Time Resolved Nuclear Resonant Scattering from ^{119}Sn Nuclei Using Synchrotron Radiation

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Nuclear resonant scattering of synchrotron radiation from ^{119}Sn nuclei at 23.87 keV has been observed. The combination of a novel nested crystal monochromator and a $^{119}SnO_2$/Pd based grazing incidence antireflection film was used to investigate the Mössbauer parameters of a tin containing powder sample in a forward scattering experiment.

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The x-ray resonant scattering from nuclei bound in a solid presents some unique aspects that are not common in scattering from atoms or individual nuclei. The broadening of energy response for Bragg diffraction and for forward scattering, or the corresponding "speedup" in time domain experiments are manifestations of coherent excitation of nuclei. Also, for magnetically split samples, observation of quantum beats provides a new way of measuring hyperfine interactions. Recently there are attempts to build experimental stations dedicated to nuclear resonant scattering in various synchrotron radiation facilities around the world. Here, we present a new way to monochromatize synchrotron radiation for nuclear resonant scattering studies. The coherent nuclear resonant scattering of synchrotron radiation from Mössbauer nuclei provides a unique source of highly collimated and very monochromatic hard x rays with energy resolution $\Delta E/E$ of 10^{-10}-10^{-13}. The brilliance of this radiation is a few orders of magnitude higher than that of radioactive sources. In contrast to forward scattering experiments, angle-resolved diffraction experiments with conventional radioactive sources are difficult and time consuming due to the limited brilliance. The synchrotron radiation based beams combine a high degree of polarization, a well defined time structure, and the high brilliance with the energy resolution of Mössbauer transitions [1].

Thus far, the technique has been successfully applied to ^{57}Fe [2–5] and ^{169}Tm [6] nuclei. Here, we present experimental results of the observation of nuclear resonant scattered photons from ^{119}Sn nuclei at 23.87 keV. This experiment extends the method to a Mössbauer transition in the medium energy range above 20 keV. In our setup a high-resolution, large-angular-acceptance nested Si crystal monochromator was followed by a $^{119}SnO_2$/Pd based grazing incidence antireflection (GIAR) film. With the beam from this arrangement, we performed a forward scattering experiment to measure the hyperfine interaction parameters of a ^{119}Sn-containing sample in the time domain. It should be noted that ^{119}Sn, unlike ^{57}Fe, does not have a suitable parent to do timing experiments. The 11/2 spin state has a lifetime of 245 days, and hence does not lend itself to timing experiments.

The experiments were performed at the F2 24-pole wiggler beam line of Cornell High Energy Synchrotron Source, CHESS. The critical energy of the wiggler source is 24 keV at a positron energy of 5.5 GeV. The experimental setup, shown in Fig. 1, consists of three consecutive monochromators followed by a time-resolving coincidence detector.

The first monochromator is a water cooled Si(111) double crystal monochromator specially designed to handle the high heat load of the wiggler radiation [7]. The second monochromator consists of two channel-cut crystals placed in a nested geometry. The first face of an asymmetrically cut Si(333) crystal (asymmetry factor $b=-7.3$) accepts 5.7 μrad divergence of the incoming radiation and reduces it to 0.8 μrad. This is followed by a symmetrical Si(555) channel-cut crystal, reducing the energy bandpass to 50 meV. Finally the x-ray beam impinges on the second face of the Si(333) channel-cut crystal with reciprocal asymmetry factor. The principles of this monochromator were described by Ishikawa *et al.* [8] and a working model has been constructed [9]. For the angular adjustment of the crystals we used rotation stages driven by piezoelectric inchworms (Burleigh Instruments, RS-75) with an angular resolution of 0.1 μrad. Rotation angles were independently measured by incremental angle encoders (Heidenhain, ROD 800 with an angular resolution of 0.17 μrad). The third monochromator is a $^{119}SnO_2$/Pd GIAR film. It should be noted that the excited state of a ^{119}Sn nucleus lies 23.87 keV above the ground state. The natural mean lifetime is 25.6 nsec, giving an energy width of 25.7 neV. Compared to an incident beam bandpass of 50 meV, this would represent a signal-to-noise ratio of 5×10^{-7} in a forward scattering experiment (if the absorber has one natural

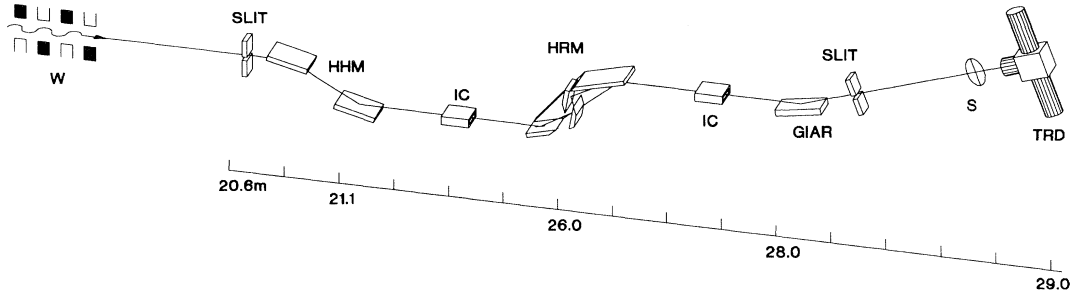

FIG. 1. The experimental setup: *W*, 24-pole wiggler; HHM, high heat load Si(111) double crystal monochromator; IC, ionization chamber; HRM, high energy resolution nested monochromator; GIAR, SnO_2/Pd grazing incidence antireflection film; *S*, the SnO-containing sample; and TRD, time-resolved coincidence detector.

linewidth). In order to alleviate overloading of the detector, additional optics are necessary to improve the signal-to-noise ratio. It was proposed that a GIAR film containing resonant nuclei may reduce the electronic reflectivity by a few orders of magnitude, while maintaining substantial nuclear reflectivity [10–12]. The design principles and characterization of such films have been described for ^{57}Fe [13,14] and ^{119}Sn [15,16].

The GIAR film in this experiment consisted of a superpolished Zerodur substrate with a surface area of 30 $\times$150 mm^2, coated with 13 nm Pd and 43.2 nm $^{119}SnO_2$. The surface microroughness of the substrate was 0.3 nm and the long range flatness was better than 50 nm. The layers have been produced in a sputter deposition process, in which the $^{119}SnO_2$ coating was deposited by reactive sputtering from a metallic ^{119}Sn target. The rocking curve of the electronic reflectivity is shown in Fig. 2. The expected minimum is at 2.09 mrad with the electronic reflectivity reduced to 2.2%. For the theoretical simulation of the rocking curve (solid line), a microroughness of 0.3 nm had to be assumed for the SnO_2/Pd boundary and the SnO_2 top surface. The calculated nuclear resonant reflectivity of the GIAR film set at 2.09 mrad, the angle of minimum electronic reflectivity, is shown as an inset in Fig. 3. Here the nuclear resonant reflectivity is plotted as a function of the energy deviation from resonance energy in units of the natural linewidth. The energy bandpass (FWHM) is 3.2 μeV, which corresponds to 123 Γ. This relatively large bandpass results from a "speedup" of nuclear decay under the grazing incidence condition that increases the number of nuclei participating in the coherent decay process [11]. The energy broadening makes it possible to use this medium as a bandpass filter or monochromator to carry out spectroscopy on samples of interest. Figure 3 shows the measured time response of the GIAR film. As expected from the bandpass of 123 Γ, the decay is strongly sped up in comparison to the mean lifetime of the nuclear level. The delayed quanta are nearly completely hidden under the prompt peak (due to nonresonant scattering), and only a small part is visible in the time interval between 5 and 10 nsec.

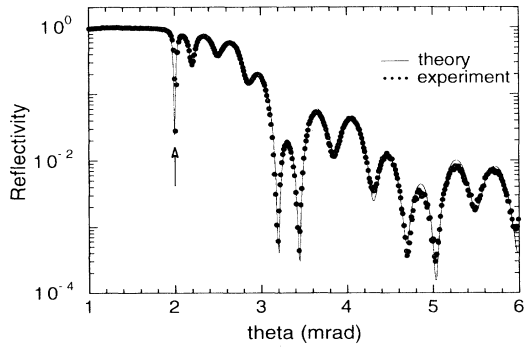

FIG. 2. The electronic reflectivity of $^{119}SnO_2$/Pd GIAR film measured at 23.87 keV. The film was operated at 2.09 mrad as indicated to suppress nonresonant radiation. The solid line is a theoretical fit. The low angle position was chosen to keep the nuclear reflectivity and the energy bandpass high.

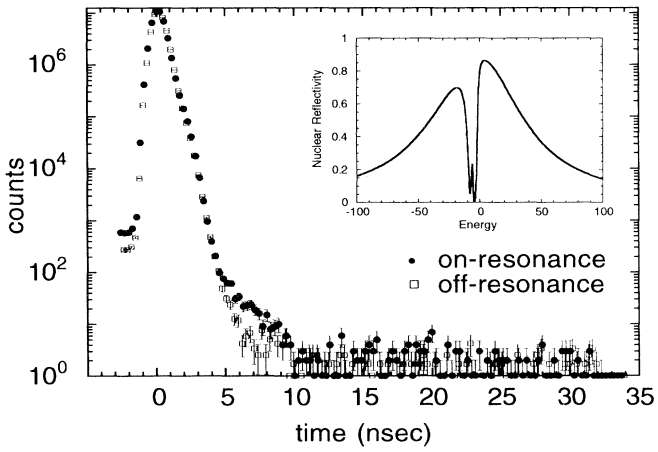

FIG. 3. The time response of the $^{119}SnO_2$/Pd GIAR film, measured using the high energy resolution crystal monochromator. Solid dots are on-resonance and empty squares are off-resonance spectra. The inset shows the calculated nuclear reflectivity in the energy domain. The integrated reflectivity is 123 Γ, leading to an energy bandpass of 3.2 μeV.

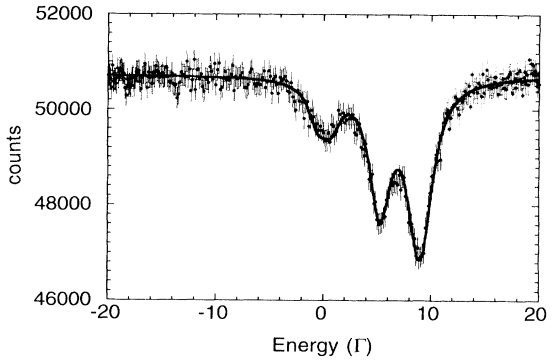

FIG. 4. Conventional ^{119}Sn Mössbauer spectrum of the sample measured in this study.

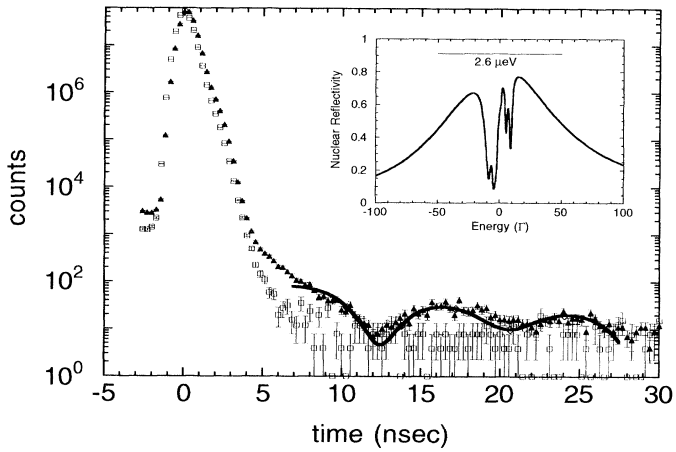

FIG. 5. The time-resolved nuclear resonant spectra of the SnO-containing sample, measured using a high energy resolution crystal monochromator and a SnO_2/Pd GIAR nuclear monochromator. Solid triangles are on-resonant and empty squares are off-resonance spectra recorded over a 2 h period. The solid line is a theoretical fit, providing a new way of performing Sn Mössbauer spectroscopy to measure hyperfine interaction parameters with synchrotron radiation. The inset is the combined response of the GIAR film and the absorber in the energy domain.

The photons were detected using a time-resolving coincidence detector [17]. Two Hamamatsu H3241 photomultipliers are optically connected to a Bicron 420 plastic scintillator. The detector fully recovered after 3.2 nsec from the prompt pulse even when the incident intensity exceeded 10^5 Hz indicating the suitability of this detector for this energy range.

In order to demonstrate that spectroscopy is indeed possible, we performed a forward scattering experiment [18] using the beam emanating from the SnO_2/Pd GIAR film. An absorber was prepared by chemical reduction of SnO_2 which was enriched to 90% in ^{119}Sn. It contained mostly divalent Sn in the form of SnO, which has an isomer shift with respect to $BaSnO_3$ of $\delta=7.2$ Γ (1 Γ=0.32 mm/sec), and an electric quadrupole splitting of $\Delta=3.8$ Γ. The transmission Mössbauer spectrum, shown in Fig. 4, reveals small contributions from unreduced SnO_2 with $\delta=0.02$ Γ and $\Delta=1.25$ Γ, and metallic Sn with $\delta=7.89$ Γ and $\Delta=0.25$ Γ. Furthermore, a conversion electron Mössbauer spectrum (CEMS) of the GIAR film was measured which revealed hyperfine parameters in the $^{119}SnO_2$ layer to be $\delta=0$ and $\Delta=3.2$ Γ. These contributions are properly included in the calculations. On the basis of these data, the GIAR film reflectivity and the transmission through the absorber have been calculated as a function of energy. The product of both functions is the energy response of the combined setup; it is shown in the inset of Fig. 5. The time spectrum of the delayed quanta observed in the experiment is shown in Fig. 5. In the time window of 3.2–30 nsec, a quantum beat pattern is clearly visible. The background was less than 0.2 Hz, and the signal in this time window was 2.1 Hz when the prompt count rate was about 60 kHz.

In order to provide further evidence that the delayed quanta are resonant photons, we performed two separate measurements: First, the Si(555) channel-cut crystal was tuned 50 meV away from the resonance and a time spectrum was recorded. This is also shown in Fig. 5. The time beats can no longer be seen. Second, the rocking curve width of the Si(555) against the Si(333) was measured with both the prompt and the delayed photons. This result is shown in Fig. 6. The rocking curve measured with prompt photons reflects the dispersive broadening of both reflections in this setup, whereas the one with the delayed photons (with a width of about 0.7 μrad) represents the intrinsic width of Si(555) at 23.87 keV.

To evaluate the time spectrum in Fig. 5, we used the CONUSS program package [19] for the calculation of the absorber spectrum and we followed the theory given by Hannon *et al.* [10,11] for calculation of the GIAR film response. Due to limited statistical accuracy of the data, we restricted the number of variable parameters to isomer shift and the Mössbauer-Lamb factor. The quantum

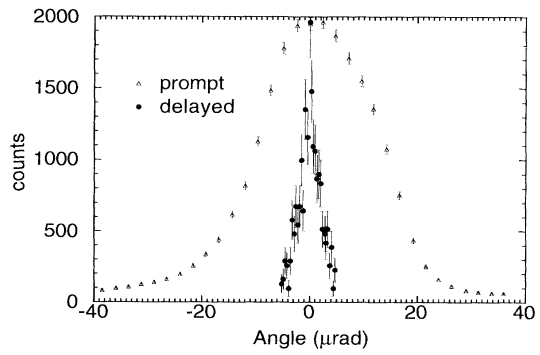

FIG. 6. Rocking curve of the Si(555) crystal measured with delayed and prompt photons at 23.87 keV.

beat period is essentially determined by the isomer shift between the ^{119}Sn in the GIAR film and the absorber. From the evaluation of the time spectrum we obtain a relative isomer shift of $-3.1(6)$ Γ. Furthermore, the Mössbauer-Lamb factor of the Sn in the GIAR film is determined to be 0.42(1).

In conclusion, we have observed time-resolved nuclear resonant scattering from ^{119}Sn nuclei. The performance of novel nested crystal monochromators combining symmetric and asymmetric reflections to achieve high energy resolution with minimum sacrifice of the angular acceptance has been demonstrated. A nuclear forward scattering experiment with a powder sample was performed by introducing a GIAR film nuclear monochromator. The nuclear reflectivity of the GIAR film itself is rather simple, and the following forward scattering experiments benefit from this simple structure. The strength of this method is that the response of the nuclei in the GIAR monochromator is well understood and calculable, and hence enables rather accurate measurement of the hyperfine parameters and recoil free fraction of samples.

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