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New Applications of the Mössbauer Effect

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Abstract—Results from a Mössbauer experiment to observe acoustic oscillations induced by pulsed laser excitation in MgO:$^{57}Fe^{2+}$ crystal are presented. Time-domain spectra are satsifacorily described by the theory of the frequency modulation of Mössbauer radiation transmitted through a vibrating resonance medium. It is proposed that the $D_{4\Omega}/D_{2\Omega}$ ratio of the fourth and second Fourier harmonics of the modulated radiation be used to measure the amplitude of nuclear oscillations.

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INTRODUCTION

Mössbauer spectroscopy has been successfully used to obtain information on the microstructure, magnetic ordering, and chemical bonds between resonance atoms and their immediate environments. Among the many other useful applications of the Mössbauer effect that allow us to obtain information about the dynamic processes induced in a target, those of radio-frequency (RF) magnetic fields, ultrasound, and optical radiation hold a special place [1]. Interesting effects of the impact external fields have on resonance nuclei include the ultrasound [2] and magnetostrictive modulation [3–5] of gamma radiation, the RF collapse of magnetic hyperfine structure [6], quasienergy splitting of Mössbauer lines upon the excitation of nuclear magnetic resonance on the ground and excited states of the resonance nucleus [7–9], the rotation of the hyperfine magnetic field on a nucleus [10], and gamma echoes [11]. These phenomena markedly improve the efficiency of applying the Mössbauer effect to study dynamic processes and allow us to obtain information that is inaccessible using traditional experimental schemes.

For example, the ultrasound modulation of the Mössbauer radiation is effectively used to study solid-state ultrasound phenomena [12]; the RF collapse of hyperfine structure allowed the authors of [13] to recover the distribution of quadrupole splittings in alloys of soft magnetic materials; and the quasienergy splitting of Mössbauer lines was used in [14] to determine the strength of alternating magnetic fields on nuclei of resonance atoms and the coefficient of the hyperfine enhancement of an RF field on nuclei in magnetically ordered materials.

At the same time, there has been much interest in studying phenomena induced by laser radiation. This interest is due to the possibility of extending the concepts of quantum optics with respect to interference effects in multi-level coherently excited electron systems to the regions of nuclear systems and gamma-range radiation [15, 16]. Considerable changes in Mössbauer spectra, including the suppression of the resonance absorption, changes in the width of lines and their shifts, and the emergence of additional lines and the splitting of existing lines were predicted in [15]. Experimental studies performed on optical crystals doped with ^{151}Eu and ^{57}Fe nuclei revealed considerable changes in the shape of the Mössbauer spectrum of ^{151}Eu ions and the emergence of additional hyperfine structure in the case of ^{57}Fe ions subjected to optical pumping, due to the redistribution of the population of electronic sublevels under the action of resonance laser radiation [16].

Excitation of the solid state by pulsed laser radiation can produce effects associated with the generation of mechanical acoustic oscillations of a studied sample. Different characteristic elastic modes can arise when a pulsed laser beam falls on a solid surface as a function of the depth of radiation penetration, thermal diffusion, the elastic and geometric properties of the material, and the parameters of the laser pulse (e.g., intensity, shape, duration, and focal spot size). The action of high-intensity pulsed laser radiation results in strong thermal and hydrodynamic deviation from the equilibrium state of the irradiated region of the medium [17]. For the average optical energy densities released in the medium, the main factor in generating sound is the thermal expansion of the irradiated volume of the material. Other mechanisms for the optical generation of ultrasound waves in the region of units of GHz and higher using schemes of excitation with crossed pulsed lasers are also known [18].

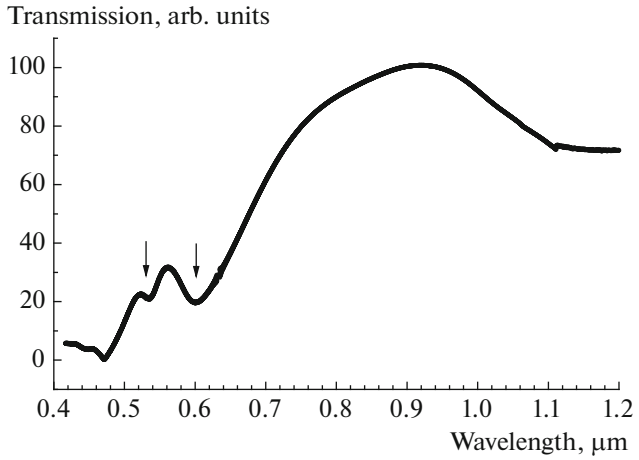

Fig. 1. Optical absorption spectrum of MgO:$^{57}Fe^{2+}$.

The study of ultrasound oscillations induced by the pulsed laser radiation allows us to obtain useful information about the physical properties of a material, along with the dynamics of phase transitions and the photoinduced chemical reactions caused by optical radiation. Acoustic signals are usually measured using piezoelectric detectors, optical circuits, and induction coils. Mössbauer spectroscopy has yet to be used to study ultrasound oscillations caused by the optoacoustic effect. In earlier experiments, the ultrasound modulation of Mössbauer radiation was mainly performed by exciting piezoelectric or magnetostrictive oscillations via radio frequency fields. At the same time, there is a whole class of optoacoustic phenomena that could be effectively studied using the Mössbauer effect.

In this work, we present the results from an experiment to observe the modulation of Mössbauer radiation using pulsed laser excitation.

EXPERIMENTAL

Our experiment was performed in transmission geometry with a ^{57}Co source in a Rh matrix. A rectangular plate of MgO:$^{57}Fe^{2+}$ with a wide optical absorption band in the region of ~530 and ~600 nm was used as the absorber. Figure 1 shows the optical absorption spectrum of a sample. The arrows indicate the positions of the absorption peaks. Optical pumping of the sample was performed using a pulsed Nd:YAG laser (Verdi-V18, Coherent) with a radiation wavelength of 532 nm. The laser beam power was 1.85 W, and the optical pulse duration was around 1 μs at the repetition frequency of 10 kHz.

Figure 2 shows the Mössbauer spectrum of (a) an unperturbed MgO:$^{57}Fe^{2+}$ sample and (b) the spectrum obtained from the pulsed laser radiation's effect on it. It is seen that the action of the pulsed laser radiation resulted in considerable broadening of the absorption

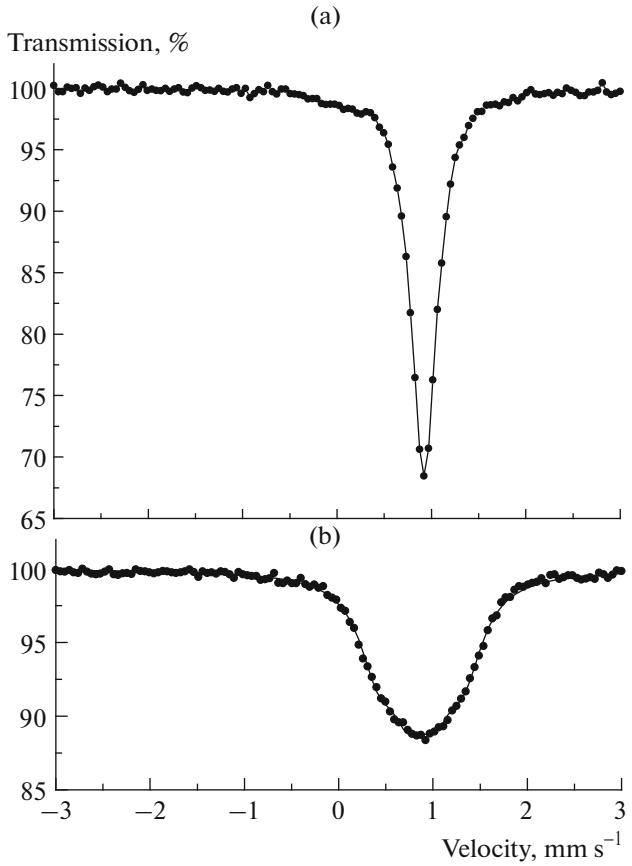

Fig. 2. Mössbauer spectrum of the MgO:$^{57}Fe^{2+}$ sample (a) with no laser pumping and (b) upon its impact.

line. This was due to the optoacoustic effect in the Mössbauer spectrum. The absorption of the optical radiation energy and the subsequent thermal expansion of the irradiated volume led to crystal deformation and the excitation of acoustic oscillations.

It is known that the amplitude of an acoustic signal is proportional to the variable part of the light flux [17]. To excite acoustic oscillations, we used laser pulses of exponential shape with a decay constant of around 160 ns. The rectangular MgO: $^{57}Fe^{2+}$ plate was fixed in the holder of the sample so that it could excite its own mechanical oscillations under the action of pulsed laser radiation. Under these conditions, nuclear oscillations with frequency Ω were observed in the Mössbauer spectra in the form of sets of symmetric satellites at a distance of $\pm n\hbar\Omega$, where n is an integer, from the central initial absorption line. The amplitude of the satellites depends on that of the nuclear oscillations (i.e., modulation index m). When all nuclei of the sample oscillate coherently with the same amplitude, the intensity of the satellites is determined by a quadratic Bessel function of the first kind, $J_n^2(m)$. When the amplitude of the nuclear oscillations obeys the

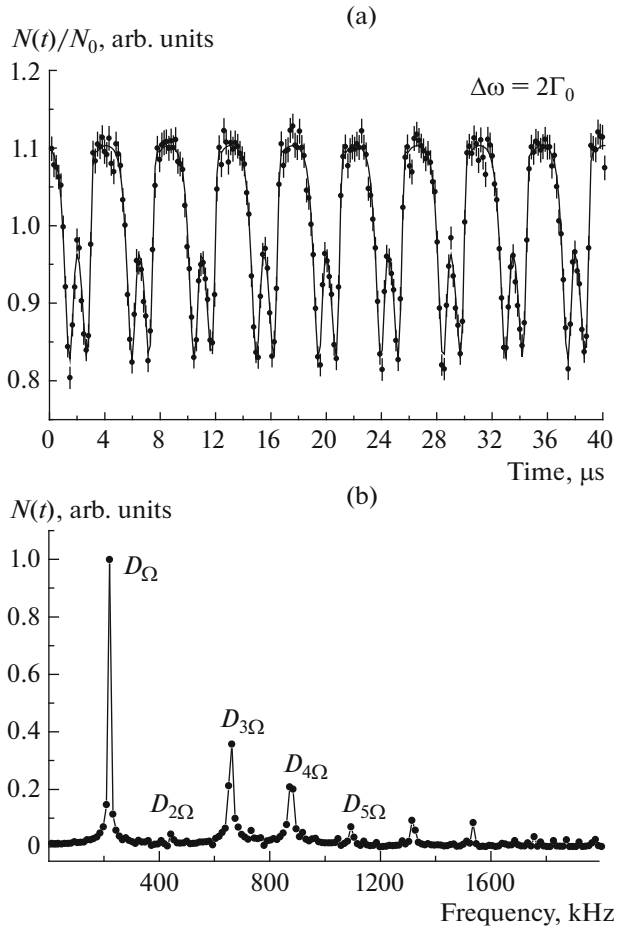

Fig. 3. Time-domain dependence of (a) the normalized intensity of the resonance radiation transmitted through a vibrating absorber and (b) its Fourier spectrum. $D_{n\Omega}$ is the Fourier component of the time domain spectrum. The detuning from resonance is $\Delta\omega = 2\Gamma_0$.

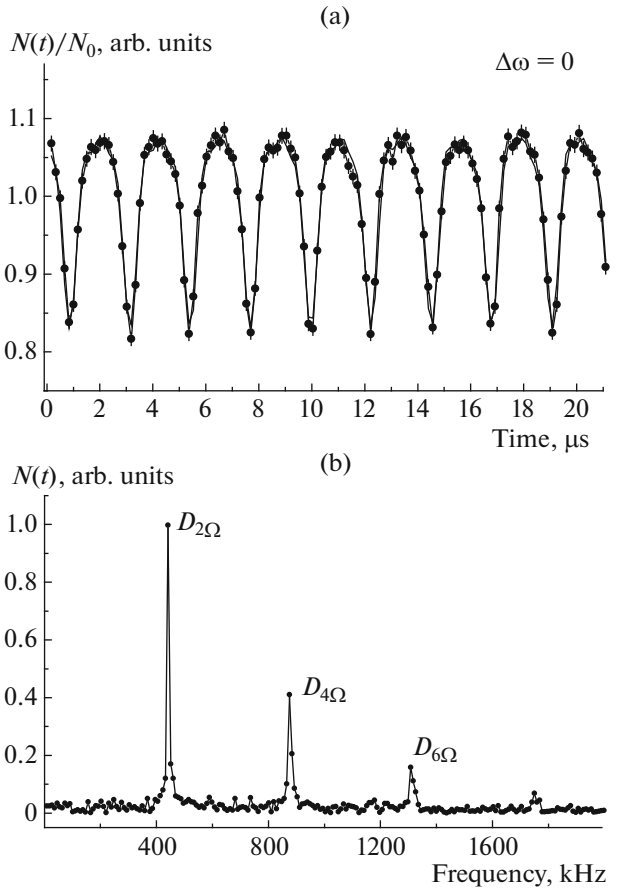

Fig. 4. Time-domain dependence of (a) the normalized intensity of resonance radiation transmitted through a vibrating absorber and (b) its Fourier spectrum. The radiation of the source was tuned to the resonance with the absorption line of the absorber ($\Delta\omega = 0$).

Rayleigh distribution, that of the satellites is proportional to modified Bessel functions $e^{-m^2} I_n(m^2)$.

If frequency Ω of nuclear oscillations is less than the natural line width of resonance transitions Γ_0 (for ^{57}Fe nuclei, $\Gamma_0 \approx 1.1$ MHz), modulation of the resonance radiation is apparent in the Mössbauer spectrum only as absorption line broadening (Fig. 2b). This greatly reduces the statistical accuracy of determining the frequency and amplitude of oscillations induced by the pulsed laser radiation when making traditional Mössbauer measurements. In this situation, delayed-coincidence time-domain measurements, in which the number of resonance gamma-photons transmitted through a vibrating absorber is recorded as a function of time elapsed since the sample's excitation by pulsed laser radiation, are more informative. Time-domain measurements were made using a time–amplitude converter operating in the start–stop mode. The basic scheme of the equipment was given in [19]. The start signals for the converter were formed by a photodiode at the time of laser irradiation, and the stop signals at the time the resonance (14.4 keV) photon was recorded.

Figures 3 and 4 show the time-domain spectra of the normalized intensity of radiation transmitted through a absorber subjected to pulsed laser radiation. Figure 3a shows the time-domain spectrum obtained upon moving the source with a velocity detuned from the resonance by $\Delta\omega = 2\Gamma_0$. The spectrum shown in Fig. 4a was recorded at the exact resonance between the energy of the source radiation and that of the sample's resonance absorption ($\Delta\omega = 0$).The experimental dependence has a strictly periodic character, allowing us to determine the frequency of nuclear oscillations upon pulsed laser excitation with a high degree of accuracy.

The relative intensity of the resonance radiation transmitted through an absorber vibrating with frequency Ω was determined using the expression [20]

$$N(t)/N_0 = \mathrm{Re}\left\{1 - 2f_S b F_+(t)\int_{-\infty}^{t} dt' \frac{J_1\left[2\sqrt{b(t-t')}\right]}{F_+(t')\sqrt{b(t-t')}} + 2f_S b^2 e^{-\Gamma_a t}\int_{-\infty}^{t} dt' \frac{F_-(t')J_1\left[2\sqrt{b(t-t')}\right]}{\sqrt{b(t-t')}}\int_{-\infty}^{t'} dt'' \frac{J_1\left[2\sqrt{b(t-t'')}\right]}{F_+(t'')\sqrt{b(t-t'')}}\right\}, \quad (1)$$

where N_0 is the radiation intensity far from the resonance; f_S and f_A are the Lamb–Mössbauer factors of the source and absorber nuclei, respectively; $F_\pm(t) = \exp[-(\Gamma_S \pm \Gamma_A)t/2 - i\Delta\omega t + i\varphi(t)]$; $\Delta\omega = \omega_A - \omega_S$ is the detuning from the resonance; and $b = T_M\Gamma_0/4$ is a parameter that depends on the effective Mössbauer thickness of the absorber, $T_M = nf_A\sigma_0$. Here, n is the number of resonance ^{57}Fe nuclei per unit area, and σ_0 is the cross section of resonance absorption.

The solid lines in Figs. 3a and 4a show the results from approximating the experimental spectra using expression (1). Mathematical processing of the spectrum revealed that the Mössbauer nuclei oscillate with a frequency of 221 kHz, which is very close to that of the intrinsic mechanical oscillations of the rectangular plate, determined by the formula [21]

$$\Omega = Cc_1 h\left[(1/a)^2 + (1/b)^2\right], \quad (2)$$

where c_1 is the speed of the sound of longitudinal oscillations; a and b are the transverse dimensions of the plate; h is the plate thickness; and C is the coefficient of proportionality, determined by the condition of the plate fixing.

Integral expression (1) satisfactorily describes the experimental spectra. However, it is not sufficiently clear or convenient for determining the amplitude of the Fourier harmonics of the radiation transmitted through the vibrating absorber. The amplitudes of the Fourier harmonics of the time-domain spectrum contain useful information on the amplitude of nuclear oscillations and modulation index m. To derive the analytical dependence of the amplitude of a spectrum's Fourier harmonics on the value of the modulation index, let us consider the expression for the normalized intensity of transmitted radiation given in [22]:

$$N(t)/N_0 = \sum_{n,k=-\infty}^{+\infty} J_n(m)J_k(m)e^{i(n-k)\Omega t}B_{nk}(\Omega),$$

where

$$B_{nk}(\Omega) = \frac{\Gamma_S}{2\pi}\int_{-\infty}^{+\infty} d\omega \frac{e^{-\frac{b}{\Gamma_A/2+i(\omega+n\Omega-\Delta)}-\frac{b}{\Gamma_A/2-i(\omega+k\Omega-\Delta)}}}{(\Gamma_S/2)^2+\omega^2}. \quad (3)$$

Let us consider a case where the energy of the source radiation is in resonance with that of the absorber absorption; i.e., $\Delta\omega = 0$. Using the familiar relations between the Bessel functions and symmetry in wriring the integral in terms of coefficients $B_{nk}(\Omega)$, it is easy to show that the amplitude of the second harmonic is given by

$$D_{2\Omega}(m,\Omega) = -J_1^2(m)B_{-11}(\Omega) + \sum_{n=0}^{+\infty} J_n(m)J_{n+2}(m)\left[B_{n,n+2}(\Omega) + B_{-(n+2),-n}(\Omega)\right], \quad (4)$$

and the below expression holds for the amplitude of the fourth harmonic:

$$D_{4\Omega}(m,\Omega) = J_2^2(m)B_{-2,2}(\Omega) - J_1(m)J_3(m)[B_{-1,3}(\Omega) + B_{-3,1}(\Omega)] + \sum_{n=0}^{+\infty} J_n(m)J_{n+4}(m)\left[B_{n,n+4}(\Omega) + B_{-(n+4),-n}(\Omega)\right]. \quad (5)$$

Figure 5 shows dependences $D_{4\Omega}(m)$, $D_{2\Omega}(m)$ and their ratio $D_{4\Omega}(m)/D_{2\Omega}(m)$ on modulation index m under the conditions of exact resonance ($\Delta\omega = 0$). These dependences are useful for determining the modulation index and measuring the amplitude of resonance nuclear oscillations using time-domain spectra. This is possible because of the extreme sensitivity of the time-domain spectrum's harmonic composition to even small changes in the modulation index; i.e., to the amplitude of nuclear oscillations x_0, since $x_0 = m\lambda$.

The experimental spectra presented in Figs. 3a and 4a were approximated using the fixed parameters: $f_S = 0.76$, $\Gamma_S = \Gamma_0$, $\Gamma_A = 2.1\Gamma_0$, and $T_M = 2.2$. The best fit was achieved when variable parameter Ω was 221 kHz, and modulation index $m \approx 16$. The obtained value of Ω is in good agreement with the frequency of the first harmonic of the Fourier spectrum (Fig. 3b) obtained for the detuning from the resonance equal to $\Delta\omega = 2\Gamma_0$, and with the value of the frequency of intrinsic mechanical oscillations of the MgO plate, calculated using formula (2). Fourier harmonic $D_{2\Omega}$ with frequency 2Ω is the fundamental harmonic in the time-domain spectrum (Fig. 4a) and its Fourier decomposition (Fig. 4b) if the energy of the source radiation coincides with that of the sample's absorption line ($\Delta\omega = 0$). The time-domain dependence of the transmitted radiation determined using formula (1) has a strictly harmonic form at low amplitudes of

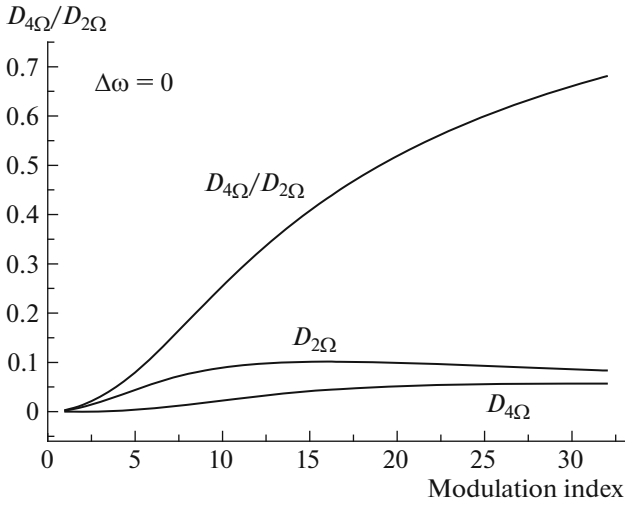

Fig. 5. Dependence of even harmonics $D_{2\Omega}(m)$, $D_{4\Omega}(m)$ and their ratio $D_{4\Omega}(m)/D_{2\Omega}(m)$ on modulation index m for the exact resonance between the energy of the source's radiation and the absorption energy of the sample ($\Delta\omega = 0$).

nuclear oscillations ($x_0/\lambda = m < 1$). Upon an increase in the amplitude of oscillations, however, the shape of the spectrum is distorted and higher-order harmonics appear (Fig. 4b). Detuning from the resonance by $\Delta\omega \neq 0$ leads to the emergence of odd harmonics Ω, 3Ω, and 5Ω in the spectrum, along with even harmonics. The larger this deviation, the greater the contribution from fundamental harmonic Ω. These changes are due to the incident radiation interfering with the components of radiation scattered forward [22, 23].

It was shown earlier that using the ratio $D_{2\Omega}/D_{\Omega}$ of the Fourier components, we can determine the energy shift of Mössbauer lines with a high degree of accuracy [19]. Our studies show that ratio $D_{4\Omega}/D_{2\Omega}$ of the Fourier components can be used to determine the amplitude of nuclear oscillations. With exact resonance ($\Delta\omega = 0$), the dependence of ratio $D_{4\Omega}/D_{2\Omega}$ of Fourier components on modulation index m has a monotonic character (Fig. 5). The dependence given in this figure was calculated for frequency of oscillations $\Omega = 221$ kHz and thickness parameter $b = T_M \Gamma_0/4 = 0.572$. Experimental ratio $D_{4\Omega}/D_{2\Omega}$ of the amplitude of even Fourier harmonics of the spectrum shown in Fig. 4b is 0.43. Using this value of $D_{4\Omega}/D_{2\Omega}$ and the dependence given in Fig. 5, we find that the modulation index is in this case 16.

It should be noted that the proposed way of determining the amplitude of nuclear oscillations can also be used to study more complicated optoacoustic and magnetoelastic interactions induced by pulsed laser radiation. The gamma-resonance technique of measuring the frequency and amplitude of induced oscillations allows us to obtain additional information on the physical properties of objects and processes resulting from laser irradiation.

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