Interference of nuclear transitions in pure nuclear diffraction of 14.4-keV gamma rays by hematite

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The Mössbauer spectra of Bragg scattering of 14.4-keV resonant γ rays from Fe^{57} were measured under conditions of pure nuclear diffraction for all (from the first through the ninth) odd orders of reflection from the system of (111) planes of single-crystal α–Fe_2O_3. Interference was observed between the scattered γ quanta for different transitions of the hyperfine structure of the scatterer, as manifest in an asymmetry of the Mössbauer peaks. The measurement results are compared with theoretical calculations for ideal- or mosaic-crystal models.

Two types of interference, which appear when resonant Mössbauer γ rays interact with matter, have recently been observed in experiment and have become the subject of detailed study: 1) interference of elastic coherent scattering of γ rays by nuclei and electrons[1-3], 2) interference between the photoeffect and internal conversion[4-6]. We report here observation of a new type of interference, namely between various nuclear transitions between sublevels of the magnetic hyperfine structure in the diffraction of resonant γ rays.

Assume that the γ quanta are scattered by a crystal containing resonantly-scattering nuclei with a hyperfine level structure, under conditions when the Rayleigh scattering by the electrons is fully suppressed from structural considerations. In this case, pure nuclear diffraction is observed, and the amplitude of the scattering of the quanta by the unit cell of the crystal is described, apart from a constant factor, by the expression[7]

$$A \sim \sum_{MM_0} \left(2\frac{E-E_{MM_0}}{\Gamma}+i\right)^{-1} \sum_{j} \exp[i(\mathbf{k}_\beta-\mathbf{k}_\alpha)\rho_j]P(n_{jMM_0}). \qquad (1)$$

Here $\exp[i(\mathbf{k}_\beta - \mathbf{k}_\alpha)\cdot\rho_j]$ and $P(n_{jMM_0})$ are respectively the nuclear structural and polarization factors; M and M_0 are the magnetic quantum numbers of the excited and ground states of the nucleus; the summation with respect to j is only over the resonantly-scattering nuclei in the unit cell, while the summation with respect to M and M_0 is over all the hyperfine-structure transitions. The remaining notation is standard[3,7].

In our case there is no interference between the nuclear and electron scattering, and the spectral line contours are symmetrical in first-order approximation[8]. It follows from (1), however, that each γ-quantum energy E corresponds to different, generally nonzero amplitudes for scattering by all transitions of the hyperfine structure of the scatterer, so that interference should take between the scatterings on these transitions, and should manifest itself in the form of a line asymmetry in the energy dependence of the intensity of the diffracted radiation.

In mixed electron-nuclear diffraction, when the form of the spectrum is determined principally by the strong interference between the Rayleigh and resonant nuclear scattering, the referred-to interference between the nuclear transitions does not manifest itself in practice[3]. It turns out, however, that under conditions of pure nuclear diffraction this type of interference can be observed in explicit form.

The experiment was performed at room temperature with a Mössbauer diffractometer[9]. Gamma rays from a one-line Mössbauer source (Co^{57} in chromium, 200 μCi) were incident on single-crystal α-Fe_2O_3 (Fe^{57} content 85%, ~ 30″ mosaic) mounted in one of the positions of the symmetrical Bragg reflection (nnn). The crystal was in an external magnetic field ~1 kOe, which oriented the internal magnetic fields H_n at the iron nuclei parallel to the crystal surface in the scattering plane. The detector was a scintillation counter with NaI(Tl) crystal measuring $5 \times 10 \times 0.08$ mm.

We measured the dependence of the integral intensity of the Bragg reflections on the source velocity for all the existing odd orders of reflection (Fig. 1). The times of measurement of the individual spectra were approximately the same (~100 hours). The good effect/background ratio, reaching ~80 in the case of the high orders of reflection, was due to the absence of the diffraction of the 14.4-keV γ radiation by the hematite electrons and to the large scattering angle, which greatly suppressed the scattering of quanta having other energies by the hematite and by the diffractometer parts situated under the beam, and also by the high quality of the shielding and by the use of a thin NaI(Tl) crystal. The detector's own background in a 14-keV window was about 4 counts/hour.

The interference of the γ-quantum scattering by various transitions of the hyperfine structure was observed in all the obtained spectra. It was manifest most obviously in the fact that the internal slopes of the central pair of peaks, those facing zero velocity, are much steeper than the external ones. Figure 2 shows, in an enlarged scale, one of the central peaks of the (999) spectrum and the interference term separated from it, having the form of a dispersion curve. The value of the asymmetry (the ratio of the asymmetry of the dispersion curve to the height of the peak) amounts here to ~5%.

The sign of the asymmetry is determined by the ratio of the signs and magnitudes of the amplitudes for the γ-quantum scattering by all the transitions in the spectrum. For the outermost peaks, the experimental errors exceed the asymmetry, the value of which as predicted by the theory should be about 1.5%. The outer slopes of the peaks should then be steeper than the inner ones.

The value of the interference term decreases rapidly with increasing distance between resonances; for hematite these distances are larger by tens of times than the line width (~30 Γ_{nat} between the central peaks

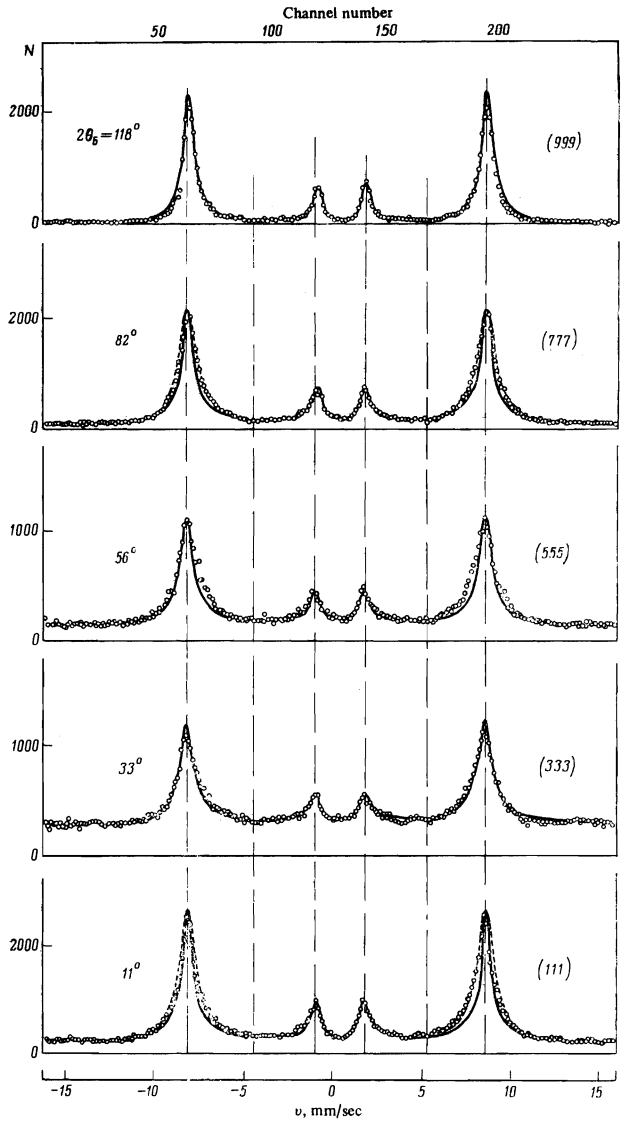

FIG. 1. Intensity of diffracted γ rays vs. the source velocity for odd orders of Bragg reflection (nnn) from single-crystal α-Fe_2O_3. The ordinates represent the number of counts in the analyzer channel. The vertical dashed lines show the positions of the nuclear resonances measured in transmission geometry. The solid lines are the theoretical curves calculated by the dynamic theory of the interaction of the γ quanta with ideal crystal; the dotted curves show the results of calculations for a mosaic crystal.

and ~70 Γ_{nat} from the outer peaks to the central pair). Nonetheless, the high sensitivity of the shape of the resonance lines, measured under conditions of pure nuclear diffraction, makes it possible to separate the interference term reliably.

Notice should be taken also of one other interesting detail in the spectra. For nuclear transitions with $\Delta m = 0$ (second and fifth resonances in the usual Mössbauer spectra of the iron), structural extinction of the nuclear scattering is observed, analogous to x-ray extinction. The experimental spectra have therefore no scattering peaks with $\Delta m = 0$. The absorption remains, however, and small dips appear at the locations of the resonances with $\Delta m = 0$, in all orders of reflection on the scattering "tails" from the remaining hyperfine-structure transitions. One of them is shown enlarged in Fig. 3.

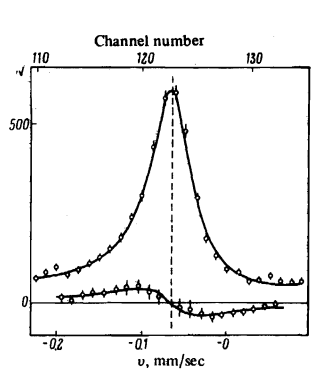

FIG. 2

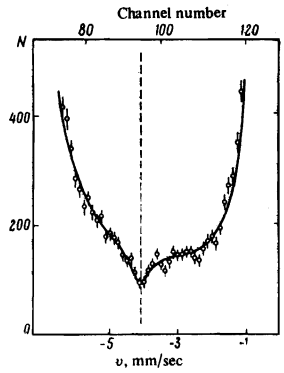

FIG. 3

FIG. 2. Interference of the scattering of γ quanta by various hyperfine-structure transitions, using a section of the (999) spectrum as an example (see Fig. 1). The lower curve shows the asymmetrical part of the resolution of the peak and represents the interference term. The solid curves were calculated theoretically from the dynamic theory.

FIG. 3. Section of the (999) spectrum in the region of the resonance with $\Delta m = 0$. A small dip is seen, due to the absorption of the γ quanta scattered by other transitions of the hyperfine structure. The solid theoretical curve was calculated from the dynamic theory.

The solid lines in Figs. 1—3 are the theoretical curves calculated with a computer from the dynamic theory of the interaction of γ rays with ideal crystals[7]; the main data on the calculation procedure and on the comparison with experiment are given in our earlier paper[3]. It is seen from the figure that the theoretical curves describe well all the characteristic features of the measured spectra. Some of the quantitative discrepancies observed in Fig. 1 can be ascribed to insufficient perfection of the crystal.

In the case of pure nuclear diffraction, the dynamic character of the interaction influences mainly the width of the resonance curve. The actual change of the width depends on the ratio of the extinction length to the absorption length. The difference between the curves calculated for the models of ideal and mosaic crystal should be largest when the depth of penetration is determined mainly by the extinction, and absorption plays a minor role. In our case, however, the situation is as a rule reversed, and the curves should be close to each other. Nonetheless, we calculated the curves for the reflection of resonant γ rays from a mosaic crystal. As expected, these curves also agree well with experiment. The dotted curves in the (111) and (777) reflection spectra (Fig. 1) show the results of the calculations for a mosaic crystal. For these spectra, the discrepancy between the two model is maximal. In the other cases the difference between the curves does not exceed the line thickness.

We note in conclusion that an analogous interference should be observed also under conditions of pure nuclear quadrupole diffraction[10].

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