Active pumping of ^{229}Th nuclear clock isomer by synchrotron radiation-based nuclear resonant scattering

A nuclear system is one of the ideal quantum states because it is well isolated from external disturbance by the surrounding electrons, and thus it is highly irrefrangible. If a nucleus can be operated using a coherent light source laser, one can expect to realize a research field of quantum electronics based on nuclei instead of atoms or molecules. However, the accessible laser energy is up to only ~10 electron volts (eV) and it is impossible to excite nuclei using a laser because the transition energy of nuclei is usually higher than the keV or MeV range.

The only exception is the ^{229}Th nucleus. This nucleus has a first excited state of extraordinarily low energy. This state is called an isomeric state or isomer. The energy level of the isomeric state is only ~8 eV, which corresponds to the vacuum ultraviolet (VUV) or light with ~150 nm wavelength; therefore, the transition between the ground state and the isomeric state is accessible by a laser technique. Although this fact was first pointed out in the 1970s, the existence of the isomeric state was confirmed only recently. In 2007, the isomer energy was reported to be 7.8 ± 0.5 eV on the basis of indirect gamma-ray spectroscopy [1], and in 2016, deexcitation from the isomeric state via internal conversion was observed [2]. However, the energy of the isomeric state is still ambiguous, and the lifetime of the isomeric state is also unknown. Furthermore, no optical transition between the two states has been reported.

Even though the details are not yet clear, many applications based on this laser-accessible nuclear transition have been proposed. The most famous example is the ultrahigh-precision frequency standard: the "nuclear clock." An atomic clock which is used as a current frequency standard is a base technology in both science and society. The current precision of the atomic clock is about 1 part in 10^{18}. A nuclear clock based on ^{229}Th nuclei is expected to be much more precise, 10^{19}–10^{20}. A ^{229}Th nuclear clock may provide many applications, for example, in relativistic geodesy, in the search for dark matter, and in the observation of possible variation of the fundamental constants of physics [3].

Our group aims to observe the VUV photon emission via deexcitation from the isomeric state to accurately determine the energy and lifetime of the isomeric state. Figure 1 shows the principle of our scheme. First, the X-ray beam is used to irradiate ^{229}Th nuclei in the ground state so that the nuclei undergo a transition to the second excited state

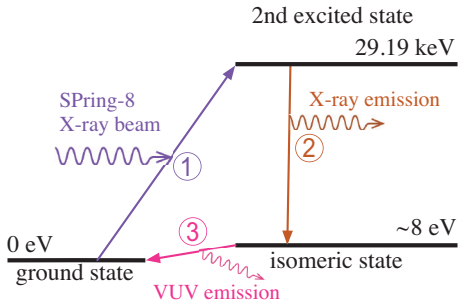

Fig. 1. Transitions used in the experiment. The three horizontal lines represent the three lowest ^{229}Th nuclear levels.

(process 1 in Fig. 1). Then, the nuclei spontaneously predominantly decay to the isomeric state; hence, the population of the isomeric state is increasing during beam irradiation (process 2). Finally, we observe the VUV photons emitted during the deexcitation from the isomeric state to the ground state (process 3).

A key point of the scheme is the preparation of the isomers. All the past experiments that reported positive results used ^{233}U α-decay as an isomer source. Since the α-decay causes radioactivity that disturbs the measurement, the ground state of ^{229}Th is preferred as an isomer source. To produce the isomers from the ground state, we need to pump up the nuclei actively. However, no attempts at active production have succeeded to date due to ambiguity of the isomeric state, such as the lack of knowledge of the precise energy and lifetime. On the other hand, our method should enable ^{229}Th nuclei to be actively pumped up to the isomer state regardless of the uncertainties. The combination of processes 1 and 2 is usually called synchrotron radiation-based nuclear resonant scattering (NRS), which is widely used in materials science.

In this work [4], we carried out the NRS measurement at SPring-8 **BL09XU** and **BL19LXU**. A schematic view of the experiment is shown in Fig. 2. The beam operation mode was A-mode in which the 203 bunches are equally spaced with a time interval of 23.6 ns. The X-ray beam was monochromatized by two silicon monochromator pairs and the bandwidth of the beam was ~0.1 eV. It was focused by a compound refractive lens array and irradiated to the target of thorium oxide. A total of 0.24 μg ^{229}Th was deposited on a thin graphite plate with a diameter of 0.4 mm. At the downstream end of the beam, we placed an

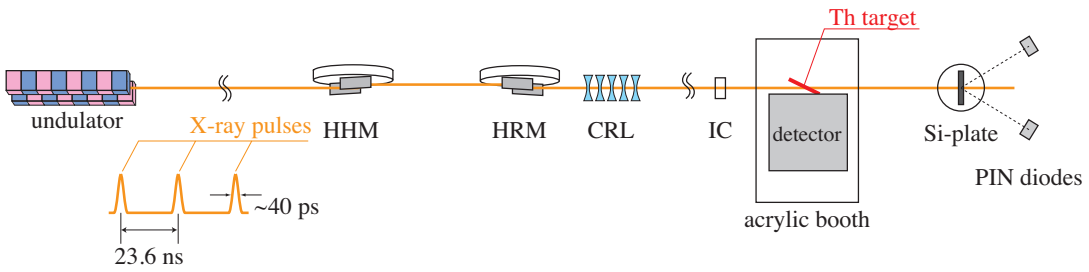

Fig. 2. Schematic of the experiment. The X-ray beam is generated at the undulator and propagates to the right side.

absolute X-ray beam energy monitor that can measure the beam energy with an accuracy of 0.07 eV.

In the NRS measurement, we scanned the X-ray beam energy and searched for the NRS peak. To confirm that NRS occurred, we detected the X-ray photons emitted via the transition in process 2 in Fig. 1. In comparison with the NRS signal rate, the background rate due to X-ray fluorescence following photoelectric absorption was 10^6–10^7 times higher; we developed a dedicated fast and energy-sensitive X-ray detection system to overcome the poor signal-to-noise ratio. This system consists of silicon avalanche photodiodes and peripheral fast circuits [5]. The energy and timing of each X-ray photon from the thorium target were measured simultaneously using this system. It also measured the pulse width of each analog pulse. The subsequent analysis based on these three parameters can increase the signal-to-noise ratio.

Figure 3 shows the NRS peak and the temporal profile of the NRS signal. Using the NRS data, we determined the accurate energy and the half-life of the second excited state to be 29,189.93 ± 0.07 eV and 82.2 ± 4.0 ps, respectively.

This work is the world's first realization of active pumping to the isomeric state. The production rate was ~25 k counts per second. This scheme provides a more efficient and cleaner method for isomer production than ^{233}U α-decay. This method also enables us to control the isomer production. It is expected to become a key technique for VUV photon detection.

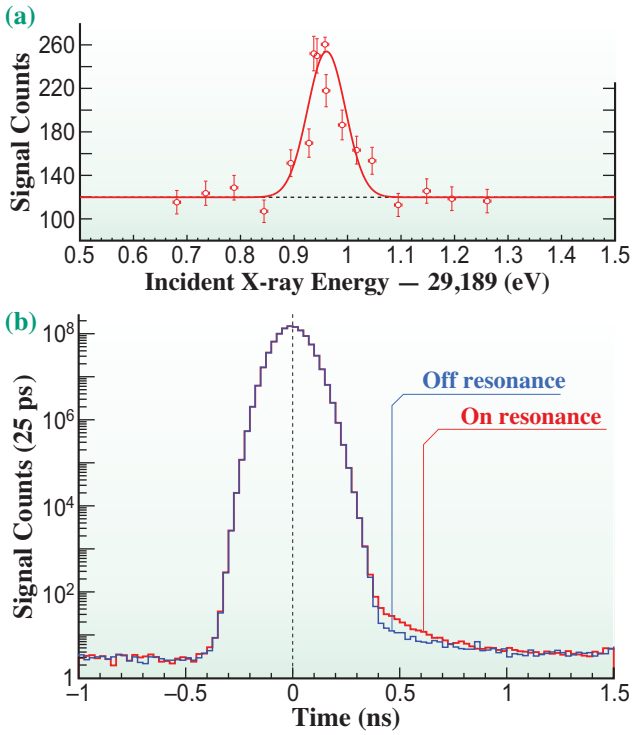

Fig. 3. **(a)** NRS peak and **(b)** temporal profile of the NRS signal.

T. Masuda*, T. Hiraki, A. Yoshimi and K. Yoshimura

Research Institute for Interdisciplinary Science, Okayama University

*Email: masuda@okayama-u.ac.jp

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