# Coherent transient effects in the Mössbauer radiation of $^{57}Fe$

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Coherent transient effects have been observed experimentally during the stepped phase modulation of a $^{57}Co(Pd)$ $\gamma$ source by means of a piezoelectric transducer. The effects take the form of periodic pulses in the intensity of the 14.4-keV resonant $\gamma$ radiation transmitted through the absorber. The pulses have a short rise time (15–30 ns), a length less than 100 ns, and a peak intensity exceeding the steady-state level of the resonant effect. The use of these coherent transient effects to study the time evolution of inelastic resonant scattering is demonstrated.

The study of coherent transient effects is a new direction in Mössbauer spectroscopy, of considerable theoretical and experimental interest. The possibility of observing these effects was predicted theoretically for the 93.3-keV resonance of the isotope $^{67}Zn$ ($\tau_0 = 13.2$ $\mu$s), and it was for this resonance that these effects were first demonstrated.[1,2] It has been shown that an in-phase Doppler modulation will, by causing a rapid (in comparison with the level lifetime $\tau_0$) disruption of the resonance of the $\gamma$ source and the absorber, give rise to interference changes in the time-frequency composition of the intensity of the $\gamma$ radiation: oscillations, jumps in the intensity, an increased transmission, etc. Analogous effects have been observed in NMR[3] and laser spectroscopy.[4]

Because of the high Debye-Waller factors ($f \approx 0.7$–$0.8$) and the brief lifetime ($\tau_0 = 140$ ns), the 14.4-keV Mössbauer radiation of $^{57}Fe$ has some clear advantages over that of $^{67}Zn$. The possibility of achieving a high time resolution (less than 5 ns)[5] and the realization of a rapid in-phase motion of the sources (or absorbers)[6,7] lay the groundwork for successful experiments. The possibility of observing coherent transient effects for $^{57}Fe$ through a modulation of the absorber was demonstrated in Ref. 7.

In the present letter we report an experimental study of the coherent transient effects for $^{57}Fe$ during rapid stepped modulation of the $\gamma$ source.

The basic features of the coherent transient effects can be described well by the classical theory of a time-dependent transmission of phase-modulated Mössbauer radiation.[2]

We consider the case of a $\gamma$ source and a resonant absorber with single Lorentzian lines with central frequencies $\omega_s$ and $\omega_a$ and half-widths $\Gamma_s$ and $\Gamma_a$, respectively, where a stepped phase shift $\varphi(t) = \varphi_0 + a\theta(t)$ is caused by an instantaneous mechanical displacement of the source with respect to the absorber at the time $t = 0$. The displacement is by a distance $\Delta x$ ($a = \Delta x/\lambda\!\!\!^{-}$, where $2\pi\lambda\!\!\!^{-}$ is the wavelength of the $\gamma$ radiation). The frequency shift of the lines, $\Delta\omega = \omega_s - \omega_a$, before and after the phase step $a$ is assumed to be steady. If modulated radiation of this type passes through a resonant absorber with a Mössbauer thickness $T_M$, changes occur in the phase relations between the two field components corresponding to the wave trains (1) emitted at random times and (2) emitted at the time of the stepped phase shift. As a result, a pulse appears in the intensity transmitted by the absorber. Our calculations show that the time-frequency dependence of the intensity can be described by

$$I(t, \Delta\omega) = 2f_s e^{-\Gamma_a t}[(1-\cos a)\,\mathrm{Re}(AB) - \sin a\,\mathrm{Im}(AB)]\theta(t),$$

where

$$A = \sum_{n=0}^{\infty} J_n(\sqrt{T_M\Gamma_a t})(-1)^n\left(\frac{4t}{T_M\Gamma_a}\right)^{n/2}[i\Delta\omega + (\Gamma_s - \Gamma_a)/2]^n,$$

$$B = \sum_{n=1}^{\infty} J_n(\sqrt{T_M\Gamma_a t})(-1)^n\left(\frac{T_M\Gamma_a}{4t}\right)^{n/2}[i\Delta\omega + (\Gamma_s + \Gamma_a)/2]^{-n},$$

$\mathrm{Re}(AB)$ and $\mathrm{Im}(AB)$ are the real and imaginary parts of $(A, B)$, $J_n$ is the Bessel function of the first kind of index $n$, and $f_s$ is the Debye-Waller factor of the $\gamma$ source.

The corresponding expression in Ref. 2 holds for $T_M < 4$ and $\Delta\omega = 0$, so that expression is of limited applicability in the relevant cases of thick ($T_M \approx 8$) $^{57}$Fe absorbers with $\Delta\omega \neq 0$.

Analysis of this expression shows that the step phase shift is accompanied by an abrupt change in the intensity, which reaches values of $4f_s$ at $\Delta\omega = 0$ and $8f_s$ at $\Delta\omega \neq 0$ in the case of a thick absorber; i.e., this jump exceeds the steady-state resonant effect, $\epsilon_0$, by factors of four and eight. The intensity decay after the step can be described approximately by an exponential function with a decay constant $\tau_n$, which depends on $T_M$ and $\Gamma_a + \Gamma_s$. As $T_M$ is increased, the pulse becomes considerably shorter than $\tau_0$. The pulse decay terminates in oscillations, whose amplitude increases and whose peaks shift toward $t = 0$ with increasing $T_M$.

Experiments on a stepped phase modulation have been carried out for the combination of a $^{57}$Co(Pd)$\gamma$ source and a $K_4\,^{57}Fe(CN)_6 \cdot 3H_2O$ ($T_M \approx 8$), with $\epsilon_0 = 0.29$. The source was modulated by a piezoelectric transducer (an $x$-cut quartz disk 0.25 mm thick) cemented to the source. Periodic square voltage ($U$) pulses with short rise and decay times ($\sim$15–20 ns) were applied to this transducer. The line shift $\Delta\omega$ was set by a slow ($\sim$10-Hz) Doppler shift of the $\gamma$ source.

The time evolution of the intensity of the $\gamma$ beam transmitted by the absorber with respect to the rise time of the exciting pulse was measured by a Mössbauer time-resolving spectrometer with a time-to-amplitude converter.[5] The resolving time of the apparatus for the detection of 14.4-keV $\gamma$ rays is 4.8 ns.

The experiments confirm that it is possible to generate intense periodic pulses of 14.4-keV $^{57}$Fe $\gamma$ rays with a short rise time ($t_{0.1\text{–}0.9} = 20$–30 ns) and a short decay time (with $\tau_n$ between 35 and 100 ns) from a beam of $\gamma$ rays emitted at random over time (Fig. 1). These pulses are synchronized with the fronts of the exciting signal. The intensity of the pulses increases significantly with increasing excitation amplitude $U$ (Fig. 1a), reaching $L_{\max} = 1.88$ times the intensity at $t < 0$, which is $2.2\epsilon_0$ on this scale.

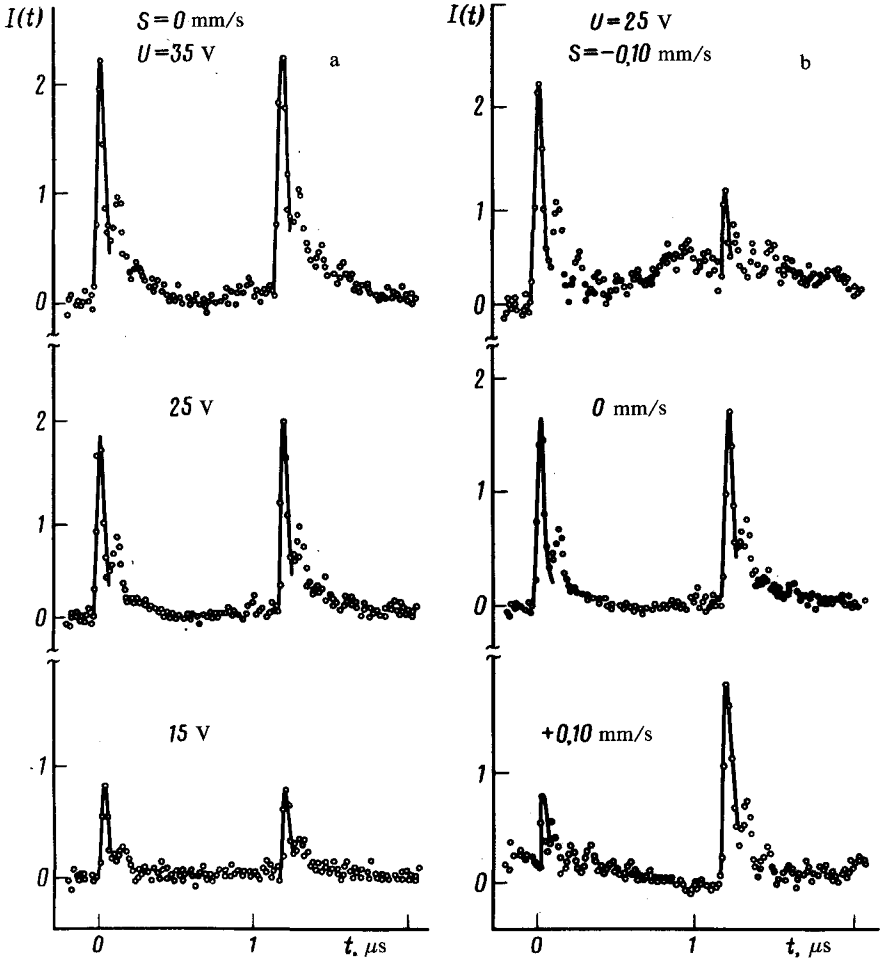


FIG. 1. Time evolution of the intensity of the 14.4-keV $\gamma$ radiation of $^{57}$Fe transmitted through a $K_4$ $^{57}Fe(CN)_6 \cdot 3H_2O$ absorber ($T_M \approx 8$) during stepped modulation of the $^{57}$Co(Pd) $\gamma$ source. a—For $S = 0$ and various amplitudes of the exciting pulses, $U$; b—for $U = 25$ V and various isomer shifts $S$. The intensities $I(t)$ are given in units of $\epsilon_0$.

The increase in the intensity jump above $\epsilon_0$ and the decrease in the duration $\tau_p < \tau_0$ are evidence that we are observing coherent transient effects in the isotope $^{57}$Fe. As expected, the overall pictures of the changes in the intensity are identical for modulation of the source and modulation of the absorber (in Ref. 7).

It can be seen from Fig. 1 that the pulse shape is distorted slightly by the imperfect motion of the modulator. The finite duration of the phase shift and the response of the piezoelectric transducer reduce the peak intensity, increase the effective value of $\tau_p$ in comparison with the theoretical predictions, and give rise to afterpulses.

The ratio of the peak intensities ($I_+/I_-$) corresponding to phase steps of different signs (the rise and decay of the exciting pulse), which depends strongly on the isomer

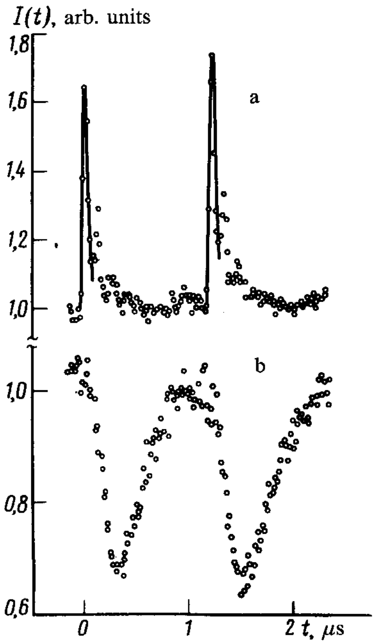


FIG. 2. Time evolution of the intensity, $I(t)$ during stepped modulation of a $^{57}Co(Pd)$ $\gamma$ source with $S=0$ and $U=28$ V. a—Detection of the $\gamma$ rays transmitted through a $K_4$ $^{57}Fe(CN)_6 \cdot 3H_2O$ absorber by a NaI-Tl scintillation detector; b—detection of scattered conversion electrons by a $^{57}$FeAl resonant detector.

shift $S \sim \Delta\omega$ (Fig. 1b), can be exploited for precise measurements of isomer or temperature shifts. We found $I_+/I_- \sim kS$, where $k = 3.95$ s/mm in the region[7] $|S| \leqslant 0.15$ mm/s; the coefficient $k$ increases significantly with decreasing amplitude $U \sim a$.

The coherent transient effects of the $^{57}$Fe $\gamma$ radiation can be used successfully to study the time evolution of resonant scattering. The time dependence of the scattered conversion electrons (Fig. 2), which we measured with a resonant $^{57}$FeAl detector for a stepped phase modulation of a $^{57}$Co(Pd) $\gamma$ source, is characteristic of inelastic resonant scattering. The shape of the curve is qualitatively the same as that of the corresponding curves found in measurements by the delayed $\gamma$-$\gamma$ coincidence method[8–10] and predicted theoretically.[11] The high peak intensity, the short pulse length, and the universal applicability give the method of shaping pulses of resonant $^{57}$Fe $\gamma$ radiation based on the coherent transient effects some clear advantages over modulators of other types (that of Ref. 12, for example).

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