Resonance Energy Shifts during Nuclear Bragg Diffraction of X Rays

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We have observed dramatic changes in the time distribution of synchrotron x rays resonantly scattered from ^{57}Fe nuclei in a crystal of yttrium iron garnet, which depend on the deviation angle of the incident radiation from the Bragg angle. These changes are caused by small shifts in the effective energies of the hyperfine-split nuclear resonances, an effect of dynamical diffraction for the coherently excited nuclei in the crystal. The very high brightness of the synchrotron x-ray source allows this effect to be observed in a 15-min measurement.

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When the 14.4-keV Mössbauer resonance in ^{57}Fe in a Bragg-diffracting crystal is excited by a short, broadband pulse of radiation from a synchrotron light source, the time distribution of the diffracted radiation exhibits a beat pattern due to interference between the coherently excited nuclear hyperfine levels.[1-4] Analysis of this time distribution gives a very accurate determination of the hyperfine fields in the crystal.

We have used an undulator beam line at the PEP storage ring at Stanford Synchrotron Radiation Laboratory (SSRL) to resonantly scatter x rays from ^{57}Fe in a nearly perfect crystal of yttrium iron garnet (YIG). The high brightness of this x-ray source allowed us to obtain a time distribution with excellent statistics in less than 15 min, with a data collection rate nearly 2 orders of magnitude higher than in previous experiments of this type.[2,3,5-7] Along with the beat pattern due to the hyperfine splitting of the nuclear resonance, we observed dramatic changes in the time distribution that depend sensitively on the deviation of the incidence angle of the radiation from the Bragg angle. These changes are due to small shifts in the effective resonance energies of the collection of Fe nuclei in the crystal lattice, an effect of dynamical diffraction, and are the subject of this Letter.

Analysis of the dynamical diffraction theory for a perfect lattice of simple resonant nuclei shows that the resonance energy plays a role complementary to that of the diffracting wave vector, since the crystal reflectivity depends strongly on both the angular deviation from the Bragg angle and the energy deviation from the resonance energy. For both resonant and nonresonant scattering, the reflectivity of a thick crystal in Bragg geometry is maximized when the quantity $[(\alpha-2g_0)^2-4g_K^2]^{1/2}$ is minimized,[8-10] where α is proportional to the deviation of the incident radiation from the Bragg angle, g_0 is the scattering amplitude in the forward direction, and g_K is the scattering amplitude in the reflected direction (this assumes that the crystal is centrosymmetric). The scattering amplitude for nonresonant electronic scattering is

$$g_{eK}=\frac{\lambda^2}{\pi V}F_e(K)\,, \tag{1}$$

where V is the unit-cell volume and $F_e(K)$ is the complex electronic scattering length of the unit cell, including the geometrical structure factor, electronic form factors, imaginary electronic absorption factors, and Debye-Waller temperature factors. In contrast, the energy-dependent scattering amplitude for pure resonant nuclear scattering from a single Mössbauer transition is[11]

$$\begin{gathered} g_{nK}(E)=\frac{G_{nK}}{2(E-E_0)/\Gamma+i}\,, \\ G_{nK}=-\frac{\lambda^3}{2\pi^2 V}\frac{3}{2}\frac{\Gamma_\lambda}{\Gamma}\frac{\eta}{2I_0+1}C^2P_{0K}F(K)f(k_0)f(k_1)\,, \end{gathered} \tag{2}$$

where Γ_λ is the effective energy width for radiative decay, Γ is the total width, η is the isotopic enrichment factor, $2I_0+1$ is the number of ground-state levels, C is the Clebsch-Gordan coefficient coupling ground and excited states, P_{0K} is the appropriate polarization factor coupling the incident and scattered beams, $F(K)$ is the geometrical structure factor, $f(k_0)$ and $f(k_1)$ are the Lamb-Mössbauer temperature factors for the absorption and emission processes, and E_0 is the resonance energy.

Just as the angular dependence of the crystal reflectivity at fixed energy yields a Darwin plateau with its center offset from the Bragg angle, the energy dependence of the reflectivity of a crystal of resonant scatterers at fixed angle yields a range of energy values with high reflectivity and an offset of the center of this range from E_0. For a pure nuclear reflection, with electronic scattering forbidden, the offset or shift of the resonance energy can be shown[12,13] to be equal to the real part of the complex energy shift

$$\Delta_0(\alpha)=\frac{\Gamma|G_{n0}|}{\alpha-2g_{e0}}\,, \tag{3}$$

where $\alpha=-2(\theta-\theta_B)\sin(2\theta_B)$, and g_{e0} is the electronic forward scattering amplitude. [Equation (3) applies to a reflection for which electronic scattering is forbidden, but the electronic forward scattering amplitude does not vanish, and it contributes an index of refraction effect.] For the YIG crystal discussed below, the resonance energy shift can be expressed in terms of the resonance width

Γ (4.67×10^{-9} eV) as

$$\mathrm{Re}\{\Delta_0(\delta\theta)\} = -3.18\Gamma C^2 F(0)\frac{\delta\theta}{(\delta\theta)^2+3.00}\,, \qquad (4)$$

with $\delta\theta = \theta - \theta_B + \mathrm{Re}(g_{e0})/\sin(2\theta_B)$, the angular deviation from the Bragg angle corrected for the index of refraction, expressed in μrad. The sign of the energy shift depends on the sign of $\delta\theta$, and its magnitude is greatest when the value of $\delta\theta$ is slightly greater or less than zero. Exactly at the Bragg angle, there is no shift. The maximum value of the shift is limited by the imaginary part of g_{e0}, the electronic absorption.

The range of energies over which the crystal exhibits a high reflectivity is given by twice the real part of

$$\Delta_K(\alpha) = \frac{\Gamma\,|G_{nK}|}{\alpha - 2g_{e0}}\,. \qquad (5)$$

When the crystal is near the Bragg angle, this energy range can be much broader than the resonance width of an isolated nucleus, leading to the speedup effect, an enhanced rate of decay of the crystalline excited nuclear state. Enhancement of the coherent radiative decay channel results in a smaller fraction of the excitations decaying via internal conversion.[14,15]

These properties of the energy dependence of the crystal reflectivity do not imply a change in the nuclear physics or static fields at the nuclei. Rather, they indicate that the collective response of a large number of coherently excited identical nuclei, positioned on a lattice with definite geometric phase relationships which depend upon the deviation angle, differs somewhat from the individual response of an isolated nucleus.

The line broadening and resonance shifts, though very small, have been observed in a careful Mössbauer diffraction experiment using a highly collimated beam from a conventional radioactive source.[16] These effects are much more prominent when the crystal is excited by a synchrotron pulse, as they modify the time distribution of the scattered radiation. The time-dependent reflectivity amplitude for the pure nuclear reflection described above, excited by a very short pulse at $t=0$, is[12,13]

$$R(t>0) = i\frac{G_{nK}}{|G_{nK}|}\exp\left[-i\frac{tE_0}{\hbar} - \frac{t\Gamma}{2\hbar} + i\frac{t\Delta_0}{\hbar}\right] \times \frac{J_1(t\Delta_K/\hbar)}{t}\,. \qquad (6)$$

The speedup due to the broadened energy width, expressed by the Bessel-function factor, is easily observed in a nuclear diffraction experiment using a synchrotron source.[17] The resonance energy shift has a more subtle influence on the time distribution, and its observation requires the use of interference between multiple nuclear levels. The effects of the resonance energy shift were noted in Ref. 17. However, due to the antiferromagnetic arrangement of the internal fields at the scattering nuclei in that experiment, the observed effects were very small.

If the nuclear resonance is split by fields in the crystal into several closely spaced hyperfine levels, the levels can be excited coherently by a synchrotron pulse, and the reflectivity will exhibit a beat pattern characteristic of the hyperfine energy splitting.[1,2,4] For a typical magnetic material containing ^{57}Fe, the hyperfine energy splitting is about an order of magnitude larger than the diffraction-induced energy shifts. However, under certain conditions the shifts can have a striking effect on the beat pattern.

We observed the time distribution of photons diffracted from YIG (002) planes in symmetric Bragg geometry, when illuminated by short (<1 ns) pulses of synchrotron x rays from the 1B undulator at the PEP storage ring at SSRL.[18] A double-crystal silicon (111) premonochromator was used to produce an incident beam with bandwidth of about 2 eV at 14.4 keV, and divergence of approximately ±9 μrad. At the sample, an alignment field (of about 100 G) nearly parallel to the incident beam and to the (100) crystal axis oriented the internal moments and provided a quantization axis, so that the hyperfine structure for each nuclear site consisted of two $\Delta J_z = +1$ resonances excited by one circular-polarization component of the incident light, and two $\Delta J_z = -1$ resonances excited by the opposite-polarization component.[19] The incident radiation was highly linearly polarized, providing equal amounts of left-hand-circular (LHC) and right-hand-circular (RHC) light. The observed time distribution of the scattered radiation therefore consisted of a superposition of an intensity pattern with beats from the LHC component, and an intensity pattern with beats from the RHC component. [Each of these beat patterns was further modified by a slow modulation due to electric quadrupole fields in the crystal. These fields introduce slight resonance energy shifts between alternating planes in the (002) direction, thereby allowing nuclear scattered radiation to be observed from what would otherwise be a crystallographically forbidden reflection.] The diffracted x rays were detected with time resolution of 3 ns FWHM using a plastic scintillator and photomultiplier tubes. This experimental arrangement was similar to that used in some experiments performed at Hamburger Synchrotronstrahlungslabor.[2,3,5]

The Clebsch-Gordan coefficients for the transitions to the excited states with $J_z = \pm\frac{3}{2}$ are larger than the coefficients for the transitions to the excited states with $J_z \pm \frac{1}{2}$. Within each of the diffracting crystal planes, each circular-polarization component excites one transition of each type. Since the magnitudes of the energy shifts introduced by a deviation angle are proportional to the Clebsch-Gordan coefficients, the different transitions experience different shifts. It turns out that a deviation angle which causes the effective energy values for the

LHC resonances to move closer together causes the effective values for the RHC resonances to move farther apart, and vice versa. Therefore, a change in the deviation angle causes the periods of the two overlapping beat patterns to be changed relative to each other.

The time distribution of the diffracted radiation changes dramatically when the superimposed beat patterns shift against each other in response to a change in deviation angle. Of particular interest is a comparison of the time distributions observed at corresponding deviation on either side of the Bragg angle. The electric quadrupole interaction in YIG introduces a small difference between the LHC and RHC beat periods which is independent of deviation angle. The angle-dependent shifts add to or subtract from this difference, producing different time distributions for opposite deviation angles. As van Bürck *et al.* have pointed out,[17] an asymmetry of this type gives immediately the sign of the electric quadrupole interaction.

Figure 1(a) shows the time distribution obtained when the deviation angle $\delta\theta$ was small. The most prominent features of this distribution are the large speedup giving very little intensity after about 140 ns, and the fast-beat structure due to the interference of the hyperfine-split resonances. A large peak at $t=0$, due to nonresonant electronic scattering, is not shown. No background or other corrections have been made to these data.

Figures 1(b) and 1(c) show the time distributions obtained when the YIG crystal was slightly rotated, introducing the indicated deviation angles. With such deviation angles, the overall intensity is reduced, the speedup effect is smaller, and a slow-beat pattern with period of about 130 ns, due to the electric quadrupole interaction, is clearly visible. Moreover, there are striking differences in the fast-beat patterns. The energy shifts in the $+31$-μrad case are such that the LHC and RHC beat patterns have peaks and valleys that nearly coincide during the time period 10–100 ns, whereas in the -33-μrad case the peaks of the LHC beat pattern tend to fall upon the valleys of the RHC pattern during this time period.

The solid curves were calculated by numerically Fourier transforming the energy-dependent reflectivity amplitude. Whereas the reflectivity amplitude for a simple resonance can be transformed analytically, giving the time response shown in Eq. (6), the multiple hyperfine-split resonances of the YIG case require numerical transformation. Since the diffractometer angle setting accuracy was about ± 5 μrad, the values for $\delta\theta$ shown in Fig. 1 were determined by fitting the calculations to the data. The statistical error associated with the fitting procedure was ± 1 μrad. The calculations were averaged over a Gaussian profile with FWHM of 18 μrad to account for incident-beam divergence and crystal imperfections. No correction for time resolution was needed. It was found that the hyperfine magnetic field in our sample was 370 ± 4 kG, 7.5% smaller than the published value of 400 kG.[19] This was confirmed in an independent

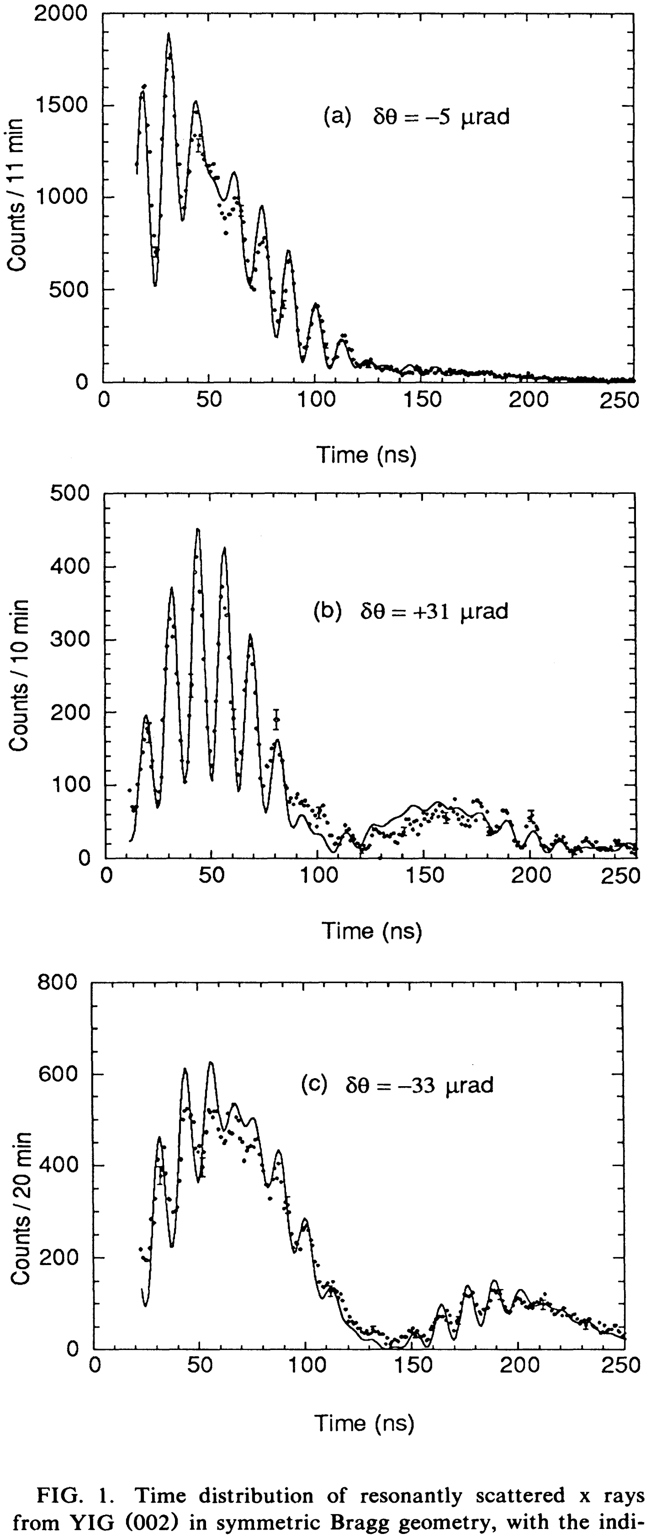

FIG. 1. Time distribution of resonantly scattered x rays from YIG (002) in symmetric Bragg geometry, with the indicated deviation angles from the Bragg angle (corrected for refraction due to the electron density). Each solid curve gives the intensity of the Fourier transform of the dynamical theory calculation for the multilevel energy-dependent reflectivity amplitude.

Mössbauer absorption experiment. The reduced field is believed to be due to a substantial amount (4%) of Pb impurity introduced in order to minimize strains in the ^{57}Fe-enriched YIG crystal grown epitaxially on a gadolinium gallium garnet substrate.[20]

The fast-beat patterns of the two polarization states have periods of about 12.3 and 12.8 ns in Fig. 1(b), whereas in Fig. 1(c) the periods are about 12.0 and 13.1 ns. The small changes in the periods, due to the small shifts in effective resonance energies by about $\pm 0.5\Gamma$ or $\pm 2\times 10^{-9}$ eV, have a clearly visible effect on the time distribution.

Previous observations of diffraction-induced resonance energy shifts by van Bürck *et al.*,[16] using a strong radioactive Mössbauer source, required several weeks of continuous counting. In the present experiment, the time required to accumulate the data for one of the beat patterns was typically less than 15 min. Peak counting rates of nuclear-scattered x rays were about 500 Hz. This high data rate was made possible by the PEP undulator source, which produces an extremely bright x-ray beam at 14.4 keV with an angular divergence only slightly larger than the ± 9-μrad acceptance of the premonochromator crystals.

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