Observation of Electromagnetic Radiation from Deexcitation of the ^{229}Th Isomer

G. M. Irwin
Department of Physics, Idaho State University, Pocatello, Idaho 83209

K. H. Kim
Department of Physics, North Carolina Central University, Durham, North Carolina 27707
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Experimental measurements of ultraviolet and visible emission from two samples containing ^{233}U suggest the deexcitation of a reported 3.5 eV isomeric level in ^{229}Th. The unique phenomenon of ultraviolet γ-ray emission was observed from this level. Results are consistent with theoretical arguments suggesting that the nucleus can also deexcite via an inelastic electronic bridge mechanism. These observations indicate the exotic phenomenon of nuclear emission in the optical band, and open new areas of physics related to nuclear-atomic interaction. [S0031-9007(97)03785-X]

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High precision γ-ray spectroscopy measurements utilizing ^{233}U sources have suggested the existence of a low-lying $I^\pi = \frac{3}{2}^+$ level (assigned $\frac{3}{2}$ [631] Nilsson state) in the nucleus ^{229}Th only 3.5 ± 1.0 eV above the $I^\pi = \frac{5}{2}^+$ ground state (assigned $\frac{5}{2}$ [633] Nilsson state) [1]. These measurements involved taking appropriate sums and differences of γ-ray energies of tens and hundreds of keV observed from ^{229}Th following α decay of the ^{233}U nucleus. This low excited state is the lowest known of all nuclei, and is of importance as it represents the only case in which nuclear excitation energies are similar to atomic or molecular excitation energies, suggesting nuclear deexcitation by emission of optical photons via an $M1$ transition. This level opens unique possibilities for studies of atom-nuclear interactions including nuclear excitation via atomic electrons. The purpose of this Letter is to report the results of experimental measurements designed to observe the emission of optical photons from samples of ^{233}U.

The low-lying excited state in ^{229}Th is populated approximately 2% of the time in the α decay of ^{233}U through γ-ray transitions and internal conversions from higher levels [2]. Theoretical considerations have suggested that such a low-lying nuclear level can deexcite via an inelastic electronic bridge mechanism [3], which has been observed as a small effect in the decay of the 30.7 keV ($T_{1/2} = 13.6$ yr) level in ^{93}Nb, which deexcites mainly by internal conversion of an $M4$ transition [4]. Calculations of the branching ratio between the direct γ-ray emission and the electronic bridge mechanism for ^{93}Nb agree with the experimental results to within an order of magnitude [5]. The electronic bridge mechanism has also been observed in the decay of the 80.3 keV isomeric level in ^{193}Ir [6]. Details of the calculations involved in the electronic bridge mechanism can be found in Refs. [3–5], and references therein. In the case of ^{229}Th, the second-order effect of internal conversion is apparently not allowed for such a low level [3], as the ionization potential of the thorium atom is about 6.1 eV, so radiative processes should dominate (it is not clear, however, how internal conversion may proceed via electronic states in a chemical compound.) Figure 1 shows the Feynman diagrams for the electronic bridge process. It is essentially like the internal Compton effect, except that the electron is excited to a discrete atomic level instead of the continuum of levels. The researchers in Ref. [3] suggest that the deexcitation of a 3.5 eV level in ^{229}Th can proceed by exciting the thorium atom from the $6d_{3/2}$ ground state to the $7p_{1/2}$ state, with an energy of 1.28 eV. The redshifted γ rays would then have an energy $E_\gamma \approx 3.5$ eV $-$ 1.28 eV $= 2.22$ eV. Such photons would have a wavelength of about 560 nm, which occurs in the green part of the visible spectrum. Direct γ emission, on the other hand, would result in photons of about 354 nm, in the near ultraviolet.

The experimental program to detect optical photons from the decay of the isomer is complicated by the need for optically suitable sources and the confounding fact that many uranium compounds (e.g., those containing uranyl ions) are fluorescent, emitting a well-known greenish fluorescence as observed in antique canary glass. Hence it is necessary to distinguish between such effects and

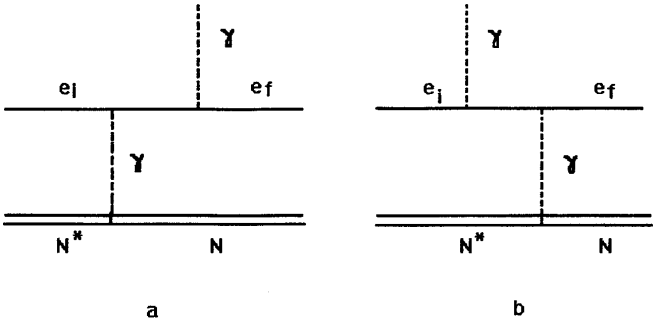

FIG. 1. Feynman diagrams for the inelastic electronic bridge mechanism. (a) Direct diagram; (b) exchange diagram. Double lines indicate the deexciting nucleus (N), solid lines indicate an atomic electron (e), and dashed lines indicate the photon (γ).

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the electronic bridge effect. Therefore part of this work involved attempts at exciting fluorescence in uranyl nitrate and uranium glass using ultraviolet light and intense γ radiation from a ^{57}Co source. Similar excitation experiments were attempted on the ^{233}U samples. These samples had been prepared for γ-ray measurements and were not necessarily the best for this study, but were available from the Idaho National Engineering Laboratory (INEL, operated by Lockheed Martin-Idaho Technologies, Inc.), where the low-lying level was discovered [1]. Two samples, one 100 μCi (sample 1) and one 300 μCi (sample 2) were used in this study. Sample 1 consisted of a deposit of ^{233}U containing a brownish material on Kaptan and covered with transparent tape. Sample 2 consisted of a light-yellowish deposit on a filter paper (probably $UO_4 \cdot 2H_2O$), taped down to Kaptan. The Kaptan films were taped to an aluminum support card. The chemical composition of these materials was not certain at the time of this writing, but as part of this study it was determined that they are not fluorescent (see below). The transparent tape covering the samples had good transmission in the visible part of the spectrum, but not in the ultraviolet. Therefore measurements were performed with the source material exposed by removing the protective tape, with appropriate care taken to prevent contamination.

Figure 2 shows a block diagram of the experimental apparatus used in this work. The Bausch & Lomb high intensity monochromator contained a grating with 1350 grooves per mm and a 300 nm blaze wavelength, with a reciprocal dispersion of 6.4 nm/mm. The Oriel photomultiplier had a responsivity of about 3×10^4 A/W over the range of interest. Measurements were made by simply monitoring the photomultiplier current as the monochromator slowly scanned (20 nm/min) from 850 to 100 nm. An Oxford PCA-III multichannel analyzer card was used in sample voltage list mode to monitor the analog output of the Oriel 7070 readout for data collection. The monochromator was usually used with very wide (6 and 3 mm) slits for maximum light gathering capability, and the samples (which were extended over a diameter of about 1.5 cm) were simply placed near the entrance slit in the acceptance cone of the monochromator (f# 3.5) with no intervening optics. The monochromator had a calibrated wavelength scale which was confirmed with a HeNe laser and mercury lamp. For purposes of ultraviolet irradiation of samples an Ultraviolet Products, Inc. Blak-ray long-wave ultraviolet lamp was used by allowing the uv to illuminate the samples in a chamber. A 1 mCi ^{57}Co source was used to irradiate samples with γ radiation by placing the small source (consisting of a 1 cm diameter palladium foil implanted with the isotope) directly behind the samples. Samples consisting of uranyl nitrate, ground uranium glass, and zinc sulfide were also prepared for irradiation studies.

Figure 3 shows the results of measurements made on ^{233}U sample 1. As postulated, optical emission in the ultraviolet part of the spectrum was observed. The measurements involved several separate runs which individually took 2250 s during which the Oriel 7070 output was

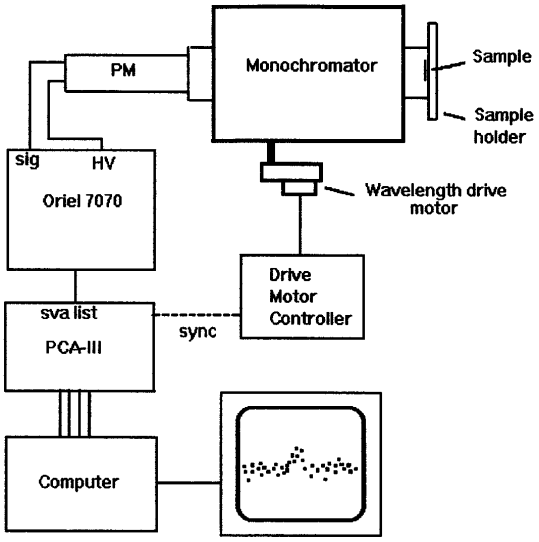

FIG. 2. Block diagram of the optical spectroscopy apparatus for investigating the deexcitation of the low-lying level in ^{229}Th.

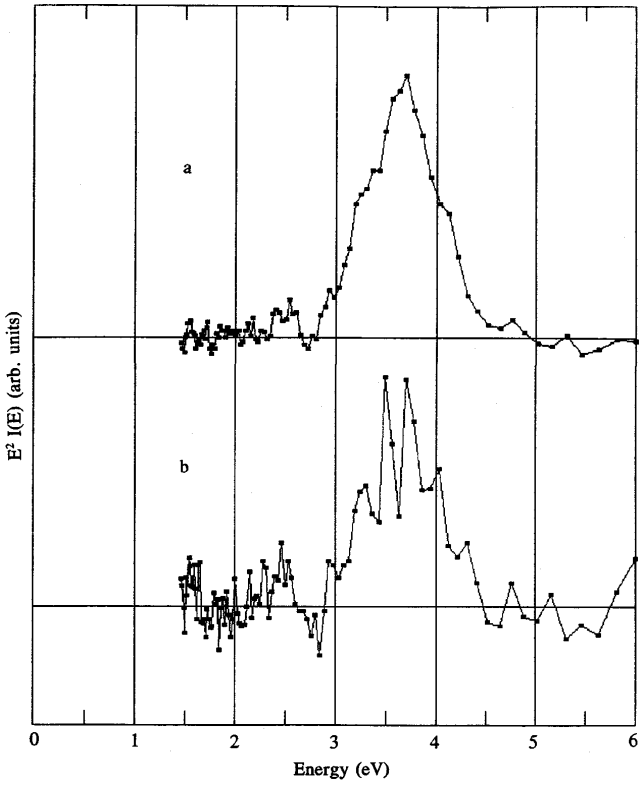

FIG. 3. Results of optical measurements on ^{233}U sample 1. (a) 6 mm entrance slit; (b) 3 mm entrance slit. Solid lines are drawn between adjacent data points to guide the eye.

sampled every 400 ms, resulting in 5625 data points from 850 to 100 nm. These data were then reduced to 112 data points by simply averaging in groups of 50 points. Note that the data plotted are $E^2I(E)$ as a function of energy in eV, where $I(E)$ is the photocurrent per unit energy. Figure 3(a) shows the spectrum of the 100 μCi sample 1 with a 6 mm entrance slit, resulting from 16 separate runs which have been added, showing a peak near 3.5 eV in agreement with the results of Helmer and Reich [1]. We note that the researchers in Ref. [1] purposefully doubled the uncertainty in their determination of the energy separation of the excited state and ground state to account for possible unrecognized systematic errors, and so a value of 3.5 ± 0.5 eV for the isomer is not too conservative and our results are still in good agreement. Figure 3(b) shows the spectrum for the same sample with a 3 mm entrance slit resulting from 16 separate runs. This spectrum, obtained with better resolution but a poorer signal-to-noise ratio, suggests that the ultraviolet part of the spectrum consists of a series of lines, indicating some sort of fine structure. This width to the spectrum may also limit the precision of the techniques used in Ref. [1] for determination of the energy of the low-lying level. Also note a weak peak near 2.4 eV which may be due to the inelastic electronic bridge mechanism.

Figure 4 shows the results for ^{233}U sample 2. Data are the result of 16 separate runs. As with sample 1, sample 2 shows emission in the ultraviolet near 3.5 eV. Note the prominent emission near 2.4 eV, much larger than in sample 1. The various components in the ultraviolet part of the spectrum in Fig. 4(b), which are at the same energies as in sample 1, are possibly correlated with somewhat broader components at 1.3 eV lower energy in the visible part of the spectrum, in agreement with theoretical suggestions by Strizhov and Tkalya from Ref. [3] that the nuclear deexcitation takes place via an inelastic electronic bridge in which the $6d_{3/2}$ thorium atomic ground state is excited to the $7p_{1/2}$ state. Confirmation of this correspondence will require better data with finer resolution and a better signal-to-noise ratio. Note that one would also expect to see photons with energy 1.28 eV following deexcitation of the thorium atom, but such a wavelength (about 970 nm) is not observable with the present apparatus.

The spectra in Figs. 3 and 4 have not been corrected for system efficiency, which is maximum in the uv, and so integrated intensities do not directly indicate branching ratios. By correcting for the efficiency of the monochromator grating and the responsivity of the detector, we have made rough estimates of the branching ratios $\zeta = W^{(3)}/W^{(1)}$ (third-order electronic bridge to first-order direct γ-ray emission) for the two samples. Sample 2 has $\zeta \approx 4$, so that the electronic bridge process dominates, as predicted in Ref. [3]. Sample 1, with the very weak visible emission, has a value $\zeta \approx 0.2$, suggesting that direct uv γ emission dominates. However, these values do not include possible effects due to different attenuation of the uv and visible photons in the two samples, which appear to be chemically different. For example, it was found that the total emission from sample 1 is about $\frac{1}{6}$ of that from sample 2, as compared with $\frac{1}{3}$ as one would expect from the relative α activities, suggesting that about half as many photons, mainly visible, are able to escape from sample 1 as compared to sample 2. Hence it may be that the inelastic electronic bridge mechanism dominates in both cases. It should be noted that samples 1 and 2 apparently had different molecular forms, and the exact energy of the $7p_{1/2}$ state in the thorium atom may depend on the molecular environment, thus leading to the slightly different peaks in the two samples, as well as different branching ratios for direct γ-ray emission and the inelastic electronic bridge.

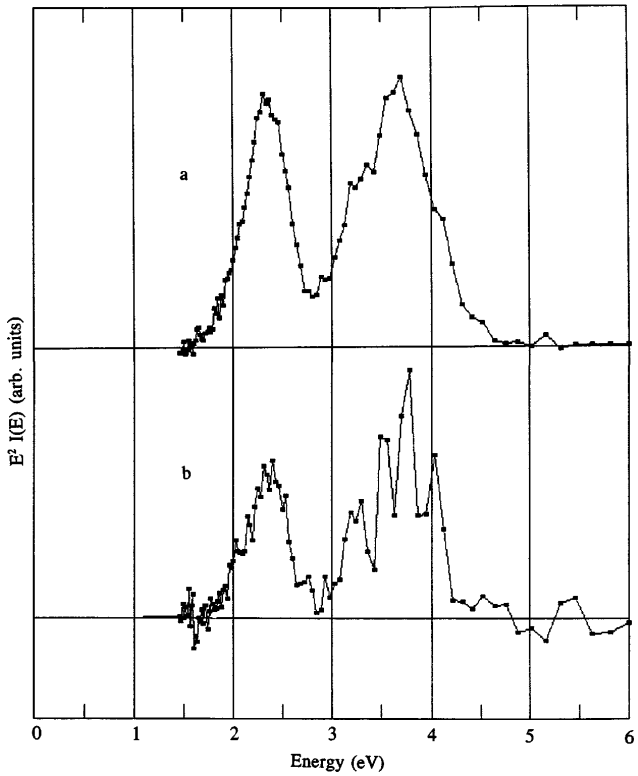

FIG. 4. Results of optical measurements on ^{233}U sample 2. (a) 6 mm entrance slit; (b) 3 mm entrance slit. Solid lines are drawn between adjacent data points to guide the eye.

One might argue that the optical emission observed from samples 1 and 2 is simply some form of fluorescence or scintillation, e.g., from uranyl fluorescence. Experiments designed to account for the optical emission from sources other than the rather exotic mechanisms postulated were attempted. First, it was found that samples 1 and 2 could *not* be excited to fluoresce in ultraviolet light, although such excitation was easily observed in uranyl nitrate, uranium glass, and zinc sulfide, all of which have a greenish fluorescence. Further, both the uranyl nitrate and

zinc sulfide samples could easily be made to scintillate under γ irradiation from the 1 mCi ^{57}Co source, which had no effect on samples 1 and 2 nor on a sample "yellow cake" ($UO_4 \cdot 2H_2O$). We have been unable to observe any optical emission from other uranium-containing materials with activities a substantial fraction of sample 1 and 2. The observed emission is also not consistent with atomic thorium lines as observed with arc and park methods, which lead to lines between 268 and 319 nm, nor with uranium which is known to have many lines throughout the ultraviolet and visible [7].

Clearly further studies are necessary, utilizing sources of similar activity and chemical composition but with other uranium isotopes, such as ^{234}U which has a half-life of 2.4×10^5 yr compared with 1.6×10^5 yr for ^{233}U. Also needed is a detailed chemical analysis of samples 1 and 2 used in this work. Such studies, as well as studies of various chemical compounds of uranium, are planned, utilizing photon-counting methods and a detection system with better resolution and sensitive to longer wavelength photons. With photon counting techniques, it may be possible to coincidence the optical counts with the weak γ emission from the 29.1 keV level in ^{229}Th, which accounts for most of the population of the low-lying level. This would provide a confirmation that the observed optical emission is in fact due to the decay of the isomeric level and not some other source, but the technique would not work if the lifetime of the low-lying level is very long. We have observed changes in the visible portion of the spectra of both samples at liquid nitrogen temperatures, and we plan more detailed cryogenic experiments to investigate these effects (we note that no such effects were observed in the uranyl nitrate sample in liquid nitrogen, supporting our conclusion that the emission in the visible part of the spectrum is not due to uranyl fluorescence). Also studies in which the excited level in ^{229}Th is populated from the ground state are planned, which may lead to measurements of the lifetime of the level, which is expected to depend strongly on the environment of the atom [3]. This may be possible via the inverse electronic bridge mechanism, in which the nuclear level is excited by exciting appropriate electronic transitions in the atom, or by direct nuclear excitation with intense lasers or other light sources [8]. It has also been suggested that the α-decay rate of ^{229}Th may be greater by a factor of 3 in the isomeric level [9]. Such effects could be observable, in principle, if sufficient numbers of isomeric nuclei could be produced.

In conclusion, we have observed ultraviolet and visible photons from samples of ^{233}U suggesting the deexcitation of a 3.5 eV isomeric level in ^{229}Th. Visible emission is consistent with theory predicting that the isomeric level can also deexcite via the inelastic electronic bridge mechanism. This exotic nuclear level is the lowest known of all nuclei, and opens new areas of physics related to atom-nucleus interaction.

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