Controlling ^{229}Th isomeric state population in a VUV transparent crystal†

T. Hiraki,[*1] K. Okai,[*1] M. Bartokos,[*2] K. Beeks,[*2] H. Fujimoto,[*3] Y. Fukunaga,[*1] H. Haba,[*4] Y. Kasamatsu,[*5] S. Kitao,[*6] A. Leitner,[*2] T. Masuda,[*1] M. Guan,[*1] N. Nagasawa,[*7] R. Ogake,[*1] M. Pimon,[*2] M. Pressler,[*2] N. Sasao,[*1] F. Schaden,[*2] T. Schumm,[*2] M. Seto,[*6] Y. Shigekawa,[*4] K. Shimizu,[*1] T. Sikorsky,[*2] K. Tamasaku,[*8] S. Takatori,[*1] T. Watanabe,[*3] A. Yamaguchi,[*9] Y. Yoda,[*7] A. Yoshimi,[*1] and K. Yoshimura[*1]

^{229}Th has an extremely low nuclear isomeric state (^{229m}Th) at approximately 8.4 eV.[1)] The radiative lifetime of ^{229m}Th is of the order of 10^3 s.[2)] This state can be excited by a vacuum ultraviolet (VUV) laser[3)] and can be used as an excited state for clock transitions. In contrast to conventional atomic clocks, ^{229}Th nuclear clocks can be operated when ^{229}Th atoms are doped in VUV-transparent crystals with a wide bandgap host such as CaF_2. In this work, we excite ^{229}Th nuclei doped in CaF_2 to the isomeric state via the second excited state using a high brilliance X-ray beam. We successfully observed VUV photons from ^{229m}Th and discovered that the lifetime of ^{229m}Th during X-ray beam irradiation is significantly shorter than that after irradiation.

This experiment was conducted at the BL19LXU beamline in SPring-8. In the upstream of the beamline, three silicon monochromator sets were installed to monochromatize the X-ray beam to a full-width at half-maximum of approximately 30 meV. The X-ray beam energy was set to approximately 29190 eV, which is close to the nuclear second excited state of ^{229}Th. High concentration ($\mathcal{O}(10^{18})/\mathrm{cm}^3$) of ^{229}Th-doped CaF_2 crystals were prepared by TU Wien.[4)] VUV photons from the crystal target were focused by a parabolic mirror and a MgF_2 lens, and were measured by a solar-blind PMT (VUV PMT). Four dichroic mirrors were installed in the middle of the optical path to reduce background photons from the crystal. Another PMT (Veto PMT) was installed behind the first dichroic mirror that can significantly reduce radioluminescence background events by timing coincidence between the two PMTs.

By taking data under on- and off-resonance conditions near the second excited state, the decay curve of the radiative decay signal was obtained. From fitting the difference, the radiative half-life of ^{229m}Th was estimated to be $T = 447 \pm 16$ (stat.) ± 20 (syst.) s.

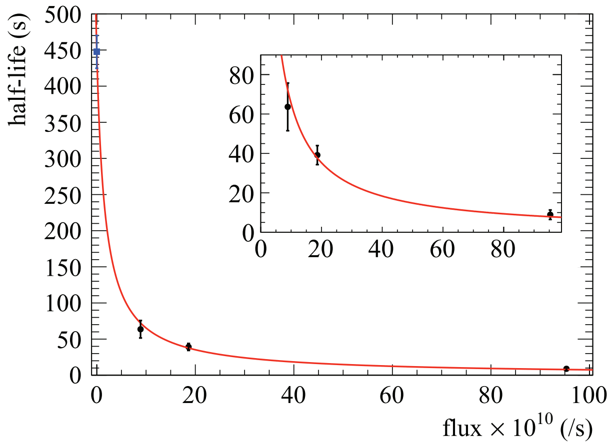

Fig. 1. X-ray beam flux dependence of half-life during X-ray beam irradiation. The blue point represents the half-life measured after beam irradiation.

Wavelength measurements of the VUV signal from ^{229m}Th were conducted by using optical bandpass filters in the VUV region. A total of six bandpass filters with different transmission spectra were prepared and mounted in a motorized wheel installed in front of the VUV PMT. The transmittance of the VUV signal was obtained by taking the ratio of the signal intensity. By comparing signal transmittance with the reference transmittance spectra obtained in our laboratory, the wavelength of the VUV signal was obtained to be 148.18 ± 0.38 (stat.) ± 0.19 (syst.) nm.

The half-life of ^{229m}Th during X-ray beam irradiation (T_{irr}) was estimated by measuring the dependence of the signal intensity on the irradiation time. Under the nominal condition, T_{irr} was estimated to be approximately ten times shorter than T. The dependence of T_{irr} on the X-ray beam intensity was measured, revealing that T_{irr} decreases under more intense beam irradiation, as shown in Fig. 1.

References

1) S. Kraemer *et al.*, Nature **617**, 706 (2023).
2) A. Yamaguchi *et al.*, Nature **629**, 62 (2024).
3) J. Tiedau *et al.*, Phys. Rev. Lett. **132**, 182501 (2024).
4) K. Beeks *et al.*, Phys. Rev. B **109**, 094111 (2024).

† Condensed from the article in Nat. Commun. **15**, 5536 (2024)

*1 Research Institute for Interdisciplinary Science, Okayama University
*2 Institute for Atomic and Subatomic Physics, TU Wien
*3 National Metrology Institute of Japan, National Institute of Advanced Industrial Science and Technology
*4 RIKEN Nishina Center
*5 Graduate School of Science, Osaka University
*6 Institute for Integrated Radiation and Nuclear Science, Kyoto University
*7 Precision Spectroscopy Division, Japan Synchrotron Radiation Research Institute
*8 SPring-8 Center, RIKEN
*9 Center for Advanced Photonics, RIKEN

DOI:10.34448/RIKEN.APR.58-258