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P. HELISTÖ, E. IKONEN, T. KATILA, J. JÕGI, E. REALO, S. REIMAN

TIME DEPENDENCE OF INELASTIC MÖSSBAUER SCATTERING WITH STEPWISE PHASE MODULATION

(Presented by K. K. Rebane)

The time dependence of inelastic Mössbauer scattering in the case of stepwise phase modulation of recoilless gamma radiation is treated theoretically as well as experimentally. In a thin scatterer approximation a formula describing conversion electron yield for different center shift and phase change values is derived. The stepwise phase modulation of the 14.4-keV resonant radiation is performed by an x-cut quartz piezotransducer to which a ^{57}Co: Pd gamma source is cemented. By using a resonance detector with thin ^{57}FeAl-alloy scatterers to detect conversion electrons an acceptable time resolution (< 30 ns) is achieved. A reasonable agreement between theory and experiments is obtained.

1. Introduction

The discovery of coherent transient effects in Mössbauer spectroscopy [1, 2] has given a new method for studying the interaction between recoilless gamma radiation and solids. Phase switching of resonant radiation by means of piezotransducers has been applied to the investigations of ^{67}Zn [2, 3] and ^{57}Fe [4, 5] Mössbauer transmission transients. Contrary to the delayed coincidence techniques, where the time reference is provided by previous nuclear events, the present method with an external time reference from periodic exciting signals is not limited to low source activities. This is important for precision Mössbauer scattering studies.

The earlier experiments on the time dependence of Mössbauer scattering have been performed, as a rule, by using gamma-gamma (^{57}Fe) or x-ray-gamma (^{119}Sn) coincidence and by detecting the scattered gamma radiation [6], emitted x-rays [7] or conversion electrons [8–10]. The only exception is a recent work [11], where the time dependence of ^{57}Fe Mössbauer diffraction has been measured by using a periodic magnetic gamma shutter. The application of resonance detectors to conversion electron detection [8–10] has been advantageous in obtaining a high statistical accuracy owing to the large values of internal conversion coefficients of the gamma transitions considered.

Here we present a theory of time-dependent inelastic Mössbauer scattering in the case of stepwise phase modulation of source radiation and the results of the corresponding experiments with an ^{57}FeAl scatterer incorporated in the resonance detector.

2. Theory of phase-modulated inelastic Mössbauer scattering

In this section we present a classical derivation of the time dependence of inelastic Mössbauer scattering (e. g. emission of conversion electrons) from resonant atoms illuminated by phase-modulated recoilless gamma

radiation. General features of the derivation and its application to thin electron scatterers with stepwise phase modulation are given. In the derivation, nonresonant effects are neglected. A more comprehensive treatment applicable also to elastic scattering processes will be published elsewhere.

Previously the time dependence of scattered x-rays [7] and gamma rays [6] has been calculated in connection with Mössbauer coincidence measurements. These derivations are based on the classical theory of time dependence of resonantly filtered gamma rays [12]. A series expansion has been derived for the Mössbauer transmission to interpret the quantum-beat spectra [13]. The dependence of the transmitted intensity on phase modulation of an arbitrary shape and frequency has been derived both classically [2] and semiclassically [3]. So far, no calculations have been presented on the time dependence of scattered radiation in phase modulation experiments.

Let us assume that the source field $E_s(\omega, t_0)$ (due to an excited state formed at time t_0) strikes perpendicularly to the plane of a scatterer foil of thickness d. The scattering amplitude $\psi(\omega, x)$ caused by an atom at depth x is then [6, 7]

$$\psi(\omega, x) \approx \frac{\exp\left(\dfrac{i\beta x}{\omega - \omega_a - i\Gamma_a/2}\right)}{\omega - \omega_a - i\Gamma_a/2} E_s(\omega, t_0) = S(\omega, x) E_s(\omega, t_0), \qquad (1)$$

where ω_a and $\Gamma_a/2$ are the center frequency and half width of the scatterer resonance, and $\beta = f_a n \sigma_0 \Gamma_0/4$. In this expression f_a is the recoilless fraction and n is the number density of the Mössbauer nuclei in the scatterer, σ_0 is the absorption cross section and Γ_0 is the natural width of the resonance. Small variations in the resonant frequencies may cause line broadening: $\Gamma_a > \Gamma_0$. The complex exponential factor in Eq. (1) takes into account the effect of the resonant medium of thickness x on the source field. The response of the scatterer nucleus to the driving field is proportional to the factor $(\omega - \omega_a - i\Gamma_a/2)^{-1}$ in the harmonic oscillator approximation, if $\omega_a \gg \Gamma_a$.

The time dependence of the scattered intensity is obtained by squaring the inverse Fourier transform of Eq. (1) and by integrating over x and t_0:

$$N(t) = \int_0^d dx \int_{-\infty}^{\infty} dt_0 \left| \int_{-\infty}^{\infty} dt' S(t - t', x) E_s(t', t_0) \right|^2, \qquad (2)$$

where $S(t, x)$ and $E_s(t, t_0)$ are the inverse Fourier transforms of $S(\omega, x)$ and $E_s(\omega, t_0)$, respectively. Because the electrons (or x-rays) are emitted inelastically, the intensities of single scatterers can be summed [7]. Eq. (2) can be used for conversion electrons as well as for x-rays and inelastically scattered gamma rays following resonant absorption.

We use the following time dependence for the source field [2]

$$E_s(t, t_0) = \exp\left[-\Gamma_s(t - t_0)/2 + i\omega_s(t - t_0) + i\Phi(t) - i\Phi(t_0)\right]\Theta(t - t_0), \qquad (3)$$

where ω_s and $\Gamma_s/2$ are the resonance frequency and half width of the emitted field and

$$\Phi(t) = \Delta\Phi\Theta(t) = \frac{\Delta x}{\lambda}\Theta(t) \qquad (4)$$

is the phase modulation due to source displacement by Δx at $t = 0$ ($\lambda = c/\omega_s$).

Assume that the parameter $b = \beta d \ll \Gamma_a$. Then, after some calculations,

the following expression for the time dependence of the total yield of conversion electrons is obtained:

$$N(t) = N_0 f_s \frac{\alpha}{\alpha+1} 2b\Gamma_a \operatorname{Re}\left\{\frac{1}{\Gamma_+(\Gamma_+ + \Gamma_-)} - \right.$$
$$\left. - (1 - e^{i\Delta\Phi}) \frac{e^{-\Gamma_+ t}(1 - e^{-\Gamma_- t})}{\Gamma_+ \Gamma_-} \Theta(t)\right\}. \tag{5}$$

A similar expression can be derived for the emission of x-rays or Auger electrons after resonance absorption. Here α is the internal conversion coefficient, N_0 and f_s are the intensity and recoilless fraction of the source, correspondingly,

$$\Gamma_\pm = (\Gamma_a \pm \Gamma_s + b)/2 \pm i\Delta\omega \tag{6}$$

and $\Delta\omega = \omega_a - \omega_s$.

For $\Gamma_s \approx \Gamma_a$ and $\Delta\omega = 0$ the time dependence of Eq. (5) approaches $-t\exp(-\Gamma_a t)$ as $b \to 0$. In Mössbauer coincidence measurements, where t_0 is measured, the corresponding time dependence is $t^2\exp(-\Gamma_a t)$ [6, 7]. Even a relatively small center shift can change considerably the shape of the scattering transient. This is demonstrated in Fig. 1 for various values of the phase change $\Delta\Phi$, when $\Delta\omega = \Gamma_0/2$. The curves are plotted for both forward and backward motion of the source. When $\Delta\Phi = \pi/3$, the transient is positive, i.e. counting rates above the steady-state level are obtained. At $\Delta\Phi = \pm\pi$ or $-2\pi/3$, the intensity rapidly drops to less than half the steady-state value and returns back within $\sim 4/\Gamma_0$. In Fig. 2 theoretical curves for electron scattering are shown at several values of $\Delta\omega$, when $\Delta\Phi = \pi/2$. The time dependences corresponding to opposite signs of $\Delta\Phi$ (or of $\Delta\omega$) are considerably different. Oscillations are observed as $\Delta\omega$ is increased. Both figures are plotted assuming

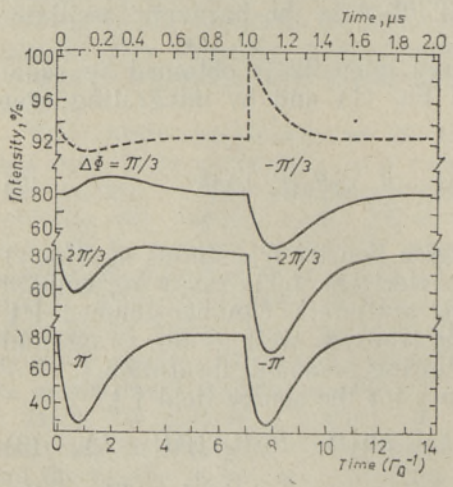

Fig. 1. Theoretical transmission (dashed curve) and scattering (solid) curves with phase changes $\Delta\Phi = \pm\pi/3$, $\pm 2\pi/3$ and $\pm\pi$. The scattering curves are scaled vertically so that 100% corresponds to the steady-state intensity at $\Delta\omega = 0$. The upper time scale refers to ^{57}Fe. The following parameter values were used: $\Gamma_s = \Gamma_a = \Gamma_0$, $\Delta\omega = 0.5\ \Gamma_0$ and $b = 0.05\ \Gamma_0$.

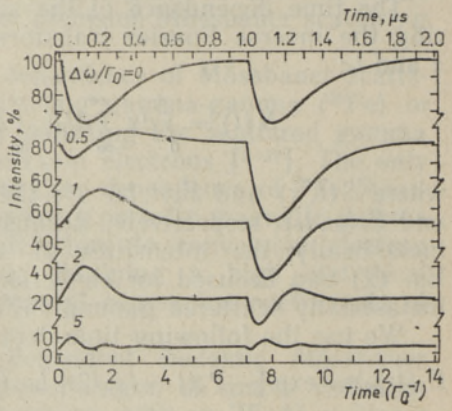

Fig. 2. Theoretical scattering curves corresponding to center shifts $\Delta\omega/\Gamma_0 = 0$, 0.5, 1, 2 and 5 when $\Delta\Phi = \pm\pi/2$. The shape of the phase modulation and other parameter values are as in Fig. 1.

natural linewidths. Additional line broadenings appear mainly as a compression of the transients and an associated scaling of the $\Delta\omega$-value.

Effects as large as $\sim 75\%$ relative to the maximum steady-state intensity can be realized with a thin scatterer when $\Delta\omega = 0$ and $\Delta\Phi = \pi$. The change in the corresponding transmission intensity would be only $0.2 f_a$ ($b = 0.05\Gamma_0$). The increase in the counting rate for nonzero $\Delta\omega$ can be understood by studying the corresponding transmission curve: a decrease in the scattering intensity is accompanied by a positive transmission peak and vice versa as shown in Fig. 1. In some cases the scattering transients decay considerably slower than the transmission transients.

So far only electron scattering due to resonant absorption has been considered. The effect of the Rayleigh scattering is negligible. Photoabsorption produces a small background effect. The attenuation of conversion electrons and nonideal detection geometry mainly decrease the counting rate. Considering these effects, measurements of gamma transients in scattering geometry can be effectively performed with resonance detectors by utilizing the large conversion coefficient of ^{57}Fe.

Detection of the oscillations demonstrated in Fig. 2 does not require a well-defined displacement amplitude of the source. They should be observable also if there exists a distribution in the values of $\Delta\Phi$ owing to experimental nonidealities. It is worth stressing that the scattered radiation does not react instantaneously to changes in the phase of the source field, contrary to the transmitted radiation. Thus the requirements on the motion of the source and on the time resolution of the electronics are not as stringent as in the case of transmission measurements.

3. Experimental

The time dependence of the yield of conversion electrons from an ^{57}Fe resonant scatterer due to the absorption of phase-modulated gamma radiation was measured by means of a resonance detector [14]. The detector was a methane-filled proportional counter with two cathodes (scatterers) made of 95% enriched ^{57}FeAl alloy. The thicknesses of the ^{57}FeAl layers evaporated on mica foils were slightly larger than the track length (~ 90 µg/cm^2) of the 7.3-keV conversion electrons. A thin W anode wire was placed between the parallel scatterers spaced at a distance of 5 mm. In all experiments a collimated (5 mm in diameter) beam of 90° incidence gamma radiation struck at the center of the scatterer. Anode pulses of the detector were amplified by a fast pulse-shaping amplifier and fed for timing to a constant fraction discriminator.

A conventional Mössbauer spectrum of the detector showed a single broadened line of width 0.34 ± 0.01 mm/s and a background level of 8.5%. A small contribution from photoeffect ensured the low background level. A measurement performed in transmission geometry also confirmed the additional broadening of the scatterer line. The linewidth of the transmission spectrum was 0.38 ± 0.01 mm/s and the resonant absorption was equal to $\varepsilon_0 = 0.05$. As the linewidth of the ^{57}Co : Pd source was estimated to be $\Gamma_s \approx 0.11 \pm 0.01$ mm/s $= 1.1\Gamma_0$, we can conclude that $\Gamma_a \geqslant 0.20$ mm/s $= 2\Gamma_0$. The additional broadening is caused by the inhomogeneities of the scatterer material.

The time resolution of the resonance detector was determined from coincidence measurements by using a ^{22}Na source. A plastic scintillator coupled to a fast EMI9813B photomultiplier served as a start detector for a time-to-amplitude converter (TAC). The energy selection of the

start detector was set near 250 keV on the Compton distribution of the 511 keV annihilation radiation. The resonance detector provided stop pulses corresponding to the emission of photoelectrons. The energy window was limited to the one used for the detection of conversion electrons. The prompt curve had a full width at half maximum of 28±2 ns. Though the time resolution of the resonance detector was much lower than that obtained with the NaI(Tl) scintillation detector (4.8 ns for 14.4 keV) [15], it can be considered acceptable for scattering experiments. The stepwise phase modulation of the source radiation was realized by means of a piezotransducer. A $^{57}Co:Pd$ source foil was cemented to an x-cut quartz disk and used as an electrode. A massive Al plate formed the other electrode. For modulation periodic 3.0 μs rectangular voltage pulses with 15 ns rise and decay times were applied to the transducer. The amplitude of the exciting pulses could be selected in the interval 0 — 40 V with an accuracy of ±0.2 V. The pulse generator also provided synchronizing start pulses for timing. The time distribution of resonance detector pulses with respect to these start pulses was measured by means of the TAC. Its output was recorded by a multichannel analyzer. Only the transients corresponding to the positive phase change, i.e. forward displacement of the source, were recorded.

4. Results and discussion

Due to the short lifetime (141 ns) of the Mössbauer state of ^{57}Fe, the nonidealities of source motion with stepwise modulation usually cannot be neglected. We have evaluated the characteristics of actual displacements from measurements in transmission geometry.

A detailed study of coherent transmission transients was performed by using the source-transducer assembly described above, an enriched $K_4Fe(CN)_6 \cdot 3H_2O$ absorber ($b/\Gamma_0 = 1.75 \pm 0.25$), and a fast scintillation detector [16]. The experimental results were reasonably well described assuming a cosinusoidal radial distribution in the values of the phase change $\Delta\Phi$ and a finite inertia-limited duration of mechanical displacements.

Such transmission transients measured at $U = 15$ V are presented in Fig. 3*a* for a zero center shift and in Fig. 3*b* for $\Delta\omega = -0.20$ mm/s, (upper curves). In Fig. 3*a* the corresponding theoretical result is also shown (solid curve). A positive transient at $\Delta\omega = 0$ has a peak-to-peak intensity of $1.07\,\varepsilon_0$ ($\varepsilon_0 = 0.36 \pm 0.02$ is the conventional absorption) and a width of 80 ns. At $\Delta\omega = -0.20$ mm/s the positive transmission peak is followed by a zero-crossing and a negative undershoot.

In Fig. 3*a* and *b* $^{57}FeAl$ scattering transients for various excitation voltages U and two different center shifts $\Delta\omega = 0$ and $\Delta\omega = -0.20$ m/ms, respectively, are presented. At $\Delta\omega = 0$ the intensity of transients increases up to 50% relative to the steady-state level, while U increases up to 20 V. A further enhancement of U results in a small decrease of the peak-to-peak intensity, e.g. 40% at $U = 40$ V. The corresponding values are 10% higher, when background correction is taken into account. The transients reach their minima within 150 ns and have total durations of about 600 — 650 ns. With non-zero center shift oscillatory behaviour is seen (Fig. 3*b*). In the lowermost curves zero-crossing at 200—300 ns is followed by a positive overshoot. As far as the opposite-sign behaviour of intensities is concerned, the measured scattering curves are in good accordance with the transmission time dependences. It should be noted that the transmission and scattering transients in Fig. 3 cannot be compared in detail because the absorber and the scatterer differ considerably

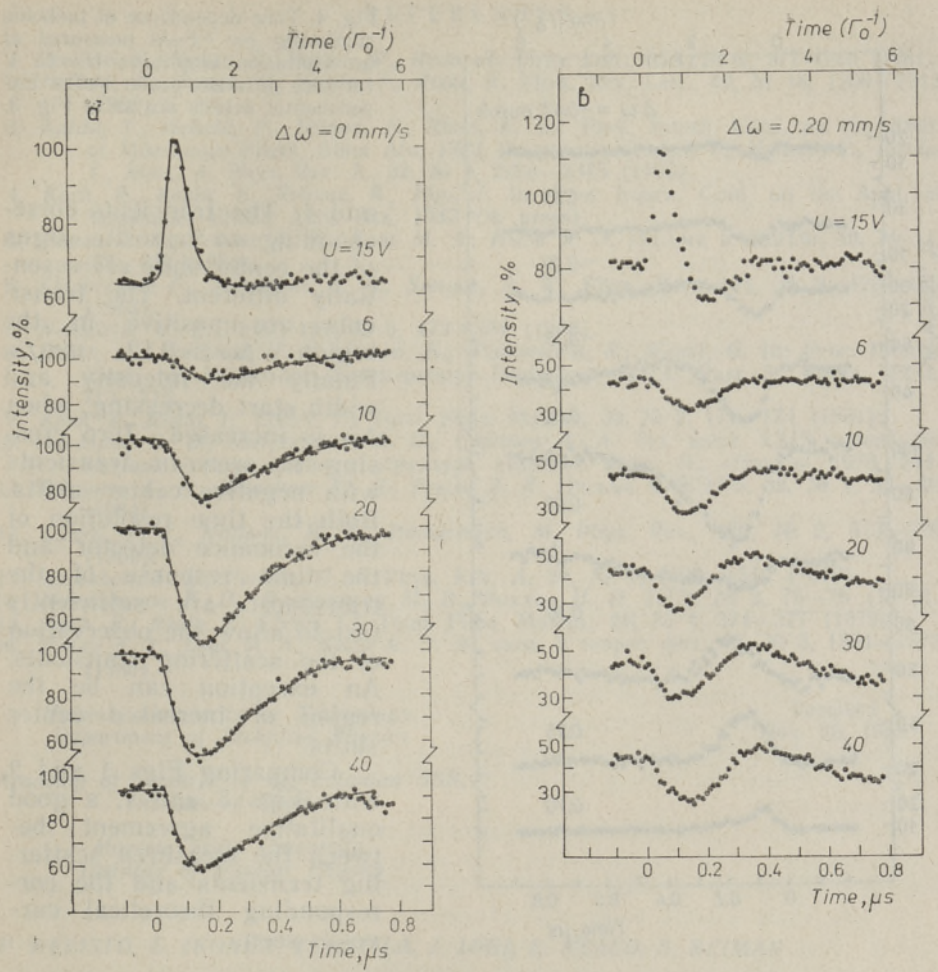

Fig. 3. Transmission transients measured with a $K_4{}^{57}Fe(CN)_6 \cdot 3H_2O$ absorber ($b \approx 1.75\ \Gamma_0$) at an excitation amplitude $U=15$ V (uppermost curves). The other ten curves show the time dependence of electron emission intensity from ^{57}FeAl scatterers measured by using stepwise modulation at five different excitation amplitudes $U=6$, 10, 20, 30 and 40 V for two center shifts: $\Delta\omega=0$ mm/s (a) and $\Delta\omega=-0.20$ mm/s (b). The curves correspond to positive phase changes. The gamma source is a ^{57}Co : Pd foil cemented to the x-cut quartz transducer and the scatterer consists of two ^{57}FeAl layers. The scattering intensity is scaled as in Fig. 1. Solid curves are calculated from Eq. (5), with $\Gamma_s=\Gamma_0$ and $\Gamma_a+b=1.25\Gamma_0$.

from each other. At the voltages $U>20$ V a decrease and curvature of the steady-state level are observed, caused by the excitation of resonances of the transducer. To minimize these distortions the amplitude of exciting pulses should be kept low.

A strong center shift dependence of the scattering transients, which follows from the theory presented above, is also observed experimentally. In Fig. 4 a series of scattering transients measured at a constant voltage $U=15$ V for various center shifts in the interval from —0.46 mm/s to 0.70 mm/s are shown. The steady-state level follows conventional Mössbauer scattering intensity as a function of the center shift. A nonresonant background amounting to 8.5% is not subtracted in Figs 3

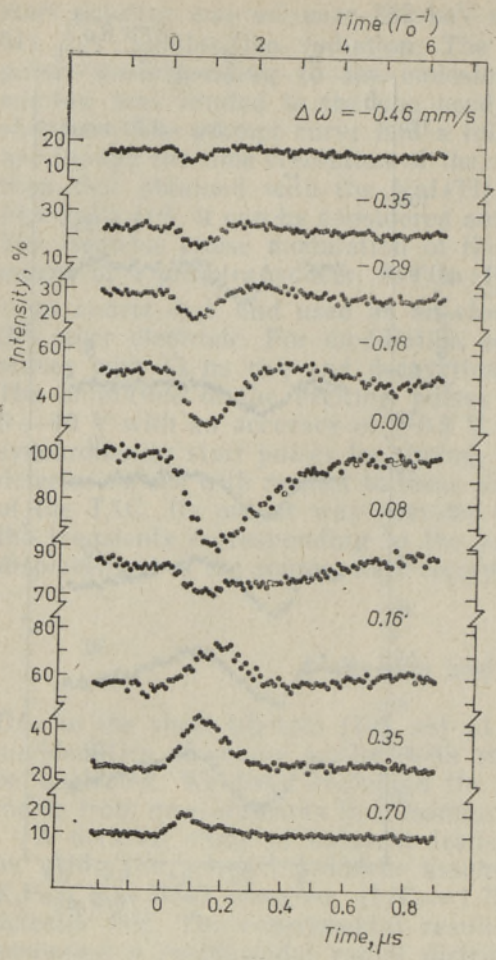

Fig. 4. Time dependence of inelastic scattering for ^{57}FeAl measured at an excitation amplitude $U = 15$ V for nine different center shifts. Experimental details are as in Fig. 3.

and 4. The transients corresponding to opposite signs of the center shift are essentially different. The transients are positive in the region $\Delta\omega > 0.12$ mm/s. Finally the intensity and width start decreasing, when $\Delta\omega$ is increased. Zero crossing is seen in transients with negative center shifts. Both the time resolution of the resonance detector and the time response of the transducer are sufficiently fast to allow the observation of the scattering transients. An exception can be the region of increased center shifts.

Comparing Figs 1 and 2 with Figs 3 and 4, a good qualitative agreement between the measured scattering transients and the corresponding theoretical curves is seen.

5. Summary

We have shown that the phase modulation of recoilless gamma radiation can be effectively applied to time-dependent electron scattering. The main results of the theory developed to describe the time dependence of inelastic Mössbauer scattering in the case of stepwise phase modulation are consistent with the corresponding experiments using ^{57}Fe. A high signal-to-background ratio and sufficient time resolution are obtained owing to the use of a resonance detector with ^{57}FeAl scatterers for the detection of emitted conversion electrons. Further research of different scatterers with a better time resolution and improved phase modulation is needed.

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Helsinki University of Technology,
Department of Technical Physics

Academy of Sciences of the Estonian SSR,
Institute of Physics

Moscow State University,
Institute of Nuclear Physics

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P. HELISTÖ, E. IKONEN, T. KATILA, J. JÕGI, E. REALO, S. REIMAN

MITTEELASTSE MÖSSBAUERI HAJUMISE AJASÕLTUVUS ASTMEKUJULISE FAASIMODULATSIOONI JUHUL

Mitteelastse Mössbaueri hajumise ajasõltuvust resonantsgammakiirguse astmekujulise faasimodulatsiooni korral uuriti teoreetiliselt ja eksperimentaalselt. Tuletati valem, mis õhukese hajutaja lähenduses kirjeldab konversioonelektronide saagist joonenihete ja faasimuutuste erinevate väärtuste juhul. Eksperimentaalselt tekitati 14,4 keV resonantskiirguse astmekujuline faasimodulatsioon ^{57}Co (Pd) gammaallika külge liimitud piesomuundi (x-lõike kvartsi) abil. Õhukeste ^{57}FeAl-sulamist hajutajatega resonantsdetektori kasutamisel konversioonelektronide registreerimiseks saavutati piisav ajaline eraldusvõime (< 30 ns). Teoreetilised sõltuvused on heas kooskõlas eksperimendi tulemustega.

П. ХЕЛИСТЕ, Э. ИКОНЕН, Т. КАТИЛА, Я. ЙЫГИ, Э. РЕАЛО, С. РЕЙМАН

ВРЕМЕННАЯ ЗАВИСИМОСТЬ НЕУПРУГОГО МЕССБАУЭРОВСКОГО РАССЕЯНИЯ ПРИ СТУПЕНЧАТОЙ ФАЗОВОЙ МОДУЛЯЦИИ

Проведено теоретическое и экспериментальное исследование временной зависимости неупругого мёссбауэровского рассеяния в случае ступенчатой фазовой модуляции резонансного гамма-излучения. В приближении тонкого рассеивателя выведена формула для описания выхода электронов конверсии при различных величинах изомерных сдвигов и скачков фазы. Экспериментально ступенчатая фазовая модуляция резонансного излучения 14,4 кэВ осуществлялась с помощью приклеенного к гамма-источнику ^{57}Co (Pd) пьезопреобразователя из кварца x-среза. При использовании резонансного детектора с тонкими рассеивателями из сплава ^{57}FeAl было достигнуто временное разрешение <30 нс для детектирования электронов конверсии. Получено хорошее согласие теоретических зависимостей и результатов эксперимента.