

## Search for a low-lying 3.5-eV isomeric state in $^{229}\text{Th}$

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**Abstract** We have extracted freshly produced  $^{229}\text{Th}$  atoms from 200 mg isotopically pure  $^{233}\text{U}$  in order to search for the radioactive decay of a 3.5 eV isomeric state in  $^{229}\text{Th}$ . A UV-sensitive photocathode, cooled and temperature stabilized within a controlled environment is used to search for the photons emitted in the radioactive decay of this state. No evidence of the state was found. From this experiment we have concluded that the half-life of the isomeric state  $^{229}\text{Th}^m$  must be either shorter than 5 minutes or longer than 115 days, assuming that the energy of the isomer is within  $3.5 \pm 1.0$  eV and that it is populated with a 2% branching ratio from the  $\alpha$  decay of  $^{233}\text{U}$ . Furthermore, we have set  $3\sigma$  limits to the possible branching ratio as a function of isomer half-life for a range of isomer state energies defined by the sensitivity of our system.

High-precision  $\gamma$ -ray spectroscopy revealed the existence of an unusually low-lying isomeric state of  $^{229}\text{Th}$ , at an excitation energy of  $3.5 \pm 1.0$  eV above the nuclear ground state [1]. Independent corroboration of the existence of a low-lying state has been achieved experimentally in the reaction  $^{230}\text{Th}(d,t)^{229}\text{Th}$ , placing an upper bound on the energy of the state at 7 keV [2]. Considering nuclear excitation energies are typically in the  $10^4 - 10^6$  eV range, this would be by far the lowest known nuclear excitation energy, and a unique case in which the energy is below the atomic ionization energy. This state, if confirmed, could be used in a number of interesting applications including the study of interactions between the nucleus and its environment and novel ways to achieve stimulated nuclear excitation [3].

Thorium-229 is produced in the alpha decay of  $^{233}\text{U}$ , with an estimated 2% of this decay populating the isomeric level through  $\gamma$ -ray transitions and internal conversions from higher levels [4]. The ground state of  $^{229}\text{Th}$  undergoes alpha decay with a half-life of 7880 years [5], while the half-life of the isomer for  $\alpha$ -decay is estimated to be about 2-4 times smaller [6]. Theoretical predictions estimate the radiative lifetime of the isomer to be of order a few hours [7, 8]. In addition, if the isomer excitation energy were found to

be greater than 6.3 eV, the ionization potential of  $^{229}\text{Th}$ , the nucleus would rapidly decay to the ground state via emission of internal conversion electrons. It has also been suggested that because of the very low excitation energy, the isomer half-life may vary with the chemical state of thorium [8].

A number of recent experiments [9-12] intending to detect de-exciting photons in the range of 300-500 nm have failed to confirm the existence of the isomer. These experiments highlighted the difficulties in attempting to distinguish radiative photons from direct nuclear de-excitation to fluorescence caused by contaminant daughters or from the alpha decay of uranium. A more recent publication by Browne *et al* [13] describes a method that does not require the detection of radiation from the isomeric decay. Rather they measure the growth in the disintegration rate of  $^{229}\text{Th}$  due to its delayed population from the 3.5-eV isomer. They conclude that the  $^{229}\text{Th}^m$  half-life must be either  $< 6$  hours or  $> 20$  days at a 99% confidence level.

This experiment was designed to test the validity and clarify discrepancies seen in recent publications. The aim of this experiment was to provide limits not only on the half life and energy of the isomer, but also on the branching ratio from the decay of  $^{233}\text{U}$ . The method involves extracting freshly produced  $^{229}\text{Th}$  atoms (and its 3.5-eV isomer) from 200 mg isotopically pure  $^{233}\text{U}$  ( $^{232}\text{U}/^{233}\text{U} < 10^{-9}$ ). The  $^{229}\text{Th}$  accumulated for 19.3 hours before being chemically separated from the uranium, and dissolved in 0.1M hydrochloric acid (HCl). The solution was stored in a 1.5 ml volume quartz cell for observation. By measuring the  $\gamma$ -rays the alpha activity of the source was inferred and, more importantly,  $^{229}\text{Th}$  activity in the sample could be confirmed from the presence of  $^{225}\text{Ra}$  K x-rays. The freshly produced thorium had a total  $\alpha$ -activity of  $22 \pm 5$   $\alpha/s$ . The presence of  $^{229}\text{Th}$  in the sample was checked by measuring its  $\gamma$ -ray spectrum with a  $2 \text{ cm}^2 \times 1 \text{ cm}$  Low Energy Photon Spectrometer (LEPS) system. The Ra k- $\alpha$  line of 88.47 keV was used to determine the  $^{229}\text{Th}$  activity.

The detection system was based on passively watching for the radioactive decay of the isomeric state with a UV-sensitive photon counting head, Hamamatsu H8259-01 series. The photon detector was placed in a hermitically sealed dark container as shown in Fig. 1. A slow bleed of nitrogen kept the container pressurized at 1-psi above atmosphere. The photon detector was cooled to  $\sim 5^\circ\text{C}$  and temperature controlled to  $0.1^\circ\text{C}$  with a combination of Peltier elements and a copper water jacket. This reduced the dark count rate of the detector from 80 counts/s at room temperature to an operating level of 10 counts/s. Exposure to a background photon rate as low as a few tens of kHz resulted in “settling” times of the photocathode of more than an hour. Therefore, to maintain stable conditions, the photon head was cooled and in operation for several hours prior to the experiment.

The spectral response of the photocathode covered a wavelength range from 185-850 nm, with both the anode and cathode sensitivity measured by Hamamatsu. The

measured cathode sensitivity of the detector exceeded the typical specifications quoted in the literature by a factor of  $\sim 1.2$ , and so the typical quantum efficiencies of the tube were taken as accepted values. Sensitivity to photons with a wavelength of 185 nm (corresponding to an isomer energy of 6.7 eV) was unattainable due to 100% absorption of photons with wavelengths of 200 nm and below in the quartz cell. The decision to use hydrochloric acid as the dissolving agent for the thorium resulted from absorption measurements as a function of wavelength in different solutions. By folding in the solid angle for detection,  $8 \times 10^{-3}$ , the photocathode quantum efficiency and the absorption of light both in the quartz and 0.1M HCl, the detection efficiency of the experiment as a function of photon energy can be calculated. The peak sensitivity of the detector is at energy of 4 eV (0.17% of all photons with this energy can be detected), reducing to 0.09% at 6 eV and 0.002% at 1.5 eV. For a decaying isomer emitting photons with energy 3.5 eV the detection efficiency is 0.17%.

The two most critical parameters of the experiment are as follows: firstly to ensure sensitivity to the shortest possible isomer lifetime, extraction of the thorium from the uranium column must be completed in a short a time as possible; secondly, the light level must be kept to a minimum during loading of the sample to reduce the exposure “settling time” of the photocathode. The time between extraction, loading into the system and acceptance of data was 22 minutes. The source was mounted on a pneumatic sled as shown in Fig. 1. Data were taken at a rate of 1 Hz, and every alternate minute the sled moved the source out of the viewing range of the photocathode for a background measurement. The experiment ran for a total of 22 hours, after which the background data was subtracted from the source data. Fig. 2 shows the raw data after background subtraction binned in 10-minute intervals. It is clear that the count rate is non-zero and photons emitted from the solution have been detected. A chi-squared analysis routine fitting an exponential decay to the data with free parameters yielded a reduced chi-squared value of 0.98, whilst fitting a straight line yielded a marginally different value of 0.99. The half-life extracted from the exponential fit agrees with the previously measured photocathode “settling” time of 40 minutes after minimum light exposure during sample loading. This value was confirmed with the same radioactive solution two days later after repeating the experiment. With the conclusion of having seen no radioactive decay, a range of possible exponential curves are fitted to the data with a constraint that the background must be greater than or equal to zero. This ensures that a realistic non-negative value is maintained for fluorescence induced in the solution from alpha decay.

Fig. 3 shows the results of the chi-squared analysis fits as a function of initial photon count rate versus isomer half-life. Each exponential fit to the data assumes that the minimum value of the reduced  $\chi^2$  occurs when the number of photons emitted at the start of the scan is zero, in other words assuming that there is no isomeric decay. For each chosen isomer half-life, limits can then be assigned to the initial photon count rate as

shown by the three curves in Fig. 3. From the known amount of uranium in the column and assuming a 2% branching ratio to the isomeric state, we can estimate the expected photon count rate as function of half-life, 22 minutes after extraction. The peak of the expected photon distribution as a function of both isomer half-life and energy (folding in the detection efficiency) is at 3 hours and 4-eV respectively. Assuming the emitted photons are not absorbed in the radioactive solution, we would expect to see a photon rate as high as 1.6 kHz after background subtraction.

This is clearly not the case as can be seen from the binned data in Fig. 2. Therefore by comparing the expected photon count rate with the results obtained from the experimental fits to the data, limits can be set on the branching ratio as a function of isomer half life as shown in Fig. 4. For the various isomer energies indicated, the region within the parabolic curves defined by the branching ratio and half-life excludes the  $^{229}\text{Th}^m$  at a confidence level of 99.7%. The displacement of the parabolas along the y-axis is simply attributed to the product of the photocathode quantum efficiency and the wavelength transmission through the quartz and HCl solution. In fact, the parabola for an isomer energy of 3.5-eV coincidentally overlaps with the parabola for an isomer energy of 4.5-eV. There is little difference in sensitivity in energy until that of 1.5-eV which is dominated by the rapidly decreasing photocathode sensitivity to light with wavelength greater than 800 nm. The dashed line indicates the position of the estimated branching ratio to the isomeric state from the decay of  $^{233}\text{U}$  [4].

The possibility exists that photons emitted from the decay of the isomer are absorbed in the solution. This has been tested with a 1 nCi  $^{56}\text{Co}$  gamma ray source fixed to an inorganic  $\text{BaF}_2$  scintillator. The crystal scintillates with a fast component at a maximum peak wavelength of 220 nm, and a slow component at a peak wavelength of 310 nm [14]. With no radioactive solution between the scintillator and detector an average count rate of 87 counts/s was measured. With the thorium solution between the detector and scintillator the signal size decreased to an average of 73 counts/s. This reduction in signal is attributed to the absorption of light in the surfaces of the quartz sample cell demonstrating that the solution does not absorb ultraviolet light.

In conclusion, the results of our search for the low-lying isomer of  $^{229}\text{Th}$  have limited the half-life of the isomer to be either shorter than 300 seconds or longer than 115 days at a 99.7% confidence level. This assumes the energy of the isomer is at  $3.5 \pm 1.0$  eV above the nuclear ground state [1] and that it is populated with a 2% branching ratio from the decay of  $^{233}\text{U}$ . Furthermore, by relaxing the assumption of a fixed branching ratio we have plotted the distribution of branching ratio as a function of isomer lifetime at the 3 $\sigma$  confidence level for a range of energies defined by the sensitivity of our system. A more abundant amount of  $^{233}\text{U}$  would make this experiment sensitive to shorter half-life values of  $^{229}\text{Th}^m$ , however it is believed that no significant gains can be made on the speed of the extraction process.

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### Figure Captions

**Fig. 1.** Schematic of the thorium fluorescence detection system.

**Fig. 2.** Photon counts detected in the system after background subtraction and subsequent binning of the data.

**Fig. 3.** Fitting the raw data with an exponential decay as a function of  $^{229\text{m}}\text{Th}$  half-life yields values for the initial photon count rate. The assumption that the minimum value of the reduced chi-squared occurs when the initial photon count rate is zero enables statistical limits to be set on the initial rate as shown by the three curves.

**Fig. 4.** Using the results calculated from the chi-squared analysis routine and by comparing to the expected photon count rate from the known starting amount of  $^{233}\text{U}$ , limits can be set on the branching ratio as a function of isomer half life for the energy range accessible to the system.

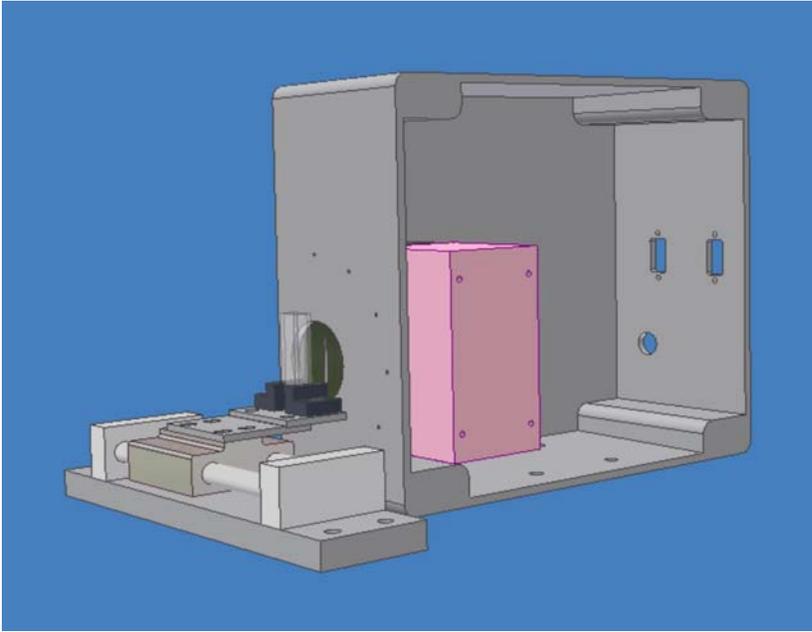


Fig. 1

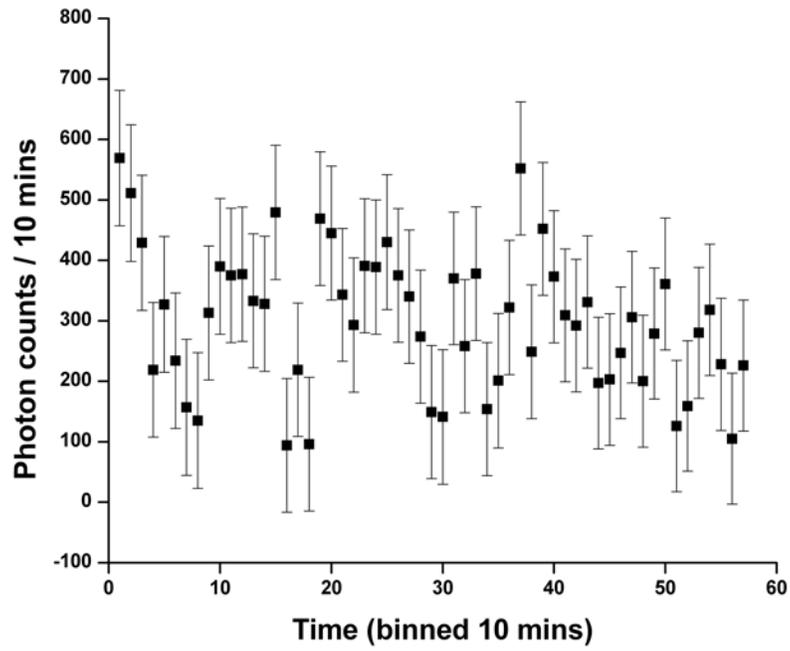


Fig. 2

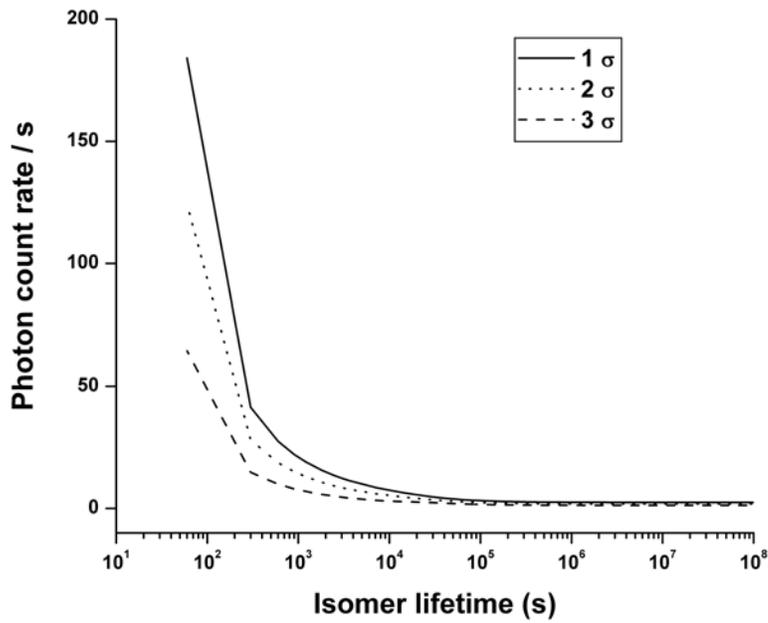


Fig. 3

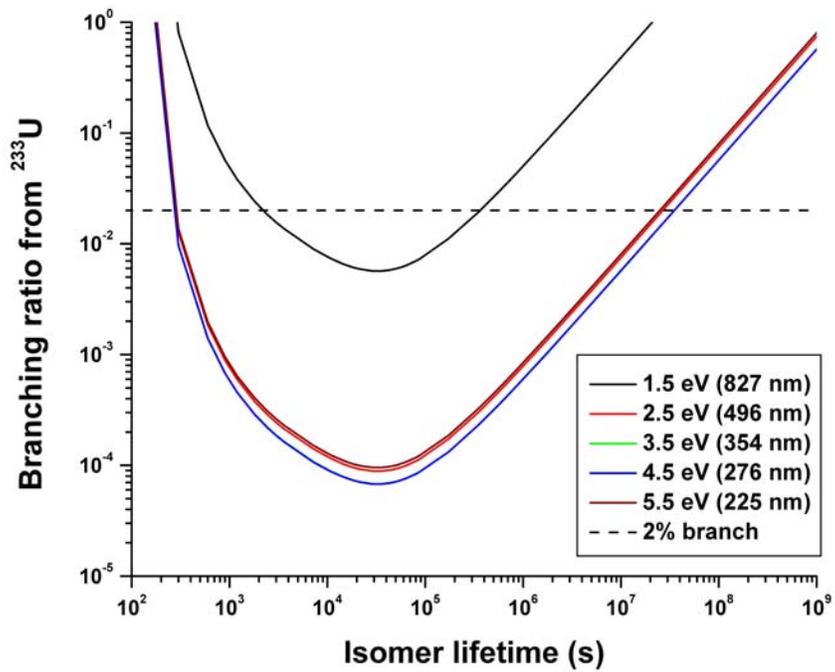


Fig. 4