Decay of the Cerium-139 Isomers

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The decay of the 140-day ground state of Ce^{139} has been reinvestigated. A 4π scintillation spectrometer has been used to show that K-electron capture transitions between ground states is less than one percent of all K-electron captures. The same instrument has been used to measure the fraction of all transitions which terminate at the 166-kev level due to K-electron capture, and also has been used to determine the conversion coefficient of the 166-kev transition. The K-conversion coefficient was found to be 0.22 ± 0.01. K-electron capture accounts for 73% of all transitions to the 166-kev level. The energy available for decay to the excited level is 104 ± 6 kev as computed from the experimental capture ratio. A value of 0.88 ± 0.01 was obtained for the K-fluorescence yield at Z of 57 from beta-ray spectrometer measurements and coincidence counting. An isomeric level of 740 ± 5 kev above the ground state of Ce^{139} has been found. The half-life of the transition is 55 ± 3 sec and its K-conversion coefficient is 0.08 ± 0.02. Mass and element assignments have been established and approximate cross sections for activation of each level in Ce^{139} by the (n,γ) reaction on Ce^{138} in the graphite reactor have been determined.

I. INTRODUCTION

THE decay of the ground state of Ce^{139} by electron capture through the 166-kev level in lanthanum has been investigated by Pruett and Wilkinson.[1] They have determined the K-conversion coefficient of the 166-kev level to be 0.20 ± 0.05 and the $K/(L+M)$ conversion ratio to be 6.6 ± 0.3. An excited level of 163-kev energy follows beta decay of the 85-min Ba^{139}. The K-conversion coefficient for this transition has been recorded as 0.20 ± 0.01 by Nussbaum and van Lieshout[2] and as 0.22 by Mitchell and Hebb.[3] The latter investigators also report the $K/(L+M)$ conversion ratio to be 7.0. Gerholm and de Waard[4] have reported the half-life for the decay of the 163-kev transition following the beta decay of Ba^{139} to be 1.5×10^{-9} sec; consistent with this value is the observation[1] that the transition following Ce^{139} decay has a half-life less than 10^{-8} sec. Since the values reported for the characteristics of the radiations following the decay of Ce^{139} and Ba^{139} are in agreement, it seems probable that the same transition is being observed. Although no direct evidence for a ground-state to ground-state transition in the decay of Ce^{139} has been reported, Pruett and Wilkinson indicate that a small amount is probable. The fraction of the electron-capture transitions to the excited level due to K-electron capture is given[1] only as a function of the branching ratio to the ground state. Finally, a value, 0.94 ± 0.02, reported[1] for the K-fluorescence yield at Z of 57 is said to be in disagreement with the curve of Broyles *et al.*[5]

Hill[6] has looked for an isomeric state of Ce^{139} and reports that none exists with a half-life longer than one day. Since the expected isomeric state in Ce^{137} has been reported[7] recently, it is of interest to find this level in Ce^{139} to show the effect of the added neutrons on the level spacings.

In the present studies a 4π-scintillation spectrometer and a xenon-filled proportional-counter spectrometer used as a critical detector have been employed in addition to the usual magnetic lens spectrometer and coincidence equipment with energy discrimination. With these instruments it has been possible to measure the K-conversion coefficient of the 166-kev transition and the K-electron capture fraction with accuracies of 5%. A maximum limit has been placed on the ground-state to ground-state K-capture transitions. The K-fluorescence yield has been redetermined. A new isomer of Ce^{139} has been found and its energy, half-life, and conversion coefficient have been determined. A mass assignment, element assignment, and the reactor production cross section measurement for each isomer has been made.

II. ELECTRON CAPTURE OF Ce^{139}

A. Irradiations and Source Preparations

The 140-day Ce^{139} activity was prepared both by the (p,n) reaction on La^{139} and by the (n,γ) reaction on the natural barium isotopes. The proton irradiations were made using the 22-Mev proton beam of the ORNL 86 inch cyclotron. The irradiation of barium was made on the Berkeley 60-inch cyclotron using 45-Mev alphas.

Cerium-free lanthanum for irradiation was prepared by precipitating zirconium iodate from a solution of lanthanum in which the trace of cerium had been oxidized by sodium bromate. Ceric iodate was carried down by the zirconium iodate. The lanthanum was then precipitated as the fluoride and converted to the perchlorate. Finally, it was precipitated as the oxalate and converted to the oxide. The cerium produced by

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irradiation was separated from the lanthanum target by precipitation with zirconium iodate. Cerium was separated from zirconium by fluoride precipitation and the final separation from traces of other rare earths was made on a Dowex 50 ion exchange column.[8] Beta spectrometer sources with low total solids were prepared on a Formvar-polystyrene film of 50-μg/cm^2 thickness which was made electrically conducting by an evaporated 50-μg/cm^2 gold film.

The barium fluoride which was irradiated with alphas contained a very small amount of cerium impurity. After irradiation this cerium was carried on one hundred micrograms of pure erbium precipitated from a solution of the target salt. The final purification was made on a Dowex 50 column. Satisfactory beta spectrometer sources of about 50-μg/cm^2 could be prepared from this material.

B. 4π Scintillation Spectrometer Measurements

When an isotope decays by electron capture to an excited level, the conversion coefficient α_K, and the capture ratio ϵ, can in principle, be computed[1] from spectrometer and coincidence counting data by means of the following relationships:

$$\alpha_K = R_4/(2R_3 - R_2), \tag{1}$$

$$(1+\epsilon) = \left[\frac{1}{\alpha_K} + \frac{1+R_1}{R_1}\right]\frac{1+\beta}{2R_3-1}, \tag{2}$$

where $1+\epsilon$ is the ratio of electron captures from all shells to K-electron captures, R_1 is the $K/(L+M)$ conversion ratio, R_2 is the ratio of K-Auger electrons to K-conversion electrons, $2R_3-1$ is the ratio of K captures to K conversions, R_4 is the ratio of K x-rays to gamma rays, and β is the ratio of ground-state to excited-state K captures. The ratio of K captures to K conversions, $2R_3-1$, can be determined[1] from the relationship

$$R_3 = \frac{N_{x\gamma}}{N_x N_\gamma} \Big/ \frac{N_{xx}}{N_x' N_x''}, \tag{3}$$

where N_x and N_γ are singles counting rates of x-rays and gamma rays and N_{xx} and $N_{x\gamma}$ are x-ray—x-ray coincidence and x-ray—gamma-ray coincidence counting rates. The counting rates N_x, N_x', and N_x'' are not necessarily equal since the detection efficiencies which obtain in the detectors used for each type of coincidence measurement may be different.

It is very difficult to make a precise determination of the conversion coefficient of the 166-kev transition or the capture ratio ϵ, in the decay of Ce^{139} from coincidence counts and spectrometer measurements by the application of these equations. Each of the four experimental measurements required to determine α_K by means of

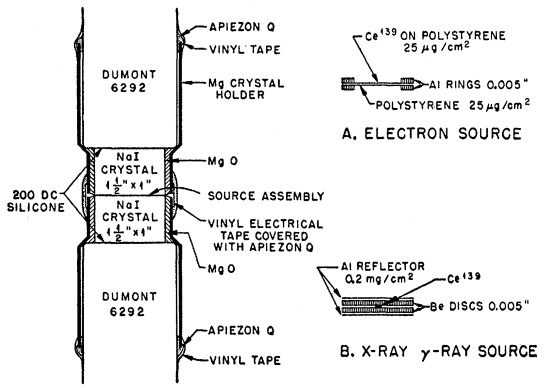

FIG. 1. 4π scintillation spectrometer and source assemblies.

Eqs. (1) and (3) is subject to rather large errors, which can be reduced only with great difficulty. Pruett and Wilkinson have stated the problem concerning the determination of the ratio of K x-ray to gamma rays R_4, and the difficulty of quantitative determinations of low-energy Auger electrons is well known. The precision of the determination of the ratio of K captures to K conversions, $2R_3-1$, from coincidence measurements is limited by uncertainties in the knowledge of scattering effects and detection efficiencies which are required to compute random coincidence corrections if these efficiencies are not very low. Therefore it is desirable to measure directly the values of α_K, ϵ, and β.

Precise determination of the conversion coefficient of the transition from the excited state following electron capture in Ce^{139} cannot be made at the present time by directly measuring the ratio of K x-rays to gamma rays with a scintillation spectrometer which has low detection efficiencies. Precise knowledge of the detection efficiencies and also the values of ϵ and β is required. However, if the efficiencies are high, as is the case with a 4π scintillation spectrometer, the precision of the determination of α_K is greatly increased. Also with such an instrument the detection of capture K x-rays which are not coincident with other radiations should permit the value of β to be measured directly. ϵ can be determined using a 4π scintillation spectrometer as has been shown by der Mateosian[9] who has made such measurements on I^{125} and Cd^{109} by growing scintillation crystals which contain these activities.

Since a 4π scintillation spectrometer of very high resolution is required for the present problem, an attempt was made to use commercially available crystals. The detector used in this work consisted of a matched pair of 1-in.$\times 1\frac{1}{2}$-in. thallium activated sodium iodide crystals mounted between a matched pair of DuMont 6292 photomultiplier tubes as shown in Fig. 1. The entire assembly was compressed between two aluminum base plates by means of four $\frac{3}{8}$-in. threaded aluminum rods. Whereas the detector could be dis-

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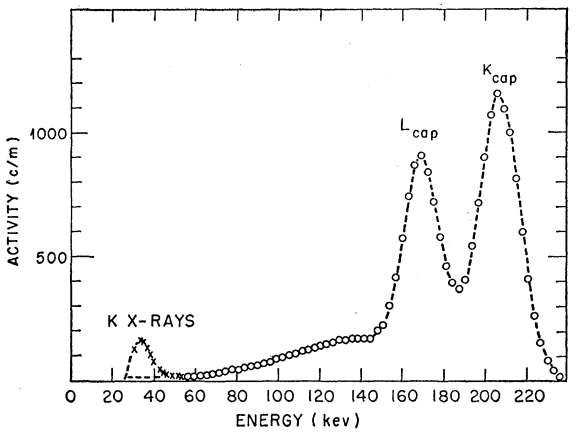

FIG. 2. 4π scintillation spectrometer electron source spectrum.

assembled easily in order to change sources, the spectra were observed to be stable for several weeks. The gains of the photomultipliers were controlled from a single high-voltage source using a voltage divider to match the output pulse amplitudes. The output signals were fed into a single preamplifier, through a double delay-line differentiated linear amplifier[10] and a sixty-channel pulse-height analyzer. The full width at half-height for the photoelectric peak from the 661-kev gamma ray of Cs^{137} was 7%.

Since the decay of the excited state of Ce^{139} is fast[4] compared to the integrating time of the detection system, radiations due to a capture process and the succeeding decay of the excited state will produce a single pulse. If a thin source such as is shown in Fig. 1(A) is used, Auger electrons and photoelectrons as well as x-rays and gamma rays will be detected. The spectrum from such a source should consist of a peak at the gamma-ray energy plus the K binding energy when K-electron capture occurs and all radiations are detected. Capture of any less tightly bound electron should result in a peak at essentially the gamma-ray energy. If there is K-electron capture to the ground state a peak at the K-shell binding energy should be observed. Figure 2 shows an energy spectrum recorded with this type of Ce^{139} source. Details of the source assembly are shown in Fig. 1(A). The three predicted peaks are observed. The broad distribution between 50 kev and 150 kev is thought to be due to partial energy loss by some of the conversion electrons. Since about 3% of the gamma rays escape detection in crystals of the size used a correction must be applied to the intensity of the K x-ray peak in order to obtain the intensity of the K x-rays due to K-capture branching to the ground state. After this correction has been made it is concluded that the ratio of ground-state to excited-state K-electron captures β in Ce^{139} is zero within 1%. Further analysis of this spectrum does not seem warranted since the distribution of pulses beneath the capture peaks due to energy loss of conversion electrons is not readily calculable.

If sufficient absorber is placed on each side of a Ce^{139} source in the 4π detector so that neither Auger electrons nor conversion electrons reach the crystals, peaks corresponding to capture x-rays coincident with gamma rays should be observed as above. In addition a peak due to x-ray—x-ray coincidences should result from K capture followed by L conversion and L capture followed by K conversion. A fourth peak should be observed due to K capture followed by K conversion. Figure 3 shows the spectrum observed with a Ce^{139} source mounted between Be absorbers. Details of the source mount are shown in Fig. 1(B). From the energies at which the peaks A and B occur it is evident that with this detector L x-rays are not contributing to the pulse height appreciably. This is not surprising if the nature of the crystal surfaces and the large absorption coefficient of these x-rays in sodium iodide are considered. However, complete or partial failure to detect the L x-rays does not interfere with the determination of α_K and ϵ from the spectrum. After corrections, which will be discussed below, either of the peak area ratios B/D or A/C will be the K-conversion coefficient α_K, and either of the peak area ratios C/D or A/B will be the capture ratio ϵ.

The corrections which must be applied arise as a result of failure to detect a K-shell vacancy or a gamma ray. These losses cause pulses which should be observed in peaks D and B to be observed in peak C or peak A. A K-shell vacancy will not be observed if an Auger event occurs or if a K x-ray is lost in the beryllium used to absorb the conversion electrons. A gamma ray may be lost in the absorber or may pass through the crystal undetected. Since the largest correction results from losses due to the Auger effect, the fluorescence yield must be known. Uncertainty of the correct value led to a new determination of the fluorescence yield described below. The x-ray absorption losses were computed from known absorption coefficients and geometries. The method of applying these corrections in order to determine the number of events of each

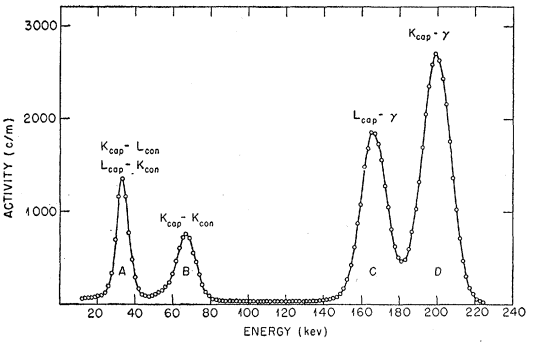

FIG. 3. 4π scintillation spectrometer x-ray-γ-ray source spectrum.

[10] Details of this instrument will be published soon in the Review of Scientific Instruments by E. Fairstein.

type in the decay process is summarized in the following equations.

$$N_{K\text{cap-}\gamma}=\frac{D}{P_K P_\gamma}, \tag{4}$$

$$N_{L\text{cap-}\gamma}=\frac{C}{P_\gamma}-\frac{D(1-P_K)}{P_K P_\gamma}, \tag{5}$$

$$N_{K\text{cap-}K\text{con}}=\frac{B}{P_K{}^2}, \tag{6}$$

$$N_{L\text{cap-}K\text{con}}=\frac{A}{P_K}-\frac{D(1-P_K)}{P_K P_\gamma}-\frac{2B(1-P_K)}{P_K{}^2}-\frac{B}{RP_K{}^2}. \tag{7}$$

$N_{K\text{cap-}\gamma}$ is the number of K-captures followed by gamma-ray emission and $N_{K\text{cap-}K\text{con}}$ is the number of K captures followed by K-conversion electron emission. A, B, C, and D are the areas of peaks A, B, C, and D of Fig. 3; P_K and P_γ are the probabilities of detection of a K-shell vacancy and a gamma ray, respectively. R is the K/L conversion ratio. The relative importance of the corrections can be derived from the following relative peak areas and efficiency factors:

A	B	C	D	P_K	P_γ	R
0.22	0.2	0.62	1.0	0.848	0.953	7

The importance of absorption corrections and assembly technique was ascertained by observing the spectra of seven different source assemblies. The value of ϵ from the areas of peaks C and D is 0.37±0.02 and the value of α_K from the areas of peaks B and D is 0.22±0.01. Due to the fact that many different types of events produce pulses which are observed in peak A, as is indicated in Eq. (7) above, values of ϵ and α_K computed from the ratios A/B and A/C are not precisely known, though consistent with the values recorded above.

C. Coincidence Measurements

In order to demonstrate the consistency of the values of ϵ and α_K by means of Eq. (2), the x-ray—x-ray and x-ray—gamma-ray coincidence measurements required to determine R_3 by Eq. (3) were made. For these measurements two separate scintillation spectrometers with crystals 1 in.×1½ in. were used in coincidence. Measurements were made with several geometrical arrangements to establish the absence of scattering effects or erroneous random coincidence corrections. The value of $2R_3-1$ obtained is 4.25±0.2, consistent with the value reported by Pruett and Wilkinson.[1] ϵ is computed by Eq. (2) to be 0.35±0.08 if this value of $2R_3-1$ is used together with 0.22±0.01 for the value of α_K. This value of ϵ is consistent with the value determined by the 4π scintillation spectrometer method but does not agree with Pruett and Wilkinson's value of 0.21 if β is taken as zero.

TABLE I. Summary of experimental results.

Ratio	Measured value	Notation
K/L conversions	7.2 ±0.5	R
L/M conversions	3.7 ±0.3	···
$K/(L+M)$ conversions	5.7 ±0.3	R_1
K Augers/K conversions	0.62±0.04	R_2
K captures/Kconversions	4.25±0.2	$2R_3-1$
K-conversion coefficient	0.22±0.01	α_K
Electron capture ratio	0.37±0.02	ϵ
K capture to ground state	<1%	···

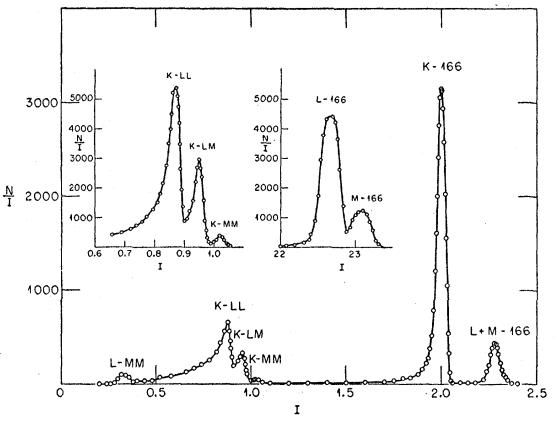

FIG. 4. Beta-ray lens spectrometer spectra.

D. Magnetic Spectrometer Measurements

Values of the fluorescence yield and the $K(L+M)$ conversion ratios are needed in order to make the necessary corrections to the data obtained with the 4π scintillation spectrometer. Some discrepancy exists among reported values of the fluorescence yield.[1,5] The value of the ratio R_1 is reported by Mitchell and Hebb[3] to be 7.0 from studies of the decay of Ba^{139} in agreement with Pruett and Wilkinson's value of 6.6±0.3.

Sources of the order of 50-μg/cm² thickness were measured with a thin magnetic lens spectrometer. The counter window cutoff was at about 1.5 kev so no window correction was required in the region of the K-Auger electrons. The spectrum is shown in Fig. 4. The inserts show data for the Auger electron region and the L- and M-conversion electron region taken at higher resolution with a different source. The results of these measurements are summarized in Table I along with observed and derived results from 4π scintillation spectrometer and coincidence measurements.

From these measurements the fluorescence yield can be computed by using the relationship

$$f_K=1-(R_2/2R_3).$$

The value obtained is 0.88±0.01 which is in agreement with the smooth curve of Broyles, Thomas, and Haynes.[5]

III. ISOMERIC TRANSITION IN Ce^{139}

Since the Ce^{139} nucleus contains eighty-one neutrons, oen less than a filled shell, the energy difference between the $h_{11/2}$ and $d_{3/2}$ levels is expected to be small enough so that the transition between them would have a measurable half-life. The transition energies of the isomeric states of other isotopes with eighty-one neutrons, Te^{133}, Xe^{135}, and Ba^{137}, have been plotted against their proton numbers and the extrapolation indicates that the $h_{11/2}$ level of Ce^{139} should fall about 800 kev above the $d_{3/2}$ ground state. An isomeric transition of this energy would be expected to have a half-life of the order of minutes and the conversion coefficient would be so low that the unconverted gamma ray should be sought rather than conversion electrons or x-rays.

A rapidly decaying energetic gamma ray was detected when a sample of enriched Ce^{138} was irradiated in the ORNL graphite reactor for one minute and its radiations were observed with an activated sodium iodide scintillation spectrometer. The gamma-ray energy was measured as 740±5 kev when compared with the 661-kev gamma of Cs^{137} and the 768-kev gamma of Nb^{95} on a scintillation spectrometer. The half-life was measured to be 55±3 sec. The K-conversion coefficient was measured with a scintillation spectrometer and multichannel analyzer to be 0.08±0.02. The x-ray and gamma-ray efficiencies of this detector were determined with samples of 140-day Ce^{139} and 35-day Nb^{95}, whose disintegration rates were known from coincidence counts. Enriched cerium isotopes were used to establish the mass number as 139. The x-rays from conversion were shown to be cerium x-rays by comparison of the escape peaks in a xenon proportional counter spectrometer with those observed in the decay of Ce^{137m} and the 140-day Ce^{139} which emit x-rays of cerium and lanthanum, respectively. These spectra will be shown in a forthcoming article[11] on La^{137} and Ce^{137}.

IV. ACTIVATION CROSS-SECTION MEASUREMENTS

The cross sections for production of each of the isomers of mass 139 by the (n,γ) reaction in the neutron distribution existing in the graphite reactor have been determined. The value obtained for production of the 55-sec isomer is 7 mb as determined from comparison of its x-ray intensity with that of the 8-hr ground-state transition of Ce^{137} whose production cross section has been measured.[11] The cross section to yield the 140-day isomer of Ce^{139} is about one barn as determined by assaying the amount of 166-kev gamma relative to the 145-kev gamma of Ce^{141} in a source of cerium irradiated in the reactor. The cross section of Ce^{141} was taken as 0.6 barn. In order to resolve the 166-kev gamma present in so low an intensity relative to the 145-kev gamma its photopeak was accentuated by observing x-ray—gamma-ray coincidence. The coincidence counter was calibrated with a sample of 140-day Ce^{139} of known disintegration rate.

The cross-section values of 7 mb and 1 b for production of the isomeric state and the ground state are known to within about a factor of two. The latter value is not considered to be in disagreement with the value 9 b given by Pomerance[12] since his value is known only to ±100%. The 1 b figure is consistent with the reported[13] ratio $\sigma_{138}/\sigma_{140}$ of 1.4.

V. DISCUSSION

A decay scheme which is consistent with all the above observations is shown in Fig. 5. The ground-state spin of La^{139} has been measured[14] and found to be 7/2. Using the single particle shell model, this ground state would be expected to be $g_{7/2}$. The K/L conversion ratio 7.2±0.5 for the 166-kev transition of La^{139} is in agreement with the computed value[15] for an $M1$ transition within the uncertainty. The half-life of this transition also indicates[4] the magnetic dipole characterization. However, although the conversion coefficient reported here agrees with the value 0.23 computed by Rose *et al.*[15] for an $M1$ transition of this energy and atomic number, it cannot be used to distinguish between an $M1$ and $E2$ transition in this case. The $d_{5/2}$ assignment to the excited level is consistent both with the observations and the shell theory. Ambler *et al.*[16] have reported that the spin sequence $3/2^+ \rightarrow 5/2^+ \rightarrow 7/2^+$ is consistent with their observations of the gamma-ray anisotropy of aligned Ce^{139} nuclei. Also they have stated that the 166-kev gamma-ray transition is $M1$ with 4% admixture of $E2$. The small effect of this mixing upon the K/L conversion ratio is within the experimental error of our observations.

The isomeric transition of Ce^{139} appears to be an $M4$ transition since its conversion coefficient value 0.08 ±0.02 agrees with the theoretical value[15] of 0.075 for this energy and atomic number. The observed partial lifetime for gamma-ray emission is 86 sec which is consistent with the lifetime of 94 sec computed from the lifetime *vs* energy relationship obtained by Goldhaber and Sunyar[17] from a fit to more than thirty $M4$ transitions. The energy of the transition is about that expected from the extrapolation of the curve of energy *vs* proton number for metastable states of other eighty-

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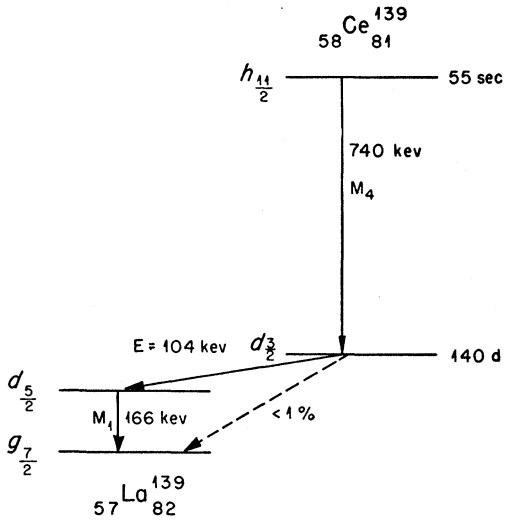

FIG. 5. Decay scheme of the Ce^{139} isomers.

one neutron nuclei. Again the $h_{11/2}$ and $d_{3/2}$ assignments to the isomeric level and the ground state are consistent with the spin and parity change required for an $M4$ transition and also with the shell model predictions.

The spin assignments for the 140-day ground-state decay made here are the same as those previously given.[1,4] On the basis of these spins and parities the decay of the 140-day ground state to the excited state of La^{139} should be allowed. However, the ground-state to ground-state transition should be second forbidden. Therefore, it is not unexpected that the value of β, the ratio of ground-state to excited-state K captures, should be essentially zero.

As was indicated above the values reported here for the conversion coefficient, the capture ratio ϵ, the $K/(L+M)$ conversion ratio, and the ratio of K captures to K conversions are consistent. However, a discrepancy exists between the value of the capture ratio 0.37 ± 0.02 reported here and 0.21, which would be the value of Pruett and Wilkinson[1] if the ground-state to excited-state K-capture ratio β is taken as zero. Also, the $K/(L+M)$ conversion ratio 5.7, reported here is lower than the values reported by Mitchell and Hebb[3] and Pruett and Wilkinson. This latter discrepancy is not understood. However, because R_1 is large relative to α_K this discrepancy has very little effect on the value of ϵ computed from Eq. (2). The cause of the discrepancy between the values of the capture ratio appears to lie in the large error in ϵ resulting from relatively small errors in the K-conversion coefficient α_K, and the ratio of K captures to K conversions R_3.

The large value of the observed capture ratio relative to the ratio of L_1 to K-electron densities at the nucleus, computed by Rose and Jackson[18] to be 0.124 for Z of 57, is taken to indicate that the energy available for electron capture is not large relative to the K binding energy. As has been stated above the capture transition to the excited level in La^{139} is allowed on the basis of what now appears to be well-established level assignments. Therefore the electron capture transition energy can be computed[19] from the value of ϵ, the electron densities at the nucleus and the electron binding energies.[20] The K-shell and L-subshell radial wave functions required for this computation were obtained from the work of Brysk and Rose.[21] In order to approximate the M_1 and N_1 shell-capture contributions, the Hartree self-consistent field wave-function values[22] for Cs^+ were used. The capture transition energy to the excited level so obtained is 104 ± 6 kev. The corresponding $\log ft$ value is 5.3 ± 0.1 if the half-life value, 140 ± 1 days, reported by Pool and Krisberg[23] is correct. The energy value is reasonable according to the disintegration energy systematics of Way and Wood[24] and the $\log ft$ value is consistent with the prediction that electron capture to the excited state is allowed.

ACKNOWLEDGMENTS

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