# MÖSSBAUER ABSORPTION BY THICK FERROMAGNETS IN RADIO-FREQUENCY MAGNETIC FIELD

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**UDC 539.32:621.315.592**
**‰ 2002**

The dynamical scattering theory is developed for transmission of the Mössbauer radiation through a ferromagnetic absorber of arbitrary thickness whose magnetization periodically reverses under the influence of an external radio-frequency (RF) magnetic field. The thickness dependence of the Mössbauer absorption spectrum as well as the time dependence and energy distribution of the transmitted beam are analyzed. The transmitted spectrum as a function of the frequency of transmitted $\gamma$-quanta, reveals a sideband structure separated by twice the frequency of the RF field, which collapses to a single line at high frequencies.

## Introduction

Many theoretical papers were devoted to analysis of the Mössbauer absorption in soft ferromagnets placed in a RF magnetic field [1 – 6]. Unfortunately, all they restrict themselves by the kinematical approximation, when rescattering of $\gamma$-quanta inside an absorder is ignored. Such an approach is valid only for thin absorbing films, for which

$$n_0 \sigma_a << 1, \qquad (1)$$

where $n_0$ is the number of Mössbauer isotopes per unit area, $\sigma_a$ is the absorbing cross-section for $\gamma$-quanta. In most experiments, this constraint breaks, therefore one needs to build the dynamical scattering theory, taking into account the multiple scattering of $\gamma$-quanta in a crystal.

Let a soft ferromagnet be located in an external RF magnetic field with circular frequency $\Omega$. This field forces the crystal magnetization to reverse with the same frequency. Such reversals ensure the corresponding periodic jumps of the magnetic field at the Mössbauer nucleus between two walues $+\mathbf{h}_0$ and $-\mathbf{h}_0$. This is the main idea of the step-like reversals model [1 – 6], which explained such peculiarities of the Mössbauer spectra as the collapse of its hyperfine structure to a single line or doublet at high frequencies $\Omega$ as well as the existence of sidebands spaced by $\Omega$ at intermediate values of $\Omega$. It was observed first by Pfeiffer [7, 8] (see also surveys [9 – 11]). There are two reasons for the formation of these sidebands: first, the exchange by modulation photons $\hbar\Omega$ between $\gamma$-quanta and magnetostrictive vibrations which are generated in the ferromagnet by the RF magnetic field; second, the exchange by RF photons with frequency $\Omega$ between $\gamma$-quanta and the alternating magnetic field. In the recent experiment [12], we analyzed the role of the latter process, as long as the magnetostrictive vibrations were suppressed. This was achieved by taking a very thin absorbing permalloy film with thickness $\approx 0.6\,\mu$m, that worthened the resonant conditions for the vibrations. It was found that the step-like model excellently explained our observations. In principle, the magnetic field at different nuclei alters its direction at different moments of time. But in very soft ferromagnets, like $FeBO_3$ used in experiment [13, 14], these jumps occur practically simultaneously over all the crystal. Starting from such an assumption, we explained [15] the observations published in [13, 14]. Therefore, we shall again assume that the magnetic fields at all the nuclei throughout the crystal jump instantaneously at the moment $t = 0$ from the value $\mathbf{h}_0$ to $-\mathbf{h}_0$, and, in the backward direction, a jump occurs in time $T/2$. Such reversals are repeated with the period $T = 2\pi/\Omega$.

## 1. Scattering Amplitude

Let us direct the axis $z$ perpendicularly to the slab, which occupies the space $0 \le z \le D$, where $D$ is the slab thickness. Assume that the wave vector of incident $\gamma$-quanta $\mathbf{k}_0 = \{0, 0, k_0\}$ is parallel to this axis. Further, we suppose the field $\mathbf{h}_0$ to lie along the axis $x$, that is typical of the Mössbauer experiments in RF fields [7 – 14]. The $\gamma$-quanta with linear polarization $\mathbf{e}_x$ generate the transitions $M_g \to M_e = M_g \pm 1$ in a nucleus $^{57}Fe$; and those with polarization $\mathbf{e}_y$ are absorbed if $M_e = M_g$. Hereafter, $I_\kappa$ and $M_\kappa$ are the spin and its projection on the direction $+\mathbf{h}_0$ in the ground $(\kappa = g)$ and excited $(\kappa = e)$ states. Incident $\gamma$-quanta with frequency $\omega = E/\hbar$, that are scattered to the forward direction by nuclei in the RF field, give rise to $\gamma$-quanta with the wave vectors $\mathbf{k}_n = \{0, 0, k_n\}$, where $n$ is an integer, $k_n = \omega_n/c$, and the frequencies $\omega_n = \omega - n\Omega$. The general expression for the coherent Raman scattering amplitude of $\gamma$-

quanta by the $j$-th nucleus in a RF field was derived in [4]. For the particular case of straight-forward scattering, it reduces to

$$f_{\text{coh}}^{(n',n)}(\mathbf{k}_n, \mathbf{e}_\alpha; \mathbf{k}_{n'}, \mathbf{e}_\alpha)_j^N = -p_j e^{-2W_a^j(\mathbf{k})} \frac{1}{2I_g+1} \times$$

$$\times \sum_{m=-\infty} \sum_{M_e, M_g} \frac{|\langle e| j_\alpha^N(\mathbf{k})| g\rangle|^2 a_{eg}^*(m-n')\, a_{eg}(m-n)}{E - E'_0 - m\hbar\Omega + i\Gamma/2}, \tag{2}$$

where $p_j$ is the fraction of the Mössbauer isotope in the $j$-th site, $e^{-2W_a}$ is the Debye – Waller factor for the absorber, $E'_0$ and $\Gamma$ are, respectively, the energy and width of the resonant level of the scattering nucleus; $|\kappa\rangle = |I_\kappa M_\kappa\rangle$ are the stationary wave functions of the nucleus in the field + $\mathbf{h}_0$; the operator

$$j_\alpha^N(\mathbf{k}) = \mathbf{e}_\alpha \int d\,\mathbf{r}\, e^{i\mathbf{kr}}\, \mathbf{j}_N(\mathbf{r}), \tag{3}$$

and $\mathbf{j}_N(\mathbf{r})$ is the current density operator of the nucleus. The Fourier coefficients $a_{eg}(n)$ are as follows [4]:

$$a_{eg}(n) = \exp\left[-i\left(\frac{x_{eg}+n\pi}{2}\right)\right] \times$$

$$\times \sin\left(\frac{x_{eg}+n\pi}{2}\right) \frac{2x_{eg}}{x_{eg}^2 - (n\pi)^2}, \tag{4}$$

where

$$x_{eg} = \frac{\alpha_{eg} T}{2}, \quad \alpha_{eg} = (\gamma_g M_g - \gamma_e M_e)\, h_0/\hbar. \tag{5}$$

The amplitude $f^{(n)}$ describes the Raman scattering of γ-quanta with emission $(n > 0)$ or absorption $(n < 0)$ of $n$ modulation quanta $\hbar\Omega$. The reflection $M_e, M_g \to -M_e, -M_g$ does not change the factor $|\langle e| j_\alpha^N(\mathbf{k})| g\rangle|^2$, where $\mathbf{k}$ is perpendicular to $\mathbf{h}_0$. Then, for $E \approx E'_0$ and $\hbar\Omega >> \Gamma/2$, one finds after the substitution of (4) in (2) that the scattering amplitude $f_{\text{coh}}^{(n)}(\mathbf{k}_0, \mathbf{e}_\alpha; \mathbf{k}_n, \mathbf{e}_\alpha) = 0$ for odd $n$. In other words, γ-rays can emit or absorb only even number of photons with frequency Ω. The reason is that the contributions of transitions $M_e \to M_g$ and $-M_e \to -M_g$ into $f^{(n)}$ compensate each other as $n = \pm 1, \pm 2, \ldots$

## 2. Electromagnetic Waves

Let us consider the propagation of γ-quanta with one of the eigenpolarizations $\mathbf{e}_\alpha$ $(\alpha = x$ or $y)$ through the crystal far from the Bragg condition. Then the initial state of the quantized field is $|\mathbf{k}_0, \mathbf{e}_0\rangle$. Such an incident γ-quantum may be described by the electric field strength [16]

$$\mathbf{E}^{(\alpha)}(\mathbf{r}, t) = \langle 0| \hat{\mathbf{E}}(\mathbf{r}, t)| \mathbf{k}_0, \mathbf{e}_\alpha\rangle = \mathbf{E}_0^{(\alpha)} e^{ik_0 z - i\omega t}, \tag{6}$$

where $|0\rangle$ stands for the vacuum state of the electromagnetic field, $\hat{\mathbf{E}}(\mathbf{r}, t)$ is the electric field strength operator, $\mathbf{E}_0^\alpha = i(2\pi\hbar\omega)^{1/2}\mathbf{e}_\alpha$ is the amplitude of the wave incident on the crystal. Since the RF field is uniform in the crystal, the wave vectors of electromagnetic waves inside the crystal $\mathbf{K}$ do not depend on the number $n$:

$$\mathbf{K}_\mu^{(\alpha)} = \{0, 0, k_0 + \delta_{\mu,\alpha}(\omega)\}. \tag{7}$$

If $0 \le z \le D$, the electromagnetic wave may be written as a superposition of plane waves

$$\mathbf{E}^{(\alpha)}(\mathbf{r}, t) = \mathbf{E}_0^{(\alpha)} \sum_{n=-\infty}^{\infty} \sum_{\mu=-\infty}^{\infty} C_n^{(\mu,\alpha)}(\omega) e^{iK_\mu^{(\alpha)} z - i\omega_n t}. \tag{8}$$

Matching waves (6) and (8) at the face surface of the slab $(z = 0)$ provides the boundary condition for the amplitudes:

$$\sum_\mu C_n^{(\mu,\alpha)} = \delta_{n0}. \tag{9}$$

The wave transmitted through the crystal will be

$$\mathbf{E}^{(\alpha)}(z,t)_{\text{tr}} = \mathbf{E}_0^{(\alpha)} \sum_{n=-\infty}^{\infty} \sum_{\mu=-\infty}^{\infty} C_n^{(\mu,\alpha)}(\omega)\, e^{i\delta_{\mu,\alpha}(\omega) D} e^{-i\omega_n \tilde{t}}, \tag{10}$$

where $\tilde{t} = t - z/c$ is the retarded time. Expression (10) is obtained in the approximation $\Omega D/c << 1$, which is well fulfilled for the typical situation when the frequency $\nu = \Omega/2\pi \le 100$ MHz and $D \approx 10\,\mu$m [7 – 11].

The general system of equations to define amplitudes and wave vectors inside a crystal was derived in [17]. Far from the Bragg condition when the alternating field is uniform in a crystal, this system reduces to

$$(K^2 - k_0^2)\, C_{n'}^{(\alpha)} =$$

$$= \frac{4\pi}{\upsilon_0} \sum_{n=-\infty}^{\infty} F^{(n'-n)} (\mathbf{k}_n, \mathbf{e}_\alpha; \mathbf{k}_{n'}, \mathbf{e}_\alpha)\, C_n^{(\alpha)}, \qquad (11)$$

where $\upsilon_0$ is the volume of an elementary cell, $F^{(n)}$ is the coherent Raman scattering amplitude of $\gamma$-quanta by an elementary cell to the forward direction ($\mathbf{k}_n$ are directed along the axis $z$):

$$F^{(n)} (\mathbf{k}_n, \mathbf{e}_\alpha; \mathbf{k}_{n'}, \mathbf{e}_\alpha) =$$

$$= F (\mathbf{k}_n, \mathbf{e}_\alpha; \mathbf{k}_{n'}, \mathbf{e}_\alpha)^R \delta_{n0} + F^{(n)} (\mathbf{k}_n, \mathbf{e}_\alpha; \mathbf{k}_{n'}, \mathbf{e}_\alpha)^N, \qquad (12)$$

where $F^R$ and $F^N$ are, respectively, the Rayleigh and nuclear coherent scattering amplitudes to zero angle. In particular,

$$F^{(n)} (\mathbf{k}_n, \mathbf{e}_\alpha; \mathbf{k}_{n'}, \mathbf{e}_\alpha)^N = \sum_j f_{\text{coh}}^{(n)} (\mathbf{k}_n, \mathbf{e}_\alpha; \mathbf{k}_{n'}, \mathbf{e}_\alpha)_j^N, \qquad (13)$$

where summation is carried out over all the nuclei within an elementary cell.

Taking into account that $|\delta| << k_0$ after the substitution of (8) in (11), one gets

$$\delta = \delta^R + \delta^N, \qquad (14)$$

where

$$\delta^R = \frac{2\pi}{k_0 \upsilon_0} F (\mathbf{k}_n, \mathbf{e}_\alpha; \mathbf{k}_{n'}, \mathbf{e}_\alpha)^R, \qquad (15)$$

and $\delta^N$ together with $C_n$ are the eigenvalues and eigenvectors of the matrix

$$\frac{2\pi}{k_0 \upsilon_0} F^{(n'-n)} (\mathbf{k}_n, \mathbf{e}_\alpha; \mathbf{k}_{n'}, \mathbf{e}_\alpha)^N. \qquad (16)$$

It is convenient to introduce the dimensionless parameters

$$x = 2 (E - E'_0)/\Gamma, \quad \beta = 4b/\Gamma, \quad \Delta x = 2\hbar\Omega/\Gamma, \qquad (17)$$

where $b$ is the thickness parameter of the absorber,

$$b = \frac{\sigma_0 \Gamma}{4} e^{-2W_a} n, \qquad (18)$$

and $\sigma_0$ is the resonant absorption cross-section. Then the algebraic equations for $\delta^N D$ and $C$ may be written as

$$\sum_{n=-\infty}^{\infty} A_{n'n}^{(\alpha)} C_n^{(\alpha)} (x) = \delta_\alpha^N (x)\, D\, C_{n'}^{(\alpha)} (x) \qquad (19)$$

with the matrix

$$A_{n'n}^{(\alpha)} = - \sum_{m=-\infty}^{\infty} \frac{\beta_{n'n}^{(\alpha)} (m)}{x - m\Delta x + i}, \qquad (20)$$

where

$$\beta_{n'n}^{(x)} (m) =$$

$$= \frac{\beta}{16} \{3 [a^*_{-3/2,-1/2} (m - n') a_{-3/2,-1/2} (m - n) +$$

$$+ a^*_{3/2,2/1} (m - n') a_{3/2,1/2} (m - n)] +$$

$$+ a^*_{1/2,-1/2} (m - n') a_{1/2,-1/2} (m - n) +$$

$$+ a^*_{-1/2,1/2} (m - n') a_{-1/2,1/2} (m - n)]\},$$

$$\beta_{n'n}^{(y)} (m) =$$

$$= \frac{\beta}{4} \{a^*_{-1/2,-1/2} (m - n') a_{-1/2,-1/2} (m - n) +$$

$$+ a^*_{1/2,2/1} (m - n') a_{1/2,1/2} (m - n)\}. \qquad (21)$$

At high frequencies of the RF field ($\Omega \to \infty$), we have

$$\beta_{n'n}^{(\alpha)} (m) \to \frac{\beta}{2} \delta_{mn'} \delta_{mn}, \qquad (22)$$

since

$$a_{eg} (n) \to \delta_{n0} \qquad (23)$$

in this limiting case (see also [4]). As a consequence, the matrix $A_{n'n}^{(\alpha)}$ takes the diagonal form under these conditions:

$$A_{n'n}^{(\alpha)} \to - \frac{\beta/2}{x - n\Delta x + i} \delta_{n'n}. \qquad (24)$$

Combining this with the boundary constraint (9), we see that, in the regime of RF collapse, there will be only a single transmitting wave

$$\mathbf{E}_\alpha (z, t) = \mathbf{E}_0^{(\alpha)} e^{iKz - i\omega t}, \quad 0 \le z \le D, \qquad (25)$$

with the wave vector

$$\mathbf{K} = \mathbf{k}_0 + \frac{2\pi F^{(0)} (\mathbf{k}_0, \mathbf{e}_\alpha; \mathbf{k}_0, \mathbf{e}_\alpha)}{k_0 \upsilon_0} \mathbf{e}_z. \qquad (26)$$

## 3. Transmitted Beam

If incident γ-quanta had frequency ω and one of the eigenpolarizations $\mathbf{e}_x$ or $\mathbf{e}_y$, then the intensity of the transmitted beam would be $\sim |\mathbf{E}(z,t)|^2$ with $\mathbf{E}(z,t)$ given by Eq. (10). This quantity should be averaged over the phononless energy distribution of incident γ-quanta

$$w_e^{(0)}(\omega) = \frac{\Gamma}{2\pi}\frac{e^{-2W_e}}{(E-E_0)^2+(\Gamma/2)^2}, \quad (27)$$

where $E_0$ and $e^{-2W_e}$ are the resonant energy and Debye – Waller factor for the emitter. Then the instantaneous flux density of the transmitted beam in units $j_0 e^{-\mu_e D}$ ($j_0$ is the flux density of incident γ-quanta and $\mu_e$ is the electron absorption coefficient) is given by

$$j^{\alpha}(t) = B + \frac{1}{\pi}\int_{-\infty}^{\infty} dx \frac{e^{-2W_e}}{(x-x_0)^2+1}\times$$

$$\times \left| \sum_{n,\,\mu=-\infty}^{\infty} C_n^{(\mu,\alpha)}(x)\, e^{i\delta^N_{\mu,\alpha}(x)D} e^{in\Delta x\tau/2}\right|^2, \quad (28)$$

where $\tau = \Gamma\tilde{t}/\hbar$ is the time in units of the nuclear lifetime $\hbar/\Gamma$, and the first term $B = (1-e^{-2W_e})$ is responsible for the background. In addition, we average (28) over polarizations. For an unpolarized incident beam,

$$j(t) = \frac{1}{2}(j^{(x)}(t) + j^{(y)}(t)). \quad (29)$$

The intensity of the transmitted beam averaged over time reads

$$j = \langle j(t)\rangle = B +$$

$$+\frac{1}{2}\sum_{\alpha=x,y}\int_{-\infty}^{\infty} dx\, w_e^{(0)}(x)\sum_{n=-\infty}^{\infty}\left|\sum_{\mu=-\infty}^{\infty} C_n^{(\mu,\alpha)}(x)\, e^{i\delta^N_{\mu,\alpha}(x)D}\right|^2. \quad (30)$$

The function $j$ vs relative velocity $\upsilon$ of the emitter and absorber determines the Mössbauer absorption spectrum taken with an absorber of arbitrary thickness.

## 4. Electromagnetic Wave Packets

The incident γ-quantum, emitted by the nucleus being excited at the moment $t_0$, is described by the wave packet

$$\mathbf{E}(z,t) = \mathbf{E}_0\int_{-\infty}^{\infty} g(\omega)\, e^{ikz-i\omega t} d\omega, \quad (31)$$

where

$$g(\omega) = -\frac{1}{2\pi i}\frac{e^{i\omega t_0}}{E-E_0+i\Gamma/2}, \quad E = \hbar\omega. \quad (32)$$

Substitution of (32) in (31) gives the well-known formula for the incident wave:

$$\mathbf{E}(z,t) = \mathbf{E}_0\, e^{-i\omega_0(t-t_0-z/c)-\Gamma(t-t_0)/2\hbar}\,\Theta(t-t_0), \quad (33)$$

where

$$\Theta(x) = \begin{cases} 1, & x>0, \\ 0, & x<0. \end{cases} \quad (34)$$

The transmitted electromagnetic wave packet will be as follows:

$$E_{\rm tr}^{(\alpha)}(z,t) = \mathbf{E}_0^{(\alpha)}\int_{-\infty}^{\infty} d\omega\, g(\omega)\times$$

$$\times\sum_{n,\mu=-\infty}^{\infty} C_n^{(\mu,\alpha)}(\omega)\, e^{i\delta_{\mu,\alpha}(\omega)D} e^{-i\omega_n\tilde{t}}. \quad (35)$$

On the other hand, it can be written as

$$E_{\rm tr}^{(\alpha)}(z,t) = \mathbf{E}_0^{(\alpha)}\int_{-\infty}^{\infty} d\omega'\, g_{\rm tr}^{(\alpha)}(\omega')\, e^{-i\omega'\tilde{t}}. \quad (36)$$

Equating (35) and (36), one finds the energy representation of a transmitted coherent wave as

$$g_{\rm tr}^{(\alpha)}(\omega') = \sum_{n,\,\mu=-\infty}^{\infty} g(\omega'+n\Omega)\, C_n^{(\mu,\alpha)}(\omega'+n\Omega)\times$$

$$\times\, e^{i\delta_{\mu,\alpha}(\omega'+n\Omega)D}. \quad (37)$$

The frequency distribution of these transmitted wave packets for unpolarized incident beams will be

$$w_{\rm tr}(\omega') \approx \sum_{\alpha=x,y} |g_{\rm tr}^{(\alpha)}(\omega')|^2. \quad (38)$$

Its averaging over the initial moments $t_0$, yields the final results for the energy distribution of a transmitted beam:

$$w_{\rm tr}(\omega') \approx \sum_{n=-\infty}^{\infty} w_e^{(0)}(\omega'+n\Omega)\times$$

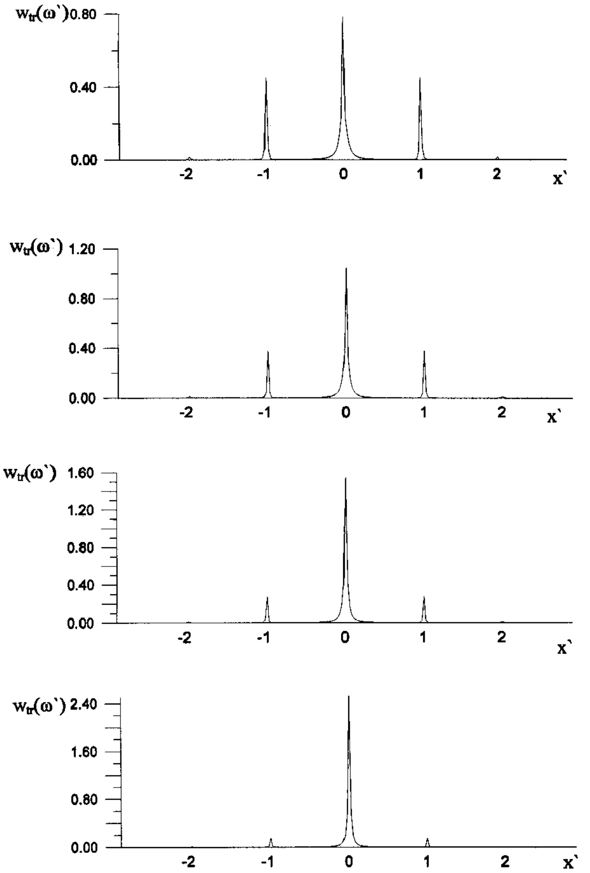


Fig. 1. Frequency distribution of the Mössbauer radiation transmitted through a soft ferromagnet with reversing magnetization

$$\times \sum_{\alpha = x,y} \left| \sum_{\mu=-\infty}^{\infty} C_n^{(\mu,\alpha)} (\omega' + n\Omega) \, e^{i\delta_{\mu,\alpha}(\omega' + n\Omega) D} \right|^2 . \quad (39)$$

The distribution $w_{tr}(\omega')$, as a function of the dimensionless parameter $x' = 2\hbar(\omega' - \omega_0)/\Gamma$, is plotted in Fig. 1 for the case $\omega_0 = \omega_0'$, when the incident radiation is tuned just to the central peak. The frequency $\nu = 30$ MHz. The thickness parameter $\beta$ runs the values 2, 4, 6, 8.

In [14], the experiment was reported, where the magnetic field at Mössbauer nuclei was changed by sudden application of the external magnetic field. The reversed state of magnetization was maintained for 400 ns and the period of reversals was 2 $\mu s$. There was observed the broadening of resonant lines which was explained in [14] by the influence of vibrations. At the same time, Shvydko et al. [14] ignored completely the role of field reversals. But, as shown in [2, 5, 12], both mechanisms (reversals and vibrations) are responsible for the shape of the observed spectra. Moreover, at low frequencies, the amplitude of magnetostrictive vibrations is small [10, 11]. Therefore, we tried to explain the results due to [14] only by the influence of reversals, by assuming that the amplitude of vibrations $x_0 = 0$. The results of our calculations are compared with the data [14] in Fig. 2. We use the parameters $\beta = 174.4$, $\Gamma = 0.097$. Following [5, 12], we take the asymmetry parameter $R = 0.6$.

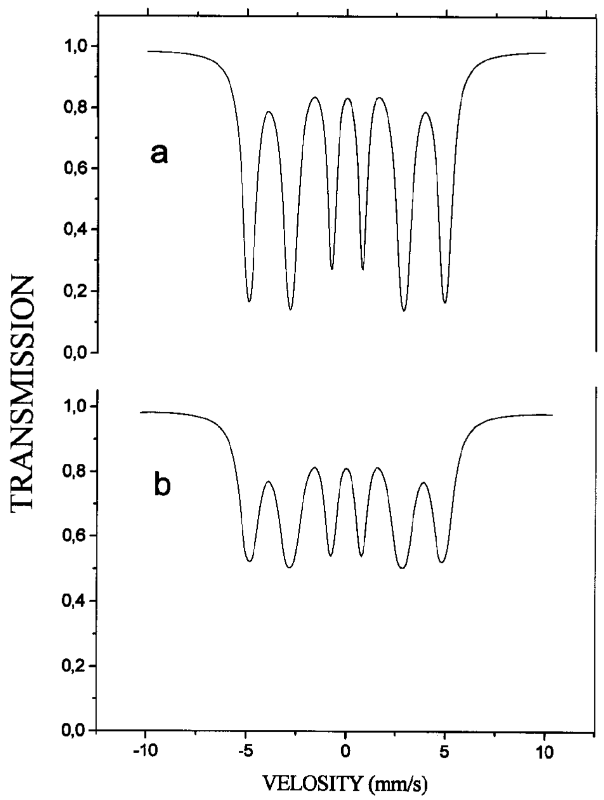


Fig. 2. Effect of low-frequency reversals with $T = 2$ μs on the shape of the Mössbauer spectrum: *a* — our calculations, *b* — experimental data

## Discussion

Raman scattering of γ-rays by nuclei exposed to a RF field inside the crystal gives rise to electromagnetic waves with different wave vectors and frequencies. Their coupling is determined by the system of algebraic equations (19), (20). In numerical calculations, we truncated them at $|n| = 20$. These calculations are illustrated by the Figs. 1 – 3. We assumed only the periodic reversals of the magnetic field which are synchronized at all the nuclei of the absorber and did not take into account any magnetostrictive vibrations. Therefore, our calculations may be directly applied in experiments with damped vibrations. It is worth noting that the heighboring peaks in the frequency spectrum of transmitted radiation are separated by $2\Omega$

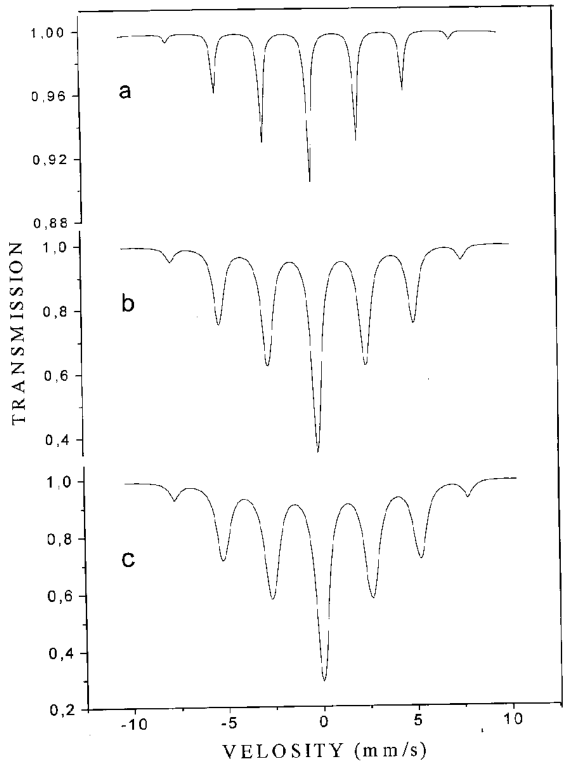


Fig. 3. Thickness dependence of the Mössbauer absorption spectrum ( $\beta = 0.1$ (*a*), $\beta = 50$ (*b*) and $\beta = 100$ (*c*))

but not by $\Omega$ as in absorption Mössbauer spectra. In other words, $\gamma$-quanta scattered to the forward direction may absorb or emit only even number of RF photons if the incident beam of radiation is perpendicular to the RF magnetic field. Note also that the Mössbauer lines of the absorption spectrum broaden due to the field reversals, but their relative intensity does not alter with the growing thickness of the absorber.


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Received 6.07.01