Vacuum Tltraviolet Search from Thorium-229 Isomer in Crystal Toward Solid-State Nuclear Clock

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Abstract—Thorium-229 (^{229}Th) is a laser-excitable nucleus owing to its extremely low first excitation energy in the order of electron volts. Nuclear clocks, which utilize the resonance frequency of this unique nuclear isomer transition of ^{229}Th, are expected to demonstrate relatively higher accuracy compared to conventional atomic clocks. Particularly, solid-state nuclear clocks using ^{229}Th-doped crystals are expected to have a variety of applications for both fundamental physics and practical use in society as compact and precision clocks. However, laser excitation of ^{229}Th remains challenging due to the energy and lifetime uncertainty of the ^{229}Th isomer state. To address this challenge, we developed an original excitation method that does not attempt direct excitation to the first isometric state but instead excites to the second excitation state with transition properties and generates the isomer state through a de-excitation process from the second excitation state. We are currently applying this method to ^{229}Th-doped crystals to search for de-excitation vacuum-ultraviolet (VUV) light from the first isomeric state. This paper presents the current status of our search.

Keywords— Thorium-229, nuclear clock, solid-state clock

I. Introduction

The thorium isotope ^{229}Th is a laser-excitable nucleus with an extremely low first excitation state (isomer state) of approximately 8 eV [1–4]. Given that quantum control of nuclei is feasible, this nucleus is expected to emerge as the next-generation quantum frequency standard [5]. For example, it is expected to have applications in fundamental physics, such as the verification of the invariance of physical constants, and the variations of the strong coupling constants for dark matter searches [6,7]. However, laser excitation has not yet been achieved because of uncertainties in the energy and lifetime of the isomeric state, which are necessary to enable nuclear excitation through a laser.

Direct search experiments with de-excited vacuum-ultraviolet (VUV) light from the isomeric state are crucial for determining the energy with greater precision. Consequently, this project aimed to investigate the de-excitation process from the isomeric state to the ground state and to detect VUV light with high precision. In 2023, the VUV photon along with the isomeric transition was observed for the first time at the ISOLDE facility at CERN, in which the ^{229}Th isomer state was produced from the radioactive decay [8]. However, direct excitation from the ground state using synchrotron radiation or lasers has yet to be achieved. Therefore, we attempted to detect VUV light via X-ray excitation from the ground state via the second state of ^{229}Th.

The nuclear clock employs ^{229}Th captured in a multi-charged ionic state in ion traps or solids [9,10]. In this study, we employed calcium fluoride crystals doped with ^{229}Th (^{229}Th:CaF_2), which are candidates for solid-state nuclear clocks owing to their large band gap and high transmission in the VUV region (~ 50 %). We excited the ^{229}Th in the crystals to the second excitation state of known energy using X-rays from the ground state and then created isomeric states through the de-excitation process.

Herein, we present the detailed experimental setup to facilitate the search for VUV light from ^{229}Th isomeric state and the current status of our search.

II. METHODS

A. X-RAY PUMPING OF THE ^{229}TH NUCLEAR CLOCK ISOMER

To detect VUV light directly from the isomeric state of ^{229}Th, we have developed a novel excitation method that excites ^{229}Th using synchrotron X-ray irradiation to the second excitation level of a well-known energy level (approximately 29 keV), thereby producing the isomeric state of ^{229}Th via the de-excitation process shown in Fig. 1. Based on this method, excited ^{229}Th isomers can be confirmed by detecting the nuclear resonance scattering (NRS) signal [11] emitted by the de-excitation process from the second excited state. In 2019, we obtained the NRS signal and quantitatively determined the resonant energy, lifetime, and branching ratio of the second excited state of ^{229}Th with the highest precision [12]. In addition, by controlling the resonant and non-resonant synchrotron X-ray irradiation, we are able to search for the VUV light emitted when the isomeric state is de-excited to the ground state by subtracting the background with high precision.

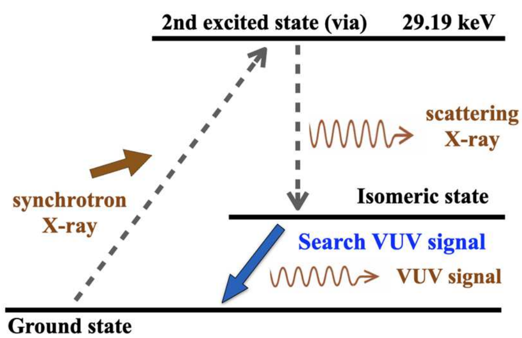

Fig. 1. Nuclear-level structure of ^{229}Th. Isomeric ^{229}Th is populated by X-ray pumping into the second excited state. The decay energy, lifetime, and branching ratio were measured using X-ray pumping method [12].

B. SETUP FOR VUV SEARCH FOR ^{229}TH ISOMER

Since 2020, the aforementioned active-pumping method has been applied to $^{229}Th:CaF_2$ crystals to search for de-excited VUV signals. The experiments were performed at the BL19LXU beamline [13] of SPring-8, a synchrotron radiation facility in Japan. A schematic of the experimental setup is shown in Fig. 2.

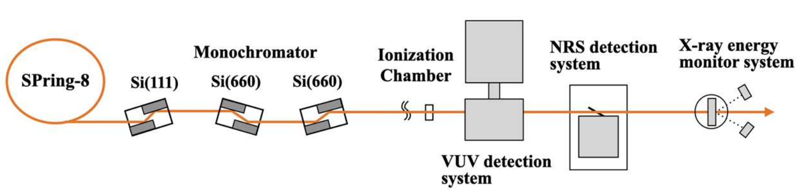

Fig. 2. A schematic of the experimental setup of the VUV search.

A high-brightness X-ray beam is monochromatized with Si(111) and two Si(660) crystals to approximately 29 keV in energy and 50 meV at the full width at half maximum (FWHM). The X-ray flux is monitored using an ionization chamber. The target crystal ($^{229}Th:CaF_2$) is installed in the VUV detection system and placed upstream of the X-ray energy monitoring system. The absolute X-ray irradiation energy of the second excited state of the ^{229}Th nucleus is confirmed by detecting the NRS signal, and the experiment is performed under on- and off-resonance conditions. In the NRS detection system, a high-density solid ^{229}Th target (1.8 kBq; approximate diameter: 0.7 mm) fabricated through the dry-up method is placed in the focusing position of the incident X-ray beam. After irradiation, the fluorescent X-rays emitted by the solid ^{229}Th target were detected using a 9-channel silicon avalanche photodiode (Si-APD) sensor [14]. An X-ray energy monitoring system [15] installed downstream monitored the incident beam energy with an accuracy of better than 0.1 eV.

C. VUV DETECTION SYSTEM

The VUV detection system consists of an irradiation chamber and a measurement chamber as shown in Fig. 3 The ^{229}Th crystal target was attached to a target holder. The crystal target was irradiated with X-rays to produce ^{229}Th isomers. The target holder was moved from the irradiation chamber to the measurement chamber using a pneumatic moving stage.

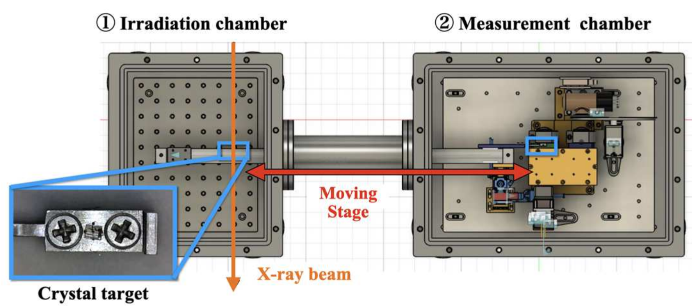

Fig. 3. Overview of the experimental setup. The VUV detection system consisted of an irradiation chamber and a measurement chamber. After irradiation, the crystal target was moved from the irradiation chamber to the measurement chamber using a pneumatic moving stage.

A photograph of the measurement chamber is shown in Fig. 4.

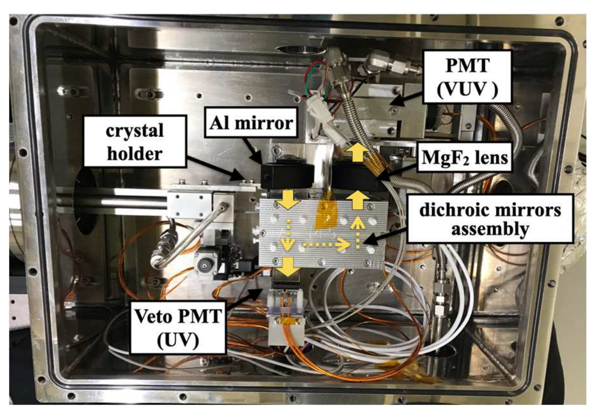

Fig. 4. Optical system overview of the measurement chamber. Light from the crystal is focused using an aluminum-coated mirror placed in front of the crystal holder. The focused light is reflected using a dichroic mirror assembly, refocused by an MgF_2 lens, and directed toward a VUV PMT.

First, light from the crystal target is focused using a 40-mm diameter aluminum-coated spherical mirror (OptoSigma) placed in front of the target holder. The reflectance of the mirror in the VUV region is approximately 80 %.

The focused light is then reflected using dichroic mirrors. The dichroic mirrors consist of four coated prisms, and the prism (a UV-fused silica right-angle prism, PS612, Thorlabs) features a coating applied by OptoSigma that reflects a specific wavelength region, thereby functioning as a VUV band-pass filter. Light from the dichroic mirror is refocused by a 40-mm-diameter MgF2 lens (Pier Optics) and directed into the PMT (R10454, Hamamatsu). The quantum efficiency of around 150 nm of the VUV PMT is approximately 20%. To reduce the dark current background, the VUV PMT is cooled to −30 ℃ using a Peltier temperature controller (TECSource 5305, Arroyo Instruments). A veto PMT (R11265-203, Hamamatsu) is also mounted to reduce the radiation-induced background, mainly caused by the radioactive decay of ^{229}Th. The veto PMT is sensitive to the UV region and detects both UV and visible light transmitted through the prism. By synchronizing the detection timing of the VUV PMT, we could reject radioluminescent background events.

The preliminary total detection efficiency of the experimental setup is shown in Fig. 5. The total detection efficiency includes the ideal crystal transmittance, the geometric efficiency, the reflectance of the spherical mirror, the reflectance of the four dichroic mirrors, the transmittance of the MgF_2 lens, and the quantum efficiency of the PMT. The optical reflectance is measured using a deuterium lamp (D200F-HV, Heraeus) and a spectrometer (VMK200, Vacuum & Optical Instruments). The geometric efficiency is estimated using an optical simulation. The total detection efficiency covers a wavelength region of 148.71 nm, as reported in the ISOLDE experiment [8].

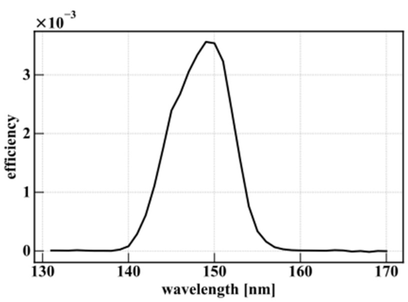

Fig. 5. Preliminary total detection efficiency of experimental setup.

D. Crystal Target

The calcium fluoride crystals doped with ^{229}Th (^{229}Th:CaF_2) are used as targets in our experiments. The crystal target developed by the Wien group [16] is shown in Fig. 6. The size of the crystal was approximately 1 mm cubic, and its density was approximately 10^{15} /mm^3.

Characterization studies were performed using X-ray absorption fine structure spectroscopy (XAFS) methods [17] on another ^{229}Th:CaF_2 crystal, which was produced in the same technique as the crystal target. Based on the absorption edge peak spectrum, the valence state of ^{229}Th in the crystal is determined to be 4+. This spectrum is in good agreement with the structural model of a single crystal of calcium fluoride substituted from Ca ions to Th ions, thereby validating that it was doped at the expected position (Ca^{2+}).

Fig. 6. (Left) Crystal target of ^{229}Th:CaF_2. The size of the crystals is approximately 1 mm cubic. (Right) crystal holder. The crystal was attached to a target holder using a stainless steel fixing wire.

E. Experimental procedure for background reduction

In VUV-search experiments, on-resonance and off-resonance data are alternately acquired under the same experimental conditions to subtract the background signal. Fig. 7 shows the experimental scheme. X-ray irradiation is controlled for on-resonance and off-resonance conditions using the nuclear resonant scattering (NRS) detection system [12] and the X-ray energy monitor [15]. First, the crystal target is irradiated in the irradiation chamber for 600 s under the on-resonance condition, generating the isomeric state of ^{229}Th. Thereafter, the crystal target attached to the stage is moved to the measurement chamber in about 1 s, and the data are acquired for 1800 s. The crystal is then irradiated in the irradiation chamber again under the off-resonance condition, and data is acquired in the same manner. The VUV PMT and Veto PMT signals were acquired with an oscilloscope (PXIe-5162, National Instruments).

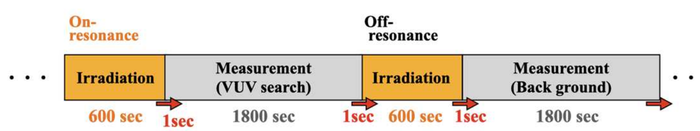

Fig. 7. Example of experimental procedure for one dataset. On-resonance and off-resonance data were alternately obtained for background reduction.

III. Results and Discussion

After background removal, preliminary results from subtracting off-resonance set data from on-resonance set data implied a difference. A detailed analysis of the isomeric parameters is currently in progress.

Upon scanning the X-ray energy, an excess of VUV signal over the background was indicated only at energies consistent with the resonance energy of the second excited state. We plan to improve the beam energy resolution by improving the monochromator.

IV. Conclusions

^{229}Th is a laser-excitable nucleus owing to its extremely low first excitation energy. For the laser excitation of the ^{229}Th isomer, the nuclear clock transition energy and lifetime are being actively investigated. Direct search experiments are important for precisely determining the energy. We searched for a de-excited VUV signal in the first isometric state of ^{229}Th directly. Moreover, we developed an original excitation method that does not attempt direct excitation from the ground state to the first isometric state but instead excites via a de-excitation process from the second excited state. We are currently applying this method to a ^{229}Th-doped crystal in the search for VUV light. Notably, both the comparison of on-resonance and off-resonance datasets and the X-ray beam energy scanning have indicated an excess of VUV signal.

We are working to achieve precise observations by improving the sensitivity of the measurements to determine isomer parameters such as wavelength and lifetime.

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