Nuclear Resonance Excitation by Synchrotron Radiation

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We have excited the 14-keV level of the Fe^{57} nucleus using synchrotron radiation, and observed the conversion electrons emitted in the decay of that state. We believe this to be the first observation of nuclear excitation via synchrotron-produced x rays.

Synchrotron radiation (SR) from electrons in storage rings has recently been used for a wide variety of experiments in physics, chemistry, and biology.[1,2] Increasing demand for synchrotron radiation sources has led to the construction of a number of new facilities throughout the world, and a further increase of experiments involving the interaction of synchrotron-produced x-rays with electrons can be anticipated. Experiments based on the interaction of synchrotron radiation with nuclei have been widely proposed,[3-5] but until this report, so far as we are aware, there has been no experimental success. The equipment and research described in this Letter demonstrate the feasibility of some of those concepts, and also exemplify a new approach to the detection of small signals in the presence of heavy overloads.

Synchrotron radiation produces an intense (e.g., at Stanford University, at 10 keV, under optimal conditions, 2×10^{11} photons eV^{-1} sec^{-1} in a 0.2-cm^2 spot) highly collimated, pulsed beam of x radiation, and it should in principle be easy to excite low-lying nuclear states with these x rays.[6] However, the experiments are in fact extremely difficult, because at the energy levels available from current synchrotrons and monochromators, all of the accessible nuclear levels are extremely narrow (typically 10^{-8} eV). The SR is emitted in a continuum which can be monochromatized to only about a 1-eV width, so that only $\sim 10^{-8}$ of the incident photons can be absorbed by the nuclear levels. Thus, the nuclear fluorescence excited by the SR is far weaker than radiation nonresonantly scattered by the electronic processes, since the latter arise from the entire 1-eV width of the monochromatized SR.

Two refinements have been proposed to deal with this problem. The first, the use of nuclear Bragg scattering, effectively enhances the ratio of nuclear to electronic cross sections, and is currently being pursued by two groups.[5,7] The second, first outlined by Ruby,[3] uses the relatively long lifetime of the nuclear state, and the pulsed nature of the SR, to discriminate between the nuclear and electronic events. It is this approach, described in detail below, that we have used here.

The experiment configuration is shown in Fig. 1(a). The SR is monochromatized by a channel-cut crystal permanently mounted in the SSRL (Stanford Synchrotron Radiation Laboratory) beam line, and radiation of the appropriate energy is absorbed by the Fe^{57} nuclei in the iron foil target (1.85 mg/cm^2, enriched to 90.6% in Fe^{57}). The 14-keV level excited by this process decays with a 100-nsec half-life, primarily by emitting conversion electrons of ~7 and ~13 keV. These con-

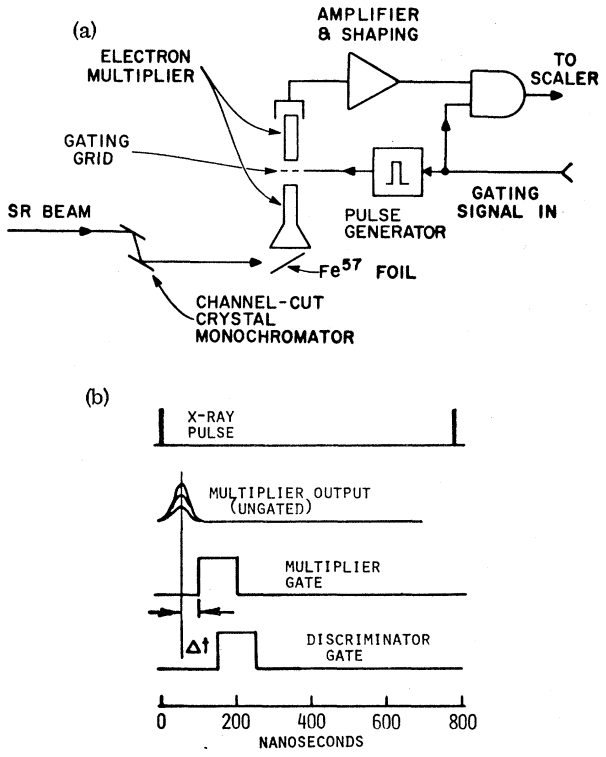

FIG. 1. (a) Schematic drawing of gated conversion-electron detector used at the Stanford storage ring. (b) Timing diagram for the gating circuits shown in (a). The timing sequence is repeated at the 1.28-MHz rotational frequency of the synchrotron.

version electrons enter a continuous-dynode Channeltron[8] electron multiplier which is divided into two stages. The multiplier is gated by a grid between the two stages. The electron-multiplier output pulse is amplified by a fast charge-sensitive preamplifier, and then further amplified and delay-line clipped to provide pulses ~50 nsec wide into a gated discriminator. Timing circuits provide for both the detector and discriminator to be gated off during the time that the SR pulse arrives at the detector, and then turned on ~100 nsec later. This delay allows for the photoelectrons, Compton electrons, and low-energy secondary electrons to dissipate before the detector is turned on. Very low-energy electrons, which could have flight times as long as 100 nsec, are rejected by a grid and retarding potential in the electron-multiplier entrance. The detector and discriminator are gated on for 100 nsec, and then turned off to wait for the next SR pulse [Fig. 1(b)]. The sequence is repeated for every SR pulse, at a 1.28-mHz rate.

The reasons for these gating arrangements are apparent when the signal and interference rates are considered. Under typical operating conditions,[9] about five photoelectrons are detected for each SR pulse. These photoelectrons have an energy spectrum identical to that of the conversion electrons we are trying to observe. Under typical conditions,[9] approximately twenty nuclear excitations are produced per second, and after we account for the electron detector solid angle and detection efficiency, and the 25% gating fraction, the anticipated counting rate is 0.2/sec due to the nuclear events. Thus we are trying to detect single-electron "true" events occurring at ~0.2/sec in a background of five electron events occurring at 1.28 MHz. Electron-multiplier afterpulsing on the photoelectrons will tend to appear as "delayed counts" indistinguishable from the conversion-electron counts we are looking for. Thus afterpulsing levels on the order of 10^{-6} must be achieved, and this is substantially better than can be obtained with conventional electron-multiplier structures, where afterpulsing levels of ~5% are normally obtained.[10]

The detector we have developed to deal with these demanding conditions is shown in Fig. 1(a).[11] It consists of a two-stage, continuous, semiconducting-dynode electron multiplier[8] with gating by a grid between the two stages. The grid is gated "off" during the prompt photoelectron pulse, and the amplification of the photoelectrons is thus terminated at the end of the first stage. This procedure minimizes the total charge that passes through the electron multiplier, and thus removes the main cause of afterpulsing, which has been established as ionization of residual gas by the multiplied electrons.[10] It would be difficult to gate the input end of the multiplier by grids or retarding potentials, because the photoelectron energy spectrum extends to 14 keV.

Figure 1(b) shows the timing of the detector operation. The discriminator gate provides better time definition than the electron-multiplier gate, which is turned off gradually over about 150 nsec.

The detector has been used at SSRL in a standard beam line, with a Si⟨220⟩ channel-cut crystal monochromator. The monochromator is scanned over the energy of the Fe^{57} nuclear resonance, and counting rate is plotted as a function of x-ray energy (Fig. 2). The nuclear resonance is inherently 5×10^{-9} eV wide, and is broadened to 5×10^{-7} eV by the magnetic hyperfine structure in the iron foil. This width is negligible compared to the x-ray monochromator width, ~3 eV, so that the scan actually profiles the monochromator transmission function by sweeping over the nuclear resonance, which is effectively a δ function by comparison. The width and shape of

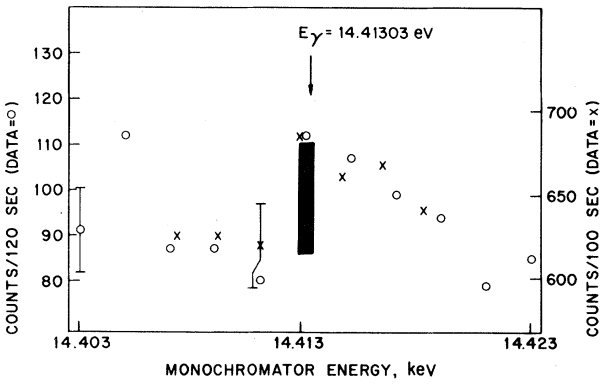

FIG. 2. Resonance curves observed by scanning the x-ray monochromator over the nuclear excitation energy. The hyperfine-broadened nuclear resonance is 5×10^{-7} eV wide, and so the shape of the observed resonance actually represents the monochromator pass function (see text). Actual numbers of counts are shown, for 100 sec (×) or 120 sec (○) counting times, for two different runs. The vertical scales have been selected to merge the data points. Changes to the detector between the two runs account for the reduced background and slightly lower efficiency in the second run. The vertical bar shows the position and calculated size of the resonance under these operating conditions.

the resonance curve thus should reflect the shape of the monochromator output. This shape function is a convolution[12] of the Darwin width of the diffracting crystal and the vertical angular divergence of the SR beam, determined by the combination of the slit function and vertical deviation (from the mean orbital plane) of the electron orbits. The last term appears to be dominant. In the SSRL beam line, the monochromator output would be a symmetric, approximately triangular function, about 3 eV wide at the base. Additional broadening and deviations from symmetry, as appear to be observed in Fig. 2, could be caused by geometric misalignment of the monochromator or defining slit, or excessive divergence of the electron orbits.

Figure 2 shows that the detector sensitivity to nonnuclear events has been reduced to a small, but nonnegligible, value, and the nuclear resonance counts were observed above this background. The background arises from photoelectric events, and would be expected to be smooth over the narrow energy range scanned, since there are no absorption edges in this energy region.

Although only a weak resonance was observed in the measuring time available (only 33 minutes for all the data shown, as a result of limited running time and the stochastic nature of beam availability), it is unequivocally exactly of the anticipated energy and the right intensity. The expected nuclear resonance amplitude can be calculated from the x-ray flux at 14 keV, measured by an ionization chamber in the beam line ahead of the detector, and the gated detector sensitivity is known to be 0.01 counts per resonantly absorbed photon, determined from transmission Mössbauer experiments using a radioactive source. The resonance amplitude calculated from these values is shown by the vertical black bar in Fig. 2. The nuclear resonance energy is known to be $14.413\,03 \pm 0.000\,08$ keV.[13] The monochromator energy calibration has been established (in the first run) by reference to features in the L_{III} edge of Pt metal,[14] and in the second run by an absolute measurement of the Bragg-scattering angle and the known lattice constant of Si. These calibrations are seen in Fig. 2 to be consistent with the observed resonance curve. We therefore believe that we have demonstrated that it is possible to excite, and observe, nuclear states by synchrotron radiation.

The approach we have described could be immediately used for studying profiles of Bragg-scattering monochromators. In the next few years, photon fluxes 10^4 times those used here will be available at a number of synchrotron facilities, and our approach could be used for hyperfine interaction or other solid-state studies. The low-afterpulsing, high-repetition-rate instrumentation developed here could also be useful in neutron time-of-flight experiments.

We thank all of the staff of SSRL for assistance in interfacing our apparatus to existing facilities, often under difficult circumstances. We especially express our thanks to G. S. Brown for establishing the monochromator energy calibration, and to J. A. Kirby for help with the on-line data accumulation routines. We thank B. M. Kincaid for helpful comments. Part of this work was performed at SSRL, which is supported in part by National Science Foundation Grant No. DMR 73-07692 in cooperation with the Stanford Linear Accelerator Center and U. S. Department of Energy.

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[7]See S. L. Ruby, Stanford University Synchrotron Radiation Laboratory Report No. 77/11 (unpublished), p. 30; P. Flinn, *ibid.*, p. 62.

[8]Made by Galileo Electro-Optics, Sturbridge, Mass. We are indebted to Mr. Paul Henkel of Galileo for helpful discussions on the properties of these multipliers.

[9]Stanford storage ring at 3.5 GeV, 30 mA of electron current, $\sim 2 \times 10^9$ 14-keV photons/sec incident on 1-cm-wide detector foil, with monochromator passband of about 3 eV.

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Polarization Analyzing Power $A_y(\theta)$ in pp Elastic Scattering at 643, 787, and 796 MeV

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Data have been obtained for the polarization analyzing power $A_y(\theta)$ in pp elastic scattering from near 30° to 90° (c.m.) at 643, 787, and 796 MeV. Relative uncertainties are typically ±0.003 with an overall normalization uncertainty of $(^{+1}_{-0.5})\%$. Data are not consistent with existing phase-shift analyses.

Below 400 MeV the proton-proton interaction proceeds essentially through the elastic channel with recent phase-shift analyses[1] indicating that the data are reasonably consistent. In the intermediate energy range from 400 MeV to 1 GeV considerable uncertainty still exists in our knowledge of the phase shifts.[2,3] With the advent of a polarized ion source at the Clinton P. Anderson Meson Physics Facility (LAMPF), a program of experiments is underway to determine uniquely the proton-proton phase shifts at energies in the 400- to 800-MeV range to complement the recent impressive results appearing from TRIUMF.[4] This paper describes measurements of the polarization analyzing power $A_y(\theta)$ in pp elastic scattering that are almost an order of magnitude more precise than earlier data above 500 MeV.

Figure 1 compares our polarization analyzing power measurements at 796 MeV with a calculation made by extrapolating from 750 to 800 MeV the energy-dependent phase-shift solution of MacGregor, Arndt, and Wright, paper XIII[2] (shown as a dashed line) and a calculation with the imaginary part of the $\delta(^1D_2)$ increased by 30° (shown as a solid line). Although the fit is improved by this change, in agreement with what we found in fitting our elastic scattering differential cross sections at this energy reported earlier,[5] this should only be taken as an indication of the large uncertainties in our knowledge of the phase shifts at this energy. Measurements at 643 MeV show a similar shape but the polarization analyzing power at the maximum is increased to 0.55. Figure 2 shows a plot of the polarization analyzing power near its maximum value [about 40° (c.m.)] as a function of proton energy in the intermediate range. Our three data points at 643, 787, and 796 MeV are generally consistent with, but much more precise than, previous data near these energies.

The technique for measuring polarization analyzing power is well known.[7] In this experiment a beam of protons with transverse polarization up to 0.92 was obtained from the LAMPF accelerator and focused (typically 4 mm diam) onto a CH_2 target. Data were taken at three beam energies, two of which were measured by the High Resolution Spectrometer (HRS) to be 796 ± 2 and 787 ± 2 MeV; the third energy was estimated to be 643 ± 4 MeV from accelerator operating param-