UCRL 3860

UNIVERSITY OF CALIFORNIA

Radiation Laboratory

BERKELEY, CALIFORNIA

DISCLAIMER

UCRL-3860

UNIVERSITY OF CALIFORNIA
Radiation Laboratory
Berkeley, California

Contract No. W-7405-eng-48

PLUTONIUM-237m, ANOTHER CASE OF E3 ISOMERISM FOR NEUTRON NUMBER 143

F.S. Stephens, Jr., Frank Asaro, Saadia Amiel and I. Perlman

Printed for the U.S. Atomic Energy Commission

PLUTONIUM-237m, ANOTHER CASE OF E3 ISOMERISM FOR NEUTRON NUMBER 143

F.S. Stephens, Jr., Frank Asaro, Saadia Amiel* and I. Perlman

Radiation Laboratory
University of California
Berkeley, California

July 9, 1957

An "E3 isomer" of U^{235} has been sought and recently found;[1,2] the isomeric state has spin 1/2 and lies only a fraction of a kilovolt above the spin 7/2 ground state. The search for the isomer was initiated when it became clear from the alpha spectrum of Pu^{239} that the lowest observable level of U^{235} that is populated has spin 1/2[3-5] whereas direct measurement of the U^{235} ground state showed its spin to be 7/2.[6-8] Because these two levels lie almost on top of each other in U^{235}, there would seem to be a good chance that the same states would not be widely separated in neighboring nuclei with the same neutron number and consequently that one or more similar E3 isomers might be found. (It should be mentioned that the appearance of isomerism in regions of spheroidal nuclear shape follows a quite different pattern than that noted just before the closure of major shells. Particle states in spheroidal nuclei are only two-fold degenerate,[9] hence, in general, the pair of states defining the isomers will be the two lowest-lying states only for a single nucleon number. In contrast to this, similar isomers occur over a considerable range of nucleon numbers near the filling of a major shell in a manner nicely explained by shell theory.[10] Also it might be pointed out that the appearance of isomers in spheroidal nuclei seems to be somewhat accidental in the sense that it is not possible to predict the exact place where two states of sufficient spin difference will be near-lying and without intervening states.)

The known species which, like U^{235}, have 143 neutrons are Th^{233}, Pu^{237}, and Cm^{239}. Experimentally, Pu^{237} appeared most attractive because it is the only one that is the decay product of a reasonably long-lived parent (1% α-branching of 35-day, electron-capture, Cm^{241}.[11,12]) More important was the indication from fragmentary decay data of Cm^{241} and Pu^{237} that spin 7/2 and 1/2 states

*On leave from Israel Atomic Energy Commission at the Weizmann Institute of Science, Rehovoth, Israel.

exist in Pu^{237} in much the same relation as in U^{235}. The data suggesting this similarity will be discussed below. Finally, Pu^{237} lends itself to rapid separation from its parent by virtue of the alpha-recoil collection method.

The Cm^{241} used in this study was prepared by irradiating 60 mg of Pu^{239} with 373 μ a-hr of 32-Mev alpha particles in the Crocker Laboratory cyclotron. The curium fraction was isolated by a series of precipitation and ion-exchange reactions and vacuum sublimed onto a 0.002-inch-thick platinum plate in order that the deposit be sufficiently thin and uniform to permit recoil collection with high efficiency. In all, the Cm^{241} intensity produced was $\sim 5 \times 10^6$ electron-capture disintegrations per minute (5×10^4 alpha disintegrations). Alpha-energy analysis showed that Cm^{240} and Cm^{242} were present, but the observable gamma-ray spectrum was only that which accompanies the electron-capture decay of Cm^{241}. The even-even alpha emitters have gamma-rays that are well characterized but are in very low abundance. In any case, the presence of even-even species does not interfere with the measurements of interest.

The first experiment showed that any Pu^{237} isomer resulting from alpha-decay of Cm^{241} must have a half life longer than several days or shorter than 2 sec. The recoil nuclei were caught on a 6-mil aluminum plate held one-eight inch above the Cm source and charged 300 to 600 volts negative by means of batteries. The sample was placed as quickly as possible into a windowless proportional counter, and no activity was observed.

In the search for a shorter half life, the following system was employed. A closed loop was made of a 5-ft length of 2 1/4-in. -wide paper tape, and this was friction-driven as an endless belt (by a rubber stopper mounted on the shaft of a 60-rpm motor) so that recoils collected on the tape were continuously conveyed through the counter. The tape itself closed the bottom of the methane proportional counter, and, at a variable distance before its entry into the counter, it ran under the Cm^{241} source and over a lead brick that was maintained at several hundred volts negative potential causing the recoils to deposit on the tape.

With the source 6 cm from the counter and with the tape traveling at 20 cm/sec, a counting rate of 2000 cpm above background was observed. The background was measured with everything functioning except that the recoil-collection potential was removed. The half life of the collected recoils was determined by varying at 2-cm intervals the distance between the counter and point of collection. The plot so

obtained is shown in Fig. 1 and corresponds to a half life of 0.18 ± 0.02 sec. This half life was checked with 0.25-mil aluminum foil between the counter and the tape.

The same conveyance system was employed for determining the energy of the 0.18-sec transition. This time a 1 1/2-by 1-inch NaI crystal was used as the detector and the energy spectrum was displayed with a 50-channel pulse-height analyzer. A 2-inch-thick lead brick was used to shield the crystal particularly from the intense 470-kev gamma ray that accompanies the electron-capture decay of Cm^{241}.[13] The background was determined this time merely by inserting a card under the source to intercept the recoils. The spectrum as shown in Fig. 2 included a prominent peak of L x-rays, a gamma ray of 145 ± 5 kev and a possible K x-ray peak at ~105 kev. These photons had the same half life as the electrons measured with the proportional counter.

The multipolarity of the transition was determined from the conversion coefficients and the lifetime. The K x-ray and 145-kev photon intensities were compared to give a K-shell conversion coefficient ≤ 0.3 which, as seen from Table I, rules out magnetic transitions of all multipolarities. The L-shell conversion coefficient was obtained by comparing L x-ray peaks with a source of Cm^{242} which has a known amount of L-shell vacancy.[17] (A small error may be introduced in this comparison because the L-shell fluorescence yields may be somewhat different for the two cases.) The L conversion coefficient determined and shown in Table I is 39 ± 8, which is unique for an E3 transition.

The radiative transition lifetime for the 145-kev transition is estimated to be ~13 sec on the basis of the measured L-shell conversion coefficient and theoretical values for M and N shells for an E3 transition.[18] It is seen that this value also agrees only with an E3 assignment and the agreement is good when one recalls that all known E3 transitions are retarded by the order of 10^2 to 10^3 from the single-particle calculations.

The orbital assignments for the ground state of Pu^{237} and its isomeric state, as well as that for the ground state of Cm^{241}, are shown in Fig. 3. The assignments listed are identical with the corresponding case of Pu^{239} in its decay to U^{235}.[1] The arguments upon which these assignments are made are as follows.

Table I

E3 Assignment of the 145-kev gamma ray

	E1	E2	E3	E4	M1	M2	Exp
α_K [14]	0.16	0.19	0.20	0.18	7.0	27	≤ 0.3
α_L [15]	0.03	2.9	43	450	3.0	21	39 ± 8
$T_{1/2}$(sec) [16]	4×10^{-14}	5×10^{-8}	8×10^{-2}	2×10^{5}	8×10^{-12}	2×10^{-6}	~ 13

The terms α_K and α_L refer to K and L conversion coefficients respectively. The half lives shown are for the radiative transitions; that shown as the experimental value is estimated as explained in the text.

The electron capture of Cm^{241} leads predominantly to a level of Am^{241} believed to have spin 3/2 and not to the ground state which has spin 5/2.[13] This is hardly an argument for assigning Cm^{241} the spin 1/2 but is consistent with the working hypothesis that it is the same as Pu^{239}, which has the same neutron number. The gross alpha-decay half-life of Cm^{241} indicates that the main transition is almost unhindered,[13] a situation thought to occur only between levels in which the odd particles are in identical states, in this case the same Nilsson levels[9] of spheroidal nuclei. (Radiation intensity considerations showed that the main alpha decay of Cm^{241} leads to the isomeric state.) The isomeric state of Pu^{237} is therefore the same as the ground state of Cm^{241} which has been taken to have spin 1/2. Since it deexcites by an E3 transition, the ground state has spin 7/2 and opposite parity. The principal experimental check on this assignment for the ground state of Pu^{237} is the fact that in its electron-capture decay it goes almost entirely to the ground state of Np^{237} [19] which has the assignment 5/2+.[20] The assignments in terms of spin, parity, and asymptotic quantum numbers are summarized in Fig. 3. They are identical with those of U^{235}, but the isomeric level has moved to 145 kev, whereas in U^{235} it is less than 1 kev above the ground state.

The appearance of this isomeric state changes the previously accepted alpha-decay energy of Cm^{241} by 145 kev and largely eliminates a notable discrepancy in the systematic trend of alpha energies.[20]

This work was performed under the auspices of the U.S. Atomic Energy Commission.

LEGENDS

Fig. 1. Decay rate of Pu^{237m} measured in a proportional counter by moving-tape technique.

(Abscissa is the distance of recoil collection from counter with conveying tape moving 20 cm / sec).

Fig. 2. Photon spectrum of Pu^{237m} isomeric transition. (The possible peak at 105 kev is the position of K x-rays).

Fig. 3. Partial decay scheme of Cm^{241} showing level assignments in terms of Nilsson asymptotic quantum numbers. I is the spin; Π, the parity; N, the principal oscillator quantum number; n_Z, the Z-axis oscillator quantum number; and Λ, the nucleon orbital-angular-momentum component along the symmetry axis.

REFERENCES

1. Frank Asaro and I. Perlman, Phys. Rev. (In press).
2. Huizenga, Engelkemeir, and Tomkins, reported at Washington APS meeting, April 1957 in conjunction with paper: Bull Am. Phys. Soc. 2, 198 (1957).
3. Bohr, Fröman, and Mottelson, Dan. Mat. Fys. Medd. 29, No. 10 (1955).
4. Frank Asaro and I. Perlman, Phys. Rev. 88, 828 (1952).
5. Gol'din, Tret'yakov, and Novikova, Conf. Acad. Sci. USSR on Peaceful Uses of Atomic Energy, Phys. Math. Sci,, p. 226 (1955).
6. K.L. Vander Sluis and J.R. McNally, Jr., J. Opt. Soc. Amer. 45, 65 (1955).
7. Hutchison, Jr., Llewellyn, Wong, and Dorain, Phys. Rev. 102, 292 (1956).
8. J.O. Newton, Nuc. Phys. 3, 1957 (in press).
9. S.G. Nilsson, Dan. Mat. Fys. Medd. 29, No. 16 (1955).
10. M. Goldhaber and A.W. Sunyar Chapter XVI in Beta and Gamma Ray Spectroscopy , K. Siegbahn, Ed. (Interscience, New York 1955).
11. G.H. Higgins, Ph.D. Thesis, UCRL-1796 (June 1952).
12. Glass, Carr, Cobble and Seaborg, Phys. Rev. 104, 434 (1956).
13. Asaro, Amiel, Stephens, and Perlman, unpublished data (1957).
14. L.A. Sliv and I.M. Band, privately circulated copy of a compilation appearing partially in J. Exp. Theor. Phys. (USSR) 31, 134 (1956).
15. Rose, Goertzel, and Swift, privately circulated tables and Appendix IV of Siegbahn (see Ref. 10).
16. Calculated from formulas given by S.A. Moszkowski, Chapter XIII in Siegbahn (see Ref. 10).
17. Asaro, Thompson, and Perlman, Phys. Rev. 92, 695 (1953).
18. M.E. Rose, privately circulated tables of M-shell conversion coefficients.
19. R.W. Hoff and M. Kalkstein (private communication).
20. See discussion by I. Perlman and J.O. Rasmussen "Alpha Radioactivity" UCRL-3424 (June 1956); to be published in Handbuch der Physik, Vol 42, (Springer-Verlag, Berlin).

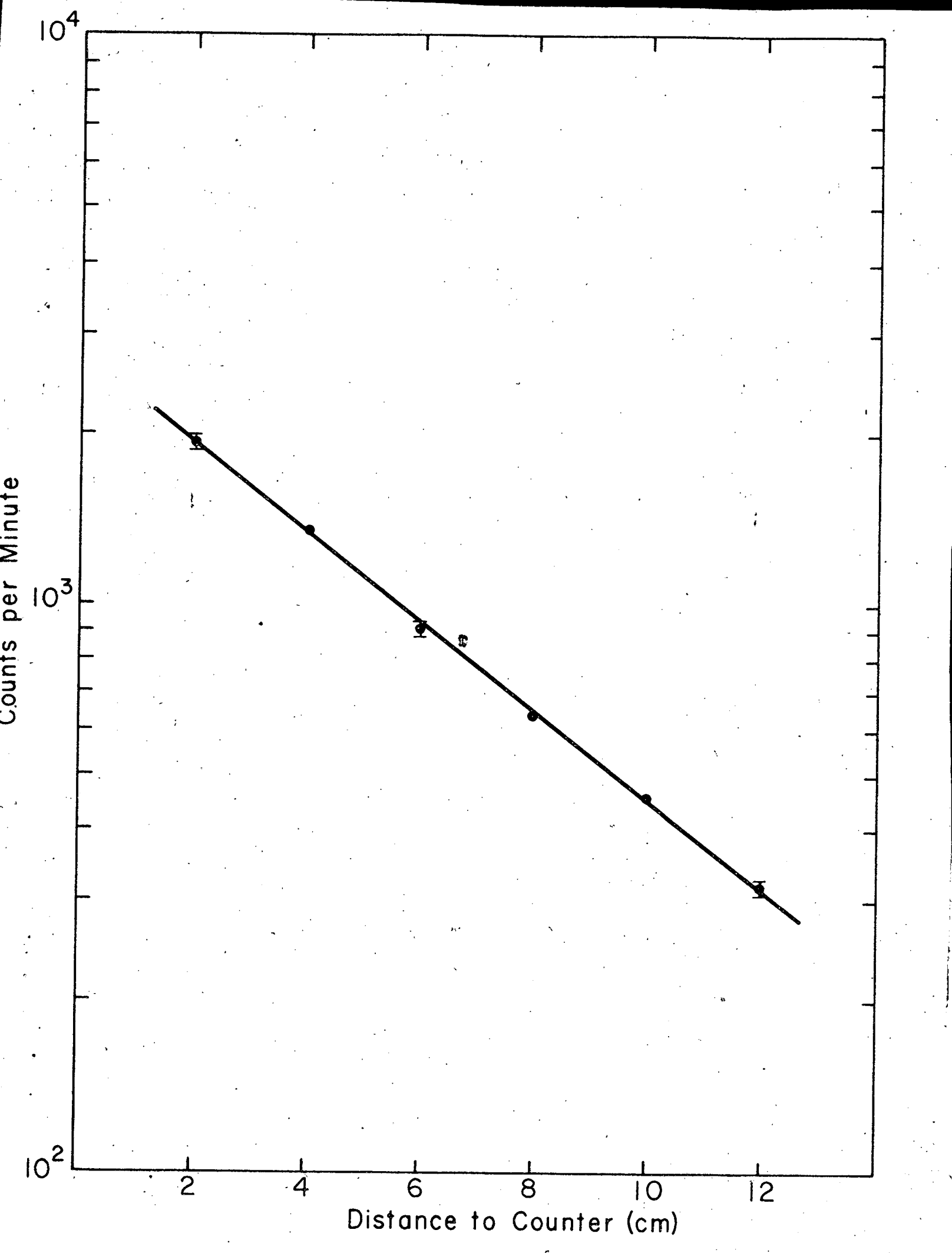
10^4
10^3
10^2
Counts per Minute
2
4
6
8
10
12
Distance to Counter (cm)

Fig 1

51661-1

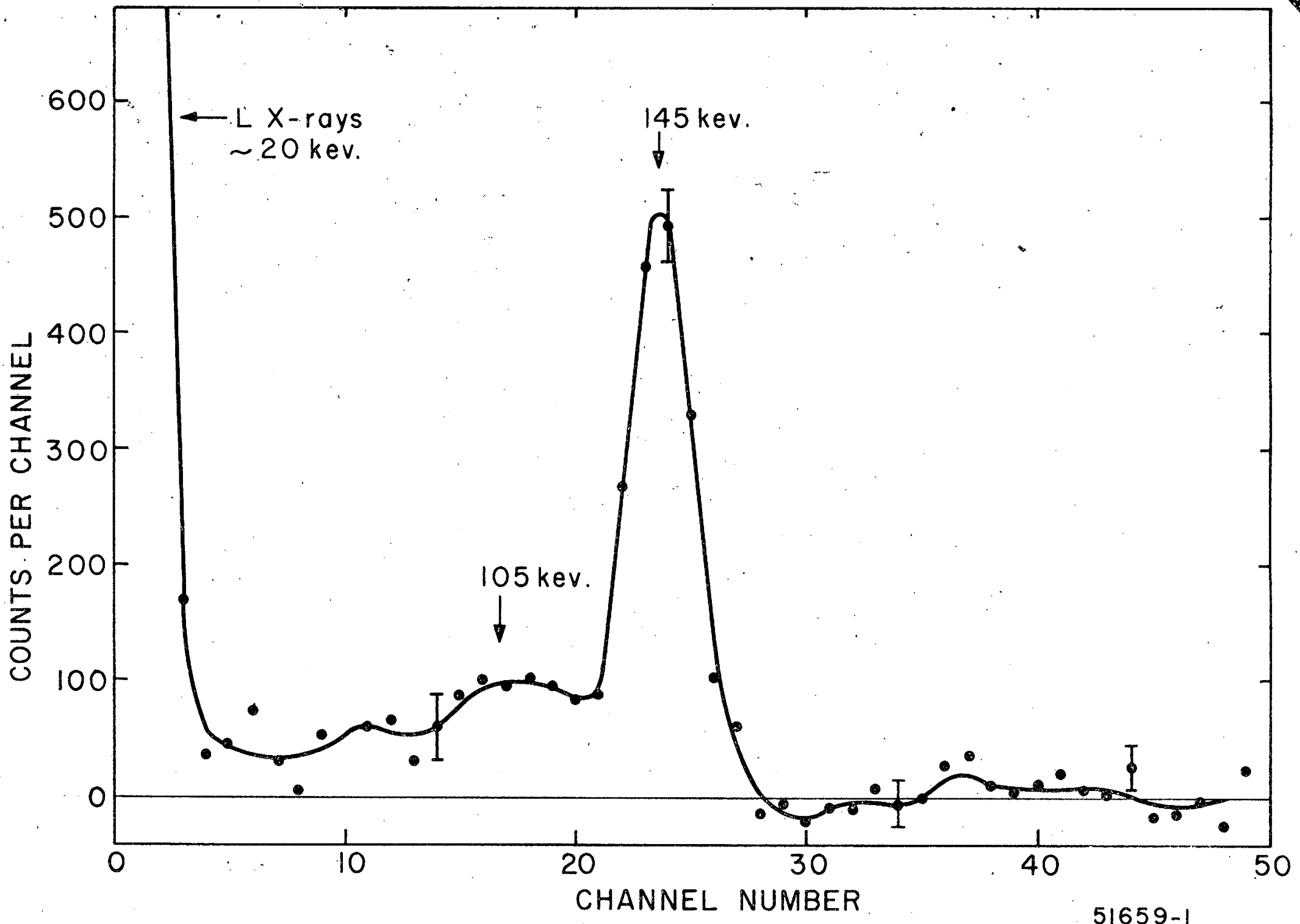
600
500
400
300
200
100
0
COUNTS PER CHANNEL
L X-rays
~20 kev.
145 kev.
105 kev.
0
10
20
30
40
50
CHANNEL NUMBER
51659-1

Fig 2

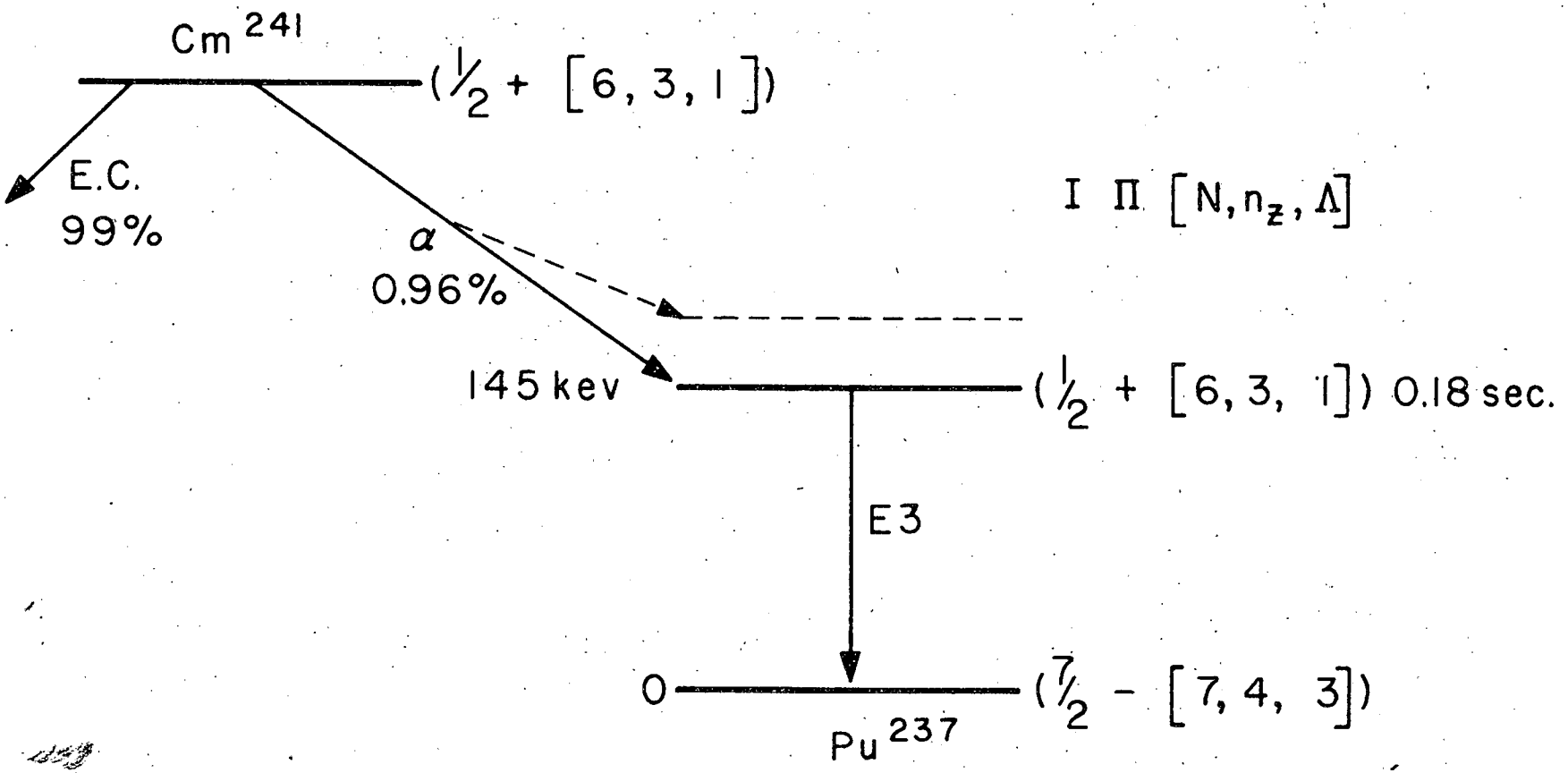
Cm 241
(1/2 + [6, 3, 1])
E.C.
99%
α
0.96%
I Π [N, n_z, Λ]
145 kev
(1/2 + [6, 3, 1]) 0.18 sec.
E3
0
(7/2 - [7, 4, 3])
Pu 237

Fig 3

51660-1