Accelerated decay of a nuclear excitation in a crystal during resonant γ scattering

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The time dependence of the coherent scattering of resonant γ rays by nuclei in a crystal has been measured. For the first time, it has been found possible to observe an acceleration of the decay of an excited nuclear state along the direction of the Bragg angle.

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The duration of the resonant interaction of a γ ray with a nucleus is determined by the lifetime of the excited state which is formed after the capture of the γ ray. This time is very long for the low-lying isomer states which are characteristic of Mössbauer transitions. For the interaction of a γ ray with a system of resonant nuclei forming a crystal it seems natural to assume that the decay of each individual excited nucleus occurs independently, especially since the nuclei essentially do not interact with each other.

As far back as the early 1960's it was suggested that under certain conditions we might expect a mutual effect of nuclear oscillators in an emission process, resulting in changes in the decay characteristics of the excited state.[1]

The problem was first studied systematically by Kagan and Afanas'ev,[2] who introduced the idea of a delocalized collective excitation in a system of identical nuclei in a crystal (a nuclear exciton). They showed that a coherence-conserving decay of a collective excitation occurs only along the direction of the primary γ ray and at the Bragg angle. It follows directly from the expressions which they derived for probabilities that there would be a substantial acceleration of the decay through coherent channels and that this phenomenon could be observed in scattering experiments. The theory of the delay of γ rays in resonant Bragg scattering was developed further in Ref. 3.

It is important to note that the acceleration of the decay can be observed only in a coherent elastic channel, since in other cases—spin-flip scattering, the production of a conversion electron, etc.— there would always be some indication of a specific nucleus that has undergone decay; i.e., the collective nature of the interaction would be lost, and we would evidently see manifestations of the decay of an individual nucleus.

Although this problem has continued to attract considerable interest, attempts to observe the theoretically predicted effect have so far been unsuccessful.

In order to study accelerated decay we need to identify a time which marks the beginning or end of the excitation of the nuclei in the scattering crystal in some way, and we need to study the time evolution of the subsequent emission of γ rays.

In principle, we could use the delayed-coincidence method, which has been used previously in experiments on the time evolution of incoherent scattering in polycrystalline samples (see Refs. 4 and 5, for example). However, this method places a limit on the source activity (which cannot be higher than 10^6 Bq because of the danger of false coincidences), so it cannot be used if there is a hard collimation of a γ-ray beam.

Another possibility is to use pulsed synchrotron radiation. This approach, however, runs into the formidable difficulty of filtering out from a white spectrum a Mössbauer component at a relative intensity $\sim 10^{-14}$. The first results of experiments by Chechin *et al.*[6] on the coherent pulsed excitation of Fe^{57} nuclei by synchrotron radiation have recently been published.

In the present experiments we used a method involving the rapid opening of a shutter blocking the beam of Mössbauer γ rays, so that we could determine the time at which the resonant radiation arrives at the system of scattering nuclei. In this approach the accuracy at which the origin on the time scale is set is determined by the rise time of the γ beam, τ_r, i.e., by the velocity properties of the shutter. Exceedingly stringent demands must be met by the shutter. The rise time τ_r must be much shorter than the lifetime of the excited nuclear state, τ_0 ($\tau_0 = 142$ ns for the Fe^{57} nucleus). We were able to solve the shutter problem by making use of the rapid magnetization reversal[7] of $FeBO_3$ crystals. High-quality iron borate crystals can undergo a change in magnetization state in a time of 2–5 ns in pulsed magnetic fields ~ 5 Oe. The use of these crystals in a special shutter (described in detail in Ref. 8) allowed us to produce approximately square γ pulses with a rise time $\tau_r \leqslant 15$ ns. This method has the important advantage that Mössbauer sources of any attainable activity can be used; this flexibility is required in diffraction experiments, with their low luminosity.

This shutter has been used for the first study of the time dependence of the coherent scattering of Mössbauer γ rays. We used the emission of Fe^{57} nuclei and an ideal $Fe^{57}BO_3$ crystal as a scattering object.

Figure 1 shows the basic experimental apparatus: the Co^{57} Mössbauer source in chromium; the γ shutter, which consists of two $Fe^{57}BO_3$ crystals, a Helmholtz-coil magnetizing device, and a current pulse generator; the $Fe^{57}BO_3$ crystal whose scattering was studied; the detector; and the time-resolving spectrometer.[9]

The Mössbauer spectrum of iron-57 in the crystal $Fe^{57}BO_3$ consists of six lines of a magnetic hyperfine splitting. All the experiments described here were carried out with the source line tuned to the resonant nuclear transition $+1/2 \rightarrow +1/2$ in the crystal.

The experimental geometry was determined by the directions of the wave vectors $\mathbf{k}_0$ (the incident beam) and $\mathbf{k}_1$ (the scattered beam), which define the scattering plane, and by the goniometer axis 00′, which runs perpendicular to the scattering plane. The test crystal of iron borate was held by the goniometer.

The weakly ferromagnetic $Fe^{57}BO_3$ crystals were thin single-crystal wafers with surfaces corresponding to the (111) plane: the easy-magnetization plane. The first two crystals were each 22 ± 2 μm thick, while the third was 150 μm thick. The degree of enrichment in the resonant isotope Fe^{57} was 95%. All the crystals were of high structural quality.

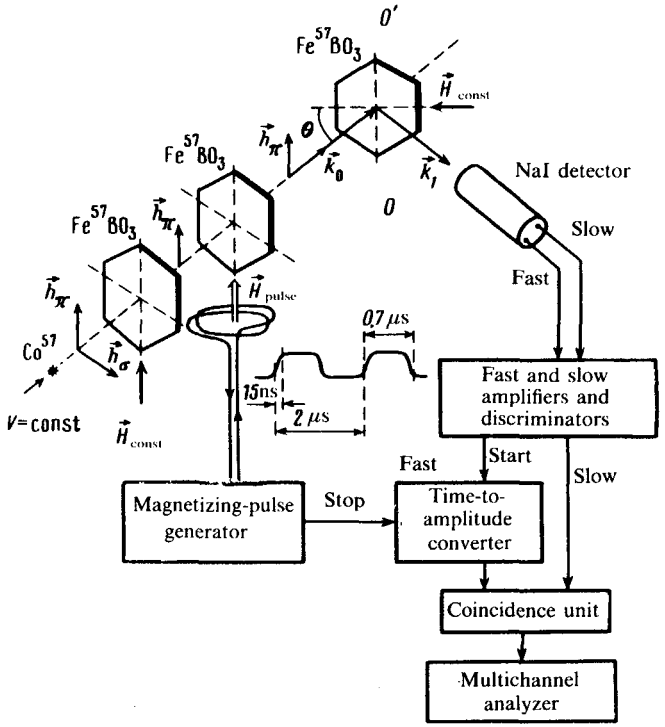

FIG. 1. The experimental apparatus.

The crystals used in the shutter were positioned perpendicular to the γ beam. The first crystal was kept magnetized at all times by an external field $\mathbf{H}_{const}$ perpendicular to $(\mathbf{k}_0,\mathbf{k}_1)$. This crystal served as a polarizer and transmitted primarily the π-polarized component of the radiation. The next iron borate crystal through which the radiation passed was subjected to a pulsed magnetic field $\mathbf{H}_{pulse}$ directed along $\mathbf{H}_{const}$. This crystal was switched from an unmagnetized, multidomain state (opaque to the π component) to a magnetized single-domain state which transmitted the π component. Pulses of the π-polarized component of the γ radiation were produced in the course of these transitions.

Figure 2a shows the time dependence of the intensity of the transmitted γ beam. Each channel is 7.2 ns wide. The results clearly show a step with a rise time $\tau_r \leqslant 15$ ns (we are not concerned here with the return to the original state). This rise time turns out to be short enough for studying the time evolution of the nuclear-scattering processes of interest.

The third crystal, whose scattering was studied, was magnetized constantly in the $(\mathbf{k}_0,\mathbf{k}_1)$ plane, so that only the chopped π-polarized component of the radiation could interact with the nuclei in it. To study the coherent scattering put the crystal in the orientation corresponding to Bragg nuclear reflection. We used the strongest, purely nuclear scattering for the $\Delta m = 0$ resonance in the $Fe^{57}BO_3$ crystal: (222), with[10] $\theta_B = 10.26°$.

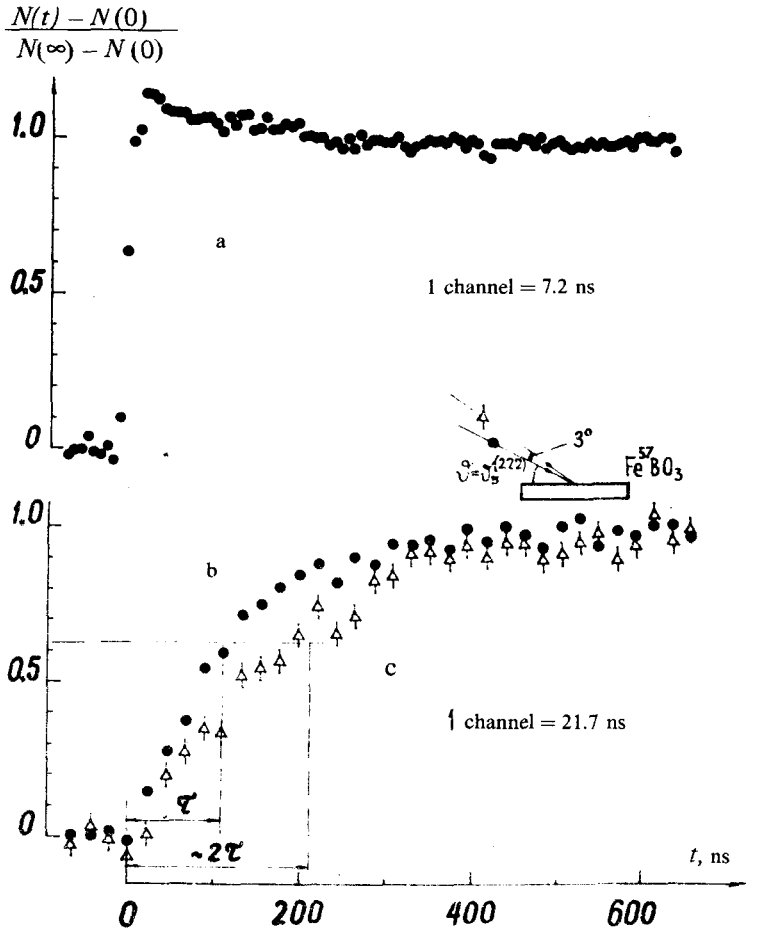

FIG. 2. a—Time dependence of the γ intensity beyond the magnetic-resonance shutter; b—time dependence of the Bragg scattering (filled circles); c—time dependence of the incoherent scattering (triangles).

Curve b in Fig. 2 shows the measured time evolution of the Bragg scattering. Each point here represents 21.7 ns. As expected, the resonant scattering is delayed with respect to the incoming γ-radiation front: The steep step is transformed into a gentle rise. The point of primary interest, however, is the way in which the intensity rises, since the behavior carries information about the lifetime of the excited state of the nuclear system.

To identify the acceleration effect in the decay of the collective excitation, we also measured the time dependence of resonant incoherent scattering. For this purpose the crystal was rotated 3° away from the Bragg position. No coherent scattering occurs in this position, and the γ rays arriving at the detector are due exclusively to incoherent scattering, e.g., scattering involving a flipping of the nuclear spin in the ground state or an excitation (or absorption) of a phonon. We have discussed this problem previous-

ly.[11] In the measurements of the time evolution of the incoherent scattering, the detector was brought to within ~ 4 cm of the crystal to increase the solid angle. The average angle of the γ scattering in these measurements was 90°. Curve c in Fig. 2 shows the result of the measurements of the time evolution of the non-Bragg scattering.

A quick comparison of curves b and c shows that the Bragg scattering occurs much more rapidly than the non-Bragg scattering, for scattering by the same system of nuclei in an ideal crystal. A given intensity level, e.g., the level of $1 - e^{-1} = 0.63$ used in the construction in Fig. 2, is reached approximately twice as fast under coherent-scattering conditions as under incoherent-scattering conditions. *In this result we can thus see direct evidence of an acceleration of the decay of a collective excitation of nuclei in a crystal during coherent resonant scattering of γ rays.* The time evolution of the two processes is substantially different even if the incident beam is poorly collimated (with a divergence $\sim 1°$ in the present experiments); the difference should be much more apparent in the case of a hard collimation near the Bragg angle.

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