Development of a photon measurement apparatus for observing the radiative decay of ^{229m}Th produced from ^{229}Pa

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The first excited state in the ^{229}Th nucleus (^{229m}Th) has an excitation energy of ∼8.3 eV (150 nm),[1] which potentially enables an ultraprecise nuclear clock. We have been aiming to observe the radiative decay (γ rays) of ^{229m}Th and to determine its radiative half-life, which is an essential parameter to develop the nuclear clock, by doping a CaF_2 crystal with ^{229}Pa, which decays to ^{229m}Th with negligibly small recoil energy.[2] In this study, we developed a photon measurement apparatus for observing the γ rays of ^{229m}Th. We also investigated the background photons originating from the decay of ^{229}Pa ($T_{1/2}$ = 1.5 d) and other impurities such as ^{232}Pa ($T_{1/2}$ = 1.31 d) and ^{230}Pa ($T_{1/2}$ = 17.4 d).

Figure 1 shows a schematic view of the developed apparatus. Two photomultipliers are placed inside a vacuum chamber: one is for measuring the γ rays of ^{229m}Th in the vacuum ultraviolet (VUV) range (PMT1, Hamamatsu R10454), and the other is for measuring scintillation photons produced from ^{229}Pa and other isotopes in a CaF_2 crystal (PMT2, Hamamatsu R7154). The events of PMT1 that coincided with those of PMT2 correspond to high-energy radiation that can produce scintillation photons, and thus, such events can be excluded from the analysis of the γ-ray events of ^{229m}Th. Band-pass (BP) filters for photons of 151 ± 20 and 171 ± 20 nm (eSource Optics) are placed between PMT1 and the CaF_2 sample. The filters can be switched using a linear drive driven by a stepper motor; the γ rays of ^{229m}Th are expected to be detected only for the 151-nm BP filter. PMT1 and PMT2 can be cooled to −25°C using a Peltier cooler, reducing the dark count rate to 0.09 s^{-1} for PMT1 and 0.25 s^{-1} for PMT2. Considering the half-life of ^{229m}Th (10^3–10^4 s), the ^{229}Pa-doped CaF_2 can be rapidly introduced to the measurement position (∼10 min) as follows without the leakage of the large vacuum chamber (Fig. 1). First, the sample fixed on a linear drive is

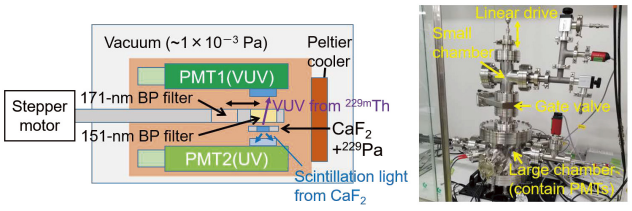

Fig. 1. Schematic view of the developed apparatus (left) and a photograph of the sample loading system (right).

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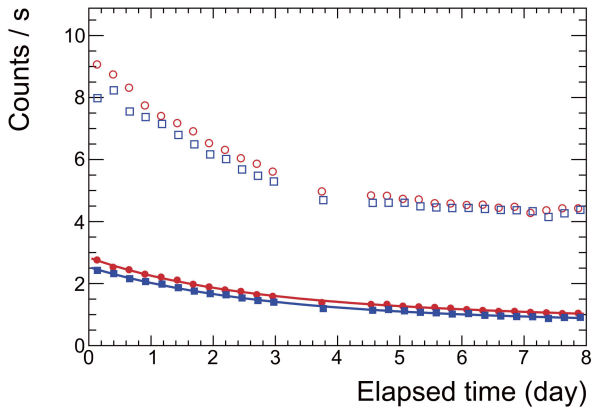

Fig. 2. Count rate of photons measured using PMT1 for the 151-nm (circle) and 171-nm (square) BP filters with (closed) and without (open) anticoincidence using the events measured by PMT2. The sum of the exponential decay functions of ^{229}Pa and ^{230}Pa is fitted to the count rate with anticoincidence for each filter (solid lines).

placed inside a small vacuum chamber, which is then rapidly evacuated. Next, a gate valve is opened, and the sample is moved to the large chamber for photon measurement.

We investigated the background photons originating from ^{229}Pa and other isotopes, which may interfere with the observation of the γ rays of ^{229m}Th. The production and chemical separation of ^{229}Pa were performed similarly to those in a previous study.[2] First, two ^{232}Th metallic foils (total 138 mg/cm^2) were irradiated with 1 μA of a 30-MeV proton beam for 10 h at the RIKEN AVF cyclotron. Next, the foils were dissolved with concentrated HCl and fed onto an anion-exchange column (Muromac 1X8, 100–200 mesh, ∼1.0 mL). After pouring concentrated HCl, 6 M HCl, and 8 M HNO_3 to the column, Pa isotopes were eluted with 9 M HCl/0.1 M HF. In this study, we performed an additional anion-exchange process to reduce radioactive impurities such as ^{97}Zr as follows. First, Pa isotopes were dissolved in 0.1 M HCl/0.1 M HF and fed onto an anion-exchange column (Muromac 1X8, 100–200 mesh, ∼0.5 mL). After pouring 0.1 M HCl/0.1 M HF, Pa isotopes were eluted with 0.4 M HCl/0.1 M HF. The ratio of radioactivity of ^{229}Pa isotopes to that of other radioactive elements was ∼10^5 after the chemical separation. Thereafter, we dropped the ^{229}Pa solution on a CaF_2 crystal, annealed it, and started a photon measurement four days after the proton irradiation. Compared with the previous measurement,[2]

the amount of ^{229}Pa was dominant over other isotopes (^{229}Pa 48(3) kBq, ^{232}Pa 2.51(6) kBq, and ^{230}Pa 6.6(4) kBq); thus, we could evaluate the background photons produced by high-energy radiation from ^{229}Pa more precisely.

As shown in Fig. 2, the count rate of photons detected by PMT1 for the 151-nm BP filter was $\sim$9 s^{-1} at the start of the measurement. Anticoincidence using PMT2 reduced the count rate to $\sim$1/3. For each BP filter, the sum of the exponential decay functions of ^{229}Pa and ^{230}Pa was well fitted to the data, indicating that the photons originate from ^{229}Pa and ^{230}Pa. The ratio of photons from ^{229}Pa to those from ^{230}Pa was $\sim$1. In the presence of these background photons, if we use a CaF_2 sample doped with 100 kBq of ^{229}Pa, our simulation indicated that we can observe the γ rays of ^{229m}Th without the mass separation of ^{229}Pa and determine its half-life with a relative error of $\sim$25%; the development of the apparatus for measuring the γ rays of ^{229m}Th is nearly complete.

References

1) B. Seiferle *et al.*, Nature **573**, 243 (2019).
2) Y. Shigekawa *et al.*, RIKEN Accel. Prog. Rep. **54**, 143 (2021).