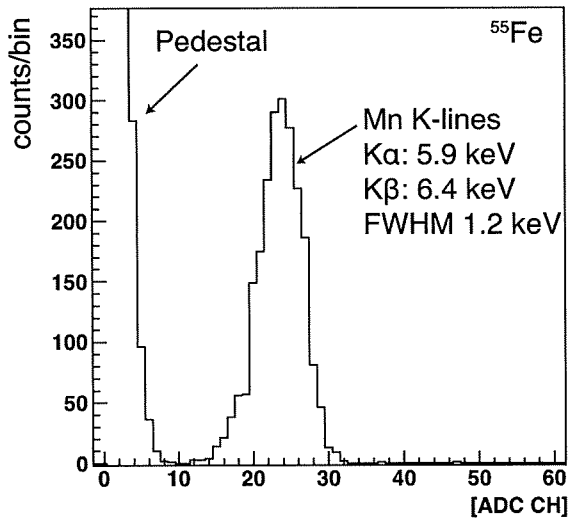

Fig. 10. ^{55}Fe X-ray spectrum obtained with the Al-pad/CdTe/Pt detector using VA32TA6. At operating temperature of −20°C, 400 V of bias voltage was applied. The pedestal level was corrected, and the common mode was subtracted by using the data from the Common Mode Detection Unit.

spectral performance. Gamma-ray imaging spectroscopy with a position resolution of 400 μm was demonstrated by using the 2.6 cm CdTe DSD. The energy resolution of 1.8 keV (FWHM) at 59.54 keV was obtained with the 1.3 cm CdTe DSD.

For semiconductor detector readout, we have developed a new analog ASIC, VA32TA6, characterized by a main new feature of including an on-chip ADC. By constructing CdTe pad detectors, we tested the functions and performance of VA32TA6. The ADC worked properly and good noise performance was obtained.

We are developing the next version of the CdTe DSD, that will use VA32TA6s as the readout ASICs. By using VA32TA6s, the readout system may be simplified, because it is possible to control the ASICs and read data by only using digital signals. Such a new configuration would offer improvements in performance as required for the the ASTRO-H (NeXT) Hard X-ray Imager and future X-ray/gamma-ray telescopes.

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Experimental Results of the Gamma-Ray Imaging Capability With a Si/CdTe Semiconductor Compton Camera

Shin'ichiro Takeda, Hiroyuki Aono, Sho Okuyama, Shin-nosuke Ishikawa, Hirokazu Odaka, Shin Watanabe, Motohide Kokubun, Tadayuki Takahashi, Kazuhiro Nakazawa, Hiroyasu TajimaHiro, and Naoki Kawachi

***Abstract*—A semiconductor Compton camera that combines silicon (Si) and Cadmium Telluride (CdTe) detectors was developed, and its imaging capability was examined with various kinds of gamma-ray targets such as a point source, arranged point sources and an extended source. The camera consists of one double-sided Si strip detector and four layers of CdTe pad detectors, and was designed to minimize the distance between a scatterer and the target. This is because the spatial resolution with Compton imaging improves as the target approaches the scatterer. This new camera realizes a minimum distance of 25 mm. By placing the target at a distance of 30 mm from the detector, resolving power better than 3 mm was demonstrated experimentally for a 364 keV (^{131}I) gamma-ray. Positional determination with accuracy of 1 mm was also demonstrated. As a deconvolution method, we selected the iteration algorithm (called List-Mode Expectation-Maximizing Maximum Likelihood), and applied it to several kinds of experimental data. The Compton back projection images of the arranged point sources and an extended object were successfully deconvolved.**

***Index Terms*—CdTe detector, Compton camera, gamma-ray imaging, silicon strip detector.**

I. Introduction

COMPTON imaging is an attractive technology used in gamma-ray detection for various applications. In high energy astrophysics, high-sensitivity gamma-ray observation is required for studying nucleosynthesis and particle acceleration, the phenomena of which are widely observed throughout the vast universe. The technology is also applicable to nuclear medical imaging and/or non-destructive inspection [1], [2], since it can directly locate the position of gamma-ray signals with a wide field of view, as well as a wide energy band.

We have developed a Compton camera—the Si/CdTe Compton camera [3]–[6]—based on the combination of Silicon (Si) and Cadmium Telluride (CdTe) semiconductor detectors. It consists of many layers of thin Si and CdTe position-sensitive detectors. The design leads to a high-angular resolution thanks to the high-energy and position resolution of these semiconductor detectors, including dedicated low-noise analog ASICs. Si is a suitable scatterer material, given its smaller Doppler broadening effect than that of any other semiconductor devices [7]. This effect is caused by the non-zero momenta of electrons, and imposes a theoretical limit on angular resolution. CdTe works very nicely as an absorber thanks to its large atomic numbers (48, 52) and high density (5.8 g/cm^3).

In our previous work, we developed a prototype Compton camera that was primarily designed for future balloon-borne astrophysics experiments [8], and verified its performance. The Compton reconstruction was successfully performed and the gamma-ray images of point sources were obtained from 662 keV down to 59.5 keV. The typical resolution of scattering angle (the Angular Resolution Measure or ARM) was 3.5° Full-Width-at-Half-Maximum (FWHM) and 2.5° (FWHM) at 356 keV and 511 keV, respectively.

A Compton camera with an angular resolution of a few degrees is an attractive detector applicable not only to astrophysics, but also to nuclear medical imaging. Such applications require spatial resolution at the mm level. In this paper, we report the experimental results of Compton imaging using a new detector designed for high spatial resolution in short-distance imaging. Fig. 1 shows a photo of the detector system. Section II describes our approach to obtaining high spatial resolution. Section III describes the key detectors mounted on the Compton camera and the camera's design. Section IV describes the procedure of Compton imaging, and Section V summarizes the imaging results.

II. Design Approach to High Spatial Resolution

In order to achieve the high spatial resolution of a short-distance target by using Compton imaging, designing the arrangement of detectors requires special care. Fig. 2 shows the schematic of a Compton camera consisting of two scatterers and one absorber. A gamma-ray photon emitted from a target is detected through two major scenarios: (a) scattered in the 1st scatterer or (b) in the 2nd scatterer, and then absorbed in the absorber

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S. Takeda, H. Aono, S. Ishikawa, H. Odaka, S. Watanabe, M. Kokubun and T. Takahashi are members of the Institute of Space and Astronautical Science, Japan Aerospace Exploration Agency, Kanagawa 229-8510, Japan, and also the Department of Physics, University of Tokyo, Tokyo 113-0033, Japan (e-mail: takeda@astro.isas.jaxa.jp; aono@astro.isas.jaxa.jp; ishikawa@astro.isas.jaxa.jp; odaka@astro.isas.jaxa.jp; watanabe@astro.isas.jaxa.jp; kokubun@astro.isas.jaxa.jp; takahasi@astro.isas.jaxa.jp).

S. Okuyama and K. Nakazawa are with the Department of Physics, University of Tokyo, Tokyo 113-0033, Japan (e-mail: okuyama@amalthea.phys.s.u-tokyo.ac.jp; nakazawa@amalthea.phys.s.u-tokyo.ac.jp).

H. Tajima is with the Stanford Linear Accelerator Center, Menlo Park, CA 94025 USA (e-mail: htajima@slac.stanford.edu).

N. Kawachi is with the Quantum Beam Science Directorate, Japan Atomic Energy Agency, Gunma 370-1292, Japan (e-mail: kawachi.naoki@jaea.go.jp).

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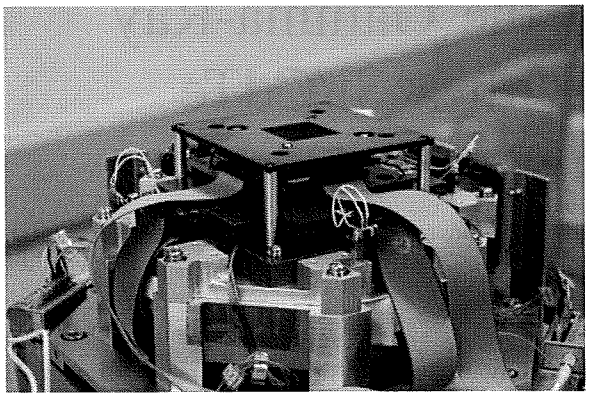

Fig. 1. Photograph of the Compton camera.

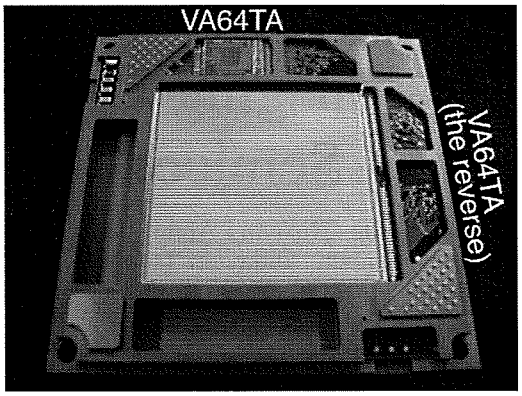

Fig. 3. Photograph of a DSSD board.

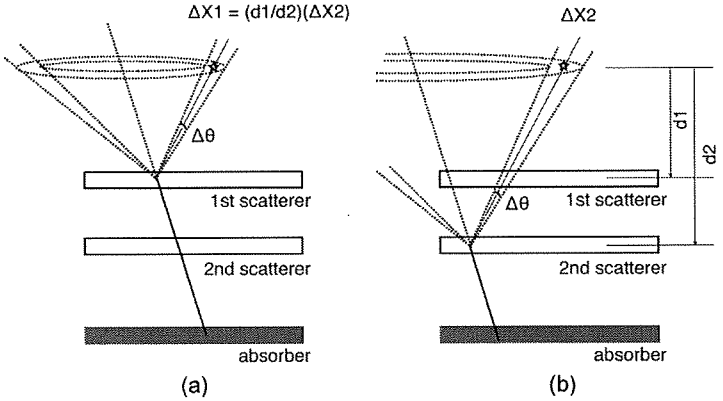

Fig. 2. Schematic of a Compton camera consisting of two scatterers and one absorber. Case (a) has a factor of (d1/d2) better spatial resolution, as derived from the angular resolution multiplied by the distance.

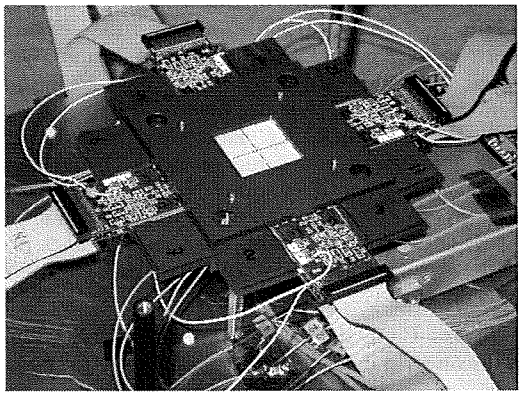

Fig. 4. Photograph of a 4-layer stacked CdTe module.

detector. The direction of incident photons is obtained as a cone in the sky (Compton cone) based on information about the interaction positions and related energy deposits in both the scatterer and absorber. By simply assuming that cases (a) and (b) have the same angular resolution, case (a) has a factor of (d1/d2) better spatial resolution, as derived from the angular resolution multiplied by the distance. Generally speaking, the spatial resolution in Compton imaging gradually deteriorates as the distance increases between the target and scatterer.

In our previous Compton camera [8], we used a Si scatterer consisting of four layers of a double-sided silicon strip detector (DSSD described in Section III-A) in order to improve detection efficiency [8], [9]. For astrophysical imaging, only the angular resolution and detection efficiency are essential since the targets are located virtually indefinitely in the far distance. Conversely, in a short-distance application, the 2nd to 4th layer of the scatterer degrades the spatial resolution, given the increasing distance to the target. Therefore, we employed one DSSD layer for the scatterer in the Compton camera described in this paper. It is also important to minimize the distance between 1st scatterer and a target. When considering the few degrees of angular resolution, the interval must be limited within a few tens of mm to obtain mm-scale spatial resolution. In our case, a minimum distance of 25 mm to a target was achieved.

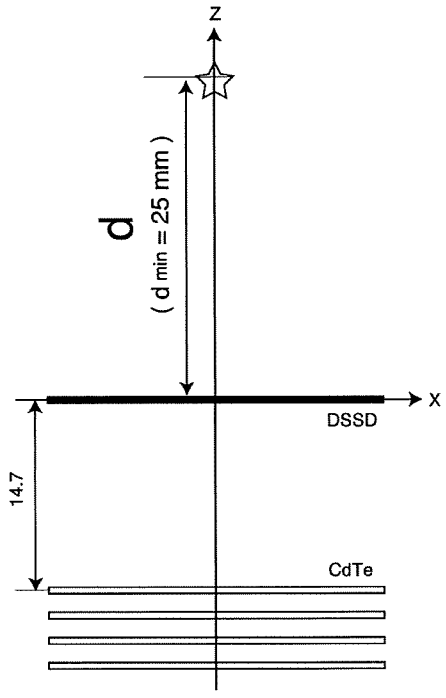

Fig. 5. Cross-sectional view of the Compton camera.

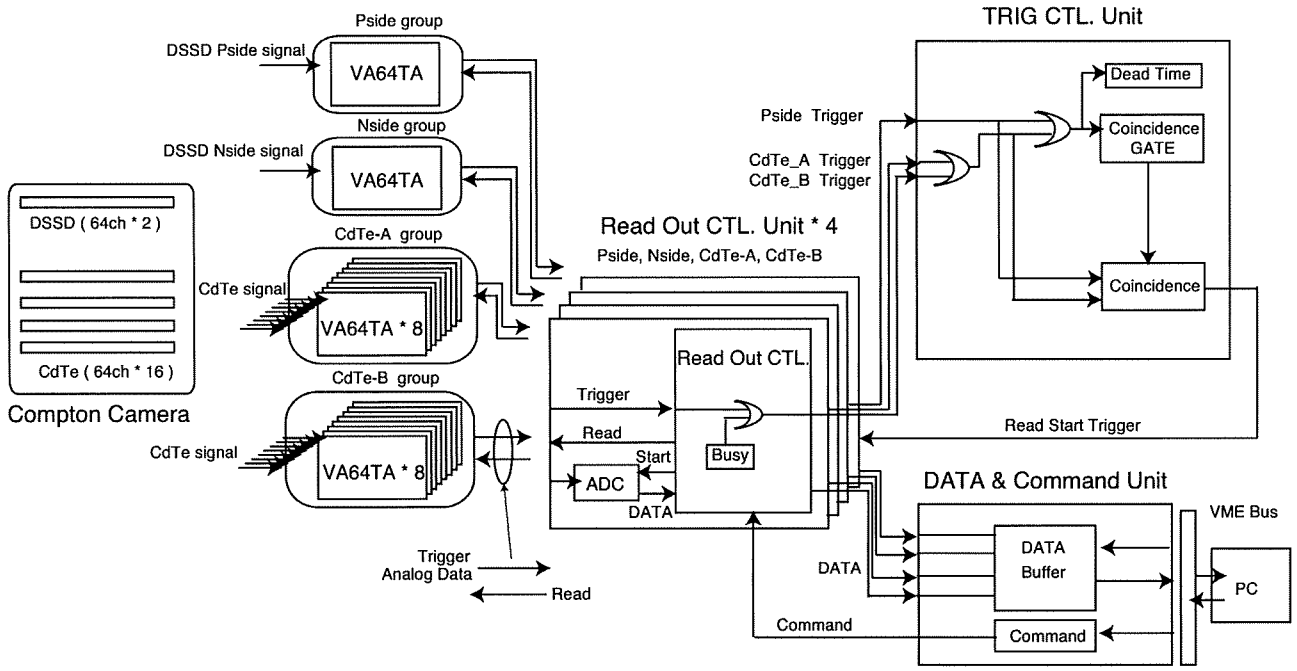

Fig. 6. Schematic diagram of the readout system.

III. Compton Camera Setup

A. Double-Sided Silicon Strip Detector (DSSD)

As a scattering part of the Compton camera, we developed a low-noise double-sided silicon strip detector (DSSD) [9]–[12]. Fig. 3 shows a photograph of the DSSD board. It consists of one DSSD and two low-noise, low-power consumption analog VLSIs (VA64TA1s [13] developed in collaboration with Gamma-Medica Ideas) and mounted on a small circuit board. The DSSD used here is a 2.56-cm wide and 500-μm thick device developed in conjunction with Hamamatsu Photonics, Japan. The strip pitch is 400 μm with a strip gap of 100 μm on each side. The N-side has a floating p-implantation between the strips to isolate the adjacent strips. The Al electrodes are directly coupled on each strip with ohmic contact.

In order to extend the imaging band down to low-energy gamma-rays, low-noise read-out to achieve a lower detection threshold is needed on both the P-side and N-side strips of the DSSD. This is because we need both the P-side and N-side signals to obtain two-dimensional positional information. Thus, a DC-coupled readout is employed for both sides. To apply a reverse bias, the N-side circuit board is biased as a whole. The applied bias voltage is 250 V. The decoupling is held on the digital interface after the analog signal is digitalized by the ADC. The energy resolution at 59.5 keV is found to be 1.6 keV (FWHM) for the P-side and 2.8 keV for the N-side. From the noise level measured on the N-side, we calculated the possibility of incorrectly identifying the position due to random noise, and found that it is less than 10% for a 5 keV energy deposit for this device.

B. 4-Layer Stacked CdTe Module

The absorber consists of a total 16 CdTe pad detectors based on the technology of a high-resolution CdTe Schottky diode [14]–[16]. The production and pixelation of CdTe were performed by ACRORAD, Japan. Each detector is 13.2-mm wide and 500-μm thick. In the CdTe detector, a common electrode made of Indium (In) is used as the anode, and pixelated electrodes divided into 64 pads made of Platinum (Pt) are used as the cathode. The size of each pad of the cathode electrode is 1.35 mm^2, with a gap of 50 μm between the electrodes. A guard-ring electrode surrounds the pads. Due to a high Schottky barrier and the guard-ring electrode, leakage current as low as 10 pA per pad is obtained under bias voltage of 700 V. Good energy resolution is realized thanks to the higher efficiency of charge collection.

Fig. 4 shows the 4-layer stacked CdTe module with a stack pitch of 2 mm [16]. One layer consists of four CdTe pad detectors configured in 2 × 2 geometry and two FECs (Front End Cards) on which four VA64TA1s are mounted. Energy resolution of 2.0 keV (FWHM) is achieved at 81 keV and that of $\Delta E/E \sim 1\%$ (FWHM) is at 511 keV under temperature of $-20°C$, and bias voltage of 600 V. Under these conditions, no significant degradation in spectrum performance was observed during continuous operation of up to two weeks.

C. Setup of the Compton Camera

We constructed the new Compton camera (shown in Fig. 1) by combining the DSSD and 4-layer stacked CdTe module. Fig. 5 shows a cross-sectional view of the camera. The 4-layer stacked CdTe module is arranged 14.7 mm underneath the DSSD. The minimum distance to the target (d_{min}) is 25 mm due to the entrance window provided for thermal decoupling.

Fig. 6 shows a schematic diagram of the readout system. The readout VA64TA1s are divided into four groups: DSSD Pside, DSSD Nside, CdTe-A, and CdTe-B. In the CdTe-A and CdTe-B groups, the eight VA64TA1s are arranged in a daisy chain configuration. The readout sequence of each group is controlled by

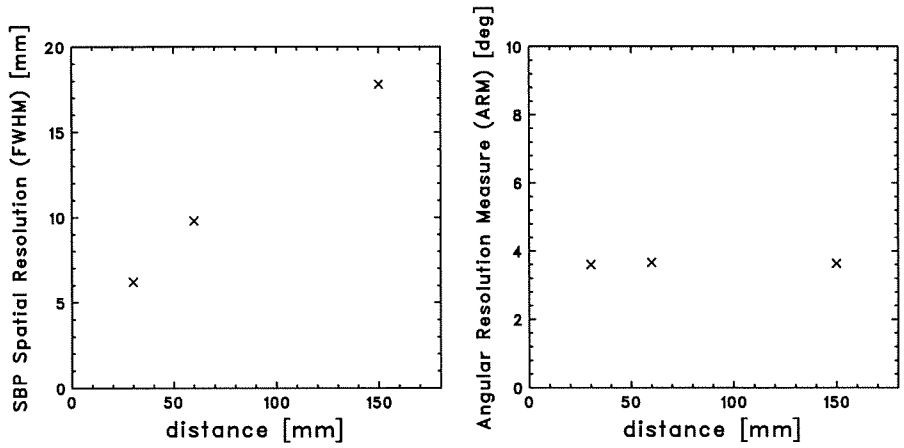

Fig. 7. Left panel showing the distance dependence of spatial resolution of the SBP image; right panel showing that of angular resolution defined by FWHM of the ARM distribution for a 356 keV gamma-ray.

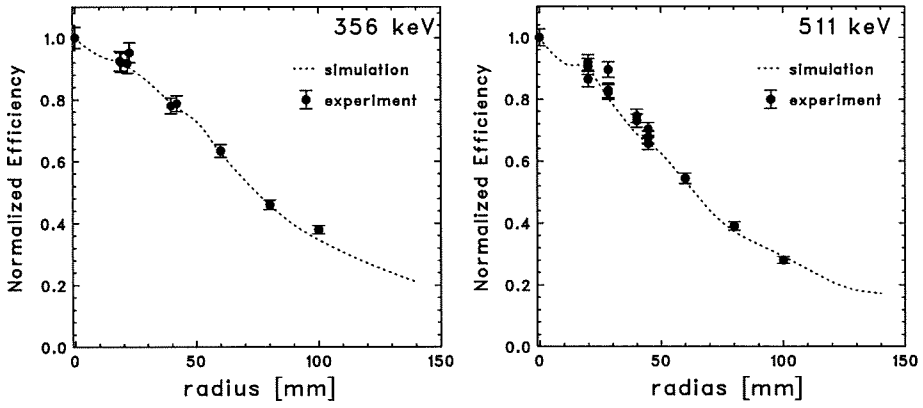

Fig. 8. The position dependence of detection efficiency for 356 keV and 511 keV at d = 60 mm plane. The efficiency at the center position (x, y, z) = (0, 0, 60) in Fig. 5 is set to 1. The radius denotes the distance from (x, y) = (0, 0). The radius 100 mm corresponds to a 120° field of view.

a dedicated Read Out CTL Unit (ROU). The ROU starts the readout sequence by using an external read start trigger. The event trigger from the Compton camera is fed to the TRIG CTL Unit (TCU) that consists of NIM modules in this version. In the TCU, a coincidence judgment is made between the trigger of DSSD and that of CdTe for the effective acquisition of data on Compton events. The coincidence judgment output is then returned to the ROU as a read start trigger. Since this trigger is sent to all ROUs, the pulse heights of all channels are acquired for a coincidence event. The pulse heights converted into digital data in the ROU are then sent to DATA&Command Unit controlled by a PC via the VME bus.

IV. Procedure for Compton Imaging

Data reduction and image reconstruction are performed for Compton imaging. First we select "two hit events", meaning one hit in a DSSD and one in a CdTe detector. Here, the energy threshold to determine a hit is set to 10 keV for both the DSSD and CdTe detectors. Obtaining the Compton events of incident gamma-rays entails the selection of "two hit events" as follows:

- The sum of energy deposits in Si (E_{dssd}) and CdTe (E_{cdte}) is equal to the incident gamma-ray energy (E_{in}), that is, $E_{dssd}+E_{cdte} = E_{in}$. By considering the energy resolution of the detectors, the events where $E_{dssd}+E_{cdte}$ equal $E_{in}\pm$ 5 keV are selected.
- The events where E_{dssd} is from 20 keV to 35 keV are excluded. This region is contaminated by fluorescence X-ray events, which are X-rays from Cd (K_α: 23.1 keV, K_β: 26.1 keV) and Te (K_α: 27.4 keV, K_β: 31.0 keV) escaping from the CdTe detectors and then absorbed in the DSSD.

With these selected events, the Simple Back Projection (SBP) image was obtained. We assumed that incident gamma-rays are scattered in the DSSD and absorbed in the CdTe detector. Based on this assumption, the direction of incident photons is calculated as;

$$\cos\theta = 1 - m_e c^2 \left(\frac{1}{E_{cdte}} - \frac{1}{E_{dssd} + E_{cdte}} \right). \quad (1)$$

The back projection of the Compton cone into the RI source plane is performed event by event. The SBP image is the simple accumulation of Compton cones over all events. The overlapping position of the Compton cones is the origin of the gamma-rays.

Conversely, in the SBP image, most parts of a cone not overlapping with a source become the background. Imaging capability should be improved using a deconvolution method that properly accounts for the response of the Compton camera. We selected a deconvolution algorithm called List-Mode Expectation-Maximizing Maximum Likelihood (LM-EM-ML). Briefly,

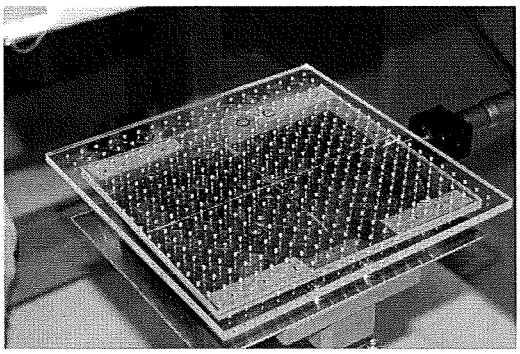

Fig. 9. Acrylic plate with 2 mm diameter holes arranged regularly. The liquid ^{131}I (364 keV) sources were mounted at 20-mm pitch.

this is an iteration algorithm between image space and data space.

$$\lambda_j^{l+1} = \frac{\lambda_j^l}{s_j} \sum_i \frac{t_{ij}}{\sum_k t_{ik} \lambda_k^l} \tag{2}$$

where, λ_j^l denotes the image-bin content at iteration level l, s_j the probability that an event emitted from j is measured, and t_{ij} the probability that a photon emitted from j is measured with the parameters of event i (response). From image space, estimated data space is calculated with a response matrix derived from list-mode data. By comparing the expected data and measured data, corrections are made to image space. A detailed description of the LM-EM-ML method is given in [17], [18].

V. Results

A. Distance Dependence

As mentioned in Section II, the spatial resolution is a function of the distance between the scatterer and target. To demonstrate this concept, we obtained point source data in three cases of $d = 30$ mm, 60 mm and 150 mm. Here, d denotes the distance between the DSSD and the point source, as shown in Fig. 5. The 356 keV gamma-ray source is placed on the Z-axis penetrating the center of the DSSD.

The left panel in Fig. 7 shows the distance dependence of spatial resolution of the SBP image; the right panel shows that of the angular resolution defined by FWHM of the ARM distribution. We evaluated the spatial resolution as FWHM of the SBP image by fitting using a two-dimensional Voigt function. The function is a convolution of Gaussian and Lorentzian distribution, and offers good approximation of a point spread function. We also determined the angular resolution as the ARM distribution in FWHM, by using a one-dimensional Voigt function. The obtained angular resolution was 3.6° and independent of the distance. The spatial resolution (proportional to distance) was about 6 mm and 18 mm at $d = 30$ mm and $d = 150$ mm, respectively. This relation is consistent with the description given in Section II.

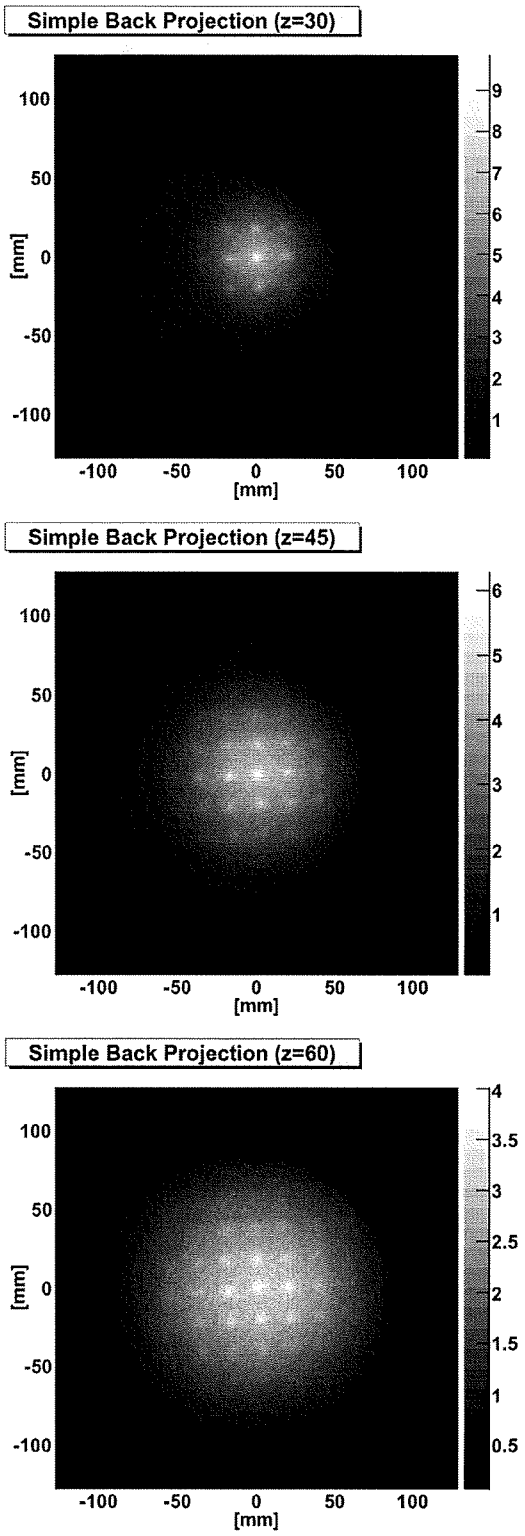

Fig. 10. SBP image of the grid sources in case of $d = 30$ (top), 45 (center) and 60 (bottom) mm. The liquid ^{131}I (364 keV) sources were mounted with a pitch of 20 mm.

B. Field of View

One feature of Compton imaging is a wide field of view. By using 356 keV and 511 keV gamma-ray point sources, we investigated the position dependence relative to detection efficiency. The point source was placed on the plane of $d = 60$ mm (see Fig. 5), and measurements were performed for several positions on the plane. The detection efficiency for each position is obtained from the counts of events where the total energy deposit

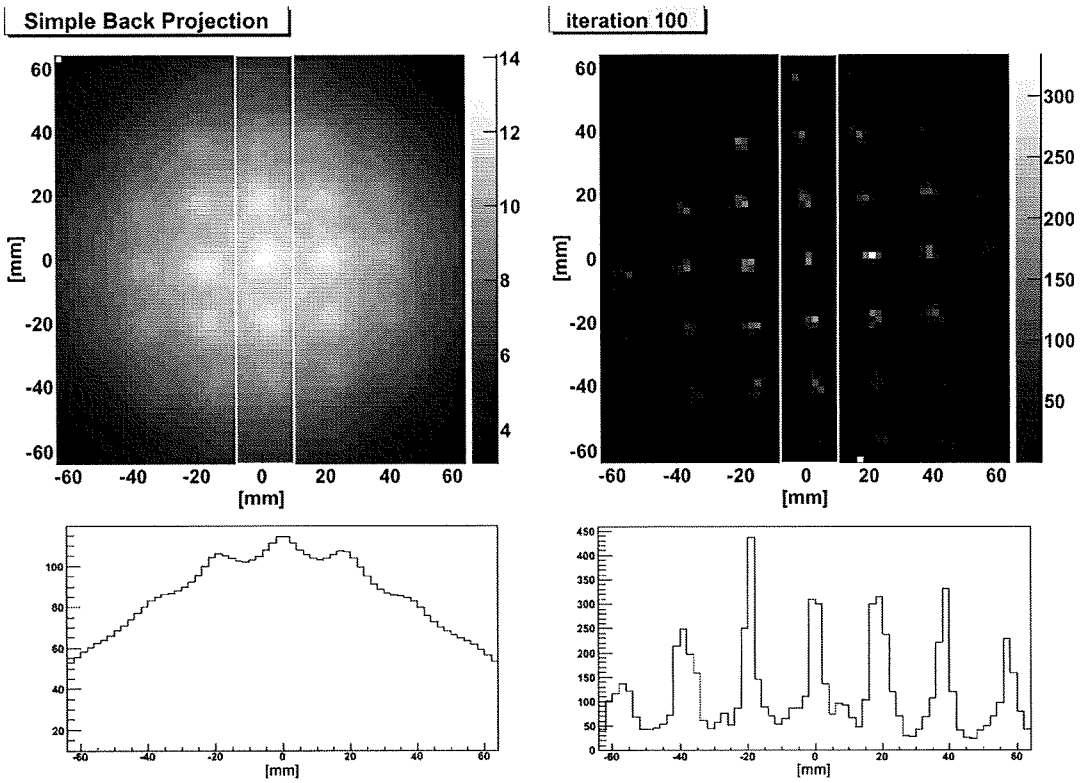

Fig. 11. Deconvolution results for experimental data (d = 60 mm in Fig. 10). The left panel shows the SBP image; the right panel shows the deconvolved image after the 100th iteration. The bottom panel is the projection of the rectangular region of each image to the Y-axis.

$(E_{DSSD} + E_{CdTe})$ is within 356 keV ± 5 keV or 511 keV ± 5 keV.

Fig. 8 shows the position dependence of detection efficiency normalized at the center position of $(x, y, z) = (0, 0, 60)$ for the 356 keV (left) and the 511 keV (right) gamma-rays. The radius denotes the distance from $(x, y) = (0, 0)$. The normalized efficiency at a radius of 100 mm (corresponding to a 120° field of view) is 40% for 356 keV and 30% for 511 keV.

For a detailed evaluation of detection efficiency, we developed a simulator [19] based on the Geant4 toolkit. In the simulation, we included the G4LECS [20] extension in order to estimate the effect of Doppler broadening. The mass model is constructed not only with the detector material, but also with various passive support structures. The energy and position resolutions of the detectors are parameterized based on the experimental results. Fig. 8 shows the normalized efficiency calculated by the simulator, together with the experimental data. The simulation reproduced the experimental results very well. The absolute efficiency derived from the simulator is on the order of 10^{-2}% at the center position for 356 keV gamma-rays.

C. Imaging of Grid Sources

To investigate the resolving power and accuracy of positional determination, an imaging test was conducted using an acrylic plate with 2 mm diameter holes arranged regularly (Fig. 9). The liquid ^{131}I (364 keV, 85 kBq/hole) sources were mounted at 20-mm pitch. The variability of strength of each source is less than 10%. Imaging is performed in three cases: $d = 30, 45$ and 60 mm.

Fig. 10 summarizes the SBP images. The point sources were clearly resolved. The outside sources become visible as d increases, since the sources enter the field of view. Resolving power better than 20 mm was demonstrated up to a distance of $d = 60$ mm, and larger than an 80° field of view as well.

We performed image deconvolution by using the LM-EM-ML method. A total of 1×10^5 events are utilized in this process. A correction factor (s_j) in (2) is derived from the position dependence of detection efficiency calculated by the simulator mentioned in Section V-B. Fig. 11 shows the deconvolution results for the experimental data obtained at $d = 60$ mm. The left panel shows the SBP image; the right panel shows the deconvolved image after the 100th iteration. In the deconvolved image, the point sources placed at the edge become apparent, while only sources located around the central region are resolvable in the SBP image. The bottom panel is the projection of the rectangular region shown in each image to the Y-axis in Fig. 11. Following deconvolution, the peak area of each source becomes stable, thanks to the deconvolution technique that included the correction of efficiency.

It is also important to study the accuracy of positional determination. Therefore, we first derived the center position of the sources from the deconvolved image shown in the right panel in Fig. 11. For the 25 sources arranged in 5 × 5, the center position of each source was determined by calculating the weighted center around the largest pixel. The center positions are plotted as shown Fig. 12 with open squares. Then based on the assumption that the sources are at the intersection of cross lines at a right angle, we fit the center positions. The red grid in Fig. 12 shows the fitting results. The bottom panel of Fig. 12 summarizes the

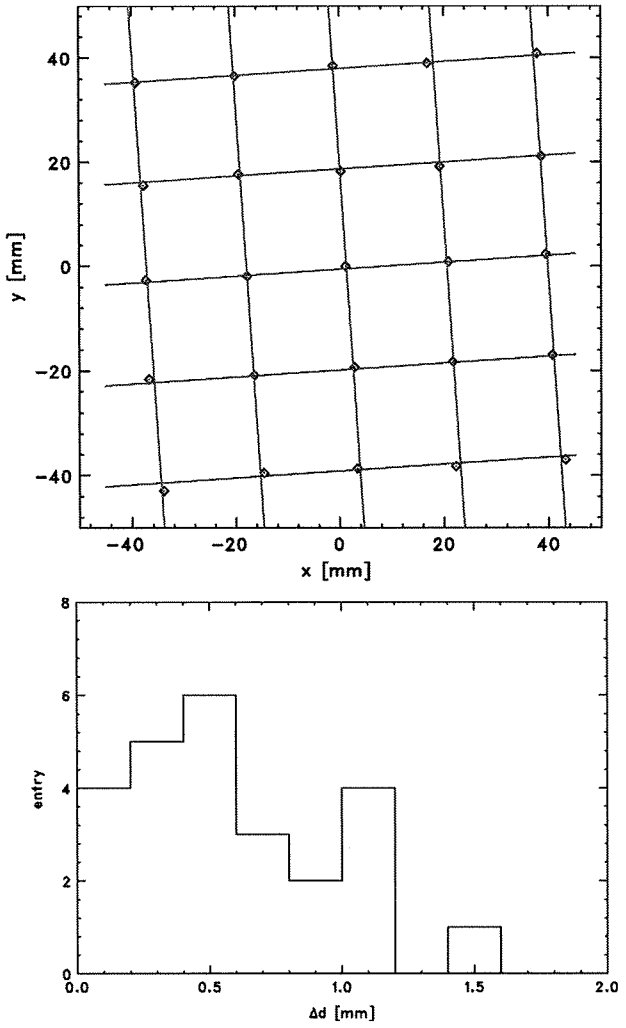

Fig. 12. Top: measurement values of source positions (open square) derived from the deconvolved image shown in the right panel of Fig. 11 in the grid (cross lines). Bottom: distribution of the difference of all 25 sources.

differences of these positions of all 25 sources as distributed within ~1 mm. Thus, this experiment demonstrates positional determination accuracy of 1 mm for the point source.

D. Extended Object

Since the ability to image extended object is a key requirement for both astrophysical and medical applications, we prepared an extended target as shown in Fig. 13. The liquid ^{131}I source (364 keV, 1.6 MBq) is soaked in paper and shaped like an inverted "C" with a gap of 3 mm. The target was placed on the $d = 30$ mm plane.

Fig. 14 shows the images obtained. The top panel shows the SBP image; the bottom panel shows the 10th iteration using the LM-EM-ML method. The shape with a gap of 3 mm is recognized even in the SBP image. With the LM-EM-ML method, the shape is successfully deconvolved. Thus, resolving power better than 3 mm is demonstrated experimentally.

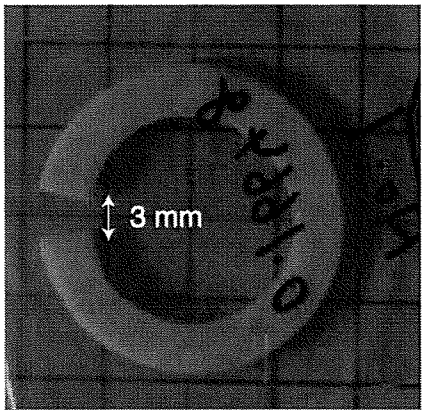

Fig. 13. Photograph of the extended target. The liquid ^{131}I source in soaked in paper, and shaped like an inverted "C" with a gap of 3 mm.

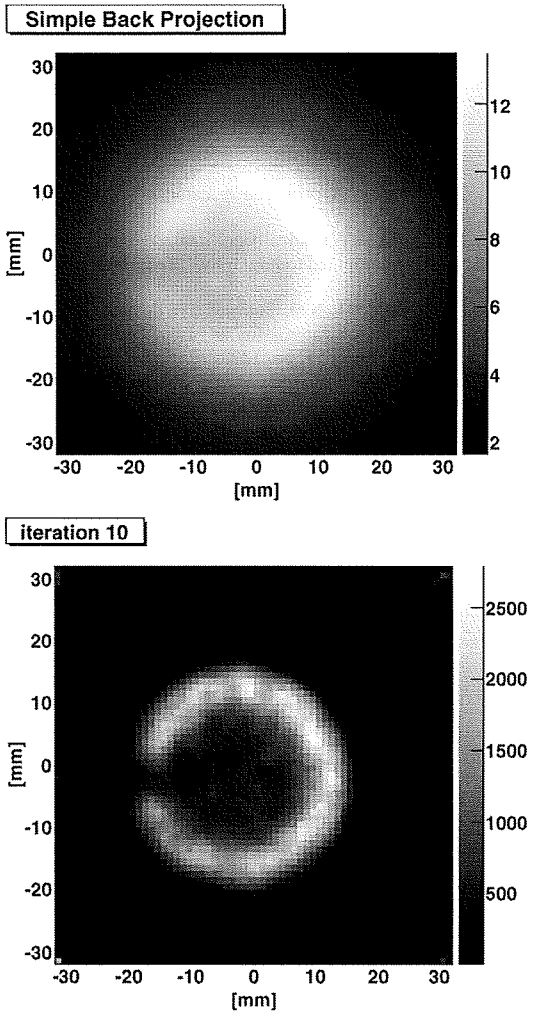

Fig. 14. Results of extended target imaging. The distance to the target is d = 30 mm. The top panel shows the SBP image; the bottom panel shows the deconvolved image after the 10th iteration.

VI. Conclusion

By combining the DSSD and a 4-layer stacked CdTe module, we developed a new Compton camera designed to achieve high spatial resolution. As explained in Section II, the spatial resolution gradually deteriorates as the distance increases between the target and scatterer. Therefore, the camera is designed to

minimize this distance. A minimum distance of 25 mm was realized. Studies were conducted on the field of view, spatial resolution, the accuracy of positional determination, and resolving power. Using the experimental data, resolving power better than 3 mm was demonstrated for a 364 keV (^{131}I) gamma-ray placed 30 mm away from the sensor. Positional determination accuracy of 1 mm was also demonstrated experimentally. Furthermore, the imaging of an extended object was also successfully performed.

As the deconvolution method, we selected an iteration algorithm called List-Mode Expectation-Maximizing Maximum Likelihood (LM-EM-ML). It was applied to several kinds of experimental data. The Compton back projection images derived from the arranged point sources and the extended object were successfully deconvolved.

The results of this imaging test suggest that the Si/CdTe Compton camera is an attractive detector not only for astrophysics application, but also for medical imaging. The data obtained by the experiments also proved effective in investigating the detailed response of a Compton camera used in astrophysics.

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PHYSICAL REVIEW C **79**, 034313 (2009)

Search for a "3.5-eV isomer" in ^{229}Th in a hollow-cathode electric discharge

T. T. Inamura and H. Haba
RIKEN Nishina Center, Wako, Saitama 351-0198, Japan
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A hollow-cathode electric discharge, a well-established source in optical spectroscopy, was used to populate the "3.5-eV isomer" in ^{229}Th with use of nuclear excitation by electron transition (NEET). The radiochemically purest ^{229}Th sample was loaded into the hollow cathode in which the electric discharge excited the ^{229}Th to atomic states some of which could be expected to lie close to the excitation energy of the sought isomer. Although there remain some uncertainties, our experiments indicate that the isomer was populated by NEET and its α decay observed after switching off the electric discharge with a corresponding isomer half-life 1 min $\lesssim T^m_{1/2} \lesssim$ 3 min. From the present NEET condition, the isomer appears to lie between 3 eV and 7 eV. The probability of the isomer population by NEET is discussed.

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I. INTRODUCTION

Because of its uniquely low excitation in the nucleus, the isomer ^{229}Th^m is important in its own right and in understanding its nuclear structure. But its existence has yet to be established in spite of numerous efforts made experimentally and theoretically for the last three decades. We report an experiment that is expected to significantly advance this study of ^{229}Th^m, Nuclear Excitation by Electron Transition (NEET) in a hollow-cathode electric discharge.

The extremely low-lying isomer at an energy of <100 eV in ^{229}Th was first postulated by Kroger and Reich [1] who studied γ rays following the α decay of ^{233}U. Eighteen years later, Helmer and Reich [2] revised that excitation energy to 3.5 ± 1.0 eV. Since then, the claimed isomer has often been called "3.5-eV isomer" in ^{229}Th. Meanwhile, Burke *et al.* [3] suggested the existence of such a low-energy isomer at $\leqslant$5 eV by observing angular distributions of the ^{230}Th$(d, t)^{229}$Th reaction. Their results are, however, not direct evidence to support the existence of such an extremely low-lying nuclear isomer ^{229}Th^m. Later, several attempts were made to obtain direct evidence. Because the excitation energy corresponds to the uv, mostly optical measurements were attempted to detect the possible $M1$ isomeric transition that was expected to ensue from the α decay of ^{233}U [4–7]. None of them was successful.

Recently, two different attempts were made to determine the lifetime of ^{229}Th^m with use of radiochemical techniques designed to avoid the huge background inevitably caused by the parent nucleus ^{233}U. Browne *et al.* [8] searched for the α decay of the isomer to ^{225}Ra using "fresh" ^{229}Th chemically separated from the parent nucleus ^{233}U. They measured the change with time of the 193-keV γ-ray intensity from ^{225}Ra, and concluded that the half-life of the isomer must be either <6 h or >20 d. Mitsugashira *et al.* [9] measured α-decay spectra of the reaction products ^{230}Th$(\gamma, n)^{229}$Th, ^{229}Th^m. They concluded that the half-life of the isomer was 14 ± 3 h. Their measurements, however, were not self-consistent, and neither gave clear evidence for the existence of ^{229}Th^m. More recently, two new measurements were reported by the Mitsugashira's group [10,11]: First, they searched for photons from an atomic process suggested in theoretical studies [12–14] as well as for the $M1$ transition from the isomer after rapid chemistry to separate the parent nuclide ^{233}U; second, search for the α decay of ^{229}Th^m that is expected to be populated by the β^- decay of ^{229}Ac produced by the ^{232}Th$(\gamma, p2n)$ reaction. In the latter, one can assume higher population for the isomer than in the α decay of ^{233}U. Again, convincing results were not obtained. We also note the work of Gangrsky *et al.* [15] in which the isomer was expected to be populated by nuclear deexcitation of ^{229}Th irradiated with bremsstrahlung from a microtron. No trace of the isomer was detected in the time range between 2 min and 10 h.

There are three extensive γ-ray spectroscopic studies of ^{229}Th: Gulda *et al.* [16] and Ruchowska *et al.* [17] used the β^- decay of ^{229}Ac that was obtained from the decay chain of ^{229}Fr produced by a spallation reaction in a ^{232}ThC$_2$ or ^{238}U target; and Barci *et al.* [18] who relied on the α decay of ^{233}U. However, these inferred only indirectly evidence of ^{229}Th^m. Guimaräes-Filho and Helene [19] reexamined the excitation-energy data of Helmer and Reich and other previous γ-ray studies. They deduced a value of 5.5 ± 1.0 eV. In a recent experiment Beck *et al.* [20] measured the excitation energy E^m by studying the ^{229}Th γ rays with use of the NASA x-ray spectrometer (XRS): Their high-resolution result, $E^m = 7.6 \pm 0.5$ eV, is significantly greater than the currently accepted value of 3.5 ± 1.0 eV. Neither value is a direct measurement of the $M1$ isometric transition, but is deduced from γ cascade-crossover combinations which are considered to involve the isomer. The latter value is corroborated by independent measurements with use of Ge detectors [2,16–18]. The discrepancy in the two values is presently not understood.

Theoretical studies of ^{229}Th^m have been made mainly by Karpeshin *et al.* [12,21] and by Tkalya *et al.* [13]. (Earlier related papers are given in their references.) The latter proposed an experiment to populate ^{229}Th^m with use of synchrotron radiation from the Advanced Photon Source at Argonne National Laboratory, but this experiment has not been reported so far. The experiment by Gangrsky *et al.* mentioned above is similar in principle. Karpeshin *et al.* [12] also proposed an experiment to populate the isomer with use of laser excitation. In practice this would require a precise

knowledge of E^m. This also holds for an experimental proposal of Kálman and Bükki [14], who made a critical study of the theoretical work of Karpeshine *et al.* From the experimental viewpoint, however, these may all be moot without direct evidence for the existence of $^{229}Th^m$.

Karpeshin *et al.* [22] proposed an interesting experiment to ascertain the existence of $^{229}Th^m$ by making use of hydrogen-like ^{229}Th and $^{229}Th^m$ at GSI in Darmstadt. The advantage of this proposal is in the fact that $M1$ transitions between hyperfine structure components of this isomer and the ground state would be enhanced by several orders of magnitude compared to ones in normal atomic ^{229}Th. No actual experiment has been reported. A yet different attempt to get direct evidence of this isomer by collinear laser spectroscopy has been reported by Tordoff *et al.* [23].

II. EXPERIMENT

A. Feasibility study

Two feasibility studies of NEET in a hollow-cathode electric discharge were made by Inamura *et al.*, the first, measurement of photons [24], the second, of α particles [25]. We review these briefly.

In a hollow-cathode electric discharge one can observe atomic excited states in neutral atoms and up to the continuum in singly charged ions. A test was carried out on thorium and it was confirmed that all atomic states in natural Th and Th^{1+} up to $\approx$10 eV could be excited. A first critical test was to confirm that after switching off the discharge there is no afterglow from atomic excited states as delayed photons attributable to the "3.5-eV isomer" were sought. Experiments described in Ref. [25] switched to the detection of the α decay of $^{229}Th^m$, taking advantage of its better S/N ratio.

The optical measurement, made with a uv spectrometer, gave an estimate $N \approx 10^{13}$ for the number of atoms per second at a given excited state [24,25]. By assuming a NEET probability $P_{\mathrm{NEET}} \sim 10^{-10}$ [12], the population of $^{229}Th^m$, $N \cdot P_{\mathrm{NEET}}$, was expected to be $\approx 10^3$ atoms/s. Consequently, a total counting rate of α particles from $^{229}Th^m$ was estimated to $\lambda_\alpha N^m \approx 1.3 \times 10^{-3}$ s^{-1}, i.e., 4–5 counts/h [25] for an isomer half-life of 14 h [9] and for the ^{229}Th source excited in the electric discharge continuously for ten hours. Further, we expected a considerable enhancement factor that could reach even the order of 10^{11} [12,25] as explained below. The feasibility of this experiment was thus deemed possible.

The excitation and deexcitation processes of the NEET are shown in Fig. 1. The enhancement factor arises from the fact that this process is repeated continuously in the dc electric discharge during the expected lifetime of the isomer.

B. Hollow-cathode electric discharge tube

The hollow-cathode tube for the present study was described in Ref. [25]. However, without changing its geometry, we made it possible to install inside an ambient light-tight solid-state detector (SSD) (ORTEC B-type Si detector) with 150-mm^2 active area and 300-μm depleted depth to detect α particles as rapidly as possible after switching off the discharge. In the present study we used the hollow-cathode tube with or without the SSD depending on the assumed half-life of the isomer. It is shown in Fig. 2 with the SSD in place. The cathode consists of an aluminum module with a hollow and its base made of copper.

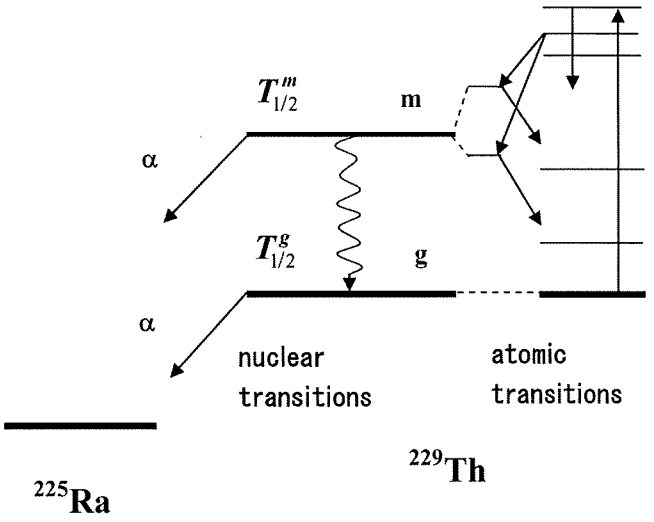

FIG. 1. Schematic excitation and deexcitation processes of $^{229}Th^m$ in the hollow-cathode electric discharge: nuclear ground and isomeric states are denoted by g and m, respectively. Possible intermediate states produced by coupling atomic and nuclear states are denoted by the two short lines.

The thorium sample (as ThO_2—see Sec. II C) is placed inside the hollow from where atoms are sputtered by the discharge-supporting argon gas atoms. The argon pressure was 200 Pa, the discharge current 200 mA dc, and the voltage $\approx$300 V (constant current mode). It is expected that sputtered molecules and atoms are mostly ThO^{1+}, Th, and Th^{1+} [26,27].

C. Source preparation of ^{229}Th

Our ^{229}Th source ($T_{1/2} = 7340$ y) was obtained in May 1980 from the mother ^{233}U sample in a HNO_3 solution by an anion-exchange method. The isotopic impurity ^{232}U in the mother sample was known by α spectroscopy to be 3 ppm. At the beginning, this impurity caused contamination of the separated ^{229}Th by $^{228}Th(T_{1/2} = 1.913$ y), with the ^{228}Th α activity about fifteen times that of ^{229}Th. Today, the ^{228}Th has effectively decayed away to such an extent that it is undetectable. Radiochemically, our ^{229}Th source is almost 100% pure. The ^{229}Th source has been kept in 8 M HNO_3, the concentration of which is 10 mg/L.

Before loading it into the hollow cathode by direct- or electrodeposition, the ^{229}Th source was further purified by the anion-exchange method with use of a chromatographic column (5 mm diameter $\times$40 mm height) filled with an anion-exchange resin (Dowex 1X8, 200–400 mesh): First, 1 mL of the 8 M HNO_3 solution of ^{229}Th was passed through the column to adsorb ^{229}Th by the resin; second, the resin was washed with 4 mL of 8 M HNO_3 to remove impurities such as its daughter elements; and last, the ^{229}Th thus purified was rapidly eluted with pressurized 4 mL 2 M HCl. The resulting elution was

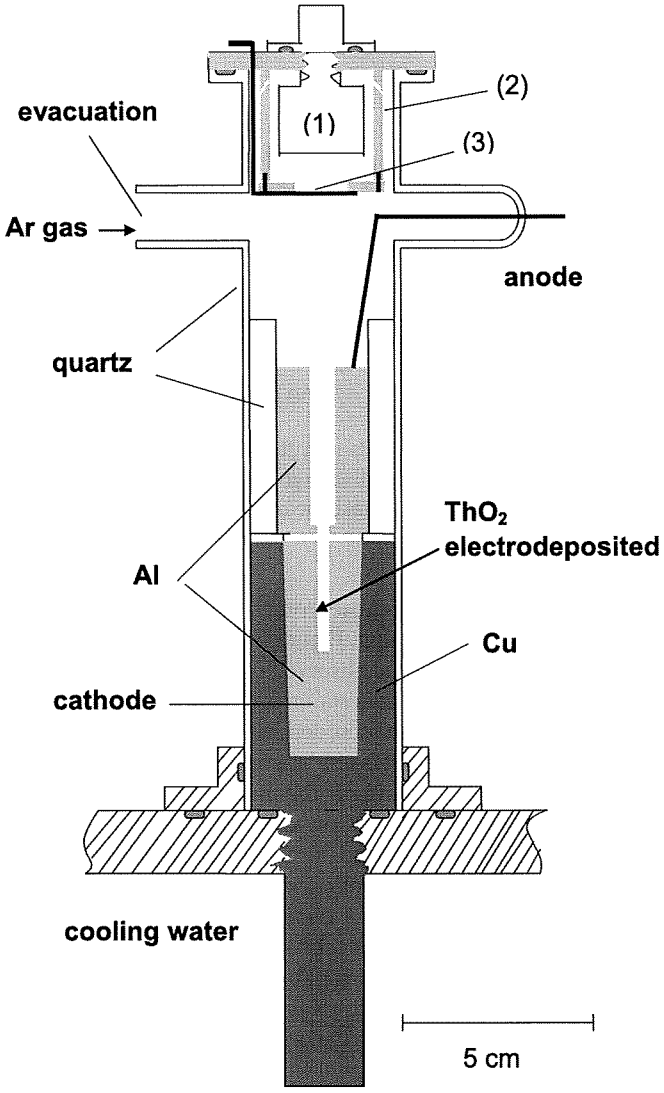

FIG. 2. Cross-sectional view of the hollow-cathode tube with an α-measurement assembly: (1) SSD, (2) aluminum light-shield housing, and (3) a movable shutter to protect the SSD from intense light coming from the hollow-cathode electric discharge. The geometry of the tube is the same as given in Ref. [25].

deposited into the hollow cathode. Alternatively, the elution was dried and then dissolved in 0.5 mL of 0.01 M HNO_3. The concentration of ^{229}Th in the last solution was carefully adjusted by measuring α particles from the ^{229}Th ground state to provide the required source activity.

When the molecular plating method (electrodeposition) [28] was used, a drop (10 μL) of the ^{229}Th in HNO_3 was mixed with 350 μL of 2-propanol to get the appropriate solution. The volume of the hollow, 490 μL, was reduced to $\approx$400 μL after insertion of the 1-mm diameter platinum wire (positive electrode) placed at the center of the hollow and its holder. In this way we obtained the desired uniformly deposited ^{229}Th source inside the hollow cathode, and one in which outgassing is significantly reduced during the discharge.

The electrodeposition was completed in 15 to 30 min. The solution was then washed out with additional 2-propanol and the final efficiency of electrodeposition was found to be greater than 90%.

Before starting the discharge inside the hollow-cathode tube, the aluminum cathode module, loaded directly or by electrodeposition, was baked at 250°C in vacuum for more than two hours. The chemical form of ^{229}Th loaded inside the hollow cathode is considered to be ThO_2 just before the discharge.

D. Measurements

Measurements were made separately in search of ^{229}Th^m depending on its assumed half-life $T^m_{1/2}$.

1. Measurement for $T^m_{1/2} \gtrsim 1$ h

First, we intended to confirm the half-life that Mitsugashira *et al.* [9] reported previously: $T^m_{1/2} = 14 \pm 3$ h. About 1 kBq of ^{229}Th (in a HCl solution as described in Sec. II C) was deposited directly around the middle of the inside wall of the aluminum cathode module. The ^{229}Th source thus prepared was subjected continuously for three hours to the electric discharge in the hollow-cathode tube. After switching off the discharge, the hollow-cathode tube was disassembled rapidly to take out the cathode module and the ^{229}Th source was dissolved with 8 M HNO_3. This solution was passed through the anion-exchange column as described in Sec. II C to purify the thorium source which was supposed to contain ^{229}Th^m populated during the discharge. The thorium elution was co-precipitated with a small amount of samarium as hydroxide with an excess amount of aqueous NH_3. The precipitate filtered on a filter paper (Whatman, Anodisc25) 18 mm in diameter was sintered on the hot plate. This method has proved to provide an excellent α source of ^{229}Th/^{229}Th^m [11].

Twenty minutes after switching off the discharge, we started measurements of α spectra with use of the SSD as a function of time for 17 h. We did not observe significant time-dependent α yields (see Fig. 3). We could not confirm the half-life as was reported by Mitsugashira *et al.* [9]. Next, as described in the following subsection, we tried to make measurements for a short-lived isomer, for which we could adapt our experiment.

Figure 3 shows α spectra observed for the first 10 min, twenty minutes after switching off the discharge and for the last 10 min, seventeen hours after the switching off the discharge. Four clear peaks are from the α decay of the ground state $5/2^+[633]$ of ^{229}Th [29]: the most intense one is mainly of the 4845-keV α transition (56.2), the second is 4901-keV (10.2), the third 4968-keV (6.0) and 4978-keV (3.2), and the fourth 5053-keV (6.6) (the intensities given in the parentheses are percentages of the total α decay of the ground state; the remaining intensities come from α transitions sitting on the low-energy side of the main peak).

2. Measurement for 3 min $\lesssim T^m_{1/2} \lesssim 1$ h

We tried several times to do rapid chemistry in order to shorten the time between starting the measurement and

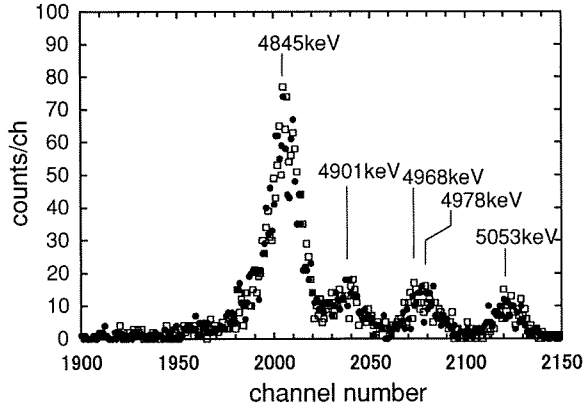

FIG. 3. Comparison of ^{229}Th α spectra recorded each for 10 min, 20 min (•), and 17 h (□) after switching off the dc electric discharge (a period of 3 h) in the hollow-cathode tube loaded with ≈1 kBq of ^{229}Th.

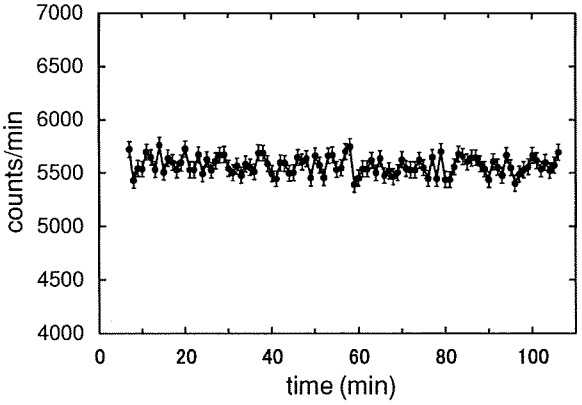

FIG. 5. Total ^{229}Th α yields as a function of time for 100 min, 6 min after switching off the discharge (a period of 30 min). Rapid chemistry was carried out to make the α source.

switching off the discharge, while retaining good energy resolution, 22-keV full width at half maximum (FWHM), for 8376-keV α particles from a daughter nucleus, ^{213}Po, in the decay chain of ^{229}Th. First, 14 min after switching off the discharge, we made measurement of α spectra for 2-min intervals successively for 100 min. Second, 8 min after the discharge, we made the measurement for 5-min intervals successively for 100 min. Finally, we succeeded in making two measurements 6 min after the discharge, and we recorded α spectra for 1-min intervals successively for 100 min. For each measurement the ^{229}Th source was freshly electrodeposited inside a fresh aluminum cathode module as described in Sec. II C, and its activity was increased to ≈5 kBq. The discharge period was 1/2 h and the chemical procedure to get the α source was the same as described in the previous subsection.

Figure 4 shows an example of α spectra from the daughters of ^{229}Th as well as from ^{229}Th. It is seen that α particles from ^{229}Th are well separated from those of the daughters. There is no possibility for the α particles from the daughters to affect the energy range of those from ^{229}Th and to produce any spurious delayed component. Figure 5 shows the total ^{229}Th α yields as a function of time observed 6 min after switching off the discharge. Statistical uncertainties are indicated on the data points. The energy spectra recorded are excellent, but presently no time-dependent α yields are apparent. To further shorten the time between switching off the discharge and starting the measurement, we used the hollow-cathode discharge tube with the SSD inside, as described in Sec. II B, and measured α particles directly from the hollow cathode. This enabled us to start the measurement half a minute after switching off the discharge and evacuating the tube.

Figure 6 shows the total ^{229}Th α yields as a function of time thus observed after being subjected to the discharge for one hour: measurements were made at 5-min intervals over a period of 150 min. The data presented in Fig. 6 are the sum of the first two measurements with the same ^{229}Th source. The source activity was increased to ≈25 kBq. Although statistical fluctuations are large for the entire time range, there appears to be a time-dependent component with a short half-life. To

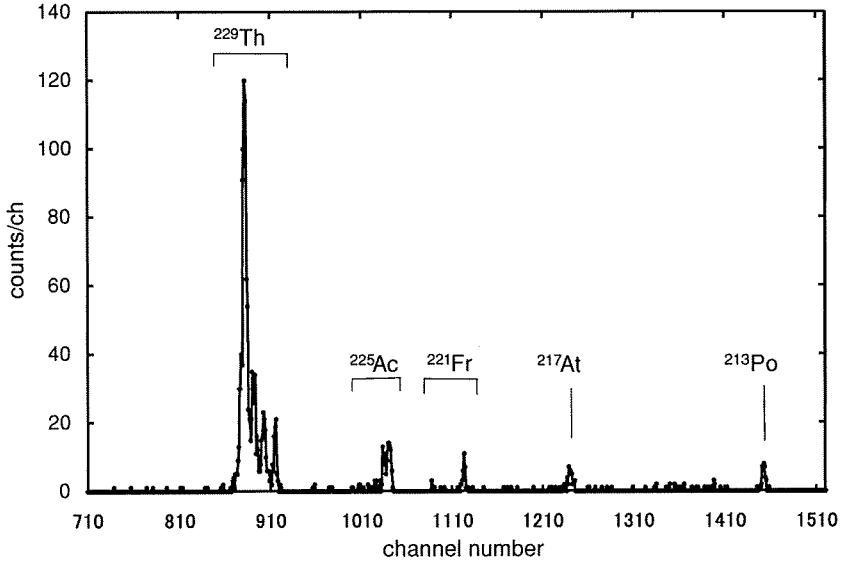

FIG. 4. α spectra from the daughters of ^{229}Th as well as from ^{229}Th observed for 2 min after being subjected to the discharge for 30 min and chemically separated. The activity of the ^{229}Th source was ≈5 kBq.

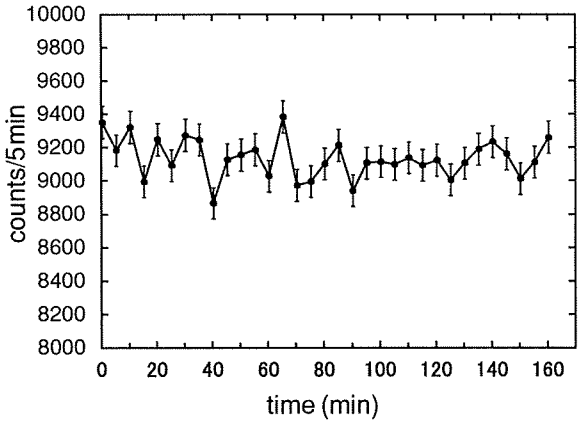

FIG. 6. Total ^{229}Th α yields as a function of time observed inside the hollow-cathode tube after being subjected to the discharge for 1 h. The measurement started half a minute after switching off the discharge and the activity of ^{229}Th source electrodeposited inside the hollow cathode was ≈25 kBq.

check further, we made measurements with 1-min intervals as described in the next subsection.

3. *Measurement for* 0.5 min $\lesssim T_{1/2}^{m} \lesssim$ 5 min

The ^{229}Th source was electrodeposited and its activity was increased further to ≈40 kBq. Ten measurements were made with this source. The discharge period was 5 min each time. α spectra were recorded with the setup described in Sec. II B half a minute after switching off the discharge and evacuating the tube. Measurements were made at 1-min intervals successively over 30 min for the first three runs and over 15 min for the rest. Figure 7 shows α yields from the ^{229}Th source as a function of time for the ten measurements which were added successively: (a) the total α yields from ^{229}Th and possible ^{229}Th^{m}; (b) the total daughters' α yields for comparison. The important observation is the decrease with time seen in Fig. 7(a) but not in Fig. 7(b). This trend improves with the better statistics in the first 10 min, as seen in the top data. If we assume that the trend is probably due to the decay of ^{229}Th^{m}, we will try to extract its half-life as described in the next section.

III. DATA ANALYSIS

To extract the half-life $T_{1/2}^{m}$ of ^{229}Th^{m} from the data [top curve in Fig. 7(a)], we have to examine the background level

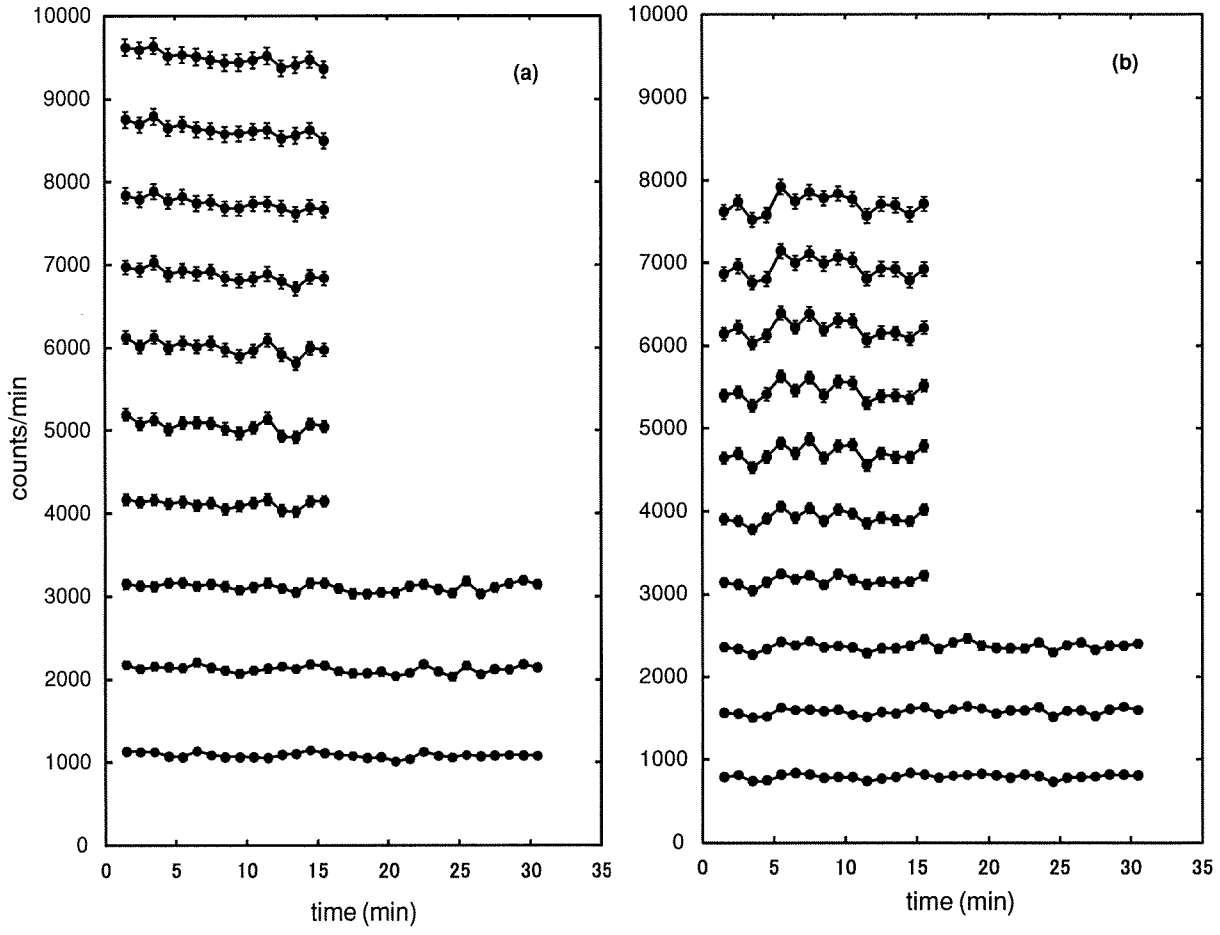

FIG. 7. Total α yields as a function of time for a shorter time range: (a) ^{229}Th and possible ^{229}Th^{m}; (b) daughters for comparison. The measurement was made as presented in Fig. 6 but with a discharge period of 5 min. The lowest curves are for the first measurement after being subjected to the first discharge. The data were successively added up to the tenth measurement (top curves). The activity of the ^{229}Th source was ≈40 kBq.

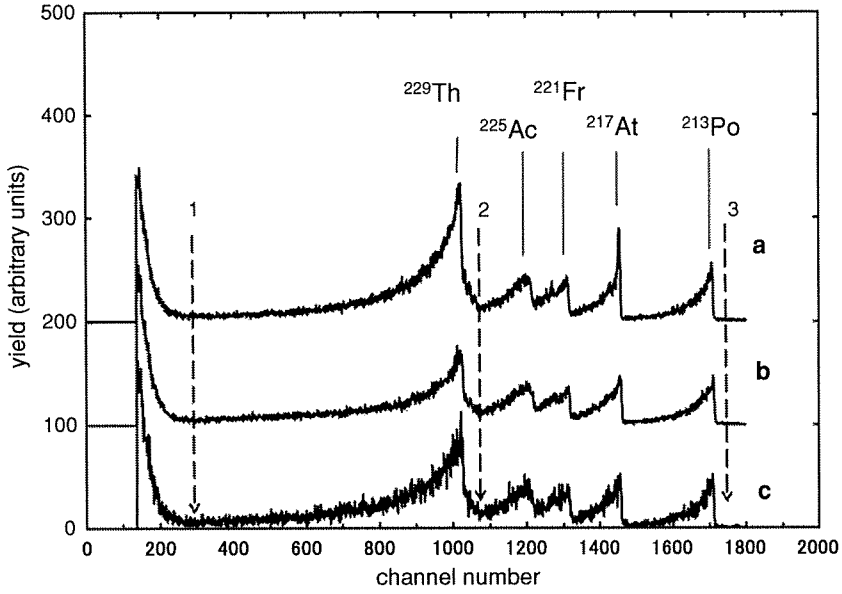

FIG. 8. α spectra observed for 50 min before (a) and after (b) being subjected to the discharge. For comparison, spectrum (c) is presented, which was obtained by integrating measurements over time (0.5–15.5 min) for the first run (see Fig. 7). Spectra (a) and (b) are normalized to 15-min measurements, respectively. Dashed arrow lines 1 and 2 indicate the region of interest to integrate to obtain the total α yields from ^{229}Th and possible ^{229}Th^m as a function of time [see Figs. 6 and 7(a)]. Dashed arrow lines 2 and 3 indicate the α-energy range of daughters which resulted in Fig. 7(b).

due to the α decay of the ^{229}Th ground state. The decay of the ground state ($T^g_{1/2} = 7340$ y) is time-independent in the range of interest. Nevertheless, we have to examine the effects of α emitting daughters, all of which have $E_\alpha > 5100$ keV, because individual α lines might have spread significantly on their lower energy side into the α-energy range of ^{229}Th as a result of the deteriorated energy resolution of α spectra recorded inside the hollow-cathode tube. The ^{229}Th source position was seen obliquely by the SSD (see Fig. 2) and the energy resolution significantly diminished (see Figs. 3, 4, and 8 for comparison).

The α emitting daughters in the decay chain are [29]: ^{225}Ra (14.8 d) $-\beta^-\rightarrow$ ^{225}Ac (10.0 d) $-\alpha\rightarrow$ ^{221}Fr (4.9 min) $-\alpha\rightarrow$ ^{217}At (32.3 ms) $-\alpha\rightarrow$ ^{213}Bi (45.6 min) $-\beta^-\rightarrow$ ^{213}Po (4.2 μs) $-\alpha\rightarrow$ ^{209}Pb (3.25 h) $-\beta^-\rightarrow$ ^{209}Bi. Figure 8 shows a comparison of α spectra observed before and after the discharge in the third measurement (Sec. II D 3). There is no significant difference among the α spectra except for a loss of ^{229}Th caused by sputtering during the discharge. The loss of the ^{229}Th source is significant, but does not affect time-dependence of α yields.

As is seen from Fig. 8, the individual α lines are spread out over the lower energy side and prevented the identification of any weak α transitions attributable to the decay of the isomer. It also means that any possible α transitions from the isomer are definitely less than ten percent of the total α-decay rate of the ^{229}Th ground state (see the known relative intensities of its components given in Sec. II D 1); we note that the three weak α lines from the ground state were in fact observed. While the energy spectra may overlap, we recall Figs. 7(a) and 7(b) that indicate a significant difference as a function of time. Therefore, we conjecture that ^{229}Th^m was populated by NEET during the discharge and it disappears in minutes.

With this conjecture the total counting rate of α particles from ^{229}Th and ^{229}Th^m at time t is written as

$$n_\alpha(t) = \lambda^m_\alpha N^m(t) + \lambda^g_\alpha N^g(t), \tag{1}$$

where λ^m_α is the α-decay constant for ^{229}Th^m, and for the ground state ^{229}Th $\lambda^g_\alpha \equiv \lambda^g = \ln 2/T^g_{1/2}$. We take $t = 0$, the time when the discharge is switched off. At time t, the number of ^{229}Th^m atoms, $N^m(t)$, and the number in the ground state ^{229}Th, $N^g(t)$, are given, respectively, by

$$N^m(t) = N^m(0)e^{-\lambda^m t}, \tag{2}$$

$$\frac{dN^g(t)}{dt} = \lambda^m_p N^m(t) - \lambda^g N^g(t), \tag{3}$$

where λ^m_p is the total photon-decay constant for the isomer. Noting $\lambda^m_p \approx \lambda^m = \ln 2/T^m_{1/2}$ because $\lambda^m_\alpha \ll \lambda^m_p$, we obtain from Eqs. (2) and (3)

$$N^g(t) = N^m(0)\frac{\lambda^m}{\lambda^g - \lambda^m}\left(e^{-\lambda^m t} - e^{-\lambda^g t}\right) + N^g(0)e^{-\lambda^g t}, \tag{4}$$

the number of ^{229}Th atoms before the discharge is given, approximately, by $\overline{N} = N^m(0) + N^g(0)$.

Noting that $T^g_{1/2} = 7340\,y$, and $\lambda^m \gg \lambda^g, t \ll T^g_{1/2}$ or $\lambda^g t \ll 1$, we have from Eq. (4)

$$N^g(t) \approx \overline{N} - N^m(0)e^{-\lambda^m t}. \tag{5}$$

Combining Eqs. (1) and (5), we obtain

$$n_\alpha(t) = \left(\lambda^m_\alpha - \lambda^g\right) N^m(0)e^{-\lambda^m t} + \lambda^g \overline{N}. \tag{6}$$

The second term gives the background, i.e., α particles from the ground state. When we make successive measurements at intervals Δ following a time Δ_0 after switching off the discharge, we can reduce Eq. (6) to

$$n_\alpha(t_i) = Ce^{-\lambda^m t_i} + \lambda^g \overline{N}\Delta, \tag{7}$$

where $t_i = \Delta_0 + i\Delta$, $i = 1, 2, 3, \ldots$ and $C = [(\lambda^m_\alpha - \lambda^g)/\lambda^m]$ $N^m(0)(e^{\lambda^m \Delta} - 1)$. In fact, the second term should also include the environmental background.

In principle, it should be possible to fit the data to a function of the form Eq. (7) in order to extract the isomer decay constant λ^m. Unfortunately, statistical uncertainties are not sufficiently small to allow a linear least-squares fit to the residue after background subtraction on the logarithmic scale. Instead, we examine the decay properties by non-linear least-squares of

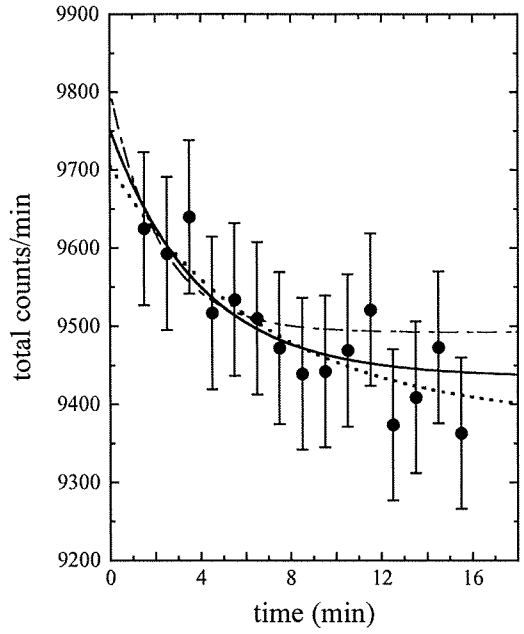

FIG. 9. Least squares-fits to the top data in Fig. 7(a) with different background constraints: solid curve for 9435, dash-dotted for 9492, and dotted for 9378 (see the text for details).

Eq. (7) for three probable background levels obtained in the following way.

Comparing the three independent measurements (see Figs. 5, 6, and 7), we assume that the α yields as a function of time become the background level about 10 min after switching off the discharge. In Fig. 7(a) we take this to be 10.5 min and the background level, including environmental, is estimated to be 9435 ± 57 by averaging the yields [top curve in Fig. 7(a)] over the range 10.5–15.5 min. Figure 9 shows the top data with nonlinear least-squares fits for the three background levels: the solid curve corresponds to 9435, the dash-dotted curve to $9435 + 57$, and the dotted curve to 9435–57. In the actual least-squares fit to the data, Eq. (7) is renormalized to include the detection efficiency Ω: $n_\alpha(t_i)$ and $\lambda^g \overline{N} \Delta$ are replaced by actual counts, respectively, and the constant C is replaced by $C' = \Omega C$, where $\Omega = (3.5 \pm 0.4) \times 10^{-4}$, the ratio of the total ^{229}Th α yield observed inside the hollow-cathode tube to that from the ^{229}Th source activity electrodeposited. The results of the fits are summarized in Table I. We note that without background-level constraints the fitting is not possible because the number of points is too small to determine the background as a free parameter. Mention should also be made of the fact that the smaller value of χ^2 does not necessarily mean the better fit in the present case, and we do not deduce a value unique to λ^m, but rather consider the probable *largest* limit of $\lambda^m = 0.452 + 0.447 \approx 0.9$ min^{-1}, i.e., the probable *shortest* half-life $\approx$1 min, and, as discussed below, the probable *longest* half-life.

For the probable *longest* limit of the half-life, we consider the second measurement shown in Fig. 5 for the interval 3 min $\lesssim T^m_{1/2} \lesssim$ 1 h (Sec. II D 2) and the constant C' rather than the probable *smallest* value of λ^m in Table I that gives a half-life $\approx$140 min. For the present purpose we take $C' = 320 \pm 170$,

TABLE I. Least-squares fits to the data with the form Eq. (7). See Fig. 9 and the text for details.

2nd term	λ^m (min^{-1})	C'	χ^2
9492	0.452 ± 0.447	311 ± 338	5.87
9435	0.253 ± 0.147	317 ± 167	2.93
9378	0.146 ± 0.064	327 ± 108	2.48

the median of the fits. This corresponds to ten times the average total α yield from ^{229}Th^m per minute at $t = 0$. Now we consider $R_i = C'_i/(\lambda^g \overline{N} \Delta)'_i$, $i = 2, 3$, the ratio of the total probable α yield from ^{229}Th^m to that from the ground state ^{229}Th in the second measurement $i = 2$ (Fig. 5) and in the third measurement $i = 3$ [top data in Fig. 7(a)] at $t = 0$. (The prime indicates the renormalized values as described in the preceding paragraph.) Generally, we expect $R_2 \gtrsim R_3$ where R_2 depends on the isomer lifetime because the isomer should be populated by the same NEET, and the discharge period $T_2 > T_3$. Since we are interested here only in the *shortest* measurable half-life of the isomer in the second measurement that will be the probable *longest* half-life limit of the isomer in the third measurement, we can assume $R_2 = R_3$, otherwise R_2 will give even a shorter half-life. We found $(\lambda^g \overline{N} \Delta)'_3$ to be 6400 ± 140 after measuring the environmental background without the aluminum cathode module loaded with ^{229}Th. We thus find $R_3 = 0.050 \pm 0.027$.

The average net counts per minute in the second measurement (see Fig. 5) is $\approx$5600. This should be equal to $(\lambda^g \overline{N} \Delta)'_2$, but C'_2 is unknown. If it could be observed, the probable total α yield from the isomer should be comparable at least to or larger than the statistical uncertainty ($\pm$75) of that average counts. The probable total α yield per minute from the isomer is given by $C'_2 = 5600 R_2$. From these considerations, the α yield from the isomer with a half-life of 3 min would be marginal to be observed 6 min after switching off the discharge. Consequently, the upper limit of the isomer half-life $T^m_{1/2}$ is reckoned at $\approx$3 min and the probable range is 1 min $\lesssim T^m_{1/2} \lesssim$ 3 min.

IV. DISCUSSION

It is of interest to see whether the present results are consistent with the feasibility study that was made independently of the present measurement.

The time dependence of the number of atoms $N^m(t)$ during the discharge is given by

$$\frac{dN^m(t)}{dt} = \eta N P_{\mathrm{NEET}} - \lambda^m N^m(t), \tag{8}$$

where η is an enhancement factor given by the ratio of the mean lifetime ($\tau^m = 1/\lambda^m$) to the NEET cycle period ($\tau_c \sim 0.1$ μs) [12,25]. Hence the number of atoms after the continuous discharge is on for a period of T, is

$$N^m(T) = \eta \frac{N P_{\mathrm{NEET}}}{\lambda^m} \left(1 - e^{-\lambda^m T}\right). \tag{9}$$

For the present measurement (Sec. II D 3), $T = 5$ min, and $N^m(T = 5\ \mathrm{min})$ gives the number of ^{229}Th^m atoms when the discharge is switched off, i.e., $N^m(T = 5\ \mathrm{min}) \equiv N^m(t = 0)$,

as defined in the previous section. Here we assume the median half-life $T^m_{1/2} = 2$ min so that $\lambda^m = 0.347\ \text{min}^{-1}$ and we have

$$N^m(0) \approx 1.4 \times 10^2 \eta N P_{\text{NEET}}, \tag{10}$$

where $\eta \approx 1.7 \times 10^9$.

Next, we extract $N^m(0)$ from the constant C given in the previous section. According to Tkalya *et al.* [13], λ^m_α and λ^g obey the relation $2 \leqslant \lambda^m_\alpha/\lambda^g \leqslant 4$. For simplicity, we assume $\lambda^m_\alpha = 3\lambda^g$, their mean value. Since $\lambda^g_\alpha = \lambda^g = 1.79 \times 10^{-10}\text{min}^{-1}$, we have $(\lambda^m_\alpha - \lambda^g)/\lambda^m \approx 1.04 \times 10^{-9}$. Thus we find $N^m(0) \approx 2.1 \times 10^{14}$ atoms from the present observation. Comparing this value with Eq. (10), we obtain the experimental isomer-population rate by NEET, $N P_{\text{NEET}} \approx 8.6 \times 10^2$ atoms/s, which is very close to the value 10^3 atoms/s that was given in the feasibility study [25]. This agreement is fortuitously good in view of the fact that the feasibility study and the present measurement were made independently.

We electrodeposited ≈40 kBq of ^{229}Th into the hollow cathode, which corresponds to $\approx 1.5 \times 10^{16}$ atoms. This would imply that the ground state was pumped to the isomer $^{229}\text{Th}^m$ at a rate of $1.4 \pm 0.8\%$ (statistical error only) through the hollow-cathode discharge. This is to be tested by measuring delayed photons (afterglow) from $^{229}\text{Th}^m$ (see Fig. 1). Observation of the afterglow will provide direct evidence for NEET to $^{229}\text{Th}^m$ without observing α lines characteristic of its decay. If one could measure energies (or frequencies) of the afterglow accurately, for instance with an optical spectrometer, then it would be possible to determine the isomer excitation energy E^m. We add that there is an uncertainty in the sputtering rate for ThO_2 to produce Th and Th^{1+}, from which we can expect NEET to $^{229}\text{Th}^m$, compared with the rate for the metallic natural thorium foil used in the feasibility study. This uncertainty will affect the number of ^{229}Th atoms available for NEET. One can expect that the production of Th and Th^{1+} will be ≈60% of that for the metallic Th foil (see Sec. II A). We note that Kasamatsu *et al.* [10] suggested the possibility of measuring a half-life of a few minutes or even shorter of $^{229}\text{Th}^m$ by photon detection after a rapid chemical separation of the isomer from the parent nucleus ^{233}U.

Finally, we evaluate the limits of E^m from the NEET mechanism shown in Fig. 1. First, the electric discharge at 300 V could hardly excite atomic states higher than 10 eV, as was found in the feasibility study [24,25]. This provides a limit $E^m < 10$ eV. Further, we consider a calculation of atomic states in Th^{1+} made by Karpeshin *et al.* [12]. These included $8p_{1/2}$, $7p_{3/2}$, and $7p_{1/2}$ electrons which may play a part in NEET to $^{229}\text{Th}^m$ and its deexcitation via atomic states, i.e., a new type of internal conversion. In the first feasibility study [24], we confirmed that there are atomic transitions as predicted by their calculation. Obviously, E^m must be lower than the atomic state from which NEET starts and higher than the possible lowest atomic state. This implies that $3\ \text{eV} \lesssim E^m \lesssim 7$ eV. As noted, this must be tested by measuring the afterglow.

The NEET process is inverse internal conversion, as was pointed out by Morita [30] who, first, showed theoretically that such a process is possible. It was confirmed experimentally in ^{189}Os by Otozai *et al.* [31] five years later. When there is NEET, there should always be internal conversion as an inverse process, or vice versa. The underlying interaction Hamiltonians are exactly the same: electromagnetic interaction between atomic electrons and the nucleus. Whether electrons are involved explicitly or not is totally dependent on the available energy in the interaction. The name "electronic bridge" (EB) was proposed initially by Krutov and Fomenko [32] and has been used often ever since. Because of the added use of "electron-beam bridging" in polymer physics and chemistry, we call the excitation of $^{229}\text{Th}^m$ via atomic states exclusively NEET or IIC (inverse internal conversion) and its deexcitation via atomic states IC (internal conversion).

Ruchowska *et al.* [17] obtained $T^m_{1/2} = 10^{+18}_{-5}$ h for $E^m = 3.5 \pm 1.0$ eV with use of the reduced transition probability $B(M1) = 0.025\ \mu_N^2$ that was given by the quasiparticle-plus-phonon model (QPPM). That half-life is considerably longer than the present observation, but it is reasonable because the

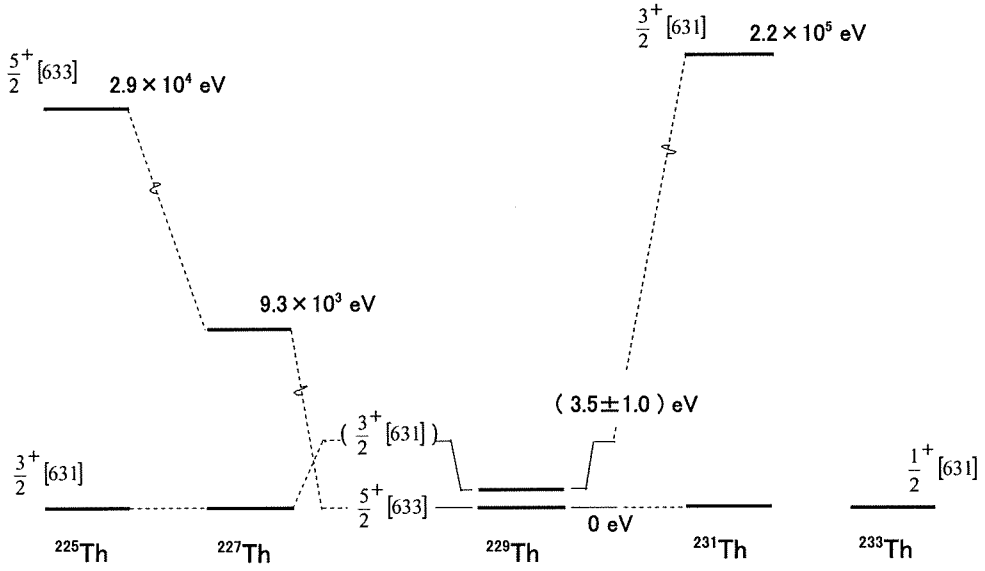

FIG. 10. Low-lying level systematics of odd-mass Th isotopes in the neighborhood of ^{229}Th.

isomer is likely to be populated by NEET and eventually there is IC that should be dominant in deexciting the isomer: these were not included in the QPPM calculation. As Karpeshin *et al.* [33,34] pointed out, IC from ^{229}Th^m could be three orders of magnitude larger than its direct nuclear transition. Our lifetime observation is in agreement with such a theoretical prediction. The present information on the isomer lifetime could serve to find which electron wave functions are dominant in NEET and IC associated with the isomer, and also to evaluate E^m theoretically.

From the systematics of Nilsson states $5/2^+[633]$ and $3/2^+[631]$ in the neighboring odd-mass Th isotopes, shown in Fig. 10, it is likely that the $3/2^+[631]$ state exists as the first excited state in ^{229}Th, although its energy is not yet known exactly. There is a characteristic feature of crossing of the $5/2^+[633]$ and $3/2^+[631]$ states at ^{229}Th. This is consistent with the Nilsson diagram for the neutron number $N > 126$ [35]. We expect that ^{229}Th^m is the Nilsson state $3/2^+[631]$ as first suggested by Kroger and Reich [1].

V. SUMMARY

We started the measurement by means of the hollow-cathode electric discharge, assuming the half-life of ^{229}Th^m to be 14 ± 3 h, a value given by Mitsugashira *et al.* [9] and cited by the National Nuclear Data Center (USA) [36]. But we have come to the conclusion that its half-life is much shorter. We have probably populated ^{229}Th^m by NEET during the electric discharge and found its half-life 1 min $\lesssim T^m_{1/2} \lesssim 3$ min. Based on our NEET condition, we are led to an excitation energy 3 eV $\lesssim E^m \lesssim 7$ eV. Had we been able to identify individual α lines of the decay of ^{229}Th^m, that would have certainly made our observation convincing. At present our observation seems to provide only possible evidence for NEET to ^{229}Th^m, and for its population relative to the ground nuclear state of the order of 10^{-2}.

ACKNOWLEDGMENTS

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RIKEN Gas-filled Recoil Ion Separator (GARIS) as a Promising Interface for Superheavy Element Chemistry—Production of Element 104, ^{261}Rf, Using the GARIS/Gas-jet System—

Hiromitsu Haba,*[1] Daiya Kaji,[1] Yukiko Komori,[2] Yuki Kudou,[1] Kouji Morimoto,[1] Kosuke Morita,[1] Kazuhiro Ooe,[2] Kazutaka Ozeki,[1] Nozomi Sato,[1] Atsushi Shinohara,[2] and Akira Yoneda[1]

[1]*Nishina Center for Accelerator Based Science, RIKEN, Wako 351-0198*

[2]*Graduate School of Science, Osaka University, Toyonaka, Osaka 560-0043*

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An isotope of element 104, rutherfordium (^{261}Rf), produced in the ^{248}Cm(^{18}O, 5*n*)^{261}Rf reaction was successfully extracted to a chemistry laboratory using a gas-jet transport system coupled to the RIKEN gas-filled recoil ion separator GARIS. The present system is a promising interface to explore new frontiers in superheavy element chemistry.

Chemical characterization of superheavy elements (SHEs) with atomic numbers $Z \geq 104$ is an extremely interesting and challenging subject in modern nuclear and radiochemistry.[1,2] A most important and interesting question is to clarify chemical properties of these newly synthesized heavy elements and to elucidate the influence of relativistic effects on chemical properties of these heaviest elements.[1,3,4] SHEs are produced in accelerators in heavy-ion-induced nuclear reactions. Extremely low production yields and short half-lives of SHEs force us to conduct rapid and efficient on-line chemical experiments with *single atoms*. Using gas-jet coupled chemistry apparatuses, chemical properties of SHEs have been studied for elements 104 (Rf) to 108 (Hs) and recently element 112.[1,2,5] At the same time, many of these successful experiments have clearly demonstrated the limitations of the applied techniques. Large amounts of background radioactivities from unwanted reaction products become unavoidable for SHEs with higher *Z*. High-intensity beams from advanced accelerators give rise to a problem in that the plasma formed by the beam in a target chamber significantly reduces the gas-jet transport efficiency. To overcome these limitations, the concept of physical preseparation of SHE atoms has been proposed.[1,6] The pioneering experiment with the recoil transfer chamber coupled to the Berkeley gas-filled separator (BGS) was very successful.[7] The isotope of ^{257}Rf physically separated from the large amount of β-decaying products was identified with a liquid scintillator after a liquid–liquid solvent extraction. However, the very short half-life of ^{257}Rf ($T_{1/2} = 4.7$ s) produced in the cold fusion reaction of ^{208}Pb(^{50}Ti, *n*) imposes stringent time limits on the gas-jet transport as well as the chemical separation.[8] In the RIKEN linear accelerator (RILAC) facility, the gas-jet transport system for the SHE chemistry was installed at the focal plane of the gas-filled recoil ion separator (GARIS).[9] The performance of the system has been investigated using ^{206}Fr ($Z = 87$), ^{245}Fm ($Z = 100$), and ^{255}No ($Z = 102$) produced in the ^{169}Tm(^{40}Ar, 3*n*), ^{208}Pb(^{40}Ar, 3*n*), and ^{238}U(^{22}Ne, 5*n*) reactions, respectively.[9,10]

In order to produce SHE nuclides with longer half-lives for chemical experiments, hot fusion reactions based on actinide targets such as ^{244}Pu and ^{248}Cm should be considered. However, very small recoil velocities of evaporation residues (ERs) produced by such asymmetric reactions cause serious problems in the operation of the gas-jet system coupled to the gas-filled separator. The transport efficiency of the separator drastically decreases with decreasing recoil velocity due to the multiple small-angle scattering in the filling gas. A vacuum window foil, which separates the gas-jet chamber from the separator, should be thin enough to allow ERs to pass though and has to withstand a pressure difference of ca. 1 bar. Since the last experiment with ^{255}No, we have developed a new gas-jet chamber having a large focal plane window of 100-mm diameter to efficiently collect ERs. A rotating target system for the use of a radioactive ^{248}Cm material was also installed. A chemistry laboratory was constructed behind the focal plane of GARIS, shielded with 50-cm concrete from the target room. In this work, the most desirable nuclide for Rf chemistry, ^{261}Rf ($T_{1/2} = 68$ s), produced in the very asymmetric ^{248}Cm(^{18}O, 5*n*) reaction was successfully extracted to the chemistry laboratory after the physical separation by GARIS.

A schematic of the experimental setup is shown in Figure 1. The $^{18}O^{5+}$ ion beam was extracted from RILAC. A $^{248}Cm_2O_3$ target of 280 μg cm^{-2} thickness was prepared by electrodeposition onto a 0.90 mg cm^{-2} Ti backing foil. The eight arc-shaped targets were mounted on a rotating wheel of 100 mm in diameter. The wheel was rotated during the irradiation at 1000 rpm. The beam energy was 95.5 MeV at the middle of the target, and the average beam intensity was 5 particle μA. GARIS was filled with helium at a pressure of 33 Pa. The magnetic rigidity of GARIS was set at 1.73 Tm. The evaporation residues of interest were separated in-flight from the beam and the majority of the nuclear transfer products by GARIS and then guided into the gas-jet chamber of 100-mm i.d. × 20-mm depth through a Mylar window of 0.5-μm thickness which was supported by a circular-hole (2.0-mm diameter) grid with 78% transparency. The ^{261}Rf atoms were stopped in helium gas, attached to KCl aerosol particles, and were continuously transported through a Teflon capillary (2.0-mm i.d. × 10-m length) to the rotating wheel apparatus MANON for α spectrometry. The flow rate of the helium gas was 2.0 L min^{-1}, and the inner pressure of the gas-jet chamber was 49 kPa. In MANON, the aerosol particles were deposited on 200-position Mylar foils of 0.5-μm thickness placed at the periphery of a 420-mm diameter stainless steel wheel. The wheel was stepped at 30-s intervals to position the foils between seven pairs of Si PIN photodiodes (Hamamatsu S3204-09). Each detector had an active area of 18 × 18 mm^2 and a 38% counting efficiency for α particles. The energy resolution was 60 keV FWHM for the detectors which look at the sample from the collection side. All events were registered in an event-by-event mode.