Effect of background asymmetry in conversion-electron Mössbauer spectroscopy with total external reflection

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An unusual behavior of the baseline has been observed experimentally in the Mössbauer spectra of conversion electrons in an arrangement of total external reflection. The observed asymmetry of the spectral tails is explained on the basis that the photoelectron yield acquires a resonant dependence.

The methods of grazing-incidence of Mössbauer optics have been attracting increasing interest. The primary reason is that there is the real possibility of achieving a resonant "supermonochromatization" of synchrotron radiation through the use of multilayer coatings and Mössbauer mirrors.[1,2] A second, and no less important, part of the research consists of attempts to develop a new method for studying surfaces. At grazing incidence, and under conditions of total external reflection, Mössbauer spectroscopy presents the possibility of obtaining information on the spatial profiles of hyperfine interactions in ultrathin surface layers and films, ~1–50 nm thick. The depth selectivity of conversion-electron Mössbauer spectroscopy, as the glancing angle is varied, is at least an order of magnitude better than that which can be achieved through energy analysis of the emitted electrons, as has been confirmed by some recent experiments.[3,4]

The interaction of Mössbauer radiation with resonant nuclei becomes substantially more complex near a surface under conditions of total external reflection, giving rise to some nontrivial features in the conversion-electron Mössbauer spectra. We have experimentally observed one such feature. It is the topic of the present letter.

Figure 1 shows experimental conversion-electron Mössbauer spectra. Frames *a* and *b* correspond to total external reflection, and frames *c* and *d* to specular reflection, from a film of metallic iron enriched to ~90% in ^{57}Fe. This film was ~40 nm thick and was grown by vacuum deposition on a beryllium substrate. The spectra shown here correspond to two glancing angles of the incident radiation: $\theta = 3.5$ and 4.2 mrad. These measurements were carried out on an SM 2101 TER spectrometer. The experimental apparatus is described in detail in Ref. 4. Since these spectra were recorded over a large velocity interval, one can easily see that the baseline is asymmetric on the right and left of the resonant region. This asymmetry changes sign when a critical

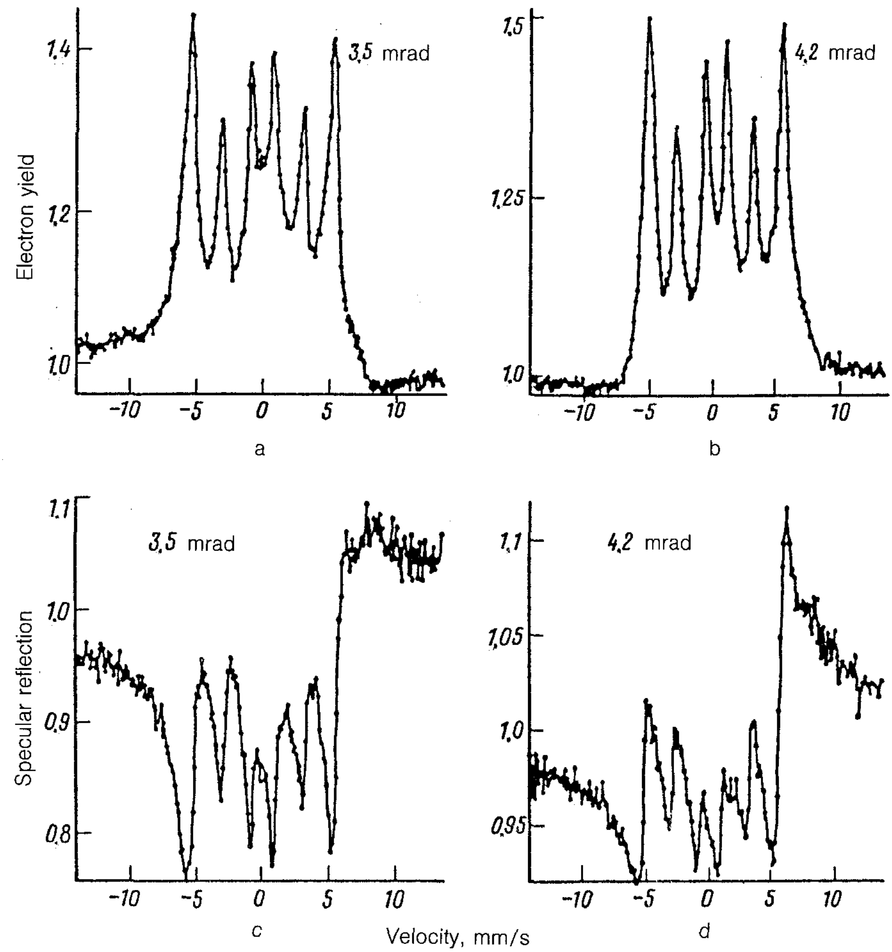

FIG. 1. Experimental conversion-electron Mössbauer spectra. a, b—Under conditions of total external reflection; c, d—specular reflection. The glancing angles are $\theta = 3.5$ and 4.2 mrad. The spectra have been normalized to the velocity of light at $v = \infty$.

angle θ_c is crossed (for metallic iron, it is $\theta_c \simeq 3.8$ mrad). With a further increase in θ, the asymmetry of the baseline disappears.

A previous calculation of the resonant spectrum of the yield of conversion electrons under conditions of total external reflection[4] correctly describes the general changes in the shape of the spectrum with the glancing angle, but it does not describe this asymmetry of the tails on the experimental spectrum.

The unusual effect observed here can be explained by arguing that the nonresonant background acquires a resonant dependence. In Mössbauer spectroscopy, the background can usually affect only the relative yield, having absolutely no effect on the shape of the spectrum. Under conditions of total external reflection, the situation changes radically. The distribution of the radiation field in the medium depends on the amplitude of the specularly reflected wave, having a strong, resonant dependence. Consequently, the photoelectron yield also acquires a resonant dependence.

A calculation has been carried out for a simple model of a single-layer medium with an isolated resonance. The linewidth was $\Gamma \simeq 6\Gamma_0$, where Γ_0 is the natural linewidth; the amplitude of the line was $A \simeq 0.3A_0$, where A_0 is the amplitude of the unsplit line; and the other parameter values correspond to metallic iron. The result of this calculation confirm the observed behavior of the tails on the Mössbauer spectra of the conversion electrons under conditions of total external reflection, as shown in Fig. 2, a–d.

At $\theta < \theta_c$, the depth to which the radiation penetrates into the medium is definitely smaller than the film thickness and also the photoelectron emission depth. Consequently, all the radiation which is not reflected can be converted into detectable sec-

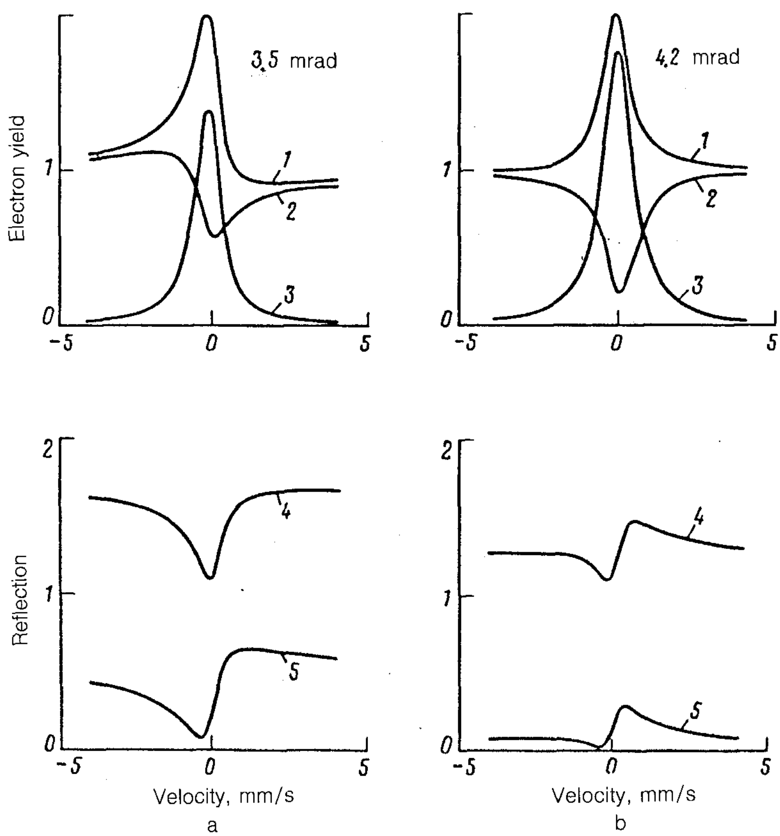

FIG. 2. Model-based calculations of the energy dependence of (1) the total electron yield, (2) the yield of photoelectrons, (3) the yield of conversion electrons, (4) the field amplitude at the surface, and (5) the specular-reflection coefficient for glancing angles (a) $\theta = 3.5$ and (b) 4.2 mrad.

ondary radiation. The energy dependence of the yield of nonresonant photoelectron in this case looks like the energy dependence for specular reflection upside down: The photoelectron yield is higher where the specular-reflection coefficient is lower. This effect had been seen previously in Refs. 5 and 6 on the angular dependence of the yield of fluorescence (secondary emission from a large depth) in the region of total external reflection.

At $\theta > \theta_c$, the radiation penetrates to a sharply greater depth—to a depth comparable to the photoelectron emission depth and the film thickness. The resonance curve of the photoelectron yield becomes more sensitive to the amplitude of the radiation field near the surface. In other words, the photoelectron yield becomes higher in the region in which the field amplitude at the surface is higher. The asymmetry of the tails changes sign. A similar dependence of the yield of photoelectrons with a small emission depth on the field amplitude at the surface (an increase near θ_c on the angular dependence) had been observed in Refs. 7 – 9.

Although the particular features of the yield of secondary emission during total external reflection of x radiation are well understood today,[5–10] these results have not been used to interpret the conversion-electron Mössbauer spectra under conditions of total external reflection. The reason is that the photoabsorption cross section is much smaller than the resonant-absorption cross section.

The magnitude of the asymmetry of the tails depends strongly on the relation between the yields of conversion electrons and photoelectrons. It is difficult to calculate this relation theoretically for multicomponent systems, with allowance for the multiplication of electrons of various energy groups. In the calculations, whose results are shown in Fig. 2, it was assumed that the photoelectron yield is a tenth of the conversion-electron yield exactly at resonance.

The magnitude of the asymmetry of the tails in the experimental spectrum is evidence that the yield of photoelectrons cannot be ignored in studies of conversion-electron Mössbauer spectra under conditions of total external reflection, even for enriched samples. In the case of a coherent interaction of Mössbauer radiation with a resonant medium for total external reflection or dynamic Mössbauer diffraction, the resonant dependence of the background may cause a serious distortion of the spectral shape, and it may change the interpretation of the results.

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