BRIEF REPORTS

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Neon radioactivity of uranium isotopes

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We report a new measurement of the rate of spontaneous emission of monoenergetic Ne nuclear clusters from ^{232}U, which results in a revision of rates previously reported for Ne radioactivity of ^{232}U, ^{234}U, and ^{235}U. The Ne emission rate goes up a factor of 4.6 for ^{232}U, goes down a factor of 4.8 for ^{234}U, and may have been marginally detected (1.9σ) for ^{235}U.

Soon after the pioneering experiments on ^{14}C emission from various radium isotopes [1,2], spontaneous emission of neon nuclear clusters was discovered [3]. Among neon-emitting nuclides, ^{232}U is the one with the highest decay probability, due to strong shell effects in the structure of the daughter nucleus, the doubly magic ^{208}Pb.

This decay was first observed in 1985 at Berkeley [3] by means of polyethylene terephthalate nuclear track-recording plastic films. That experiment yielded a branching ratio relative to the alpha decay of 1.83×10^{-12}. In 1989 the measurement was repeated in Milano using a track-recording phosphate glass detector [4]. Although that experiment was primarily aimed at measuring other ^{232}U decay modes such as spontaneous fission and Mg emission, neon tracks were also recorded, and a branching ratio of 8.68×10^{-12} was determined. This factor-of-four discrepancy between the two measurements is somewhat greater than the discrepancy between measured cluster decay rates and rates predicted by models of cluster decays, several of which have been quite successful in predicting rates for even-even nuclides.

Because of the large branching ratio for neon emission from ^{232}U, the presence of ^{232}U impurities in sources consisting of other uranium isotopes may give rise to a background of neon emission that must be accurately known and accounted for. We decided to make a third measurement of the neon emission rate of ^{232}U and to reevaluate the neon emission rate of ^{234}U and ^{235}U, both of which had been studied before, using sources containing a non-negligible concentration of ^{232}U.

We obtained a 15 ± 0.7 MBq ^{232}U source from the Chemistry Division of the Atomic Energy Authority, Harwell, U.K., and used a PSK-50 phosphate glass plate, 5 cm$\times$5 cm in area, in contact with the source, to measure the neon tracks. At the end of 38 days of exposure, the glass received 7×10^{12} alphas cm^{-2}, well below the limiting value of 1×10^{14} at which radiation damage causes the performance of the detector to deteriorate [2]. At this stage we carefully checked the source activity. We performed two different gamma-ray countings with different geometries, which yielded consistent results. The uncertainty in source strength includes both the uncertainties in detector geometry and in branching ratios of the levels being counted.

We subsequently etched the plate in 70% nitric acid at 65 °C for 25 min and scanned as carefully as possible for neon tracks using two independent observers. The method was to scan the entire plate at 200$\times$ in transmitted light and then to measure the geometry of the tracks at 1000$\times$ as done in previous experiments [2–4]. We found a total of 90 tracks with zenith angles between 0° and 65°. To calculate the branching ratio relative to alpha decay, we had to take into account both the decrease in recording ability of the glass detector at very large zenith angles and the inaccuracy in measurement of zenith angle at near vertical incidence. These effects were corrected for experimentally by plotting the distribution in solid angle as a function of zenith angle and excluding

the few (eight out of 90) tracks at angles smaller than 20° and greater than 55°. The distribution of tracks with $20° \leq \theta \leq 55°$ was consistent with a constant number per unit solid angle, as it should be for isotropic emission from a thin source. After dividing the number of detected tracks in the interval 20° to 55° by $(\cos 20° - \cos 55°)/2$, the fraction of the full 4π solid angle, we obtained a branching ratio of $(9.16 \pm 1.1) \times 10^{-12}$. This result is quite consistent with the one obtained in 1989 (Ref. [4]) and a factor of nearly 5 greater than the one obtained in 1985 (Ref. [3]). We believe the most likely explanation for the large discrepancy is that in the first experiment the source was obtained from a commercial firm and its activity was assumed to be that specified by the manufacturer rather than independently checked. Calculating the weighted average between the results of the second and third experiments, we obtain the value $B(\mathrm{Ne}/\alpha) = (8.88 \pm 0.71) \times 10^{-12}$, which we will use in the rest of this paper.

We next discuss the implication of the present result for previous experiments on neon radioactivity of ^{234}U and ^{235}U and present a revision of the related decay rates.

In Ref. [5] the Berkeley-Livermore Collaboration reported the first measurement of the neon radioactivity of ^{234}U (not isotropically resolved; assumed to be due to ^{24}Ne and ^{26}Ne). They used a ^{234}U source that contained 0.002% of ^{232}U by weight. Using the (incorrect) neon emission rate from the 1985 measurement, they estimated the contribution of ^{24}Ne emitted from ^{232}U at 18% of the total. However, using the present result, the ^{24}Ne emission rate is a factor of 4.61 more probable. The contribution from the ^{232}U impurity therefore increases to 83%, and the branching ratio for Ne emission from ^{234}U decreases from $(4.36 \pm 0.5) \times 10^{-13}$ to $(9.06 \pm 6.60) \times 10^{-14}$.

Tretyakova *et al.* [6] have recently measured the neon emission rate of ^{234}U using polyethylene terephthalate detectors. Their ^{234}U source contained 0.0012% by weight of ^{232}U. Using the new rate of neon emission by ^{232}U, we estimate that their branching ratio for ^{24}Ne emission from ^{234}U decreases from $(3.89 \pm 1.01) \times 10^{-13}$ to $(9.93 \pm 9.93) \times 10^{-14}$.

In both of the ^{234}U experiments, branching ratios for Mg emission relative to alpha decay were also measured. Since the emission rate of Mg by ^{232}U is negligible relative to the Ne emission rate, the Mg/α branching ratio in ^{234}U is unaffected by the present result. By using the revised decay probabilities for neon emission, we recalculate the branching ratios $R = \lambda_{\mathrm{NE}}/\lambda_{\mathrm{Mg}}$ to be 0.66 for the experiment reported in Ref. [5]. For the experiment in Ref. [6] the error is too large for the derived value of R to be meaningful.

We now consider the neon emission rate of ^{235}U, which was studied by Tretyakova *et al.* [6], using a source containing some ^{234}U impurity. In their polyethylene terephthalate detectors they found seven Ne tracks which, on the basis of their branching ratios for ^{234}U, were all attributed to emission from that isotope. They therefore inferred a null result for ^{235}U and only set an upper limit for the related branching ratio. Clearly, if the neon emission rate of ^{234}U is lowered, the contribution of ^{234}U impurities to neon decays from the ^{235}U source (together

TABLE I. Comparison of experimental results and theoretical predictions for branching ratio for Ne emission, $B = \lambda_{\mathrm{Ne}}/\lambda_\alpha$, and relative emission rate, $R = \lambda_{\mathrm{Ne}}/\lambda_{\mathrm{Mg}}$, for Ne and Mg emission.

	^{232}U		^{234}U			^{235}U	
	B	B	R	B	R	B	R
Previous results	$(1.93 \pm 0.50) \times 10^{-12}$ (Ref. [3]) $(8.68 \pm 0.93) \times 10^{-12}$ (Ref. [4])	$(4.36 \pm 0.5) \times 10^{-13}$ (Ref. [5])	3.16 ± 0.7	$(3.9 \pm 1) \times 10^{-13}$ (Ref. [6])	1.7 ± 0.7	$< 5 \times 10^{-12}$ (Ref. [6])	
Present results	$(8.88 \pm 0.71) \times 10^{-12}$	$(9.06 \pm 6.6) \times 10^{-14}$	0.66 ± 0.5	$(9.9 \pm 9.9) \times 10^{-14}$		$(8.06 \pm 4.32) \times 10^{-12}$	$> 10^{a}$
Predictions from [2]	5×10^{-12}	1.8×10^{-13}	0.6	1.8×10^{-13}	0.6	all Ne 1.1×10^{-13}	~ 1.0
Predictions from [7]	3.98×10^{-12}	1.25×10^{-13}	0.79	1.25×10^{-13}	0.79	^{24}Ne 2.69×10^{-14} ^{25}Ne 5.37×10^{-15}	0.14
Predictions from [8]	4.00×10^{-12}	2.00×10^{-13}	0.77	2.00×10^{-13}	0.77	^{24}Ne 2.59×10^{-14} ^{25}Ne 2.40×10^{-12}	2.1

[a]This limit assumes the validity of both the upper limit on the Mg emission rate from Ref. [6] and the value $B = 8.06 \times 10^{-12}$ obtained in the present analysis. To bring the value of R into agreement with the predictions of Refs. [2] and [7] would require a reduction of B by at least an order of magnitude.

with ^{234}U impurities) must also be smaller. From the experimental conditions reported in [6], we calculate that, of the seven neon tracks recorded, the most probable value for the number of tracks due to the ^{234}U impurity is only 1.6. Therefore, the branching ratio for neon emission from ^{235}U becomes $(8.06\pm4.32)\times10^{-12}$.

Tretyakova *et al.* [6] detected three Mg tracks that were emitted from their ^{235}U source. From this observation, after correcting for Mg from the ^{234}U impurities, they estimated an upper limit of 8×10^{-13} for the branching ratio $B(\mathrm{Mg}/\alpha)$. If we accept the validity of this upper limit, and if we also accept the value $B(\mathrm{Ne}/\alpha)=8.06\times10^{-12}$ found in the previous paragraph, the branching ratio $R(\mathrm{Ne/Mg})$ for ^{235}U must be greater than 10. This value is a factor of at least 10, 70, and 5 higher than predicted by the models in Refs. [2], [7], and [8], respectively. The discrepancy is far outside that due to the statistics of three Mg events. A possible explanation is that the value of $B(\mathrm{Ne}/\alpha)$ for ^{235}U has been overestimated in the subtraction procedure. We conclude that a convincing case for the detection of Ne emission by ^{235}U has not yet been made.

The current theories of heavy-ion radioactivity are classified as either fission models or cluster models. It is interesting to compare the predictions of three of the most successful models [2,7,8] with the branching ratios obtained in the present work.

In Table I we see that all three models still differ by a factor of 2 from the present updated result for ^{24}Ne emission from ^{232}U, although the rates are now low rather than high. For ^{234}U the results of the three models are now in much better agreement with the revised experimental rates for neon emission as well as with the branching ratios Ne/Mg. Of course, one must keep in mind that the errors implied by the subtraction procedures are larger than just those due to counting statistics.

In the case of ^{235}U, the three models fail by 2 orders of magnitude to predict the Ne/α ratio and by one or more orders of magnitude to predict the Ne/Mg ratio if we take seriously the subtraction procedure and if we consider only the contribution of ^{24}Ne to the measured neon events. Note, however, that, due to the favorable Q value, ^{25}Ne is also a possible contributor. The Q value for ^{25}Ne emission is 57.8 MeV, to be compared with 57.36 MeV for ^{24}Ne emission. Since both the energy resolution and the isotopic resolution of track detectors are insufficient to discriminate between the two neon isotopes, both isotopes must be considered when comparing with theoretical predictions. Consider first the simple square-well model [2], with three arbitrary parameters, one of which is a constant hindrance factor for decays of odd-A nuclides. The fission model of Poenaru *et al.* [7] predicts $B=5.37\times10^{-15}$ for ^{25}Ne, which is even smaller than the one for ^{24}Ne, and is therefore not large enough to improve the agreement with experiment. In the case of the cluster model of Blendowske and Walliser [8], the situation seems, at least in principle, more promising. In this model, provision is made for distinguishing between even and odd emitters. For odd emitters, a decreased spectroscopic factor is introduced (S odd). This permits an explanation of the hindered transitions which have been known for a long time for odd-A emitters, and which have also been confirmed in the case of cluster radioactivity [2].

In the cluster model, the physical picture is that the difficulty in assembling an even cluster out of an odd nucleus would considerably slow down the transition to the ground state of the daughter nucleus. However, this need not be true in the case of an odd cluster, such as ^{25}Ne, from an odd parent.

While the above case has not yet been explicitly treated by any cluster model, an encouraging starting point is to calculate the $^{235}\mathrm{U}\rightarrow{}^{25}\mathrm{Ne}+{}^{210}\mathrm{Pb}$ decay rate by using, within the model of Ref. [9], the spectroscopic factor appropriate for unhindered transitions (S even). Following this approach, we obtain the value $B=2.4\times10^{-12}$, in rather good agreement with the experimental value obtained by subtraction as discussed above.

One should, however, keep in mind that the Ne result on which this discussion has been based is positive only by 1.9σ, and the Ne/Mg ratio is poorly accounted for. It would be important to try to reduce the background neon emission rate by working with a ^{235}U source containing a smaller ^{234}U content, and to collect a larger sample of Ne and Mg events.

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