directly. There are a number of variations of the distorted wave treatment which are being developed at the present time to handle such cases. It is to be expected that at extremely high velocities the problem again becomes tractable by semiclassical methods, as in the case for the stopping powers of alpha particles passing through matter. Unfortunately high-temperature thermal collisions and collisions resulting in chemical reactions are usually in the intermediate velocity range which is most difficult to handle.

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Radioactive Decay of La^{137} and Ce^{137}

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A long-lived radioactive isotope of lanthanum has been produced by neutron bombardment of natural cerium. It decays with emission of Ba x-rays and has been assigned as La^{137}. The K-electron capture half-life is $(6\pm2)\times10^4$ yr calculated from the Ba K x-ray intensity and the yield of mass 137 atoms in the bombardment. Approximate measurements of neutron cross sections of Ce^{136} gave 6.3 barns for activation of Ce^{137} and 0.6 barn for activation of Ce^{137m}. A previously unobserved 10-kev $M1$ transition has been discovered in the decay of Ce^{137}. A decay scheme is proposed which has a $g_{7/2}$ ground-state spin assignment for La^{137}. Electron capture to the $d_{3/2}$ ground state of Ba^{137} on the basis of this assignment is second-forbidden.

I. INTRODUCTION

CHUBBOCK and Perlman[1] estimated that the half-life of La^{137} must be longer than 400 yr. Their estimate was based on the K x-ray counting rate of a Ce^{137} sample, the x-ray detection efficiency of their counter, and the fact that they found no evidence of radioactivity which could be attributed to a lanthanum daughter. In some recent work on the Ce^{137} isomers, the present authors[2] estimated that the La^{137} half-life must be longer than 10^5 yr if it decayed by K electron capture. However, spin assignments consistent with the experimental data indicated that the transition between the La^{137} and the Ba^{137} ground states should be allowed.

In the present work, La^{137} has been identified and found to decay by K electron capture. Since the half-life was found to be very long for an allowed transition with enough energy to permit K capture, further work was done on the Ce^{137} decay scheme in order to understand the apparent anomaly. This work has led to the discovery of a low-energy transition and to the addition of a new energy level below the ground state in the previous Ce^{137} decay scheme. On the basis of the spin assigned to the new ground state, electron capture decay of La^{137} would be second-forbidden.

II. IDENTIFICATION OF La^{137}

La^{137} was produced by bombardment of 880 mg of CeO_2 in the ORNL graphite reactor with an integrated flux of 1.3×10^{23} neutrons/cm^2. After dissolving the CeO_2 in nitric acid, a known amount of La^{140} tracer was added. Most of the cerium was removed from the lanthanum by extraction with tributyl phosphate. Final purification of the lanthanum fraction was accomplished by adsorption on a cation exchange column and elution under conditions which gave essentially complete separation of lanthanum from cerium and other rare earths.[3] Measurement of the La^{140} activity in the final sample showed that it contained 60% of the original lanthanum.

After the La^{140} tracer had decayed, scintillation spectrometer measurements showed the presence of x-rays with an energy of about 32 kev and the absence of gamma rays of higher energy. The intensity of these x-rays has remained constant for a period of more than six months, indicating a half-life longer than twenty years. The proportional counter pulse-height spectra in Fig. 1 were used to identify the radiation from the long-lived lanthanum source as barium K x-rays. These spectra were taken with a xenon filled counter and show the "escape"[4] peaks that result from critical absorption of x-rays in xenon with escape of Xe, $K\alpha$, or $K\beta$ x-rays from the counter.

In the upper curve of Fig. 1 taken with a Pm^{145} source, which emits Nd K x-rays, both the $K\alpha$ and the $K\beta$ lines are critically absorbed in xenon. In each case either xenon $K\alpha$ or $K\beta$ x-rays may escape resulting in a pulse height spectrum with four escape peaks. The most intense peak at 7.6 kev results from Nd $K\alpha$ absorption followed by Xe $K\alpha$ escape. The peak at 8.6 kev which results from Nd $K\beta$ absorption with Xe $K\beta$ emission is not resolved from the peak at 7.6 kev and hence only

[1] J. B. Chubbock and I. Perlman, Phys. Rev. **74**, 982 (1948).
[2] A. R. Brosi and B. H. Ketelle, Phys. Rev. **100**, 169 (1955).
[3] B. H. Ketelle and G. E. Boyd, J. Am. Chem. Soc. **69**, 2800 (1947).
[4] S. C. Curran, *Beta and Gamma Ray Spectroscopy*, edited by K. Siegbahn (Interscience Publishers, Inc., New York, 1955), p. 176.

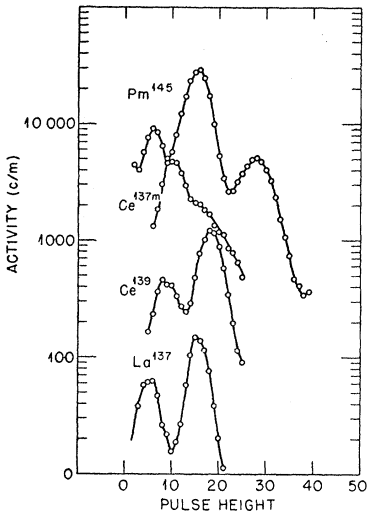

FIG. 1. Xenon-filled proportional counter spectrometer K x-ray "escape" spectra. An aluminum absorber was used to remove L x-rays.

three peaks are seen. Absorption of Nd $K\alpha$ x-rays followed by Xe $K\beta$ escape gives a peak at 3.5 kev and Nd $K\beta$ absorption with Xe $K\alpha$ escape gives a peak at 12.8 kev.

In the case of Ce^{137m} the prominent peak results from critical absorption of Ce $K\alpha_1$ x-rays in the counter and emission of Xe $K\alpha$ x-rays. This spectrum is complicated, because the 34-hour isomeric transition is in equilibrium with the 8.7-hour ground state which decays by electron capture and therefore emits La x-rays. Although about 60% of the x-ray quanta emitted are from La, the escape peaks from La x-rays do not stand out in this spectrum because only the relatively low intensity $K\beta$ La x-rays are critically absorbed in xenon. The shoulder on this curve results from the fact that absorption of the $K\beta$ x-rays of Ce and La followed by Xe $K\alpha$ escape gives peaks at 9.8 kev and 8.3 kev, respectively, which are not resolved.

In the electron capture decay of Ce^{139}, La x-rays are emitted. Since the La $K\alpha$ x-rays do not have sufficient energy to be critically absorbed in xenon, the escape spectrum has two peaks at 8.0 kev and at 4.2 kev, which result from absorption of La $K\beta$ x-rays followed by the escape of Xe $K\alpha$ and of Xe $K\beta$ x-rays, respectively.

Barium $K\beta$ x-rays have about 1.5 kev less energy than lanthanum $K\beta$ x-rays. It can be predicted that they will be critically absorbed in xenon and give an escape spectrum the same as that from lanthanum x-rays except that the peaks will be shifted 1.5 kev to lower energies. Since the spectrum observed from the x-rays emitted by the long-lived lanthanum fraction is the same as the spectrum predicted for Ba K x-rays it is concluded that the lanthanum decays by K electron capture. An attempt was made to obtain the escape spectrum of Ba $K\beta$ x-rays from Cs^{137}. This proved to be impossible with the equipment used, because the 660-kev gamma ray gave pulses which completely obscured the low-energy x-ray escape spectrum.

Although it has not been established that the long-lived lanthanum isotope which decays by electron capture is La^{137}, it seems very improbable that it is any other lanthanum isotope. The only other La isotope that could be produced by an (n,γ) reaction on Ce is La^{139} which is known to be stable. The flux of neutrons energetic enough to produce (n,p) or $(n,2n)$ reactions on cerium isotopes was several orders of magnitude lower than the slow-neutron flux. In addition, the presence of any of the lanthanum isotopes, other than La^{137}, that might be produced by these reactions could be excluded because of the absence of their known radiations in the gamma-ray spectrum of the sample. It has been concluded, therefore, that the long-lived lanthanum produced in the cerium bombardment is almost certainly La^{137}.

The intensity of Ba K x-rays emitted by the lanthanum source and the number of mass-137 atoms produced in the neutron bombardment have been used to estimate the K electron capture half-life of La^{137}. The x-ray intensity was determined by comparison of the scintillation counter pulse-height spectrum with the Ba K x-ray spectrum from a Cs^{137} source of known disintegration rate. The K electron capture decay rate, corrected for chemical yield, is given in Table I along with other data needed to calculate the La^{137} half-life.

Uncertainty in the Ce^{136} neutron cross section contributed a much greater error to the half-life than any of the other quantities used in the computation. Because of this, the yield of 8.7-hr Ce^{137} from bombardment of Ce^{136} in the ORNL graphite reactor was redetermined. The neutron flux was monitored with Co^{59}, and Ce^{137} was counted with a scintillation counter which had been calibrated for La x-rays by using a Ce^{139} source of known disintegration rate. A 6.3-barn cross section was found for neutron activation of the 8.7-hr ground state. The cross section for activation of the 34.5-hr isomer was found to be one-tenth that for activation of the ground state giving a total cross section for production of Ce^{137} of 7 barns. These cross sections are the thermal-neutron activation cross sections provided $1/v$ absorption occurs. However, since pile bombardment conditions for La^{137} production were essentially the same as for the Ce^{137} yield measurement, resonance absorption of neutrons should not lead to an appreciable error in the half-life. The data in Table I give a La^{137} half-life of 6×10^4 yr. This is somewhat shorter than the mini-

TABLE I. Computation of La^{137} half-life.

Weight of CeO_2 bombarded	880 mg
Atoms of Ce^{136} bombarded	5.9×10^{18}
Cross section of Ce^{136}	$(7\pm1)\times10^{-24}$ cm^2
Integrated neutron flux	$(1.3\pm0.3)\times10^{23}$ neutrons/cm^2
Atoms of mass 137 produced	$(5.4\pm1.5)\times10^{15}$
Disintegrations/sec of La^{137}	$(2.0\pm0.2)\times10^3$ K captures/sec
Half-life of La^{137}	$(6\pm2)\times10^4$ yr

mum half-life estimated earlier[2] because the new cross section used in the computation is smaller by a factor of three than the cross section used in the estimate.

The K electron capture half-life obtained from the data in Table I could be much longer than the total electron capture half-life if the ratio of K electron capture to electron capture from other shells were small. The relative intensity of the L and K x-rays emitted by La^{137} was compared with the relative intensity of the L and K x-rays emitted by a Ce^{139} source. The L to K x-ray intensity ratio of La^{137} was about one-half the L to K ratio[5] of Ce^{139}. This is the intensity ratio expected if La^{137} has the theoretical[6] L/K electron capture ratio and the disintegration energy is large compared to the K electron binding energy.

III. Ce^{137} DECAY SCHEME

In earlier work on Ce^{137}, the La^{137} ground state was given a $d_{5/2}$ assignment in a decay scheme which was consistent with all experimental observations. Since the ground state of Ba^{137} is known to be a $d_{3/2}$ level, electron capture decay of La^{137} should be allowed if the $d_{5/2}$ ground state spin assignment is correct. In the work on La^{137} reported above, the electron capture half-life was found to be long even though the disintegration energy appeared to be large compared with the K electron binding energy. Because of this discrepancy, the Ce^{137} decay scheme has been studied further. Since the energy differences between the $d_{5/2}$ and the $g_{7/2}$ levels of odd-Z nuclides in this region are known to be small, this work has taken the form of a search for a low-energy transition which would be indicative of a low-lying energy level.

A source of high specific activity, made by bombardment of CeO_2 enriched in Ce^{136} in the Low-Intensity Training Reactor at ORNL, was measured in a thin magnetic-lens beta-ray spectrometer with a 10-$\mu g/cm^2$ window on the counter. The electron momentum spectrum obtained is shown in Fig. 2. In addition to the

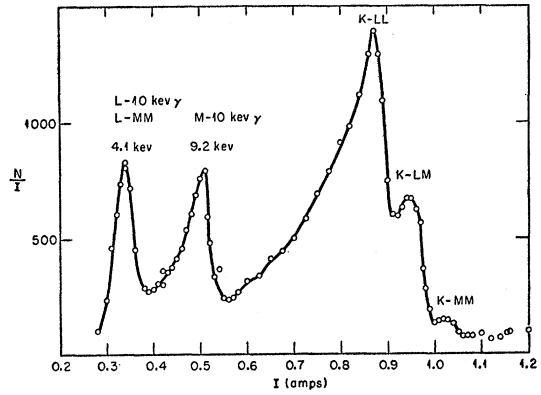

FIG. 2. Conversion and Auger electron spectrum of Ce^{137}.

[5] Ketelle, Thomas, and Brosi, Phys. Rev. **103**, 190 (1956).
[6] M. E. Rose and J. L. Jackson, Phys. Rev. **76**, 1540 (1949).

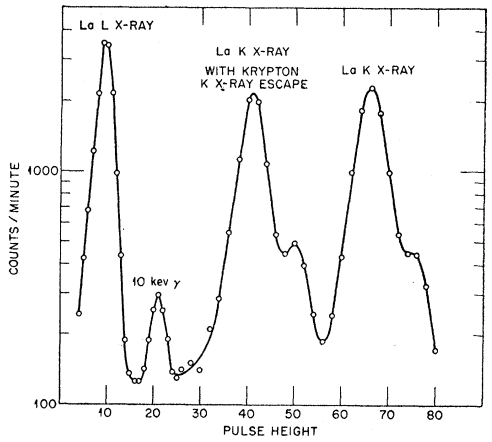

FIG. 3. Krypton-filled proportional counter spectrum showing the x-rays and the 10-kev gamma ray emitted in Ce^{137} decay.

K and L Auger electrons, this spectrum shows the presence of 9.2-kev electrons which are interpreted as the M conversion electrons of a 10-kev gamma ray. The intensity of the 4.2-kev peak relative to the K Auger peak is greater than would be expected from the L/K x-ray intensity ratios when compared with the Ce^{139} spectrum. Therefore, part of the intensity at 4.2 kev from the Ce^{137} source is attributed to L conversion electrons of a 10-kev gamma ray. Both of the conversion electron peaks decay with the same half-life as Ce^{137} within an experimental error of about 10%.

The low-energy photon spectrum of Ce^{137} was measured with a krypton-filled proportional counter and is shown in Fig. 3. This spectrum shows the presence of a 10-kev gamma ray in addition to the K and L x-rays of lanthanum. By measuring coincidences between a scintillation counter and a proportional counter, both the K x-rays and the 445-kev gamma ray of Ce^{137} were shown to be in "prompt" coincidence with the 10-kev gamma ray. Since the 10-kev gamma ray is emitted in coincidence with the other radiations of Ce^{137} and since it decays with the Ce^{137} half-life, it is concluded that this gamma ray is emitted in the decay of Ce^{137}. It can also be concluded that the lifetime of the initial state of the 10-kev transition is shorter than 10^{-7} sec because no delayed coincidences were observed in measurements with a 4×10^{-7} sec coincidence resolving time.

Because of large and uncertain electron absorption and scattering losses in the source and counter window, conversion electron intensities from the data in Fig. 2 cannot be used with confidence to estimate a conversion coefficient. However, if it is assumed that all Ce^{137} disintegrations occur through the 10-kev level, then a total conversion coefficient, $(N_L+N_M)/N_\gamma$, can be computed from the relative intensities of the unconverted 10-kev gamma ray and the K x-rays from electron capture. Support for this assumption was obtained by comparing the L/K x-ray intensity ratio of Ce^{137} with that of Ce^{139}. The relative L x-ray intensity of Ce^{137} was

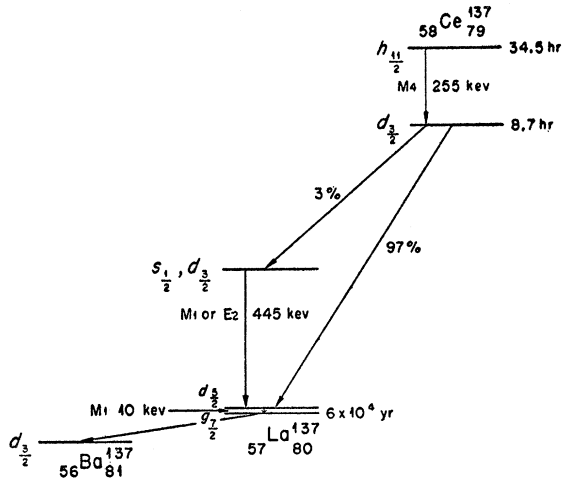

FIG. 4. Ce^{137} and La^{137} decay schemes.

about three times that of Ce^{139}, in approximate agreement with the relative intensity calculated for the case where every capture in Ce^{137} is followed by L conversion.

The intensity of the K x-rays emitted by a Ce^{137} source was measured on a counter calibrated with a Ce^{139} source whose x-ray emission rate was determined by coincidence counting. The 10-kev gamma intensity from the same Ce^{137} source was measured with a krypton-filled proportional counter at a known geometry. The counting efficiency was calculated to be 50% from the krypton absorption coefficient for 10-kev gamma radiation. These data gave a conversion coefficient N_e/N_γ of 140 with an uncertainty of a factor of two.

IV. DISCUSSION

The decay scheme proposed for Ce^{137} and La^{137} is shown in Fig. 4. The Ce^{137} scheme differs from the one proposed earlier[2] by inclusion of the 10-kev transition discovered in this work. The 10-kev transition is placed after the 445-kev transition because coincidences were found between the 10-kev gamma ray and the 445-kev gamma ray. In addition, the intensities of the conversion electrons and the L x-rays indicate that there are almost as many 10-kev transitions as captures, whereas only a few percent of the disintegrations are through the 445-kev transition. This indicates independent capture to the 10-kev level, placing it below the 445-kev transition. The relative intensities of the radiations emitted in Ce^{137} decay also indicate that the fraction of captures directly to the ground state of La^{137} is small.

The level assignments are based on the single-particle model and the systematics of energy level spacings. The ground-state level assignments of odd-proton even-neutron nuclei just after the 50-proton shell is filled are in general $d_{5/2}$ for low-mass-number and $g_{7/2}$ for high-mass-number nuclides of a given atomic number. In the case of atomic number 53, the shift from $d_{5/2}$ to $g_{7/2}$ occurs between mass numbers 127 and 129. At a Z of 55, the shift is between mass numbers 131 and 133. This trend suggests that at atomic number 57 and mass number 137 the two levels will be close together and that the $d_{5/2}$ level may be above the $g_{7/2}$ level.

Although the experimental data do not permit definite spin assignments, they are consistent with the assignments given in Fig. 4. The maximum limit placed on the lifetime of the 10-kev level indicates that the multipole order of the transition is low. This is consistent with the assignments which show an $M1$ transition. Although a large extrapolation is required to obtain the theoretical conversion coefficients[7] for a 10-kev gamma ray, the differences between multipole orders are so large that the experimental value indicates rather definitely an $M1$ transition. A $d_{5/2}$ assignment of the La^{137} ground state would require electron-capture decay to the $d_{3/2}$ ground level of Ba^{137} to be allowed. This is inconsistent with the experimental work reported here, which shows that the K electron capture half-life is 6×10^4 yr even though the disintegration energy appears to be considerably larger than the K electron binding energy. On the other hand, if the La^{137} ground state assignment is $g_{7/2}$, electron-capture decay is second-forbidden which is consistent with the long half-life observed experimentally. This assignment also makes electron capture to the La^{137} ground state in Ce^{137} decay second-forbidden. This is consistent with the conclusion, drawn on the basis of intensity measurements, that only a small fraction of the electron captures occur directly to the La^{137} ground state.

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[7] M. E. Rose (privately distributed L conversion coefficients).