

# A Nuclear Transition in the Optical Range: Search for the 3.5 eV Gamma Emission from Thorium-229m

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**Abstract.** The reported photon emission from relaxation of the 3.5 eV excited nuclear state of Th-229 (1,2) has been proven to be incorrect (3-6). The status of current experiments to determine the energy of this nuclear isomeric state is described, and plans for a more direct excitation of the state are discussed.

## INTRODUCTION

An anomalously low-energy, excited level for the  $^{229}\text{Th}$  nucleus was reported some years ago by Helmer and Reich (7). Analysis of gamma transition energies to the ground and first excited states in  $^{229}\text{Th}$  yielded the range of 3.5  $\pm$  1.0 eV for the energy of this level (7). Four research teams have published results of optical measurements of  $^{229}\text{Th}$  in the last three years. The low energy isomeric level of  $^{229}\text{Th}$  is populated approximately 2% of the time in the alpha decay of  $^{233}\text{U}$ . A structured photon emission for a sample of  $^{233}\text{U}$  in the near UV optical range was observed by two groups in 1997 and 1998 (1,2). The  $^{229}\text{Th}$  basis for these optical spectra was questioned by Young et al. (3) in 1998, and Utter et al. (4), Shaw et al. (5) and Young et al. (6) in 1999. Alternative explanations for the spectral emission from  $^{233}\text{U}$  samples were proven in the later work: namely, the near-UV emission is from nitrogen in the air (5), luminescence of uranyl ions (6), and fluorescence of the silica tube (6), all excited by the respective sample alpha activity.

The anomalously low-energy isomeric level in  $^{229}\text{Th}$  has generated considerable interest because such a level should have some unusual properties, such as for example, an optical range gamma ray and an accompanying electron bridge decay route

(8). The electron bridge raises the possibility of a highly effective nuclear excitation by optical photons (9); this could lead to a change in the alpha decay rate of  $^{229}\text{Th}$  via population of the 3.5 eV level under low intensity laser radiation (10).

The decay channels of the 3.5 eV level and the half-life should depend strongly on the chemical environment (8). In an isolated Th atom, the 3.5 eV level should decay predominantly via the electron bridge channel (8). This is a third order process, which is essentially unobserved in other nuclear transitions. Within a metal sample, nonradiative decay via conduction electrons should dominate (11). This second order one-photon process is the inverse of the process of inelastic electron scattering by nuclei and can be viewed as the analog of internal conversion for these conduction electrons. In a wide energy-gap dielectric, such as  $^{229}\text{ThO}_2$ , the first order process of nuclear gamma emission should be possible (12,13).

The 3.5 eV level in  $^{229}\text{Th}$  is a "bridge" between low energy nuclear physics and many other fields of physics, such as optics, laser physics, solid state physics, physics of surfaces, and nuclear isomer interactions.

## **EXPERIMENTAL**

Earlier experimental work concerned with locating the energy position of Th-229m emission was reported at the last RIS conference (4). Further emission studies of various compounds of isotopically pure  $^{233}\text{U}$  (< 0.1 ppm  $^{232}\text{U}$ ) have been carried out. Weak emissions were recorded over the 250 to 1000 nm wavelength range. The samples have included  $^{233}\text{U}_3\text{O}_8$ ,  $^{233}\text{UO}_3$ ,  $^{233}\text{UO}_4 \cdot 2\text{H}_2\text{O}$  and a  $^{238}\text{UO}_2(\text{NO}_3)_2$  solution spiked with  $^{225}\text{Ac}$ . The last solution was prepared to have an alpha activity equivalent to that for a 10 ppm  $^{232}\text{U}$  impurity. The samples were contained in one of three types of  $\text{SiO}_2$  tubes. Commercial grade  $\text{SiO}_2$  from an unidentified vendor was used for the first tests. Later Suprasil II or S-300 silica (Technical Glass Products, Mentor, OH) was used. The first two samples were sealed in the sample tubes under vacuum after flushing with He. The uranyl peroxide and nitrate solution samples were sealed in air with a teflon ball fixed into place with Torr Seal epoxy (Varian Vacuum Products, Lexington, MA). Details of the emission spectroscopic instrument have been described previously (4,5,6). The sample image was focused on the entrance slit of a Spex polychromator (Model HR 460); a 150 groove/mm or in a few cases a 1500 groove/mm diffraction grating, blazed at 500 nm, was the dispersing element in the spectrometer. The dispersed image was focused onto a 1024 by 256 pixel, liquid nitrogen-cooled, UV coated, CCD detector (Model EEV CCD15-11). All of these items were available from Instruments SA, Edison, MA. Data were collected via Spectramax (version 1.1D), a DOS program also available from Instruments SA. The program was modified to allow multiple averaged exposures or a single exposure for periods of time up to 4 days.

## RESULTS AND DISCUSSION

We have carried out emission spectral studies of several  $^{233}\text{U}$  oxides and have seen no reproducible emission that could be attributable to a gamma ray emission. The oxides have included  $\text{UO}_4 \cdot 2\text{H}_2\text{O}$ , a compound thought to be the sample (1) that was used in the original study and which gave a structured emission in the ultraviolet region. This structured emission was later identified as  $\text{N}_2$  and  $\text{N}_2^+$  emission in air, excited by alpha radiation from the sample (5,6). A broad visible emission in the 520 nm region was also observed for this sample (1,2); that was later proven to be  $\text{UO}_2^{++}$  luminescence (6).

The various uranium oxide samples we have studied and the emission results observed are summarized in Table 1. As can be seen from the table, emission from the  $\text{SiO}_2$  container was seen in all the samples. Of note is that an atomic oxygen line (O I) was observed for the uranyl peroxide sample,  $\text{UO}_4 \cdot 2\text{H}_2\text{O}$ . This sample was sealed in air, but so was a sample of  $\text{UO}_3$ . Atomic oxygen was not observed in the latter sample. It would appear that O I is generated in the  $\text{UO}_4$  sample, being a transient species coming from the radiation-induced decomposition of the  $\text{O}_2 =$  moiety. Our earlier observation of  $\text{N}_2$  emission (5), originating in the air above the sample, was seen above either  $\text{UO}_4 \cdot 2\text{H}_2\text{O}$  or  $\text{UO}_3$  when they were sealed in air, but was not observed when imaging only the solid in these cells.

**TABLE 1.** Summary of  $^{233}\text{U}$  Oxide Emission Experiments<sup>(a)</sup>

Compound	Color	Sample atmosphere	Type of $\text{SiO}_2$ container	Comments
$\text{UO}_4 \cdot 2\text{H}_2\text{O}$	orange	air	Commercial $\text{SiO}_2$	$\text{SiO}_2$ emission O I emission
$\text{UO}_3$	yellow	air	Suprasil II	$\text{SiO}_2$ emission
$\text{UO}_3$	yellow	vacuum	Suprasil II	$\text{SiO}_2$ emission
$\text{U}_3\text{O}_8^{(b)}$	black	vacuum	Commercial $\text{SiO}_2$	$\text{SiO}_2$ emission 314 nm emission
$\text{U}_3\text{O}_8^{(b)}$	black	vacuum	Suprasil II	$\text{SiO}_2$ emission
$\text{U}_3\text{O}_8^{(b)}$	black	vacuum	S-300	$\text{SiO}_2$ emission

<sup>a</sup> All spectral studies involved averaged 1 hour integrations for more than 18 hour total.

<sup>b</sup>The identical  $\text{U}_3\text{O}_8$  powder sample was used in all three studies.

An apparently artifactual emission was seen at 314 nm in one sample of  $^{233}\text{U}_3\text{O}_8$  sealed in an evacuated commercial  $\text{SiO}_2$  container. The emission was sharp, but not

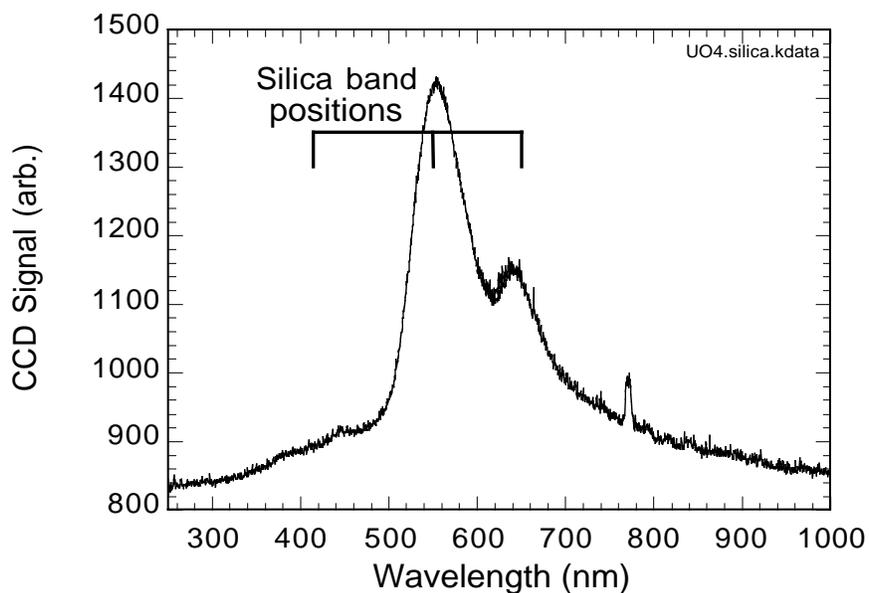
slit-limited when observed using a 0.2 mm entrance slit in the polychromator. As noted in the table, when that same sample powder was observed in a sealed evacuated Suprasil II or S-300 (4 ppm OH concentration) sample container, the emission was not observed. Based on chemical analysis of that  $U_3O_8$  sample, it contained, perhaps, unusual amounts of Al, (20 wt %); Pt, (2 wt %); and Nd, (5%). The amount of Al and Pt is unusually large and was thought to be present because of the sampling handling. None of these three elements, however, would appear to be the basis for the single emission peak seen, however. Keep in mind, further, that  $U_3O_8$  is visually black and would therefore be expected to absorb UV photons.

In all experiments with long-term, multi-hour, observations,  $SiO_2$  emissions were seen. These emissions were most probably excited by alpha decay from  $^{233}U$ ,  $1.6 \times 10^5$  year half-life. A typical  $SiO_2$  emission spectrum is shown in Figure 1. This spectrum corresponds to a sample of  $^{233}UO_4 \cdot 2H_2O$  sealed in air in a commercial silica tube. Note the broad emission peaks at 415, 550 and 650 nm. The energy position of these three bands correspond to the three bands reported for the cathodoluminescence of silica (14). The relative intensities of these bands do not match those seen in that study. They are, however, consistent with the view that they arise from silica (15); the relative intensities would be expected to vary with composition of silica and the type of excitation source. Note that the most intense  $SiO_2$  peak is seen near 550 nm for this sample and silica type.

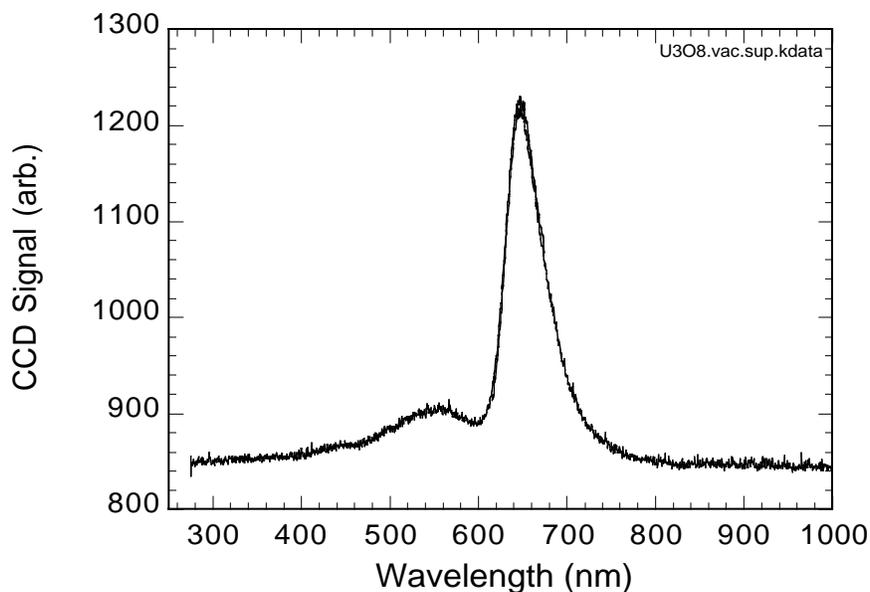
The emission spectrum of Suprasil II  $SiO_2$  is shown in Figure 2. Note that the emission in the 420 nm region is almost nonexistent; the peak at 550 nm is weak; and the most intense peak is at 620 nm. The sample in this case was  $^{233}U_3O_8$  sealed in a vacuum. A similar emission spectrum was recorded for  $^{233}UO_3$  in an evacuated Supersil II tube.

As pointed out in Table I, an unidentified emission peak was observed at 314 nm in one sample of  $^{233}U_3O_8$  sealed in an evacuated commercial  $SiO_2$  tube. In this sample of  $U_3O_8$ , an emission was also seen in the 620 nm region, but a more intense broad emission was seen that extended from 350 to 600 nm. Whether the composition of this tube was the same as that used in the study shown in Figure 1 is unclear. As noted in the table of results, however, when this same sample was later contained in an evacuated Supersil II tube, the emission peak at 314 nm was not observed. Further the emission spectrum of  $SiO_2$  seen was essentially identical to that seen in Figure 1. It seems to be established that the emission of Suprasil II is reproducible with  $^{233}U$  sample excitation.

The same sample of  $^{233}U_3O_8$  was next transferred to and sealed in an evacuated S-300  $SiO_2$  tube. As noted earlier, S-300 contains a very low concentration of OH radicals (4 ppm). In our loading process, the  $SiO_2$  was sealed by heating in a gas flame. We had hoped that the emission at 620 nm, thought to be related to OH radicals, would be minimal. This was not the case and the assumption can be made



**FIGURE 1.** Emission spectrum of  $^{233}\text{UO}_4 \cdot 2\text{H}_2\text{O}$  sealed in a commercial grade  $\text{SiO}_2$  tube. The data was obtained by averaging one hour observations totaling more than 18 hours. The  $\text{SiO}_2$  emission band positions (14) are indicated. The sharp emission at 777 nm is due to O I



**FIGURE 2.** Emission spectrum of  $^{233}\text{U}_3\text{O}_8$  in an evacuated Suprasil II  $\text{SiO}_2$  tube. The data was obtained by averaging one hour observations totaling more than 50 hours.

that sealing such SiO<sub>2</sub> with a gas flame introduces OH into the SiO<sub>2</sub>. The peak at 314 nm was not seen. It was only observed in a commercial SiO<sub>2</sub> tube and must be considered to be the result of that material.

## CONCLUSIONS AND FUTURE WORK

No UV-visible emission has yet been observed that can be attributed to a gamma emission from <sup>229m</sup>Th when that isomeric state is generated from <sup>233</sup>U alpha decay. We have, however, observed other emissions from various <sup>233</sup>U compounds, and most of these can be ascribed to SiO<sub>2</sub> emissions excited by alpha decay of the sample contained therein.

A different approach is needed to investigate the presence of the isomeric state of <sup>229</sup>Th. Proposals for a better experiment have been suggested in the literature by Tkalya (12,13) and in an presentation by Young and Shaw (16). The approaches are similar in several respects. Both choose to study <sup>229</sup>ThO<sub>2</sub> as the sample material, probably as a dried rather than as an ignited oxide. ThO<sub>2</sub> is transparent in the region below 6 eV (17). Both approaches would use a synchrotron source to excite the isomeric state from ground state <sup>229</sup>Th. Both would look for the gamma emission from the isomeric state as the goal of the study. A theoretical basis for this approach has been presented by Tkalya (12). He proposed to excite the intermediate nuclear levels 5/2+ (29.19 KeV) and 7/2+ (71.78 KeV) by monochromatic synchrotron radiation. From the 5/2+ level, then, the isomeric state could be populated and observed. In the Young and Shaw approach (16), the <sup>229</sup>ThO<sub>2</sub> contained in an evacuated Suprasil II container would be excited by broad band synchrotron energy from 4.5 to 2.5 eV, or broader if necessary. Observation of the gamma emission would be observed after an irradiation time or between excitation cycles. Once the emission has been identified, an attempt to excite the isomeric level with a carefully tuned UV laser could be made. Much useful information about isomeric nuclei and methods of excitation can be realized if laser coupling can be accomplished from the isomeric state that nature has provided.

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