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### Publication Date

1957-05-01

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UCRL-3774

UNIVERSITY OF CALIFORNIA

Radiation Laboratory  
Berkeley, California

Contract No. W-7405-eng-48

AN ISOMERIC STATE OF URANIUM-235

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May 1957

Printed for the U. S. Atomic Energy Commission

## AN ISOMERIC STATE OF URANIUM-235

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There has been an apparent inconsistency in the data concerning the energy levels of  $U^{235}$  ever since it was noted that:

- (1) The ground state spin of  $U^{235}$  determined from optical spectrum hyperfine structure is  $7/2$ ,<sup>1,2</sup> and this is in agreement with Coulomb excitation studies.<sup>3</sup>
- (2) The lowest lying state of  $U^{235}$  populated in the alpha-decay of  $Pu^{239}$  apparently has spin- $1/2$ , since it and the sequence of levels above it have the characteristics of a Bohr-Mottelson rotational band with spin- $1/2$  base state.<sup>4,5,6</sup>
- (3) There is no obvious gamma-ray transition from the  $1/2$  state to the  $7/2$  state, even though virtually all  $Pu^{239}$  alpha-transition populate directly or indirectly to the spin- $1/2$  state.

This information suggests that  $Pu^{239}$  decays to an isomeric state of  $U^{235}$  which is either very long lived (to explain the failure to find the abundant  $1/2 \rightarrow 7/2$  gamma-transition) or two states lie so close together that the radiation is not observable by usual methods. For an E3 transition in a heavy element it is improbable that the lifetime can be long, since, as will be pointed out at the end of this note, no matter how low the energy becomes, the conversion coefficient becomes correspondingly large. Nevertheless, on the supposition that the radiation was measurable but long lived, careful gamma-ray and conversion electron measurements<sup>7</sup> were made in this laboratory on a sample of  $Pu^{239}$  which had aged for several years and failed to show any radiations other than those previously seen. In addition, a chemical separation of uranium was made; this also gave no evidence for the isomeric transition.<sup>8</sup> Very recently Huizenga, Engelkemeir and Tompkins<sup>15</sup> have looked at the optical spectrum of  $U^{235}$  removed from  $Pu^{239}$  and found only the  $7/2$  state, setting a lower limit of six months for the half-life of an isomer of spin  $1/2$ . The alternate explanation that the radiation was too soft to be detected under the means employed therefore seemed more attractive; some evidence for this appeared to come from the measurement of the conversion electron spectrum of Shliagin<sup>9</sup> who found 2 kev electrons which were fairly prominent. As will

appear below, we have found the isomeric state decaying by the emission of very soft electrons, but these seem to be of even lower energy than those reported by Shliagin.

The isomeric state confirming these conclusions was found by collecting recoils from the alpha-decay of  $\text{Pu}^{239}$  and counting the  $\text{U}^{235m}$  in a windowless proportional counter. The samples were first collected on aluminum foils, using 600 volt negative potential to deposit the ions which had dissipated their recoil energy in air. Several samples so collected contained about 3000 counts per minute, which decayed to the background of 20 counts with a half-life of  $26.5 \pm 0.2$  min. In one experiment a chemical separation of uranium from plutonium yielded a sample with the same decay characteristics, but the apparent yield was low because of self-absorption of the extremely soft electrons. By the recoil method from a sample of  $\text{Pu}^{239}$  which had a surface density of 1.5 micrograms/cm<sup>2</sup>, the yield of soft electrons from the  $\text{U}^{235m}$  corresponded with 1 electron for each five  $\text{Pu}^{239}$  alpha-emission events. A sample of plutonium somewhat thicker ( $\sim 7 \mu\text{g}/\text{cm}^2$ ) gave an intensity of soft electrons of only 1/5, so it is entirely reasonable that every  $\text{Pu}^{239}$  decay event goes through this isomeric transition.

A rough measure of the energy of the electrons was made by an absorber technique in which recoils were caught on thin plastic films and counted, with the sample alternately facing upward and downward. The "range" so determined indicated that the energy of the electrons is less than 1 kev. A plastic film of 2.5 micrograms/cm<sup>2</sup> thickness cut down the counting rate by a factor greater than 100, so there does not seem to be an appreciable number of electrons with energy even as high as 2 kev.

An examination of possible assignments of the two  $\text{U}^{235}$  levels according to Nilsson particle states<sup>10,11</sup> in a deformed well, suggests that the most likely assignments consistent with all information are  $1/2+$  (asymptotic quantum numbers  $N = 6$ ,  $n_z = 3$ ) and  $7/2-$  ( $N = 7$ ,  $n_z = 4$ ). The isomeric transition would accordingly be E3.

The transition energy is deduced to be 2.2 kev or less assuming  $N_{II}$ ,  $N_{III}$  conversion according to the following reasoning: If the electrons are taken to be 1 kev and they resulted from M shell conversion (largely  $N_{II}$  and  $N_{III}$  levels), there should also be sizeable numbers of electrons from M shell

conversion; these would have energies of about 5 kev. Neither Shliagin's electron spectrum nor our absorption measurements gave evidence for such electrons. Consequently, the soft electrons must come from conversion in N shell or higher. With less confidence the similar argument can be made that the electrons are not converted in the N shell because the N and O edges differ by  $\sim 1$  kev. If the observed electrons come from O shell conversion, the transition energy would be 1.2 kev or less. Obviously, more information must be obtained before the isomeric transition energy can be decided.

Interestingly enough, the calculated lifetime for this transition is almost independent of decay energy in the energy range of interest. The gamma-ray emission half lives based on the single proton transition relations of Mosakowski<sup>12</sup> are shown in Table I for a series of gamma energies, and also listed are internal conversion coefficients obtained by extrapolating Rose's<sup>13</sup> M shell coefficients and assuming 3-fold decrease for the N shell and for each successive shell. It is seen that the estimated decay half-lives change by an order of magnitude for a change in energy of a factor of 10. If we assume that for the  $U^{235m}$  case the measured half-life should be 1 second, this  $E_3$  transition is then slower than the calculated single proton transition by a factor of  $10^3$ . Such a degree of retardation is quite normal for  $E_3$  transitions (see Goldhaber and Sunyar<sup>14</sup>) although the great extrapolations involved in the present case probably mean that the agreement is fortuitous.

Table I  
Calculated half-lives and conversion coefficients  
 $E_3$  transition in uranium

Energy (kev)	Partial $\gamma$ -decay half-life (sec)	Conversion coefficient	Total half-life (sec)
10	$1.1 \times 10^7$	$10^8$	.1
5	$1.4 \times 10^9$	$10^{10}$	.1
1	$1.1 \times 10^{14}$	$10^{14}$	1
0.5	$1.4 \times 10^{16}$	$10^{16}$	1
0.1	$1.1 \times 10^{21}$	$10^{20}$	10

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