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Resonant Diffraction of Synchrotron Radiation by a Nuclear Multilayer

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ABSTRACT

We report the observation of nuclear resonant diffraction of synchrotron radiation by a synthetic multilayer. The nuclear period of the [^{57}Fe(22Å)/Sc(11Å)/Fe(22Å)/Sc(11Å)] x 25 multilayer was chosen to be twice the electronic period to obtain a pure nuclear Bragg reflection. Strong enhancement of the radiative scattering channel provided a fast time response for the ^{57}Fe resonant scattering at the Bragg peak, with a decay time of 4 ns. The nuclear multilayer will be useful as a narrow bandpass monochromator for synchrotron radiation.

Much of the progress in studying nuclear resonant diffraction of x-radiation has been accomplished with perfect single crystals having a naturally occurring structure [1, 2]. Quite recently, artificial layered structures fabricated of Mössbauer isotopes have begun to be studied [3-6]. Synthetic layered structures are attractive tools for studying nuclear diffraction because their structure and composition may be varied within broad limits, and tailored to the experimental goals. They permit control of important parameters of the coherent x-radiation fields, including spatial arrangement, energy spectrum, time evolution, and polarization.

In particular, nuclear layered structures have an important application for the generation of highly monochromatic beams of x-radiation. X-ray monochromators utilizing Bragg reflections from silicon crystals can (by using high-order reflections) produce beams with energy bandwidths of a few meV [7]. Much narrower bandwidths (<1 μeV) can be achieved using resonant nuclear scattering from single crystals, but the magnetic crystal structure required to give a pure nuclear reflection introduces hyperfine splitting of the nuclear resonance, and gives a scattered beam with a complicated energy structure [2]. Artificial layered structures can be prepared with differing nuclear and electronic structures [8, 9], giving rise to pure nuclear diffraction of synchrotron radiation without magnetic hyperfine splitting. The very small scattering angles typical of reflections from

artificial layered structures result in wide nuclear rocking curves. This makes it possible to achieve precise Bragg conditions throughout the divergence of a synchrotron radiation beam, and to achieve thereby a large enhancement of the radiative scattering channel [10], and thus a fast time response (strong speedup [11]) and high reflectivity over an energy band that is large compared to the standard nuclear resonance width, but much smaller than the bandpass of a crystal monochromator.

Two types of artificial layered structures have been proposed as nuclear monochromators: nuclear multilayers and grazing incidence antireflection (GIAR) films. GIAR films employ specular reflection of x-rays by nuclei. They are bilayer structures incorporating resonant nuclei in one of the layers [5, 6, 9]. The top layer thickness and the layer materials are chosen to suppress electronic scattering, but the effect of the nuclear interaction gives strong scattering over the nuclear resonance energy range. The so-called damping-stabilized GIAR films operate at very small angles of incidence (typically 2-3 mrad), which gives a very strong radiative enhancement effect and hence good nuclear reflectivity over an energy range of up to 0.5 μeV. Off-resonance electronic reflectivity values for these films are typically 0.03-0.05.

Nuclear multilayers are diffracting optics involving alternating layers of differing isotopes, creating a nuclear periodicity different from the electronic diffracting periodicity. The layer thickness is chosen to give a pure nuclear Bragg reflection at an angle well above the region of high specular reflection. The radiative enhancement factor is somewhat smaller than for GIAR films, giving high reflectivity over an energy range of about 0.2 μeV, but the off-resonance electronic reflectivity is much lower (10^{-3} for the device discussed below). The goal of the present work was to observe and study resonant nuclear diffraction under the conditions of extreme radiative enhancement provided by a nuclear multilayer, and to explore the use of the nuclear multilayer as a monochromator for use in nuclear resonant scattering experiments.

The [^{57}Fe(22Å)/Sc(11Å)/Fe(22Å)/Sc(11Å)] x 25 multilayer used in these experiments was prepared at the Nizhnii Novgorod Applied

6.0×10^{-3} Torr of argon. Fifty periods of Fe(21.8Å) and Sc(10.9Å) were deposited onto an extremely smooth glass substrate (6 cm by 3 cm). Alternate iron layers were deposited using an enriched (95%) target of ^{57}Fe, with special care taken to insure that the enriched and natural iron layers were the same thickness. The multilayer unit cell for nuclear resonant scattering is thus twice as large as that for electronic scattering, providing a nuclear allowed, but electronically forbidden Bragg reflection.

This multilayer was investigated previously [4] using a conventional $^{57}Co(Cr)$ radioactive source at the Moscow Kurchatov Institute. The conversion electron spectrum of the multilayer was approximately an unresolved doublet with a total width of about 0.8 mm/sec (including a source width of 0.2 mm/sec). This important simplification of the hyperfine spectrum of ^{57}Fe is a result of partial diffusion of scandium atoms into the iron layers [12]. A pure nuclear reflection peak at a Bragg angle of 7 mrad was observed with incident radiation in resonance with the ^{57}Fe nuclear levels. A Mössbauer diffraction spectrum of the pure nuclear reflection showed coherent broadening of the resonance to a width of about 40 Γ_0 (where Γ_0 is the natural line width of ^{57}Fe, about 4.7×10^{-9} eV or 0.097 mm/sec), with an average reflectivity of about 15% over this range. The reflectivity was about 10^{-3} when the source energy was Doppler shifted far from the resonance energy of the multilayer.

The work described here was performed at the Cornell High Energy Synchrotron Source (CHESS) at the 24 pole wiggler beamline F2. Beamline optics included a double-crystal Si(111) premonochromator and a 4-bounce precision monochromator using nested Si(422) and Si(10 6 4) channel-cut crystals [13], which produced a source beam with a 12 meV bandwidth at the nuclear resonance energy, 14.413 keV. The photons reflected by the multilayer were detected by an avalanche photodiode (APD) detector [14] with timing electronics synchronized to the storage ring.

Electronic and nuclear rocking curves for the multilayer were measured simultaneously in the vicinity of the pure nuclear reflection (Fig. 1). The nuclear curve was determined by integrating counts over the time interval between 3 and 200 ns after the synchrotron pulse.

The nuclear rocking curve width of about 700 μrad greatly exceeds both the beam divergence (20 μrad) and the peak position uncertainty due to inhomogeneity in the layer thicknesses and substrate curvature (70 μrad). The prompt electronic scattering shows no Bragg peak in the vicinity of the nuclear peak, only a low reflectivity level of about 10^{-3} modified by a Kiessig beat pattern [15] due to interference between waves scattered by the top and bottom boundaries of the multilayer.

Figure 2 shows the time evolution of the radiation reflected from the multilayer at the nuclear Bragg position. There is clear evidence of speedup of the nuclear decay due to enhancement of the radiative nuclear transition probability (an effect of the coherent excitation of a nuclear lattice [16-18]); the lifetime during the interval 2-13 ns (when about 85% of the scattered radiation was detected) is 4.0 ns. For an isolated ^{57}Fe atom in an excited nuclear state, electron internal conversion is the dominant decay mode, with a lifetime of 158 ns. The radiative decay lifetime is 1300 ns, and the combined effects of both decay modes give a natural lifetime of $\tau_0=1/\Gamma_0=141$ ns. For the nuclear multilayer on Bragg, radiative decay has been so enhanced as to become very much the dominant mode. The ratio of the radiative decay rate for the multilayer to the radiative rate for an isolated nucleus is about 325.

The observed slow beats of the time evolution are related to multiple scattering of radiation within the nuclear multilayer. Experimental data were fitted using the dynamical theory of nuclear resonant scattering [11, 19]. This represents the most extreme case of Bragg reflection radiative enhancement to which the dynamical resonant scattering theory has been applied.

The spectral properties of the radiation diffracted by the nuclear multilayer were investigated using forward scattering [20] through a 12 μm pure ^{57}Fe foil. The foil was magnetized to orient the nuclear hyperfine magnetic field perpendicular to the photon propagation direction and parallel to its polarization. Under these conditions only hyperfine nuclear transitions with $\Delta m=\pm 1$ (Δm is the change in magnetic quantum number between the ground and excited substates) were excited (lines 1,3,4, and 6 of the Mössbauer spectrum). The time distribution of the radiation scattered forward through the foil was measured both using relatively broadband source radiation supplied by

the silicon monochromator (Fig. 3a), and source radiation filtered by the nuclear multilayer (Fig. 3b).

The time evolution of the forward nuclear scattering excited by broadband radiation (Fig. 3a) exhibits quantum beats [21, 22] originating from interference between the four coherently-excited nuclear transitions (the lines 1, 3, 4, 6). The dominant beat period from the strong outer transitions $2\pi\hbar/(E_6 - E_1)$, equal to 8 ns [18], is seen well. The same beat period is observed with the iron foil being excited by the nuclear multilayer radiation (Fig. 3b). This shows that all four nuclear transitions, including those separated by the energy distance 110 Γ_0 (1 and 6), are excited to some extent by the nuclear multilayer radiation.

However, the effective bandpass (FWHM) of the multilayer is smaller than the separation between lines 1 and 6. This can be concluded from the shift of about 4 ns in the structure of the beat pattern, visible in Fig. 3. This shift is related to the phase difference of the scattered radiation components with energies E_6 and E_1, since these energies lie on opposite sides of the nuclear multilayer energy distribution [5]. A value for the multilayer energy bandpass derived from fitting dynamical diffraction calculations to the time distribution data shown in Fig. 2 is 41 Γ_0, about 0.2 μeV. Lines 1 and 6 in the transmission sample are excited by the tails of the multilayer radiation distribution. The calculated reflectivity of the multilayer for lines 1 and 6 is about 10% of the peak nuclear reflectivity.

This experiment should be regarded as a first step both in the study of resonant nuclear scattering from artificial layered structures, and in the search for an effective μeV monochromator. It demonstrates that the dynamical theory of resonant nuclear scattering can be applied in cases of extreme speedup and to multicomponent scattering systems, and it demonstrates that a nuclear multilayer can be used to produce a ~μeV bandwidth x-ray beam with good out-of-band rejection. It is expected that further increases in the effective bandpass and decreases in the electronic, nonresonant scattering rate can come from the use of an antiferromagnetic multilayer [23] with an antireflection coating on top of the multilayer [24].

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REFERENCES

[1]G. V. Smirnov, Hyperfine Interactions **27**, 203 (1986).

[2]R. Rüffer, Synch. Rad. News **5**, 25 (1992).

[3]R. Röhlsberger, E. Witthoff, E. Lüken, and E. Gerdau, in *Physics of X-Ray Multilayer Structures* , Jackson Hole, 1992, Optical Society of America Technical Digest Vol. 7, p. 178.

[4]A. I. Chumakov, G. V. Smirnov, S. S. Andreev, N. N. Salashchenko, and S. I. Shinkarev, Pis'ma Zh. Eksp. Teor. Fiz. **55**, 495 (1992) [JETP Lett. **55**, 509 (1992)].

[5]R. Röhlsberger, E. Gerdau, M. Harsdorff, O. Leupold, E. Lüken, J. Metge, R. Rüffer, H. D. Rüter, W. Sturhahn, and E. Witthoff, Europhys. Lett. **18**, 561 (1992).

[6]E. E. Alp, T. M. Mooney, T. Toellner, W. Sturhahn, E. Witthoff, R. Röhlsberger, E. Gerdau, H. Homma, and M. Kentjana, Phys. Rev. Lett. **70**, 3351 (1993).

[7]D. P. Siddons, J. B. Hastings, and G. Faigel, Nucl. Instrum. Meth. **A266**, 329 (1988).

[8]G. T. Trammell, J. P. Hannon, S. L. Ruby, P. Flinn, R. L. Mössbauer, and F. Parak, in *Workshop on New Directions in Mössbauer Spectroscopy*, Argonne, 1977, edited by G. J. Perlow, AIP Conf. Proc. No. 38, p. 46.

[9]J. P. Hannon, G. T. Trammell, M. Mueller, E. Gerdau, R. Rüffer, and H. Winkler, Phys. Rev. **B32**, 6374 (1985).

[10]A. M. Afanas'ev and Y. Kagan, Zh. Eksp. Teor. Fiz. **48**, 327 (1965) [Sov. Phys. JETP **21**, 215].

[11]Y. Kagan, A. M. Afanas'ev, and V. G. Kohn, J. Phys. C **12**, 615 (1979).

[12]D. H. Ryan, J. O. Ström-Olson, and W. B. Muir, Phys. Rev. B **40**, 11208 (1989).

[13]T. S. Toellner, T. M. Mooney, S. Shastri, and E. E. Alp, in *Optics for*

High-Brightness Synchrotron Radiation Beamlines, edited by J. Arthur, Proc. SPIE 1740, 218 (1993).

[14]S. Kishimoto, Nucl. Instrum. Methods **A309**, 603 (1991); A. Q. R. Baron and S. L. Ruby, to be published.

[15]H. Kiessig, Ann. Phys. **10**, 769 (1931).

[16]G. T. Trammell, in *Chemical Effects on Nuclear Transformations* (International Atomic Energy Agency, Vienna, 1961), p. 75.

[17]Y. Kagan, A. M. Afanas'ev, and I. P. Perstnev, Zh. Eksp. Teor. Fiz. **54**, 1530 (1968) [Sov. Phys. JETP **27**, 819].

[18]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).

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

[20]J. B. Hastings, D. P. Siddons, U. van Bürck, R. Hollatz, and U. Bergmann, Phys. Rev. Lett. **66**, 770 (1991).

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

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

[23]N. Hosoito, S. Araki, K. Mibu, and T. Shinjo, J. Phys. Soc. Japan **59**, 1925 (1990).

[24]J. P. Hannon, G. T. Trammell, M. Mueller, E. Gerdau, H. Winkler, and R. Rüffer, Phys. Rev. Lett. **43**, 636 (1979).

FIGURE CAPTIONS

Fig. 1. Electronic (a) and nuclear (b) rocking curves near the pure nuclear reflection. The nuclear curve was determined by integrating between 3 and 200 ns after the synchrotron pulse, while the electronic curve was integrated over all time but is dominated by the prompt scattering. Solid line in (b) is a theory fit. Solid line in (a) is to guide the eye. The average prompt intensity shown in (a) corresponds to an electronic reflectivity of about 10^{-3}.

Fig. 2. Time response of the multilayer at the nuclear Bragg position. Solid line is a dynamical diffraction theory fit. Dashed line indicates the initial decay with a lifetime of 4 ns.

Fig. 3. Quantum beat pattern from forward scattering through a 12 μm ^{57}Fe foil, observed using relatively broadband radiation supplied by the silicon monochromator (a) and with radiation that was additionally monochromatized by the nuclear multilayer (b). Solid lines are dynamical diffraction theory fits. The shift in peak positions is due to the phase difference acquired by radiation at the energies of the ^{57}Fe hyperfine lines upon scattering from the multilayer.

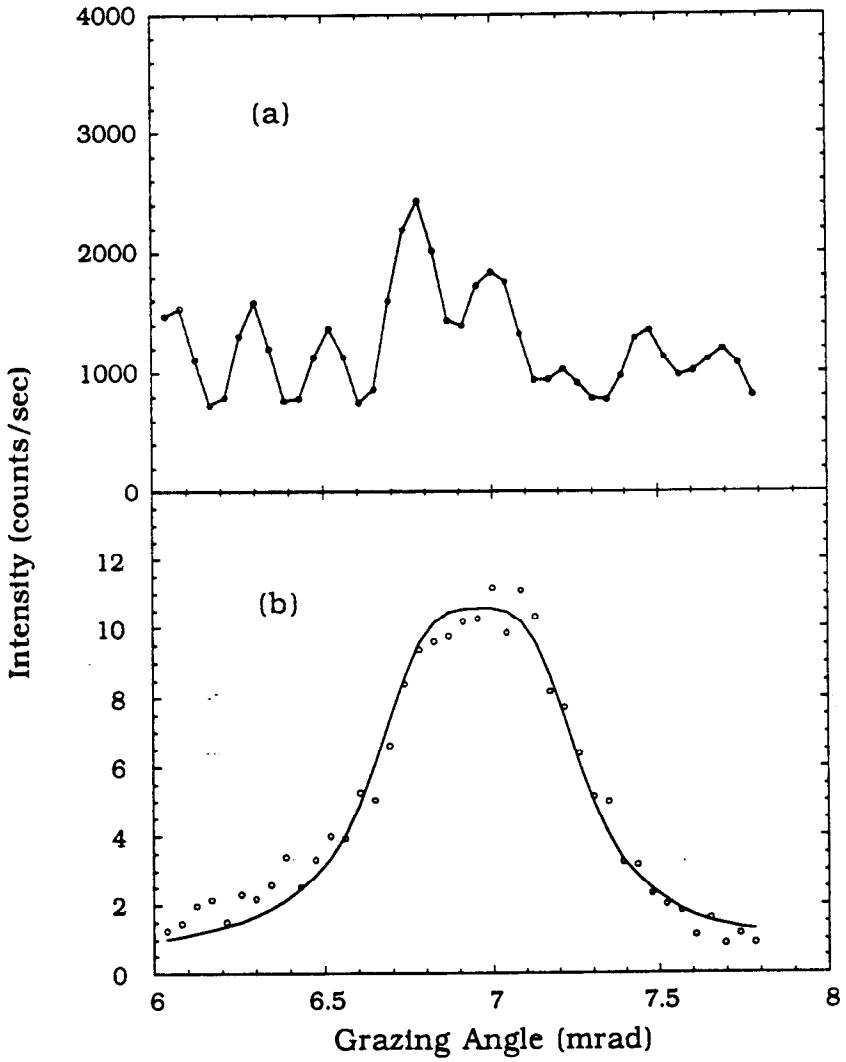
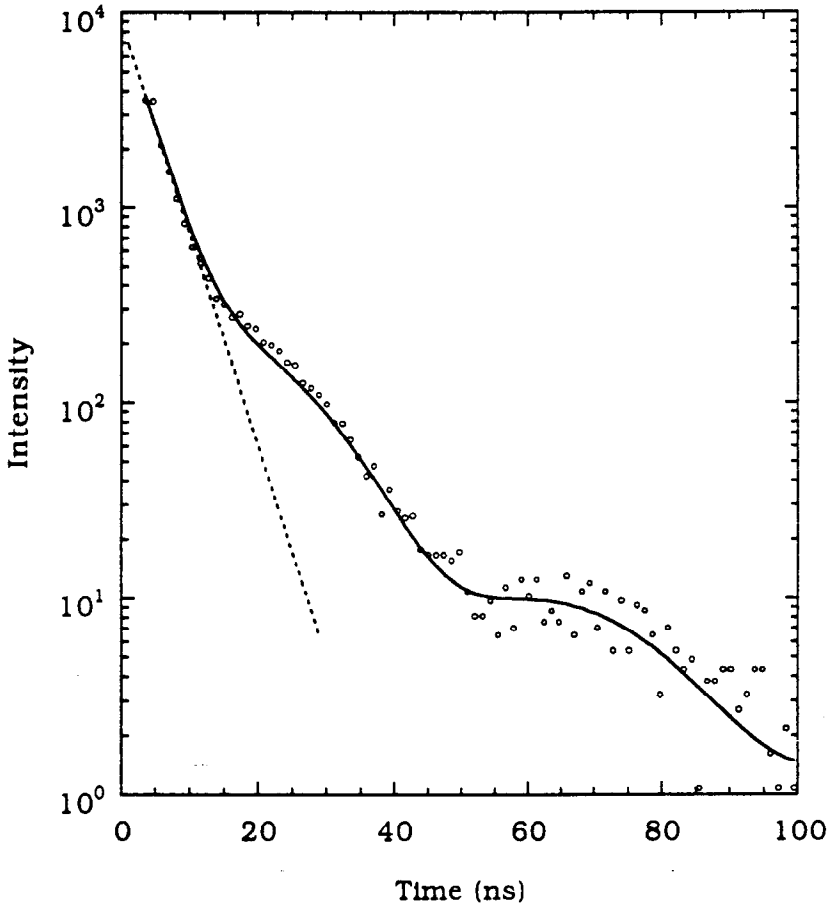


Fig. 1. Electronic (a) and nuclear (b) rocking curves near the pure nuclear reflection. The nuclear curve was determined by integrating between 3 and 200 ns after the synchrotron pulse, while the electronic curve was integrated over all time but is dominated by the prompt scattering. Solid line in (b) is a theory fit. Solid line in (a) is to guide the eye. The average prompt intensity shown in (a) corresponds to an electronic reflectivity of about 10^{-3}.



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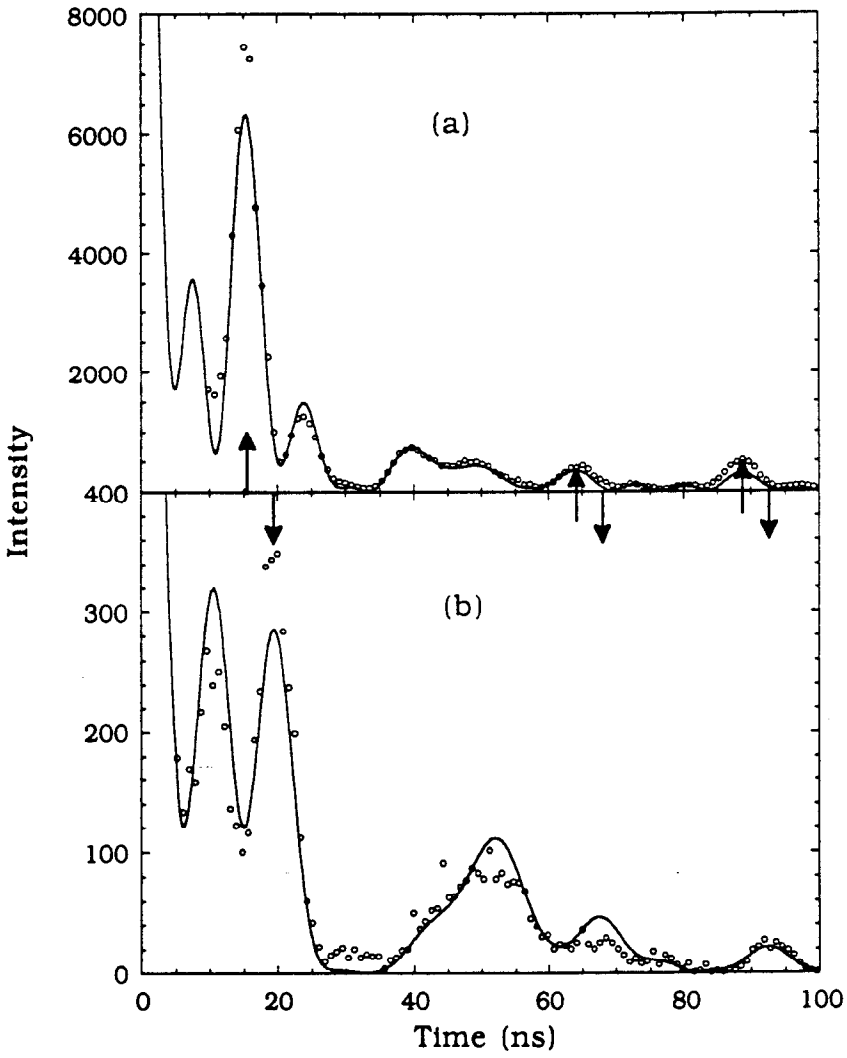

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