations by means of magnetic electron spin waves.

INTRODUCTION

If ultrasonic vibrations of arbitrary frequency Ω are excited in a crystal which is a source or absorber of resonance γ radiation, then in the emission or absorption spectrum, in addition to the main line corresponding to the frequency k_0 of the nuclear transition, additional satellites appear whose frequencies are determined by the relation $k_0 \pm N\Omega$ ($N = 1, 2, \ldots$), and the intensities fall with increasing N. Early theoretical studies[1,2] discussed the effect of ultrasound on resonance γ transitions of nuclei in nonmagnetic crystals in the case when the source or absorber is characterized by the same transition line. The principal results of these studies have been confirmed experimentally by Ruby and Bolef,[3] and Cranshaw and Reivari,[4] who excited ultrasonic vibrations in a γ-ray source with the same emission line (Co^{57} in a nonmagnetic matrix). In more recent experiments[5,6] with a similar source, ultrasound was excited in a magnetically polarized absorber (Fe^{57}) by a radio-frequency field (by means of magnetostrictive coupling). The observed pattern of the absorption spectrum turned out to be more complex than predicted by theory[1,2] for the case of a nonmagnetic matrix, and still has not yet been satisfactorily explained even qualitatively.

However, these experiments established that the satellites, in all probability, have an acoustical nature and that the ultrasound cannot be considered only as a source of modulation of the nuclear motion, for in magnetic crystals its action is more complex. This leads to the necessity of searching for additional couplings of ultrasound with nuclear degrees of freedom.

In the present article we will not attempt to analyze all the possible means by which ultrasound might affect the resonance absorption and emission of γ rays. Moreover, this cannot yet be done on the basis of existing experimental data. We will consider only the situation in which, in addition to acoustic modulation of the motion of the center of mass of the nuclei of a magnetic crystal, there occurs also a qualitatively different phenomenon: resonance absorption of ultrasound by the nuclear spin system when the frequency Ω of the ultrasound is comparable with the frequency of the Zeeman splitting of the nuclei in a constant or hyperfine magnetic field H_0 (nuclear acoustic resonance). Nuclear acoustic resonance affects the characteristics of resonance absorption and emission of γ rays by nuclei. In fact, as is well known, transitions of nuclei between Zeeman sublevels induced by ultrasound lead to a change in the angular state of the nuclear spin system. Since the nature of resonance absorption and radiation of γ rays depends substantially on the angular state of the nuclei, the need naturally arises of taking into account the effect of Zeeman nuclear transitions induced by ultrasound on the intensity and spectrum of the γ radiation. In order to take into account the effect of ultrasound on the nuclear spin system, it is necessary to know the mechanism of interaction between them. In the theory of nuclear acoustic resonance[7] of magnetic atoms of ferromagnetic materials, several means of transfer of ultrasonic energy to the spin system have been recently discussed. So far there is no agreement on this question. We will discuss the Silverstein mechanism,[8] the sense of which is that acoustic energy is transferred to the electron spin system by means of magnetostriction, and from it through the hyperfine interaction to the nuclear spin system. The authors of the previously cited experimental studies[5,6] are also inclined toward this means of sonic energy transfer to the nuclear spin system, on the basis of a careful analysis of the results obtained by them.

In the present article, for the example of a ferromagnetic crystal whose nuclei possess hyperfine (Zeeman) structure in the excited and ground states, we will discuss the effect of ultrasound on the spectrum and intensity of resonance Mossbauer radiation. The principal attention will be devoted to the case in which the frequency of ultrasound becomes comparable with the frequency ω_e of nuclear spin precession in the excited state (without difficulty the theory can be generalized to the case of nuclear acoustic resonance in the ground state).

1. EQUATIONS OF MOTION OF NUCLEI UNDER CONDITIONS OF γ AND ACOUSTIC EXCITATIONS

Let us suppose that nuclei in a crystal which have spin $\mathbf{F} = \mathbf{I}_g$ in the ground state and $\mathbf{F} = \mathbf{I}_e$ in the excited states emit γ rays characterized by the same unit polarization vectors $\mathbf{e}^0_\eta$ and wave vectors $\mathbf{k}_i$. For simplicity we will assume that all the vectors $\mathbf{k}_i$ are parallel. If the distribution of $\mathbf{k}_i$ is continuous, we can introduce a frequency distribution function $u(k_i)$ (here and subsequently we will take $\hbar = c = 1$).

Interaction of the nuclear spin with a constant (external or static hyperfine) magnetic field H_0 leads to a Zeeman splitting of the energy levels of the ground and excited states of the nucleus with energies

$$E_\mu = \mu\omega_g I_g^z, \quad E_m = k_0 + m\omega_e I_e^z,$$

where $\omega_g = \gamma_g H_0$ is the Larmor frequency of the ground state, $\omega_e = \gamma_e H_0$ that of the excited state, $\gamma_{g,e}$ is the gyromagnetic ratio of the nucleus, μ is the magnetic quantum number characterizing the magnetic sublevels of the ground state, m is that of the excited state (in what follows the magnetic quantum numbers of the excited state will be designated also by other Latin letters: l, P, n, and the ground state by Greek letters ζ, ξ); k_0 is the frequency of the γ transition, and the energy of the nuclear ground state in the absence of H_0 is taken as zero.

If we neglect the spin-spin and quadrupole interactions of the nucleus with its surroundings, the Hamiltonian of the problem which interests us here can be represented in the form

$$\mathscr{H} = \mathscr{H}_0 + \mathscr{H}_z + V_{F\gamma} + V_{Fp}, \tag{1}$$

where $\mathscr{H}_0$ describes the energy of the free nucleus, the field of γ radiation, and the lattice vibration energy, $\mathscr{H}_Z$ is the Zeeman energy of the nucleus, $V_{F\gamma}$ is the Hamiltonian of the interaction of the nucleus with a γ ray characterized by a unit polarization vector $\mathbf{e}_\eta = -\frac{1}{2}\eta(\mathbf{e}_x + i\eta\mathbf{e}_y)$ ($\eta = 1$ corresponds to a right-handed polarization, and $\eta = -1$ to a left-handed polarization with respect to the wave vector $\mathbf{k}$):[9]

$$V_{F\gamma} = \left(\frac{2\pi}{\mathscr{L}k}\right)^{1/2} [V(\mathbf{k}\mathbf{e}_\eta)a_k + V^+(\mathbf{k}\mathbf{e}_\eta)a_k^+]. \tag{2}$$

Here

$$V(\mathbf{k}\mathbf{e}_\eta) = -\frac{e}{2m_0}\sum_n \{2g_l^n \mathbf{p}_n\mathbf{e}_\eta + g_s^n \mathbf{s}_n[\nabla\mathbf{e}_\eta]\} e^{i\mathbf{k}\mathbf{r}_n},$$

$\mathscr{L}$ is the volume occupied by the radiation field, a_k^+ and a_k are the photon creation and annihilation operators, g_l^n is the orbital g factor, g_s^n is the spin g factor, $\mathbf{r}_n$ is the radius vector of the n-th nucleon, and $\mathbf{p}_n$ and $\mathbf{s}_n$ are the momentum and spin of the nucleon.

V_{Fp} is the Hamiltonian of the indirect interaction of the nuclear spin with acoustic vibrations. According to Silverstein[8] for a circularly polarized sound wave characterized by frequency Ω and wave vector $\mathbf{q}$, this Hamiltonian has the form[1)] (in the semiclassical representation)

$$V_{Fp} = -\frac{1}{2}\gamma_F H_{Fp}[F_+e^{i(\mathbf{qr}-\Omega t)} + F_-e^{-i(\mathbf{qr}-\Omega t)}],$$
$$H_{Fp} = \frac{A_F}{\omega_\mathbf{q}}\langle S_z\rangle[e_{qz}q_x(\omega_M+\omega_A) + e_{qx}q_z(\omega_M-\omega_A)], \tag{3}$$

where $F_\pm = F_x \pm iF_y$, $\mathbf{r}$ is the radius vector of the center of gravity of the nucleus, H_{Fp} is the amplitude of the radio-frequency field induced by ultrasound in the nucleus, $\langle S_z\rangle$ is the z component of the total angular momentum of the magnetic electrons, A_F is the hyperfine interaction constant, $\gamma_F = \gamma_e$ for $F = I_e$ and $\gamma_F = \gamma_g$ for $F = I_g$, $\omega_M = \gamma_0 G/M_0$ and $\omega_A = 2\gamma_0 K_1/M_0$, γ_0 is the gyromagnetic ratio of the electron, M_0 is the saturation magnetization, G is the magnetostriction constant, K_1 is the anisotropy constant, $\omega_\mathbf{q}$ is the spin wave energy, and $\mathbf{e}_\mathbf{q}$ is the unit polarization vector of the sonic wave.

The ultrasound and γ radiation produce transitions between states of the unperturbed Hamiltonian $\mathscr{H}_0 + \mathscr{H}_Z$. The effect of these perturbations on the behavior of the aggregate system is found by means of the equation

$$i\frac{\partial}{\partial t}|\psi\rangle = \mathscr{H}|\psi\rangle. \tag{4}$$

The interaction of the nuclear spins with the field H_{Fp} can be taken into account accurately by transferring to a system of coordinates rotating with frequency Ω around the direction of H_0. This transfer is accomplished by means of the unitary transformation $|\psi\rangle^* = \exp\{i\Omega F_z t\}|\psi\rangle$, which gives

$$i\frac{\partial}{\partial t}|\psi\rangle^* = \{\mathscr{H}^* + V_{F\gamma}(t)\}|\psi\rangle^*, \tag{5}$$
$$\mathscr{H}^* = \mathscr{H}_0 + \mathscr{H}_z - \Omega F_z + \omega_1\{F_x\cos qr - F_y\sin qr\},$$
$$V_{F\gamma}(t) = e^{i\Omega F_z t}V_{F\gamma}e^{-i\Omega F_z t},$$

where $\omega_1 = -\gamma_F H_{Fp}$. Now the operator $\mathscr{H}^*$ describes the energy of the system with inclusion of the interaction of the spins with the ultrasound and $V_{F\gamma}(t)$ can be considered as a perturbation producing transitions between states of this operator. Then, going over in (5) to representation of the interaction, we obtain

$$i\frac{\partial}{\partial t}|\psi^\tau\rangle = V_{F\gamma}^\tau(t)|\psi^\tau\rangle, \tag{6}$$
$$|\psi^\tau\rangle = U|\psi\rangle, \quad U = e^{i\mathscr{H}^* t}e^{+i\Omega F_z t}, \quad V_{F\gamma}^\tau(t) = UV_{F\gamma}U^{-1}.$$

The further transformations associated with Eq. (6) are well known.[10] Assuming that the state vectors of the operator $\mathscr{H}_0 + \mathscr{H}_Z$ form an orthonormal basis, we represent $|\psi^T\rangle$ in the form of a series in these states:

$$|\psi^\tau\rangle = \sum_{\mu,\alpha_s^0} b_{\mu\alpha_s 0}|_g\mu;\alpha_s^0\rangle \tag{7}$$
$$+ \sum_{i,m,\beta_s} b_{m\beta_s}^{-\mathbf{k}_i}|I_e m; -\mathbf{k}_i\beta_s\rangle + \sum_{\eta,\mathbf{k},i,\mu\alpha_s} b_{\mu\alpha_s\eta}^{\mathbf{k}-\mathbf{k}_i}|I_g\mu; \mathbf{k}-\mathbf{k}_i, \mathbf{e}_\eta\alpha_s\rangle,$$

$|I_g\mu;\alpha_S^0\rangle$ is the state vector of the system at the initial moment of time, i.e., when the nucleus is in the μ-th sublevel of the ground state and the lattice in a state characterized by a quantum number α_S^0. In absorption of a γ ray the nucleus transfers to the m-th sublevel of the excited state and the lattice to some intermediate state characterized by a quantum number β_S. This state of the system is described by the vector $|I_e m; -k_i\beta_S\rangle$. Finally, $|K_g\mu; \mathbf{k}-\mathbf{k}_i, \mathbf{e}_\eta\alpha_S\rangle$ describes the state to which the system transfers after spontaneous radiation by the nucleus of a γ ray characterized by a wave vector $\mathbf{k}$ and polarization $\mathbf{e}_\eta$.

Substitution of (7) into (6) leads to a system of equations for the coefficients b of the expansion:

$$i\dot{b}_{\mu\alpha_s{}^0} = \sum_{im\beta_s}\langle I_g\mu;\alpha_s|V_{F\gamma}^T(t)|I_e m; -\mathbf{k}_i\beta_s\rangle b_{m\beta_s}^{-\mathbf{k}_i}, \tag{8a}$$
$$i\dot{b}_{m\beta_s}^{-\mathbf{k}_i} = \sum_{\mu\alpha_s^0}\langle I_e m; -\mathbf{k}_i\beta_s|V_{F\gamma}^T(t)|I_g\mu;\alpha_s^0\rangle b_{\mu\alpha_s^0}$$

1) The effect of the oppositely rotating component can always be neglected if the amplitude of the radio-frequency field $H_{Fp} < H_0$. In ferromagnetic materials the hyperfine field is $\sim 10^5$ oersteds, and the radio frequency field does not exceed a few oersteds.

$$+\sum_{\mu k\eta\alpha_s i}\langle I_e m;\,\beta_s|V_{F\gamma}^{T}(t)|I_g\mu;\,\mathbf{k}\eta\alpha_s\rangle\, b_{\mu\eta\alpha_s}^{\mathbf{k}-\mathbf{k}_i}, \tag{8b}$$

$$i\dot{b}_{\mu\eta\alpha_s}^{\mathbf{k}-\mathbf{k}_i}=\sum_{m\beta_s}\langle I_g\mu;\,\mathbf{k}\eta\alpha_s|V_{F\gamma}^{T}(t)|I_e m;\,\beta_s\rangle\, b_{m\beta_s}^{-\mathbf{k}_i}. \tag{8c}$$

The equations (8) are written for the case in which one nucleus is taking part in the radiative processes. If the effects associated with γ excitation of the nuclei are incoherent, then the equations for the probability amplitudes can be written for each nucleus individually. This means that averaging of (8) over the ensemble of nuclei does not change the form of these equations.

Considering $V_{F\gamma}$ as a small perturbation, we can solve the system (8) with the necessary degree of accuracy, and thereby find the values of the probability amplitudes b. However, our final goal will be calculation of the intensity of γ fluorescence resulting from action on the nucleus in an excited state of the ultrasound which induces the spin transitions. It is convenient to solve this problem in the density matrix representation, and this indicated by the following. The fluorescence intensity can be determined as the change in the populations of the nuclear sublevels of the ground state as the result of spontaneous decay. However, the diagonal elements of the density matrix immediately determine the populations of the states. On the other hand, to describe the magnetic resonance it is necessary to know not only the values of the coefficients b_μ but also the cross-product terms of the type $b_\mu b_{\mu'}^*$, and so forth (coherence), which are nondiagonal elements of the density matrix of the nuclear spin system. Therefore we will turn from the Eqs. (8) to the equations for the density matrix, having defined the latter as

$$\rho_{\mu\mu'\eta}^{\mathbf{k}\mathbf{k}_i}=\Big\langle\sum_{\alpha_s^0} b_{\mu\alpha_s^0}\, b_{\mu'\alpha_s^0}^{*}\Big\rangle+\Big\langle\sum_{\alpha_s} b_{\mu\eta\alpha_s}^{\mathbf{k}-\mathbf{k}_i}\, b_{\mu'\eta\alpha_s}^{\mathbf{k}-\mathbf{k}_i *}\Big\rangle \tag{9}$$

for nuclei in the ground state and the γ-radiation fields

$$\rho_{mm'}^{\mathbf{k}_i}=\Big\langle\sum_{\beta_s} b_{m\beta_s}^{-\mathbf{k}_i}\, b_{m'\beta_s}^{-\mathbf{k}_i *}\Big\rangle \tag{10}$$

for nuclei in an excited state. The angle brackets denote averaging over the ensemble of nuclei. The first term on the right-hand side of Eq. (9) represents the density matrix of nuclei in the ground state before absorption of a γ ray. The second term is the density matrix describing the nuclei after their return from the excited as the result of spontaneous decay.[2)] In terms of this second term the intensity of γ fluorescence characterized by a wave vector $\mathbf{k}$ and polarization $\mathbf{e}_\eta$ can be written in the form

$$S_F(\mathbf{k}\mathbf{e}_\eta)=\text{const}\sum_{\substack{i,\,\alpha_s\\ \mu=\mu'}}\frac{d}{dt}\langle b_{\mu\eta\alpha_s}^{\mathbf{k}-\mathbf{k}_i}\, b_{\mu'\eta\alpha_s}^{\mathbf{k}-\mathbf{k}_i *}\rangle=\text{const}\sum_{i,\,\alpha_s\mu=\mu'}\frac{d^{(\prime)}}{dt}\rho_{\mu\mu'\eta}^{\mathbf{k}\mathbf{k}_i}. \tag{11}$$

The prime on the time derivative indicates that Eq. (11) takes into account the change in population of the Zeeman sublevels of the ground state due only to spontaneous transitions.

Thus, the problem now reduces to finding the equations for $d^{(\prime)}\rho_{\mu\mu'\eta}^{\mathbf{k}\mathbf{k}_i}/dt$, which is not hard to do by means of (8). We multiply Eq. (8c) by $b_{\mu'\alpha_s\eta}^{\mathbf{k}-\mathbf{k}_i *}$ and add the expression obtained to its complex conjugate, in which it is necessary to perform the substitution $\mu \rightleftarrows \mu'$. Then we will average over the final intermediate states of the lattice and also sum over i. The equation obtained in this case will be given below (Eq. (19)). At this point we wish to note that the right-hand part of this equation depends, as should be expected, on the density matrix $\rho_{mm'}$ of the excited state, in which, incidentally, spin transitions can be induced if the ultrasonic frequency is appropriate. We will discuss at first the derivation of the expression for $\rho_{mm'}$.

2. DENSITY MATRIX FOR NUCLEI IN AN EXCITED STATE

In derivation of the equation for $\rho_{mm'}$ we will take into account that the binding forces in the crystal are very weak in comparison with the intranuclear forces. Therefore we will assume that they affect only the motion of the center of gravity of the nucleus and do not affect its internal degrees of freedom. This means that the state vector of the entire system can be represented in the form of the product of the vectors describing the lattice state and the internal degrees of freedom of the nuclei. Taking into account this fact and also Eqs. (8b) and (8c), we obtain in second order perturbation theory for $V_{F\gamma}$ the following equation:[3)]

$$\begin{aligned}\dot{\rho}_{mm'}=&-\Gamma\rho_{mm'}+\sum_{i\mu\mu'nn'PP'}\int_0^\infty d\tau e^{-\Gamma\tau/2}G_{n'\mu'}^{n\mu}\exp\{-it[\Omega(n-n')-\omega_g(\mu-\mu')\\&+\omega_f(P-P')]\}\cdot[\exp\{-i\tau(k_0-k_i-n\Omega+\mu\omega_g-P\omega_f)\}\langle\langle e^{-i\mathbf{k}_i\mathbf{u}(\tau)}e^{i\mathbf{k}_i\mathbf{u}(0)}\rangle\rangle\\&+\langle\langle e^{-i\mathbf{k}_i\mathbf{u}(0)}e^{i\mathbf{k}_i\mathbf{u}(\tau)}\rangle\rangle\cdot\exp\{i\tau(k_0-k_i-n'\Omega+\mu'\omega_g-P'\omega_f)\}]\\&\times\langle m|P\rangle\langle P|n\rangle\langle n'|P'\rangle\langle P'|m'\rangle\rho_{\mu\mu'}(t-\tau),\end{aligned} \tag{12}$$

$$G_{n'\mu'}^{n\mu}=\langle I_e n|V_{F\gamma}|I_g\mu\rangle\langle I_e n'|V_{F\gamma}|I_g\mu'\rangle^{*},$$

where $\langle P|m\rangle = D_{Pm}^{I_e}(qr,\theta,0)$ is the Wigner function,

$$\theta=\operatorname{arctg}\frac{\omega_1}{\omega_e-\Omega},\qquad \omega_f=[\omega_1^2+(\omega_e-\Omega)^2]^{1/2},$$

$\mathbf{u}(\tau)$ is the displacement of the nucleus from its equilibrium state in the interaction representation, Γ is the decay constant of the excited states, and the brackets $\langle\langle\ldots\rangle\rangle$ indicate averaging over lattice states. The second term of the right-hand side of Eq. (12) involves the phonon correlators, which can be transformed to the form[1]

$$\langle\langle e^{-i\mathbf{k}_i\mathbf{u}(\tau)}e^{i\mathbf{k}_i\mathbf{u}(0)}\rangle\rangle=e^{-2W-X_i}\exp\Big\{\frac{X_i}{2}[h(\Omega)e^{i\Omega\tau}+(h(\Omega)+1)e^{-i\Omega\tau}]\Big\}, \tag{13}$$

where $\exp\{-2W\}$ is the Debye-Waller factor, and

$$X_i=(R/N_0\Omega)(\mathbf{e}_q\mathbf{k}_i^0)^2[h(\Omega)+1/2] \tag{14}$$

is an additional term due to the ultrasound, R is the energy of the recoil nucleus, N_0 is the number of atoms in the sample, $\mathbf{k}_i^0$ is a unit vector in the direction of propagation of the γ excitation, and $h(\Omega) \gg 1/2$ is the number of quanta of ultrasound at frequency Ω.

Using the expansion

$$\exp(X_i\cos\alpha)=\sum_{N=-\infty}^{\infty}I_N(X_i)\exp(iN\alpha), \tag{15}$$

[2)]The matrix elements $\rho_{m\mu}$ are a measure of the coherence created by the γ radiation and can be different from zero only for the condition of nuclear excitation by coherent γ radiation. [11]

[3)]It is assumed that the thermal relaxation time is much greater than the lifetime of the excited state.

where $I_N(X_i) = (-i)^N J_N(iX_i)$ is a Bessel function with imaginary argument, and converting from the summation over i to an integral, we obtain

$$\dot{\rho}_{mm'} = -\Gamma\rho_{mm'} + \sum_{\zeta'\zeta PP'll'N'} W_{l\zeta'P'}^{l\zeta P}(X_i N') \exp\{-it[\Omega(l-l') - \omega_g(\zeta-\zeta') + \omega_f(P-P')]\}\langle m|P\rangle\langle P|l\rangle\langle l'|P'\rangle\langle P'|m'\rangle\rho_{\zeta\zeta'}(t). \quad (16)$$

Here

$$W_{l'\zeta'P'}^{l\zeta P}(X_i N') = \int u(k)dke^{-2W-x^i} I_{N'}(X_i) G_{l'\zeta'}^{l\zeta}\left[\left\{\frac{\Gamma}{2} - i(k - k_0 + l\Omega - \zeta\omega_g + P\omega_f + N'\Omega)\right\}^{-1} + \{\Gamma/2 + i(k - k_0 + l'\Omega - \zeta'\omega_g + P'\omega_f + N'\Omega\}^{-1}\right] \quad (17)$$

determines the probability of excitation of nuclei by γ rays hitting the crystal. Since the density matrix $\rho_{\zeta\zeta'}$ of the ground state remains essentially unchanged during the life of the nucleus, Eq. (16) can easily be integrated:

$$\rho_{mm'}(t) = \sum_{ll'\zeta\zeta'PP'N'} W_{l'\zeta'P'}^{l\zeta P} \frac{\exp\{-it[\Omega(l-l') - \omega_g(\zeta-\zeta') + \omega_f(P-P')]\}}{\Gamma - i[\Omega(l-l') - \omega_g(\zeta-\zeta') + \omega_f(P-P')]} \times \langle m|P\rangle\langle P|l\rangle\langle l'|P'\rangle\langle P'|m'\rangle\rho_{\zeta\zeta'}. \quad (18)$$

This is a very general expression describing the stationary motion of nuclei in an excited state. Its structure reflects the double role of ultrasonic excitation in the crystal. In the first place, the ultrasound produces induced cyclic oscillations of the centers of gravity of the nuclei with frequency Ω, which eventually leads to appearance of additional lines (satellites) in the γ-ray absorption spectrum (W). In the second place, the radio-frequency field stimulated by the ultrasound induces radio-frequency transitions between the magnetic sublevels of the nuclear excited state, if its frequency Ω is of the order of the Zeeman splitting frequency ω_e. This leads to a change in the populations of the sublevels of the excited state and consequently also a change in the intensities of the individual γ-transition lines. If the ultrasonic frequency Ω differs significantly from ω_e ($\omega_e - \Omega \gg \Gamma$), there will be no sound absorption, $\langle m|P\rangle = \delta_{mP}$, and the matrix $\rho_{mm'}$ will depend only on W.

3. γ FLUORESCENCE UNDER ULTRASONIC EXCITATION

It was noted above that to calculate the γ-ray intensity it is necessary to find the change in the populations of the Zeeman sublevels of the nuclear ground state due to spontaneous nuclear transitions or, in other words, to find the expression for $d^{(\prime)}\rho^{\mathbf{k}}_{\mu\mu'\eta}/dt$. Omitting the calculations, which are identical to those carried out to find $\rho_{mm'}$, we give the final equation

$$\dot{\rho}^{\mathbf{k}}_{\mu\mu'\eta} = \sum_{N=-\infty}^{\infty} e^{-2W-X_e} I_N(X_e) \sum_{nn'KK'mm'} G_{n\mu}^{n'\mu'}(\mathbf{k}e_\eta) \exp\{it[\Omega(n-n') - \omega_g(\mu-\mu') + \omega_f(K-K')]\}\langle m'|K'\rangle\langle K'|n'\rangle\langle n|K\rangle\langle K|m\rangle\rho_{mm'} \cdot \{\delta(k-\tilde{k}_0) + \delta(k-\tilde{k}_0')\}, \quad (19)$$

where

$$X_e = \frac{R}{N_0\Omega}\left(e_q\frac{\mathbf{k}}{|\mathbf{k}|}\right)^2\left[h(\Omega) + \frac{1}{2}\right],$$

$\tilde{k}_0 = k_0 - n\Omega + \mu\omega_g - K\omega_f - N\Omega$, $\tilde{k}_0' = k_0 - n'\Omega + \mu'\omega_g - K'\omega_f - N\Omega$. It is easy to show that the right-hand side of Eq. (19) is proportional to the total probability for transition of the nucleus from the excited state to the ground state per unit time. It is different from zero when the frequency of the emitted γ ray is $\tilde{k}_0$ or $\tilde{k}_0'$, i.e., for the condition that the energy conservation law is satisfied.

Substitution of (19) into (11) now enables us to obtain a more general expression for the intensity of γ fluorescence in a given direction. We will give it here for the particular case of nuclear excitation by circularly polarized γ rays, for example, right-polarized. If we take into account that in this case it is necessary to set $l = l'$ in Eq. (18), we obtain

$$S_F\left(\frac{\mathbf{k}e_\eta}{|\mathbf{k}|}\right) = \text{const}\sum_{nn'\mu PP'l\zeta|k|} G_{n\mu}^{n'\mu}(\mathbf{k}e_\eta) \sum_{N,N'=-\infty}^{\infty} e^{-2W-X_e} I_N(X_e) W_{l\zeta P'}^{l\zeta P}(N'X_i) \times \frac{\langle n|\rangle\langle P|l\rangle\langle l|P'\rangle\langle P'|n'\rangle}{\Gamma - i(P-P')[\omega_1^2 + (\omega_e-\Omega)^2]^{1/2}} e^{i\Omega(n-n')t}\sigma_{\zeta\zeta}\{\delta(k-\tilde{k}_0) + \delta(k-\tilde{k}_0')\}, \quad (20)$$

where $\sigma_{\zeta\zeta'}$ is the density matrix of the ground state in the laboratory system of coordinates, which is related to $\rho_{\zeta\zeta'}$ by the equation $\sigma_{\zeta\zeta'} = \exp\{i\omega_g(\zeta-\zeta')t\}\rho_{\zeta\zeta'}$. In obtaining Eq. (20) we considered the fact that the nondiagonal elements of $\sigma_{\zeta\zeta'}$ are absent since the only source introducing coherence to the nuclear system—ultrasound—acts only on the excited state. This is equivalent to saying that the components transverse to the magnetic field of the magnetization of the nuclear spin system of the ground state are zero in thermal equilibrium.

For nuclei in an excited state this is no longer true. Interaction of these nuclei with ultrasound leads to establishment of definite phase relations of the excited states within the ensemble of nuclei, which is well known to be a necessary condition for experimentally observable interference of nuclear states. In the present case the interference appears as beats of the γ-fluorescence intensity with a frequency $\Omega(n-n')$. Observation of these beats requires special apparatus. The usual radiation detectors used in Mossbauer experiments record the average intensity value which is represented in Eq. (20) by terms with $n = n'$.

4. DISCUSSION OF RESULTS

Ultrasonic vibrations excited in a magnetic crystal lead to a rather complex expression for the γ-fluorescence intensity. First of all, the spontaneous radiation of the nuclei is represented not only by the principal transition lines but also by additional lines (the sum over N in (20)) which are separated from each other in frequency by an amount Ω. Recently these lines have been observed in Fe^{57} nuclei by Heiman et al.[5] (unfortunately, this is only cited in the article). The intensity of each line depends not only on the expression $I_N(X_e)\exp -X_e$ determined by the ultrasonic power, but also on the expression characteristic of nuclear magnetic resonance, which depends on the frequency and amplitude of the radio-frequency field induced by the ultrasound in the nucleus:

$$\frac{\langle n|P\rangle\langle P|l\rangle\langle l|P'\rangle\langle P'|n\rangle}{\Gamma - i(P-P')[\omega_1^2 + (\omega_e-\Omega)^2]^{1/2}}$$

For this reason the intensity of the main and satellite lines has a resonance dependence on the frequency Ω:

it is maximal for $\Omega = \omega_e$ and minimal when $\omega_e - \Omega \gg \Gamma$. It is easy to see that the dependence of the intensity on the ultrasonic frequency has a Lorentz shape with a width Γ. This dependence can be used successfully for detection and study of nuclear magnetic resonance and nuclear magneto-acoustic resonance in magnetic crystals. Here one interesting feature calls itself to our attention: spin transitions of nuclei, detected by means of γ radiation, can be observed also in the case when the Zeeman splitting frequency is much smaller than the width Γ of the excited state.[12]

As is well known, with Mossbauer's discovery an extraordinarily sensitive method has appeared in solid-state γ spectroscopy for studying the energy spectra of nuclei. However, even here, as in every spectroscopic method, the sensitivity is limited by the natural width of the excited state. Therefore, for $\omega_e < \Gamma$ it becomes completely unsuitable for the purposes mentioned. Combination of a radio-frequency technique with the Mossbauer method permits the limits of applicability of the latter to be considerably extended, in particular, to the region of very weak magnetic fields acting on nuclei.

If the ultrasonic frequency is far from the resonance frequency ω_e, there will be no sound absorption and Eq. (20), as should be expected, will go over to the well known expression given by Abragam.[1] In the absence of ultrasonic excitation $\langle n|P\rangle = \delta_{nP}$, $I_0(X_e) = 1$, $I_N(X_e) = 0$, for $N \neq 0$ the satellites disappear, and in spontaneous radiation γ rays are present whose frequencies are determined by the relation $\tilde{k}_0 = k_0 - n\omega_e + \mu\omega_g$, where n takes on the values of the magnetic quantum numbers of the excited state, and μ—those of the ground state. In other words, ordinary Mossbauer radiation occurs of γ rays with energy and polarization allowed by the selection rules.

The nature of the spontaneous radiation of γ rays is substantially affected by the conditions of nuclear excitation. In fact, S_F depends on W—the probability for absorption of γ rays whose distribution in frequency is characterized by the function $u(k_i)$. If the width Δ of the excitation spectrum is much greater than the distance between the excited sublevels, then, as follows from Eq. (17), the nuclei will be excited on absorption of γ rays with energies corresponding to the energies of the main and satellite transitions. If Δ is much less than ω_g and $\omega_e \sim \Omega$, then

$$W \sim e^{-2W - X_i} I_{N'}(X_i)\{{}^1/_2\Gamma - i(k_i - k_0 + l\Omega - \zeta\omega_g + P\omega_f + N'\Omega)\}^{-1} \quad (21)$$

represents the probability of selective excitation of a nucleus to a definite sublevel whose energy is found from the condition

$$k_i = k_0 - l\Omega + \zeta\omega_g - P\omega_f - N'\Omega. \quad (22)$$

For definite values of l, ζ, and P, the latter expression determines the location of the satellites in the γ-ray absorption spectrum, which depends on the amplitude of the radio-frequency field at the nucleus through the quantity $\omega_f = [\omega_1^2 + (\omega_e - \Omega)^2]^{1/2}$. If $\omega_e = \Omega$ and $\omega_1 < \Gamma$, then the effect of the radio-frequency field H_{Fp} is unimportant. However, if $\omega_1 \sim \Gamma$, the effect of the field must already be taken into account, for it appears in the shift of the lines. This tendency was noted by Heiman et al.[5] without any quantitative evaluations. In the absence of ultrasonic absorption, Eq. (22) goes over to the form

$$k_i = k_0 - l\omega_e + \zeta\omega_g - N'\Omega, \qquad N' = 0, \pm 1 \ldots \quad (23)$$

For $N' = 0$ this condition determines the location of the main lines, and for $N' = \pm 1, \pm 2 \ldots$ that of the satellite lines. The pattern of the spectrum appears as follows. The most intense lines, which correspond to central transitions with definite values of l and ζ, have on their left and right satellite lines at a distance $N'\Omega$. The intensity of the latter is determined by the expression $I_{N'}(X_i) \exp -X_i$ and falls off with increasing N'. This is well substantiated by the recent experimental work of Asti et al.

We have discussed the effect of sonic vibrations on absorption and radiation of γ rays by the nuclei of a magnetic crystal, without specifying the means of ultrasonic excitation. We will note only that if the sonic vibrations are induced by a radio-frequency field applied to the sample (due to the magnetostrictive interaction), then for samples whose thickness is of the order of the skin depth, the action on the nuclei of the radio-frequency field induced by the ultrasound can be neglected, and the problem reduces to solution of Eq. (4), in which V_{Fp} must be replaced by the Hamiltonian of the indirect interaction of the nuclei with the variable radio-frequency field. All of the remaining calculations will be the same as those carried out in this article.

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