

MÖSSBAUER ABSORPTION BY SOFT FERROMAGNETS IN RADIO-FREQUENCY MAGNETIC FIELD

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The simultaneous influence of periodical magnetic field reversals at the nucleus between the values $\pm h_0$ and of magnetostrictive vibrations on the shape of the Mössbauer absorption spectrum is analyzed. The effect of a constant external magnetic field is taken into account by assuming unequal durations of the states $-h_0$ and $+h_0$. It is shown that such asymmetric reversals of the magnetic field lead to splitting of the absorption lines into Zeeman patterns corresponding to the time-averaged magnetic field $h_0 R$, where R is the asymmetry parameter of the reversals. The calculations agree well with experiment.

1. INTRODUCTION

As is well known (see the surveys in [1–4]), an external radio-frequency (RF) magnetic field generates magnetostrictive vibrations in ferromagnets. The corresponding Mössbauer absorption spectrum for hard ferromagnets [1–12] consists of a central Zeeman pattern and additional lines (sidebands) shifted by $n\Omega$, where n is an integer and $\Omega = 2\pi\nu$ is the circular frequency of the alternating magnetic field. The corresponding theory is the standard one for Mössbauer absorption in a vibrating crystal [11, 15]. In the case of soft ferromagnets with low anisotropy fields H_a , the absorption spectrum is a fringe of equidistant lines (doublets if the quadrupole interaction is important, i.e., $Q \neq 0$). It collapses to single or double lines when the frequency Ω greatly exceeds the Larmor frequency Ω_L . Such RF collapse has been qualitatively explained by Pfeiffer [1, 16], who assumed that the crystal magnetization $\mathbf{M}(t)$, induced by the RF magnetic field and the magnetic field $\mathbf{h}(t)$ at the nucleus, related to \mathbf{M} , periodically reverse direction, so that at high frequencies the nucleus feels only a zero average magnetic field and the absorption spectrum degenerates to a single line. In the opposite case of vanishing Ω the spectrum [1–4] coincides with a typical spectrum for constant magnetic field.

Two main model approaches to this problem are known: the harmonic model of Olariu et al. [17], in which $\mathbf{h}(t) = h_0 \cos \Omega t$ is assumed, and second, the coherent stepwise model [18, 19], assuming that $\mathbf{h}(t)$ performs periodical instantaneous jumps between the values $+h_0$ and $-h_0$. General formulae for both the absorption and scattering spectra, taking into account time-dependent effects, were derived in [20]. In particular, it was shown that the harmonic model of Olariu [17] leads to incorrect result in the stationary limit, when $\Omega \rightarrow 0$.

The stepwise model only qualitatively reproduces the observations, yielding collapse at high frequencies and a stationary Zeeman spectrum at vanishing frequencies. In the intermediate case $\Omega \sim \Omega_L$, this model predicts a much more rapid drop in satellite intensities with the order

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n than is found experimentally [1, 16]. But experimental data for Permalloy with damped magnetostrictive vibrations are well described by the stepwise model [21]. So it is natural to attribute the great discrepancy between calculations and experimental data to the contribution of magnetostrictive vibrations. An attempt to take into account simultaneously the effect of both the magnetic field reversals and magnetostrictive vibrations has been described in [19]. Unfortunately, the final result was stated without any derivation, and as will be shown below, it contains some mistakes.

Hence one goal of this paper is to build a theory simultaneously treating both reversals of the magnetic field and magnetostrictive vibrations. Another goal is to analyze the effect of a superimposed constant magnetic field \mathbf{H}_0 on the Mössbauer absorption in an RF magnetic field.

There are clear indications [22, 23], that soft ferromagnets represent cluster structure. Every such cluster behaves as a superparamagnetic particle, within which all the spins are strongly coupled. Meanwhile, the intercluster interaction is weak. The main role of an external RF field is probably to destroy this intercluster cooperation [19]. The magnetization \mathbf{M}_c of the superparamagnetic cluster may be oriented along the easy magnetization axis or in the opposite direction. Consequently, its potential energy W has the form of two potential wells separated by a potential barrier [24]. The minima of these potential wells correspond to the two values of the magnetization, \mathbf{M}_c and $-\mathbf{M}_c$. If the easy magnetization axis is parallel to the external magnetic field $\mathbf{H}_{RF}(t)$, then the additional potential energy of the cluster will be $V(t) = -\mathbf{M}_c \mathbf{H}_{RF}(t)$. Hence the complete potential energy $W + V(t)$ will be an asymmetric time-dependent curve. In a strong field $\mathbf{H}_{RF}(t)$ one of the potential wells vanishes at some point, which forces \mathbf{M}_c to jump into the opposite potential well. Another jump occurs in the backward direction at time $T/2$.

When we superimpose a constant magnetic field \mathbf{H}_0 parallel to $\mathbf{H}_{RF}(t)$, the superparamagnetic cluster receives an additional contribution to the potential energy, $M_c H_0$ or $-M_c H_0$. The static field \mathbf{H}_0 itself without $\mathbf{H}_{RF}(t)$ ensures the asymmetry of the potential curve, causing the potential well corresponding to \mathbf{M}_c oriented along \mathbf{H}_0 to be deeper. The switched on field $\mathbf{H}_{RF}(t)$, being larger than \mathbf{H}_0 , will again produce jumps of the magnetization, but in this case the time T_1 spent by the cluster in the potential well with \mathbf{M}_c parallel to \mathbf{H}_0 will be greater than the time T_2 spent in that with \mathbf{M}_c antiparallel to \mathbf{H}_0 .

We do not touch on any stochastic problems here (see also [19]) and suppose the crystal magnetization to be completely governed by the external magnetic field, i.e., we deal only with coherent reversals of the magnetization at definite times, which imply corresponding coherent reversals of the magnetic field at the nucleus.

2. WAVE FUNCTIONS

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

$$\mathbf{h}(t) = \mathbf{h}_0 f(t), \quad f(t) = f(t + T), \quad (1)$$

where T is the period of the RF field and $\Omega = 2\pi/T$ is the circular frequency. We suppose that the reversals of the magnetic field occur as abrupt jumps from $+\mathbf{h}_0$ to $-\mathbf{h}_0$ and vice versa. Let one of such jumps from $+\mathbf{h}_0$ to $-\mathbf{h}_0$ be at $t = 0$. If we have $\mathbf{H}_0 \neq 0$, then the time T_1 during which $\mathbf{h}(t) = \mathbf{h}_0$ holds is greater than the time T_2 when $\mathbf{h}(t) = -\mathbf{h}_0$. Then

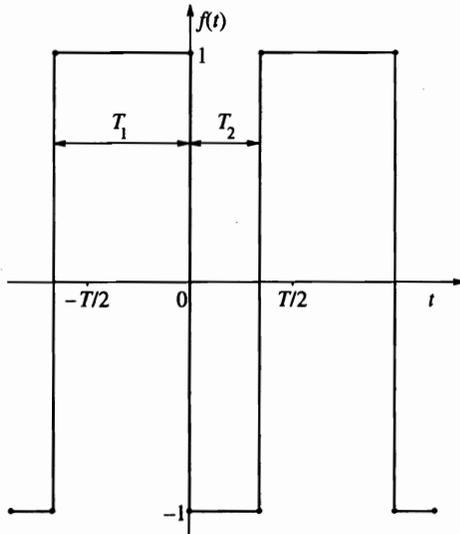


Fig. 1. Time-dependence of the magnetic field at the nucleus

$$f(t) = \begin{cases} 1, & -T_1 < t < 0 \text{ or } T_2 < t < T_1 + T_2, \\ -1, & 0 < t < T_2, \end{cases} \quad (2)$$

as shown in Fig. 1.

In order to describe such asymmetric reversals, we introduce the dimensionless parameter

$$R = \frac{T_1 - T_2}{T_1 + T_2}, \quad (3)$$

which varies in the interval $0 \leq R \leq 1$. The value $R = 0$ corresponds to symmetric reversals at equal times $T/2$, and $R = 1$ to the constant field $+\mathbf{h}_0$ during the whole time. From (3) it follows that

$$T_1 = \frac{1 + R}{2}T, \quad T_2 = \frac{1 - R}{2}T. \quad (4)$$

The Floquet wave function of the nucleus in periodical field (1), (2) may be written as

$$\Psi_{I_\kappa M_\kappa}^N(t) = |I_\kappa M_\kappa\rangle \Phi_{I_\kappa M_\kappa}^N(t) e^{-i\mathcal{E}_{M_\kappa}^N t/\hbar}, \quad (5)$$

where $|I_\kappa M_\kappa\rangle$ is the stationary wave function of the nucleus in the κ -th state ($\kappa = g$ for the ground state and $\kappa = e$ for the excited one) with spin I_κ and projection M_κ in the direction of \mathbf{h}_0 ; the periodic function of time $\Phi^N(t)$ may be defined in the interval from $-T/2$ to $T/2$ as

$$\Phi_{I_\kappa M_\kappa}^N(t) = \begin{cases} \exp[i\gamma_\kappa M_\kappa h_0(1 - R)t], & -T/2 \leq t \leq 0, \\ \exp[-i\gamma_\kappa M_\kappa h_0(1 + R)t], & 0 \leq t \leq T_2, \\ \exp[i\gamma_\kappa M_\kappa h_0(1 - R)(t - T)], & T_2 \leq t \leq T/2. \end{cases} \quad (6)$$

The corresponding quasi-energies are

$$\mathcal{E}_{M_\kappa}^N = E_\kappa^N - \gamma_\kappa M_\kappa h_0 R + Q [3M_\kappa^2 - I_\kappa(I_\kappa + 1)], \quad (7)$$

where $E_g^N = 0$ and $E_e^N = E_0'$ is the energy of the unsplit resonant level, γ_κ specifies the gyromagnetic ratio, and Q is the quadrupole constant. The complete set of quasi-energies is obtained by adding $n\hbar\Omega$ to (7), where n is an integer; the corresponding functions are given by (6) multiplied by $\exp(in\Omega t)$.

The RF magnetic field also induces magnetostrictive vibrations in a ferromagnetic crystal. If the crystal magnetization induced by the RF field coherently changes direction at definite moments of time, then the magnetostrictive vibrations occur with frequency twice as large as Ω [17, 19]. Strictly speaking, the wave function of such a lattice in the harmonic approximation is represented by a product of wave functions to describe the quantum oscillations of uncoupled oscillators about their instantaneous equilibrium positions, which perform classical vibrations [14, 15]. The displacement of Mössbauer atom owing to such classical vibrations is

$$X(t) = A \cos(2\Omega t + \varphi_0), \quad (8)$$

where A and φ_0 are the amplitude and initial phase of these magnetostrictive vibrations, respectively.

3. MÖSSBAUER ABSORPTION

A simple generalization of the method developed in [14, 15, 20] gives the following result for the phononless absorption cross-section of γ -quantum with energy E by Mössbauer nucleus:

$$\sigma_a(E) = \frac{\sigma_0 \Gamma^2}{2} e^{-2W_a} \sum_{n=-\infty}^{\infty} \sum_{M_e, M_g} \frac{J_{eg}(\vartheta) |a_{eg}(n)|^2}{(E - \mathcal{E}_e^N + \mathcal{E}_g^N - n\hbar\Omega)^2 + (\Gamma/2)^2}, \quad (9)$$

where σ_0 is the resonant cross-section, Γ is the width of the resonant level, e^{2W} is the Debye-Waller factor, and the functions $J_{eg}(\vartheta)$ determine the relative intensities of the lines versus the angle ϑ between the wave vector \mathbf{k} of the γ -quantum and \mathbf{h}_0 :

$$\begin{aligned} \frac{1}{3} J_{\pm 3/2, \pm 1/2}(\vartheta) &= J_{\pm 1/2, \pm 1/2}(\vartheta) = \frac{1}{16} (1 + \cos^2 \vartheta), \\ J_{\pm 1/2, \mp 1/2}(\vartheta) &= \frac{1}{4} \sin^2 \vartheta. \end{aligned} \quad (10)$$

The quantities $a_{eg}(n)$ stand for the following Fourier coefficients:

$$a_{eg}(n) = \frac{1}{T} \int_{-T/2}^{T/2} dt e^{-in\Omega t} \Phi_e^N(t)^* \Phi_g^N(t) \exp[ikA \cos(2\Omega t + \varphi_0)]. \quad (11)$$

The cross-section (9) must be averaged over the phononless energy distribution of incident γ -quanta

$$w_e(E) = e^{-2W_e} \frac{\Gamma/2\pi}{(E - E_0 - s)^2 + (\Gamma/2)^2}, \quad (12)$$

where $s = (v/c)E_0$ is the Doppler shift and v is the velocity of the emitter relative to the absorber. This averaging gives for ^{57}Fe

$$\sigma_a(s) = \frac{\sigma_0 \Gamma^2}{2} e^{-2W_c - 2W_a} \sum_{n=-\infty}^{\infty} \sum_{M_e, M_g} \frac{J_{eg}(\vartheta) |a_{eg}(n)|^2}{\{s - \Delta - \hbar\alpha_{eg}R - Q[3M_e^2 - 15/4] - n\hbar\Omega\}^2 + \Gamma^2}, \tag{13}$$

where $\Delta = E'_0 - E_0$ is an isomer shift; the quantities

$$\hbar\alpha_{eg} = (\gamma_g M_g - \gamma_e M_e) \hbar_0 \tag{14}$$

determine the Zeeman splitting in the constant field \mathbf{h}_0 . Furthermore, $\hbar\alpha_{eg}$ multiplied by R determines the magnetic hyperfine structure provided by the time-averaged field $\langle \mathbf{h}(t) \rangle = \mathbf{h}_0 R$.

In order to calculate $a_{eg}(n)$ we use the familiar expansion

$$e^{ix \cos t} = \sum_{n=-\infty}^{\infty} i^n J_n(x) e^{int}, \tag{15}$$

where $J_n(x)$ is the Bessel function of order n . Then

$$a_{eg}(n) = \sum_{m=-\infty}^{\infty} i^m e^{im\varphi_0} J_m(\mathbf{kA}) b_{eg}(n - 2m), \tag{16}$$

where the coefficients $b_{eg}(n)$, given by (11) with $\mathbf{A} = 0$, may be written as

$$b_{eg}(n) = b_{eg}^{(+)}(n) + b_{eg}^{(-)}(n), \tag{17}$$

$$b_{eg}^{(+)}(n) = \frac{1}{T} \int_0^{(1-R)T/2} dt \exp[-in\Omega t - i\alpha_{eg}(1+R)t],$$

$$b_{eg}^{(-)}(n) = \frac{1}{T} \int_{-(1+R)T/2}^0 dt \exp[-in\Omega t + i\alpha_{eg}(1-R)t].$$

A simple calculation yields

$$b_{eg}(n) = \frac{2x_{eg}}{[(1-R)x_{eg} - n\pi][(1+R)x_{eg} + n\pi]} \times \times \sin \left\{ \frac{1+R}{2} [(1-R)x_{eg} - n\pi] \right\} \exp \left\{ i \frac{1+R}{2} [(1-R)x_{eg} - n\pi] \right\}, \tag{18}$$

where we have used the notation

$$x_{eg} = \frac{\alpha_{eg} T}{2}. \tag{19}$$

The forced vibrations are important only for $\mathbf{kA} \neq 0$, i.e., when \mathbf{A} is not perpendicular to \mathbf{k} . In all the experiments the beam of incident Mössbauer radiation is perpendicular to the absorber surface and the external alternating magnetic field $\mathbf{H}_{RF}(t)$ is parallel to it. Therefore the original magnetostrictive vibrations, being generated along $\mathbf{H}_{RF}(t)$, are perpendicular to \mathbf{k} . As pointed out in [16], these vibrations are scattered by defects of the crystal, giving rise

to vibrations along \mathbf{k} , which manifest themselves in experiment. Then such vibrations have random phases φ_0 . Averaging $|a_{eg}(n)|^2$ over φ_0 one gets

$$\overline{|a_{eg}(n)|^2} = \sum_{m=-\infty}^{\infty} J_m^2(kx_0) |b_{eg}(n-2m)|^2, \quad (20)$$

where $x_0 = \mathbf{kA}/k$ is the amplitude of vibrations along \mathbf{k} . Besides, we must average (20) over the distribution of the amplitudes x_0 . We shall take the Rayleigh distribution, which describes Mössbauer data most accurately:

$$P(x_0) = \frac{x_0^2}{\bar{x}_0^2} \exp\left\{-\frac{x_0^2}{2\bar{x}_0^2}\right\}, \quad (21)$$

where \bar{x}_0 corresponds to the maximum of the distribution. Then the average cross-section may be written as

$$\sigma_t(s) = \sum_{n=-\infty}^{\infty} e^{-m^2} I_n(m^2) \sigma_t(s - n2\hbar\Omega)|_{\bar{x}_0=0}, \quad (22)$$

where $I_n(x)$ is the modified Bessel function, $m = k\bar{x}_0$ represents the so-called modulation index, and the cross-section $\sigma_t(s)|_{\bar{x}_0=0}$ for the case without vibrations is given by

$$\sigma_t(s)|_{\bar{x}_0=0} = \frac{\sigma_0 \Gamma^2}{2} e^{-2W_e - 2W_a} \sum_{k=-\infty}^{\infty} \sum_{M_e, M_g} \frac{J_{eg}(\vartheta) |b_{eg}(k)|^2}{[s - \Delta - \hbar\alpha_{eg}R - Q(3M_e^2 - 15/4) - k\hbar\Omega]^2 + \Gamma^2}. \quad (23)$$

Previous calculations [19] for $R = 0$ were based on an equation similar to (22), which contained the incorrect factor $I_{2n}(m^2)$ instead of $I_n(m^2)$. Therefore we first analyzed the case of symmetric reversals with $R = 0$. Using Eqs. (22) and (23) we found the best fit to Pfeiffer's data [1, 16] using the modulation index $m = 0.05$ for $\nu = 106$ MHz, $m = 0.6$ for $\nu = 61$ MHz, $m = 2.4$ for $\nu = 39$ MHz, and $m = 4$ for $\nu = 31$ MHz. As can be seen from Fig. 2, the agreement of these calculations with experiment is good. However, Pfeiffer [1, 16] proposed the following frequency dependence of the modulation index:

$$m^2 \propto 1/\nu^{3.5}. \quad (24)$$

Calculations employing this law are shown in Fig. 2c. In this case the fitting parameters are $m = 0.28$ for $\nu = 106$ MHz, $m = 0.74$ for $\nu = 61$ MHz, $m = 1.6$ for $\nu = 39$ MHz, and $m = 2.4$ for $\nu = 31$ MHz. Here the agreement with experiment is worse, so that the law (24) is doubtful.

4. LIMITING CASES

In the high-frequency case

$$\lim_{\Omega \rightarrow \infty} b_{eg}(n) = \delta_{n0}. \quad (25)$$

The average amplitude \bar{x}_0 of magnetostrictive vibrations decreases with growing frequency Ω . For $\Omega \gg |\alpha_{eg}|$ one therefore has

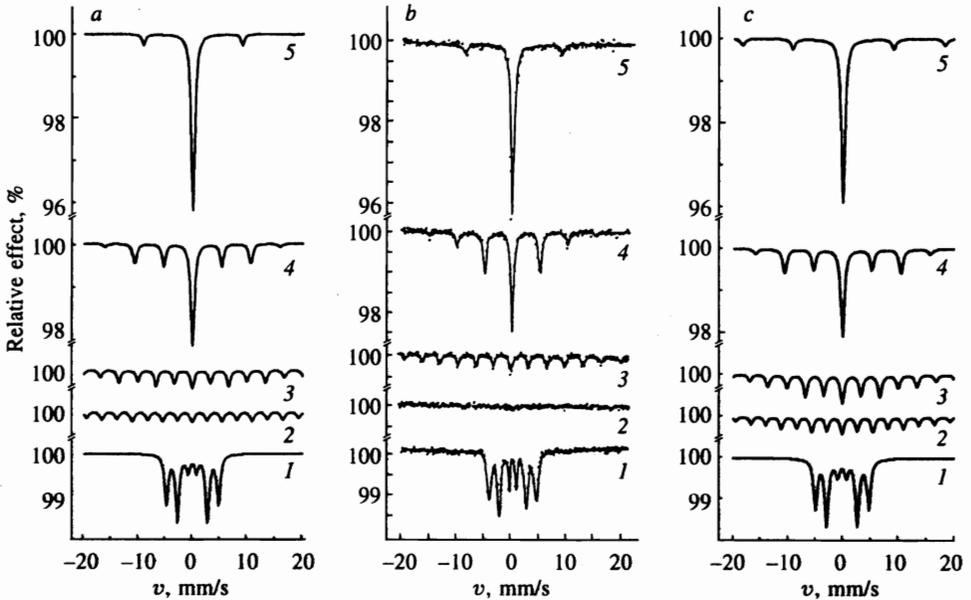


Fig. 2. Simultaneous effect of magnetic field reversals and magnetostrictive vibrations on the shape of Mössbauer spectra: a) our calculations without any constraints to the modulation index; b) Pfeiffer's data [16]; c) our calculations using the frequency dependence (24) of the modulation index. Curves 1, 2, 3, 4, and 5 correspond to $\gamma = 0, 32, 39, 61,$ and 106 MHz

$$|a_{eg}(n)|^2 \simeq \delta_{n0}. \tag{26}$$

Thus, at such high frequencies the absorption cross-section becomes

$$\sigma_t(s)_{av} = \frac{\sigma_0 \Gamma^2}{2} e^{-2W_e - 2W_a} \sum_{M_e, M_g} \frac{J_{eg}(\vartheta)}{[s - \Delta - \hbar \alpha_{eg} R - Q(3M_e^2 - 15/4)]^2 + \Gamma^2}. \tag{27}$$

Hence we can state that the hyperfine structure of the Mössbauer spectrum collapses to a single line at $Q = 0$ or a quadrupole doublet for $Q \neq 0$ only in the case of symmetric reversals with $R = 0$. When the magnetic field reverses asymmetrically with $R > 0$, the nucleus feels the high-frequency reversing magnetic field as a stationary one with magnitude $\langle h(t) \rangle$ smaller than h_0 by the factor R . Then the spectrum retains its hyperfine structure corresponding to the field $\langle h(t) \rangle$.

In the opposite low-frequency case, $(1 - R)T \rightarrow \infty$, using the definition of the ζ -function [25] one finds

$$b_{eg}^{(\pm)} \approx \pm \frac{1}{iT} [n\Omega \pm (1 \pm R)\alpha_{eg} \mp i\eta]^{-1}, \tag{28}$$

where $\eta \rightarrow +0$. Putting $n\Omega = \omega_n$ and $\Omega = \Delta\omega$, one can treat the sums over n as integrals,

$$\frac{1}{T} \sum_{n=-\infty}^{\infty} \rightarrow \frac{1}{2\pi} \int_{-\infty}^{\infty} d\omega, \tag{29}$$

in the limit $T \rightarrow \infty$. Substitution of (28) and (29) into (13) gives after a contour integration the six-line pattern produced by the constant magnetic field h_0 (see the curves 1 in Fig. 2).

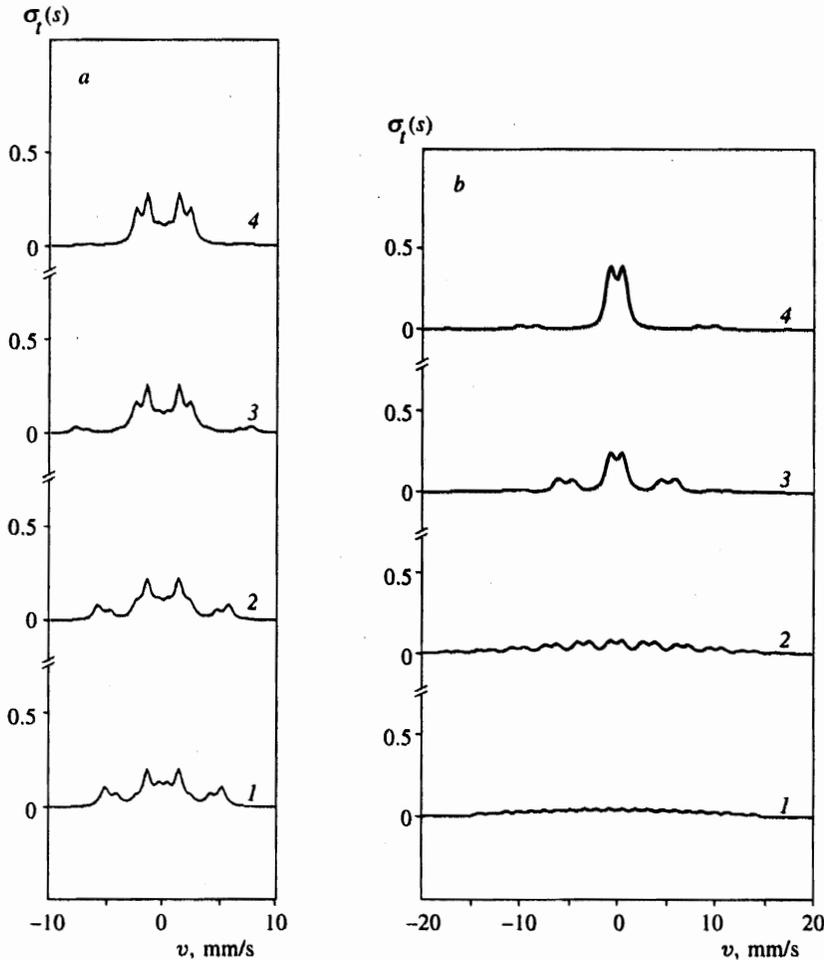


Fig. 3. Hyperfine structure of Mössbauer absorption lines due to the asymmetry of the magnetic field reversals: *a*) vibrations are absent ($Q = m = 0$) and $R = 0.5$; *b*) the modulation indices are the same as in Fig. 2*a* and $R = 0.2$; $\sigma_t(s)$ is in units $(\sigma_0 \Gamma^2 / 2) \exp(-2W_e - 2W_a)$; curves 1, 2, 3, and 4 correspond to $\nu = 32, 39, 62,$ and 106 MHz

5. DISCUSSION

An external RF magnetic field influences the nucleus in a soft ferromagnet via the reversing magnetic field $h(t)$ at the nucleus and magnetostrictive vibrations. For the constant magnetic field $H_0 = 0$ the nucleus has infinite sets of quasi-energetic levels separated by the interval $\hbar\Omega$ both in the ground and excited states. In the absence of quadrupole interaction ($Q = 0$) each such level is degenerated with respect to the magnetic quantum number M_κ . Such quasi-energetic structure produces transitions of the nucleus, absorbing γ -quanta, from the ground state to any quasi-energetic level corresponding to the excited nuclear state. As a consequence, the Mössbauer spectrum will consist of a set of equidistant lines (doublets if $Q = 0$). There n -th line (doublet) is associated with the transitions from the ground state with quasi-energy $\mathcal{E}_{M_g}^N$ to excited states with quasi-energies $\mathcal{E}_{M_e}^N + n\hbar\Omega$, which are specified by the quantum

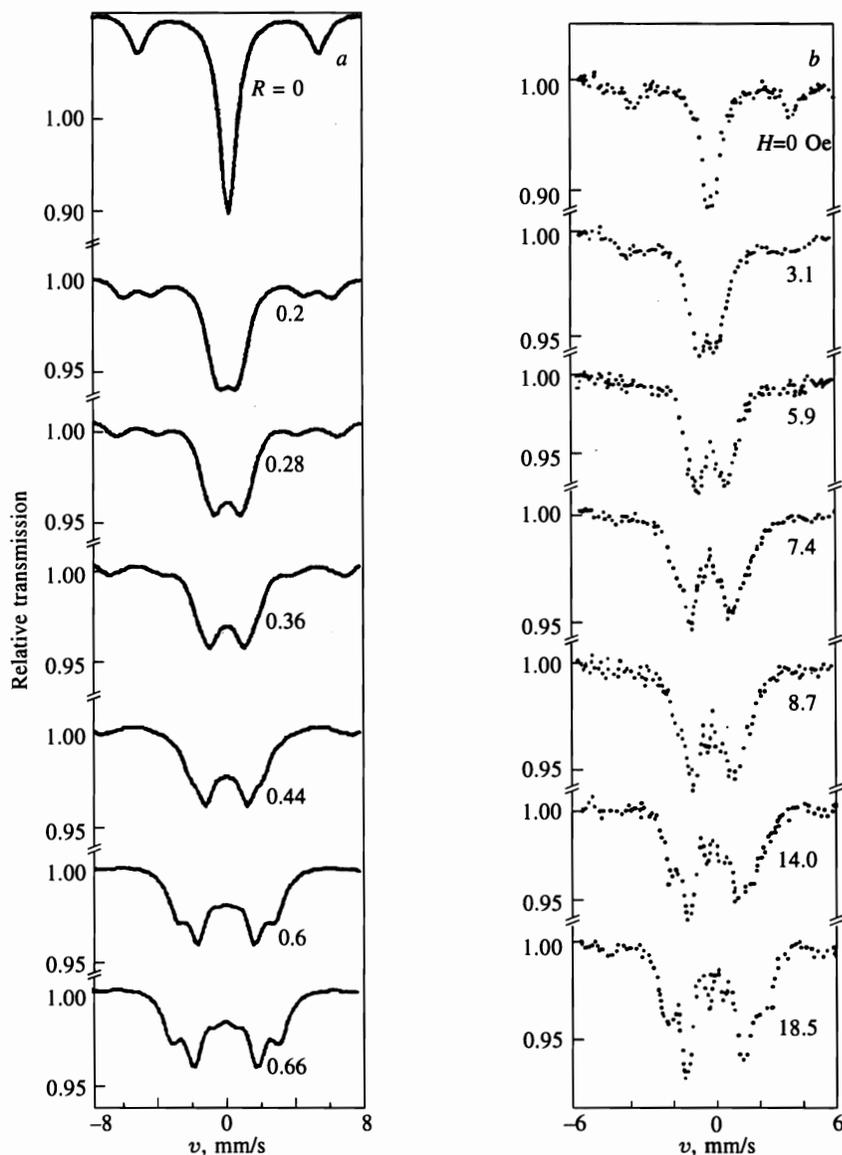


Fig. 4. Splitting of the collapsed line due to a superimposed static magnetic field: *a*) our calculations; *b*) experimental data [26]

number M_e . In high- and low-frequency cases such spectra approach single or double lines and a standard sextet, respectively.

Our calculations well agree with Pfeiffer's data on Permalloy [16] (see Fig. 2) supporting the point of view that the RF magnetic field causes both reversals of the magnetization and magnetostrictive vibrations. Note that Julian and Daniels [19], who have been using the incorrect formula, predicted unreasonably high modulation indices fitting the same data: for 32 MHz they used $m = 100$.

When the constant magnetic field is superimposed, the quasi-energetic sublevels split as indicated by Eq. (7). It is of interest that such split quasi-energies coincide with the energies of the nucleus placed in the constant magnetic field \mathbf{h}_0R . Thus, the quantum system not only feels the oscillations of the magnetic field exchanging photons with frequency Ω , but also sees its time-averaged value $\langle \mathbf{h}(t) \rangle = \mathbf{h}_0R$, where the asymmetry parameter R depends on the magnitude of the external field H_0 . The splitting of quasi-energies leads to a corresponding splitting of the Mössbauer lines. Such a splitting arises at all frequencies and is typical both for the central line and for sidebands. This effect is illustrated by Fig. 3a, showing $\sigma_t(s)$ in units $(\sigma_0\Gamma^2/2)\exp(-2W_e - 2W_a)$ for $R = 0.5$ and $Q = m = 0$. The changes owing to the magnetostrictive vibrations are shown in Fig. 3b, where we used the same modulation indices as in Fig. 2a and took $R = 0.2$. Our calculations with $Q = m = 0$ are compared with data [26] obtained at $\nu = 62$ MHz with different gradually increasing values of H_0 for amorphous $\text{Fe}_{78}\text{Si}_9\text{B}_{13}$ alloy. In Fig. 4 we can see satisfactory agreement with these observations when the line splits under the influence of the static magnetic field. From top to bottom the fitting parameters are $R = 0, 0.2, 0.28, 0.36, 0.44, 0.6, 0.66$. In Figs. 2 and 3 we used $\Gamma = 0.4$ mm/s, and in Fig. 4 $\Gamma = 0.6$ mm/s.

It is of great interest to observe such a splitting, caused by an external constant magnetic field H_0 , not only for the collapsed line but also for the sidebands existing at intermediate frequencies. It would be the most direct evidence for the nuclear quasi-energetic picture (7) and respectively to the cluster structure of soft ferromagnets.

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