Coherent excitation of Mössbauer nuclei by synchrotron radiation

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Diffraction of synchrotron radiation by Mössbauer nuclei has been observed for the first time. The measured temporal distribution of the resonantly scattered γ rays is radically different from that from the decay of isolated nuclei. The decay of excited nuclei in a crystal is directional in nature and occurs much more rapidly.

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Combining synchrotron radiation with the Mössbauer effect provides a unique opportunity for several interesting experiments. Kagan *et al.*[1] examined the temporal characteristics of Mössbauer diffraction and predicted in particular, that they would be radically different from those for the decay of isolated nuclei.

The time evolution of diffraction has not been observed previously for any type of radiation. The primary difficulties in Mössbauer synchrotron-radiation experiments stem from the extremely low ratio of the number of Mössbauer γ rays to the total flux (10^{-13} for ^{57}Fe). We have previously suggested that purely nuclear reflections from a single crystal might be used to resolve this problem.[2] Other versions of this experiment have not yet met with success.[3,4]

The present experiments were carried out in the synchrotron-radiation beam from the superconducting "snake" of the VÉPP-3 electron storage ring of the Institute of Nuclear Physics, Novosibirsk (the electron energy was $E_e = 1.94$ GeV; the field in the

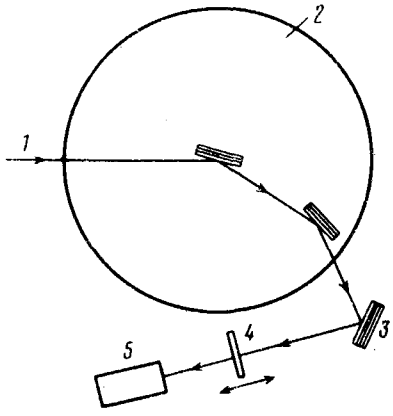

FIG. 1. Experimental arrangement. 1—Synchrotron radiation beam; 2—monochromator [two Si single crystals in the dispersive position; the (111) reflection]; 3—enriched α-$^{57}Fe_2O_3$ hematite single crystal; 4—absorber of polycrystalline hematite (10 mg/cm^2); 5—scintillation detector, NaI(Tl) with an FÉU-85 photomultiplier.

snake was $H = 29$ kOe; the critical energy of the synchrotron-radiation spectrum was $\varepsilon_c = 7.3$ keV; the average electron current was $I_e = 10$ mA; the length of the synchrotron-radiation pulse was 1 ns; and the pulse repetition period was 250 ns; Ref. 5). The experimental arrangement is shown in Fig. 1. The synchrotron-radiation beam passes through a two-crystal monochromator with $\Delta E/E = 1.4\times10^{-4}$ [Si (111) single crystals in the dispersive position], adjusted beforehand to the Mössbauer transition energy $E_M = 14.4$ keV with the help of a ^{57}Co source. The relative number of resonant γ rays in the beam emitted from the monochromator was 2×10^{-9}. This beam was incident on an α-$^{57}Fe_2O_3$ single crystal (85% enrichment, mosaic angle $\sim$30") oriented in the position corresponding to the (777) purely nuclear reflection, in which an electron reflection is structurally forbidden.[6] The use of the purely nuclear reflection made it possible to suppress the nonresonant coherent background to the extent that the detector count rate was determined primarily by the fluorescence of iron atoms in the hematite crystal and by other incoherent processes. The background was lowered even further by the high degree of polarization of the synchrotron radiation; a reflection with an angle of nearly 90° was selected [$2\theta_B(777) = 82°$]. The solid angle of the hematite detector was $\sim10^{-4}$ sr. The search for the purely nuclear reflection was carried out in a region calculated from the measured positions of the allowed (666) and (888) reflections. At each angular point we measured the intensity of the radiation at the detector with a thick Mössbauer absorber of polycrystalline α-$^{57}Fe_2O_3$; this Mössbauer absorber was either at rest (N_0) or moving at an "infinite" velocity (N_∞). A special arrangement for cyclically switching the absorber velocity maintained a balance between the N_0 and N_∞ channels within 0.1%.

A resonant-diffraction peak was detected in two independent series of measurements, between which we carried out a complete readjustment and calibration of the x-ray–optics system. In the (777) diffraction peak which we observed the ratio of the count rates in the cases of a moving absorber and an absorber at rest was

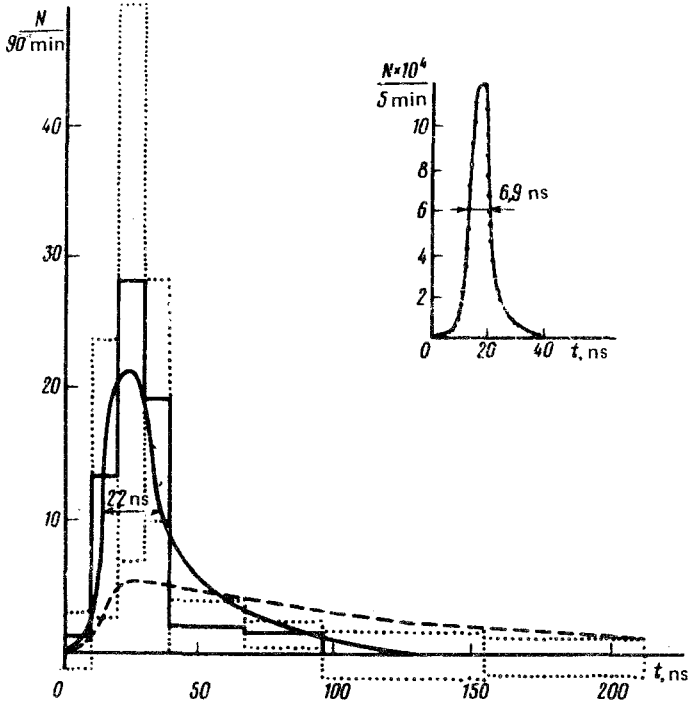

FIG. 2. Time distribution of the resonant γ rays diffracted by the hematite single crystal. Solid curve—calculated for a nonexponential decay[1] ($\chi^2 = 7$ for six degrees of freedom); dashed curve—calculated for an exponential decay of isolated ^{57}Fe nuclei ($\chi^2 = 14.55$ for six degrees of freedom). The points on the histogram show the systematic measurement errors. The inset shows the exciting synchrotron-radiation pulse, measured for the instantaneously scattered nonresonant γ rays (this is the instrumental function).

$N_\infty/N_0 = 1.29 \pm 0.16$ in the first series of measurements and 1.18 ± 0.09 in the second; the sum over the two series of measurements was $N_\infty/N_0 = 1.20 \pm 0.08$.

In addition to measuring the intensity, we measured the temporal distribution of the γ rays in the N_0 and N channels at each angular point. The detector and the time-evolution electronics provided a resolution of 6.9 ns (Ref. 7). The inset in Fig. 2 shows the exciting synchrotron-radiation pulse measured for the instantaneously scattered nonresonant γ rays (the instrumental function). Figure 2 shows the temporal distribution of the resonant γ rays, $N_r(t) = N_\infty(t) - N_0(t)$, in the diffraction peak. The solid curve is a convolution of the instrumental function with a nonexponential decay law[1] [for $t > 10$ ns, the decay is described by $(1/\tau)\exp(-\tau)$, where $\tau = t/\tau_I$, t is the time, and $\tau_I = 142$ ns is the lifetime of the ^{57m}Fe excited level]. A curve calculated in the approximation of a "black" absorber gives a rather good description of the experimental points: $\chi^2 = 7$ for six degrees of freedom (the adjustable parameter is the intensity). A similar procedure was followed for an exponential decay law $\exp(-\tau)$ ($\chi^2 = 14.55$ for six degrees of fredom). For a more reliable hypothesis, the experimental data were divided into four intervals (28.8 and 3×57.6 ns), and a procedure similar to that described above was carried out. For the exponential decay law we found $\chi^2 = 18.34$

with two degrees of freedom. On this basis we can very confidently reject this hypothesis. (The probability for obtaining $\chi^2 = 18.34$ with two degrees of freedom as a result of statisfical fluctuations is $\approx 10^{-4}$.) For the nonexponential decay law we found $\chi^2 = 2.9$ with two degrees of freedom, so that we find no contradiction of the hypothesis.

In summary, we have been able to obtain a beam of Mössbauer γ rays from synchrotron radiation with a continuous spectrum. The intensity of the γ rays amounts to 10^{-13} of the total synchrotron-radiation flux. We have measured the temporal distribution of the Mössbauer γ rays diffracted by a single crystal. The decay of the excited nuclei in the crystal is a directional process, in contrast with the isotropic decay of isolated nuclei, and it occurs much more rapidly, implying that the process is of a collective nature, in agreement with the theoretical predictions.[1]

It may be said that these new time-varying experiments open up a new research field: time-varying γ-ray diffraction.

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