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Gamma-resonance solid-state spectroscopy under high-frequency excitation conditions

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Experimental and theoretical studies of high-frequency excitations in solids by γ-resonance spectroscopy are reviewed. Both the resonance-type and non-resonance-type alterations in Mössbauer spectra that are produced by high-frequency ac fields are elucidated. A considerable part of the review is devoted to problems of modulation of γ radiation by ultrasonic vibrations. Various mechanisms of exciting sound by a radiofrequency field are studied, in particular, magnetostriction in ferromagnets. A new effect found by γ-resonance spectroscopy is discussed: collapse of the hyperfine structure upon fast remagnetization of a ferromagnet. A number of new nonlinear resonance phenomena are discussed that amount to combining the Mössbauer effect with the methods of radiospectroscopy (NMR, EPR, etc.). The possibility is discussed of applying these phenomena for studying the properties of solids.

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CONTENTS

1. Introduction 741
2. The Ultrasonic Doppler Effect 742
3. Magnetostriction Ultrasonic Vibrations in Ferromagnets 746
4. Remagnetization of Ferromagnets by Radiofrequency Fields. Collapse of Hyperfine Structure 749
5. γ-Magnetic and γ-Acoustic Resonances 751
6. Mössbauer Spectra in Electron Paramagnetic Resonance and in Optical Excitations 754
7. Conclusion 755
Bibliography 756

1. INTRODUCTION

Application of the Mössbauer effect in various fields of solid-state physics and chemistry has facilitated the rapid growth of γ-resonance spectroscopy (GRS). It has become most fruitful to use GRS to study the hyperfine interaction of Mössbauer nuclei in crystals. Owing to the high energy resolution of GRS, these investigations have become an effective way of studying solids. Methods have become widespread of studying the structures and properties of solids by applying static external perturbations, as well as of studying the relaxation processes that arise from the motion of nuclei, spins, or fast chemical reactions.

It has recently become possible to control dynamic

processes in matter by using external high-frequency fields. Consequently, new possibilities of applying GRS have arisen. We note in this regard that the characteristic lifetimes of the excited states of Mössbauer nuclei amount to 10^{-6}–10^{-9} sec. Hence the pertinent frequencies are comparable with or greater than 10^6–10^9 Hz. The dynamic processes engendered by high-frequency fields in this frequency range encompass the ranges of nuclear magnetic, quadrupole, electron paramagnetic, and optical resonances, as well as the ranges of different types of elementary excitations in solids: phonons, magnons, plasmons, etc. These processes cause quantum transitions between the energy levels of the nucleus and of the electron cloud of the ion, they alter the electric and the magnetic fields at the nucleus, and they give rise to crystal-lattice vibrations. In turn, the changes in the state of the Mössbauer nucleus are reflected in the type of interaction of the latter with resonance γ radiation that gives rise to corresponding distortions in the Mössbauer spectrum.

An example of use of GRS under high-frequency excitation conditions that is simple, yet broad in its applications, is the method of ultrasonic modulation of γ radiation. As studies have shown, the intensities and positions of the new spectral lines that arise here are sensitive to the conditions of generation and propagation of the sound vibrations in the specimen.

These properties have made possible a series of studies on the mechanisms of excitation of sound in solids. The most fruitful studies here have proved to be those on the effect of magnetostriction in ferromagnets.

Increase in the power and the frequency of the radiofrequency field has facilitated observation of a new GRS effect in ferromagnets, viz., radiofrequency collapse of the hyperfine structure. This effect is due to remagnetization of the specimen at a frequency exceeding many-fold the nuclear Larmor frequency. It is essentially a coherent analog of the collapse of hyperfine structure involving fast electronic relaxation.

A quite different character is shown by the excitation of nuclear magnetic and electron paramagnetic resonances (NMR and EPR) and by optical resonances that excite quantum transitions between the energy sublevels of a Mössbauer ion. The effect of the latter on the Mössbauer spectrum is more complicated and varied than in the previous phenomena. Yet these studies are mainly theoretical in nature. An exception is the phenomenon of γ-magnetic resonance (NMR + the Mössbauer effect), the study of which has entered an experimental phase.

A number of interesting theoretical studies have recently been made on applying a resonance radiofrequency field for studying the inhomogeneous and dipole-dipole widths of Mössbauer lines of long-lived isotopes. Apparently the experimental realization of these ideas will be of great importance in solving the problem of creating a source of stimulated γ radiation.

Thus we can assuredly say that the study of high-frequency excitations considerably expands the field of application of GRS, while furnishing in a number of cases information that is extremely hard to get by other methods. The set of problems that can be solved by GRS under high-frequency-excitation conditions is very broad and topical. Several review articles have recently appeared on individual problems of solid-state GRS under high-frequency-excitation conditions (see[1–6]). However, there has been as yet no presentation of the problem as a whole. We shall try in this paper to treat these problems as fully as possible.

2. THE ULTRASONIC DOPPLER EFFECT

A. Ultrasonic modulation of γ-radiation

Even in his first experiments, R. Mössbauer used the linear Doppler effect for displacing the frequency of the γ radiation. In this effect, the γ radiation of frequency ω_γ from a source moving at the velocity $\mathbf{v}$ with respect to the laboratory system of coordinates will have the following frequency in the latter:

$$\omega'_\gamma = \omega_\gamma \left[1 + \frac{(\mathbf{vn})}{c}\right]. \tag{2.1}$$

Here $\mathbf{n}$ is a unit vector in the direction of propagation of the γ radiation, and c is the velocity of light.

If the velocity of the source varies periodically, the observer will detect frequency-modulated γ radiation. Ruby and Bolef[7] first performed such experiments in the United States in 1960 on ^{57}Fe nuclei by exciting ultrasonic vibrations in the source or the absorber. The experiment was repeated a little later on ^{119}Sn nuclei.[8] Let us take up the classical theory of ultrasonic modulation of γ radiation.[9] If we assume that the specimen vibrates as a whole ($qL \ll 1$, where L is the size of the specimen, and $q = \Omega/v_{\text{sound}}$ is the wavenumber of the sound wave, Ω and v_{sound} being the frequency and the velocity of sound in the specimen), we can write an expression for the electric field strength of the γ-wave in the presence of the ultrasonic vibrations:

$$\mathbf{E} = \mathbf{E}_0 \exp\left\{i\left[\omega_\gamma t + \frac{\mathbf{an}}{\lambda}\cos(\Omega t + \Phi)\right]\right\}. \tag{2.2}$$

Here $\mathbf{E}_0$ is the amplitude of the electric field strength in the absence of the ultrasonic vibrations, $\mathbf{a}$ and Φ are the amplitude and phase of the ultrasonic vibrations, and $\lambda = 2\pi\lambda\!\!\!^{-}$ is the wavelength of the γ radiation. Upon using the expansion

$$e^{iz\cos\theta} = \sum_{n=-\infty}^{\infty} J_n(z)\, e^{in[\theta-(\pi/2)]}, \tag{2.3}$$

where $J_n(z)$ is a Bessel function of order n, we can reduce (2.2) to the following form:

$$\mathbf{E} = \mathbf{E}_0 e^{i\omega_\gamma t} \sum_{n=-\infty}^{\infty} J_n\left(\frac{x_0}{\lambda}\right) e^{in[\Omega t+\Phi-(\pi/2)]}, \tag{2.4}$$

where $x_0 = \mathbf{a}\cdot\mathbf{n}$.

If we further consider the intensity of the γ radiation to be proportional to $|\mathbf{E}|^2$, we find with the aid of (2.4) that the spectrum consists of symmetrically arranged lines ($\omega_\gamma^{\text{res}} = \omega_0 + n\Omega$, with $n = 0, \pm 1, \pm 2, \ldots$) having in-

tensities determined by $J_n^2(x_0/\lambda\!\!\!^{-})$ (see also[10]).[1)]

One can also calculate the ultrasonic modulation of γ radiation by the quantum method by introducing the coherent quantum states of the ultrasonic field.[1,4] However, the specifics of observation of the Mössbauer effect most often leads to the situation that the structural inhomogeneities inside the specimen and at the boundary are comparable with the wavelength of the ultrasonic vibrations. Then the phase of the ultrasonic vibrations becomes indeterminate, and the vibrations are incoherent. One can use the formalism of thermal phonons heated to a very high temperature in order to describe the incoherent ultrasonic vibrations. Such a treatment of ultrasonic modulation of γ radiation has been carried out by Abragam,[14] who showed that here the intensity of the lines is proportional to $\exp[-x_0/\lambda\!\!\!^{-})^2] \times I_n[x_0/\lambda\!\!\!^{-})^2]$, where I_n is a modified Bessel function of order n. He also showed that this expression for the intensity of the lines can be derived from the expression for coherent ultrasound by assuming that the amplitude of the latter obeys a Rayleigh distribution[15]

$$\mathcal{P}^{\mathrm{Rayl}}(x) = \frac{1}{(x_0)^2}\, x \exp\left[-\frac{1}{2}\left(\frac{x}{x_0}\right)^2\right]. \qquad (2.5)$$

Integration of $J_n^2(x/\lambda\!\!\!^{-})$ with respect to x by using this distribution gives $\exp[-(x_0/\lambda\!\!\!^{-})^2] I_n((x_0/\lambda\!\!\!^{-})^2)$. For an arbitrary amplitude distribution of the ultrasonic vibrations, the probability of emission or absorption of a γ quantum has the form[4]

$$W'(\omega_\gamma) = \sum_{n=-\infty}^{\infty} W(\omega_\gamma - n\Omega) \int_0^\infty \mathcal{P}(x)\, J_n^2\left(\frac{x}{\lambda\!\!\!^{-}}\right) dx. \qquad (2.6)$$

Here $W(\omega_\gamma)$ is the probability of absorption or emission of a γ quantum in the absence of the ultrasonic vibrations, and $\mathcal{P}(x)$ is the amplitude distribution of the ultrasonic vibrations, which is a δ-function or a Rayleigh distribution for coherent or incoherent vibrations, respectively.

B. The coherence relaxation time of ultrasonic vibrations

Mishory and Bolef[11] have characterized the two cases of coherent and incoherent ultrasonic vibrations by using the ultrasonic relaxation time τ for phonons. This is the time that it takes for the coherent ultrasonic vibrations to relax into thermal vibrations of energy kT, where T is the temperature of the specimen. However, the only stage of the process that is important for determining the amplitudes of the ultrasonic vibrations is the one during which the coherent distribution of vibrations (see, e.g.[16]) becomes incoherent and thermal, though the temperature T' corresponding to the latter distribution can yet exceed the temperature T of the specimen. The time τ' that this stage of the process takes determines the relaxation of the coherence of the ultrasonic vibrations. The values $\tau' = \infty$ and 0 correspond to coherent and incoherent vibrations. Hence we can only crudely decide on the coherence of the sound if we know the ultrasonic relaxation time τ of the phonons.

TABLE I. Comparison[11] of the intensity ratio of satellites C_n/C_0 with $I_n((x_0/\lambda)^2)/I_0((x_0/\lambda)^2)$.

$(x_0/\lambda)^2$	C_1/C_0 (exp.)	I_1/I_0 (theory)	C_2/C_0 (exp.)	I_2/I_0 (theory)	I_3/I_0 (theory)	C_3/C_0 (exp.)
0.74	0.36	0.37				
2.98	0.80	0.81	0.46	0.46		
6.70	0.92	0.90	0.72	0.75	0.49	0.46

In most cases, experiments on ultrasonic modulation of γ radiation reveal incoherent vibrations.[1,11] Table I compares the experimental data for a ^{57}Co source in stainless steel (frequency $\Omega/2\pi = 25$ MHz) with the theory of ultrasonic modulation of γ radiation by incoherent ultrasonic vibrations. As we see from the table, the theory agrees well with experiment. The incoherence of the ultrasonic vibrations is also favored by the very short ultrasonic phonon relaxation time at room temperature $\tau \approx 10^{-11}$ sec.

Yet in a number of cases, the experimental data indicate partial coherence of the ultrasonic vibrations. Experiments on ultrasonic modulation of γ radiation[4] have shown that magnetostriction ultrasonic vibrations excited by a radiofrequency field have a distribution function that differs from both the Rayleigh function and from a δ-function. Moreover, a measurement of the relaxation time of these magnetostrictional vibrations in iron gave a value of several microseconds.[17,18] These facts apparently permit one to affirm the partial coherence of the magnetostrictional ultrasonic vibrations.

C. Relationship of the intensity of the satellites to the amplitude of the ultrasound

Let us study the behavior of the relative intensities of the lines of a Mössbauer spectrum modulated by ultrasonic vibrations as a function of the amplitude of the ultrasonic field.

The intensity of the fundamental line ($n = 0$) is unity for $x_0 = 0$. It declines monotonically with increasing amplitude (incoherent vibrations) or declines and oscillates about zero (coherent vibrations). The intensity of the lateral satellites ($n \neq 0$) is zero at first for $x_0 = 0$, and then they grow with increasing amplitude as $(x_0/\lambda\!\!\!^{-})^{2n}$, and then they monotonically decline (incoherent vibrations), or decline and oscillate about zero (coherent vibrations).

Thus we see that the effect of ultrasonic modulation will occur for amplitudes of the ultrasonic displacements comparable with the wavelength $\lambda\!\!\!^{-}$ of the γ radiation. For example, for the nuclei ^{57}Fe, ^{119}Sn, and ^{67}Zn, the values of $\lambda\!\!\!^{-}$ are 0.14 Å, 0.084 Å, and 0.022 Å, re-

[1)] We note that the positions of the satellite lines, which are governed by the simple law $\omega_\gamma^{res} = \omega_0 + n\Omega$, permit one to calibrate the Mössbauer spectrum.[11,12] This is how Cranshaw and Reivari attained high accuracy in determining the nuclear g factors in iron. We also note the possibility of using ultrasonic modulation for creating a Mössbauer spectrometer.[13] Then one chooses an amplitude of the ultrasonic vibrations for which the intensity of only the satellite with $n = 1$ will be substantial. One can analyze the Mössbauer spectrum of a studied substance by varying the frequency of the ultrasonic vibrations, and thus varying the position of the satellite line.

spectively. Such amplitude values are small, and it is hard to detect them by other methods. In line with this, one can naturally use the method of ultrasonic modulation of γ radiation to detect small ultrasonic vibrations.

With increasing amplitude of the ultrasonic vibrations, the intensity of the fundamental line is no longer sharply distinguished from the intensities of the satellites. Yet the intensities of both decline, since the overall intensity of the spectrum remains constant according to the theorem of summation of Bessel functions[19]:

$$J_0^2\left(\frac{x_0}{\lambda}\right)+2\sum_{n=1}^{\infty}J_n^2\left(\frac{x_0}{\lambda}\right)=1. \qquad (2.7)$$

This fact agrees well with experiment (Fig. 1).

The effect of declining intensity of the fundamental Mössbauer line with increasing amplitude of ultrasonic vibrations has also been observed[20] in ultrasonic modulation of γ radiation in the ferroelectric crystal $LiNbO_3$ containing 0.5 mole% ^{119}Sn. Figure 2 compares the experimental data with theory. However, the voltage limit of the transducer prevented them from determining whether the behavior of the probability of the Mössbauer effect is oscillatory, and thus to determine the degree of coherence of the ultrasonic vibrations.

D. Study of vibrations in compound acoustic resonators

As the preceding discussion has implied, the intensity of the fundamental lines of the spectrum declines with increasing amplitude of the ultrasonic vibrations. This property of the Mössbauer spectrum can be used to study the propagation of sound in compound resonators (transducer plus specimen). The experiment is set up as follows: one measures the intensity of the fundamental line of the Mössbauer spectrum of the source or absorber as a function of the ultrasonic frequency while one of them is being subjected to ultrasonic vibrations.

As an example, let us treat the features of the excitation of a standing ultrasonic wave in a compound acoustic resonator. Vibrations are established in the latter whose intensities depend strongly on the coincidence of

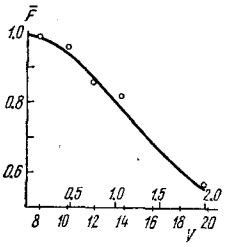

FIG. 2. Relationship of the mean probability of the Mössbauer effect (^{119}Sn in $LiNbO_3$) to the voltage applied to the ultrasonic transducer. The experimental data are compared with the theoretical curve.[20]

the vibration frequency with the mechanical-resonance frequency. Here the density of ultrasonic energy inside the specimen at mechanical resonance is larger by a factor of Q than outside it. The value of Q is as large as 10^2–10^3 under usual experimental conditions.

The positions of the mechanical-resonance frequencies of the compound acoustic resonator are determined by the thicknesses, velocities of propagation, and ultrasonic absorption values in the transducer and the specimen, respectively. These features of the propagation of sound in compound resonators have been studied in[1,21].

Bolef and his associates showed that, when the inequality $l_T \ll l_S$ is satisfied, where l_T and l_S are respectively the thicknesses of the transducer and the specimen, a compound resonator shows the usual spectrum for ultrasonic modulation of γ radiation: the mechanical-resonance frequencies are evenly spaced.

The acoustic spectrum differs in nature when $l_T \gg l_S$ (Fig. 3). In the diagram, the broad lines match the mechanical resonances of the compound acoustic resonator, and the narrow lines correspond to the mechanical resonances of the transducer. The latter are narrow in width because the absorption of ultrasonic vibrations in quartz is considerably weaker than in stainless steel.

This spectrum makes it possible to measure the phase velocity v_S in the specimen by using the formula[22]

$$v_S=(\omega^c_{m+1}-\omega^c_m)(1+2\gamma)\frac{l_S}{\pi},\quad \gamma\equiv\frac{\rho_T l_T}{\rho_S l_S}. \qquad (2.8)$$

(ω^c_m are the resonance frequencies of the compound res-

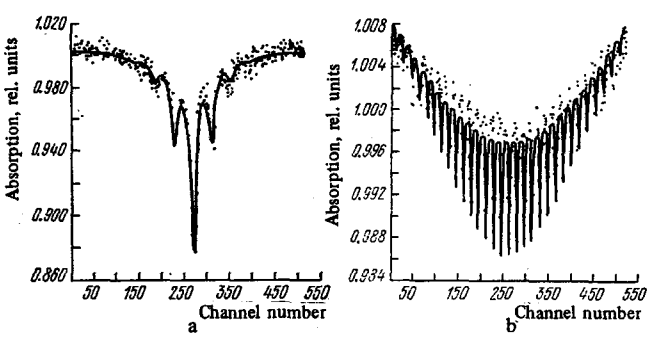

FIG. 1. Mössbauer absorption spectrum in FeAl modulated by ultrasound of frequency 16 MHz as a function of the voltage V applied to the ultrasonic transducer.[11] a—V=4 V; b—V=30 V, with velocity scale contracted by a factor of 2.5.

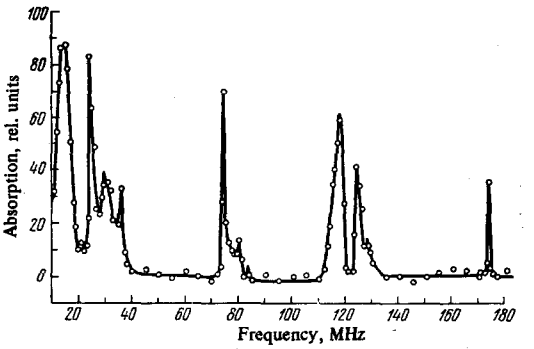

FIG. 3. The acoustic Mössbauer spectrum of an ultrasonic resonator consisting of a transducer (l_T=0.45 cm) and absorber (stainless steel, l_S=0.1 cm) at 15-MHz ultrasonic frequency.[1]

onator, and ρ_T and ρ_S are the densities of the transducer and the specimen), as well as the absorption coefficient for ultrasonic vibrations in the specimen (from the width of the lines).

E. The transverse Doppler effect

Another possible effect of ultrasonic vibrations on a Mössbauer spectrum is the transverse Doppler effect. This has been treated in[1]. Let us estimate the size of this effect. The transverse Doppler effect involves one of the conclusions of the special theory of relativity, which states that clocks run more slowly in a moving system of coordinates than in a stationary one. Then when a γ quantum of frequency ω_γ is emitted in a system of coordinates that is moving at the velocity v with respect to a stationary system, the phase of oscillation of the γ wave in the latter becomes

$$\varphi = \omega_\gamma \int_0^t \sqrt{1 - \frac{v^2}{c^2}}\, dt. \tag{2.9}$$

Let us study Eq. (2.9) for the case of ultrasonic vibrations. If we assume that v^2/c^2 is small, we can show that the frequency of the γ-radiation of the moving source will have the shift

$$\Delta\omega_\gamma = -\frac{1}{4}\frac{\Omega^2}{\omega_\gamma}\left(\frac{\bar{a}}{\lambda}\right)^2. \tag{2.10}$$

In contrast to the linear Doppler effect, the frequency shift in the transverse Doppler effect does not depend on the direction of displacement of the ultrasonic vibrations. As we see from (2.10), the most favorable conditions for observing the ultrasonic transverse Doppler effect are achieved at high energies and with a small spontaneous width Γ of the γ radiation, and high frequencies and powers of the ultrasound. Apparently, nuclei such as ^{67}Zn, ^{181}Ta, etc., are most suitable for this purpose.

Estimates for the widely used nucleus ^{57}Fe show that $\Delta\omega_\gamma/\Gamma \approx 1$ for ultrasound of frequency 10^{12} Hz and amplitude 10^{-9} cm.

Thus, the transverse effect is most accessible for application of GRS in studying ultrasonic vibrations at high frequencies closely approaching the Debye frequency.

F. The distance effect. The phase of the ultrasonic vibration

In 1961 Bömmel proposed that, according to the equivalence principle, the frequency of a source moving with an acceleration d at a distance z from the absorber should undergo a frequency shift: $\Delta\nu/\nu = zd/c^2$. The experiment was performed with a source and an absorber of γ radiation subjected to ultrasonic vibrations (with identical frequencies, amplitudes, and phases). A change in the counting rate that was linear in z was observed as the distance z was varied (up to 35 cm).

However, a distance effect may arise in the same way also from a change in the phase of the ultrasonic vibrations with distance. A theory of this phenomenon can be devised by simultaneous solution of the equation for the nuclear-density matrix in the vibrating medium and of the Maxwell equations. Consequently, the intensity of γ-radiation for a source and an absorber that vibrate along the axis Oz with frequency Ω, amplitudes a_1 and a_2, and phases Φ_1 and Φ_2, respectively, can be expressed by the following formula[24]:[2)]

$$W = \sum_{m=-\infty}^{\infty} J_m^2(R)\,\mathrm{Sp}\left\{e^{i\hat{n}^m\left(\frac{\Delta z}{\lambda}\right)}\hat{\chi}e^{-i(\hat{n}^m)+\left(\frac{\Delta z}{\lambda}\right)}\right\}; \tag{2.11}$$

Here $\hat{\chi}$ is the polarization density matrix of the source of γ-radiation,[26] the symbol + denotes the Hermitian conjugate, Δz is the thickness of the absorber, $1/\lambda$ is the wavenumber of the γ-radiation, and

$$R = 1/\lambda\sqrt{a_1^2 + a_2^2 - 2a_1a_2\cos(\Phi_2 - \Phi_1 + \Omega z/c)}\,. \tag{2.11a}$$

The tensor refractive index $n^m_{\nu\nu'}$ of the m-th harmonic of the γ radiation is expressed as follows:

$$n^m_{\nu\nu'} = -n_0 \sum_{m_e,\, m_g,\, M} \times \frac{\rho^{(0)}_{m_g}(\nu\nu')^\varepsilon D^{(L)*}_{\nu M}(\varphi,\theta,0)\, D^{(L)}_{\nu' M}(\varphi,\theta,0)\, C^2(I_g, L, I_e; m_g, M, m_e)}{i(\Gamma/2) + (\omega_\gamma + m\Omega - \omega_{m_e, m_g})},$$
$$n_0 = \pi\lambda^3 f\frac{N}{V}\frac{1+2L}{1+\alpha}. \tag{2.12}$$

Here the $D^{(L)}_{MM'}(\varphi,\theta,\psi) = e^{iM\varphi}d^{(L)}_{MM'}(\theta)e^{-iM'\psi}$ are the generalized spherical functions: the symbol * denotes the complex conjugate: the Eulerian angles φ, θ, and 0 determine the direction of the axis of quantization with respect to the laboratory system of coordinates, with the Oz axis along the wave vector of the γ wave; $\rho^{(0)}_{m_g}$ is the population of the sublevels of the ground state of the nucleus; the $C(I_gLI_e, m_gMm_e)$ are the Clebsch-Gordan coefficients for L-multipole emission; L_g, I_e and m_g, m_e are respectively the nuclear spins and magnetic quantum numbers of the ground and excited states; the parameter ε is 0 or 1 for magnetic and electric γ transitions, respectively; N/V is the number of Mössbauer nuclei per unit volume; and f is the Lamb-Mössbauer factor.

We can see from the formulas (2.11) and (2.11a) the relationship of the resonance absorption to the distance z that arises from the finite time of propagation of the signal (at the speed of light). Therefore the phase of the ultrasonic modulation at the distance z from the source is shifted by the amount $\Omega z/c$. One can show that a change over the range 0–0 cm at the frequency $\Omega/2\pi = 30$ MHz and amplitude of ultrasonic deformation $a = 10^{-8}$ cm ($a_1 = a_2 = a$, $\Phi_1 = \Phi_2$) will alter the intensity, e.g., of the fundamental line (^{57}Fe nucleus) by 20%. However, in contrast to Bömmel's experiment, the effect is quadratic in z for small z.

We can hope that extra experiments will clarify the physical pattern of this phenomenon. Let us point out now some consequences of Eq. (2.11). Thus, for a

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[2)] Recently an analogous problem has been solved on the basis of the optical theorem in the approximation of a thin absorber and small amplitudes of the ultrasonic vibrations.[25] These authors did not treat the phase of the ultrasonic vibrations nor the polarization of the γ-radiation.

stationary source or absorber,

$$W=\sum_{m=-\infty}^{\infty} J_m^2\left(\frac{a}{\lambda}\right)\mathrm{Sp}\left(e^{i(\hat{n}^m(\Delta z/\lambda))}\hat{\chi}e^{-i(\hat{n}^m(\Delta z/\lambda))}\right). \qquad (2.13)$$

Equation (2.13) supplements Eq. (2.6) for the case of polarized γ radiation with absorption of the latter in a thick absorber. Analysis of (2.13) shows that absorption of the m-th harmonic occurs analogously to the stationary absorber, but with a frequency shift of the γ radiation by $m\Omega$ and with a probability $J_m^2(a/\gamma)$ of appearance of the harmonic. Thus, passage through a vibrating thick absorber leads not only to frequency modulation, but also to amplitude modulation of the γ radiation.[27,28]

We must note that both the transverse Doppler effect and the distance effect can be useful for testing the theory of relativity in non-inertial coordinate systems, while the phase dependence of resonance absorption can be used to measure the degree of coherence of ultrasonic vibrations.

G. Decrease in the self-absorption of γ-radiation in an ultrasonic wavefield

It has turned out in the experimental study of Mössbauer spectra in an ultrasonic modulation regime that the intensity of γ emission of a thick source is increased at high powers of ultrasonic vibrations.[1] The explanation of this phenomenon proposed by the authors is that the ultrasonic vibrations shift the frequencies of the emitting Mössbauer nuclei with respect to one another in such a way that the mean frequency shift of the γ radiation is equal in order of magnitude to the spontaneous width. The self-absorption is thereby reduced.

In conclusion, we note that application of ultrasonic Mössbauer spectroscopy is especially promising at high ultrasonic frequencies, where other coherent methods of detection are not applicable. Application of this method is also highly promising for studying ultrasonic processes in solids,[3)] in particular for studying magnetostriction vibrations.

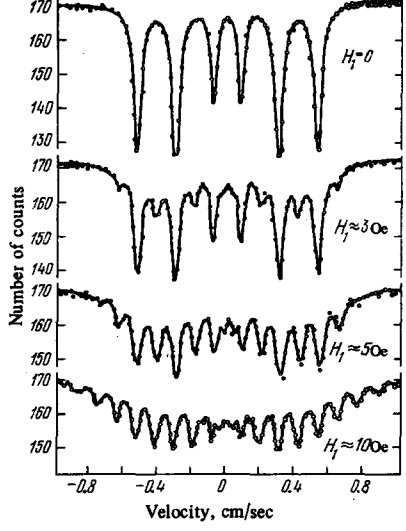

FIG. 4. The effect of an ac magnetic field of 13-MHz frequency as a function of the field intensity in the metallic absorber (iron enriched in ^{57}Fe).[30]

3. MAGNETOSTRICTION ULTRASONIC VIBRATIONS IN FERROMAGNETICS

Experimental studies of the effect of a radiofrequency (rf) field in GRS have primarily led to detecting the generation in ferromagnetics of ultrasonic vibrations. The first experiments were performed almost simultaneously in the United States[30] and in Italy.[31] These experiments showed that the action of the rf field is analogous to the ultrasonic modulation of γ radiation, and it gives rise to satellites in the Mössbauer spectrum (Fig. 4). Here the index of modulation of the latter is proportional to the amplitude of the rf field (see also[32]). They have subsequently performed a number of studies aimed at elucidating the nature of the satellites that are excited by the rf field.

A. The acoustic hypothesis of excitation of satellites

Pfeiffer[33] has performed a detailed experimental study to test the hypothesis of excitation of ultrasonic vibrations stimulated by the rf field. He studied the frequency-dependence of the index of modulation of the satellites for hematite powder (α-Fe_2O_3). Experiment showed that, first, the size of the effect increases with increasing size of the particles, and second, the effect sharply declines below a frequency of the rf field that is defined for a given size of particles. According to Pfeiffer, this critical frequency is fixed by the requirement that the acoustic half-wavelength should be shorter than the size of the particles of the powder. Otherwise, the amplitude of the acoustic wave sharply declines. Therefore, $\nu_{crit}=v/2d$, where v is the speed of sound in the specimen. The velocities v_s of shear waves and v_l of longitudinal waves differ in hematite. However, the experimental conditions are such that the lowest acoustic-mode frequency will be a mixed mode that has an effective acoustic velocity between v_s and v_l. The arrows in Fig. 5 indicate the boundaries corresponding to v_s and v_l, respectively.

A second experiment that supports the acoustic hypothesis of satellite formation was recently performed by Albanese *et al.*[34] The idea of the experiment consisted in transport of acoustic energy of a vibrating plate to the medium in which it lies. The energy density E in the plate is directly proportional to the power W generated inside it, and inversely proportional to the transport coefficient T: $E\sim W/T$; here $T=4\gamma(1+\gamma)^2$, where $\gamma=z_2/z_1$ is the ratio of the acoustic resistances ($z=\rho v$) of the two media. The coefficient depends on the elastic properties of the specimen and of the surrounding medium. For example, it is 4.3×10^{-5} for the iron-

[3)] Observation of asymmetry of quadrupole splitting induced by ultrasound in sodium nitroprusside and in siderite has been quite recently reported.[29]

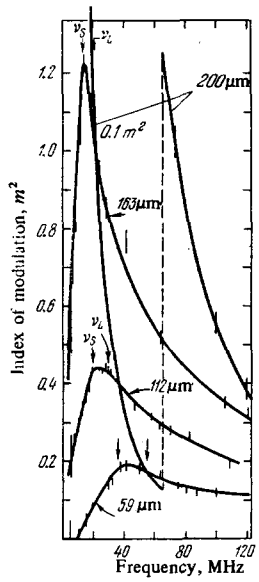

FIG. 5. Index of modulation of the satellites as a function of the frequency and of the size of the particles.[33]

air system, and 1.3×10^{-1} for the iron-oil system.

The experiment was performed with an absorber in the form of an iron foil under ordinary conditions (in air), and also with the surface of the foil covered with silicone grease. The satellite-excitation effect was absent in the latter case. The same effect of abolishing the satellites was observed when the absorber was covered with other viscous substances.

Analogous experiments have been performed quite recently to study the effect of covering the iron foil with various substances on the index of modulation of satellites excited by an rf field.[35] The studies confirmed the acoustic nature of the changes in the Mössbauer spectrum, and they indicated the great sensitivity of the method toward cleanliness of the surface. Thus, the index of modulation declined appreciably even upon touching the tested specimen with the fingers. Thus, the acoustic nature of the excitation of satellites in ferromagnetics has been rather reliably confirmed experimentally.

B. The magnetic nature of satellite excitation

A number of experiments have shown that satellite excitation involves the ferromagnetic structure of the substance.[3,4] In order to show this, the behavior of the satellites was studied in a monocrystal of $FeBO_3$ at varying specimen temperatures. $FeBO_3$ is a weak ferromagnetic with a Curie point at 348 °K. Experiment showed that the satellites vanish in the paramagnetic region. The same type of experiment was performed with a monocrystal of hematite, which is also a weak ferromagnetic, and which becomes antiferromagnetic below the Morin temperature (−13 °C). The satellites disappear in this latter state.

These experiments imply that satellites are excited only when an uncompensated magnetic moment exists.

C. The effect of a dc magnetic field

A number of experimental studies have been performed on the behavior of the satellites in dc magnetic fields.[3,4,36–40] The experiments performed with an iron-foil absorber showed a different relationship for perpendicular and parallel orientations of the dc magnetic field (with respect to the rf field direction). In the range from 80 to 320 Oe, they noted a small increase in the rms displacement of the nuclei. Then they observed a slow decline, while the intensity of the satellites rapidly approached zero beyond 1000 Oe. The intensity of the satellites declines considerably more rapidly in a parallel dc field. Lower fields are required to suppress the satellites in this case than for a perpendicular field.

D. Intra-domain magnetostriction

Excitation of ultrasonic vibrations by the rf field via an intra-domain magnetostriction mechanism has been most widely acknowledged for explaining the satellites. The theory is based on the following hypotheses[4,30]:

1) the relative deformation $\varepsilon = \delta l/l$ is described by using the static isotropic model:

$$\frac{\delta l}{l} = \frac{3}{2}\Lambda\cos^2\theta, \tag{3.1}$$

where Λ is the magnetostriction constant of the saturated ferromagnetic and θ is the angle between the direction of magnetization of the ferromagnetic and the direction in which the deformation is being measured:

2) the rf component $\mathbf{M}_1$ of the magnetization is far smaller than the static magnetization $\mathbf{M}_0$:

3) the static magnetostriction constants can be used in the rf range;

4) the acoustic vibrations generated by the rf field propagate in the polycrystalline specimen, and interact with the grain boundaries, the surface of the specimen, and the dislocations, with the result that the acoustic vibrations are scattered in directions that differ from the original direction.

Then one can show that acoustic vibrations are excited at the frequency of the rf field; consequently the index of modulation is expressed as follows:

$$m^2 \equiv \left(\frac{x_0}{\lambda}\right)^2 \approx \frac{1}{\lambda^2}\left(\frac{3}{2}l\Lambda\frac{[\mathbf{M}_0\times\mathbf{M}_1]}{M_0}\right)^2. \tag{3.2}$$

Equation (3.2) shows that satellites are excited only by the perpendicular component of the rf field. This agrees with the relationship of the intensity of the satellites to the direction of the dc magnetic field. Yet if we substitute in (3.2) the thickness of the foil absorber (several micrometers), then we get a value much smaller than what we get by experiment. If we substitute the value of the diameter of the absorber, the index of modulation attains huge values with absolute displacements of the nuclei up to 20 Å. Yet we note that, according to (3.1), magnetostriction does not lead to acoustic deformations along the direction of the γ rays (in the normal absorption geometry with the wave vector of the γ rays perpendicular to the surface of the specimen).

Additional experiments with the plane of the absorber

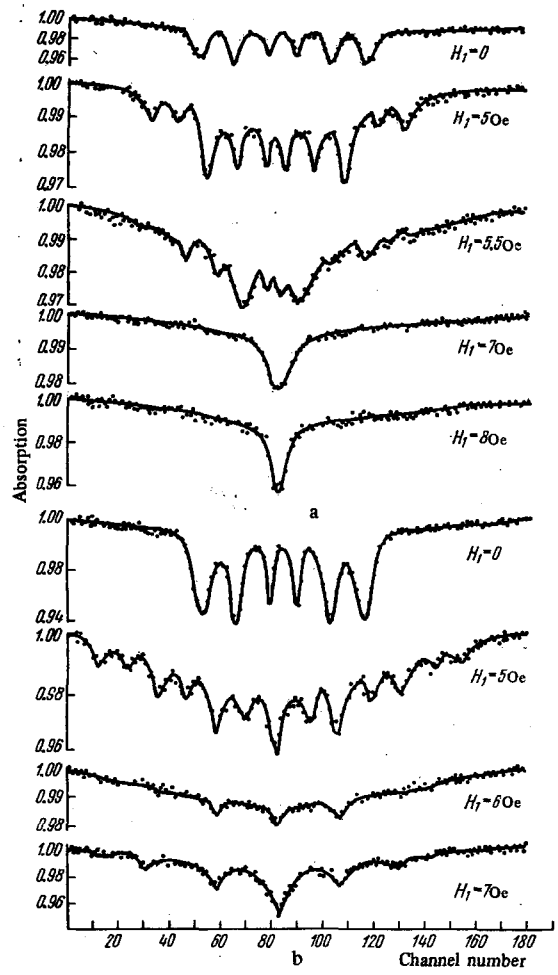

FIG. 6. Mössbauer spectra of a disordered (a) and an ordered (b) Ni_3Fe alloy at varying powers of a 40-MHz rf field.[41]

inclined showed also that the acoustic vibrations occur preferentially in the plane of the absorber. Hence, in order to explain the experimentally observed satellite intensities, they had to assume the existence of acoustic scattering in the specimen. At an amplitude of vibrations of 20 Å, one needs only several percent scattering of the energy of the acoustic wave perpendicular to the plane of the absorber.

In order to derive an expression for the index of modulation, they assumed that the Q factor of the acoustic vibrations was $Q=1$. The most recent Mössbauer experiments on the decay time of magnetostriction vibrations[17,18] have shown that $Q \approx 600$. Such a value of the Q factor gives rise to a considerably enhanced index of modulation. The latter situation works in favor of the proposed theory.

The remark made in[4] on the possible existence of spin waves in the rf range is also of interest. This hypothesis involves the fact that the rf field declines in iron to 13% of its original value at a distance of only two skin layers. Therefore one could expect the phenomenon of magnetostriction in the thin skin layer. Yet analysis of the satellite intensities in the Mössbauer spectrum shows that a substantial fraction of the nuclei in the specimen are vibrating at the imposed frequency. This favors the propagation of acoustic waves throughout the volume of the specimen.

We note also the use of GRS under rf-excitation conditions to study an entire set of magnetostrictional effects in magnetic alloys.[41] In particular, the existence has been shown of anisotropic excitation of ultrasonic vibrations arising from the texture in the rolling of the specimens. Side bands have been found in the rf excitation of an alloy of Fe+28% Ni that consists of a magnetic and a nonmagnetic phase. Here it was shown that the ultrasonic vibrations that arise in the magnetic phase propagate throughout the specimen. It was found in studying a monocrystal of Fe+4% Si that the amplitude of the ultrasonic vibrations increases substantially when the exciting rf field lies along the axis of easiest magnetization. It was also shown that the satellite intensities are substantially changed with varying degree of atomic ordering of a Ni_3Fe alloy in such a way that generation of ultrasonic vibrations is substantially facilitated in the ordered structure (Fig. 6).

Thus, application of GRS under rf-excitation conditions is apparently a convenient method of studying rf magnetostriction in magnetic alloys.

We have assumed thus far that linear magnetostriction plays the major role in exciting ultrasound. However, in certain ferromagnetics, such as the Invar alloys (Fe–Ni alloys with 30–45% Ni), ultrasonic vibrations of the nuclei have recently been observed that arise from volume magnetostriction.[39] The effect of ultrasonic modulation increased strongly with increase in the magnetic field perpendicular to $\mathbf{H}_1$ as $\mathbf{H}_0$ varied from 34 to 470 Oe. This relationship involves the increase in magnetostriction in the paraprocess region. However, an increase in $\mathbf{H}_0$ parallel to $\mathbf{H}_1$ led to a slower decline of the ultrasonic satellites than for iron foils.

E. Excitation of sound by Bloch walls

One of the mechanisms of the appearance of satellites in the Mössbauer spectrum could also be the generation of sound by Bloch walls. This theory has been proposed in[42].

Let us assume that the magnetostrictional energy of a cubic crystal is determined by the expression

$$E=\frac{B_1}{M_0^2}(e_{xx}M_x^2+e_{yy}M_y^2+e_{zz}M_z^2)+\frac{B_2}{M_0^2}(e_{xy}M_xM_y+e_{xz}M_xM_z+e_{yz}M_yM_z). \quad (3.3)$$

Here B_1 and B_2 are the magnetoelastic constants, the M_i are the components of the magnetization, and $e_{ij}=\partial u_i/\partial x_j+\partial u_j/\partial x_i$ is the deformation tensor.

By using (3.3), we can write the equation for the longitudinal vibrations of the plate as follows:[4)]

$$\frac{\partial^2 u_z}{\partial t^2}-v_t^2\frac{\partial^2 u_z}{\partial x^2}=\frac{B_2}{\rho M_0}\frac{\partial}{\partial y}(M_y\cos\theta). \quad (3.4)$$

[4)]The planes of the absorber and of the Bloch walls are respectively parallel to the planes xOy and xOz. The axis of easy magnetization lies along the Oz axis.

We see from Eq. (3.4) that excitation of vibrations of the plate along the direction of the γ radiation depends on the component of the magnetization perpendicular to the Bloch wall. Upon finding the component of the magnetization M_y by the Green's function method proposed by Janak[43] for elementary excitations of the Bloch wall and solving Eq. (3.4), one can show that the sound generated by the Bloch walls can account for excitation of satellites in ferromagnetics at amplitudes of the rf field of the order of several oersteds. Calculations show that the sound-excitation effect depends strongly on the direction of the rf field (perpendicular or parallel to the Bloch wall). The effect is negligibly small for perpendicular excitation of the Bloch wall. This fact has been confirmed by experiments on oriented specimens of yttrium iron garnet.[43] The suppression of the satellite effect with increase in the dc magnetic field can also be explained within the framework of the proposed model: the Bloch walls and the excitation of shear vibrations in the domains are abolished. The increase in the satellite effect in thin absorbers is explained by the relatively large volume of the walls in thin specimens.

A study of relaxation of ultrasonic vibrations excited by an rf field in permalloy (50% Ni) has shown recently[41,44] that the time spectrum of the decay has a complex structure. The authors suggest that it arises from magnetostriction of the Bloch walls.

F. Sound generation in metals by eddy currents

Recent studies[45] shown the possibility of generating ultrasonic vibrations in metals due to eddy currents induced by an rf field. This mechanism can be very substantial in paramagnetic metals at high temperatures and with short free flight paths of the electrons. This mechanism is manifested either through diffusional scattering of the electrons by the surface of the metals, or it involves the spatial dispersion of the electric conductivity. If $\delta/\lambda_{sound} \ll 1$, where δ is the depth of the skin layer, and λ_{sound} is the wavelength of the ultrasonic wave, then an ultrasonic modulation effect should occur in ferromagnetics having a high magnetic permeability. We note that, just as in the case of magnetostriction inside a ferromagnetic domain, the eddy currents excite elastic vibrations in an isotropic metal in the plane of the specimen. Therefore, here also, scattering of the acoustic wave by inhomogeneities of the specimen plays a large role in observation of ultrasonic modulation.

Experiments have been performed both with ferromagnetic and with paramagnetic metals, e.g., in an alloy of gold to which 7 atom % ^{57}Fe has been added.[45] The authors view these experiments as confirming the eddy-current hypothesis. However, in order to elucidate the specific contribution of the eddy-current mechanism to excitation of ultrasonic modulation of γ radiation, experiments[46] have very recently been performed in two alloys of the permalloy class that have substantially different magnetostriction coefficients, but which have similar electric conductivities. The spectrum of the alloy 50 H (50% Ni), which has appreciable magnetostriction, shows a sharply marked type of ultrasonic modulation, whereas the spectrum of the alloy 81 HM (81% Ni, 6% Mo), which has insignificant magnetostriction, lacks ultrasonic satellites. This experiment shows that the ultrasonic modulation in the given alloys arises from magnetostriction rather than from eddy currents, as the cited study[45] has pointed out.

Another experiment has recently been performed to test the eddy-current hypothesis.[47] The experiment was carried out as follows: an absorber made of stainless steel was attached on both sides to films of Ni, Ag, or Cu. Satellites were observed only in the case of Ni, which is known to have a large magnetostriction, while they were lacking for the nonmagnetic metals Ag and Cu. These experiments confirm the doubts on the effectiveness of the eddy-current mechanism in exciting satellites in ferromagnetics.

G. Parametric excitation of magnetoelastic vibrations

Up to now, the nonlinear dynamic properties of ferromagnetics in the UHF range have been studied by the ferromagnetic resonance method. However, these same phenomena can be studied also by using GRS.

One of the possible Mössbauer studies of the parametric excitation threshold of ultrasonic vibrations is the excitation of a magnetoacoustic resonance.[48] Here two transverse magnetic fields in the UHF range are applied in the plane of a ferromagnetic absorber that is magnetized perpendicular to the surface of the specimen: a homogeneous-precession field (frequency ω_1) and a pumping field (frequency ω_p). The parametric coupling of the elastic standing wave and the magnetostatic modes of the high-frequency magnetizations, causes longitudinal ultrasonic vibrations (perpendicular to the plane of the absorber) to be excited with frequency ω_2 under the condition $\omega_p = \omega_1 + \omega_2$. The study showed that a threshold value of the elastic vibrations gives rise to satellites that can be detected by the GRS technique.

It was recently reported[49] that the Mössbauer spectrum in the rf excitation of hematite shows extra satellites in addition to the usual ones. The extra satellites arise from parametric excitation with a resonance frequency of $\omega/2$, where ω is the frequency of the rf field. It was found that the intensities of these satellites reach saturation with increasing power of the rf field, and here they exceed the intensities of the usual satellites by a factor of two.

4. REMAGNETIZATION OF FERROMAGNETICS BY RADIOFREQUENCY FIELDS. COLLAPSE OF HYPERFINE STRUCTURE

A. Experiment

Pfeiffer[50] has observed an interesting phenomenon in the variation of the Mössbauer spectrum of permalloy foil (58% Fe, 42% Ni) in a strong rf field of intensity 15 Oe. Here the Mössbauer spectrum collapsed into a single line as the frequency of the rf field was increased up to 106 MHz (Fig. 7), just as happens in fast electronic relaxation.[51,52] The diagram also shows the ultrasonic satellites that are generated by the rf field. Application of a dc magnetic field parallel to the re-

magnetizing field restores the spectrum.

Pfeiffer's explanation[3,50] was that an rf field of amplitude greater than the anisotropy field remagnetizes the specimen, and the hyperfine field antiparallel to the magnetization oscillates in time with the variation in the magnetization. If the frequency of variation of the hyperfine field is far larger than the frequency of nuclear precession, then the axis of quantization at the nucleus becomes indeterminate in space, the hyperfine field averages to zero, and the Mössbauer spectrum consists of a single line—the so-called "collapse of hyperfine structure."

Analogous measurements have been performed[53] on magnetoplumbite $Ba_2Zn_2Fe_{12}O_{22}(Zn_2Y)$, which has a helical structure. This substance has a low anisotropy field of the order of 0.4 Oe. Hence they expected that remagnetization would be easily attainable in fields of 60 Oe at frequency 96 MHz.

They measured the Mössbauer spectrum simultaneously in the presence and absence of an rf field. This was done with a circuit for counting of γ quanta synchronized with the pulses from the rf generator.[54] The even channels of this circuit were opened at the instant of action of the rf field, while the odd channels recorded the quanta in its absence.

Thus they ruled out an effect of alteration of the spectrum by heating of the specimen. Comparison of these spectra showed that the spectrum with the rf power on was more strongly narrowed than that with the rf power off.

While Zn_2Y shows an appreciable magnetostriction, they did not detect satellites. Remagnetization experiments have also been performed on permalloy 81 HM (81% Ni, 6% Mo, 13% Fe),[40,46] which has an insignificant magnetostriction. The Mössbauer spectrum showed neither magnetostrictional bands generally when acted on by an rf field, nor particularly during collapse.

B. Discussion

As we know, remagnetization goes through three stages with increasing intensity of the remagnetizing field[55,56]: at first, a gradual remagnetization arising from movement of domain walls (the time of remagnetization of permalloy ≈ 1 μsec); then an inhomogeneous rotation of the magnetization vector follows; and later a homogeneous coherent rotation (the remagnetization time becomes considerably shorter than in the previous cases, becoming as small in thin films as ≈ 1 nsec).[5)]

It was shown in[53] by direct measurement that the magnetization varies coherently with the rf field in the collapse of hyperfine structure. However, the hyperfine structure can also collapse during the inhomogeneous rotation, which in a number of cases takes less than 5–10 nsec.[58] Yet the remagnetization of the specimen arising from movement of domain walls should not lead to collapse of the spectrum, owing to the slowness of the process. The process of collapse of the hyperfine structure owing to fast rotation of the magnetization somewhat resembles that of transition of a ferromagnet at the Curie point to the paramagnetic state, in the magnetization fluctuations arising from thermal motion abolish the magnetic order of the substance. The difference consists in the fact that the rf collapse can be either coherent (homogeneous rotation) or incoherent or partially coherent in nature (inhomogeneous rotation), in contrast to the phase transition, which is incoherent in nature.

The existence of the satellites in incomplete collapse shows that the magnetic moments of the atoms are partially ordered. This also agrees with the above-proposed physical picture of the collapse of the Mössbauer spectrum.

In conclusion, let us point out also the theoretical studies on remagnetization arising from movement of domain walls.[59,60] In this model, the hyperfine structure collapses at a remagnetization frequency considerably exceeding the nuclear precession frequency. They obtained a very close resemblance with Pfeiffer's results[50] upon assuming a jumpwise variation of the field at the nucleus arising from the movement of the domain walls.[60] By using a relaxation model, Perlow[59] tried to explain the effect of an rf field having a frequency of several MHz on the Mössbauer spectrum of permalloy. Yet further studies have shown[36] that an analogous

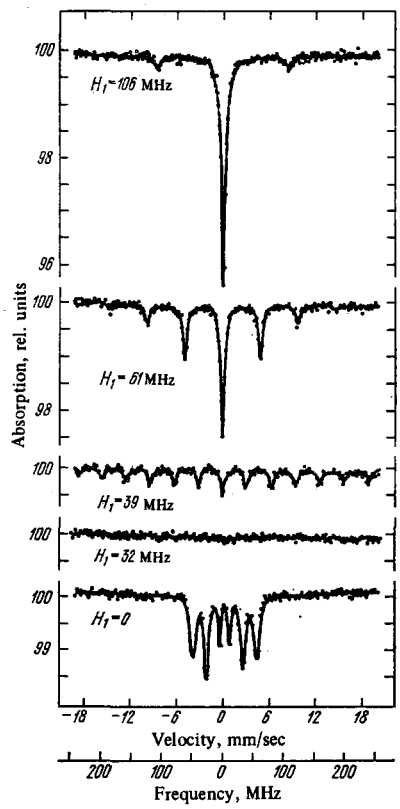

FIG. 7. The rf collapse effect as a function of the applied frequency.[50]

[5)]Therefore, analysis of the collapsing spectrum as a function of the intensity of the rf field opens up possibilities for determining the remagnetization time of the specimen, as has recently been shown in an experiment with permalloy films.[57]

form of the spectrum at the stated frequencies arises from magnetostriction. Apparently, further progress in the Mössbauer study of rf remagnetization will involve additional studies. In particular, the problem of the effect of a perpendicular dc magnetic field on the remagnetization process needs study. Such studies can help in establishing the regions of existence of the remagnetization mechanisms.[61]

One of the methods of studying the inhomogeneous rotation of the magnetization can be the remagnetizing-echo method, which is non-resonance in nature, in contrast to the paramagnetic spin echo[62] and the ferromagnetic echo.[63] As we know, application of the ferromagnetic echo in ferrites led to the discovery of the phenomenon of echo amplification.[64] We can expect analogous phenomena involving nonlinear magnetic interactions also upon excitation of the remagnetization echo.

5. γ-MAGNETIC AND γ-ACOUSTIC RESONANCES

The effect of γ-magnetic resonance (GMR) is a process of simultaneous resonance excitation of a Mössbauer nucleus by a γ-radiation field and an rf magnetic field. The former causes quantum transitions between the isomeric states of the nucleus, and the latter stimulates transitions within an isomeric state between the hyperfine nuclear sublevels. The effect of γ-acoustic resonance (GAR) differs from GMR in that the transitions between the hyperfine sublevels are excited by an ultrasonic field. Therefore we shall first take up the GMR effect.

The action of the rf field in excitation of GMR is manifested in two ways. In the case of equally populated nuclear sublevels, the fundamental effect of the rf field is a resonance-type alteration of the effective ac field at the nucleus. Yet if the population difference of the nuclear sublevels becomes substantial (either owing to cooling of the specimen to hundredths of a degree Kelvin, or by selective excitation of the nuclei[65]), then the fundamental effect becomes that of equalizing the nuclear populations by the action of the resonance rf field. In either case, the resonance should be observed at a frequency of the rf field that matches the frequency of Larmor precession of the ground or excited states of the nucleus.

The GMR effect was treated theoretically in 1966–1968 by one of the present authors.[66–68] An estimate was made of the possibility of experimental observation of this effect in ferromagnetics, and in particular, in pure iron.[66] At first, the GMR effect was treated for rf excitation at a frequency close to the Larmor precession frequency and for equally populated nuclear sublevels.[66] Then the theory was generalized to the case of exact excitation of NMR and unequal populations of the nuclear sublevels.[67,68] Among the theoretical studies in this field, we should also point out the study by Gabriel[69] and that by Hack and Hamermesh.[70]

The first experiments to detect GMR were carried out in a scattering geometry by the method of selectivity exciting the ^{57}Fe nuclei in pure iron.[71] A number of studies have now been published in which people have tried to observe GMR in other substances and with other Mössbauer isotopes.[5,72–75]

Practical application of the GMR and GAR effects will apparently be most useful for studying resonance phenomena in crystals in ac fields powerful enough, and with nuclear or electronic relaxation times short enough that the effects of NMR and NAR are no longer effective. By using the methods of GMR and GAR, which amount to a combination of the Mössbauer effect on the one hand, and of NMR and NAR on the other, it would seem that we could get varied information on the structure of solids.

A. The theory of γ-magnetic resonance. Transmission of γ-radiation

Let us now proceed to the theory of GMR under conditions of transmission of γ radiation. In establishing the theory of GMR, we can start with the method of simultaneously solving the Maxwell equations and the equations for the density matrix[76][6)] for a system of Mössbauer nuclei that lie in a dc plus an ac magnetic field.

For the sake of argument, let us study a ferromagnetic that is magnetized to saturation along the Oz' axis by a dc magnetic field $\mathbf{H}_0$. A circularly polarized rf magnetic field $\mathbf{H}_1$ of frequency ω is directed perpendicularly to the dc magnetic field. The Hamiltonian of interaction of the nucleus with the magnetic field has the form

$$\hat{\mathcal{H}}_k^0 + \hat{\mathcal{H}}^{rf} = -g_k\mu_N[(H_{hyp}+H_0)\hat{I}_{kz} + H_1(\hat{I}_{kx}\cos\omega t + \hat{I}_{ky}\sin\omega t)]. \quad (5.1)$$

Here the subscript $k = g$ or e denotes the ground and excited states of the nucleus, respectively. H_{hyp} is the hyperfine field at the nucleus, g_k is the nuclear g-factor, and μ_N is the nuclear magneton. The frequency ω of the rf field must lie near the region of excitation of NMR. Therefore the sign of ω is chosen by using the relationship sign $\omega = -\operatorname{sign} g_k$.

If we now solve simultaneously the Maxwell equations for the electromagnetic field intensity of the γ wave and the equation for the density matrix for the nuclear γ transitions in the presence of the resonance rf field, we can find the tensor refractive index of the γ wave[76][7)]

$$n_{\nu\nu'} \approx \delta_{\nu\nu'} - \frac{n_0(\nu\nu')^2}{2I_g+1} \times \sum_{\substack{m_e, m_g \\ m_{e'}, m_{g'}, M}} \frac{[d^{(I_e)}_{m_e, m_{e'}}(\beta_e)]^2 [d^{(I_g)}_{m_g, m_{g'}}(\beta_g)]^2 \Gamma/2}{i(\Gamma/2) + [\omega_\gamma - \omega_0 - a_e m_e + a_g m_g + \omega(m_{e'} - m_{g'})]} \times C^2(I_g, L, I_e; m_{g'}, M, m_{e'}) D^{(L)*}_{\nu M}(\varphi, \theta, 0) D^{(L)}_{\nu' M}(\varphi, \theta, 0). \quad (5.2)$$

Equation (5.2) makes it possible to interpret the GMR effect here as the varying interaction of the γ radiation with the system of nuclei lying in the effective ac magnetic field as a function of the frequency and amplitude of the rf field. Here there is a variation in both the quantity

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[6)] The theory of GMR is altered only slightly in studying the emission process.

[7)] We assume that the nuclear sublevels are equally populated.

$$\omega_{\text{eff}\,k} = \frac{\hbar a_k}{g_k \mu_N} = \frac{\hbar}{g_k \mu_N} \sqrt{(\omega_k - \omega)^2 + \omega_{1k}^2},$$

and in the direction of the magnetic field; the angle between the dc field and the effective field is:

$$\beta_k = \arcsin\left(\frac{\omega_{1k}}{\sqrt{(\omega_k - \omega)^2 + \omega_{1k}^2}}\right).$$

One can derive theoretically[77] an analytical expression for the intensity of resonance absorption of the γ rays at small intensities of the rf field in (5.2) for a thin absorber by using the polarization density matrix. This expression is the sum of three terms: the first defines the ordinary one-quantum Mössbauer absorption, and the second and third define two- and three-quantum combination-type transitions with absorption of a γ quantum and absorption or emission of a photon of the rf field in the ground or excited state of the nucleus.

For GMR in this approximation, the previously forbidden transitions with the selection rule $|m_e - m_g| = 2$ become possible, in addition to the ordinary γ transitions having the selection rule $|m_e - m_g| = 0, 1$ (dipole magnetic or electric emission).[66] In this regard, we note also the change in polarization of the radiation when GMR is excited.[78]

Figure 8 shows GMR transitions ($1/2\,g \rightarrow 3/2\,e$) at a frequency of the rf field that coincides with the Larmor precession frequency ($\omega = \omega_e$), as well as a GMR transition ($-1/2\,g \rightarrow 3/2\,e$) at a frequency $\omega \cong \omega_g$. Here the two-quantum process is determined by only a single intermediate level, and it consists of a sequence of two one-quantum transitions. The intensity of these transitions is maximal when $\omega = \omega_{g,e}$.[67,68] Yet if the frequency ω appreciably differs from the frequencies $\omega_{g,e}$, then a satellite is excited at the frequency $\omega_\gamma = \omega_{g,e} \pm \omega$.[66]

As the intensity of the rf field increases ($\omega_{1g,e} \simeq \Gamma$), the GMR lines broaden and shift in proportion to the intensity of the rf field.[67,68] Further increase in the intensity of the rf field causes the two-quantum approximation to fail, and in practice all transitions between sublevels of the ground and excited states of the nucleus are allowed. If the parameter $\omega_{1g,e}$ is comparable with the hyperfine splitting of the levels, then each line of the Mössbauer spectrum is split into $(2I_g+1)(2I_e+1)$ components.[69] However, if the nuclear g-factors are not equal (e.g., the nucleus ^{57}Fe in iron has the Larmor frequencies: $\nu_e = 26$ MHz, $\nu_g = 45.4$ MHz), then the Mössbauer lines split only into $2I_e+1$ or $2I_g+1$ components, depending on the frequency of the rf field.

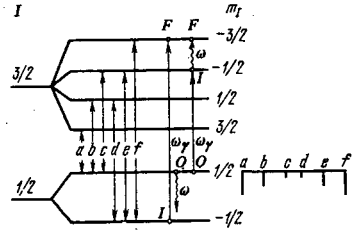

FIG. 8. Diagram of energy levels and Mössbauer absorption for ^{57}Fe in an external magnetic field. Two-quantum transitions in Mössbauer absorption. O, F, and I respectively denote the original, final, and intermediate states.

TABLE II. Minimum values of the internal magnetic field for detection of GMR.[5]

Nucleus	State	μ_N	I	Γ, keV	H, kOe
^{57}Fe	Ground	0.0902	1/2	$4.67\cdot10^{-12}$	8.1
^{61}Ni	Ground	0.746	3/2	$8.61\cdot10^{-11}$	55
^{67}Zn	Ground	0.876	5/2	$4.85\cdot10^{-14}$	0.044
^{119}Sn	Ground	1.046	1/2	$2.48\cdot10^{-11}$	3.83
^{121}Sb	Ground	3.36	5/2	$1.3\cdot10^{-11}$	30
^{129}I	Excited	2.84	5/2	$2.83\cdot10^{-11}$	7.8
^{161}Dy	Excited	0.55	5/2	$1.59\cdot10^{-11}$	23
^{181}Ta	Ground	2.36	7/2	$6.71\cdot10^{-14}$	0.031
^{197}Au	Excited	0.37	1/2	$2.54\cdot10^{-10}$	108

We also note that the nature of the GMR spectrum depends strongly on the electronic relaxation time.[79] Thus, in a theoretical model having complete inversion of the hyperfine field at the nucleus, the decrease in the electronic relaxation time enhances the effect of the rf field.

B. The size of the γ-magnetic resonance effect

As the calculations imply,[66] the ratio of the GMR cross section to that of the one-quantum Mössbauer transition is determined at low rf field intensities by the ratio $(\omega_{1k}/\Gamma)^2$. Table II gives the values of the magnetic field at the nucleus that satisfies the condition $\omega_{1k} = \Gamma$ for various Mössbauer isotopes. We see from the table that direct excitation of GMR by an rf field is possible only for ^{181}Ta and ^{67}Zn. The rest of the nuclei require amplification of the field at the nucleus.

Estimates of GMR were first performed[66] for ^{57}Fe nuclei in iron existing in a multidomain state, with account taken only of nuclei lying in the Bloch domain walls. This type of calculation was dictated by the fact the largest signal comes from the nuclei within the Bloch walls, according to NMR data on ^{57}Fe nuclei in iron.[80] The rf field at the nucleus is amplified in the Bloch walls by the oscillations of the electronic magnetization. The coefficient of amplification of the rf field at the nucleus attains values $\eta \cong 10^3$ in iron and cobalt,[81] and the enormous value $\eta \simeq 2.4\times10^5$ in a monocrystal of hematite.[82]

Another possibility is to observe GMR in the monodomain state of a ferromagnet.[83] In this case, the amplification coefficient of the rf field at the nucleus is

$$\eta = \left(1 + \frac{H_{\text{hyp}}}{H_0 + H_a}\right), \qquad (5.3)$$

where H_a is the anisotropy field.

The amplification coefficient in this case is about an order of magnitude smaller than in a Bloch wall, but the number of nuclei that participate in the process increases by an order of magnitude. Therefore, in line with the fact that the GMR effect in the two-quantum approximation is proportional to the square of the rf field at the nucleus, the multidomain state of a ferromagnetic is theoretically more favorable for observing GMR than the monodomain state.

As for antiferromagnetics, the amplification coefficient of the rf field at the nucleus is mainly determined

by Eq. (5.3), though the nature of the rf excitation here is marked by more complexity than in ferromagnetics.[84]

A possibility of observing GMR in paramagnetics has been treated in[85]. The amplification coefficient in this case is determined in order of magnitude by Eq. (5.3), with $H_a = 0$. It was also shown that electronic relaxation leads to damping of the oscillations of the nuclear moment that are excited by the rf field. Thus an imaginary component of the amplification coefficient arises.

A positive feature of paramagnetics is the absence of magnetostrictional vibrations, whose effect on the Mössbauer spectrum strongly interferes with observing GMR in ferromagnetics.

Thus, when one uses GMR, the amplification coefficient of the rf field at the nucleus can give information on the magnetic structure (domains, Bloch walls, concentration of paramagnetic ions, etc.), on electronic nuclear interactions, and on relaxation in magnetically-ordered and paramagnetic compounds.

C. Methods of observing γ-magnetic resonance. Experiment

GMR was first detected successfully by the scattering method by a group of American physicists.[71] For this purpose, they used the effect of selective excitation of Mössbauer nuclei that had been proposed by Artem'ev *et al.*[65] The principle of selective excitation consists in populating a certain sublevel of the excited state of the nucleus by using a source of Mössbauer γ-radiation moving at constant velocity. The scattered radiation is analyzed with a Mössbauer absorber that moves with constant acceleration. Consequently, scattering occurs only from a given level of the nucleus having a certain magnetic quantum number, and it corresponds to almost 100% polarization of the Mössbauer nucleus.

Application of a resonance rf field stimulates transitions between the selectively excited level and other hyperfine levels. Consequently, the population of the selectively excited level declines, and the populations of the other hyperfine sublevels of the excited state of the nucleus increase. Here the main population change will occur at field intensities $H_1 \gtrsim \hbar\Gamma/g_e\mu_N$.

As an example, let us study the selective excitation of ^{57}Fe nuclei (see Fig. 8). When the level $-3/2\,e$ is excited, the spontaneous magnetic dipole emission will consist only of the line f, whereas the spontaneous emission from the level $-1/2\,e$ will consist of the lines e and c. Excitation of GMR alters the situation.[8)] A resonance rf field ($\omega = \omega_e$) has the result that the line c appears along with the line f in the scattering spectrum upon selective excitation of the level $-3/2\,e$.

The experiment was performed on ^{57}Fe nuclei contained in a thin layer of iron powder.[71] The investigators paid special attention to the grain size of the powder, since magnetostrictional ultrasonic vibrations arise when one uses an iron foil. However, as we have seen, ultrasonic vibrations are not excited if the size of the particles is less than half the wavelength. The experiment confirmed the appearance of extra lines, in accord with the above-presented arguments. Yet the effects of broadening and splitting of the lines were not noted at intensities of the rf field up to 75 Oe. The lack of broadening in GMR with selective emission has also been shown theoretically.[90]

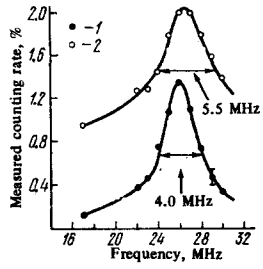

FIG. 9. Relationship of GMR (^{57}Fe in iron) to the frequency of the rf field in a dc magnetic field.[2] 1—$H_0 = 0$, 2—$H_0 = 2$ kOe.

However, the proposed methodology hinders analysis of the GMR as a function of the frequency of the rf field. Therefore a certain modification of the experiment was carried out.[2] The source and the absorber move at constant velocity, so that the absorber absorbs at resonance the radiation that is re-emitted by the scatterer. When the rf field is turned on, spontaneous emission arises also from other levels that are not in resonance for the absorber, and this diminishes the absorption of the scattered radiation. Three features have been found by this method (Fig. 9).[2] First, the GMR effect in a dc magnetic field of intensity 2 kOe is almost twofold larger than in its absence. This indicates the situation that mainly the nuclei in the domains contribute to the GMR: the domain walls in iron break down at such fields. Second, the probability of GMR remains rather high for frequencies far from NMR. Third, the resonance frequency is shifted from 26.0 to 26.5 MHz. This shift arises from the dc magnetic field.

These data show, first, that the experimental studies of GMR and NMR give opposite results. Thus, the main contribution to the NMR signal in ferromagnetics comes from the nuclei lying in the Bloch walls, whereas the contribution to GMR comes from the nuclei in the domains. Apparently the chief reason is that one uses very strong rf fields in GMR, for which the mechanism of amplification of the field at the nucleus in the Bloch walls fails. Second, excitation of GMR at frequencies far from the Larmor frequency indicates that the combination processes in GMR are substantial.[66]

Meisel[5,72] has performed an analogous experiment on hematite with use of selective excitation to observe GMR. This substance is interesting in that the amplification coefficient of the field at the nucleus attains the value $\eta = 24,000$ in the Bloch walls of the specimen, which was a thin layer of powder under the experimental conditions. However, the selective-excitation spectrum showed the presence of a weak GMR effect.

[8)] A number of studies, beginning with Abragam and Pound[86] (see also[87–89]) have discussed the problems of the effect of a resonance rf field on the angular correlation and anisotropy of γ radiation.

In order to create a population difference by cooling the specimen to superlow temperatures, one must have $\Gamma\tau_1 \ll 1$, where τ_1 is the longitudinal nuclear relaxation time. Therefore one must observe GMR arising from equalization of the populations by an rf field either with Mössbauer sources, or with the ground state of a Mössbauer nucleus. The experiments were performed[74,75] on the nuclei of a ^{57}Co source with the latter cooled to hundredths of a degree Kelvin. The NMR of the ^{57}Co nuclei causes a resonance-type diminution in the asymmetry of the Mössbauer spectrum of the ^{57}Fe nuclei that arises from cooling of the source. Observation of the GMR of the ^{57}Co nuclei in iron permitted them to determine the Knight shift.[75]

An attempt to detect GMR in an absorption geometry was first undertaken by Matthias,[87] who observed a resonance change in the counting rate of the Mössbauer spectrum of ^{57}Fe at frequencies of the rf field that equaled the Larmor precession frequencies of the nuclei in the ground or the excited states of the nucleus. However, subsequent measurements have shown[4] that the resonance effect in this experiment was due in large part to excitation of magnetostrictional satellites and the overlap of the latter with the fundamental lines of the Mössbauer spectrum.

An experiment has successfully observed GMR in an absorption geometry in ^{181}Ta nuclei,[73] which have a rather long half-life: $T_{1/2} = 6.8 \times 10^{-6}$ sec. A ^{181}W source in tungsten that was split in an 1800-Oe magnetic field was analyzed by using a ^{181}Ta absorber in tantalum. Application of a 10-Oe rf field perpendicular to the dc magnetic field gave rise to a resonance GMR effect, whereby they determined the ratio of g-factors of the ground and excited states of the nucleus.

The idea has recently arisen of applying polarization methods for observing GMR that is based on the differing nature of the effect of an rf field in stimulating NMR transitions and in magnetostrictional ultrasonic modulation. As the theoretical analysis has shown, polarization methods using nuclear Faraday effects, birefringence, etc., show a resonance dependency, and they permit one to distinguish the GMR effect from ultrasonic modulation.[77]

Thus, the fundamental inference of the theory of GMR concerning the resonance action of an rf field has been distinctly manifested in experiments performed in a scattering geometry[71,2] and in an absorption geometry.[73] Yet a number of the effects that are implied by the GMR theory: excitation of satellites and broadening and splitting of lines, requires additional experimental verification.

D. γ-Acoustic resonance

γ-acoustic resonance (GAR) differs from GMR in that the transitions between the hyperfine sublevels of the nucleus are excited by using nuclear acoustic resonance (NAR).[91,92]

A number of authors have treated the different mechanisms of exciting NAR. Thus, Bashkirov and Sadykov[93] have proposed the possible excitation of GAR by a mechanism of modulation of the quadrupole interaction by an ultrasonic field. Here the transitions that are stimulated by NAR differ from NMR in the selection rule of both $|\Delta m| = 1$ and $|\Delta m| = 2$. Recently these authors[94] have proposed using the mechanism of quadrupole interaction in ferroelectrics to observe GAR, as well as its analog involving excitation by an ac electric field.

Another mechanism of resonance action of an ultrasonic field on the nuclear spins is the mechanism of modulation of the magnetic hyperfine interaction in paramagnetics by exciting the electronic spin system. As estimates show, this mechanism is significant for paramagnetic ions having strong hyperfine spin-phonon interactions.[95]

Excitation of GAR in ferromagnetics has been proposed in[96]. According to this study, the interaction of an acoustic wave with the nuclear spin system involves the Silverstein mechanism,[97] which amounts essentially to transfer of acoustic energy by magnetostriction of the electronic system and then by the hyperfine interaction of the nuclear spin system.

Thus, the GAR effect permits one to study the spin-phonon interaction of nuclei in crystals having varied magnetic and electric structures. Apparently, the most suitable nucleus for excitation of GAR is ^{181}Ta, which has a large quadrupole interaction, and for which NAR has already been observed.[98]

E. Narrowing of lines of long-lived Mössbauer isotopes by an rf field

In connection with the problem of nuclear dipole-dipole and inhomogeneous broadening of spectral lines of long-lived Mössbauer isotopes, a number of methods have been proposed quite recently involving the narrowing of these broadenings by resonance rf fields.[99–105] Thus, they have shown that the use of a series of rf $\pi/2$ pulses,[100] as well as quasicontinuous rf cycles,[105] makes it possible to narrow the width of a line that arises from quadrupole and dipole-dipole interactions by a ratio of $6/(\Gamma_0\tau)^2$, where Γ_0 is the width of the line before averaging by the rf pulses, and τ is the period of the series of pulses. Another possibility for applying an rf field to suppress inhomogeneous broadening of Mössbauer lines is the idea of compensation of an inhomogeneous chemical shift.[99,102–104] The idea of compensation is based on the assumption that the inhomogeneous variations of the chemical shift are proportional to the inhomogeneous variation of the hyperfine splitting. The further development of the theory of rf narrowing of the lines of long-lived Mössbauer isotopes has recently acquired great importance in connection with the problem of inventing a source of stimulated γ radiation.[106]

6. MÖSSBAUER SPECTRA IN ELECTRON PARAMAGNETIC RESONANCE AND IN OPTICAL EXCITATIONS

A. Electron paramagnetic resonance

One of the influences of EPR on the Mössbauer effect is a possible alteration of the Boltzmann populations of

the electronic levels.[107] Alteration of the populations of the electronic levels by an ac magnetic field can lead to both a change in the hyperfine interaction and to polarization of the Mössbauer nuclei by dynamic orientation.[108] An appreciable change in the Mössbauer lines corresponding to the hyperfine interaction of the studied electronic levels n and m can happen only under the condition $E_n - E_m \approx kT$, where E_n is the energy of the electronic level and T is the temperature of the medium. Therefore, for the energy range of splittings 10^9 Hz $\leq (E_n - E_m)/h \leq 10^{11}$ Hz, the temperature of the specimen must lie in the range 5×10^{-2} °K $\leq T \leq 5$ °K, or lower.

Changes in the populations of the electronic levels can include both equalization and inversion. Here the amplitude of the resonance ac magnetic field $\mathbf{H}_1$ perpendicular to the dc magnetic field $\mathbf{H}_0$ and the longitudinal (T_1) and transverse (T_2) electronic relaxation times must satisfy the condition $(\gamma H_1)^2 T_1 T_2 \approx 1$, where γ is the electronic gyromagnetic ratio. Equalization can occur both in a continuous regime and in a pulsed regime (using a $\pi/2$ pulse). Yet inversion can occur only in a transition regime: by using a π-pulse or by adiabatically rapid passage through resonance (see also[109]).[9)]

Another effect is the broadening[107] or even the collapse of the Mössbauer spectrum into a single line at high powers of EPR excitation.[110] The effect amounts to stimulated relaxation, whose probability is $W \approx (\gamma H_1)^2 T_2$. An experiment can be conducted for times $T_2 \geq 10^{-6}$ sec and field intensities $H_1 \approx 1-10$ Oe. Excitation of electronic acoustic resonance gives rise to a similar effect.[111] The effect of stimulated relaxation on the Mössbauer spectrum is analogous to the effect of thermal electronic relaxation, but it differs in that one can control the length of the relaxation time by varying the power of the ac field.

Finally, we note also the effect of shifting the electronic states, which is very sensitive in certain compounds to the value of the magnetic field, e.g., in $Al_2O_3:Fe^{3+}$.[112] Modulation of the latter by an ac magnetic field can lead to considerable changes in the Mössbauer spectrum.

In summing up, we note that there has as yet been no experiment in this field,[10)] in spite of the considerable number of theoretical studies. Apparently the main difficulty in carrying out such experiments is the need of combining low temperatures with large powers of the ac magnetic field.

B. Magnetic resonances in ferromagnetics

The fundamental idea along this line is the possibility of exciting spin waves at high powers of high-frequency excitation. An experiment to observe Mössbauer absorption in polycrystalline yttrium iron garnet indicated a strong broadening of the lines of the spectrum.[114] Apparently the fundamental reason for the broadening is the excitation of spin waves that stem from scattering of the homogeneous precession by inhomogeneities of the specimen.

This statement is corroborated by recent experiments studying the effect of UHF pumping on the nuclear spin echo in yttrium iron garnet.[115] They observed a decline (by three orders of magnitude) of the transverse nuclear relaxation time, and interpreted it as a parametric excitation of spin waves.

These experiments show the possibility of Mössbauer study of the dynamics of magnetically ordered substances, and in particular, threshold and super-threshold phenomena of nonlinear ferromagnetic resonance.

C. Optical radiation

On the one hand, the effect of optical radiation on the Mössbauer effect is analogous to EPR, both in being based on a change in the populations of the optical levels and in leading to stimulated relaxation proportional to the power of the optical radiation. The former can alter the sizes of the chemical shift[116] and of the quadrupole[117] and the magnetic[67] interactions. The latter can broaden the Mössbauer lines.[67,118]

On the other hand, optical radiation can lead to modulation of γ radiation by stimulating vibration of molecules and ions. For example, γ radiation can be modulated in gases by selective excitation by a laser beam of vibrational levels of the molecules of the gas.[119] The narrow side lines excited by the laser beam make it possible to observe nuclear γ resonance in gases.

One can also expect changes in the Mössbauer spectrum during light scattering, owing to excitation of spin waves, two-magnon states, plasmons, high-frequency acoustic vibrations, etc. However, further discussion of this problem lies outside the scope of this review.

7. CONCLUSION

Thus we have discussed in this review the status of the theoretical and experimental studies in the new field of application of GRS. As the review shows, the fundamental experiments have been performed with ^{57}Fe nuclei. Yet, without doubt it is promising to study substances containing other Mössbauer isotopes, including those obtainable by Coulomb or other mechanisms of excitation of nuclei.

As we see it, applying pulsed high-frequency methodology to study the dynamics of transition processes in solids is of great interest. We note also that application of emission Mössbauer spectroscopy combined with pulsed high-frequency field technique would substantially expand the possibilities of the method in studying both various coherent processes, and the after-effects of nuclear transformations.

In the field of phenomena involving GMR, GAR, or EPR plus the Mössbauer effect, etc., we should expect a deeper study of the nonlinear resonance phenomena

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[9)] The effect of equalization of populations of electronic levels in the excitation of EPR can be used also for modulating γ radiation.[27]

[10)] An attempt has recently been undertaken to observe an effect of EPR on a Mössbauer absorption spectrum.[113] However, the statistical accuracy of the results of this experiment proved insufficient.

that arise upon applying high-power, high-frequency fields. In paramagnetics, this mainly involves the concept of the spin temperature, which particularly alters the relaxation of the spin system in the presence of a strong high-frequency field. In magnetically-ordered substances, it is of great interest to study coupled oscillations of electronic and nuclear spins, together wtih studies in the field of nonlinear ferromagnetic resonance (paramagnetic excitation of spin waves, above-threshold phenomena, etc.).

In conclusion, we also express the hope that the development of solid-state GRS under high-frequency excitation conditions will become one of the first steps in establishing the physics of nonlinear processes in the γ range.

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