Energy and time dependences of the secondary-radiation yield from different depths under the conditions of Mössbauer total external reflection of synchrotron radiation

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It is shown theoretically that the coherent radiation field, formed under the conditions of total external reflection of a synchrotron radiation pulse from a resonant sample, determines the appearance of the time-delayed response of nonresonance impurity atoms (fluorescence radiation, Auger electrons, and photoelectrons) and that the energy and time dependences of this radiation clearly characterize the depth at which it was generated

The application of pulsed synchrotron radiation for studying nuclear resonance scattering in samples containing Mössbauer nuclei marks the appearance of a new method for investigating hyperfine interactions — "time-differential Mössbauer spectroscopy" (see, for example, Refs. 1 and 2 and the references cited there). In view of the large difference in the nuclear resonance times ($\sim 10^{-8}$ s for ^{57}Fe) and the Rayleigh scattering times ($\sim 10^{-15}$ s) effective filtering of the resonance radiation occurs in time measurements. In the presence of hyperfine splitting of the nuclear levels the temporal spectra of nuclear resonance scattering exhibit quantum oscillations.[3,4] The coherent effects which arise as a result of absorption in thick absorbers, as a result of nuclear resonance diffraction by a crystalline lattice, or as a result of total external reflection, for the study of which synchrotron radiation is especially suitable, also appear in the temporal spectra in the form of dynamic oscillations.

Under the conditions of coherent interaction the temporal separation of the electronic and nuclear scattering channels becomes impossible. This is strikingly illustrated by the observation of an "interference peak" near the critical angle of total external reflection for the delayed reflection intensity from a Mössbauer mirror;[5–8] which cannot be explained by analyzing the purely nuclear scattering. Direct evidence for the existence of the excitation of the electronic subsystem of a resonant sample for times much longer than the Rayleigh scattering time of the synchrotron pulse under coherent interaction conditions could be the emission of the corresponding secondary radiation.

In the present paper we discuss the characteristic features of secondary-radiation yield (inelastic scattering), which reflects most strikingly, as we know, the structure of the radiation field in the sample, under the conditions of total external reflection from a resonance sample by analogy with the method of x-ray standing waves.[9,10] The resonance

character of the interaction in our case makes it possible to use not only the angular dependences of the secondary-radiation yield to obtain information about the localization of the emitting resonance and impurity atoms, but also the energy and, correspondingly, the time dependences.

In our analysis it makes sense to distinguish two forms of secondary radiation. I shall refer to the radiation accompanying nuclear resonance absorption (conversion electrons and subsequent x-ray radiation, Auger electrons) briefly as "resonance" secondary radiation. I shall refer to photoelectrons and the subsequent x-rays and Auger electrons, whose formation is associated with nonresonance photoabsorption, as "nonresonance" secondary radiation. From the theoretical standpoint, this separation is justified by the specific nature of their energy and time dependences. In the experiment it is undoubtedly impossible to separate, for example, the photo- and conversion electrons emitted by ^{57}Fe, but the fluorescence radiation from the impurity atoms in a resonance sample can be observed separately.

In Refs. 11 and 12 it was shown that the yield of photoelectrons (and "nonresonance" secondary radiation in general) under the conditions of total external reflection from a Mössbauer mirror becomes energy-dependent as a result of the change in the amplitude of the radiation field in the sample accompanying a change in the energy of the incident radiation in the neighborhood of a resonance. In other words, a resonance standing wave determines the characteristics of the yield of both "resonance" and "nonresonance" secondary radiations. Correspondingly, in the time domain this means that under the conditions of coherent interaction with a resonance sample the "nonresonance" secondary radiation will be observed during a time corresponding to the collective decay of the resonance system. It is important that both the energy and time dependences of both forms of secondary radiation depend directly on the localization of the atoms emitting this radiation.

Figure 1 shows the energy spectra and the corresponding temporal spectra of the "nonresonance" secondary radiation from atoms located at different depths in the resonance sample. The calculations were performed for the simplest model of a uniform semi-infinite resonance sample whose nuclear (χ_n) and electronic (χ_d) susceptibilities were chosen in the form

$$\chi_n = -\frac{F}{x+i}, \quad x = \frac{\hbar(\omega-\omega_R)}{\Gamma/2}, \quad F = 14.0\times 10^{-6}, \quad \Gamma = 0.4 \text{ mm/s}, \tag{1}$$

$$\chi_e = (-14.6+0.7i)\times 10^{-6}, \tag{2}$$

where ω_R is the resonance frequency. The width Γ of the resonance line was chosen to be much greater than the natural width of the Mössbauer level in ^{57}Fe on the basis of the fact that the unsplit resonance line can occur in an iron film with a high impurity content, which results in large inhomogeneous broadening and decreases the absolute value of the nuclear susceptibility χ_n.

The general theory of the yield of secondary radiation under the conditions of total external reflection from a Mössbauer mirror is presented in Refs. 12 and 13 . The Fourier transform for calculating the temporal spectra is calculated for the probability amplitude of the formation of secondary radiation at a depth z[1)]

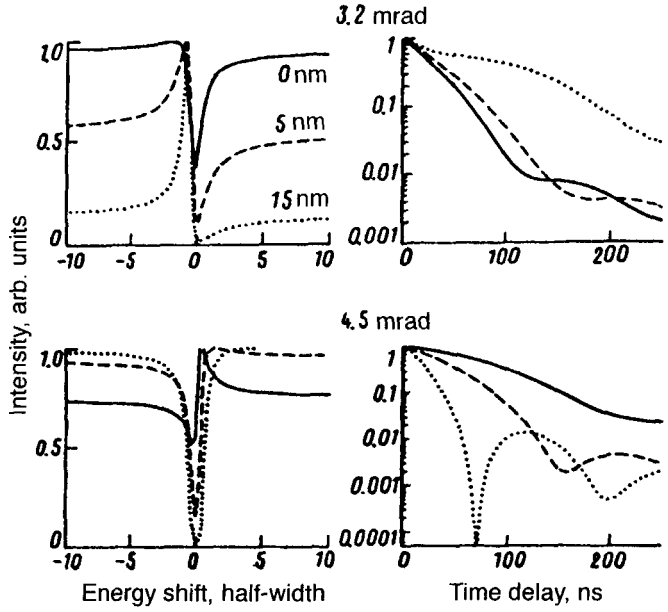

FIG. 1. Energy (left) and corresponding temporal (right) "nonresonance" spectra of secondary radiation formed at different depths (0 — solid lines; 5 — dashed lines, and 15 nm — dotted lines) in a semi-infinite resonance sample. The calculations are presented for two grazing angles of the incident radiation (3.2 and 4.5 mrad) near the critical angle of total external reflection (3.8 mrad for the chosen value of χ_e). The time-resolved spectra were normalized to $\exp(-\Gamma t)$ and to 1 at $t=+0$.

$$\mathbf{A}_s(z,t)=\frac{1}{2\pi}\int_{-\infty}^{\infty}\mathbf{A}_s(z,\omega)e^{-i\omega t}dt, \tag{3}$$

where for the cases of "resonance" ($s=n$) and "nonresonance" ($s=e$) secondary radiations, introducing normalization with respect to the intensity of the incident wave, we set correspondingly

$$\mathbf{A}_n(z,\omega)=(w_nFk/\sin\theta)^{1/2}\frac{\mathbf{E}(z,\omega)}{x+i},$$

$$\mathbf{A}_e(z,\omega)=(w_e\ \mathrm{Im}\ \chi_e k/\sin\theta)^{1/2}\mathbf{E}(z,\omega), \tag{4}$$

where the coefficients w_n and w_e determine the number of secondary particles which are of interest to us and which are produced with the absorption of one photon of the incident radiation; $k=2\pi/\lambda$; and θ is the grazing angle (for ^{57}Fe in our calculations $\lambda=0.086$ nm). The amplitude of the radiation field $\mathbf{E}(z,\omega)$ (resonance standing wave) was calculated in the simplest model using Fresnel's formula. If the secondary-radiation yield is set equal to 1 for simplicity, then the squared moduli of the amplitudes $|\mathbf{A}_s(z,\omega)|^2$ and $|\mathbf{A}_s(z,t)|^2$ are the energy and temporal spectra of the corresponding secondary-radiation yield. They are shown in Fig. 1 for the case of "nonresonance" secondary radiation.

We note that the temporal spectra of the secondary-radiation yield were analyzed in Ref. 15 , but the "nonresonance" secondary radiation, which is of greatest interest for us, was not studied there and the calculations were performed for a delayed-coincidence scheme in which the temporal structure of the exciting pulse corresponds not to a delta function, as in the case of a synchrotron radiation pulse, but rather to the natural decay of the Mössbauer nuclei in the source. The possibility of performing depth-differential investigations was not considered in Ref. 15 .

Since we are considering here, for simplicity, a single resonance line, the oscillations in the temporal spectra are due only to the dynamic interaction. The normalization chosen for the temporal spectra makes it possible to see directly the speed-up effect of the collective decay under the conditions of total external reflection. We call attention to the range of depths (0–15 nm) for which the calculations were performed — a large change in the structure of the radiation field and correspondingly in the form of the energy and temporal spectra does indeed occur in ultrathin surface layers.

The figure clearly shows that an investigation of, for example, the temporal spectrum of fluorescence radiation from nonresonance impurity atoms in a resonance sample determines uniquely the depth at which the centers are located. An ideal sample for observing the effect under discussion would be a rather thick ($\geqslant 50$ nm) ^{57}Fe film which contains at a fixed depth an ultrathin layer ($\sim 0.2-0.5$ nm) of any element whose K- or L-line fluorescence radiation can be separated from the fluorescence radiation of the iron atoms ($Z>8, Z \neq 26$). We note that samples with a rather large (several centimeters in the direction of the beam) ultrasmooth surface are required for total-reflection experiments.[6,7,10,14,16] A more realistic possible object of investigation is, for example, a thin ^{57}Fe film ($\leqslant 20$ nm) deposited on a glass substrate. If the substrate was obtained by pouring glass on liquid tin (as, for example, in Ref. 16), then the surface layer of the glass contains a substantial quantity of impurity tin atoms, and the thickness of the transitional layer, into which the tin penetrates, at the film–substrate boundary can be determined from the shape of the temporal spectrum of its fluorescence radiation. The resonance standing wave in this case must be calculated according to more general formulas which describe reflection from the system: resonance film–substrate. Interesting objects for such investigation of "nonresonance" secondary radiation are multilayer synthetic structures which consist of alternating layers of resonance and nonresonance atoms.

To increase the counting rate, the synchrotron measurements are often conducted in an integral time regime, excluding only the prompt response. In this case the angular dependence of the recorded signal yields information about the localization of the emitting atoms, just as in the case of the method of x-ray standing waves. Figure 2 shows the results of a calculation of such a dependence for "resonance" and "nonresonance" secondary radiations from atoms located at different depths. The "eating away" of the secondary-radiation intensity on the small-angle side for atoms emitting from larger depths is completely analogous to the changes in the curves over the emergence angle in grazing x-ray diffraction, where depth selection of the atoms can be achieved by changing the diffraction angle.[17] It is interesting to note that the absolute intensity of the "resonance" secondary radiation decreases with depth, whereas the intensity of the "nonresonance" radiation increases (we recall that here we ignore the absorption of the secondary radiation; for the depths ~ 10 nm considered here this is entirely justified even for

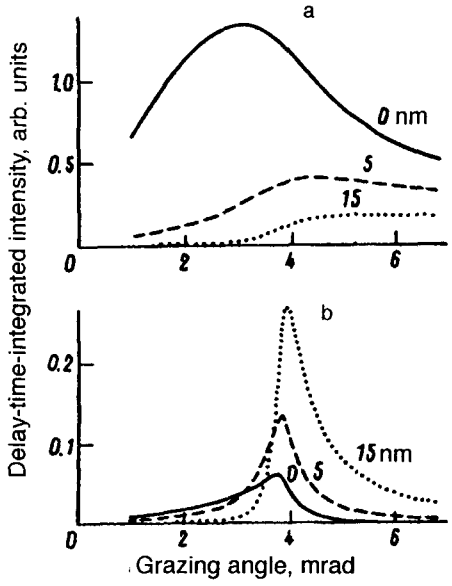

FIG. 2. Angular dependences of the integral, over the time delay (in the interval 1 — 300 ns), intensity of "resonance" (a) and "nonresonance" (b) secondary radiations (in relative units) for atoms located at different depths (0, 5, and 15 nm) in a semi-infinite resonance sample. The computational parameters are the same as for Fig. 1.

the case of secondary electrons). This attests to the fact that the conditions for collective excitation in the bulk are different from the conditions in the surface regions.

The investigations of secondary radiations which occur as a result of the interaction of synchrotron radiation with Mössbauer absorbers were initiated in Ref. 18. From our point of view, they open an extremely interesting field of structural studies and under the conditions of total external reflection they can yield unique information about the distribution profiles of different atoms in ultrathin surface layers.

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[1)]This representation is valid under the assumption that the exciting synchrotron radiation pulse is nearly instantaneous (prompt). This assumption corresponds to the experimental conditions for synchrotrons, where investigations of resonance scattering of nuclei are conducted (see, for example, Refs. 1 , 2 , 6 , 7 , and 14) and the pulse duration is equal to $\sim 0.1-0.2$ ns.

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