РАДІАЦІЙНА ФІЗИКА

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MÖSSBAUER FORWARD SCATTERING SPECTRA OF FERROMAGNETS IN RADIO-FREQUENCY MAGNETIC FIELD

The transmission of Mössbauer radiation through a thick ferromagnetic crystal, subjected to the radio-frequency (rf) magnetic field, is studied. A quantum-mechanical dynamical scattering theory is developed, taking into account both the periodical reversals of the magnetic field at the nuclei and their coherent vibrations. The Mössbauer forward scattering (FS) spectra of the weak ferromagnet $FeBO_3$ exposed to the rf field are measured. It is discovered that the coherent gamma wave in the crystal, interacting with Mössbauer nuclei, absorbs or emits only couples of the rf photons. As a result, the FS spectra consist of equidistant lines spaced by twice the frequency of the rf field in contrast to the absorption spectra. Our experimental data and calculations well agree if we assume that the hyperfine field at the nuclei in $FeBO_3$ periodically reverses and there are no coherent vibrations.

Keywords: Mössbauer spectroscopy, forward scattering spectra, iron borate, radio-frequency magnetic field.

Introduction

Ferromagnetic nanoparticles and thin films are widely used as the memory elements in electronics. Therefore intensive investigations are carried out of their magnetization dynamics [1]. The Mössbauer spectroscopy gives unique possibility to get information about such a dynamics in local vicinity of the Mössbauer isotope ^{57}Fe.

Starting from the pioneer work of Pfeiffer [2], it has been shown in numerous experimental papers (see reviews [3 - 6]), that the Mössbauer absorption spectra of soft ferromagnets, exposed to the rf field of circular frequency Ω, consist of equidistant lines spaced by Ω. When at the nucleus apart from the magnetic field $\mathbf{h}(t)$ there exists yet the electric field gradient, such lines split into doublets. At low frequencies the absorption spectrum transforms to standard Zeeman sextet. In the case of high frequencies the spectrum collapses to single line (doublet).

The external rf field $\mathbf{H}(t) = \mathbf{H}_0 \cos\Omega t$ generates in soft ferromagnet both the magnetostrictive vibrations and the periodic reversals of its magnetization $\mathbf{M}(t)$. The magnetic field at the nucleus $\mathbf{h}(t)$, which is antiparallel to $\mathbf{M}(t)$, follows these reversals jumping between the values $+\mathbf{h}_0$ and $-\mathbf{h}_0$ with the period $T = 2\pi/\Omega$. For such ferromagnets the magnetization completely repeats oscillations of $\mathbf{H}(t)$ and no static magnetization $\mathbf{M}_{st}$ is present. In this situation the magnetostrictive vibrations have the frequency 2Ω [3, 7].

The rf collapse at high frequencies is certainly due to fast reversals of the field $\mathbf{h}(t)$, when the nucleus only "sees" the averaged field $\langle \mathbf{h}(t) \rangle = 0$. The appearance of sidebands separated by the interval Ω is caused by magnetostrictive vibrations and magnetization reversals. The last mechanism has been studied in theoretical papers [7 - 14]. It was shown that the stepwise reversals of the field provide satellites, whose intensity quickly falls down with growing their order. At the same time, satellites owing to ultrasound vibrations are characterized by more slow attenuation (see, e.g., [15, 16]. Good fitting of Pfeiffer's rf absorption spectra was achieved by taking into account both the reversals and magnetostrictive vibrations [11].

If a constant magnetic field is superimposed along the rf external field, the reversals become asymmetric in time and the nucleus already "sees" along with the alternating field nonzero averaged field $\langle \mathbf{h}(t) \rangle$, which ensures quasi-Zeeman splitting of the nuclear quasi-levels. Respectively, every absorption line splits into quasi-Zeeman sextet [11, 13].

The role of stochastic jumps of the magnetization vector, driven by the applied rf field, has been studied in [17, 18].

Interesting transient effects arise in the case, when the period T of the rf oscillations much exceeds the nuclear lifetime. Then the field reversal causes oscillations of the time-dependent absorption cross section of γ-quanta [10] and strong flash of the radiation, transmitted through a ferromagnet [19, 20].

Nevertheless, a shortcoming of these calculations is that they were performed in the kinematical

approximation, i.e., they neglected rescattering of photons in the target. This is valid for extremely thin absorbers with $\sigma_0 n_0 = 1$, where n_0 is the number of Mössbauer isotopes per unit square and σ_0 is the resonant absorbtion cross section of γ-quanta. In most experiments this constraint is violated. Therefore the dynamical scattering theory has been developed of Mössbauer rays by the ferromagnet in the regime of periodical reversals of the magnetic field [21]. Another dynamical approach and preliminary straight-forward scattering rf spectra of γ-photons by the iron borate crystal $FeBO_3$ were presented in [22].

In this paper we first build detailed theory for the Raman scattering to forward direction of Mössbauer radiation by a soft ferromagnetic crystal, when simultaneously there exist periodical reversals of the magnetic field $\mathbf{h}(t)$ and coherent vibrations. This is quantum-mechanical theory, based on exact equations of multiple scattering [23], which does not involve any classical Maxwellian equations. Further we discuss our experimental results obtained on the iron borate in the rf field.

Mention also the papers [24 - 26], where the selective excitation double-resonance (SEDM) spectra of superparamagnetic particles have been observed. It was shown that stochastic reversals of their magnetization lead to appearance of satellites in such scattering spectra, which provide pure information about the frequency of magnetization reversals, not masked by the Brownian motion of the magnetization vectors and by the interparticle interaction. The corresponding scattering theory was built in [29].

All this allows us to think that forward scattering (FS) experiments will lead to better understanding of the processes in ferromagnets exposed to rf fields. Specifically, they reveal the transparency alteration [24] as well as the interference of the scattering amplitudes [25].

It is worth to note that Mössbauer FS experiments were already conducted with vibrating stainless steel [15, 16].

Scattering amplitudes

We direct the axis z perpendicularly to the crystal slab along the incident beam of γ-quanta with frequency $\omega = E/\hbar$ and wave vector $\mathbf{k} = \{0,0,k\}$. The crystal occupies the space $0 \le z \le D$, where D is the crystal thickness. The direction of the axis x is chosen along the magnetic field amplitude $\mathbf{h}_0$, which is believed to be parallel to the crystal surface.

Let the magnetic field at the Mössbauer nucleus $\mathbf{h}(t)$ periodically changes its direction to the opposite one, i.e.

$$\mathbf{h}(t) = \mathbf{h}_0 f(t), \tag{1}$$

where T is the period of the rf field and $\Omega = 2\pi/T$ is the circular frequency and the factor $f(t) = f(t+T)$ is determined by

$$f(t) = \begin{cases} -1, & 0 < t < T/2 \\ +1, & T/2 < t < T, \end{cases} \tag{2}$$

Moreover, there are coherent magnetostrictive vibrations, which ensure periodic displacements of the nuclei from the equilibrium position

$$\mathbf{R}(t) = \mathbf{A}\cos(2\Omega t + \phi_0), \tag{3}$$

where ϕ_0 is an initial phase of vibrations. We assume that these vibrations have the same amplitude A within the cross section of the incident beam of γ-quanta.

The nuclei in the periodic magnetic field (1) are described by the Floquet wave functions

$$\Psi_{M_\kappa}(t) = \psi_{M_\kappa}(t) e^{-iW_\kappa t/\hbar}, \tag{4}$$

where the periodic functions $\psi_{M_\kappa}(t) = \psi_{M_\kappa}(t+T)$ are given by

$$\psi_{M_\kappa}(t) = |I_\kappa M_\kappa\rangle \exp\left[-i\gamma_\kappa M_\kappa h_0 \int_0^t f(t')dt'/\hbar\right], \tag{5}$$

the index κ labels the ground g or excited e nuclear states, γ_κ represents the gyromagnetic ratio in the κ th state of the nucleus, $|\kappa\rangle = |I_\kappa M_\kappa\rangle$ are the stationary wave functions of the nucleus with spin I_κ and its projection M_κ on the quantization axis x, while the corresponding nuclear energies are $W_g = 0$ and $W_e = E_0'$.

The nuclei ^{57}Fe, absorbing incident photons, perform transition from the ground state $|g\rangle$ to the excited one $|e\rangle$. If k is perpendicular to h_0, γ-quanta with linear polarization $\mathbf{e}_x$ generate transitions $M_g \to M_e = M_g \pm 1$ and those with polarization $\mathbf{e}_y$ induce transitions with $M_e = M_g$. Thus, the unit vectors $\mathbf{e}_x$ and $\mathbf{e}_y$ are eigenpolarizations of photons, which are not mixed during scattering by nuclei.

Since the nuclei are exposed to an alternating magnetic field with circular frequency Ω, the

γ-quanta undergo the Raman scattering, leading to appearance of γ-quanta with shifted frequencies $\omega_n = \omega - n\Omega$, where n is an integer. In the case of straight-forward scattering far from the Bragg condition their wave vectors are $\mathbf{k}_n = \{0,0,k_n\}$ with $k_n = \omega_n / c$.

In order to find the scattering amplitude of γ-quanta by the nucleus we should calculate the matrix elements $M_{eg}(t)$ of the operator $c^{-1}\hat{j}_\alpha^N(\mathbf{k})e^{ikR(t)}$ on the Floquet wave functions (4). Here

$$\hat{j}_\alpha^N(\mathbf{k}) = e_\alpha \int d\mathbf{r} e^{i\mathbf{kr}} \mathbf{j}_N(\mathbf{r}) \tag{6}$$

represents the Fourier transform of the current density operator of the nucleus $\mathbf{j}_N(\mathbf{r})$, multiplied by the polarization vector $\mathbf{e}_\alpha$. Employing well-known expansion

$$e^{ix\cos\Omega t} = \sum_{l=-\infty}^{\infty} i^l J_l(x) e^{il\Omega t}, \tag{7}$$

where $J_l(x)$ is the Bessel function of the order l, we get the coefficients $a_{eg}(n)$ of the Fourier series for the periodic function $M_{eg}(t)$ (see also [11]):

$$a_{eg}(n) = \sum_{l=-\infty}^{\infty} i^l e^{il\phi_0} J_l(\mathbf{k}\cdot\mathbf{A}) b_{eg}(n-2l), \tag{8}$$

where

$$b_{eg}(n) = \exp\left[-i\left(\frac{x_{eg}+n\pi}{2}\right)\right]\sin\left(\frac{x_{eg}+n\pi}{2}\right)\frac{2x_{eg}}{x_{eg}^2-(n\pi)^2} \tag{9}$$

with parameters

$$x_{eg} = \frac{\alpha_{eg}T}{2}, \quad \alpha_{eg} = (\gamma_g M_g - \gamma_e M_e)h_0/\hbar. \tag{10}$$

General expression for the coherent Raman scattering amplitude of γ-quanta by j th nucleus in the rf field has been derived in [10]. For particular case of forward scattering by an unpolarized target it can be written as

$$f_{coh}^{(n'-n)}(\mathbf{k}_n,\mathbf{e}_\alpha;\mathbf{k}_{n'},\mathbf{e}_\alpha)_j^N = \frac{1}{2}\sum_{M_g=\pm 1/2} f_{gg}^{(n'-n)}(\mathbf{k}_n,\mathbf{e}_\alpha;\mathbf{k}_{n'},\mathbf{e}_\alpha)_j^N, \tag{11}$$

where $f_{gg'}^{(n'-n)}$ is the Raman scattering amplitude of γ-quanta by j th nucleus, which passes from the initial state $|g\rangle$ to the final $|g'\rangle$:

$$f_{gg'}^{(n'-n)}(\mathbf{k}_n,\mathbf{e}_\alpha;\mathbf{k}_{n'},\mathbf{e}_\alpha)_j^N = -p_j e^{-2W_a^j}\sum_{M_e}\sum_{m=-\infty}^{\infty}\frac{c^{-2}\langle e|\hat{j}_\alpha^N(\mathbf{k})|g'\rangle^*\langle e|\hat{j}_\alpha^N(\mathbf{k})|g\rangle^2 a_{eg}^*(m-n')a_{eg}(m-n)}{E-E_0'-m\hbar\Omega+i\Gamma/2} \tag{12}$$

and p_j is the fraction of the Mössbauer isotope in the j th site, e^{-2W_a} is the Debye - Waller factor for the absorber, $\Gamma = \Gamma_\gamma + \Gamma_e$ is the width of the excited level of the scattering nucleus, which consists of the partial radiative width Γ_γ and the conversion electron one Γ_e. The absorption of γ-quanta in crystals is mainly determined by Γ_e, being much larger than Γ_γ

The amplitude $f^{(n)}$ describes scattering of γ-quantum, when it emits $(n>0)$ or absorbs $(n<0)$ n modulation quanta $\hbar\Omega$. Once $\mathbf{k}$ and $\mathbf{h}_0$ are perpendicular, the reflection $M_e, M_g \to -M_e, -M_g$ does not change the factor $|\langle e|\hat{j}_\alpha^N(\mathbf{k})|g\rangle|^2$ and the product $a_{eg}^*(n'-m)a_{eg}(n-m)$ attributes the sign $(-1)^{n'-n}$, so that

$$f_{gg}^{(n)} = (-1)^n f_{-g,-g}^{(n)}. \tag{13}$$

Hence the coherent scattering amplitude $f_{coh}^{(n)} = 0$ for odd n due to interference of the terms $f_{1/2,1/2}^{(n)}$ and $f_{-1/2,-1/2}^{(n)}$.

The Raman coherent scattering amplitude of γ-rays by an elementary cell is a sum of the nuclear and Rayleigh straight-forward scattering amplitudes. The first is

$$F^{(n'-n)}(\mathbf{k}_n,\mathbf{e}_\alpha;\mathbf{k}_{n'},\mathbf{e}_\alpha)^N = \sum_j f_{coh}^{(n'-n)}(\mathbf{k}_n,\mathbf{e}_\alpha;\mathbf{k}_{n'},\mathbf{e}_\alpha)_j^N, \tag{14}$$

where summation is carried out over all sites j within the elementary cell, containing Mössbauer isotope. The Rayleigh scattering amplitude by electrons

$$F^{(n'-n)}(\mathbf{k}_n,\mathbf{e}_\alpha;\mathbf{k}_{n'},\mathbf{e}_\alpha)^R = F^R(\mathbf{k}_n,\mathbf{e}_\alpha;\mathbf{k}_n,\mathbf{e}_\alpha)\delta_{n'n}, \tag{15}$$

where F^R stands for standard Rayleigh scattering amplitude to zero angle in the absence of external periodic fields (see, e.g., [33]):

$$F^R = -r_0\sum_j Z_j + i(k/4\pi)\sigma_{pe}, \tag{16}$$

Z_j is the number of electrons of jth atom, $r_0 = e^2/mc^2$ denotes the classical radius of the electron, σ_{pe} is the photoelectric cross section.

Transmission of radiation through a crystal

Let the crystal be composed by N infinite layers of elementary cells parallel to the crystal surface. Their thickness equals d, so that the crystal thickness is $D = Nd$. Such layers are numerated by the index $m = 0, 1, 2, ..., N-1$, where $m = 0$ specifies the face layer with the coordinate $z \approx 0$ and $m = N-1$ the back layer with $z \approx D$.

The incident photon may be described by the vector potential $\mathbf{A}_\alpha^{(0)}(z,t) = \mathbf{A}_\alpha^{(0)} e^{ikz - i\omega t}$, where the vector amplitude is $\mathbf{A}_\alpha^{(0)} = (2\pi\hbar c/k)^{1/2} e_\alpha$ [32]. Introducing the retarded time $t^* = t - z/c$, we rewrite it as $\mathbf{A}_\alpha^{(0)}(t^*) = \mathbf{A}_\alpha^{(0)} e^{-i\omega t^*}$. Let us denote the off-Bragg transmitting coherent wave by $\mathbf{A}_\alpha(t^*) = \mathbf{A}_\alpha^{(0)} \psi_\alpha(t^*)$. In addition, the waves incident on the m th layer and scattered by it are denoted respectively by $\mathbf{A}_\alpha^{(m)}(t^*) = \mathbf{A}_\alpha^{(0)} \psi_\alpha^{(m)}(t^*)$ and $\mathbf{A}_\alpha^{(m)}(t^*)_{sc} = \mathbf{A}_\alpha^{(0)} \psi_\alpha^{(m)}(t^*)_{sc}$.

Scattering of photons by a crystal is determined by the system of multiple scattering equations [23]. For the elastic diffraction of Mössbauer photons they were represented in [33, 34]. And for the case considered these equations may be rewritten in the form

$$\psi_\alpha(t^*) = e^{-i\omega t^*} + \sum_{m=0}^{N-1} \psi_\alpha^{(m)}(t^*)_{sc}, \tag{17}$$

and

$$\psi_\alpha^{(m)}(t^*) = e^{-i\omega t^*} + \sum_{m' \neq m} \psi_\alpha^{(m')}(t^*)_{sc}, \tag{18}$$

Once we deal with the coherent scattering to zero angle, in (18) only the forward scattered waves with $m' = 0, 1, ... m-1$ are significant.

Let us expand the incident wave $\psi_\alpha^{(m)}(t^*)$ in the functions $\chi_n(t^*) = e^{-i\omega_n t^*}$:

$$\psi_\alpha^{(m)}(t^*) = \sum_{n=-\infty}^{\infty} b_\alpha^{(m)}(n) \chi_n(t^*). \tag{19}$$

Following [31], it can be shown that the corresponding scattered wave is given by

$$\psi_\alpha^{(m)}(t^*)_{sc} = i \sum_{n,n'} F_{n'n} b_\alpha^{(m)}(n) \chi_{n'}(t^*), \tag{20}$$

where the dimensionless forward scattering amplitude of γ quanta by the m th layer is

$$F_{n'n} = F_{n'n}^N + F_{n'n}^R,$$

$$F_{n'n}^{N(R)} = \frac{2\pi F^{(n'-n)}(\mathbf{k}_n \mathbf{e}_\alpha; \mathbf{k}_{n'} \mathbf{e}_\alpha)^{N(R)}}{k(v_0/d)} \tag{21}$$

and v_0 is the volume of elementary cell.

The matrix $F_{n'n}$, depending on the polarization of incident photons α, can be considered formally as a matrix of the operator F_α, acting in the space spanned on the basis vectors χ_n. It allows us to represent the wave scattered by the m th layer as

$$\psi_\alpha^{(m)}(t^*)_{sc} = iF_\alpha \psi_\alpha^{(m)}(t^*). \tag{22}$$

Then the equations of multiple scattering (18) are transformed to

$$\psi^{(m)}(t^*) = \chi_0 + iF \sum_{m'=0}^{m-1} \psi^{(m')}(t^*), \tag{23}$$

or in more brief form to

$$\psi_\alpha^{(m)} = (1 + iF_\alpha) \psi_\alpha^{(m-1)}. \tag{24}$$

Its solution is as follows:

$$\psi_\alpha^{(m)}(t^*) = (1 + iF_\alpha)^m \chi_0. \tag{25}$$

Since the scattering amplitudes $|F_{n'n}| = 1$, then

$$\psi^{(m)}(t^*) \approx e^{imF_\alpha} \chi_0. \tag{26}$$

The eigenvalues $\delta_{\mu\alpha} d$ of the operator F_α are determined by equation

$$F_\alpha \chi'_\mu = (\delta_{\mu\alpha} d) \chi'_\mu, \tag{27}$$

where eigenfunctions are given by the expansion

$$\chi'_\mu = \sum_n u_{n\mu} \chi_n \tag{28}$$

with the transformation matrix $u_{n\mu}$.

Using Eqs. (26) - (28), we are led to

$$\mathbf{A}_\alpha^{(m)}(z,t) = \mathbf{A}_\alpha^{(0)} \sum_{n=-\infty}^{\infty} \sum_{\mu=-\infty}^{\infty} C_n^{(\mu\alpha)} e^{i\delta_{\mu\alpha} md} e^{ik_n z - i\omega_n t}, \tag{29}$$

where the amplitudes are

$$C_n^{(\mu)} = u_{n\mu} u_{\mu 0}^{-1}. \tag{30}$$

From here it follows the boundary condition

$$\sum_{\mu=-\infty}^{\infty} C_n^{(\mu)} = \delta_{n0}, \tag{31}$$

which implies that at the surface $z = 0$ there are only incident photons with frequency ω.

Due to inequality $|\delta_\mu^N| d << 1$, the m th incident wave $\mathbf{A}_\alpha^{(m)}(\mathbf{r},t)$ practically coincides with the complete wave in the vicinity of the m th layer. Therefore inside the crystal, $0 \le z \le D$, one has

$$\mathbf{A}_\alpha(z,t) = \mathbf{A}_\alpha^{(0)} \sum_{\mu=-\infty}^{\infty} \sum_{n=-\infty}^{\infty} C_n^{(\mu\alpha)} e^{iK_{\mu\alpha} z} e^{-i\omega_n t}, \tag{32}$$

where the wave vectors of photons in the medium are $\mathbf{K}_{\mu\alpha}=\{0,0, K_{\mu\alpha}\}$ with

$$K_{\mu\alpha} = k + \delta_{\mu\alpha}. \tag{33}$$

Here the dependence of the wave vector $\mathbf{k}$ on n is neglected since $\Omega D/c << 1$.

The photons transmitted through the crystal ($z > D$) are described by

$$\mathbf{A}_\alpha(z,t)_{tr} = \mathbf{A}_\alpha^{(0)} \sum_{n=-\infty}^{\infty} B_{n\alpha}(\omega) e^{-i\omega_n t^*} \tag{34}$$

with the amplitudes

$$B_{n\alpha}(\omega) = \sum_{\mu=-\infty}^{\infty} C_n^{(\mu,\alpha)}(\omega) e^{i\delta_{\mu,\alpha}(\omega) D}. \tag{35}$$

For determination of $\delta_{\mu\alpha} D$ it is convenient to introduce the dimensionless parameters

$$x = 2(E - E_0')/\Gamma, \quad \Delta x = 2\hbar\Omega/\Gamma, \tag{36}$$

Besides, we introduce a thickness parameter of the absorber $\beta = 4b/\Gamma$, where

$$b = \frac{\sigma_0 \Gamma}{4} e^{-2W_a} n_0. \tag{37}$$

From Eq. (27) it follows that

$$\delta = \delta^R + \delta^N, \tag{38}$$

where

$$\delta^R = F^R / d, \tag{39}$$

while the product $\delta^N D$ is defined by the algebraic equations

$$\sum_{n=-\infty}^{\infty} A_{n'n}^{(\alpha)} u_n^{(\alpha)} = (\delta_\alpha^N D) u_{n'}^{(\alpha)} \tag{40}$$

Here the scattering amplitude $F_{n'n}^N$ is transformed to the matrix

$$A_{n'n}^{(\alpha)} = -\sum_{m=-\infty}^{\infty} \frac{\beta_{n'n}^{(\alpha)}(m)}{x - m\Delta x + i}, \tag{41}$$

which contains the parameters

$$\beta_{n'n}^{(x)}(m) = \frac{\beta}{6}\{3[a_{-3/2,-1/2}^*(m-n')a_{-3/2,-1/2}(m-n) + a_{3/2,1/2}^*(m-n')a_{3/2,1/2}(m-n)] + [a_{1/2,-1/2}^*(m-n')a_{1/2,-1/2}(m-n) + a_{-1/2,1/2}^*(m-n')a_{-1/2,1/2}(m-n)]\} \tag{42}$$

and

$$\beta_{n'n}^{(y)}(m) = \frac{\beta}{4}\{a_{-1/2,-1/2}^*(m-n')a_{-1/2,-1/2}(m-n) + a_{1/2,1/2}^*(m-n')a_{1/2,1/2}(m-n)\}. \tag{43}$$

Thus, our task is reduced to numerical calculation of the eigenvalues δD and eigenvectors $\{u_n\}$ of the matrix $A_{n'n}^{(\alpha)}$.

At high frequencies of the rf field $(\Omega \to \infty)$

$$\beta_{n'n}^{(\alpha)}(m) \to \frac{\beta}{2}\delta_{mn'}\delta_{mn}, \tag{44}$$

since in this limiting case

$$a_{eg}(n) \to \delta_{n0} \tag{45}$$

(see also [4]). As a consequence, the matrix $A_{n'n}^{(\alpha)}$ takes the diagonal form

$$A_{n'n}^{(\alpha)} \to -\frac{\beta/2}{x+i}\delta_{n'n}, \tag{46}$$

which means that no Raman scattering of photons occurs in the crystal and the forward scattering spectrum collapses to single line or doublet.

Spectra

The γ-quantum, emitted by a source without recoil, is described by the wave packet

$$\mathbf{A}_\alpha(z,t)_{in} = \mathbf{A}_\alpha^{(0)} \int_{-\infty}^{\infty} g_s(\omega) e^{ikz - i\omega t} d\omega, \tag{47}$$

where its Fourier transform is

$$g_s(\omega) \infty \frac{e^{i\omega t_0}}{E - E_0 - (v/c)E_0 + i\Gamma_s/2}, \tag{48}$$

while t_0 is the moment of formation of the excited nuclear state, E_0 and Γ_s are the energy and width of the excited level of the emitting nucleus, v is the velocity of the source with respect to the absorber.

The corresponding phononless energy distribution of the incident radiation is described by the Lorentzian function

$$w_s^{(0)}(E) = \frac{\Gamma_s}{2\pi} \frac{e^{-2W_s(k)}}{(E - E_0 - (v/c)E_0)^2 + (\Gamma_s/2)^2}. \quad (49)$$

According to (34), (35) the electromagnetic wave packet, scattered by the crystal to zero angle, will be

$$\mathbf{A}_{sc}(t^*) = \mathbf{A}_\alpha^{(0)} \int_{-\infty}^{\infty} d\omega' g_{sc}^{(\alpha)}(\omega') e^{-i\omega' t^*}, \quad (50)$$

where

$$g_{sc}^{(\alpha)}(\omega') = \sum_{n=-\infty}^{\infty} g_s(\omega' + n\Omega) B_{n\alpha}(\omega' + n\Omega). \quad (51)$$

When the incident beam is unpolarized the energy distribution of γ-quanta behind the target is given by

$$w_{sc}(E') = \frac{1}{2} \sum_{\alpha=x,y} \langle | g_{sc}^{(\alpha)}(\omega') |^2 \rangle, \quad (52)$$

where the brackets $<>$ denote averaging over the initial random moments t_0. The final result for the energy distribution of transmitted radiation reads

$$w_{sc}(E') = \sum_{n=-\infty}^{\infty} w_s^{(0)}(\omega' + n\Omega) \frac{1}{2} \sum_{\alpha=x,y} \left| B_{n\alpha}(\omega' + n\Omega) \right|^2. \quad (53)$$

Such a distribution is measured with the aid of the analyzing crystal with single resonant absorbtion line, which can be approximated by the Lorentz curve

$$w_{an}(E') = \frac{\Gamma_{an}}{2\pi} \frac{1}{(E' - E_0 - (v'/c)E_0)^2 + (\Gamma_{an}/2)^2}, \quad (54)$$

where $E' = \hbar\omega'$ denotes the energy of photons incident on the analyzer, v' is the velocity of the analyzer relative to the target, Γ_{an} is the width of the line. Then the experimentally measured double resonance spectrum is described by the following integral:

$$W_{sc}(v') = \int_{-\infty}^{\infty} dE' w_{an}(E') w_{tr}(E'). \quad (55)$$

Such spectrum is measured with a fixed source.

As to the absorption spectrum, depending on the source velocity v, it is given by the expression

$$W_a(v) = \int_{-\infty}^{\infty} dE' w_{sc}(E'). \quad (56)$$

Experiments

Our experimental set-up, shown in Fig. 1, consisted of the source (S) of Mössbauer rays, absorbing sample (A), analyzer (An) of the forward scattered radiation and detector (D) behind the analyzer. During measurements of forward scattering spectra the source (^{57}Co(Cr)) was at rest with respect to the sample $FeBO_3$. The width of the source line was $\Gamma_s = 0.13$ mm/s.

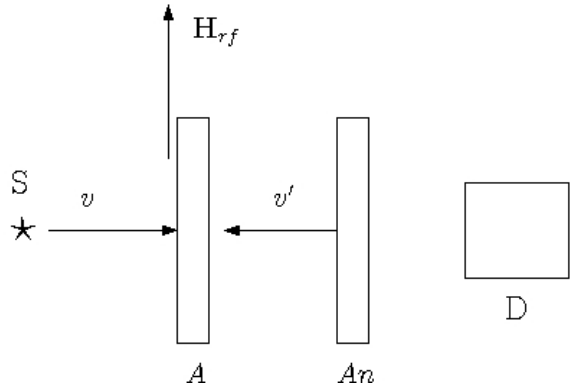

Fig. 1. Scheme of the experimental set-up for measuring forward scattering spectra, where S is the source of Mössbauer rays, A is the absorbing crystal $FeBO_3$ placed in the rf magnetic field, An is the analyzer of transmitted radiation, D is the detector.

As a target we took a $FeBO_3$ single crystal enriched with ^{57}Fe up to 95 %. $FeBO_3$ is a weak ferromagnet (canted antiferromagnet) with the Neel temperature $T_N = 348$ K. It was cut along the easy-magnetization plane (111), in which the magnetization vectors of two almost antiferromagnetic sublattices are lying. The easy plane has very low anisotropy field $H_a \infty 1$ Oe, while that along the hard axis is $6.25 \cdot 10^4$ Oe [35]. This iron borate platelet had transverse dimensions 4 mm and `thickness $D = 45\mu$ m. It was placed inside the helical contour, which produced the rf magnetic field, oscillating in the plane (111) of the target perpendicularly to incident γ rays. High-frequency generator, operating at the frequencies from 10 to 30 MHz, generated the power up to 120 W and the amplitude of the rf magnetic field H_0 achieving values up to 20 Oe. The sample temperature $T = 340 \pm 0.1$ K was kept within ± 0.1 K by means of a thermostat.

Since $H_0 \approx H_a$, the crystal magnetization $\mathbf{M}$, driven by the external rf field, periodically reverses in the easy plane. It is achieved by simultaneous reversals of the magnetization vectors of antiferromagnetic sublattices between opposite

directions. Such reversals are facilitated when the temperature T is near T_N, as in our experiment with T approaching T_N. At $T = 340$ K, even in the absence of rf field, the chaotic rotations and reversals of the magnetization M lead to narrowing and poor resolution of the sextet, whereas at $T \geq T_N$ the crystal becomes paramagnetic and the spectrum collapses into unresolved quadrupole doublet [36].

Between the absorber and detector it was placed the analyzer - another absorbing crystal, moving with constant acceleration and alternating velocity v'. It has been prepared from potassium hexacyanoferrate (II) trihydrate $K_4[Fe(CN)_6]3H_2O$.

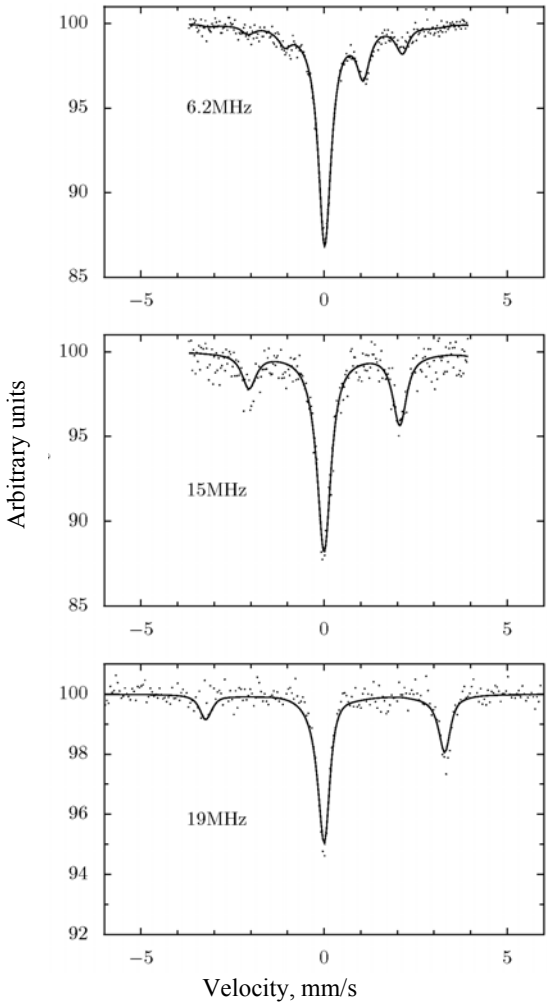

Fig. 2. Mössbauer forward scattering spectra of $FeBO_3$ in the rf field vs velocity of the analyzer. Dots are experimental data, solid line represent the calculations.

Mössbauer forward scattering spectra of $FeBO_3$ in the rf field vs velocity of the analyzer. Dots are experimental data, solid line represent the calculations, known also as potassium ferrocyanide (PFC) or yellow potassium prussiate, YPP). This compound was enriched in ^{57}Fe to 95 %. The absorption spectrum of the analyzer was fitted by a single Lorentzian line, having the width $\Gamma_{an} = 0.37$ mm/s. The Doppler modulation of the analyzer was achieved by means of a standard MS 1101 E Mössbauer spectrometer with constant acceleration mode. The detector with thin NaJ(Tl) scintillator was used.

The measured rf forward scattering spectra for $FeBO_3$ consist of the central line $(n = 0)$ and even sidebands,$(n = \pm 2, \pm 4, ...)$ separated from each other by the double driving rf frequency $\Omega = 2\pi\nu$ (Fig. 2). The satellite intensities fall down with increasing their order n much faster than in the case of ultrasound modulation [16]. Our FS spectra are asymmetric (the right-hand satellites have larger amplitude than left-hand ones) in analogy with FS spectra in the ultrasound experiment [16]). The asymmetry arises because the incident radiation is shifted with respect to the center of the Zeeman sextet due to the isomer shift $\delta = E_0' - E_0$, in our case $\delta = 0.35$ mm/s.

In addition we have measured the absorption spectrum at frequency $\nu = 19$ MHz, which is displaced in Fig. 3. In contrast to the FS spectra it contains lines of all orders $n = 0, \pm 1, \pm 2, ...$

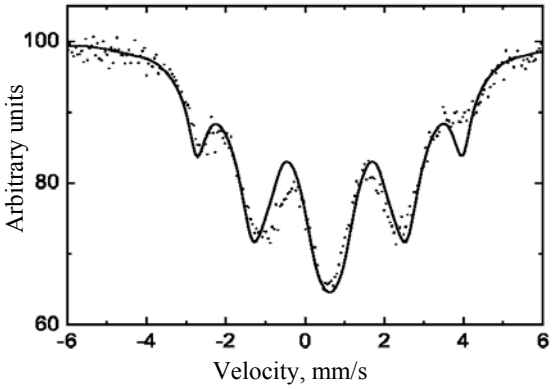

Fig. 3. Mössbauer absorption spectra of $FeBO_3$ in the rf field vs velocity of the source. Dots are experimental data, solid line represent the calculations.

Discussion

The Raman scattering of γ-rays by nuclei in rf field with the circular frequency Ω leads to splitting of their frequencies ω into $\omega_n = \omega - n\Omega$, where n is an integer. We considered their penetration through a crystal far from the Bragg condition, when the coherent waves $\mathbf{A}_\mu(z,t)$ arise with different wave vectors $\mathbf{K}_\mu$ and amplitudes C_n^μ but the same

frequency ω_n. Their coupling is determined by the eigenvalue equation (27), which is reduced to the infinite system of algebraic equations (40) with the matrix (41). The system can be truncated at some $|n|$ because the amplitudes C_n^{μ} rapidly decrease with growing $|n|$. More general system of equations to determine gamma-waves in the crystal subject to any alternating field has been derived in [31]. They describe the case when inelastic (Raman) diffraction of waves is realized in the crystal. It was shown in the same paper that during inelastic diffraction there arise the same effects (suppression of inelastic channels, pendellösung effect, etc.) as for elastic diffraction. In particular case when no Bragg condition for gamma-waves is fulfilled in the crystal, these general equations reduce to the equations derived above. It is worth to note also that in this paper we gave another and somewhat more strict derivation of the dynamical equations govering the amplitudes and wave vectors of all the waves. They take into account rescattering of waves inside the crystal, i.e., the fact that the wave incident on the mth crystal plane $\mathbf{A}_{\alpha}^{(m)}(t^*)$ is formed by the sum of the wave incident on the crystal $\mathbf{A}_{\alpha}^{(0)}e^{-i\omega t^*}$ and waves scattered by all other crystal planes. For very thin films in the kinematic approximation rescattering of waves can be ignored, then all the incident waves $\mathbf{A}_{\alpha}^{(m)}(t^*)$ are replaced by $\mathbf{A}_{\alpha}^{(0)}e^{-i\omega t^*}$. Every component with definite frequency ω_n of gamma-wave inside the crystal is a coherent sum of the waves with different amplitudes $C_{\mu\alpha}$ and wave vectors $\mathbf{K}_{\mu\alpha}$. Their interference defines attenuation in the crystal of the beam with frequency ω_n. Both radiative and conversion electron channels as well as the Rayleigh electronic scattering contribute to this attenuation.

The periodic reversals of the magnetic field and magnetostrictive vibrations are taken into account in our equations. We assumed that the field reversals occur instantly and simultaneously in the whole crystal volume. Good agreement of our model of instant reversals with the experiment says that the duration of the magnetization rotation in $FeBO_3$ is much less than the rf period T, i.e. $<< 50$ ns. Note that the estimations of [37] predict this switching time from 10 ns to 40 ns. Following [7] we believe that the frequency of vibrations is 2Ω. Therefore γ-quantum, interacting with such vibrating nucleus, can exchange its frequency ω by $n\Omega$ with even n.

Once the nucleus is exposed to the rf magnetic field, its ground and excited levels split into infinite series of quasi-energetic levels, which are spaced by the interval $\hbar\Omega$. Emission or absorption of γ-quanta by such a nucleus is accompanied by nuclear transition between quasi-energetic levels of the ground set and excited set. As a result, the nucleus absorbs and emits γ-quanta with energy shifted by any number of rf quanta $\hbar\Omega$. In other words, γ-quantum emits or absorbs rf photons. Note that in the vacuum such a process of emission or absorbtion of one photon by another is practically inhibited.

Respectively, the absorption spectrum, presented in Fig. 3, contains equidistant sidebands of all orders $n = \pm 1, \pm 2, ...$ Nevertheless, it is not the case for the coherent rf forward scattering in standard geometry with perpendicular $\mathbf{H}(t)$ and beam of γ-rays. Then the coherent wave exchanges with the rf field only by couples of the rf photons, and the forward scattering spectrum (see Fig. 2) contains only even sidebands $(n = \pm 2, \pm 4, ...)$. It is ensured by interference of the terms $f_{1/21/2}^{(n)}$ and $f_{-1/2,-1/2}^{(n)}$ in the coherent scattering amplitude (see also [21, 22]), which is destructive for odd numbers n.

Good fitting of our data for $FeBO_3$ in the rf magnetic field is obtained, assuming that $\mathbf{kA} = 0$ and taking into consideration only periodic magnetization reversals. Thus, we can conclude that no significant magnetostrictive vibrations are excited in iron borate perpendicularly to the external rf field $\mathbf{H}(t)$ and sidebands in rf Mössbauer spectra of $FeBO_3$ are caused mainly by periodic reversals of the magnetic field at the nuclei. So the magnetostrictive standing wave, which is excited in the transversal direction to the beam of γ-photons, is not scattered effectively by defects in $FeBO_3$ to give rise to vibrations along this beam. Therefore the magnetostrictive mechanism for the sideband formation in $FeBO_3$, proposed in [36], seems to be insufficient. Note that the magnetostrictive vibrations are excited with double rf frequency. If only these vibrations were responsible for appearance of sidebands, then the sidebands in the absorption spectra would be separated by the interval 2Ω, while our spectra as well as those of [36] clearly demonstrate that the absorption lines are spaced by Ω.

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СПЕКТР МЕССБАУЕРІВСЬКОГО РОЗСІЯННЯ ВПЕРЕД ФЕРОМАГНЕТИКАМИ В РАДІОЧАСТОТНОМУ МАГНІТНОМУ ПОЛІ

Вивчається проходження мессбауерівського випромінювання крізь товстий феромагнітний кристал, що перебуває в радіочастотному магнітному полі. Розвинуто квантовомеханічну динамічну теорію розсіяння, яка бере до уваги періодичні реверсії магнітного поля на ядрах та когерентні коливання ядер. Поміряно мессбауерівський спектр розсіяння вперед м'якого феромагнетика $FeBO_3$, що знаходиться в радіочастотному магнітному полі. Виявлено, що когерентна гамма-хвиля в кристалі випромінює чи поглинає тільки пари радіочастотних фотонів, унаслідок чого спектр розсіяння вперед складається із рівновіддалених ліній, розділених на подвійну частоту, у протилежність до спектра поглинання. Наші експериментальні дані та числові розрахунки добре узгоджуються, якщо припустити, що надтонке поле на ядрах $FeBO_3$ періодично реверсує без будь-яких когерентних коливань.

Ключові слова: мессбауерівська спектроскопія, спектри розсіяння вперед, борат заліза, радіочастотне магнітне поле.

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СПЕКТР МЕССБАУЭРОВСКОГО РАССЕЯНИЯ ВПЕРЕД ФЕРРОМАГНЕТИКАМИ В РАДИОЧАСТОТНОМ МАГНИТНОМ ПОЛЕ

Изучается прохождение мессбауэровского излучения сквозь толстый ферромагнитный кристалл, находящийся в радиочастотном магнитном поле. Развита квантовомеханическая динамическая теория рассеяния, принимающая во внимание периодические реверсии магнитного поля на ядрах и когерентные коллебания ядер. Измерен мессбауеровский спектр рассеяния вперед мягкого ферромагнетика $FeBO_3$, находящегося в радиочастотном магнитном поле. Обнаружено, что когерентная гамма-волна в кристале излучает или поглощает только пары радиочастотных фотонов, вследствие чего спектр рассеяния вперед состоит из равноудаленных линий, разделенных на двойную частоту, в противоположность спектру поглощения. Наши экспериментальные данные и численные расчеты хорошо согласуются, если допустить, что сверхтонкое поле на ядрах $FeBO_3$ периодически реверсирует без каких-либо когерентных коллебаний.

Ключевые слова: мессбауэровская спектроскопия, спектры рассеяния вперед, борат железа, радиочастотное магнитное поле.

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