Observation of Nuclear Excitation by Electron Transition in ^{197}Au with Synchrotron X Rays and an Avalanche Photodiode

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We have succeeded in observing nuclear excitation by electron transition (NEET) in ^{197}Au by a new method. Monochromatic x-rays of synchrotron radiation were used to ionize the K shell of gold atoms in a target foil. The internal-conversion electrons emitted from excited nuclei were detected with a silicon avalanche photodiode. At a photon energy of 80.989 keV, the NEET probability in ^{197}Au was determined to be $(5.0 \pm 0.6) \times 10^{-8}$ from a comparison of the event number per photon between NEET and the nuclear resonance at 77.351 keV.

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Nuclear excitation by electron transition (NEET) occurs during atomic inner-shell ionization if the nuclear and electron transitions have nearly the same energy and a common multipolarity [1]. For ^{197}Au, the NEET condition is satisfied in the $K \rightarrow M_1$ hole transition ($1S_{1/2}$: 80.725 keV $\rightarrow$ $3S_{1/2}$: 3.425 keV [2]) and the $3/2^+ \rightarrow 1/2^+$ nuclear transition ($0 \rightarrow 77.351$ keV; half-life: 1.91 ns [3]). The energy difference between the atomic and nuclear transitions is only 51 eV. The common multipolarity is *M1*. Calculations of the NEET probability per K-shell hole have been executed by applying different theoretical models [4–8]. The maximum probability is 3.5×10^{-5} [4] and the lowest value is 1.3×10^{-7} [8]. Fujioka *et al.* [9] have performed the only previously reported measurement on NEET in ^{197}Au. They used 100-keV pulsed electron bombardment and a stroboscopic electron spectrometer to detect 63-keV L-conversion electrons [10]. In the experiment, the NEET probability was estimated to be $(2.2 \pm 1.8) \times 10^{-4}$, which was recently corrected to $(5.1 \pm 3.6) \times 10^{-5}$ [11]. It seems that the experiment had a large uncertainty in determining the efficiency of the system and, however, a small efficiency of about 10^{-6}. The difference between the experimental value and the recent calculations is still large, more than 2 orders of magnitude. Therefore, a new experiment has been strongly desired to confirm whether the NEET phenomenon is detectable or not and to determine accurately the NEET probability.

In this paper, we report on an observation of NEET in ^{197}Au by a new method. We adopted photoionization by monochromatic x-rays of synchrotron radiation, instead of electron bombardment. The x-ray beam was tuned to an energy just above the gold K level. X-ray pulses of 50 ps long (FWHM) periodically excited the gold atoms and nuclei in a target, using a several-bunch operation of a storage ring. A simple system with a silicon avalanche photodiode (APD) recorded time spectra of the radiation (mainly L internal-conversion electrons) emitted from excited nuclei. The APD was located close to the target for a large efficiency. By tuning the x-ray energy, we also observed the nuclear resonance at 77.351 keV with the same setup as in the NEET measurement. If we use the same size of the incident beam and the same target, we can consider that the ratio of the event numbers per incident photon equals the ratio of the cross sections for NEET and the nuclear resonance. Since the NEET probability is given by the ratio of cross sections for NEET and the K photoionization, the probability can be precisely determined from the measured ratio of the event number per photon and from the known values of the cross sections for the nuclear resonance and the K photoionization. An exact estimation of the detector efficiency is not necessary for our method.

An experimental difficulty lies in the intense radiation promptly generated from the target by atomic processes. Detector pulses caused by the prompt radiation often pile up and become huge, interfering with signals of the conversion electrons that rapidly decay with a lifetime of 2.76 ns. Thus, we had to inhibit the measurement during the first few ns after the prompt pulse. For decreasing the dead time, we used the APD that had a ns-width pulse and a sub-ns time resolution [12]. Another difficulty is that a background in a time spectrum is made from the prompt radiation due to residual bunches of electrons in a storage ring. Between the main buckets of electrons, there are some buckets that are spaced 2 ns apart and are not completely empty. Although the intensity associated with these electron sub-bunches is usually less than 10^{-6} of the main bunch, the background rate is often the same as or higher than that of the weak signals due to the nuclear deexcitation. Evaluating the background is important to determine the event number of the nuclear deexcitation. We thus measured the spectrum at an energy slightly different from that at which the nuclear resonance or NEET occurs.

The time-spectroscopy experiments were carried out at undulator beam line BL09XU of the SPring-8 when the ring was operated in a 116-bunch mode (bunch

period: 42 ns). Figure 1 illustrates the experimental setup. The third-order x rays from a Si(111) double-crystal monochromator were used to obtain high energies of 77–81 keV, while the first-order x rays were attenuated to $\sim 10^{-5}$ by 26-mm-thick aluminum plates. An x-ray beam of $^{V}1.5$ mm $\times$ $^{H}1.0$ mm was passed to a vacuum chamber through a beryllium window. The beam intensity was monitored by a pair of silicon PIN photodiodes (PDs) having a 5 mm $\times$ 5 mm area of 500-μm thickness without a thick dead layer. PD currents were processed by current amplifiers and voltage-to-frequency converters. In the chamber, the x-ray beam was incident on a gold metal foil (8 mm in diameter and 3 μm thick) inclined by 30° to the beam axis. The radiation emitted from the target was detected by a silicon APD having a sensitive area of 3 mm in diameter and a depletion layer 30 μm thick. The APD was located about 2.5 mm above the target. We used a fast amplifier (bandwidth: 100 kHz–1.8 GHz, gain: 100) to obtain fast negative pulses of the APD outputs, which were processed with a constant fraction discriminator (CFD). The outputs from the CFD were used as the start signals into a time-to-amplitude converter (TAC). Here the outputs were inhibited by veto signals, except for a time region of 4.5–20 ns after the prompt radiation during a period of 42 ns. The veto signals into the CFD and the stop signals into the TAC were supplied through some divider and delay modules by 508.58-MHz signals from the accelerator. The time spectra of the resonance and NEET were recorded by the TAC and a multichannel analyzer. Before the observation of NEET, we found a nuclear resonance at 77.351 keV from a peak of delayed events by the APD while scanning the θ angle of the monochromator. A time spectrum was measured at the peak of 77.351 keV. The background spectrum was recorded at 77.455 keV. A maximum of the APD count without the veto signal was obtained at 80.989 keV and the APD count reached a minimum at 80.415 keV, in an x-ray absorption measurement around the K edge under a low beam intensity. According to this result, the time spectra were measured at 80.989 keV for the NEET observation and at 80.415 keV for evaluating the background. Except for the incident x rays, both spectra were taken under the same condition as in the resonance measurements. The beam size was not changed.

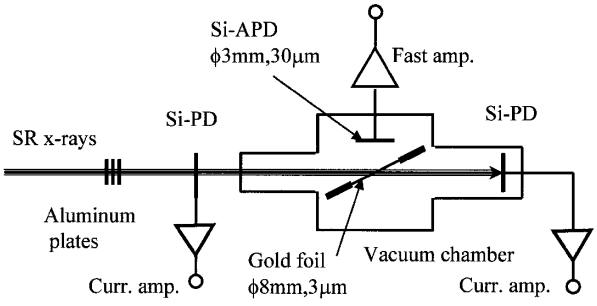

FIG. 1. Schematic of the experimental setup.

Figure 2 shows the radiation that the APD detected. The energy spectra were measured at beam line BL-14A of the Photon Factory using x rays of $\sim 10^6$ photons/s. The arrangement and the applied voltage to the APD were the same as in the time-spectroscopy experiment, although a charge-sensitive preamplifier was used instead of a fast amplifier. The CFD threshold level for the APD outputs corresponded to a pulse height of ~35-keV radiation when using the fast amplifier. Thus, the photoelectrons mainly contributed to the count of the prompt radiation while L x-rays were not counted except in the piling prompt pulses. No peaks of fluorescent K x-rays were observed even at 80.989 keV. This is due to a poor efficiency, less than 0.2%, of our APD for the K x-rays, although the fluorescence yield for the gold K shell is high, 0.964 [13]. Auger electrons increased the APD count at 80.989 keV, which was 15% over that at 80.415 keV. In an energy spectrum using 77.35-keV x rays, as a model for detecting L-conversion electrons, ~70% of L photoelectrons had energies higher than 35 keV. From an intrinsic efficiency of >80% for the electrons [14], more than 60% of the L-conversion electrons irradiated to the APD would be measured by the time-spectroscopy system.

Figure 3 shows the number of delayed events measured around 77.351 keV as a function of the monochromator energy. We approximated a peak profile at 77.351 keV to a triangle, after subtracting the background fitted by a straight line, as shown in the lower side of Fig. 3. The full width at half maximum (FWHM) of the resonance peak was 19 $\pm$ 2 eV, which corresponded to the energy width of the incident x rays from the monochromator. The time spectrum for the resonance, measured at the peak during 7466 s in one injection period, is shown in Fig. 4(a). The time of 0 ns is the timing of the prompt radiation, measured without a veto signal. The conversion electrons produced a decaying structure after the prompt radiation.

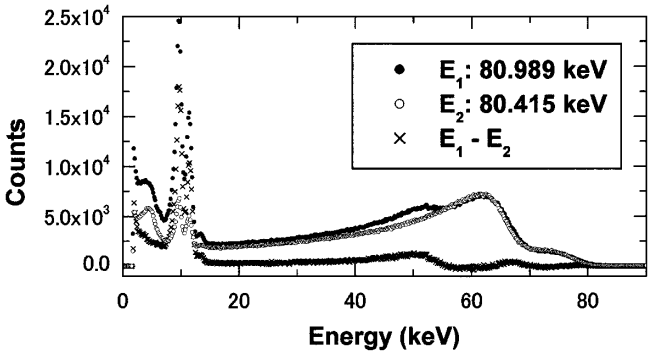

FIG. 2. Energy spectra of the radiation emitted from gold foil, measured with the APD. The energy of the incident x rays was 80.989 keV for the solid circles and 80.415 keV for the open circles. The crosses indicate the difference between them. The data were normalized to the beam intensity. M, N, and L photoelectrons (<80 keV and <69 keV) and L x-rays (9–13 keV) were observed at 80.415 keV. KLL-, KLM-Auger electrons (<57 keV and <67 keV) were additionally observed at 80.989 keV.

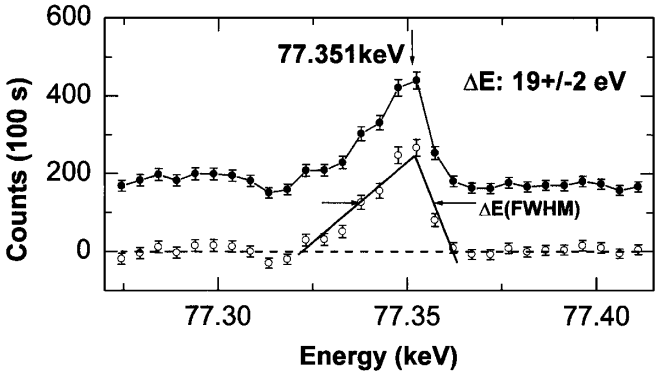

FIG. 3. Number of delayed events measured by the APD as a function of the monochromator energy: the data with the background (solid circles; upper) and the background-subtracted data (open circles; lower). The energy of the horizontal axis was determined from the peak at 77.351 keV and from the scanning angle of the monochromator. A straight line was assumed as the background structure. The peak profile was approximated to a triangle with ΔE (FWHM) $= 19 \pm 2$ eV by least-squares fit.

Figure 4(b) shows the background spectrum, recorded at 77.455 keV during 4655 s of the same run. The background consisted of a tail structure formed by the prompt radiation and of peaks with a 2-ns interval caused by the electron sub-bunches. Figure 4(c) indicates the spectrum subtracted the data in Fig. 4(b) from those in Fig. 4(a). The subtraction was carried out by considering the number of incident photons, which was obtained from the current of the PD at the backside. A correction for the absorption in the target, and a calibration to the photon rate using tungsten absorbers and a scintillation detector, was applied for the PD current. Figure 5(a) indicates the time spectrum obtained at 80.989 keV during 16 091 s of another injection period. The decaying structure is clearly seen. The spectrum, obtained at 80.415 keV during 8076 s of accumulation under the same conditions as at 80.989 keV, is shown in Fig. 5(b). The tail and the residual bunch structure exist similarly in Fig. 4(b), although the count rate in the region of 4.5–20 ns was 0.3×10^{-7}, less than 1.3×10^{-7} in Fig. 4(b), compared with that in the region involving the main bunch. We obtained the data plotted in Fig. 5(c) by subtracting the data in Fig. 5(b) from those in Fig. 5(a), considering the difference in the incident photon number. The bunch structure of a 2-ns interval slightly remains in Figs. 4(c) and 5(c). This may be due to a change in the x-ray intensity from the sub-bunches, relative to that from the main bunch, during measurements of (a) and (b) spectra.

We obtained the event numbers of conversion electrons from the background-subtracted spectra. The event numbers between 5 and 15 ns were $N_R = 9878 \pm 169$ counts during 7466 s in Fig. 4(c) and $N_{\mathrm{NEET}} = 2994 \pm 101$ counts during 16 091 s in Fig. 5(c). Here we neglected the data in the regions of 4.5–5.0 ns and 15–20 ns in order to eliminate the effects of the prompt radiation

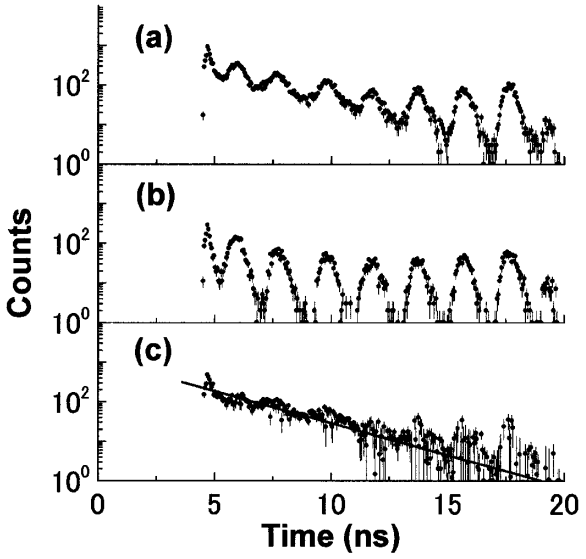

FIG. 4. Time spectra observed for (a) the nuclear resonance at 77.351 keV during 7466 s and for (b) the background at 77.455 keV during 4655 s. We obtained the data plotted in (c) by subtracting the data in (b) from those in (a), considering the number of incident photons. The solid line in (c), an exponential decay curve fitted for the data of 5–15 ns, gives a lifetime of 2.68 ± 0.14 ns.

and of low quality in statistics, respectively. The numbers of incident photons during each measuring time were given by $I_R = (5.04 \pm 0.06) \times 10^{13}$ and $I_{\mathrm{NEET}} = (10.54 \pm 0.10) \times 10^{13}$ for the resonance and NEET measurements, respectively. An exponential decay curve was fitted for data of 5–15 ns in Fig. 4(c) or for those in

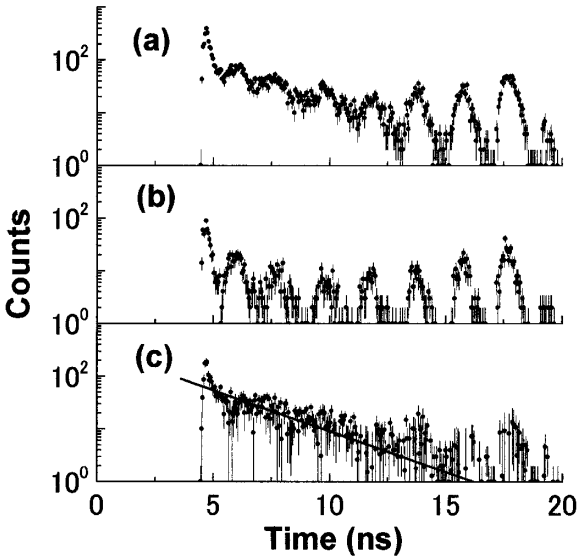

FIG. 5. Time spectra observed for (a) NEET at 80.989 keV during 16 091 s and for (b) the background at 80.415 keV during 8076 s. We obtained the data plotted in (c) by subtracting the data in (b) from those in (a), considering the number of incident photons. The solid line in (c) indicates an exponential decay curve fitted for the data of 5–15 ns, which gives a lifetime of 2.80 ± 0.29 ns.

Fig. 5(c), shown by the solid line. The lifetimes τ were obtained as $\tau = 2.68 \pm 0.14$ ns and $\tau = 2.80 \pm 0.29$ ns, respectively, both of which satisfied $\tau = 2.76$ ns for the 77.3-keV level within the error.

Using the NEET cross section σ_{NEET}, the NEET probability P_{NEET} is expressed by

$$P_{\mathrm{NEET}} = \sigma_{\mathrm{NEET}}/\sigma_K\,, \tag{1}$$

where σ_K is the photoelectric cross section of the K shell. Here σ_K is given by $(2.18 \pm 0.06) \times 10^{-21}$ cm^2 from the photoelectric cross section at the gold K-edge [$= (2.74 \pm 0.08) \times 10^{-21}$ cm^2] and from its relative K-shell contribution ($= 0.794$) [15]. Considering the narrow width of the resonance absorption, involved in the energy width of the incident x-ray beam, we define the effective cross section for nuclear resonance at 77.351 keV, σ_R, by

$$\sigma_R = (\Gamma/W) f_p \sigma_0\,, \tag{2}$$

where Γ is the width of the resonance absorption (FWHM), W is the observed energy width of the incident x rays (FWHM, 19 ± 2 eV), f_p is a factor that depends on the spectral function of the incident x rays, and σ_0 is the maximum resonance cross section [$= (3.86 \pm 0.05) \times 10^{-20}$ cm^2 [16]]. Here $f_p = 1$ if the peak shape of the incident x rays is the same Lorentz form as that of the resonance absorption. However, the spectral function of the x-ray beam was approximated to a triangular form. When the peak height and FWHM of a triangle are the same as those of a Lorentz form, the Lorentz form is larger in area than the triangle by a factor of $\pi/2$. Thus, f_p is $\pi/2$. Since we can neglect the self-absorption effect when using a thin gold foil, Γ is taken to be $\Gamma = \Gamma_0$, where Γ_0 is the natural linewidth for the 77.3-keV state [$= (2.38 \pm 0.02) \times 10^{-7}$ eV] [17]. We obtained the NEET and the resonance events for the same region of 5–15 ns, as noted above, giving N_{NEET} and N_R, respectively. When they are divided by the incident photon numbers I_{NEET} and I_R, respectively, the ratio of σ_{NEET} to σ_R is given by

$$\sigma_{\mathrm{NEET}}/\sigma_R = (N_{\mathrm{NEET}}/I_{\mathrm{NEET}})/(N_R/I_R)\,. \tag{3}$$

The probability of P_{NEET} in Eq. (1) is deduced from Eqs. (2) and (3) as

$$\begin{aligned} P_{\mathrm{NEET}} &= (\sigma_0/\sigma_K)\,(\Gamma/W) f_p (N_{\mathrm{NEET}}/I_{\mathrm{NEET}})/(N_R/I_R) \\ &= (5.0 \pm 0.6) \times 10^{-8}. \end{aligned} \tag{4}$$

The error on P_{NEET} is mostly determined by the error on W. Table I indicates the calculated and experimental values for the NEET probability on ^{197}Au. Our value is fairly close to the result calculated by Tkalya [8], though it is less than one-half of it.

In conclusion, we observed NEET in ^{197}Au by a new method. Monochromatic synchrotron x rays were used to produce holes in the K shell of gold atoms. A silicon APD enabled us to distinguish the weak conversion electrons from the intense prompt radiation with a short dead time of 5 ns in a time spectrum. By tuning the x-ray energy, we also observed the nuclear resonance of ^{197}Au at 77.351 keV with the same setup as for the NEET observation at 80.989 keV and evaluated the background due to the electron sub-bunches in the storage ring. Note that an energy of 77.351 keV is the highest level in the Mössbauer nuclides, which has been successfully excited by synchrotron radiation. From the measured ratio of the event numbers per incident photon in NEET and the resonance, the NEET probability was given by $P_{\mathrm{NEET}} = (5.0 \pm 0.6) \times 10^{-8}$. This value is smaller by 3 orders of magnitude than the previous experimental value [11] but is much closer to the recent calculations. Although our experiment does not explain why the NEET probability is so small, the present result is more reliable for examining theoretical models.

TABLE I. Calculated and experimental values of the NEET probability on ^{197}Au.

Reference	Theory	Experiment
Fujioka *et al.* [9]		$(2.2 \pm 1.8) \times 10^{-4}$
Pisk *et al.* [4]	3.5×10^{-5}	
Ljubičić *et al.* [5]	2.2×10^{-5}	
Tkalya [6]	1.4×10^{-7}	
Ho *et al.* [7]	2.4×10^{-7}	
Tkalya [8]	1.3×10^{-7}	
Shinohara *et al.* [11][a]		$(5.1 \pm 3.6) \times 10^{-5}$
Present work		$(5.0 \pm 0.6) \times 10^{-8}$

[a]The estimation of the probability was corrected by using the same experimental result as in Ref. [9].

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