Brief Reports

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Nuclear resonant scattering of synchrotron radiation by gaseous krypton

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We have observed scattering of synchrotron radiation by the 9.4-keV nuclear resonance of gaseous ^{83}Kr. In a gas, there is no recoilless absorption (no Mössbauer effect). A cell of krypton gas (natural abundance, 12% ^{83}Kr) was placed after a Si(111) monochromator. Incoherent resonant nuclear scattering, 0.3 cts/s, was separated from nonresonant electronic scattering, 7×10^7 cts/s, using the time resolution of an avalanche diode detector. The resonance energy was found to be 9403.5±1.8 eV. The time dependence of the scattered radiation agrees with a previous measurement of the excited-state lifetime.

Mössbauer's discovery[1] in 1958 of recoilless emission and absorption of nuclear radiation paved the way for many fascinating physics experiments[2] and opened the field of Mössbauer spectroscopy, now used routinely as a probe of atomic environments.[3] In the last decade, since work by Gerdau *et al.*,[4] the field of nuclear resonant scattering has expanded to make use of synchrotron radiation, which is broadband, highly collimated, polarized, and pulsed (<1-ns pulse width). The first synchrotron radiation experiments were diffraction studies of pure nuclear reflections in perfect crystals.[5] Since then, techniques have improved[6,7] making it possible to study coherent forward scattering from polycrystalline foils,[8] specular reflection from thin films,[9] small-angle diffraction from multilayers,[10] and incoherent scattering from a thin foil.[11]

The broadband, pulsed nature of synchrotron radiation allows investigation of samples that are difficult or impossible to study using radioactive Mössbauer sources. Severe broadening of the nuclear resonance, or an extremely low recoilless fraction, can make it very difficult to excite a resonance using a narrow linewidth radioactive source. In addition, the corresponding reduction in the peak resonant nuclear cross section can prevent any resonant effects from being observed above the background from nonresonant scattering processes. The large bandwidth of synchrotron radiation makes it possible to excite extremely broad lines, and the temporal definition of the pulses makes it possible to eliminate the background from nonresonant scattering. Nonresonant scattering is fast, or prompt, while scattering from the long-lived nuclear resonance is slower, or delayed.

We report observation of resonant nuclear x-ray scattering of synchrotron radiation by the 9.4-keV nuclear level of ^{83}Kr (Ref. 12) in gaseous form. This is the extreme case of a material with a vanishing recoilless fraction: there is no Mössbauer effect in a gas. Furthermore, Doppler broadening reduces the peak resonant cross section in this sample by about 6 orders of magnitude, relative to that of a solid exhibiting the Mössbauer

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effect. However, incoherent[13] nuclear resonant scattering (into 4π sr.) of synchrotron radiation from this sample may be observed because the incident radiation pulse is short compared to the nuclear excited-state lifetime.

This work was done at beamline 6-2 of the Stanford Synchrotron Radiation Laboratory (SSRL). The x-ray beam prepared by a Si(111) monochromator was allowed to fall onto a cell of krypton gas, after passing through a slit and an ionization chamber. A 1.6-cm-diam avalanche photodiode detector[14] was mounted near the cell and a horizontal scattering plane was chosen to reduce the background from prompt Rayleigh scattering (since the synchrotron radiation is largely horizontally polarized).

The krypton used in this experiment was not enriched, having the natural isotopic abundance of 12% ^{83}Kr. The sample cell, about $1\times1\times3$ cm^3, was made small to reduce the background from natural radioactive decay in the krypton sample, probably from ^{81}Kr, which has a half-life of 2.1×10^5 yr and emits a 276-keV photon.[15] This background was less than 0.05 cts/s.

The use of the large-area avalanche diode in this experiment is an important point. Although the isolation of the delayed nuclear scattering signal through time gating was suggested as early as 1962,[16] the limited dynamic range of available detectors has required that additional techniques be used to suppress nonresonant scattering in synchrotron radiation experiments (see, e.g., Refs. 4–6 and 8–11). This greatly increases their complexity. One of the goals of this experiment was to show that effective time gating could be achieved using only simple synchrotron radiation x-ray optics.

The prompt count rate due to nonresonant scattering of synchrotron radiation by the sample was about 7×10^7 x-ray photons/s, as determined from the current through the avalanche diode. The electron storage ring (SPEAR) was run in timing mode, with electron bunches providing x-ray pulses every 195 ns. Each pulse deposited an average of about 14 9.4-keV photons in the detector. By about 40 ns after the prompt flash, the detector had recovered sufficiently to detect single delayed photons.[17]

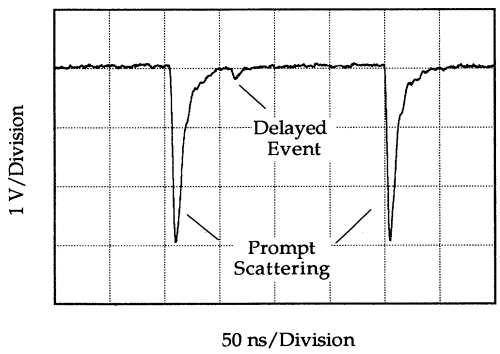

FIG. 1. Delayed single photon event between two prompt flashes from nonresonant scattering (as seen on a Tektronix TDS 620A digital sampling scope, 500 MHz, 2 Gs/s). The avalanche photodiode output has been amplified by 10^3.

Figure 1 shows an oscilloscope trace of the photodiode output, with a single delayed event bracketed by two prompt flashes from nonresonant scattering.

Figure 2 shows the delayed count rate (in the interval 62–108 ns after the prompt flash) as a function of the Si(111) monochromator position. The energy scale of the horizontal axis was determined from K-edge absorption scans of copper and zinc foils. The energy of the first inflection point in each case has been measured using an interferometrically calibrated silicon crystal.[18] The energy of the 9.4-keV transition in ^{83}Kr is found to be 9403.5 ± 1.8 eV, where the error is dominated by the uncertainty in our measurement of the zinc K-edge inflection point. This is in good agreement with earlier work[19] where the resonance energy was determined to be 9400 ± 10 eV using a Si(Li) detector.

The time window for the energy scan shown in Fig. 2 was carefully chosen. Though the electron bunches in the storage ring are nominally separated by 195-ns intervals, there are usually also some electrons in the intervening regions. The current in these "microbunches" is small (parts in 10^7 of the main bunch current), but the nonresonant scattering of the radiation from them appears in the same time window as the delayed nuclear scattering. This creates a background that can be substantially larger than the signal. The microbunches tend to cluster about the main bunches and typically limited the counting period to something like the 50 ns used above. However, during one 4-h period, the number of microbunches was reduced, making it possible to measure the signal between 60 and 180 ns after the prompt pulse. The resulting time response (background subtracted) is shown in Fig. 3, along with a curve showing the natural lifetime of 212 ns measured in Ref. 20. The data are in reasonable agreement with this decay rate ($\chi^2=25$ in 30

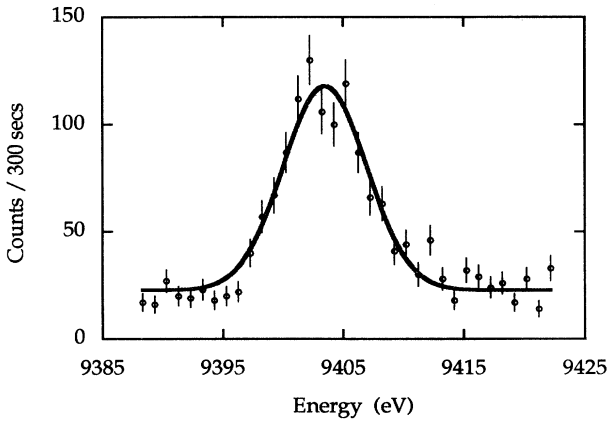

FIG. 2. Delayed count rate (in the interval 62–108 ns after the prompt flash) vs Si(111) monochromator position. The energy calibration is based on Cu and Zn k-edge absorption scans. Peak delayed rate in this time interval was 0.32 cts/s and background rate was 0.07 cts/s. The measured peak count rate of about 0.3 cts/s is consistent with the incident flux and bandwidth, the counting time interval, and the estimated detector efficiency and solid angle. The solid line is a Gaussian fit with constant background. The width of the Gaussian (7.5-eV FWHM) is the monochromator energy resolution.

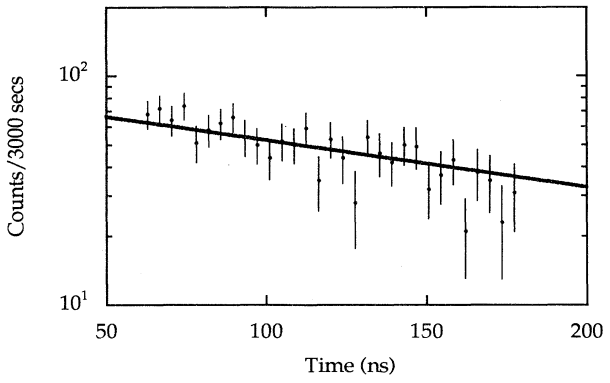

FIG. 3. Scattering rate as a function of time after the synchrotron pulse for the 9.4-keV nuclear transition in ^{83}Kr gas. The solid line corresponds to the nominal ($1/e$) lifetime of 212 ns. The background has been subtracted.

points).

The use of a gaseous sample in this work makes it significantly different than a previously published report of incoherent nuclear scattering of synchrotron radiation from an enriched iron foil.[11] The coherent nuclear forward scattering in the gaseous Kr is negligible so that, unlike in Ref. 11, there is no perturbation of the incoherent scattering due to the coherent forward scattered wave. The time response is then simply the exponential decay of a nucleus in an isolated ^{83}Kr atom.

In the absence of coherent nuclear forward scattering,[13] incoherent nuclear scattering experiments using synchrotron radiation are similar to perturbed angular correlation-distribution (PAC-PAD) experiments.[21] In fact, perturbed angular correlations have been studied previously in gaseous samples.[22] However, synchrotron radiation experiments are potentially simpler in that they involve only one nuclear excited state (up to hyperfine splitting) and have the same particle incident as scattered (thus distinguishing them from PAD experiments requiring a radioactive parent state and PAD experiments with an incident particle beam).

Finally, we note that incoherent scattering could also be used to probe velocity distributions of resonant nuclei in a sample. The bandwidth of the incident synchrotron radiation could be reduced using a high-order Bragg reflection [e.g., the Si (7 3 3) reflection[23]]. An energy dispersive arrangement of a pair of such reflections could reduce the bandwidth down to ~ 10 meV, allowing investigation of velocity scales down to the level of 300 m/s. At the meV level (~ 30 m/s Doppler shift at 9.4 keV), one can envision investigating atomic velocity distribution profiles in gases or liquids. This would complement recent work using nuclear resonant scattering to investigate the phonon density of states in polycrystalline α-^{57}Fe (Ref. 24) and the phonon excitations in a powder sample of a large macromolecule.[25]

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[1]R. L. Mössbauer, Z. Phys. **151**, 124 (1958).
[2]See reprints in H. Frauenfelder, *The Mössbauer Effect* (Benjamin, New York, 1962).
[3]See, e.g., N. N. Greenwood and T. C. Gibb, *Mössbauer Spectroscopy* (Chapman and Hall, London, 1971); *Mössbauer Spectroscopy,* edited by D. P. E. Dickson and F. J. Berry (Cambridge University Press, Cambridge, 1986).
[4]E. Gerdau, R. Rüffer, H. Winkler, W. Tolksdorf, C. P. Klages, and J. P. Hannon, Phys. Rev. Lett. **54**, 835 (1985).
[5]R. Rüffer, Sync. Radiat. News **5**, 25 (1992).
[6]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).
[7]S. Kishimoto, Nucl. Instrum. Methods Phys. Res. A **309**, 603 (1991).
[8]J. B. Hastings, D. P. Siddons, U. van Bürck, R. Hollatz, and U. Bergmann, Phys. Rev. Lett. **66**, 770 (1991).
[9]M. Grote, R. Röhlsberger, M. Dimer, E. Gerdau, R. Hellmich, R. Hollatz, J. Jäschke, E. Luken, R. Rüffer, H. D. Rüter, W. Sturhahn, E. Witthoff, M. Harsdorff, W. Pfützner, M. Chambers, and J. P. Hannon, Europhys. Lett. **17**, 707 (1991).
[10]A. I. Chumakov, G. V. Smirnov, A. Q. R. Baron, J. Arthur, D. E. Brown, S. L. Ruby, G. S. Brown, and N. N. Salashchenko, Phys. Rev. Lett. **71**, 2489 (1993).
[11]U. Bergmann, J. B. Hastings, and D. P. Siddons, Phys. Rev. B **49**, 1513 (1994).
[12]P. T. Barrett, Proc. Phys. Soc. London Sect. A **65**, 450 (1952).
[13]For discussion of some of the distinctions between coherent and incoherent nuclear scattering see J. P. Hannon and G. T. Trammell, Physica B **159**, 161 (1989); U. van Bürck and G.V. Smirnov, Hyperfine Interact. **90**, 313 (1994).
[14]A. Q. R. Baron and S. L. Ruby, Nucl. Instrum. Methods Phys. Res. A **343**, 517 (1993).
[15]*Table of Isotopes,* edited by C. M. Lederer and V. S. Shirley (Wiley, New York, 1978), p. 288.
[16]E. J. Seppi and F. Boehm, Phys. Rev. **128**, 2334 (1962).
[17]The high prompt rate also introduced a positive offset in the baseline of the output of the (capacitively coupled) detector. This would have prevented the discriminator from firing on some single photon events if it had not been corrected.
[18]J. Stümpel, P. Becker, S. Joksch, R. Frahm, and G. Materlik, Phys. Status Solidi A **124**, 565 (1991).
[19]B. Kolk, F. Pleiter, and W. Heeringa, Nucl. Phys. **A194**, 614 (1972).
[20]S. L. Ruby, Y. Hazoni, and M. Pasternak, Phys. Rev. **129**, 826 (1963).

[21]See, e.g., H. E. Mahnke, Hyperfine Interact. **49**, 77 (1989).
[22]See, e.g., H. Seyb, S. Kuberczyk, C. Ruth, L. Ziegeler, and I. Borchert, Hyperfine Interact. **77**, 301 (1993).
[23]D. E. Johnson, D. P. Siddons, J. Z. Larese, and J. B. Hastings (unpublished).
[24]M. Seto, Y. Yoda, S. Kikuta, X. W. Zhang, and M. Ando (unpublished).
[25]A. I. Chumakov (private communication).