# Study on decay of actively pumped isomeric state of $^{229}$Th nuclei

Time is a common physical parameter used in various aspects of daily life. Precise measurements of the time and frequency can be achieved using atomic transitions. High-precision frequency standards are important because they have been used in many fields such as satellite navigation, telecommunications, metrology, and fundamental science. By monitoring the transition frequencies of precise clocks, researchers can explore physics beyond the Standard Model (BSM) of particle physics, including potential temporal variations in fundamental constants. Improvements in the precision of frequency standards enable the detection of subtle deviations from established physical models, thereby providing a powerful tool for exploring BSM.

A $^{229}$Th nuclear clock is a promising candidate for a new generation of high-precision frequency standards. The $^{229}$Th nucleus has the uniquely low first excited state ($^{229m}$Th) at approximately 8.36 eV. This energy corresponds to vacuum ultraviolet (VUV) light at 148 nm, a region where narrow-linewidth lasers and established atomic-physics techniques are available. In nuclear clocks, even if the frequency precision is comparable to that of atomic clocks, the sensitivity to temporal variation of the fine structure constant is expected to be a few orders of magnitude higher. Furthermore, nuclear clocks can be operated if $^{229}$Th is doped into VUV-transparent solid-state hosts, unlike atomic clocks, because the transition frequency is significantly less sensitive to external fields than the atomic transitions.

In recent years, studies on $^{229}$Th have progressed rapidly. In 2023, deexcitation light from $^{229m}$Th was first successfully observed at the ISOLDE facility at CERN [1]. Our group also successfully observed deexcitation light from $^{229m}$Th during beamtime in May and July of the same year [2], as described below.

Our work was conducted at SPring-8 **BL19LXU**. We used 29.19 keV X-rays to actively pump $^{229}$Th nuclei to the second excited state, where the branching fraction of this state to $^{229m}$Th exceeded 50% [3]. The irradiated target was $^{229}$Th-doped calcium fluoride crystals developed by researchers at TU Wien. Recently, a high $^{229}$Th concentration of more than $10^{15}$/mm$^3$ with a high transmittance in the VUV region was produced. To mitigate reduced crystal transmittance and the quenching of $^{229m}$Th induced by X-ray beam irradiation, three silicon monochromators (Si(111), Si(660), and Si(880)) were installed upstream of the beamline for X-ray energy monochromatization.

The experimental apparatus for detecting the radiative decay signals from $^{229m}$Th is described in Ref. [4]. The experimental setup was placed in a vacuum chamber to detect VUV photons. Background events primarily consisted of scintillation photons generated inside the crystals by the decay of $^{229}$Th or daughter nuclei (radioluminescence) and those induced by X-ray beam irradiation (XEOL). The total rates of radioluminescence and XEOL were significantly higher than those of radiative decay of $^{229m}$Th. To suppress these background events, we employed four dichroic mirrors and a solar-blind photomultiplier tube (PMT). An additional PMT was installed to detect radioluminescence photons and distinguish them from the signal events. Production of $^{229m}$Th can be controlled by changing the X-ray beam wavelength

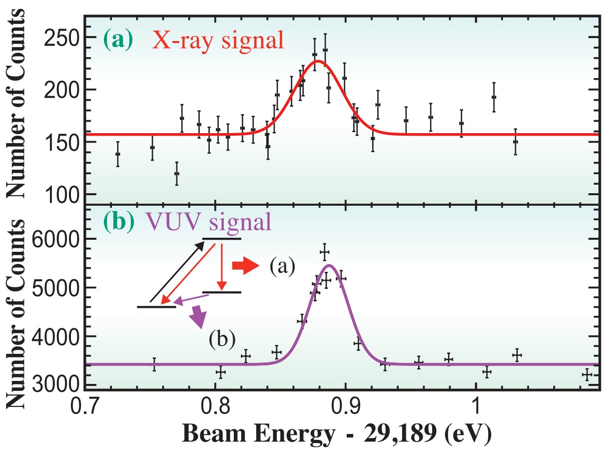


Fig. 1. Measured resonance spectra with changing X-ray beam energy. **(a)** Characteristic X-ray signals from the second excited state. **(b)** VUV photons from $^{229m}$Th.

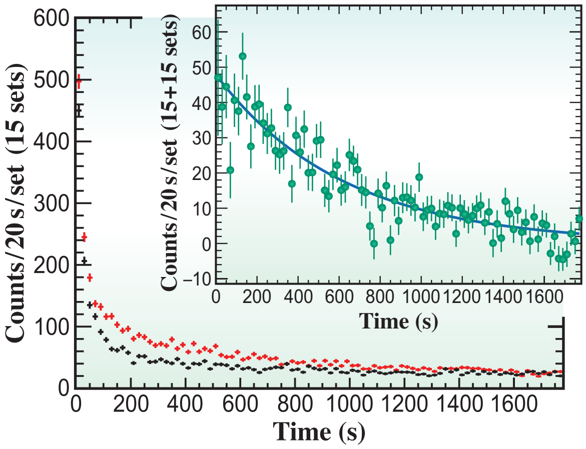


Fig. 2. Time spectra at the on-resonance condition (red) and off-resonance condition (black) averaged over 15 sets. Inset: the difference between these conditions.

near the resonant energy corresponding to the second excited state of the $^{229}$Th nuclei. The remaining luminescence events after background reduction were subtracted by comparing the data collected during the on- and off-resonance X-ray beam irradiations.

Figure 1 shows the measured resonance spectra of the nuclear resonance scattering signal (for details, see [3]) and VUV signal at the same beamtime. A peak was observed at almost the same X-ray beam energy, thereby confirming the successful observation of radiative decay photons from $^{229m}$Th.

The wavelength of the VUV signal was determined using by six VUV bandpass filters. A motorized wheel was placed in front of the PMT, and filters were inserted during this measurement. The transmittance spectra of each filter were measured in the laboratory. During this measurement, each band-pass filter was alternately inserted. By combining all the filter data, the VUV signal wavelength is determined to be 148.18 ± 0.38(stat.) ± 0.19(syst.) nm.

Figure 2 shows the temporal profile of the VUV events after stopping the X-ray beam irradiation under the on- and off-resonance conditions. By taking the difference in the spectra (Fig. 2, inset), we determined the lifetime of $^{229m}$Th by fitting this to an exponential function, $\tau$ = 646 ± 23 (stat.) ± 29 (syst.) s.

We also investigated the influence of varying the irradiation time on signal yield. The lifetime during beam irradiation (approximately 60 s) was significantly shorter than that measured after beam irradiation ($\tau$). This phenomenon is referred to as quenching of $^{229m}$Th. Subsequently, we measured the X-ray beam flux dependence of lifetime during beam irradiation. The X-ray beam flux was monitored using ionization chambers, and was changed by installing aluminum plates into the beamline or by uninstalling a Si(880) monochromator. The estimated lifetimes under different flux conditions are shown in Fig. 3. Lifetime during irradiation decreases as the beam flux increases.

In summary, we determined the lifetime of $^{229m}$Th and the wavelength of deexcitation photons. Furthermore, quenching was observed in the isomeric state. Based on a study by S. Kraemer *et al.* [1] and this study, the wavelength of a laser required to excite $^{229}$Th nuclei to the first excited state was determined with a precision of better than 0.5 nm. Finally, in 2024, the first laser excitation was achieved [5], which marked an important milestone in the realization of nuclear clocks.

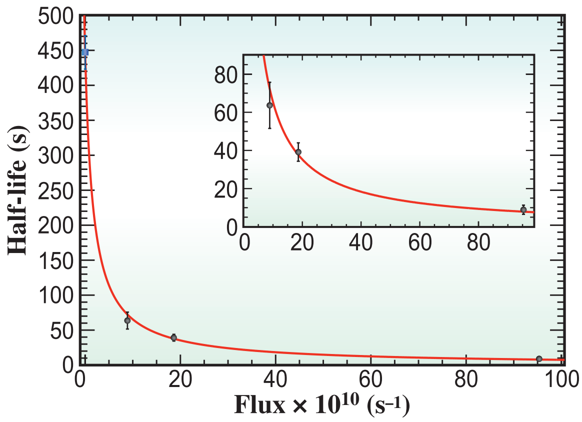


Fig. 3. X-ray beam flux dependence of lifetime during X-ray beam irradiation. The blue point represents lifetime measured after beam irradiation ($\tau$). The red curve represents the result of fitting by a function of $1/\tau' = 1/\tau$ + (const.) × flux, where $\tau'$ indicates lifetime during irradiation. Inset: enlarged view.

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