Observation of the Full Time Evolution of the Nuclear Collective-Decay Mode in Crystalline $^{57}Fe_2O_3$ Excited by Synchrotron Radiation

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(Received 10 August 1988)

The full time evolution of the coherent deexcitation of the 14.4-keV nuclear resonance, excited by synchrotron radiation in a single crystal of the simple antiferromagnet $^{57}Fe_2O_3$, has been measured. The buildup from zero decay probability at time zero following excitation, increasing to a maximum 2.4 nsec later, has been observed experimentally for the first time. The results are in good agreement with calculations based on the dynamical theory of nuclear Bragg scattering.

PACS numbers: 76.80.+y, 07.85.+n, 25.20.Dc, 78.70.Ck

The unique temporal properties of synchrotron radiation (SR) are ideally suited to the study of physical processes in the range of one to several hundred nanoseconds. For resonant nuclear Bragg scattering (NBS), this time range spans the lifetimes of several low-lying nuclear levels in isotopes which have been proposed as candidates for resonant filtering of SR. In a crystalline environment the coherent interaction between the electromagnetic field and the periodically ordered nuclear scatterers results in a drastic reduction in the effective nuclear decay time compared to that of an isolated nucleus.[1-3] Furthermore, in the case of nuclear levels split by hyperfine fields, a modulation of the time dependence occurs, resulting from interference between the various hyperfine components having different energies in the solid.[4] The existence of these effects was demonstrated for ^{57}Fe in yttrium iron garnet (YIG) and $FeBO_3$ in previous studies performed at HASYLAB.[5-7]

In this Letter we report the study of the time evolution of the Bragg scattered radiation from Fe_2O_3 (α-hematite), a simple antiferromagnet. The high angular and energy resolution of the premonochromator system built for the study of nuclear Bragg scattering[8,9] is shown to almost completely eliminate the prompt background, and as expected, has allowed the full time evolution of the deexcitation process from excitation ($t=0$) to be measured. In particular, for a simple antiferromagnet the prediction of zero intensity at $t=0$ followed by the rapid buildup and subsequent decay of the scattered intensity within 4 nsec is observed experimentally.

This result is of general significance because it demonstrates the possibility of the use of isotopes (for NBS monochromators of SR) that have significantly shorter lifetimes than that of ^{57}Fe ($T_{1/2}=98$ nsec). The use of NBS monochromators will make possible a wide range of experiments in the microvolt and submicrovolt regime; for example, inelastic x-ray scattering and interferometry with longitudinal coherence lengths of tens of centimeters. To date the rates achieved from NBS with use of ^{57}Fe are in the range of 1 to 10 photons/sec. With the use of shorter-lived isotopes (wider bandwidth), a larger fraction of the incident SR spectrum could be extracted while still maintaining a relatively narrow energy resolution, which could make a variety of new experiments feasible.

The measurements were made at the Cornell High Energy Synchrotron Source (CHESS). The six-pole wiggler installed on the storage ring provided the radiation. Machine parameters were typically 35–60 mA of stored current at 5-GeV electron energy. The experimental arrangement was the same as in our previous experiment,[8] except that the hyperpure Ge solid-state detector was replaced by a doped BaF_2 scintillation counter to provide the needed time resolution. The BaF_2 signal was input to a fast-slow coincidence network that discriminated against the high- and low-energy noise. A *p-i-n* semiconductor diode, placed so that it sampled the wiggler beam, was used to provide a $t=0$ reference time signal. The data were collected with a time-to-amplitude converter and a multichannel pulse-height analyzer. The time resolution of its combination was <1 nsec.

A 3.0×0.8-mm^2 cross section of the output beam from the high-resolution premonochromator[8] was incident on the α-hematite, 93% enriched in ^{57}Fe, set to satisfy the diffraction condition for the (777) (rhombohedral unit cell) pure nuclear antiferromagnetic Bragg peak. The crystal was placed in a static external magnetic field of ~1 kG in order to align the internal field in the sample. The magnetic field direction was parallel to the scattering plane (nuclear quantization axis perpendicular to the scattering plane). The typical resonant photon rate after the nuclear scatterer was 10–15 sec^{-1}, as measured by the Ge solid-state detector. The nonresonant background was about 0.02 sec^{-1}. Replacement of the Ge detector by the BaF_2 counter decreased the number of resonant quanta counted after the fast-slow coincidence unit to about 5 sec^{-1} and the background level increased to 0.5 sec^{-1}. The initial search for the pure nuclear reflection was carried out by use of the Ge detector in the same way as described in Ref. 8. Once set at the proper energy and Bragg angle, the Ge detector was replaced by the BaF_2 scintillator.

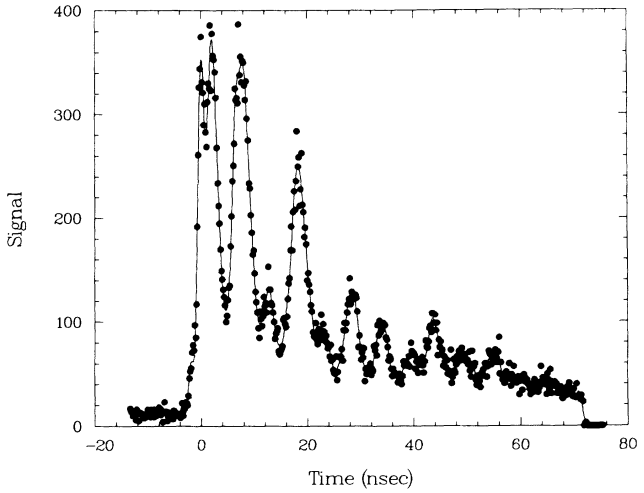

FIG. 1. The observed time evolution of the "pure-nuclear" (777) reflection from ^{57}Fe-enriched hematite. The continuous curve is derived from the data by Fourier smoothing.

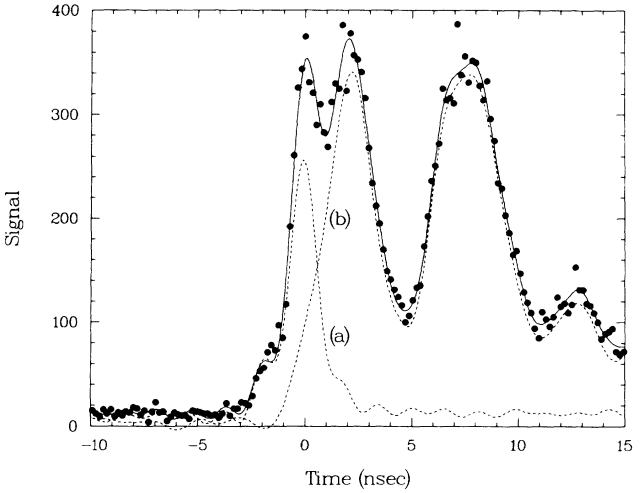

FIG. 2. The first 15 nsec of the time spectrum is shown, together with its smoothed curve. The broken curves are (*a*) the Fourier smoothed off-resonance spectrum and (*b*) the result of the subtraction of this spectrum from the smoothed resonant spectrum.

The time dependence of the deexcitation process from the $^{57}Fe_2O_3$ single crystal was measured at the peak of the rocking curve for the nuclear (777) Bragg peak. A typical spectrum is shown in Fig. 1. The solid line in the figure is a Fourier smoothed curve and is used in the analysis below to facilitate subtraction of the prompt, nonnuclear component from the observed spectrum. It is already clear from Fig. 1 that the full time evolution has been measured. The data are characteristic of a pure nuclear reflection from a simple antiferromagnet[6]; they show an alternating pattern of low and high peaks which result from the interference of the two pairs of lines of unequal strength corresponding to the $\Delta m = \pm 1$ transitions of the hyperfine-split spectrum.

An "off-resonance" spectrum was collected for use in further analysis of the resonance spectrum. This was done by our setting the energy of the probe beam off resonance by 25 meV. At this setting the only signal was a small peak at $t = 0$ presumed to be due to background scattering or magnetic scattering from electrons. This prompt peak was also Fourier smoothed and then scaled and subtracted from the smoothed resonant spectrum. The scaling was done so as to provide a smooth leading edge to the "subtracted" spectrum. For clarity of presentation, only the first 15 nsec of the result are shown in Fig. 2. The solid curve is the smoothed curve from Fig. 1 and the dashed curves are the scaled prompt component and the resultant after subtraction of this from the smoothed raw data.

Figure 3 is a comparison between the data with the "background" subtracted and a calculation. The calculation is based on the formalism of Ref. 3 including the effects of the four $\Delta m = \pm 1$ lines which contribute to the time spectrum modulation. Literature[10] values of the hyperfine field, 515 kOe, and quadrupole splitting, 0.12 mm/sec, were used. Convolution of the experimental time resolution is included. It is important to note that the above analysis assumed that the subtracted component arose only from the scattering by electrons and was not of nuclear origin, since it is nearly impossible to scale, experimentally, the "prompt" observed component off resonance with the on-resonance signal. Finally, in the calculation, the angular integration has been carried out with the assumption of a Gaussian mosaic distribution whose width, 0.6 arcsec, and position relative to $\theta = \theta_B$ of -0.14 arcsec have been adjusted to give

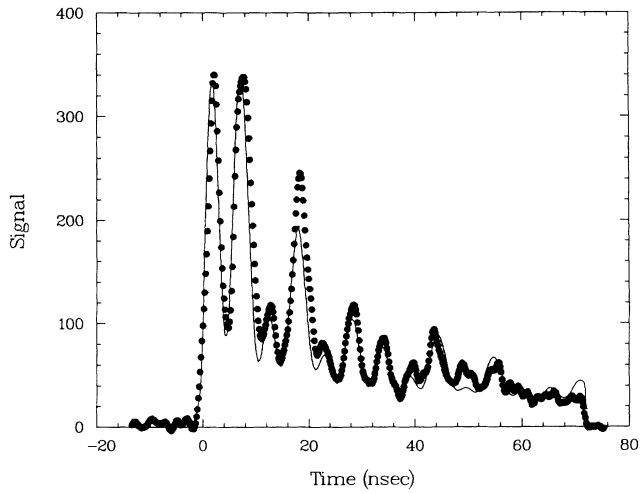

FIG. 3. The smoothed experimental data with its prompt background removed is compared with a dynamical diffraction calculation (continuous curve) described in the text.

reasonable agreement with the observed, smoothed data. The values chosen are consistent with the observed rocking-curve width of 3 arcsec and incident divergence of 0.4 arcsec.

The subtracted spectrum clearly shows the time dependence of the scattered intensity from a pure nuclear Bragg peak in a simple antiferromagnet excited by SR. The relatively broad-band incoming SR pulse excites all of the hyperfine-split levels in the ^{57}Fe nuclei simultaneously. They are consequently phase coherent at the moment of excitation. Since the crystallographic phase difference between atomic sites in the unit cell is $(2n+1)\pi$, destructive interference occurs, giving zero intensity at this time. This is evident in both Figs. 2 and 3. As one follows the process in time, the phase relationship between transitions of different energies is changing continuously. Thus the intensity increases from zero and fluctuates according to the changing phase difference between the oscillators. In the case of Fe_2O_3, the energy difference between the first $(\frac{1}{2}^+ \rightarrow \frac{3}{2}^+)$ and sixth $(\frac{1}{2}^- \rightarrow \frac{3}{2}^-)$ lines, caused by the magnetic hyperfine interaction, corresponds to a $\sim$5-nsec beat period.[6] The interference between the two inner lines $(\frac{1}{2}^+ \rightarrow \frac{1}{2}^-$ and $\frac{1}{2}^- \rightarrow \frac{1}{2}^+)$ has approximately twice this period. These two beat frequencies combine to give the alternating strong and weak maxima seen in Fig. 3. Our results show a similar behavior to that seen in the simple antiferromagnet $FeBO_3$.[7] The $FeBO_3$ data did not, however, show the early stages of the time evolution, and were thus unable to demonstrate agreement with calculation in this important region.

In summary, the complete time evolution, from $t=0$, of the deexcitation of coherent nuclear states was observed in the nuclear Bragg scattering process at the (777) pure nuclear reflection of the hematite single crystal using a highly monochromatic, collimated probe beam. The time spectrum shows a rapid buildup from zero at the instant of excitation followed by a "quantum beat" modulation characteristic of simple antiferromagnets containing ^{57}Fe. A strong speedup of the coherent decay manifests itself in increased intensity of the first peak in the spectrum at $\sim$2.4 nsec and strong damping of the quantum beat modulation. These results demonstrate that it is now technically possible to extend the study of nuclear Bragg scattering to resonant systems having faster decay rates than ^{57}Fe. Such systems may be crucial to the solution of the problem of providing the needed intensities for some of the proposed uses of resonantly filtered SR beams.

This research was carried out at Brookhaven National Laboratory under Contract No. DE-AC02-76CH00016 with the U.S. Department of Energy. Preparation of the $^{57}Fe_2O_3$ crystal by A. Cooper and J. Remeika of AT&T Bell Laboratories is gratefully acknowledge. The authors also wish to thank C. Woody of BNL for loan of the BaF_2 detector and D. Mills of CHESS for loan of the *p-i-n* diode. Hospitality and technical assistance by the CHESS facility during the performance of this research are gratefully acknowledged.

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[1]G. T. Trammel, in *Chemical Effects of Nuclear Transformations* (IAEA, Vienna, 1961), Vol. 1, p. 75.

[2]J. P. Hannon and G. T. Trammel, Phys. Rev. **186**, 306 (1969).

[3]Yu. Kagan, A. M. Afanas'ev, and V. G. Khon, J. Phys. C **12**, 615 (1979).

[4]G. T. Trammel and J. P. Hannon, Phys. Rev. B **18**, 165 (1978).

[5]E. Gerdau, R. Ruffer, R. Hollatz, and J. P. Hannon, Phys. Rev. Lett. **57** 1141 (1986).

[6]R. Rüffer, E. Gerdau, R. Hollatz, and J. P. Hannon, Phys. Rev. Lett. **58**, 2359 (1987).

[7]U. van Bürck, R. L. Mössbauer, E. Gerdau, R. Rüffer, R. Hollatz, G. V. Smirnov, and J. P. Hannon, Phys. Rev. Lett. **59**, 355 (1987).

[8]G. Faigel, D. P. Siddons, J. B. Hastings, P. E. Haustein, J. R. Grover, J. P. Remeika, and A. S. Cooper, Phys. Rev. Lett. **58**, 2699 (1987).

[9]D. P. Siddons, J. B. Hastings, and G. Faigel, Nucl. Instrum. Methods Phys. Res., Sect. A **266**, 329 (1988).

[10]O. C. Kistner and A. W. Sunyar, Phys. Rev. Lett. **4**, 412 (1960).