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Modulation gamma-resonance spectroscopy

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1. Introduction

Thanks to the Mössbauer effect, the gamma range of electromagnetic waves is being mastered more and more in both fundamental and technological areas and is beginning to compete, in terms of its complexity of problems and exactness of measurements, with the optical range with its laser light sources [1, 2]. The standard Mössbauer spectroscopy is being replaced with gamma optics, an important part of which is the modulation gamma-resonance spectroscopy. The idea behind this spectroscopy is the possibility of controlling dynamical processes in matter by modulating a gamma resonance via external varying magnetic fields with frequencies exceeding the reciprocal lifetimes of excited states of Mössbauer nuclei (i.e., in the megahertz frequency range). This frequency range of dynamical processes covers the range of high-frequency fields of nuclear magnetic resonance (NMR), quadrupole. ferromagnetic, electron paramagnetic, and optical resonances. These perturbations are detected by measuring changes in gamma-resonance responses, which manifest themselves in distortions of the Mössbauer spectrum, such as shifts and the broadening of lines, and the emergence of satellite lines, the collapse of the hyperfine structure of lines under rapid magnetization reversal of ferromagnets, as well as in the onset of quantum beats caused by the interference of nuclear states.

But the intensities of the existing sources of gamma radiation (natural or synchrotron) are much lower than the values needed for producing inverse population in isomeric nuclear states. One encouraging fact here is the recently discovered nuclear excitation of ^{197}Au caused by an electron transition stimulated by the photoionization of the K-shell by X-ray radiation from a synchrotron source [3].

The modern method of theoretical analysis of gamma-resonance processes has had a great impact on the development of modulation gamma-resonance spectroscopy. The method combines the solution of the Maxwell equations in a medium and of the equations for the nuclear density matrix and showed its usefulness in examining processes accompanying the transmission of gamma radiation [4, 5]. However, to describe gamma-resonance scattering, I used a method for solving equations for the density matrix with second quantization of the gamma-radiation field [6, 7]. This approach has made it possible to solve problems related to the effect of alternating fields and relaxation on gamma resonances.

2. Ultrasonic modulation of gamma radiation

Already in their first Mössbauer experiments, Ruby and Bolef [8] recorded frequency modulation of gamma radiation with satellites whose distances from the spectral lines were found to be integer multiples of the ultrasonic frequency of the vibrating source or absorber of gamma radiation. However, the situation with a thick vibrating Mössbauer absorber was still unclear. The theory developed in Refs [1, 9, 10] showed that when the absorber or source vibrates with a frequency Ω, the intensity of gamma radiation is given by

$$P = \sum_{n=-\infty}^{\infty} J_n^2(k_1 a)\,\mathrm{Tr}\Big\{\exp\big(\mathrm{i}k_1 z\hat{b}_n\big)\,\hat{\chi}\exp\big(-\mathrm{i}k_1 z\hat{b}_n^{\dagger}\big)\Big\}, \quad (1)$$

where k_1 is the wave number of the gamma radiation, a is the amplitude of the ultrasonic vibrations, z is the thickness of the absorber, J_n is the nth Bessel function, $\hat{b}_n$ is the tensor refractive index for a gamma wave with a shift in the gamma-radiation frequency by $n\Omega$, with Ω being the ultrasonic frequency, $\hat{\chi}$ is the polarization density matrix of the incident gamma radiation, and † stands for Hermitian conjugation.

If the source and absorber vibrate simultaneously with the same frequency and amplitude, the argument $k_1 a$ of the Bessel function can be replaced with the expression

$$w = 2k_1 a \sin\left\{\frac{1}{2}\left[\varphi_1 - \varphi_2 + \frac{\Omega}{2}\,(z_2 - z_1)\right]\right\}, \quad (2)$$

where $z_2 - z_1$ is the distance between the source and absorber and $\varphi_2 - \varphi_1$ is the difference between their ultrasonic phases.

These results were corroborated by subsequent theoretical works [11–13] and in experiments [14].

It follows from general considerations that ultrasonic vibrations cannot alter the polarization of gamma radiation. This fact, corroborated by experiments in analyzing the polarization properties of ultrasonic satellites [15], formed the basis for separating the effect of gamma magnetic resonance (GMR) from magnetostriction ultrasonic vibrations excited by a radio-frequency magnetic field (RFMF) [16]. We note that by this time (1982), ultrasonic modulation induced by an RFMF due to magnetostriction in ferromagnets became a widely accepted method in modulation gamma-resonance spectroscopy, work on which began in 1968 [17, 18]. In this connection, Baldokhin et al. [19] studied the ultrasonic modulation of the Mössbauer spectrum of the yttrium iron garnet (YIG) excited by an RFMF. The behavior of the YIG Mössbauer spectrum and its dependence on the size and direction of the constant magnetic field was explained on the basis of the assumption made in Ref. [19] that the vibrations of the walls of magnetic domains generate sound. Later, this mechanism was developed in Refs [20 – 22] and other works.

The next step in studying ultrasonic modulation was made in the experiment by Mishory and Bolef [23], in which the researchers discovered the effect of suppression of gamma-resonance self-absorption for high-powered ultrasound [23]. A theory in which the main role is played by relative (rather than absolute) strain (Fig. 1) produced results that were in good agreement with the data of this experiment (see Ref. [24]). It accounted for the spatial distribution of a standing ultrasonic wave over the thickness of the Mössbauer absorber. For a steady observation mode, it was assumed that the magnetic field strength of the gamma radiation can be represented as

$$h^p = \sum_{n=-\infty}^{\infty} h_n^p(z)\, \exp(\mathrm{i}n\Omega t)\,, \tag{3}$$

where the subscript $p = \pm 1$ determines the polarization of the left- and right-hand polarization components of the magnetic field strength of the gamma wave (the 14.4 KeV magnetic gamma transition in ^{57}Fe nuclei).

Then, if we move to the local harmonic system of coordinates via the transformation

$$h_n'^p(z) = \sum_{n'=-\infty}^{\infty} J_{n+n'}\left[k_1 a(z)\right] h_{n'}^p\,, \tag{4}$$

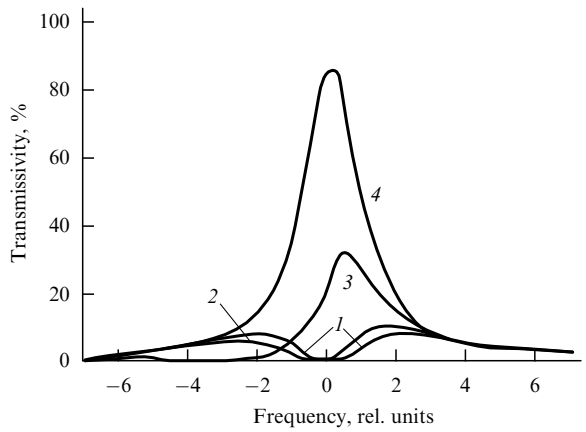

Figure 1. Frequency spectra of gamma-resonance self-absorption as functions of the ultrasound amplitude: *1*, 0; *2*, 0.5; *3*, 5; and *4*, 50 [24].

we obtain a system of equations describing the spatially inhomogeneous ultrasonic modulation of gamma radiation:

$$\frac{\mathrm{d}h_n'^p}{\mathrm{d}z} + \frac{1}{2}\, k_1 \frac{\mathrm{d}a(z)}{\mathrm{d}z}\, (h_{n+1}'^p - h_{n-1}'^p) = \mathrm{i}k_1 \sum_{p'=\pm 1} b_n^{pp'} h_n'^{-p'}\,, \tag{5}$$

where $b_n^{pp'}$ is the tensor refractive index of the nth harmonic.

The solution of system of equations (5) depends on the gradient of the index of ultrasonic modulation of gamma radiation, $k_1\, \mathrm{d}a/\mathrm{d}z$; this dependence allows developing the method known as ultrasonic Mössbauer introscopy [25, 26].

Further progress in the method of ultrasonic modulation of gamma resonance amounted to developing a new technique for detecting ultrasonic oscillations of gamma radiation [27]. A 'time – amplitude' converter was used to record quantum beats of gamma radiation. Initially, the theory proposed in [28], which dealt with the interference of gamma radiation, was described by a fairly complex formula. Later, however, this dependence was written in a form more compact (and more general) than in Ref. [28]; the formula for the intensity of the gamma radiation passing through a Mössbauer absorber also allowed for the hyperfine structure of Mössbauer nuclei and an arbitrary polarization of the incident radiation [29]:

$$P = \sum_{n,n'} \exp\left[\mathrm{i}n\,(\Omega t + \varphi)\right] J_{n'}(ka)\, J_{n'-n}(ka) \times \mathrm{Tr}\left\{ \exp\left(\mathrm{i}kz\hat{b}_{n'-n}\right) \hat{\chi} \exp\left(-\mathrm{i}kz\hat{b}_{n'}^{\dagger}\right)\right\}. \tag{6}$$

We note that much experimental work went into studies of ultrasonic quantum beats: research on ^{129}I nuclei [30], analysis of spectra in the presence of relaxation [31], determination of the hyperfine parameters of Mössbauer nuclei [32], observation of coherent transition effects [33, 34], and the study of the effect of the absorber thickness on the phase of quantum beats [35, 36].

Very short gamma radiation pulses have been used to detect the gamma-echo effect, caused by the interference of the incident radiation and the ultrasonic modulated radiation. The latter develops much more slowly and is generated in a thick absorber [37, 38].

3. Gamma magnetic resonance

Gamma magnetic resonance, or GMR, played a key role in the development of modulation gamma-resonance spectroscopy. Before this effect was discovered, Bitter [39] and Pryce [40] studied radio-optical double resonance and predicted the possibility of observing magnetic resonance on the Zeeman electronic levels of the excited states of atoms populated by optical pumping. The same researchers showed that in a strong RFMF, the electron levels split into sublevels with the energy separation proportional to the RFMF strength. This theoretical work, successful experiments that corroborated these results, and the discovery of the Mössbauer effect prompted Hack and Hamermesh [41] to calculate the probability of a radiative transition from an excited state to the single-level ground state. The researchers also confirmed the possibility of RFMF splitting of nuclear Zeeman levels, but without estimating the possibility of observing this effect in experiments. Apparently for this reason, the first experiments by Matthias [42] on the effect of a resonant RFMF on the Mössbauer spectrum were fraught with errors, since the observed effect was due to ultrasonic modulation of gamma radiation caused by magnetostriction of the sample [43].

In 1966, the theory of gamma magnetic resonance came into being [44]. Such resonance is the effect of the excitation of Mössbauer gamma-resonance transitions between the Zeeman nuclear sublevels of the ground and excited states that occur simultaneously with NMR transitions stimulated by an RFMF inside isomeric states. The theory made it possible to calculate, in the two-photon approximation, the gamma photon and the RFMF photon, the cross section of Mössbauer absorption in a fine powder of metallic iron. A key factor was an estimate of the possibility of observing the GMR effect with the hyperfine enhancement of the RFMF on a nucleus in ferromagnets taken into account. Later, Heiman et al. [45] detected the GMR effect by using the gamma-resonance scattering method with the same sample, a fine powder of metallic iron, for which the possibility of observing GMR had been predicted earlier [44]. Yakimov et al. [46] and Vagizov et al. [47] observed GMR in paramagnets. But most informative were the experiments with ferromagnets in which gamma radiation was transmitted through an absorber (^{57}Fe in α-Fe) [48 – 50] and with iron – nickel alloys [51, 52]. Here, the researchers observed not only the quasienergy line broadening [48] but also line splitting [49 – 52] (Fig. 2).

Polarization methods for detecting GMR have also been developed. An earlier theoretical paper [53] studied the change in the polarization of gamma radiation caused by GMR excitation. Later, Chugunova and I proposed using the nuclear Faraday effect, birefringence, and the Malus law to observe GMR [54 – 56], and this was implemented by Leksin et al. [16] in an experiment in which GMR was detected on ^{57}Fe nuclei in iron in the source and absorbed with split hyperfine sublevels in constant magnetic fields.

Various aspects of observing the GMR effect in paramagnets in conditions of electron relaxation were studied in Refs [57, 58]. As a result of analysis, it has been found that when the electron relaxation rate is comparable to the spontaneous Mössbauer linewidth, the GMR effect weakens substantially. Hence, the experiments in which GMR was observed in paramagnets were conducted by Vagizov et al. [47] and Leksin et al. [48] at liquid nitrogen temperatures.

Meanwhile, the theory of the GMR effect was also being developed. It was found that stimulation of GMR is accompanied by magnetic quantum beats at frequencies that are integral multiples of the RFMF frequency [59]. Later, their spectra were thoroughly analyzed in [60] (Fig. 3). To describe the effect of GMR absorption in the case of a thin absorber, the following formula was derived:

$$P \sim \mathrm{Re} \sum_{Q=-2L}^{2L} \exp\left[\mathrm{i}Q(\Omega_1 t + \varphi)\right] \mathrm{Tr}\left\{\hat{\chi}\hat{B}_{Q,0}\right\}, \tag{7}$$

$$B_{Q,0}^{pp'} = \sum_{g_1,e_1} \sum_{M=-L}^{L} (pp')^{\varepsilon} \exp\left[\mathrm{i}(p-p')\psi\right] d_{p,M-Q}^{(L)}(\theta)\, d_{p',M}^{(L)}(\theta) \times \frac{S_{g_1,e_1}^{M-Q}(\beta_g,\beta_e)\, S^M(\beta_g,\beta_e)}{\mathrm{i}[e_1 a_e - g_1 a_g + M\Omega_1 - \Delta_1] + \Gamma/2}, \tag{7a}$$

$$S_{g_1,e_1}^{M-Q}(\beta_g,\beta_e) = \sum_{g,e} d_{g_1,g}^{(I_g)}(\beta_g)\, C\,(I_g L I_e; g, M-Q, e)\, d_{e,e_1}^{(I_e)}(-\beta_e). \tag{7b}$$

Here, $a_r \sin\beta_r = \omega_{1r}$ and $a_r \cos\beta_r = \omega_{0r} - \Omega_1$, with $r = g, e$; the superscript $\varepsilon = 0, 1$ determines the electric and magnetic gamma transition, respectively; Γ is the width of the spectra

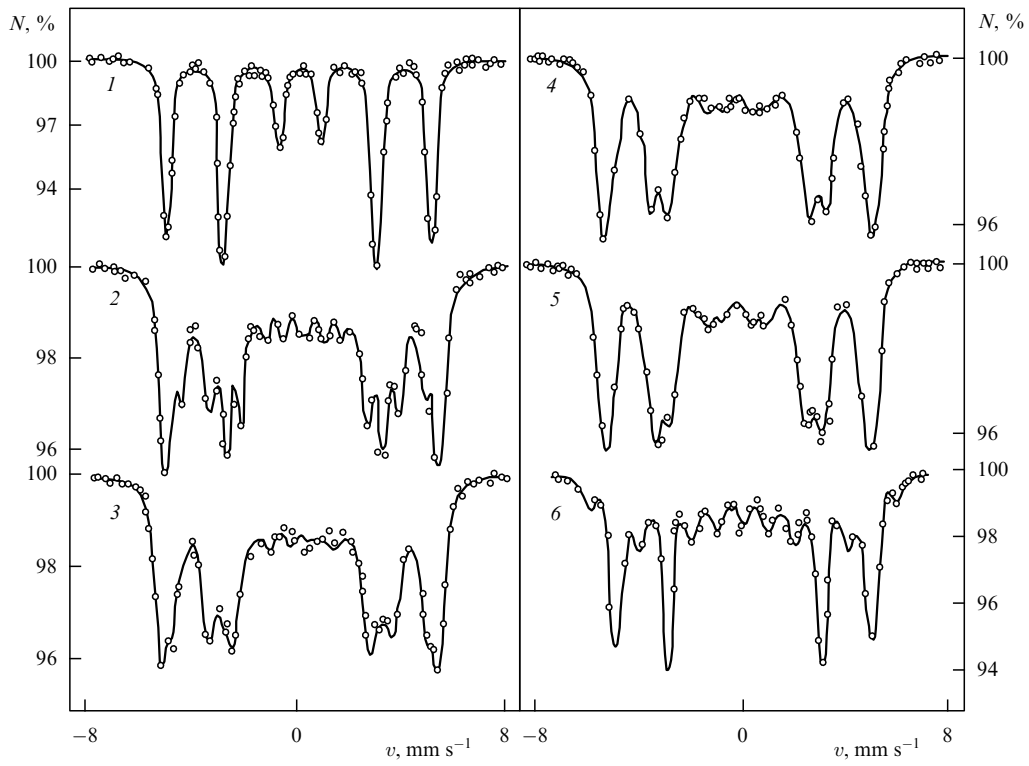

Figure 2. Mössbauer spectra for an α-Fe sample in the presence of a constant magnetic field at different frequencies [49]: *1*, $\nu = 0$; *2*, $\nu = 20$ MHz; *3*, $\nu = 22$ MHz; *4*, $\nu = 24$ MHz; *5*, $\nu = 26$ MHz; and *6*, $\nu = 36$ MHz; $H_1 \approx 25-30$ Oe; N is the relative gamma-photon counting rate.

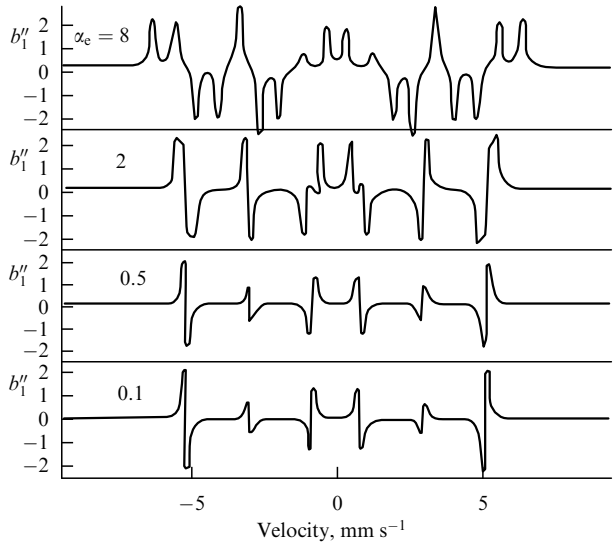

Figure 3. Doppler spectra of the imaginary part of the first GMR harmonic [60]. The dependence on the effective field on the nucleus is $\alpha_e = 2g\beta_N H_1/(\hbar\Gamma)$, and b_1'' is the imaginary part of the pseudosusceptibility of the first harmonic of the gamma-radiation intensity.

lines of the Mössbauer absorber; and the subscripts e and g denote the quantum numbers of the excited and ground isomeric states of the nucleus, respectively. The notation for other symbols is standard (e.g., see Ref. [61]). Equation (7) shows that the maximum order of the harmonics of the magnetic quantum beats in this absorber is limited by $2L$, where L is the multipole order of the gamma transition, in contrast to, generally speaking, the unlimited order of harmonics of the quantum beats in the case of ultrasonic modulation.

Equations (7), (7a), and (7b) have been derived under the assumption that the gamma radiation and the RFMF act on the ground and excited states of the nucleus simultaneously due to the coherent nature of the forward-scattered gamma wave. Hence, interference of the magnetic quasienergy isomeric state occurs. The problem was in the approximation of a resonant rotating field, and the effect should manifest itself when the g-factors of the isomeric states are of the same sign and the Rabi frequencies are comparable to the difference between the Larmor frequencies [61 – 63]. Voitovetskii et al. [64] were able to observe the isomeric interference effect in their experiments with GMR on ^{181}Ta nuclei in tantalum. Recently, it was found (see Ref. [65]) that the intensity of the first harmonic with GMR excited on ^{181}Ta nuclei is at its maximum when the RFMF frequency is selected in the interval between the values of the nuclear Larmor frequencies of the ground and excited states.

When the nuclear g-factors in the ground and excited states of the nucleus have different signs, as in the case of ^{57}Fe, isomeric interference emerges due to the components of the oscillating RFMF that rotate in opposite directions [61], with the effect caused by the quantum transitions for which the magnetic quantum numbers obey the condition $e + g = 0$. A theory of GMR scattering was developed much later than the experimental data were gathered. The first works were Refs [66, 67], but a complete theory [68–71] emerged later. A more general theory was built in Refs [70 – 72], from which the formulas describing radio-optical double resonance (among other things) follow [73]. In Refs [70, 71], I described polarization of gamma radiation and calculated the spectra of GMR scattering. Interference of the scattering amplitudes of quasienergy states has also been discovered [2, 74]. Such interference manifests itself much more strongly than the interference of the hyperfine scattering amplitudes. This research was continued by Sadykov et al. [75, 76], who studied quantum interference in the electron – nucleus system of levels.

Today, there is an upsurge of interest in the problem of using the interference of quantum states that are induced by high-frequency fields with the aim of creating conditions for resonance transparency of the medium [77]. However, in contrast to the intensities of the optical range of electromagnetic radiation, in which substantial population of the excited states can be achieved due to the existence of high-power lasers, the intensities of natural and synchrotron radiation of gamma radiation are too low for the isomeric nuclear states to become populated. At the same time, Mössbauer nuclei, which carry a magnetic moment, can interact very strongly with an RFMF by exciting GMR. The problem therefore emerged of stimulating GMR by a bi-radio-frequency coherent field with frequencies that simultaneously and resonantly excite the ground and excited isomeric states of the nucleus.

Using a ^{57}Fe nucleus in iron as an example, Roganov and I examined (see Ref. [78]) the situation where the gamma-radiation frequency was shifted by the half-sum or half-difference of the frequencies of a bi-radio-frequency field. Later, this condition was replaced by a more general one, which was determined by the structure of the symmetry group of the harmonics of the spin states of gamma transitions [79].

Analysis showed that in a thick absorber, the resonance spectrum has a much more distinct structure of the quantum-beat harmonics than in the case of time-independent detection (Fig. 4). Preliminary studies of excited GMR harmonics on ^{181}Ta nuclei in tantalum but at a single RFMF frequency also exhibit a large resonant transparency of the Mössbauer medium for quantum beats. Hence, the effect of quantum beats stimulated by GMR may find applications in integrated gamma-optics elements and, similarly to X-ray radiation [80], in developing circuits in which control and amplification are done by gamma radiation.

One of the possible areas in which GMR can be used is the study of disordered magnetic materials. The need to develop such methods stems from studies of magnetic amorphous and nanocrystalline materials whose Mössbauer spectra are characterized by strong inhomogeneous broadening and are similar for different chemical compositions. The idea of using GMR consists in the following [62, 63]. Gamma radiation from a source with a natural linewidth Γ interacts only with a small fraction of nuclei that are in an inhomogeneous hyperfine magnetic field with the width $\sigma \gg \Gamma$. The pumping RFMF H_1 acting on the nucleus forms an effective field with the amplitude

$$H_{\text{eff}} = \left[\left(H_{\text{hf}} - \frac{\hbar\Omega_1}{g_r\beta_N}\right)^2 + H_1^2\right]^{1/2}, \tag{8}$$

where H_{hf} is the hyperfine field on the nucleus, g_r is the g-factor of the excited ($r = e$) or ground ($r = g$) states, and β_N is the nuclear Bohr magneton. The frequency Ω_1 of the strong RFMF is resonant only for a specific spin packet. Hence, only for the given spin packet does there emerge a quasienergy system of levels. The weak scanning field H_2 with a frequency

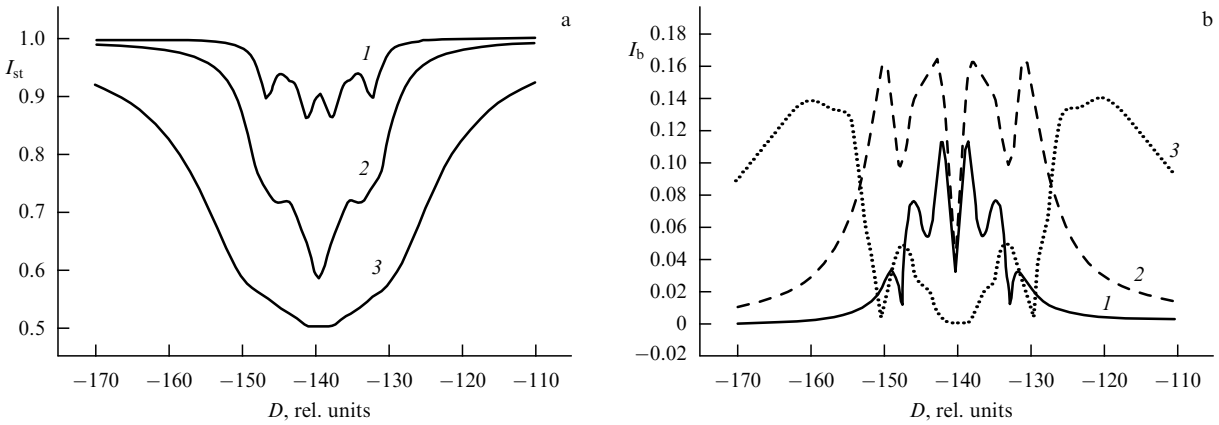

Figure 4. GMR bi-radio-frequency spectra, the symmetric case. Dependence on the thickness s of the absorber [79]: (a) time-independent intensity (curve *1*, $s = 1$; curve *2*, $s = 10$; and curve *3*, $s = 100$). (b) Intensity of the b-harmonic (curve *1*, $s = 1$; curve *2*, $s = 10$; and curve *3*, $s = 100$). D is the frequency tuning of gamma radiation in units of $\Gamma/2$.

Ω_2 is capable of exposing these quasienergy states. A further modification of this method [81] was based on using a low-frequency modulation of the amplitude of the pumping RFMF (transverse modulation) and of the constant magnetic field (longitudinal modulation). Analysis has shown, for instance, that the spectrum of the second harmonic at the frequency Ω_2 has narrow lines despite a substantial spread of the hyperfine fields [82, 83] (Fig. 5).

The results of these studies are awaiting their application in the case of synchrotron gamma radiation. Further progress is expected as these methods are extended to nanostructures.

In concluding this section, I note the possibility of using the method of GMR magnetic quantum beats for verifying the T-invariance of nuclei with mixed M1–E2 gamma transitions [84].

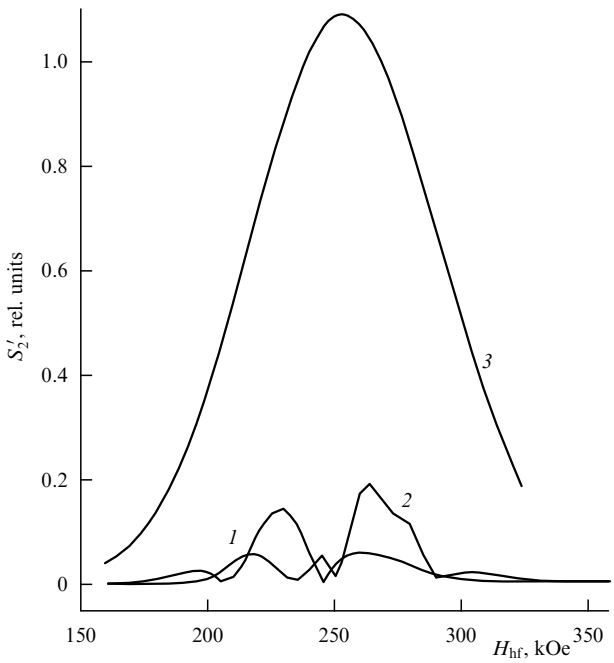

Figure 5. Distribution of the square of the absolute value of the second modulation harmonic, integrated over the frequency spectrum of gamma radiation, as a function of the hyperfine field strength H_{hf} ($\omega_{0e}/2\pi = 19.91$ MHz, $\omega_{1e}/2\pi = 2.5$ MHz, and $\Omega_2 = 3$ MHz) at $\Omega_1/2\pi = 17.91$ MHz (curve *1*) and at $\Omega_1/2\pi = 21.91$ MHz (curve *2*). Curve *3* is the Gaussian function $g(H_{hf})$ of the distribution of the hyperfine field in an amorphic metal alloy, $H_{hf}^0 = 255$ kOe [83].

4. Radio-frequency, microwave, and optical double gamma resonances

The main idea of most suggestions concerning modifications of the gamma resonance approach has amounted to simultaneous excitation of nuclear magnetic, ferromagnetic, electron paramagnetic, and optical resonances [85]. But it has been found that substantially changing a gamma resonance is fraught with many difficulties in experiments. The reason is that very powerful sources of radiation are needed to observe the effect, and low temperatures are needed to guarantee a sizable difference in populations of the ground and excited electron states. However, in ferromagnets with an RFMF whose amplitude was significantly larger than the coercive force and whose frequency was several times higher than the Larmor frequencies of the ground and excited states of the nucleus, Pfeiffer [86] observed a collapse of the hyperfine structure of the Mössbauer spectrum induced by permalloy foil in a strong RFMF field of 15 Oe. In this case, an increase in the RFMF frequency up to 106 MHz was found to lead to a collapse of the Mössbauer spectrum into a single line. The interpretation of this effect is similar to that of spectral narrowing of magnetic-resonance lines caused by molecular motion. Later, other researchers also observed the collapse of the hyperfine structure of the Mössbauer spectrum. I note that Kopcewicz et al. [87] used this effect to determine quadrupole splitting.

The effect of pulsed magnetization reversal on the gamma resonance was studied by analyzing the responses to synchrotron radiation of a $FeBO_3$ single crystal, in the geometry of Bragg scattering [88], and in the passage of radiation through the sample [89].

However, in view of the above-noted difficulties, it has proved extremely difficult to observe such effects in paramagnets by exciting EPR transitions. Nevertheless, Cherepanov et al. [90] were able to observe the broadening of K-lines of the Mössbauer spectrum of a single crystal of aluminum nitrate alloyed with iron placed in a microwave magnetic field of the frequency 9.41 GHz with the amplitude 0.7 Oe.

Earlier, theorists examined the possibility of light affecting Mössbauer spectra. The researchers assumed that such

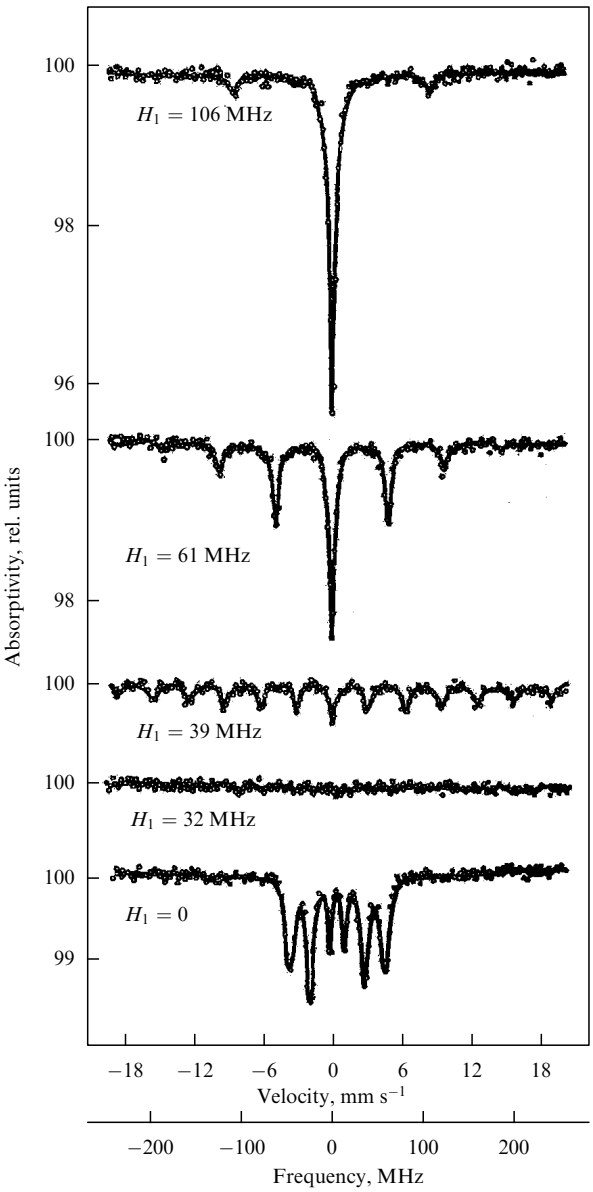

Figure 6. The RFMF collapse effect as a function of the applied frequency [86].

effects would manifest themselves in changes in the chemical shift [91], the quadrupole [92] and magnetic [93, 94] hyperfine interactions, and the presence of the laser effect of 'burning' of narrow resonances in the Doppler-broadened line of gamma transitions in gases [95].

Experimenters were able to observe an increase in the quadrupole splitting and a decrease in the isomeric shift on semiconductors [96], as well as a redistribution of intensities of the spectral lines in yttrium gallium garnets [97]. Such investigations were continued by recent experiments in which the quadrupole interaction of $^{151}Eu^{3+}$ in a number of single crystals was changed [98] and a vibrational 'ring' was recorded after a laser pulse acted on the gamma resonance of ^{57}Fe in MgO [99].

5. Conclusion

Summarizing the results of modulation gamma-resonance spectroscopy, we can say that the prospects of it becoming nonlinear gamma-resonance spectroscopy are great. However, this will not happen very soon, because powerful synchrotrons and other sources of electromagnetic radiation in the energy range from 10 to 30 keV must be built. Apparently, in the next decade, research will remain focused on the above effects of modulation gamma-resonance spectroscopy. Today, this research is being done in Russia (Kazan, Nizhni Novgorod, Moscow), USA (Texas), Belgium (Louvain), Romania (Bucharest), and other countries, which makes it possible to believe that new results and applications will come very soon.

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Methods and tools for the express immunoassay. A new approach to solving the problem

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1. Introduction

The rapid development of heterogeneous immunoassay (HetIA) techniques is facilitated by the high specificity of the antigen (Ag)–antibody (At) reaction underlain by the ability of the two molecules to recognize each other in a 'lock and key' fashion. Theoretically, it allows detecting a single molecule (particle) of the study substance in a real-time scale, provided highly sensitive assay methods are available for the purpose along with a mechanism to achieve a rapid transfer of individual Ag to the active centers immobilized at the surface. The practical solution to this problem is paramount for epidemiology, faced with the necessity of identifying causal factors of dangerous infections in multi-component environmental samples.

The proposed concept of the construction of highly sensitive systems for express immunoanalysis is first and