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Disintegration of Ce^{139}[†]

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The decay of Ce^{139} is shown to proceed by s-electron capture to a single excited state of La^{139} with the subsequent emission of an internally converted gamma ray. Magnetic beta-ray spectrometry and scintillation coincidence techniques have been used to show that the 166-kev gamma ray is magnetic dipole in character. The measured $K/(L+M)$ ratio is 6.6 ± 0.3. An estimation of the K internal conversion coefficient yields 0.20 ± 0.05 and the lifetime of the excited level is found to be less than 10^{-8} sec. An upper limit of 315 kev is found for the total disintegration energy. Although no estimate of orbital electron capture to the ground state can be made, a small amount is probable. A value of 0.94 ± 0.02 for the K fluorescent yield at $Z=57$ is obtained from the experiment.

INTRODUCTION

THE decay of Ce^{139} (140 days) occurs by orbital electron capture[1–3] and leads to an excited state of La^{139}. The subsequent gamma-ray spectrum, although simple, has not been clearly established by previous investigations. The presence of a strong internally converted 166-kev gamma ray is certain,[4–6] but there is considerable variance concerning the nature of additional radiations. Pool and Krisberg,[3] for example, have reported an 0.8-Mev gamma ray, while Keller and Cork[6] have suggested a 275-kev gamma ray in cascade with the 166-kev radiation. Further progress in the study of this element apparently has been hampered by the lack of strong sources and the problem of contaminants.

In the present paper the results of a detailed study of this element are given. Intense cyclotron bombardments and improved chemical procedures have yielded reliable and reasonably strong sources. The radiations have been studied with a small 180° shaped-field beta-ray spectrometer ($\sim$1 percent transmission, $\sim$1 percent resolution), with a flat-field permanent magnet spectrometer (126 gauss), and with a scintillation coincidence spectrometer. The decay is found to involve a single gamma ray. The multipolarity of the gamma ray has been determined from a measurement of the K/L ratio. An upper limit to the lifetime of the excited state has been obtained as well as an estimation of the internal conversion coefficient. An upper limit to the energy released in the decay has been established from the L_{I}/K capture ratio. The upper limit on the ft value which results, together with the multipole assignment of the gamma transition, allow an assignment of spins which is in agreement with the shell model. As a by-product, the K fluorescent yield of the x-rays for $Z=57$ has been obtained.

PREPARATION OF SOURCES

Ce^{139} was produced by bombarding lanthanum oxide with 11-Mev deuterons. Targets received from 500 to 1000 microampere hours. For the initial survey, the cerium was separated from the target material by oxidation to the +4 valence state and precipitated as the iodate. For quantitative measurements requiring higher specific activity the cerium in the +4 state was extracted into tributyl phosphate without the addition of cerium carrier.[7] Source material prepared in this way was not entirely carrier-free, owing to the presence of a small amount of cerium in the target material. Spectrometer sources were mounted in the usual manner on zapon (or aluminum coated zapon) films. Source thickness, including backing, ranged from 0.1 to 0.2 mg/cm^2.

MAGNETIC SPECTROMETER MEASUREMENTS

A survey of the electron spectrum from 2 kev to 2 Mev in a small shaped-field spectrometer (7.5-cm radius) yielded only the Auger electrons and the K, L, and M conversion lines corresponding to a gamma ray of energy 166.5 kev. A search for positrons gave negative results. The Auger and conversion lines as obtained with a source of about 0.1 mg/cm^2 thickness are shown in Fig. 1. The counter window cut-off occurred at approximately 1.5 kev. Calibration and window transmission in the region of the Auger electrons were checked by comparison with the well-known A and F conversion lines of ThB. The values of the $K/(L+M)$

TABLE I. Summary of experimental data.

Ratio	Notation	Measured value
K/L conversion	···	8.20 ± 0.40
L/M conversion	···	4.00 ± 0.20
$K/(L+M)$ conversion	R_1	6.60 ± 0.30
Augers/K-conversions	R_2	0.34 ± 0.03
K-captures/K-conversions	$2R_3-1$	4.40 ± 0.20
K x-rays/gamma rays	R_4	1.00 ± 0.20

[†] Supported by the joint program of the Office of Naval Research and the U. S. Atomic Energy Commission.

[1] M. L. Pool and J. D. Kurbatov, Phys. Rev. **63**, 463 (1943).

[2] D. E. Mathews and M. L. Pool, Phys. Rev. **72**, 163 (1947).

[3] M. L. Pool and N. L. Krisberg, Phys. Rev. **73**, 1035 (1948).

[4] J. B. Chubbuck and J. Perlman, Phys. Rev. **74**, 982 (1948).

[5] R. D. Hill, Phys. Rev. **82**, 449 (1951).

[6] H. B. Keller and J. M. Cork, Phys. Rev. **84**, 1079 (1951).

[7] This procedure is described in detail by J. C. Warf, Iowa State College, U. S. Atomic Energy Commission Report AECD-2524 (unpublished).

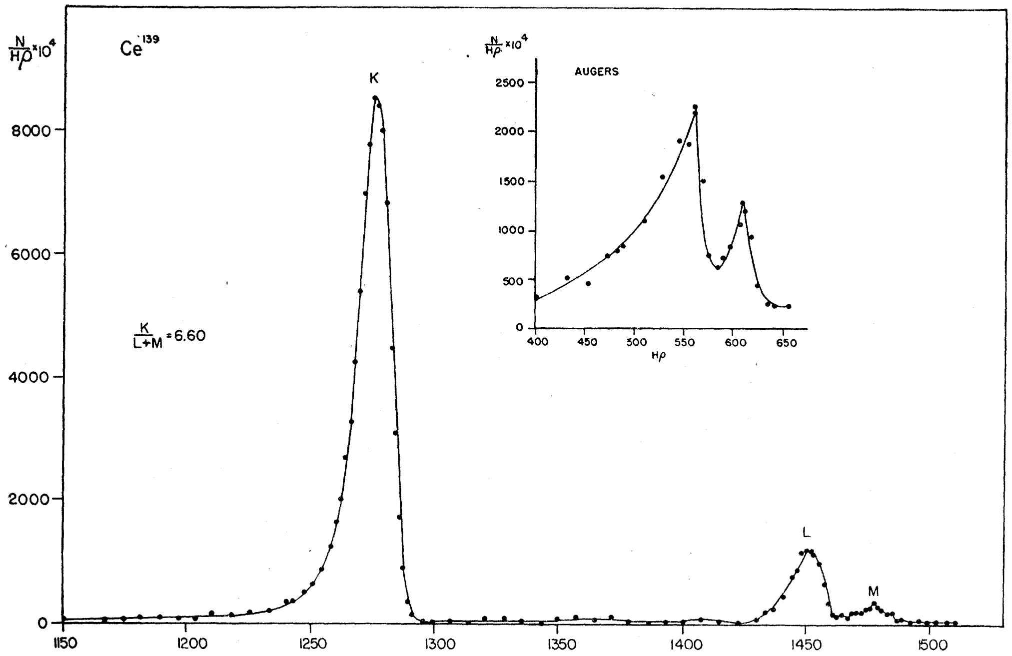

FIG. 1. Negatron spectrum of Ce^{139}.

conversion ratio R_1, and the Auger/K-conversion ratio R_2, which are listed in Table I, are the result of a number of runs.

As a further check on these results the electron spectrum was obtained in a permanent-magnet spectrometer utilizing photographic detection. A long exposure taken 10 months after the irradiation did not bring to light any additional features of the negatron spectrum in the region 0.01 to 0.5 Mev. The energy of the gamma ray obtained from these data is 165.5 kev.

SCINTILLATION SPECTROMETER MEASUREMENTS

Additional information concerning the radiations of Ce^{139} was obtained from scintillation spectrometry. Two scintillation spectrometers in a coincidence arrangement having a coincidence resolving time of 2×10^{-7} sec were used. This apparatus is a modification of one described by Roggenkamp.[8] The detectors in this case were 1-in. ×1-in. cylindrical canned NaI(Tl) crystals mounted on DuMont 6292 photomultiplier tubes.

A survey of the photoelectron spectrum in a single channel from 25 kev to 1.6 Mev showed only peaks due to La K x-rays and the 166-kev gamma ray. Higher-energy gamma rays, if present at all, have an intensity of less than 10^{-3} of the intensity of the 166-kev radiation. The pulse-height distribution taken in channel 2 is shown in Fig. 2 (squares). The Compton electron distribution is very small in this case since almost total absorption takes place in the detector at these energies.

With channel 1 set to accept only the x-ray, the coincidences which occurred as channel 2 scanned the spectrum are shown in Fig. 2 (circles). The x-x and x-γ coincidence peaks are expected if the decay of Ce^{139} occurs by s-electron capture to the 166-kev level in La^{139} followed by the emission of an internally converted gamma ray. The inset of Fig. 2 shows the coincidence peak which resulted when channel 1 was set to register the gamma ray as channel 2 surveyed the spectrum.

If the coincidence curve shown in Fig. 2 is normalized to the singles curve at the gamma ray peak, the ratio of the singles x-ray counting rate to the coincidence x-ray counting rate for corresponding pulse-height settings near the peak will not depend on the over-all efficiency factor of either counter. In terms of the coincidence rates, $N_{x\gamma}$ and N_{xx}, and the singles counting rates, N_x and N_γ, in channel 2, this ratio is

$$R_3=\frac{N_{x\gamma}}{N_\gamma}\Big/\frac{N_{xx}}{N_x}.$$

By an extension of the analysis described by Mitchell,[9] it may be shown that $(2R_3-1)$ represents the ratio of the total K-shell captures to the number of K conversions. The quantity R_3 as determined at a number of corresponding points on the curves near the peaks was found to be constant. The value given in Table I represents the average over a number of points and includes a small correction for coincidences in the neighborhood of the x-ray due to the gamma-ray Compton distribution.

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[8] Roggenkamp, Pruett, and Wilkinson, Phys. Rev. **88**, 1262 (1952).

[9] A. C. G. Mitchell, Revs. Modern Phys. **20**, 296 (1948).

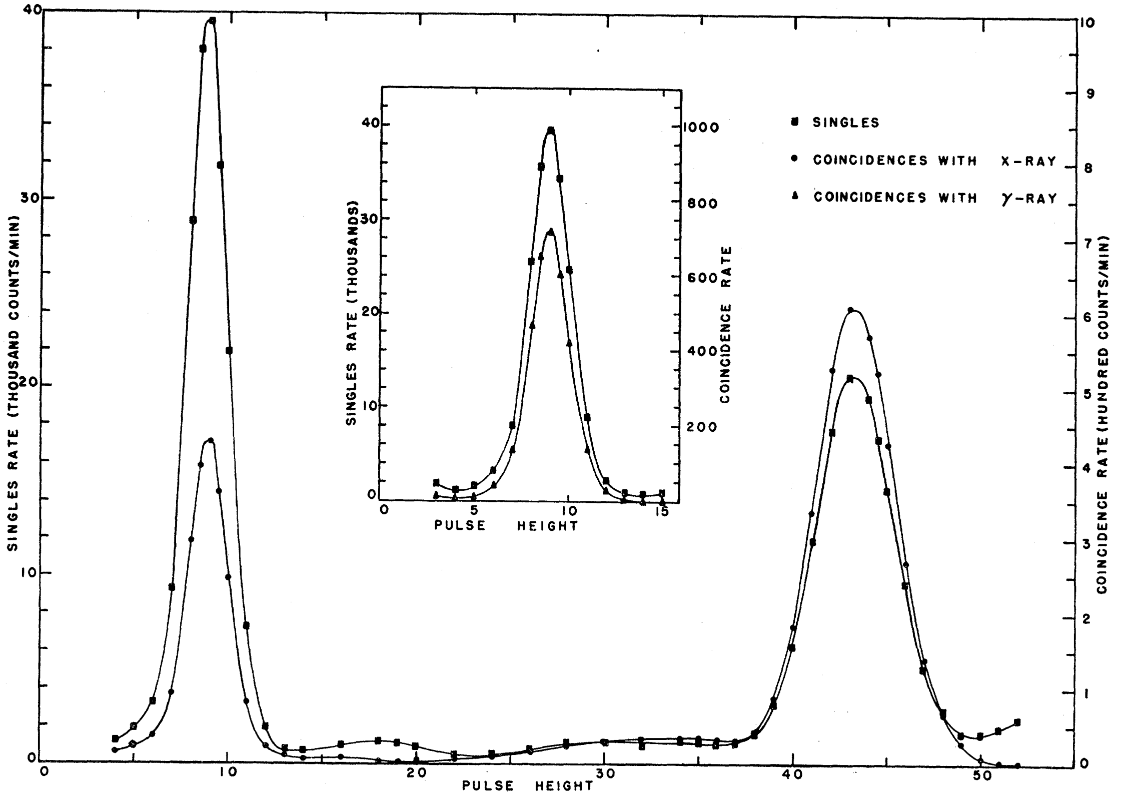

FIG. 2. A scintillation coincidence spectrum of the gamma radiation of Ce^{139}. A single-channel pulse height distribution is also plotted. Peaks are due to the 33-kev x-ray and the 166-kev gamma ray.

The ratio, R_4, of the number of K x-rays to the number of gamma rays is also of interest. The single-channel pulse-height distribution in this case was taken with a well-collimated source. The figure listed in Table I was obtained after applying corrections for energy dependence of counter efficiencies, absorption in the crystal holder, and escape of the iodine x-ray. Unfortunately, the last mentioned correction is not straightforward and consequently the number R_4 is the most uncertain of any in Table I. In fact, the K x-ray of La gives rise to a very pronounced "escape peak" in NaI. This is due to the fact that the K absorption edge of iodine lies between the energies of the $K_{\alpha 1}$ and $K_{\alpha 2}$ x-rays of lanthanum.[10] The correction applied involved the data of McGowan[11] and the known relative intensities of the La K x-ray components.

In addition to the measurements described above, a delayed coincidence experiment was performed to check the lifetime of the 166-kev excited state in La^{139}. The plot of coincidence rate *vs* artificial delay was a curve characteristic of "prompt" radiation. From the slope of the curve it is estimated that the lifetime of this state is less than 10^{-8} sec. A small asymmetry in the delayed coincidence curve was ascribed to difference in energy between the gamma ray and x-ray.

RESULTS

The data given in Table I establish most of the features of the decay scheme of Ce^{139}. The K/L ratio when compared with empirical curves similar to those of Sunyar and Goldhaber[12] incorporating the more recent data of Graham and Bell,[13] is found to lie directly on the $M1$ curve. The presence of an $E2$ admixture, if any, must be small. The upper limit of 10^{-8} sec on the lifetime of the 166-kev state, while not a definitive check on the assignment of $M1$, is consistent with it. As a further check the internal conversion coefficient may be derived from the data. It may be verified that, with no assumptions involved, the K conversion coefficient is

$$\alpha_K = R_4/(2R_3 - R_2) = 0.20 \pm 0.05.$$

Within the experimental uncertainties, especially in R_4, this is consistent with the theoretical value[14] of 0.24 for $M1$ radiation of 166-kev energy at $Z=57$. Since the $E2$ conversion coefficient differs very little from that for $M1$, no estimate of mixing can be made on this basis. The decay scheme is shown in Fig. 3. The measured spin of the ground state[15] is 7/2, hence

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[10] Hill, Church, and Mihelich, Rev. Sci. Instr. **23**, 523 (1952).

[11] F. K. McGowan, Phys. Rev. **93**, 163 (1954).

[12] M. Goldhaber and A. W. Sunyar, Phys. Rev. **83**, 906 (1951).

[13] R. L. Graham and R. E. Bell, Can. J. Phys. **31**, 377 (1953).

[14] Rose, Goertzel, Spinrad, Harr, and Strong, Phys. Rev. **83**, 79 (1951).

[15] J. E. Mack, Revs. Modern Phys. **22**, 64 (1950).

the assignment $M1$ for the gamma ray leads reasonably to the shell model predictions of $g_{7/2}$ and $d_{5/2}$ for the respective spins of the ground state and excited state. By further inference from the shell model, the orbital electron capture to the excited state should be allowed and the ground state transition 2nd forbidden since the ground state of Ce^{139} is expected to have a $d_{3/2}$ configuration.

An estimate of the energy released in the transition and ft value may be obtained by a closer examination of the quantity R_3. It may be shown that R_3 depends on the ratio of L_I to K capture to the *excited* state ϵ, the ratio of ground state to excited state K-shell capture β, the K conversion coefficient α_K, and the ratio of the total conversions to K conversions $(1+1/R_1)$. In fact, since the total orbital electron capture to the excited state is equal to the number of gamma rays plus the number of conversion electrons, then

$$(2R_3-1)(1+\epsilon)/(1+\beta)=1/\alpha_K+(1+R_1)/R_1.$$

If one uses the theoretical value of α_K for the 166-kev $M1$ transition and the data of Table I, it follows that

$$\epsilon=0.21+1.21\beta.$$

According to Rose and Jackson,[16] the value of the ratio of L_I to K electron densities at the nuclear radius is 0.124 for $Z=57$. If the energy available for electron capture to the *excited* state W_0 is large compared to the K binding energy this quantity should be essentially equal to ϵ. It will be noted that even with $\beta=0$, i.e., no K capture to the ground state, the value of ϵ is almost double Rose's theoretical value for $(\psi_{L\mathrm{I}}/\psi_K)^2$. It is to be concluded that the magnitude of the K binding energy cannot be neglected. In this case the upper limit to W_0 corresponds to $\beta=0$. If the transition to the excited state is assumed to be allowed and the value of Rose given above as well as the current electron binding energies[10] for La are used, an upper limit of $W_0=147$ kev results. The corresponding upper limit to $\log ft$ is 6.0, a somewhat large value for an allowed transition. The presence of a small amount of ground state K capture would lower both W_0 and $\log ft$. While no estimate of β can be made from these experiments it is interesting to note that if 10 percent, for example, of the K captures occur to the ground state the experimental data yield $W_0=94$ kev and $\log ft=5.3$ for the transition to the 166-kev level. The assignment of a small value of W_0 receives further support from the fact that no evidence of inner bremsstrahlung was found in the scintillation spectrum beyond the photoelectron peak of the gamma ray.

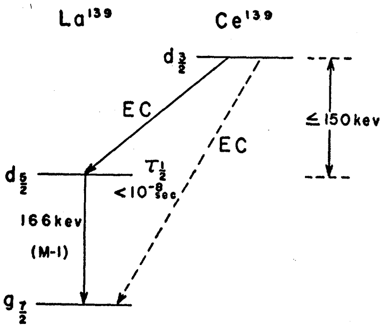

FIG. 3. Decay scheme of Ce^{139}.

The fluorescent yield f_k of the K x-rays for $Z=57$ is contained implicitly in the foregoing analysis of the data. Reference to Table I shows that this auxiliary piece of information may be obtained. It is readily seen that

$$f_k=1-(R_2/2R_3)=0.94\pm0.02.$$

It would appear that the curve of Broyles *et al.*[17] is somewhat low in this region. The spread in R_2 and R_3 necessary to bring f_k below 0.90 is considerably greater than the most generous limits which the uncertainties of the experiment allow. Evidence for this conclusion has been previously reported by Browne *et al.*[18] Their data lead to a value of 0.99 for $Z=58$ and 59 with little likelihood of experimental limitations reducing this figure below 0.90.

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[16] M. E. Rose and J. L. Jackson, Phys. Rev. **76**, 1540 (1940).

[17] Broyles, Thomas, and Haynes, Phys. Rev. **89**, 715 (1953).

[18] Browne, Rasmussen, Surls, and Martin, Phys. Rev. **85**, 146 (1952).