Reexamination of the Optical Gamma Ray Decay in ^{229}Th

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(Received 3 June 1998)

Optical measurements of a clean, 2-μC ^{233}U sample were made to verify light emission from gamma ray decay of the first excited nuclear level in ^{229}Th. The results showed that the light observed in earlier studies was likely to be caused by alpha-particle induced fluorescence of air. *In vacuo,* no light emission was discernable. The ^{229}Th system, therefore, does not appear to provide the level of access for studying atomic-nuclear interactions suggested by the previous measurements. [S0031-9007(98)08232-5]

PACS numbers: 23.20.Lv, 27.90.+b, 32.30.Jc

Interactions between the nuclear and atomic systems have attracted considerable attention because of the potential of stimulating nuclear transitions by laser excitation of atomic states. In the inelastic bridge mechanism, for example, nuclear deexcitation is coupled to atomic excitation. In particular, an electromagnetic nuclear transition couples to an atomic electron, which is elevated to an excited state, thereby reducing the energy of the final gamma ray [1–3]. The mechanism suggests that the inverse may be possible, i.e., that atomic deexcitation may induce nuclear excitations [4]. Nuclear excitation energies, however, are generally much larger than atomic excitation energies. The identification of cases where the energies are comparable is, therefore, of highest importance.

The nuclear spectroscopy of high-energy gamma rays from ^{229}Th produced in the alpha decay of ^{233}U suggests that the ground state is a closely spaced doublet separated by 3.5 eV [5–7]. The energy of the first excited level, thus, is lower than any other known excited nuclear level. Recently, Irwin and Kim reported the detection of photon emission resulting from the deexcitation of the ^{229}Th isomer [8]. The energy of the observed photons was determined to be about 3.5 ± 1.0 eV, in agreement with the value inferred indirectly from the nuclear spectroscopy of the high-energy gamma rays. Moreover, one of two samples of ^{233}U studied showed a second photon peak near 2.4 eV [8]. The observation of this peak was consistent with predictions that the inelastic bridge mechanism could cause excitation of the thorium atom from the $6d_{3/2}$ ground level to the $7p_{1/2}$ excited level. The Letter suggested that ^{229}Th provides a perfect isotope for future studies of low-energy nuclear-atomic interactions since the energy spacing is well within the realm of present-day laser capabilities.

A subsequent study by Richardson *et al.* [9] also observed emission in the UV. Employing a higher resolution spectrograph than Irwin and Kim, they were able to discern three peaks near 3.5 eV. Their result, thus, appeared to confirm the measurement of Irwin and Kim.

In the following we present experimental studies of the optical emission of a clean sample of ^{233}U decaying to ^{229}Th. We found no evidence for the optical emission attributable to the deexcitation of the predicted low-lying ^{229}Th isomer. We found light caused by alpha-particle induced fluorescence of air. In the absence of air, no light emission was discernible. Since the observation by Irwin and Kim was conducted in air, our results suggest that their observation is not that of a nuclear gamma ray, but that of air fluorescence. The ground state doublet of ^{229}Th, therefore, is an even more difficult experimental testbed for studying nuclear-atomic interactions than suggested by the earlier measurements.

The present measurements were carried out at the Lawrence Livermore National Laboratory. A 2-cm-diameter sample of pure ^{233}U was prepared in-house by electroplating a thin layer of uranium (99.92% ^{233}U) on a 2.5-cm-diameter platinum disk. The uranium was electroplated from an isopropyl alcohol solution containing about 5% 0.1 N HNO_3. The total activity of the sample was 2 μC, resulting in an optically thin layer approximately 35 Å thick. Flaming, i.e., heating by an open flame, was used to fix the uranium to the substrate and to convert the uranium deposit to the oxide. Although our sample was considerably less active than those used by Irwin and Kim, any higher activity levels would not necessarily increase the amount of optical gamma rays detected because of the higher opacity for thicker samples.

The source disk was mounted to a holder using four small screws with washers. Only a small portion of each washer lay over the edge of the platinum disk so as to not make contact with the uranium. A picture of the arrangement is shown in Fig. 1(a). The picture was taken with a charge-coupled device (CCD) camera, which was also used to measure the light emission from the sample. The camera employed a Nikon ultraviolet (quartz) objective with f/4.5 and a 105 mm focal length. The detector consisted of a 2.5×2.5 cm^2 back-illuminated, cryogenically cooled, CCD with 25×25 μm^2 pixels. Binning of

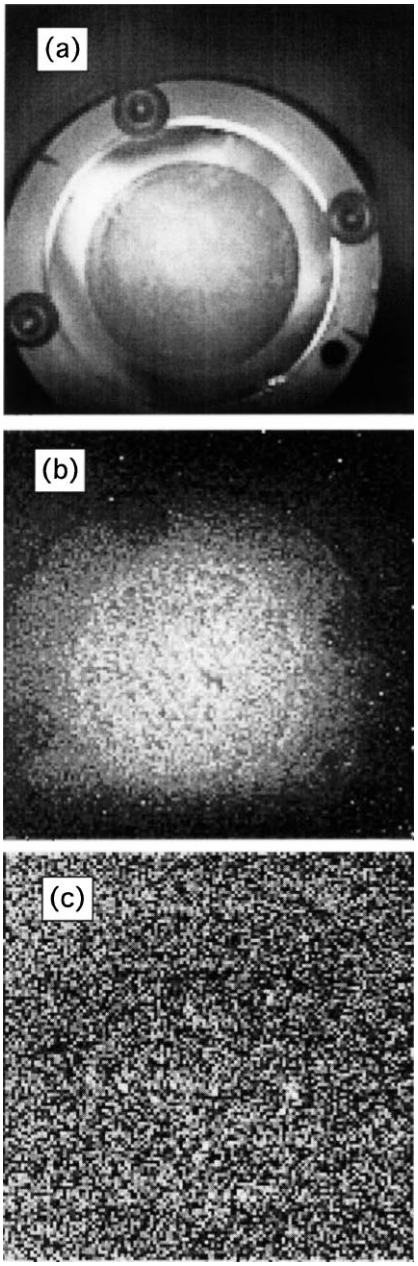

FIG. 1. Images of the ^{233}U sample: (a) Visible light image showing the 2-cm-diameter sample on a 2.5-cm-diameter Pt disk held by a mounting ring; (b) image of the photon emission generated by the sample in air; (c) image of the photon emission generated by the sample in vacuum. The intensity scale in (c) has been magnified by a factor of 5 with respect to (b). Without this rescaling (c) appears totally black. For all three images the arrangement shown in Fig. 2 was used. The images were binned to create an effective grid of 128 × 128 pixels each 200 × 200 μm^2 in area.

the pixels to an effective grid of 128 × 128 was used to increase the signal to background ratio.

A direct exposure of the ^{233}U sample immediately verified light emission, as illustrated in Fig. 1(b). All external light leaks were eliminated by placing the source inside a stainless steel vessel and sealing any light leaks with

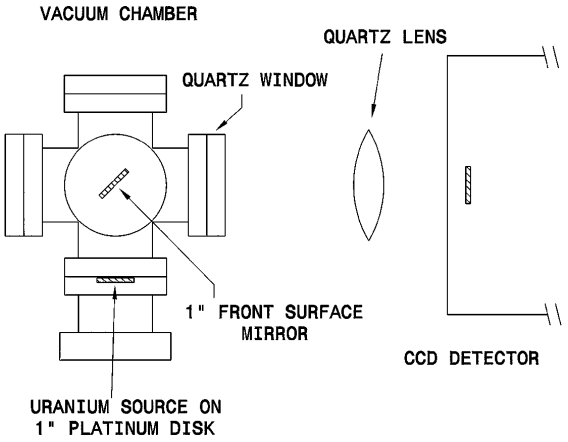

FIG. 2. Experimental arrangement for detecting light emission from the ^{233}U sample.

black tape and cloth. Tests for light leaks were made with the room lights on or off. The 30-min measurement in Fig. 1(b) clearly showed light coming from the direction of the source. In fact, enough light was produced to discern the outline of the screws and holder. It is curious to note that light did not only seem to emanate from the ^{233}U sample itself, but also from the rim of the platinum disk. The images in Fig. 1 were recorded using the mirror arrangement shown in Fig. 2 and decribed below. Stronger light emission was seen when looking at the sample along a direct line of sight.

In order to investigate the possibility that the light was produced by alpha-particle induced fluorescence, or that parts of the sample were illuminated by reflection of light off the inside walls of the vessel, we placed the source at the entrance of one of the ports of a six-way cross vacuum chamber located 90° from the camera port. To view the source a 2.5 × 2.5 cm^2 front coated mirror was mounted in the center of the chamber at a 45° angle with respect to the source, as illustrated in Fig. 2. In this manner any light emitted from the source would reflect on the mirror and be detected by the camera. Source alpha particles impinge only on the metallic surface of the mirror, or pass through to the port cap behind the mirror. This eliminates the possibility of induced fluorescence of the quartz window.

While Fig. 1(b) represents the image of the sample in air, Fig. 1(c) represents the same image in vacuum (about 20 mtorr). Again, a 30-min exposure time was used. As the figure shows, the light has disappeared. A quantitative analysis of images taken in air showed that photons were detected at a rate of 6.30 ± 0.15 (counts/bin)/min in the "brightest" region of the image, whereas in the "darkest" region the rate was 5.04 ± 0.13 (counts/bin)/min. This suggests a rate of about 1.26 (counts/bin)/min above the background. Analyzing the same regions of the images taken in vacuum the count rates were found to be 5.02 ± 0.14 (counts/bin)/min and 5.00 ± 0.09 (counts/bin)/min, respectively, for a difference of only 0.02 (counts/bin)/min: less than the statistical error of the measurement.

The comparison clearly shows that the light observed was caused by the alpha-particle induced fluorescence of air. This difference amounts to a rate of only 1.1 counts/sec at the detector. Considering the total collection efficiency of the system, the 2 μC source strength, and the $\sim$2% branching ratio to this isomeric state [10], a count rate of 4.6 counts/sec would be expected at the detector. It should be noted that much of the statistical error in the measurement arises from the readout of the CCD and therefore would be effectively reduced during longer exposures with the same binning factors. However, we show below that doing so will not actually improve the identification of the optical gamma ray emission.

Measurements of the spectral distribution of light caused by fluorescing air have been made before. A measurement of the fluorescence of N_2 and air by Davidson and O'Neil in the optical showed strong peaks at 3371, 3537, 3577, 3756, 3805, 3914, 3998, 4059, 4270, and 4278 Å [11]. These peaks match the peaks shown by Irwin and Kim and measured with their smaller-slit, higher-resolution spectrograph [8]. Most importantly, the strongest two peaks observed by Irwin and Kim at 3.50 and 3.70 eV match the stronget two peaks observed by Davidson and O'Neil at 3371 and 3577 Å, respectively. Similar results were also obtained in Refs. [12,13]. Since Irwin and Kim conducted their observations in air [14], our analysis and experiments strongly suggest that their observation is due to air fluorescence and not nuclear gamma radiation.

The measurements of Richardson *et al.* [9] were made using a liquid ^{233}U source. Fluorescence of the liquid was taken into account by subtracting a spectrum obtained from a liquid ^{232}U source. The energy of their measured UV peaks are at 3.50 $\pm$ 0.04, 3.71 $\pm$ 0.05, and 3.96 $\pm$ 0.09 eV, which again match the strongest flourescence peaks of air. In fact, the energies of the band head for the N_2 second positive system $C^3\Pi \rightarrow B^3\Pi$ are 3.46, 3.67, and 3.92 eV for the 0-1, 0-0, and 1-0 vibrational transitions, respectively [15]. This suggests that differences in the fluorescence of air in the space above the liquid sample was not taken into account.

One might argue that vacuum measurements with considerably longer exposure times than discussed above could discern the light emission of optical gamma rays from ^{229}Th, especially since there is a hint that there are more photon counts observed from the sample area than from other areas such as the holder frame and since the statistical error of the measurement could be reduced. Such a measurement is not possible for the following reason. Unless the sample is prepared *in situ* and in an ultraclean environment, there will always be monolayers of "dirt" on top of the ^{233}U [16]. This layer may fluoresce and mask any light emitted in the gamma ray decay of the first excited ^{229}Th level. To illustrate this, we present in Fig. 3 an 8-h exposure with the full 1024 $\times$ 1024 pixel spread of the CCD detector. The exposure clearly showed "hot spots" on the image of the ^{233}U sample, i.e., regions which were much brighter than the surrounding area. These hot spots were likely related to dust particles falling onto the surface during an opening of the system to check the alpha-particle count rate of the sample. Bathed in alphas, the dust specks fluoresce strongly. Some of these fluorescing specks are located outside the ^{233}U deposition area, which proves clearly that this light is not emitted by the radioactive material itself. We expect that other surface contaminants will radiate as well masking the potential light emission from the purported gamma ray decay. In fact, the count rate in the same "bright" region of the detector considered above, but excluding obvious hot spots, resulted in rates of 0.466 $\pm$ 0.006 (counts/bin)/min. A rate of 0.465 $\pm$ 0.006 (counts/bin)/min was found in the corresponding "background" region. Taking the difference of these two values and scaling by a factor of 64 to account for the different bin sizes resulted in a similar excess as above, i.e., 0.08 (counts/bin)/min. This is, however, hardly evidence for the existence of gamma ray emission given the possibility of fluorescence of surface contaminents. Irwin and Kim observed an intense spectral feature near 2.5 eV from one sample and only a very weak feature at this energy from the other [8]. While the latter is consistent with fluorescence of air [11], the former is not and we do not know its origin. A possibility is that it may have been the result of different surface or sample contaminants, especially since the two samples used by Irwin and Kim were of unknown chemical composition and visually looked different.

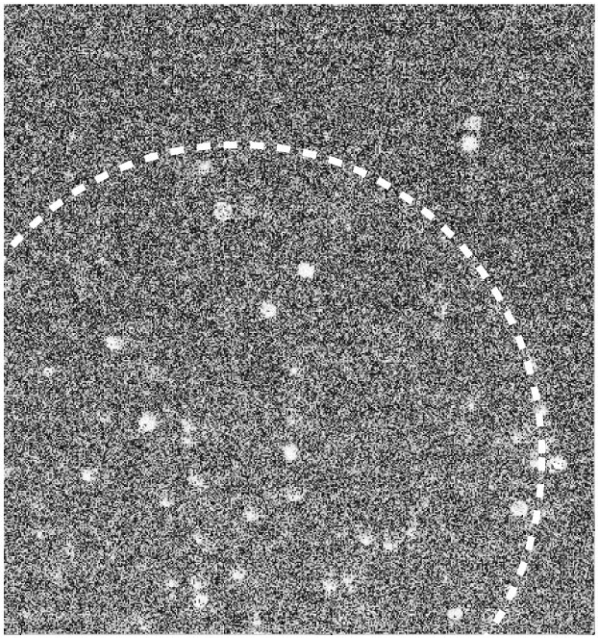

FIG. 3. Long time-exposure image of the ^{233}U sample. The exposure was taken in vacuum and shows hot spots attributed to alpha-particle induced fluorescence of dust specks on the sample. The rim of the ^{233}U-covered area is indicated by the dashed line.

In conclusion, we note that our studies of ^{233}U decaying to ^{229}Th confirmed Irwin and Kim's [8] observation of light emission. We showed that the observed light is caused by alpha-particle induced fluorescence of air. No significant light emission was discernible *in vacuo,* although light emission was found from isolated surface contaminants, which we believe to be alpha-particle induced fluorecence of dust specks. Because of the possibility of fluorescence of surface contaminants, the direct detection of optical gamma ray decay from ^{229}Th appears difficult unless samples can be prepared in an ultraclean environment. Previous studies of the fluorescence of air suggest that the lines reported at 3.5 eV correlate to the nitrogen emission lines. The approach used by Richardson *et al.* attempted to eliminate fluorescence by subtracting intense spectra from two nearly identical samples with different uranium isotopes [9]. This approach requires that even small differences, such as sample volume and gas composition in the air space, are avoided. Evidently, without the detection of any light attributable to the purported gamma ray decay, inferences of the energy of the first excited level useful for studies of atomic-nuclear coupling are not possible. The problem of direct detection of the decay of the first excited level in ^{229}Th, unfortunately, remains unresolved and requires further study to establish a reliable experimental signature of this elusive level.

This work was performed under the auspices of the Department of Energy by Lawrence Livermore National Laboratory under Contract No. W-7405-ENG-48.

Note added.—G. Tungate of the University of Birmingham informed us that he and his colleagues have confirmed our results concerning the 3.5 eV feature. Using an alpha source, they showed that the lines reported by Richardson *et al.* [9] and Irwin and Kim [8] appearing approximately at 3.5 eV are most likely to have come from alpha-induced fluorescence of atmospheric nitrogen (both N_2 and N_2^+). However, the same measurements show no evidence for lines at 2.5 eV.

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