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Mössbauer forward scattering: time-domain spectra

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Abstract The transmission of the Mössbauer radiation through an absorber being in the acoustic oscillation mode under forward scattering (FS) conditions has been analyzed. The modification of the existing models of the FS spectra (frequency and time) formation to the case of the arbitrary phase correlation of nuclear oscillations in the sample has been proposed. An adequate description of the time delayed experiments with the ^{57}Fe Mössbauer resonance using the modulation of the single-photon wave packet by acoustic field has been obtained. One has been done in the frame of the Raman scattering of Mössbauer photons. The models extended this way can be used to control the degree of phase correlation of nuclear oscillations (or other processes) induced in the sample by external fields.

Keywords Mössbauer scattering · Effect of acoustic oscillations · Time spectra · Single photon response

1 Introduction

The results of Mössbauer forward scattering experiments in conditions of external acoustic excitation of sample (target) were published first time in papers [1, 2, 4]. The frequency distribution of gamma radiation emerging from the target in the forward direction (named as FS spectrum) was found as consisting of ultrasound (US) satellites in these works. A mechanism of the formation of US satellites in the FS spectra (coherent enhancement of the forward Raman scattering) was emphasized in [4] also. The US satellite structure of FS spectra, in contrast to the US structure of Mössbauer absorption spectra [5], is formed only

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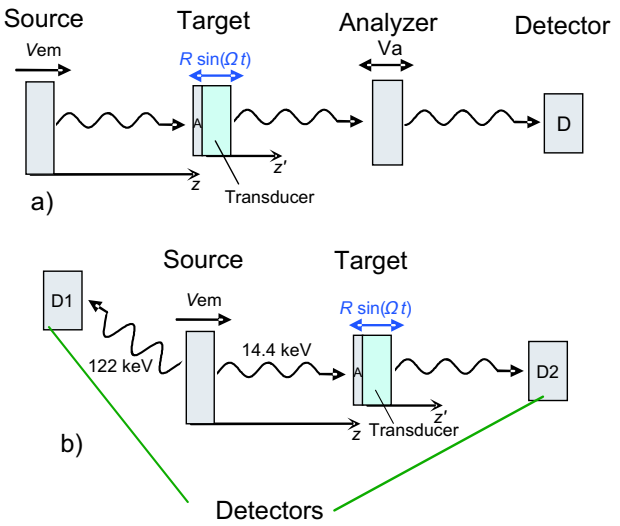

Fig. 1 The principal schemes setup for observing the FS Mössbauer spectra with a natural Mössbauer source: **a** frequency spectra and **b** time-domain spectra

for thick samples and only in the case of perfect phase correlation of nucleus oscillations. The model [4] was worked out for the case when all nuclei of the target are vibrating with the same phase, and for the case of complete dissonance, when nuclei are vibrating with arbitrary phases. The authors of [4] especially underlined the phenomenon of an unusual increase of the Mössbauer FS intensity for the strict phase correlation condition of the Mössbauer nucleus oscillations.

In this work, we propose a modification of the existing model of Mössbauer forward scattering spectra [4] to the cases of the partial phase correlation of nuclei oscillations induced in sample by the external (US) field. Such mode should be expected for samples in the general case that leads to the dependence of the shape of the Mössbauer FS spectra on the extent of the phase correlation of the corresponding processes. Last one allows to control experimentally the mechanisms of the materials excitation by external fields. We underline also another possibility of extension of the model [4]. That is the formation of the structure of the time-domain Mössbauer spectra in FS scheme as a consequence of coherent Raman scattering of gamma photons. Such spectra are shaped usually in time delayed coincidence experiments with the internal [6] or external [7] time mark. The acoustic effects under discussion are of interest as a source of additional information accessible on the basis of fitting of both types (frequency and time-domain) Mössbauer spectra. It must be noted also, the model of time-domain FS spectra is topical as a tool of simulation (fitting) of experiments on the time Mössbauer spectroscopy (as an instrument for control of the photon states in the gamma-range) [8] (Fig. 1).

We specify the above for the case of the ^{57}Fe isotope under the US excitation of the sample with a single resonance absorption line (for example stainless steel). In the expression for the oscillation of a separate nucleus, $\mathbf{u}_a(t) = \mathbf{u}_a \sin(\Omega t + \varphi_a)$, the spread of oscillation amplitudes $\mathbf{u}_a$ and of their initial phases φ_a is allowed now. The appearance of one for the US amplitudes in the sample is confirmed by numerous experiments. The satisfactory description of US Mössbauer absorption spectra has been achieved by using the distribution functions for the oscillation amplitudes of the Rayleigh [9] or Rayleigh-Rice [10, 11]

type. The Rayleigh distribution was used also at the simulation of experimental FS spectra [4]. In this work we pay special attention to the consequences of the possible spread of the initial phases (φ_a) of acoustic oscillations of nuclei. It is natural to relate the appearance of such inhomogeneity to the fluctuations caused by impurities and boundaries and also by the thermal motion in the sample. As we will see below, the phase spread may decrease the efficiency of the coherent enhancement of the forward Raman scattering intensity and leads to more considerable changes in the satellite structure of FS spectra than the amplitude spread. Quantitatively, the phase spread of nuclei oscillations in the sample can be given by a random function $\varphi_a\,(t)$ having the ergodicity property. In this work, such function is introduced by phenomenological way. We analyze the expected changes of the Mössbauer FS spectra, when the acoustic phase fluctuation process is described by the normal distribution function $R(\varphi)$

2 The effects of phase fluctuation

The random character of the initial phases of oscillations of nuclei in the absorber can be taken into account within the model of the formation of the frequency Mössbauer forward scattering spectrum [4] based on the dynamic theory of the Raman scattering of gamma-photons in the medium. According to this model, each Fourier component of the radiation of the source $E_\omega\,(0) = \varepsilon_0\,(\omega)\exp\,(-i\omega t)$ during the propagation through the oscillating medium acquires Raman satellites, the amplitudes of which $E_{\omega l}\,(y)$ vary with the target thickness y according to the system of equations (for details, see (3.19) in [4]):

$$\frac{\partial E_{\omega l}\,(y)}{\partial y} = \sum\nolimits_{l'} G_{ll'} E_{\omega l'}\,(y)\,, \tag{1}$$

with the general expression for the matrix of coefficients $G_{ll'}$

$$\begin{aligned} G_{ll'} \;=\; & -\tfrac{i2\pi f_{LM}}{c\omega\hbar(2I_g+1)} \sum_q \int \tfrac{d^3\mathbf{k}}{(2\pi)^3} \sum_{e,g} \left\langle g|\widehat{j}\,(\mathbf{k})|e\right\rangle \left\langle e|\widehat{j}^{\,*}\,(\mathbf{k}_\gamma)|g\right\rangle \\ & \times \textstyle\int_{-\infty}^{t} d\tau \exp\left[-i\left(\omega-\omega_0-q\Omega+i\Gamma_0/2\right)(\tau-t)\right] \sum\limits_a J_{q+l}\,(b_a)\,J_{q+l'}\,(b_a) \\ & \times \exp\left(-i\,(q+l)\,\varphi_a\,(t)\right) \exp\left(i\left(q+l'\right)\varphi_a\,(\tau)\right) \exp\left(i\left(\mathbf{k}_\gamma-\mathbf{k}\right)\mathbf{R}_a\right). \end{aligned} \tag{2}$$

Now however unlike [4], the oscillation phases are presented by random functions $\varphi_a\,(t)$. The matrix $G_{ll'}$ reflects the mechanism of the Raman resonance scattering of photons by nuclei $g \to e \to g$, $\mathbf{k}_\gamma$ and $\mathbf{k}$ are wave vectors of photons incident on the target and scattered in the resonance manner on nuclei, respectively, ω_0 and Γ_0 are the frequency and width of the gamma-resonance of the target, summation is performed over nuclei (a) and over the number of sound phonons of the frequency Ω participating in the Raman scattering (q), $\mathbf{R}_a$ is the equilibrium position of the a-th nucleus, $J_{q+l}\,(b_a)$ is the Bessel function. Under the equality of the oscillation amplitudes in the sample, $\mathbf{u}_a = \mathbf{u}_0$ (modulation indices $b_a = b_0$), the phase spread of oscillations of nuclei is taken into account by introducing the fluctuation of the phase for a-th nucleus as $\Delta\varphi_a\,(t) = \varphi_a\,(t) - \varphi$, where φ is the phase of the external driving field set in the time scale of the nuclear process. A fragment of expression (2) depending on $\Delta\varphi_a\,(t)$ has the form

$$\begin{aligned} & \exp\left(-i\varphi\left(l-l'\right)\right) \sum_a \exp\left(i\left(\mathbf{k}_\gamma-\mathbf{k}\right)\mathbf{R}_a\right) \exp\left(-i\,(q+l)\,\Delta\varphi_a\,(t)\right) \\ & \quad \times \textstyle\int_{-\infty}^{t} d\tau \exp\left[-i\left(\omega-\omega_0-q\Omega+i\Gamma_0/2\right)(\tau-t)\right] \exp\left(i\left(q+l'\right)\Delta\varphi_a\,(\tau)\right) \end{aligned}. \tag{3}$$

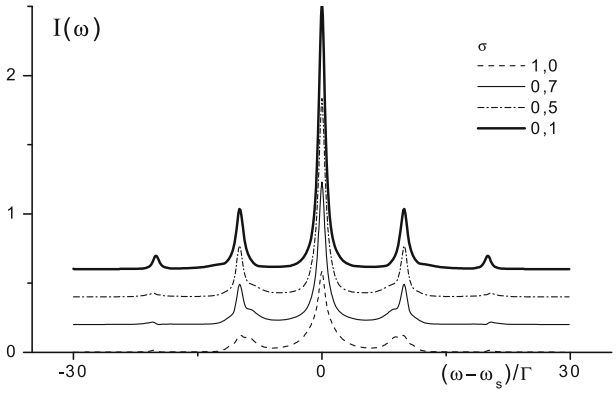

Fig. 2 Intensity of Mössbauer forward scattering $I(\omega)$ at different degrees of phase correlation of acoustic oscillations σ; $\Omega = 10$ MHz, $b = 3.05$, $\Gamma_s = 1$, $\Gamma_0 = 1$, $t_e = \sigma_0 N_0 f_{LM} Y = 50$, and $\omega_s = \omega_0$

Further we consider two modes of the time dependence of fluctuations $\Delta\varphi_a(\tau)$: slow fluctuations and fast fluctuations. The fluctuations are slow if their lifetime is larger than the lifetime of the excited nucleus. In this case, the exponent containing $\Delta\varphi_a(\tau) \simeq \Delta\varphi_a(t)$ can be taken out of integral in (3). As a result (3) becomes

$$\frac{\exp\left(-i\varphi\left(l-l'\right)\right)}{-i\left(\omega-\omega_0-q\Omega+i\Gamma_0/2\right)}\sum_a \exp\left(i\left(\mathbf{k}_\gamma-\mathbf{k}\right)\mathbf{R}_a\right)\exp\left(-i\Delta\varphi_a(t)\left(l-l'\right)\right). \quad (4)$$

We rewrite the sum in (4) introducing the average over the ensemble (triangular parentheses) as

$$\sum_a \exp\left(i\left(\mathbf{k}_\gamma-\mathbf{k}\right)\mathbf{R}_a\right)\exp\left(-i\Delta\varphi_a(t)\left(l-l'\right)\right) = (2\pi)^3 N'\delta\left(\mathbf{k}_\gamma-\mathbf{k}\right), \quad (5)$$

where

$$N' = N_0\left\langle\exp\left(-i\Delta\varphi\left(l-l'\right)\right)\right\rangle. \quad (6)$$

In the case of in-phase oscillations ($\Delta\varphi_a(t) = const$ for all a values), expression (5) is reduced to the well-known condition of the diffraction maximum (see, e.g., [12]). In the case of the phase spread ($\Delta\varphi_a(t)$) equality (5) should be considered as a phenomenological relation substantiated by the geometry (the collimation conditions) of the detection channel of the measuring setup, with the effective value N' smaller than the number of scattering centers per unit target volume N_0.

The lifetime of fast fluctuations, on the contrary, is small in comparison with the lifetime of the nucleus. It can be considered that the lifetime of the nucleus in the excited state is sufficient for $\varphi_a(\tau)$ to acquire values characteristic of a given random process. Now the average over time (over the ensemble) can be taken out of the integral in (3); as a result, we have instead of expression (5)

$$\sum_a \exp\left(i\left(\mathbf{k}_\gamma-\mathbf{k}\right)\mathbf{R}_a\right)\exp\left(-i\Delta\varphi_a(t)\left(q+l\right)\right)\left\langle\exp\left(i\Delta\varphi\left(q+l'\right)\right)\right\rangle = (2\pi)^3 N''\delta\left(\mathbf{k}_\gamma-\mathbf{k}\right);$$
$$N'' = N_0\left\langle\exp\left(i\Delta\varphi\left(q+l'\right)\right)\right\rangle\left\langle\exp\left(-i\Delta\varphi\left(q+l\right)\right)\right\rangle. \quad (7)$$

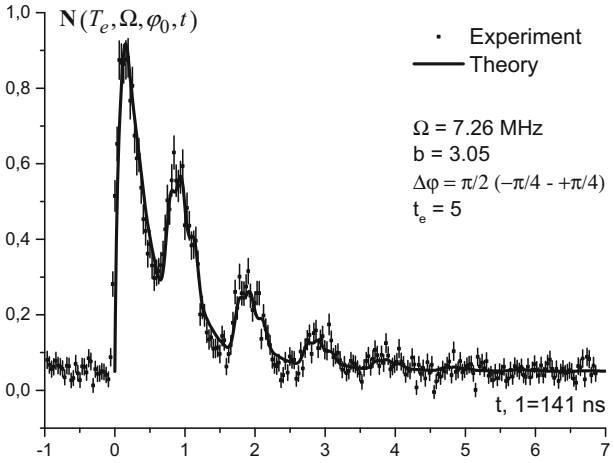

Fig. 3 Time dependence of the counting rate (see (16))$\mathbf{N}(T_e, \Omega, \varphi_0, t)$; comparison of calculations (*solid line*) with experiment (points), $\omega_s - \omega_0 = 2\Omega$, $\varphi_0 = 0$

Averaging in (5) and (7) with allowance for the stationary character of fluctuations (using the normal distribution function $R(\Delta\varphi) = \left(1 \Big/ \sigma\sqrt{2\pi}\right) \exp\left(-(\Delta\varphi)^2 \Big/ 2\sigma^2\right)$ with parameter σ) makes it possible to obtain coefficients $G_{ll'}$ in (2) for slow (a) and fast (b) fluctuations, respectively (we note that $\sigma_0$is maximum resonance cross section below):

$$G_{ll'} = -i \sum_q e^{-i(l-l')\varphi} J_{q+l}(b) J_{q+l'}(b) \frac{\sigma_0 f_{LM} N_0 \Gamma_0 e^{-(l-l')^2\sigma^2/2}}{4(\omega - \omega_0 - q\Omega + i\Gamma_0/2)}, \tag{8a}$$

$$G_{ll'} = -i \sum_q e^{-i(l-l')\varphi} J_{q+l}(b) J_{q+l'}(b) \frac{\sigma_0 f_{LM} N_0 \Gamma_0 e^{-\left((q+l)^2+(q+l')^2\right)\sigma^2/2}}{4(\omega - \omega_0 - q\Omega + i\Gamma_0/2)}. \tag{8b}$$

In view of the appearance of the phase spread of acoustic oscillations, the solution of the system of equations (1) cannot be found by the iteration method (which was used in [4]). Instead, the matrix exponential method (see [13, 14]) is used. The radiation at the absorber output ($y = Y$) due to the harmonic $\varepsilon_0(\omega)\exp(-i\omega t)$ at its input in the presence of the phase spread of oscillations of atoms can be presented as

$$E_\omega(Y, t) \sim \varepsilon_0(\omega) \sum_l e^{-il\Omega t} e^{-i\omega t} \{\exp(\mathbf{G}(\omega_a, \omega, \varphi, \sigma) \cdot Y)\}_{l0}, \tag{9}$$

where $\mathbf{G}(\omega_a, \omega, \varphi, \sigma)$ is the matrix of coefficients (8a). Using (9), one can easily determine the frequency forward scattering spectrum according to the algorithm of the calculation of such spectra (see [4, 13]). Model calculations using $|\varepsilon_0(\omega)|^2 = \left(\Gamma_s \Big/ 2\pi\right) \Big/ \left[(\omega - \omega_s)^2 + \left(\Gamma_s \Big/ 2\right)^2\right]$ with Mössbauer source parameters (ω_s, Γ_s) confirm the considerable weakening of the intensity of satellites in forward scattering spectra with increasing σ and further their complete disappearance (Fig. 2).

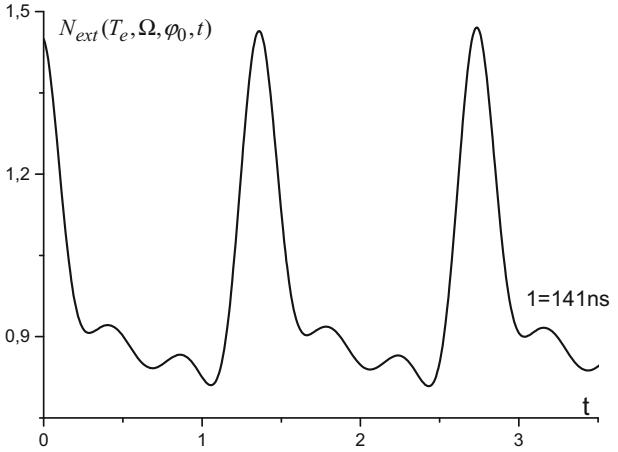

Fig. 4 The model spectrum calculated on the basis of (18) by numerical method: b=1.8, t_e =5, $\omega_s - \omega_0 = -\Omega$, $\Omega = 5.16$ MHz, $\Gamma_s = \Gamma_0 = 1$

2.1 Single-photon response experiment under acoustic excitation

Further we analyze the formation of the time-domain spectrum (the time dependence of the count rate in the detector) of the gamma radiation behind the oscillating target in time-delayed coincidence condition. It means, the mechanism of generation of Raman amplitudes of gamma radiation in the target we describe on the basis of model [4] (using (1)). It must be noted, however, that the matrix of coefficients $G_{ll'}$ in (1) must now be reconsidered for single-photon response.

The radiation field of the Mössbauer source $E(t)$ under conditions of the single-photon experiment (implemented by the delayed coincidence method) can be presented by the Fourier transformation $\bar{\varepsilon}_0(\omega)$:

$$E(t) = \Gamma_s^{1/2}\theta(t)\exp[-i\omega_s t - \Gamma_s t/2] = \int d\omega\, \bar{\varepsilon}_0(\omega)\exp(-i\omega t), \tag{10}$$

where $\bar{\varepsilon}_0(\omega) = i\theta(t)\,\Gamma_s^{1/2}(2\pi)^{-1}(\omega - \omega_s + i\Gamma_s/2)^{-1}$. The Fourier component $\bar{\varepsilon}_0(\omega)\exp(-i\omega t)$ generates in the oscillating absorber a gamma wave with the slowly varying amplitude $\mathrm{E}_\omega(y,t) = \theta(t)\sum_l \mathrm{E}_{\omega l}(y)e^{-i\Omega l t}$, the Raman terms of which satisfy the system (1) with the boundary conditions $\mathrm{E}_{\omega l}(0) = \delta_{l0}\cdot i\Gamma_s^{1/2}(2\pi)^{-1}(\omega - \omega_s + i\Gamma_s/2)^{-1}$ in accordance with the conditions of the considered experiment. For the phase correlated oscillations of nuclei (with the same amplitude) the matrix of coefficients (2) for system (1) is re-written as

$$\begin{aligned} G_{ll'} = &-\frac{i2\pi f_{LM}}{c\omega\hbar\left(2I_g+1\right)}\sum_q\int\frac{d^3\mathbf{k}}{(2\pi)^3}\sum_{e,g}\left\langle g|\widehat{j}^{\,s}(\mathbf{k})|e\right\rangle\left\langle e|\widehat{j}^{\,*s'}(\mathbf{k}_\gamma)|g\right\rangle J_{q+l}(b)\,J_{q+l'}(b) \\ &\times\int_0^t d\tau\exp[i(\omega-\omega_0-q\Omega+i\Gamma_0/2)(t-\tau)]\exp\left(i\left(l'-l\right)\varphi\right)\sum_a\exp\left(i\left(\mathbf{k}_\gamma-\mathbf{k}\right)\mathbf{R}_a\right) \\ = &-i\sum_q e^{i(l'-l)\varphi}J_{q+l}(b)J_{q+l'}(b)\,\frac{\sigma_0 f_{LM}N_0\Gamma_0\left(1-\exp\left(i\left(\omega-\omega_0-q\Omega+i\Gamma_0/2\right)t\right)\right)}{4\left(\omega-\omega_0-q\Omega+i\Gamma_0/2\right)}. \end{aligned} \tag{11}$$

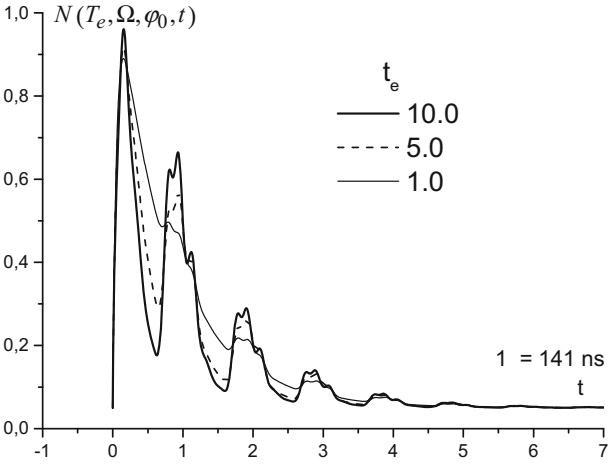

Fig. 5 Counting rate $N\,(T_e, \Omega, \varphi_0, t)$ as a function of the effective thickness t_e: $\Omega = 7.26$ MHz, $b = 3.05$, $\phi_0 = 0$, $\Delta\varphi = \pi/2$, $\Gamma_s = \Gamma_0 = 1$and $\omega_s - \omega_0 = 2\Omega$

Expression (11) is obtained by substituting the integration $\int_{-\infty}^{t} d\tau$ in (2) by $\int_{0}^{t} d\tau$. The solution of system (1) can be found by the iteration method For the harmonic $E_{\omega l}\,(Y)$ behind the absorber we have

$$E_{\omega l}\,(Y) \;=\; \bar{\varepsilon}_0\,(\omega) \sum_q \exp\left(-il\varphi\right) J_{l+q}\,(b) J_q\,(b) \exp\left(iG_q\,(\omega)\cdot Y\right),$$

$$G_q\,(\omega) \;=\; -\sigma_0 N_0 f_{L-M}\left(\Gamma_0\,/4\right) \frac{\left(1-\exp\left(i\,(\omega-\omega_0-q\Omega+i\Gamma_0/2)\,t\right)\right)}{\left(\omega-\omega_0-\Omega q+i\Gamma_0\,/2\right)}. \qquad (12)$$

We integrate $\mathrm{E}_{\omega}\,(Y, t)\exp\left(-i\omega t\right)$ over all radiation frequencies of the source in order to determine the resultant field on the detector $E'\,(Y, \Omega, \varphi, t)$:

$$\begin{aligned} E'\,(Y, \Omega, \varphi, t) &= \int d\omega\, \exp\left(-i\omega t\right) \sum\nolimits_l E_{\omega l}\,(Y) \exp\left(-il\Omega t\right) \\ &= \sum_q P_{q\{l\}}(\varphi, b, t)\cdot E''_q\,(Y, t)\,, \qquad (13) \\ P_{q\{l\}}\,(\varphi, b, t) \;&=\; \sum_l \exp\left(-il\Omega t\right) \exp\left(-il\varphi\right) J_{l+q}\,(b)\, J_q\,(b)\,. \end{aligned}$$

The counting rate of photons (with allowance for the nuclear-resonance interaction) is

$$N\,(T_e, \Omega, \varphi_0, t) = \frac{1}{\Delta\varphi} \int_{\varphi_0-\Delta\varphi/2}^{\varphi_0+\Delta\varphi/2} \left|E'\,(T_e, \Omega, \varphi, t)\right|^2 d\varphi, \qquad (14)$$

where we introduced in $E'\,(Y, \Omega, \varphi, t)$ the effective thickness of the absorber t_e by relations $t_e = \sigma_0 f_{L-M} N_0 Y$ and $T_e = t_e\Gamma_0/4$. Expression (14) sets the counting rate of pair events: the detection (by D1) of a signal photon from of 122 keV transition of the ^{57}Fe isotope at time $t = 0$ and further (at time t) the detection of the Mössbauer14.4 keV photon (see Fig. 1b). It is important to note that out of the multitude of pair events only those are accumulated, for which the sound phase φ, at the moment of detection of the signal photon is located

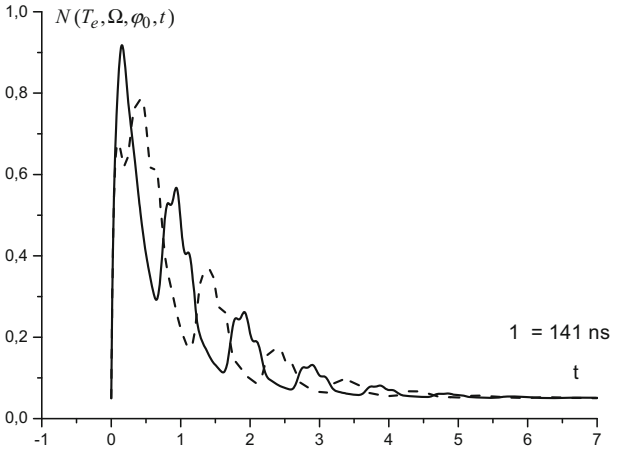

Fig. 6 Dependence of the $N\,(T_e,\,\Omega,\,\varphi_0,\,t)$ on the phase value φ_0 (solid line - $\varphi_0 = 0$, dashed line - $\varphi_0 = \pi$, see [15]): $\Omega = 7.26$ MHz, $b = 3.05$, $\Delta\varphi = \pi/2$, $\Gamma_s = \Gamma_0 = 1$, $\omega_s - \omega_0 = 2\Omega$, and $t_e = 5$

within the significantly narrow interval of $\Delta\varphi + 2\pi n$ with the center of the $\Delta\varphi$ range in φ_0. An essential feature of the $N\,(T_e,\,\Omega,\,\varphi_0,\,t)$ function is determined by integrals of the type

$$E_q''\,(Y,t) = \int d\omega\,\exp\left(-i\omega t\right)\bar{\varepsilon}_0\,(\omega)\exp\left\{-iT_e\frac{\left(1-\exp\left(i\,(\omega-\omega_0-q\Omega+i\Gamma_0/2)\,t\right)\right)}{(\omega-\omega_0-q\Omega+i\Gamma_0/2)}\right\}, \tag{15}$$

where the expression under the integral contains only one singular point unlike the integral considered in [6]. This difference is the consequence of the specification of the theoretical model [4] performed above for single-photon experiments. It is required for the description of the experimental time spectrum. The use of expression (15) in this case made it possible to achieve the correspondence between the model time spectrum calculated on the basis of (14) and the experimental time spectrum in the entire time interval (Fig. 3).

The experimental spectrum in Fig. 3 was obtained using the delayed coincidence scheme. The block scheme of the used setup is given in [15]. Stainless steel with the thickness of 25 μm served as a resonance absorber. It was glued with epoxy on the plate of a polymer piezoelectric converter (PVDF – polyvinylidene fluoride), which was excited by the high-frequency voltage with the frequency of 7.26 MHz. The oscillation amplitude of the absorber corresponded to the modulation index $b = 3.05$ that provided the maximum intensity of the second ultrasound satellite in the Mössbauer absorption spectrum. The velocity of the Mössbauer radiation source (^{57}Co in the Rh matrix) was tuned to this (second) satellite. The delayed coincidence spectrum was measured only for the photons, that appear in the source at times corresponding to the acoustic phase in the absorber in the interval from $\phi_0 - \Delta\phi/2$ to $\phi_0 + \Delta\phi/2$, where $\varphi_0 = 0$ and $\Delta\phi = \pi/2$.

We note that the time dependence of the experimental spectrum differs from the dependence $N\,(t)$ by the constant factor due to the electron absorption (with coefficient of absorption μ_e) of gamma-photons:

$$\mathbf{N}\,(T_e,\,\Omega,\,\varphi_0,\,t) = N\,(T_e,\,\Omega,\,\varphi_0,\,t)\cdot\exp\left(-\mu_e\cdot Y\right). \tag{16}$$

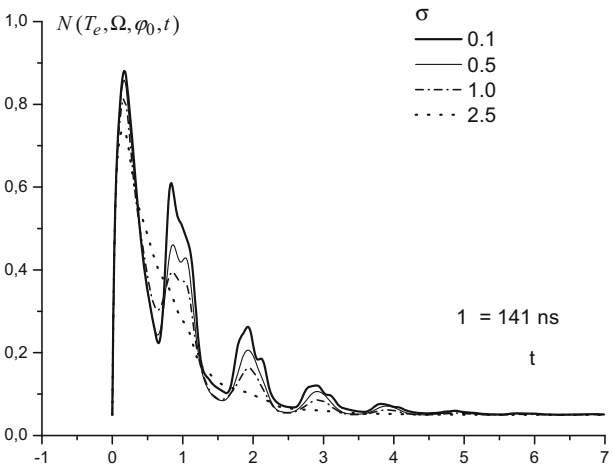

Fig. 7 Time dependence of the counting rate $N\,(T_e,\,\Omega,\,\varphi_0,\,t)$ for different values of the phase correlation of acoustic oscillations σ: $\Omega = 7.26$ MHz, $b = 3.05$, $\phi_0 = 0$, $\Delta\varphi = \pi/2$, $\Gamma_s = \Gamma_0 = 1$, and $\omega_s - \omega_0 = 2\Omega$, $t_e = 5$

Two types of time-domain Mössbauer spectra have been mentioned above: namely, with *an internal time mark* and with *an external time mark*. In the first case as a time mark a some time signal related to the radiating nucleus, usually the moment of its appearing in excited state of the Mössbauer transition may be used. Another way to do this is the use of some time-dependent parameter (say the phase) of the external field interacting with the nucleus. From this point of view, the time dependence of the count rate of gamma photons in (14) corresponds to the definition of the spectra of internal time mark type. At the same time, the parametrical dependence of expression (14) on the initial phase of acoustic oscillations φ_0 (in nuclear time scale) is caused by the external field. The external parameter-dependence of the counting rate becomes as main one if in the experimental setup the counting of Mössbauer photons is not conditioned by the time delayed coincidences (i.e. the signal photons are not registered). In this case the experiment, strictly speaking, is not a single-photon type experiment. Now the Mössbauer photons are detected irrespective of the detection of signal photons. In case of the Mössbauer source with small enough activity the count-rate time dependence $N_{ext}(t)$ (in a laboratory time scale) may be written using the expression (14):

$$N_{ext}(T_e,\,\Omega,\,\varphi_0,\,t) \;=\; \lim_{\substack{T+t\\ T\,\to\,\infty}} \frac{1}{T+t} \int\limits_{-T}^{t} d\tau\; W\,(T_e,\;\Omega,\;\varphi_0+\Omega\tau,\;t-\tau), \qquad (17)$$

$$W\,(T_e,\,\Omega,\,\varphi_0,\,t) \;=\; \left|E'\,(T_e,\,\Omega,\,\varphi_0,\,t)\right|^2.$$

We see, $N_{ext}(T_e,\,\Omega,\,\varphi_0,\,t)$ is a result of summing over pair events mentioned above (detection of signal photon and Mössbauer photon) differing from each other only by time moments of appearing of signal photon (τ) in time interval $-T<\tau<t$, where $t \geq 0$ is a moment of detection of Mössbauer photon and $T \gg \tau_n$, τ_n- nucleus life time. In Fig. 4 the model spectrum calculated on the basis of (17) by numerical method is presented.

2.2 Model calculations of single-photon time-domain Mössbauer spectra

Expression (14) was used also for model calculations illustrating the dependence of the time structure of the single-photon Mössbauer spectra on i) the effective thickness t_e of the absorber (Fig. 5), ii) the value of the sound phase φ_0 at the moment of the detection of the signal (122 keV) gamma-photon (Fig. 6) and iii) the extent of the phase correlation of oscillations of nuclei in the sample given by the parameter of the normal phase distribution σ (Fig. 7). When calculating the last spectrum, for $G_{ll'}$ the expression (11) was used supplemented by a factor taking into account the degree of phase correlation of Mössbauer nuclei oscillations in the sample in the approximation of slow fluctuations (see (8a)). As in the case of frequency spectra, the system of equations (1) was solved by the matrix exponential method.

With increasing σ the characteristic structure in the forward scattering spectra is transformed (destroyed) and the time dependence $N(t)$ approaches an exponential one (see Fig. 7).We also note that fitting of the experimental spectrum (see Fig. 3) on the basis of the model expanded to the case of partial phase correlation of nuclei in the sample showed a rather good phase correlation of acoustic oscillations in this experiment ($\sigma \leq 0.1$).

3 Concluding remarks

In this work, we have presented the theoretical models of the formation of Mössbauer (frequency, time) forward scattering spectra for a thick sample containing nuclei exposed to acoustic oscillations. The model for frequency spectra was obtained by the generalization of the model [4] to the case of arbitrary phase correlation of nuclei oscillations in the sample. The model of the time-domain FS spectra we present is of special interest for a description of acoustical effects in Mössbauer experiments performed with the timedelayed coincidence method Our model contains the mechanism of the influence of sound oscillations in the target on the shape of the wave packet of the forward scattered gamma-photon and, like the frequency model, can be used for the determination of the coherency extent of forward scattering. The importance (necessity) of this model was shown convincingly. The adequate description of an acoustical Mössbauer experiment performed by us in the time-delayed coincidence condition (single-photon experiment) was achieved.

At last the presented models should be distinguished from models of Mössbauer experiments [16, 17] using the synchrotron radiation (SR) as a source of resonance gamma photons. The application of SR caused the powerful development of Mössbauer studies. In particular, a wide class of interference phenomena which are caused by acoustic oscillations in the sample and substantially affect on the formation of the Mössbauer response (time-domain Mössbauer spectra) was studied. The presented model and experimental results are in qualitative agreement with the results of the above works (in particular, associated with acoustic methods [18–20]), supplement them, and at the same time differ from them concerning the mechanisms of the formation of the Mössbauer spectra and detection schemes inherent to the natural Mössbauer source.

The above analysis (see also [8, 13, 14, 21]) demonstrates a series of features of Mössbauer forward scattering on thick absorbers excited by external (ultrasound or radio-frequency) fields. Under these conditions we reveal the increasing of the information capability of the Mössbauer experiment. They are associated with the dependence of the interference effects (the extent of coherence of forward scattering) on the phase synchronism of processes initiated on nuclei by the external fields. The possibility to get such kind

information from the observed structure (time, frequency) of Mössbauer forward scattering spectra is confirmed by the model calculations.

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