that a quantitative development of some of the ideas associated with the collective model will considerably increase our understanding of the nuclear fission process.

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Decay Scheme of La^{138}

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The methods of scintillation spectroscopy have been applied to the naturally occurring radionuclide La^{138}. It is shown that La^{138} emits gamma radiation of energies 1.43±0.01 and 0.81±0.01 Mev. Coincidence studies show that the 1.43-Mev radiation is correlated in time with an observed Ba K x-ray (32±1 kev), but not with the 0.81-Mev gamma ray. A decay scheme is proposed in which La^{138} undergoes electron capture to the first excited level of Ba^{138} at 1.43±0.01 Mev with a partial half-life of $(2.1\pm0.1)\times10^{11}$ years and an L/K ratio of 1.4±0.25, and negatron decay to an excited level of Ce^{138} at 0.81 Mev with a partial half-life of $(2.4\pm0.2)\times10^{11}$ years. Consideration of beta decay systematics suggest that in each case the transition energy is low. The total half-life is estimated to be $(1.0\pm0.1)\times10^{11}$ years.

INTRODUCTION

THE natural radioactivity of lanthanum has been reported[1] and confirmed.[2–4] Nuclear systematics indicate that the radioactive nuclide is the rare (isotopic abundance 0.089%)[5] isotope La^{138}, since this isotope is the central member of the naturally occurring isobaric triplet Ba^{138}—La^{138}—Ce^{138}, and is odd in both N and Z. In an earlier investigation,[2] gamma radiation was detected, of energies 1.39±0.03, 0.807±0.015, and 0.535±0.015 Mev and relative intensities 1:0.65:0.3, respectively, with a total specific gamma-ray activity of 0.6 gamma quanta per second per gram of lanthanum. In addition, the observation of barium K x-radiation (32±1 kev) of specific activity 0.4 x-rays per second per gram of lanthanum led to the proposal of a decay scheme in which La^{138} undergoes K capture to levels in Ba^{138} at 0.807 Mev and 1.39 Mev. In contrast, Bell and Cassidy[3] have since reported that the gamma-ray spectrum of La^{138} comprises two components of energies 1.06 Mev and 0.545 Mev. Mulholland and Kohman[4] have detected a low-intensity negatron component (0.07 disintegration per second per gram of lanthanum) with an end-point energy of 1.0±0.2 Mev. Their study of the x-ray spectrum yielded a specific activity of 1.0 K x-rays per second per gram. Selig[6] has studied the x-ray spectrum and reports activities of 0.23±0.07 K x-rays and fewer than 0.013 L x-rays per second per gram of lanthanum with an L/K capture ratio of less than 0.5. Recent studies[7–9] of the beta and gamma-ray spectra of Cs^{138} have shown that the first excited state of Ba^{138} is at 1.426 Mev but otherwise contradict the proposed decay scheme for La^{138}. We have re-examined the gamma-ray spectrum of lanthanum with a high-resolution scintillation spectrometer in a massive lead and mercury shield and have studied the time correlation of the observed components.

SPECTRUM ANALYSIS

For the analysis of the electromagnetic spectrum of La^{138}, a well-shielded crystal of NaI(Tl) ($1\frac{1}{4}$ in. in diameter and $1\frac{1}{2}$ in. high) mounted on a DuMont K1186 photomultiplier was used. A detailed description of the massive lead and mercury shield has been published elsewhere.[10] After amplification in a high-gain, non-overloading amplifier, the pulse-height spectrum was analyzed by means of a Harwell type-1074A five-channel descriminator. The resolution of the spectrometer for gamma radiation of 0.662 Mev was

[1] Pringle, Standil, and Roulston, Phys. Rev. **78**, 303 (1950).
[2] Pringle, Standil, Taylor, and Fryer, Phys. Rev. **84**, 1006 (1951).
[3] P. R. Bell and J. M. Cassidy, Oak Ridge National Laboratory Report ORNL-782, 1953 (unpublished).
[4] G. J. Mulholland and T. P. Kohman, Phys. Rev. **87**, 681 (1952).
[5] Ingraham, Hayden, and Hess, Phys. Rev. **72**, 349, 967 (1947).
[6] H. Selig, Doctoral dissertation, Carnegie Inst. Tech. N. Y. O.-6626 (1955).
[7] Langer, Duffield, and Stanley, Phys. Rev. **89**, 907(A) (1953).
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[9] Bunker, Duffield, Mize, and Starner, Phys. Rev. (to be published).
[10] Pringle, Turchinetz, and Funt, Rev. Sci. Instr. **26**, 859 (1955).

better than 8%. The source material, 41.50 grams of lanthanum oxide powder (35.4 grams lanthanum), after treatment in an ion exchange column[11] to remove traces of rare earth metals, was firmly packed into a Lucite source holder so that the crystal was surrounded by a thickness of 0.77 cm of source. For background purposes, a dummy source of 60 grams of highly purified[11] cerous oxalate was used in the same geometry. To calibrate the energy scale, the well-known gamma rays from the following radionuclides were used: Cs^{137}(0.032, 0.662 Mev), RaD(0.0465 Mev), Zn^{65}(0.511, 1.12 Mev), Co^{60}(1.17, 1.33 Mev).

The results of the pulse-height analysis are shown in Figs. 1 and 2. The gamma-ray spectrum (Fig. 1) is seen to be composed of two components with energies 1.43±0.01 and 0.81±0.01 Mev, in agreement with the two earlier[2] gamma rays at 1.39±0.03 and 0.807 ±0.015 Mev. However, a careful study of the spectrum in the region of 0.535 Mev yielded no evidence of gamma radiation of this energy and it is now evident that in the earlier work, with a poorer spectrometer and less adequate statistics, the Compton peak for the 0.807-Mev gamma ray was interpreted as the photoline for another gamma ray. The x-ray spectrum (Fig. 2) shows a component of energy 32±1 kev. For ease of comparison the distribution for the Ba K x-radiation following the decay of Cs^{137} is indicated on the same figure, as is the location of the Ce K x-ray photoline at 34.7 kev.

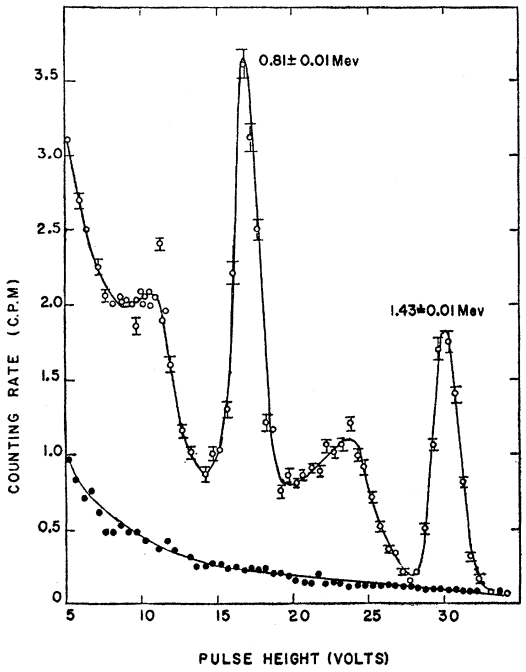

FIG. 1. Gamma-ray spectra from La and Ce (background).

[11] We are indebted to Drs. D. S. Russel and Fetch of the Chemistry Division, National Research Council, Ottawa, for their cooperation in this work.

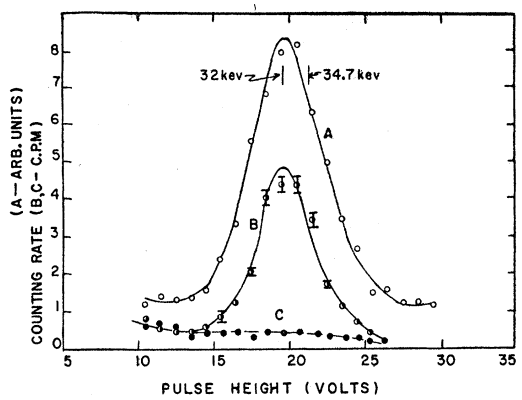

FIG. 2. X-ray spectra: *A*, Ba K x-ray following decay of Cs^{137}, *B*, x-ray following La decay; *C*, background.

The specific activity of the 1.43-Mev gamma-ray component was calculated from the area under the photoline by comparison with the photoline counting rate from a known mass of K^{40} in the same geometry, and was found to be 0.41±0.03 gamma quanta per second per gram of lanthanum, taking the specific gamma-ray activity of K^{40} to be 3.4±0.1 gamma quanta per second per gram of potassium.[12] To arrive at an estimate for this quantity for the 0.81-Mev radiation the 1.465-Mev photoline from K^{40} was normalized to the 1.43-Mev photoline from La^{138} and the level of the normalized K^{40} Compton distribution was taken as indicative of the level of the Compton distribution from the 1.43-Mev lanthanum gamma ray, after which the area under the 0.81-Mev photoline was measured. Corrections for the variation of the crystal detection efficiency and the photo fraction with gamma-ray energy were made using available data.[13]

The result of the computation is that there are emitted 0.36±0.03 gamma ray of energy 0.81 Mev per second per gram of lanthanum.

An estimate of the specific K x-ray activity from the data obtained with the aforementioned source geometry is difficult to make because of the appreciable absorption corrections that must be applied for radiation of this low energy. To simplify matters in this regard, additional counting data were obtained using a cylindrical NaI(Tl) crystal ($3\frac{1}{2}$ in. in diameter and 2 in. thick) mounted on a DuMont 6364 photomultiplier, with 10 grams of lanthanum oxide spread over a circular area of 46.53 cm^2 concentric with the axis of symmetry of the NaI(Tl) crystal. Under these circumstances, corrections for fractional geometry and absorption were easily made. There was also a small (~5%) escape peak correction for the barium K_β

[12] P. M. Endt and J. C. Kluyver, Revs. Modern Phys. **26**, 95 (1954).

[13] P. R. Bell, *Beta and Gamma Ray Spectroscopy*, edited by K. Siegbahn (Interscience Publishers Inc., New York, 1955), pp. 133–164. Maeder, Muller, and Winterstebger, Helv. Phys. Acta **27**, 1 (1954).

radiation, calculated using Williams'[14] data on the relative intensities of the components of the K-series x-radiation from barium and Axel's[15] data on the escape-peak correction for NaI(Tl) as a function of x-ray energy. The result of these corrections applied to the counting data obtained with a single channel scanning the photoline at 32 kev, is that the specific K x-ray activity of lanthanum is 0.145 ± 0.01 K x-rays per second per gram. By using the expression for fluorescence yield as a function of atomic number given by Steffen *et al.*[16] it was found that the specific K-capture activity is 0.17 ± 0.015 disintegrations per second per gram.

COINCIDENCE COUNTING

For the coincidence counting, the lanthanum oxide sample was packed into a disk $3\frac{1}{2}$ inches in diameter and $\frac{1}{4}$ inch thick which was sandwiched between two NaI(Tl) cylinders ($3\frac{1}{2}$ in. in diameter and 2 in. thick) mounted on DuMont 6364 photomultipliers. A similar disk of cerous oxalate was used for the background determinations. Pulse-height analysis for one detector was accomplished by means of a Dynatron Rádio type N-101 single-channel analyzer. The output of this unit was used to "gate" the five-channel analyzer which scanned the output of the other detector. The resolving time of the coincidence circuit was 60 microseconds.

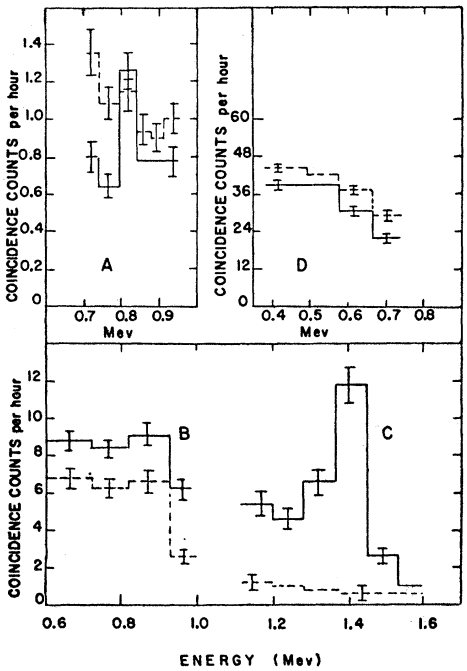

FIG. 3. Coincidence counting histograms: A(1.43−0.81 Mev) B(32 kev−0.81 Mev); C(32 kev−1.43 Mev); D(0.511 Mev−0.511 Mev). (Broken line background.)

Because of the very low counting rate and the comparative simplicity of the gamma-ray spectrum, wide gates were used throughout. The accidental coincidence counting rate due to the lanthanum activity was negligible.

The following conceivable cascades were investigated: 32 kev−1.43 Mev, 32 kev−0.81 Mev, 1.43 Mev−0.81 Mev, by fixing the single channel on the first-mentioned feature and scanning the energy region of the second feature with the gated five-channel analyzer. In addition, evidence for positron emission was sought in the form of coincidence counts between annihilation quanta. The results of the coincidence experiments are shown in Fig. 3. Parts B and C of Fig. 3 show the gamma-ray spectrum in coincidence with the 32-kev x-ray. It is clear from this figure that it is the 1.43-Mev gamma ray which is correlated in time with the 32-kev x-ray. The slight surplus coincidence counting rate in the region of 0.8 Mev is due to coincidence events involving the 32-kev x-ray and the Compton distribution from the 1.43-Mev radiation. Part A of Fig. 3 shows that there is no detectable time association between the 0.81- and the 1.43-Mev radiation. The search for evidence of positron activity, the results of which are indicated in part D of Fig. 3, yielded no measurable coincidence counting rate over the background.

DISCUSSION

The experimental results indicate that the 1.43-Mev gamma radiation is associated with K capture to the first excited level of Ba^{138} at 1.426 Mev.[9] From the measured K-capture specific activity (0.17 ± 0.015 disintegration per minute per gram) and the total electron capture activity, as indicated by the intensity of the 1.43-Mev gamma radiation (0.41 ± 0.03 disintegration per second per gram), the L/K capture ratio is calculated to be 1.4 ± 0.25. The 0.81-Mev radiation must be associated with negatron emission to an excited level in Ce^{138}. Handley and Olson[17] have reported the presence of gamma radiation of this energy following the positron decay of Pr^{138}. The observed intensity of the 0.81-Mev radiation (0.35 ± 0.03 quanta per second per gram) is too high to be associated with the 1.0-Mev negatron component (0.07 disintegration per second per gram) reported by Mulholland and Kohman.[4]

The spin of La^{138} has been recently measured[18] and is known to be 5, in agreement with the shell model which predicts that the neutron-proton configuration should be $d_{3/2}-g_{7/2}$. On this basis, the ground state of La^{138} has even parity. Scharff-Goldhaber's[19] survey of the spins and parities of the first excited states of even-even nuclei suggests that the first excited state of $Ce^{138}(N=80, Z=58)$ has $J=2(+)$. From these considerations it follows that the negatron transition to

[14] J. H. Williams, Phys. Rev. 44, 146 (1933).
[15] P. Axel, Rev. Sci. Instr. 25, 391 (1954).
[16] Steffen, Huber, and Humbel, Helv. Phys. Acta 22, 167 (1949).
[17] T. H. Handley and E. L. Olson, Phys. Rev. 96, 1003 (1954).
[18] P. B. Sogo and C. D. Jeffries, Phys. Rev. 99, 613 (1955).
[19] G. Scharff-Goldhaber, Phys. Rev. 90, 587 (1953).

the 0.81-Mev level in Ce^{138} is second-forbidden unique with $\Delta J=3$(no). If one takes a range of $\log ft=12.5-13.5$ as characteristic of transitions of this type and uses the measured half-life for the 0.81-Mev gamma ray of $(2.4\pm0.2)\times10^{11}$ years, one gets for the transition energy to the 0.81-Mev level in Ce^{138} a range of values of 5–10 kev, which is in fair agreement with the estimate of 0.2 ± 0.2 Mev based on Mulholland and Kohman's measurement of the energy of the weak β component at 1.0 ± 0.2 Mev. For a transition energy of 0.2 Mev and the present half-life, a value of $\log ft=18.1$ is obtained which is more characteristic of third-forbidden transitions. The spin of the first excited level of Ba^{138} is known[9] to be 2 and the parity is probably even since this is an even-even nucleus[20,21]; thus the selection rules appear to be $\Delta J=3$(no) for the electron capture transition to this level. Using the measured value of L/K, 1.4 ± 0.25, and the theoretical results of Brysk and Rose[22] on the L/K-capture ratio as a function of transition energy and atomic number, we find that the transition energy is 160 ± 30 kev. This is in fair agreement with the $La^{138}-Ba^{138}$ mass difference of 1.3 ± 0.4 Mev estimated from the following closed cycle of reactions: $La^{139}(\gamma,n)La^{138}$ $(Q=-8.8\pm0.2$ Mev)[23]; $Ba^{138}(d,p)Ba^{139}$ $(Q=3.0\pm0.3$ Mev)[24]; $Ba^{139}-\beta\rightarrow La^{138}$ $(E_{\beta-}=2.27\pm0.02$ Mev).[25] In conjunction with the measured half-life of $(2.1\pm0.1)\times10^{11}$ years for the electron capture activity, this value for the transition energy yields[26] $\log ft=16.9$ which appears to be somewhat large* for a second-forbidden unique transition

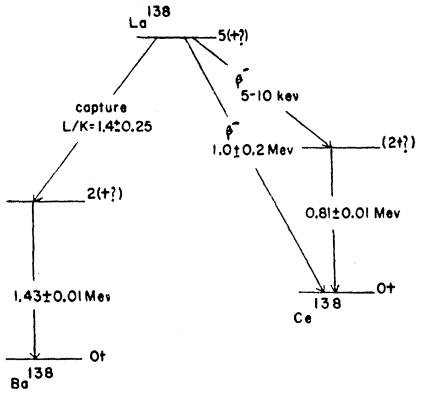

FIG. 4. Proposed decay scheme for La^{138}. The 1.0 ± 0.2 Mev negatron component is that of Mulholland and Kohman[4] while the 5–10 kev component is inferred from the experimental data and beta-decay systematics.

($\Delta J=3$(no). We are thus led to propose the decay scheme of Fig. 4 for La^{138}. The total half-life of La^{138} is calculated to be $(1.0\pm0.1)\times10^{11}$ years, if one uses the value of 1.2×10^{12} years for the half-life of the negatron transition reported by Mulholland and Kohman,[4] and the two partial half-lives mentioned above.

ACKNOWLEDGMENTS

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[20] M. J. Glaubman, Phys. Rev. **90**, 1000 (1953).
[21] I. Talmi, Phys. Rev. **90**, 1001 (1953).
[22] H. Brysk and M. E. Rose, Oak Ridge National Laboratory Report, ORNL-1830, January, 1955 (unpublished).
[23] Sher, Halpern, and Mann, Phys. Rev. **84**, 387 (1951).
[24] J. H. Harvey, Phys. Rev. **81**, 353 (1951).
[25] L. R. Shepherd and J. M. Hill, Nature **162**, 566 (1948).
[26] J. K. Major and L. C. Biedenharn, Revs. Modern Phys. **26**, 321 (1954).

* *Note added in proof.*—Glover and Watt have re-examined the β spectrum of La^{138} [R. N. Glover and D. E. Watt, Phil. Mag. (to be published)] and, finding a single component with an end point of 205 kev and $\log ft=19.0$, proceeding presumably to the 0.81-Mev level in Ce^{138}, suggest that the neutron configuration of La^{138} is $h_{11/2}$ and that the ground state of La^{138} has $J=5$ with odd parity. This would mean that the La^{138} activity is third forbidden [$\Delta J=3$ (yes)] and that $\log ft=16.9$ for the electron capture transition is of the right order of magnitude, even though it is calculated using a transition energy of 160 kev derived from the experimental L/K ratio of 1.4 and Brysk and Rose's results for second forbidden unique transitions. We are indebted to Mr. Glover and Mr. Watt of the Department of Natural Philosophy of Glasgow University for permission to quote these results in advance of publication.