^{238}U$(e, e'\alpha)$ cross section in the region of the giant quadrupole resonance*

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We performed an experiment which was much more sensitive to the ^{238}U$(e,e'\alpha)$ reaction at 13.1 MeV incident electron energy than was a previous experiment which reported surprisingly large cross sections. Our results set an upper limit for the $(e,e'\alpha)$ cross section of about 16 μb, an order of magnitude smaller than that reported earlier.

[NUCLEAR REACTIONS ^{238}U$(e,e'\alpha)$; E=13.1 MeV; measured $\sigma_{e,\alpha}(E)$.]

This paper describes an experiment designed to learn more about the unexpectedly large ^{238}U$(e, e'\alpha)$ cross sections reported[1,2] for incident electrons in the energy range from 9 to 24 MeV. Those cross sections were interpreted as indicating the presence of an $E2$ resonance in ^{238}U centered at an excitation energy of 8.9 MeV with a width of 3.7 MeV. The inferred value of the energy integrated $E2(\gamma, \alpha)$ cross section was[1,2] 28 MeV mb which is surprisingly large for two reasons. First, the entire sum rule prediction[3] for the isoscalar $E2$ resonance is only 28 MeV mb. Second, the α-decay probability of an $E2$ resonance at 8.9 MeV in ^{238}U would be expected to be very small because α emission is severely inhibited by the Coulomb barrier whereas there are many open channels for both fission and neutron emission.

The production of ^{234}Th due to the ^{238}U$(e, e'\alpha)$ reaction had been inferred[1,2] by detecting the 63 and 92.5 keV γ rays which follow the β decay of the 24.1 day ^{234}Th. We decided to produce a much more intense source of the puzzling γ rays near 63 and 92.5 keV, and to study them in the hope that we might discover a more plausible explanation of their origin.

We made a ^{238}U target that was about 1 mg/cm^2 thick and 0.7 cm^2 in area by using a collodian spreading technique[4] in which uranium nitrate was deposited on a 25.4 μm thick aluminum backing foil. We tried to remove nitrogen oxides by heating but subsequent measurement of the energy of the α particles from ^{238}U indicated that a thin film remained on the uranium. The 63 and 92.5 keV γ-ray intensities were measured with a 50 cm^3 Ge(Li) detector in a reproducible geometry. The γ-ray intensity before electron bombardment was due to the 24.1 day ^{234}Th which was in equilibrium with its 4.51×10^9 yr ^{238}U parent. After measuring these γ rays, we attempted to produce the previously reported increased activity by irradiating the foil with 70 mC of 13.1 MeV electrons. This irradiation was made in a few hours using a beam current of between 5 and 10 μA. If the $(e, e'\alpha)$ cross section were 0.1 mb, this irradiation would have produced 4.3 times the amount of ^{234}Th that was originally in the target.

Our data are shown in Fig. 1. The activity measured in the target before bombardment is shown by thw lowest set of data. The γ rays at 63 and 92.5 keV come from the ^{234}Th that was in equilibrium with the ^{238}U. The data also show some background γ rays in the energy range between 70 and 90 keV which probably come from ^{205}Pb in the shielding around the Ge(Li) detector. The intensity of these background γ rays varied during the course of our series of measurements because the shielding was rearranged.

The other spectra shown in Fig. 1 were taken at different times between 11 and 60 days after bombardment. Most of these spectra are dominated by the 59.5 and the 97.2 keV γ rays emitted following the 6.75 day β decay of ^{237}U produced by the $(e, e'n)$ reaction; even the weaker 64.8 keV γ ray from the ^{237}U decay is much stronger than the 63 keV γ rays from ^{234}Th in the early spectra. In the top spectrum, taken 11 days after irradiation, the ratio of the intensities of the 59.5 keV γ ray (from 6.75 day ^{237}U) to the 63 keV γ ray (from 24.1 day ^{235}Th) was 224 to 1. In the next to the lowest spectrum shown in Fig. 1, which was taken 60 days after bombardment, the ratio of intensities of the 59.5 to the 63 keV γ ray was about 1.7. Each of the spectra shown in Fig. 1 was measured for about 10 h; the data were normalized to a counting time of 10 h.

Our experiment was designed to measure the intensity of the γ rays corresponding to an $(e, e'\alpha)$ cross section of 0.1 mb to an accuracy of about 10%. The 10 h counting time was sufficient to detect about 1300 counts due to the 63 keV γ ray and about 2000 counts due to the 92.5 keV γ ray before bombardment. If the cross section had been 0.1 mb, there would have been about 1000 extra counts due to 63 keV γ rays 60 days after our

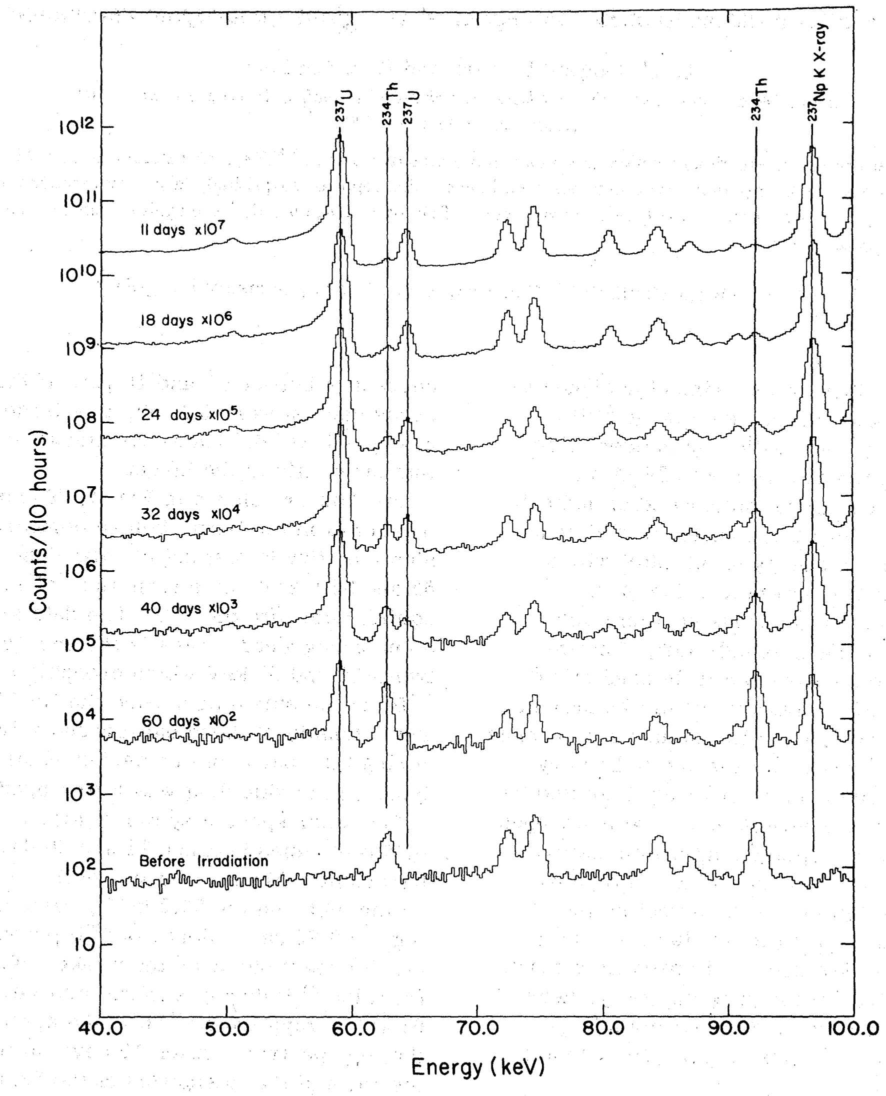

FIG. 1. Observed γ-ray activity from a ^{238}U foil before irradiation (lower spectrum) and at specified intervals after irradiation by 70 mC of charge from a 13.1 MeV electron beam (upper spectra).

bombardment, and we would have determined this increment with a statistical accuracy of about 6%. The main systematic error in the experiment was associated with the estimated 5% uncertainty in the positioning of the sample relative to the detector.

Our data do not show any experimentally significant increase in the number of 63 or 92.5 keV γ rays. The previously reported ($e,e'\alpha$) cross section near 13 MeV was about 0.2 mb (estimated from Fig. 3 in Ref. 1); the curve used to fit the reported data (in Fig. 6 of Ref. 2) indicates a cross section of about 0.16 mb near 13 MeV. Our experiment indicates that the cross section is not as large 0.016 mb at 13.1 MeV. This disagreement by an order of magnitude in the ($e,e'\alpha$) cross section is in sharp contrast to the excellent agreement in the ($e,e'n$) cross section; we measured 856 ± 47 mb, whereas Fig. 4 in Ref. 2 implies about 850 mb. This ($e,e'n$) cross section is also in very good agreement with the (γ,n) cross section[5] if the ($e,e'n$) reaction is dominated by electric dipole excitation.

There are several different ways to illustrate the gross disagreement between our data and a 0.16 mb ($e,e'\alpha$) cross section. If the cross section were 0.16 mb, the 63 and 92.5 keV γ-ray intensities 11 days after bombardment would have been 6 times as great as those observed before bombardment. However, no statistically significant increase was observed. The 63 keV intensity extracted from the 11 day spectrum by our computer program for fitting γ-ray lines was 1.05 $\pm$0.06 times the intensity before bombardment; a cross section of as little as 0.016 mb would have produced a ratio of 1.51. We are reluctant to use this spectrum to set an upper limit on the cross section because of the very large intensities of γ rays from ^{237}U and from fission products. The spectrum obtained sixty days after bombardment would have had 63 and 92.5 keV γ-ray intensities that were 2.3 times those observed before bombardment if the $(e, e'\alpha)$ cross section were 0.16 mb. No intensity increase is evident, even though the 63 and 92.5 keV γ rays are well separated from neighboring γ rays and background in the spectrum taken 60 days after bombardment as shown in Fig. 1. From this spectrum, the ratio of intensity of the 63 keV γ ray after bombardment to that before bombardment was 0.97 ± 0.04. Even if the ratio were three standard deviations above the observed value of 0.97, and if the actual ratio were 5% higher due to a sample positioning error, the implied $(e, e'\alpha)$ cross section would be only 0.016 mb.

In order to reduce the possibility that errors in positioning the sample relative to the Ge(Li) detector were obscuring increased 63 and 92.5 keV γ-ray intensities, we also analyzed our data by assuming the known $(e, e'n)$ cross section and the 24.1 day half-life of ^{234}Th, and assuming that the 6.75 day ^{237}U half-life could be used together with the observed 59.5 keV γ-ray intensity to obtain the relative γ-ray efficiencies for each spectrum taken after bombardment. Using these assumptions, the $(e, e'\alpha)$ cross section was deduced from the observed intensities of the 63 keV γ ray. The inferred $(e, e'\alpha)$ cross section was 0.006 ± 0.012 mb, (95% confidence). The examples given above are all related to the 63 keV γ ray because the previous experiment[1,2] was more sensitive to this γ ray. Our detector was about 1.5 times more sensitive to the 92.5 keV γ ray, and we did not see any experimentally significant evidence that its intensity was increased by electron bombardment.

Our experiment should have had a sensitivity to the $(e, e'\alpha)$ cross section that was 3 or 4 orders of magnitude greater than the sensitivity of the experiment of the Sao Paulo group.[1] The largest contributor to our improved sensitivity was the larger charge of 70 mC that we used to bombard the sample. In addition, we used a target whose cross sectional area was only 0.7 cm^2. The targets that were used during the previously reported measurements were about 20 cm^2. In the previous measurements the inferred cross sections depended[6] on how the efficiency of the γ-ray detector varied for different portions of the sample, and on the size of the irradiated portion of the sample. Our experiment did not have these complications. Our target, which was between 4 and 10 times as thick as the samples used in the reported studies[6], and our bigger γ-ray detector enabled us to obtain better statistical accuracy for measuring the γ-ray activity orginating from the portion of the sample that had been irradiated by electrons. The previous reports of larger $(e, e'\alpha)$ cross sections[1,2] do not include an adequate description of the careful checks that would be necessary in order to make reliable cross section measurements from such small percentage enhancements in ^{234}Th as are implied by the reported cross sections[1,2] and experimental procedures.[6]

In summary, our much more sensitive experiment with 13.1 MeV electrons contradicted previous reports[1,2] which indicated large $(e, e'\alpha)$ cross sections. We also did a less complete experiment using electrons with an energy of 14.9 MeV; it was similarly negative. Doubt has been cast on the reported $(e, e'\alpha)$ cross sections[1,2] by Hayward, Dodge, and Wolynec[7] who were unable to observe any associated α particles and also by McGeorge *et al.*[5] Our experiment supports the findings of Hayward *et al.*,[7] and additionally rules out other possible electron-induced reaction channels leading to the production of ^{234}Th from ^{238}U with a cross section larger than $\frac{1}{10}$ that reported by Wolynec *et al.* The previous reports[1,2] were based mainly on rather small enhancements of 63 keV intensities. Not enough experimental details are given to indicate how the measurements were made with the required reliability. We conclude that the reported 63 keV γ-ray intensity enhancements[1,2] were not caused by electrons interacting with ^{238}U, and that the reported[1,2] $(e, e'\alpha)$ cross sections are in error.

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