New Approach to the Study of Nuclear Bragg Scattering of Synchrotron Radiation

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Nuclear Bragg scattering of synchrotron radiation from isotopically enriched $^{57}Fe_2O_3$ has been observed at a signal-to-noise ratio of 100:1 without delayed-coincidence techniques. The use of a high-resolution premonochromator providing a 5-meV energy spread and 0.4″ collimation allows for high-resolution studies of dynamical diffraction effects. In particular, the observed intensity of the pure nuclear (7 7 7) reflection implies a dramatic decrease of the effective lifetime due to coherent scattering.

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The monochromatization of synchrotron radiation (SR) to bandwidths of 10^{-6} to 10^{-8} eV will open up new fields of research with important applications in diverse areas of physics, chemistry, and the materials sciences. The high energy resolution, polarization properties, long coherence length, and the small angular divergence of such beams will be unique. As has been pointed out in earlier papers,[1-3] the most promising methods to achieve these properties are the use of either nuclear resonant Bragg scattering or grazing-incidence nuclear total reflection of SR. Several studies have been successful in the production of resonantly filtered beams.[4,5] Delayed coincidence techniques have been employed in these works to elucidate the time evolution of the resonant scattering. From examination of the high-quality data of Gerdau *et al.*,[4] it is clear that, in the early part of the time evolution, the prompt photons coming from diffuse and inelastic electronic scattering mask the behavior of the resonant scattering. This background of unwanted radiation is unavoidable with the use of previous techniques, and limits the broad application of resonant nuclear scattering for several reasons. First, the number of isotopes which are candidates for nuclear resonant monochromators is limited by the lifetime of their excited states. This should be longer than approximately 5 nsec, since it is instrumentally rather difficult to achieve efficient gating in times shorter than this. This restriction becomes more severe when one considers the reduction of the effective excited-state lifetime predicted by the dynamical theory of resonant scattering for perfect crystals.[6-8] Second, by excluding the early part of the resonantly scattered radiation one loses that part of the spectrum which may contain the largest number of photons. Third, to prove the predictions of the dynamical theory, i.e., the collective nature of the scattering, it is important to study the time dependence of the resonantly scattered photons from the moment of the excitation.

In this Letter we demonstrate the possibility of studying the complete time evolution of nuclear Bragg scattering. A monochromator system for the study of nuclear Bragg scattering has been built with use of a bending-magnet source on the x-ray ring of the Brookhaven National Synchrotron Light Source and we have shown that it is possible to isolate resonant photons from the SR continuum without temporal suppression of prompt radiation, while providing excellent signal-to-noise ratios.

The dynamical effects in resonant scattering from a perfect crystal can best be studied by our exciting the scatterer with an incoming plane wave at (or very close to) the Bragg angle corresponding to the pure nuclear reflection. At the same time, in order to reduce the incoherent scattering, one should use the narrowest possible incoming energy width without losing those photons capable of resonant scattering. Both requirements have been satisfied quite well by the use of a six-reflection premonochromator assembly providing extremely high energy resolution and small angular width (Fig. 1). The first Si(1 1 1) double reflector isolates the subsequent high-resolution stage from the heat load produced by the SR beam. It extracts a band of radiation approximately 10 eV wide from the white SR spectrum. The next stage consists of two double reflectors of Si(10 6 4) set in dispersive mode. They determine the angular and energy width of the outgoing beam. The calculated divergence and energy bandwidth of the resultant 14.4-keV beam is

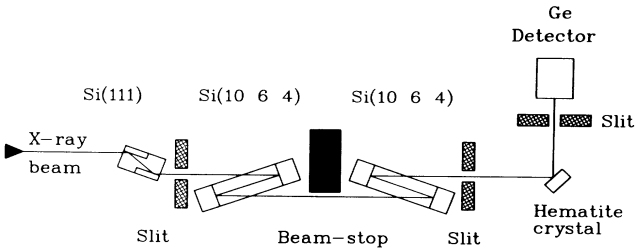

FIG. 1. Schematic view of experimental arrangement.

0.4″ and 5 meV, respectively.

This beam was then scattered by a highly perfect Fe_2O_3 (α-hematite) single crystal which was isotopically enriched with ^{57}Fe to 93%. The crystal was prepared by slow growth at high temperature from a flux of bismuth and boron oxides. It could be oriented to give scattering by the pure nuclear (777) reflection planes.[9,10] The scattered photons were detected by a high-purity Ge detector of about 400-eV resolution. A low-noise single-channel analyzer with digital adjustment was used to reduce additionally the background. The experimental station was situated 12 m from the source point. A 2×0.2-mm^2 section of the output beam from the premonochromator was incident on the Fe_2O_3, illuminating a 0.3×5-mm^2 area of the crystal. During the measurements the Brookhaven National Synchrotron Light Source x-ray ring was operated with 2.5-GeV electrons stored with currents that ranged from 50 to 200 mA. The typical photon rate incident on the Fe_2O_3 was $(3-8)\times10^4$ quanta/sec and the nonresonant background at the detector was about 0.01 quantum/sec. The number of resonant quanta received by the detector was in the range 1–2.5 sec^{-1} dependent upon the storage-ring operating conditions.

Energy calibration of the monochromator was carried out with use of a 0.2-mm-thick Si wafer cut and oriented such that the $(\bar{1}064)$ reflection could be observed in symmetrical reflection and the (111) and $(\bar{1}\bar{1}\bar{1})$ reflections observed in symmetrical transmission. This wafer replaced the Fe_2O_3 crystal and allowed the determination of the energy setting of the monochromator with a precision of 80 meV. The Bragg angle for the (777) reflection of Fe_2O_3 was accurately determined from the measurement of the electronically allowed (666) and (888) reflections.[10,11] After the initial location of the (777) peak, the precision of these calibrations reduced any subsequent searches for the pure nuclear reflection to a relatively small two-dimensional parameter space: thirty steps of 5 meV in energy and three steps of 0.9″ in θ with a counting time of 10 sec per point.

Figure 2 shows measured rocking curves of both the allowed electronic reflection, (666), and the pure nuclear (777) reflection. The calculated intrinsic width of the (666) reflection is 0.75″ and the calculated width of the energy-integrated reflectivity curve of the resonant nuclear (777) reflection is 0.4″. The observed width of 3″ in both cases shows that the crystal is not ideally perfect. X-ray topographic studies[12] of the crystal indicate, however, that the dominant source of broadening is long-range strain, and one can thus expect essentially dynamical behavior of the scattering.

It is possible that the observed intensity at the (777) position could come either from magnetic x-ray scattering, previously studied from[13] α-Fe_2O_3 or multiple Bragg scattering.[14] In order to eliminate either possibility energy scans were performed at both the (666) and (777) reflections and these are shown in Fig. 3. The width of the (666) reflection, 250 meV, is consistent with the 3″ rocking curve dominated by the crystal deformations. The (777) reflection, however, is only 5 meV wide, just the monochromator resolution function. Furthermore, measurements of the (555) pure nuclear reflection were also made. The intensity compared to the (777) reflection scaled as predicted,[12] and occurred at the same energy. The energy width and the angular range of reflection were essentially identical to those of the (777). These measurements confirm the pure nuclear origin since either magnetic x-ray scattering or multiple scattering would also reflect the crystal deformations and would have a width in energy similar to that of the (666) electronic peak.

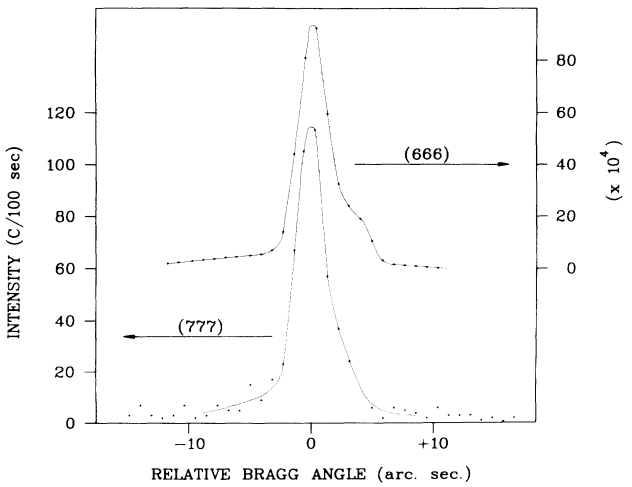

FIG. 2. The rocking curve of the electronic (666) reflection (upper curve) and the energy-integrated rocking curve of the pure nuclear (777) reflection (lower curve). The continuous lines are guides for the eye.

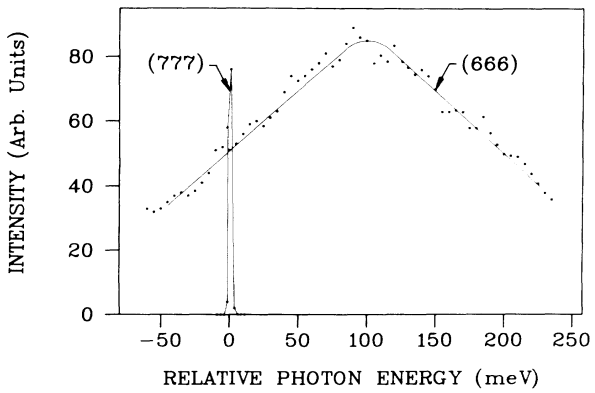

FIG. 3. Energy dependence of the scattered radiation from $^{57}Fe_2O_3$. The broad curve corresponds to the (666) reflection while the sharp curve corresponds to the (777) reflection. The continuous lines are guides for the eye.

On the basis of the intensity of the (777) peak and the energy width and intensity of the beam incident on the hematite, the expected effective linewidth was calculated.[12] The result is $(60 \pm 15)\Gamma$, where Γ is the natural width of the ^{57}Me Mössbauer transition. Applying the dynamical theory of nuclear diffraction for the (777) reflection of the hematite one calculates $\Gamma_{\text{eff}} = 50\Gamma$, which is in good agreement with the above estimate. This extremely large energy broadening reflects pronounced dynamical behavior of the crystal, as expected, and implies an effective lifetime of 3 nsec in the Fe_2O_3 crystalline environment while satisfying the Bragg condition, compared to the 138-nsec lifetime of isolated nuclei.

Furthermore, on the basis of the time-integrated signal-to-noise ratio at the (777) peak an estimate of the visibility of the first peak in the quantum-beat spectrum[4,15] can be made. Assuming that all the background intensity is prompt, we calculate 30:1 for the ratio between the integral under the first peak and the prompt background.

In summary, we have demonstrated that nuclear Bragg scattering can be observed with a time-integrated signal-to-noise ratio of 100:1 without temporal suppression of prompt scattering, thereby opening up many other nuclear resonant species for study than was possible with previous techniques. The measured parameters of the scattered beam are interpretable through the use of the dynamical theory of nuclear scattering. Straightforward improvements to the apparatus will provide greatly increased fluxes, and in the near future we will study the polarization properties and the full time dependence of the scattered radiation from hematite. We expect that this method of premonochromatization will make practical the utilization of nuclear Bragg scattering to fill the gap in energy resolution that presently occurs between existing crystal monochromators and radioactive Mössbauer sources.

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