(b) The total orbital angular momentum L may not be a good quantum number.††

Hypothesis (a) seems unlikely in view of the recent determinations[20,24] of the equivalence in energy of the excited state independent of the reaction producing it.

An experimental measurement of the angular momentum and magnetic moment of Be^7 would be of great help in choosing between various hypotheses, as also would a measurement of the electric quadrupole moment of Li^7.‡

In Table II is also included a calculation of τ_0, the characteristic time constant of beta-decay, which employs the very accurate data on $I(W)$ and $T_{\frac{1}{2}}$ which is now available.

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†† *Note added in proof.*—This hypothesis is discussed by S. S. Hanna and D. R. Inglis in a recent letter to Phys. Rev. **75**, 1767 (1949).

[24] Rasmussen, Lauritsen, and Lauritsen, Phys. Rev. **75**, 199 (1949); Zaffarano, Kern, and Mitchell, Phys. Rev. **74**, 105 (1948); F. N. D. Kurie and M. Ter-Pogossian, Phys. Rev. **74**, 677 (1948); Buechner, Strait, Stergiopoulos, and Sperduto, Phys. Rev. **74**, 1569 (1948); Fowler, Lauritsen, and Rubin, Phys. Rev. **75**, 1463, 1471 (1949).

‡ *Note added in proof.*—The sign of the electric quadrupole moment of Li^7 has been recently found (Kusch, Phys. Rev. **76**, 138 (1949)) to be positive instead of negative as expected on past nuclear models. According to recent unpublished calculations of R. Avery and C. Blanchard at the University of Wisconsin, both the magnetic moment and electric quadrupole moment of Li^7 can be accounted for correctly in sign and approximate magnitude by assuming that the ground state of Li^7 is predominately $D_{3/2}$.

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Neutron-Capture Cross Section of 85-Minute Ba^{139}

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Four samples of barium carbonate were irradiated with neutrons at three different times and at three different positions in the heavy water pile. A few days after an irradiation, lanthanum separations were made until no further La^{140} activity could be detected. The lanthanum was discarded. At intervals of 4 to 9 days the La^{140} grown from the Ba^{140} was separated and its number of atoms determined with a calibrated Geiger-Müller counter. The fluxes of thermal neutrons were obtained by experiment as far as possible; the maximum used was 2.0×10^{13} neutrons/cm²/sec. The final value for the capture cross section of Ba^{139} is 4.7 ± 1.2 barns.

I. INTRODUCTION

CROSS SECTIONS for the capture of thermal neutrons by stable elements have been measured in numerous investigations. A nucleus of mass A transforms to one of mass $A+1$ when a neutron is captured. This nucleus is radioactive in some cases and stable in others. Some methods of measuring capture cross sections that require little or no knowledge of scattering cross sections are (1) the activation method, (2) the pile reactivity method, and (3) the mass spectrographic method. The activation method is applicable when the nuclei formed by neutron-capture are radioactive. The number of thermal neutrons captured per second is equal to and in practice obtained from the number of β-disintegrations per second at saturation. This method of measuring capture cross sections has been applied, for instance, by Seren, Friedlander, and Turkel,[1] who discussed it in detail and presented a long list of measurements. When the nucleus of mass $A+1$ is stable, the number of thermal neutrons captured per second has to be determined in some other way such as by the change in the reactivity of a chain-reacting pile.[2] If the capture cross section is high, the mass spectrograph shows a measurable change in the relative abundances of the isotopes before and after a prolonged irradiation.[3] Numerical values of cross sections could be obtained in favorable cases.

The combination of high flux of thermal neutrons in a pile and the activation method permits capture cross sections to be measured even for β-active nuclei. A stable isotope of atomic number Z and mass A is irradiated with neutrons in a pile, producing β-active isotopes of masses $A+1$, $A+2$, $\cdots$ in succession. The amounts of these successive isotopes produced in a given time are in rapidly decreasing orders of magnitude. It is clear that if the neutron flux is sufficiently high, the amount of isotope of mass $A+2$ produced by neutron bombardment can, in many cases, be either directly measured, or obtained from a measurement on

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[1] Seren, Friedlander, and Turkel, Phys. Rev. **72**, 888 (1947).

[2] Anderson, Fermi, Wattenberg, Weil, and Zinn, Phys. Rev. **72**, 16 (1947).

[3] Lapp, VanHorn, and Dempster, Phys. Rev. **71**, 745 (1947). A. J. Dempster, Phys. Rev. **71**, 829 (1947).

the separated daughter substance $(Z+1, A+2)$ if it is radioactive, by the very sensitive method of β-particle detection. This may be related to the capture of neutrons by the β-active isotope of mass $A+1$. If, in addition, the amount of the latter formed in the given time is similarly measured or calculated from the known weight of isotope of mass A and its capture cross section, and if the neutron flux is known, the capture cross section of isotope of mass $A+1$ can be deduced. The method may be complicated by the formation of $(Z+1, A+2)$ by neutron-capture in $(Z+1, A+1)$, and by other β-activities if the stable element that is introduced into the pile consists of several isotopes.

An extensive knowledge of the neutron-capture cross sections of both stable and radioactive nuclei may show further interesting regularities[4] and may assist in accounting for the abundances of elements and of their isotopes found in nature.[5] The present paper is one of a series giving measurements of neutron-capture cross sections of β-active nuclei. It is concerned with Ba^{139}, a β-emitter of half-period 85 minutes, which occurs with high yield in fission initiated by slow neutrons.

II. DETAILED NUCLEAR REACTIONS

It is preferable to produce the Ba^{139} by neutron-capture in stable Ba^{138} than to separate it from mixed fission products. The pertinent nuclear reactions that occur when natural barium is irradiated in a pile are as follows:

$_{56}Ba^{138}$ (71.66 percent of natural barium)

(n,γ)

$$Ba^{139}\xrightarrow[85\text{ min.}]{\beta^-}{}_{57}La^{139}\text{ (stable)}$$

$$Ba^{140}\xrightarrow[12.8\text{ days}]{(n,\gamma);\ \beta^-}La^{140}\xrightarrow[40\text{ hr.}]{\beta^-}{}_{58}Ce^{140}\text{ (stable)};\qquad \text{(I)}$$

Ba^{130} (0.101 percent of natural barium)

(n,γ)

$$Ba^{131}\xrightarrow[12.0\text{ days}]{K}{}_{55}Cs^{131}\xrightarrow[9.6\text{ days}]{K}{}_{54}Xe^{131}\text{ (stable)};\qquad \text{(II)}$$

Ba^{132} (0.097 percent of natural barium)

(n,γ)

$$Ba^{133*}\xrightarrow[38.8\text{ hr.}]{I.T.}Ba^{133}\xrightarrow[20\text{ yr.}]{K}Cs^{133}\text{ (stable)}.\qquad \text{(III)}$$

Let P, Q, R, U, and V represent, respectively, the numbers of atoms of Ba^{138}, Ba^{139}, Ba^{140}, La^{139}, and La^{140} present at any time t during neutron irradiation at a constant flux $\rho v(\approx 10^{13}$ neutrons/cm^2/sec.). The subscripts 0, 1, 2, 3, and 4 will be used to designate the respective neutron-capture cross sections σ and transformation constants λ of these nuclei. The differential equations that apply during the irradiation are as follows:

$$dQ/dt=\rho v\sigma_0 P-\lambda_1 Q-\rho v\sigma_1 Q \approx \rho v\sigma_0 P-\lambda_1 Q; \qquad (1)$$

$$dR/dt=\rho v\sigma_1 Q-\lambda_2 R-\rho v\sigma_2 R \approx \rho v\sigma_1 Q-\lambda_2 R; \qquad (2)$$

$$dU/dt=\lambda_1 Q-\rho v\sigma_3 U; \qquad (3)$$

$$dV/dt=\rho v\sigma_3 U+\lambda_2 R-\lambda_4 V-\rho v\sigma_4 V \approx \rho v\sigma_3 U+\lambda_2 R-\lambda_4 V. \qquad (4)$$

P is assumed to be constant. At $t=0$, $Q=R=U=V=0$. At the end of the irradiation of duration T, the desired formulas are as follows:

$$Q_T=\frac{\rho v\sigma_0 P}{\lambda_1}(1-e^{-\lambda_1 T}); \qquad (5)$$

$$R_T=\frac{(\rho v)^2\sigma_0\sigma_1 P}{\lambda_1\lambda_2}(1-e^{-\lambda_2 T})-\frac{(\rho v)^2\sigma_0\sigma_1 P}{\lambda_1(\lambda_1-\lambda_2)}(e^{-\lambda_2 T}-e^{-\lambda_1 T}); \qquad (6)$$

$$U_T=\frac{\sigma_0 P}{\sigma_3}(1-e^{-\rho v\sigma_3 T})-\frac{\rho v\sigma_0 P}{\lambda_1-\rho v\sigma_3}\times(e^{-\rho v\sigma_3 T}-e^{-\lambda_1 T})\approx\rho v\sigma_0 PT; \qquad (7)$$

$$V_T\approx\frac{(\rho v)^2\sigma_0\sigma_3 PT}{\lambda_4}+\frac{(\rho v)^2\sigma_0\sigma_1 P}{\lambda_4\lambda_4}(1-e^{-\lambda_4 T})-\frac{(\rho v)^2\sigma_0\sigma_1 P}{\lambda_1(\lambda_4-\lambda_2)}(e^{-\lambda_2 T}-e^{-\lambda_4 T}). \qquad (8)$$

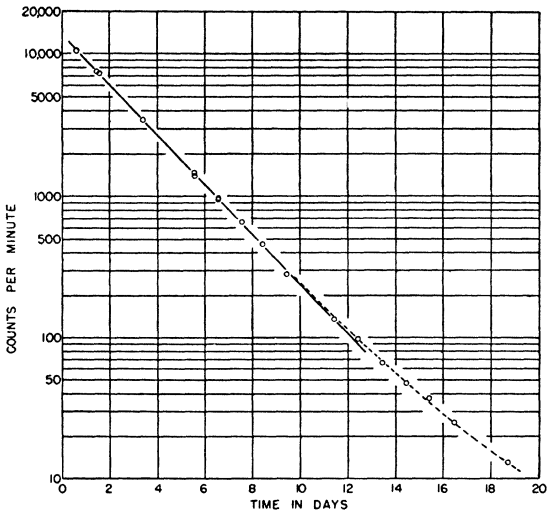

FIG. 1. Decay of separated lanthanum. Half-period is 41 hours when allowance of 10 counts per minute (i.e., 0.1 percent of initial activity) is made for barium contamination.

[4] M. G. Mayer, Phys. Rev. 74, 235 (1948).
[5] R. A. Alpher, Phys. Rev. 74, 1577 (1948); R. A. Alpher and R. C. Herman, Phys. Rev. 74, 1737 (1948).

In formula (8) the first term arises from the neutron-capture by La^{139} and the other terms from the β-decay of Ba^{140}.

An examination of formulas (5)–(8) suggests that the following four methods should be considered in determining σ_1, the neutron-capture cross section of Ba^{139}.

1. Determine the ratio of the β-counting rates of Ba^{140} and Ba^{139} at the end of the irradiation. (Formulas (5) and (6).) This method has the advantages of being independent of σ_0, the capture cross section of Ba^{138}, which is not accurately known, and of the solid angle subtended at the Geiger-Müller counter.
2. Determine the absolute β-counting rate of Ba^{140} at the end of the irradiation. (Formula (6).)
3. Determine the absolute β-counting rate of La^{140} separated at the end of the irradiation. (Formula (8).)
4. Determine the absolute β-counting rate of La^{140} that grows from the Ba^{140} after the irradiation has stopped.

Methods 1 and 2 cannot be used owing to the Ba^{131} that is formed in nuclear reaction II. The similarity in the half-periods of Ba^{140} and Ba^{131} and the complexity of their decay chains make it difficult, if not impossible, to determine their separate activities. Method 3 is ruled out by the fact that the La^{140} formed from La^{139} (first term, formula (8)) is much greater than the La^{140} formed from Ba^{140} (second and third terms, formula (8)). In fact, assuming $\sigma_1=\sigma_3$, the ratios of these contributions are 1220 after one day, 350 after 10 days, and 450 after 30 days irradiation. Consequently, method 4 was adopted. At the end of the irradiation, lanthanum was separated from barium and discarded. The La^{140} grown from Ba^{140} during successive intervals was separated and measured. While the La^{140} formed during the irradiation, mainly from La^{139} of known capture cross section, could have been used to determine the neutron flux, this was not done as a La^{139} impurity of even one part per million in the original barium sample would have caused a large error.

Suppose the La^{140} is separated at a time t_1 measured from the end of the irradiation and new La^{140} grows during the subsequent time t_2. The formula for the new La^{140} separated at this time is

$$V=\lambda_2/(\lambda_4-\lambda_2)R_T e^{-\lambda_2 t_1}(e^{-\lambda_2 t_2}-e^{-\lambda_4 t_2}), \qquad (9)$$

which is used in the calculation of σ_1.

III. EXPERIMENTAL PROCEDURE

Barium carbonate (analytical reagent grade) was irradiated in high flux positions in the heavy water pile. To free the barium from lanthanum after irradiation, the procedure was in each case to dissolve the sample in dilute hydrochloric acid, and take an aliquot from which the barium content was determined as barium sulfate. About 300 mg lanthanum nitrate was added to the main sample and a hydroxide precipitation was made. This step was repeated, lanthanum carrier being added each time, until no La^{140} activity could be detected in the precipitate. The lanthanum hydroxide samples were discarded.

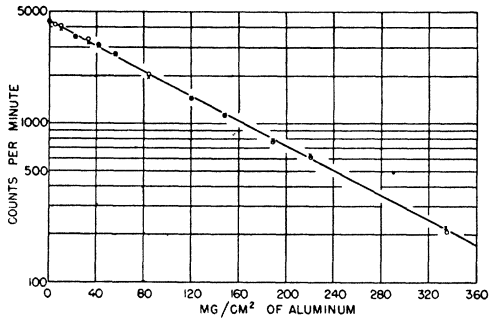

FIG. 2. Absorption of β-rays of La^{140} in aluminum. Crosses refer to lanthanum extracted from irradiated barium, circles to spectroscopically pure lanthanum after irradiation in the pile.

For collecting and separating the lanthanum that grows subsequently, the barium-containing solution was acidified and a weighed amount (about 300 mg) of lanthanum nitrate was added as a carrier. At a known time later (4 to 9 days), lanthanum hydroxide was precipitated from the solution. The precipitate was dissolved in dilute hydrochloric acid, barium chloride carrier added, and barium sulfate precipitated. Barium and caesium hold-back carriers were added to the centrifugate, and lanthanum hydroxide was again precipitated. The precipitate was washed with dilute

TABLE I.

Sample $BaCO_3$	Time in pile, days	Thermal neutron flux (neutrons/cm²/sec.) $\times 10^{-12}$	Elapsed time to first La separation, days
D	27.30	3.5	1.3
E	10.95	18	6.2
F	17.97	20	4.1
G	17.97	20	4.1

Note.—Samples *F* and *G* were irradiated together. The La^{140} separated at the time after irradiation given in column 4 was discarded.

TABLE II.

La^{140} sample measured	Gm $BaCO_3$	La^{140} growth time, days	La^{140} atoms formed $\times 10^{-8}$	$\sigma_1(Ba^{139})$, barns	
*D*1	5.38	5.6	2.23		5.12
*D*2	5.19	5.9	1.42		4.58
*D*3	5.19	6.1	1.11		4.93
*E*1	1.20	4.0	5.53		4.40
*F*1a	1.32	5.9	15.2	5.56	
*F*1b	1.32	5.9	14.5	5.30	5.15
*F*1c	1.32	5.9	12.6	4.60	
*F*2a	1.28	6.05	9.03	4.70	
*F*2b	1.32	6.05	6.92	3.49	3.87
*F*2c	1.29	6.05	6.59	3.39	
*F*2d	1.29	6.05	7.54	3.89	
*G*1	8.88	8.97	85.2		5.02
*G*2	7.72	3.96	41.2		4.37
*G*3	7.96	5.97	37.8		4.63
*G*4	8.08	8.00	24.8		4.31
*G*5	7.85	6.00	19.6		5.18
*G*6	7.59	7.04	13.2		5.09
				Average	4.72

Note.—The samples of La^{140} grown and measured are numbered in their natural order. Samples *F*1 and *F*2 were split into smaller samples *F*1a, *F*1b, etc., and the La^{140} separated and measured separately.

barium nitrate, dissolved in hydrochloric acid, barium carrier added, and the sulfate precipitated again. This cyclic process was repeated five times. The last barium sulfate precipitate was tested for activity, and none was found. The lanthanum hydroxide was precipitated, converted to the formate, dried, and the La^{140} activity measured.

To ensure that this precipitation of lanthanum had been complete, 300 mg lanthanum nitrate was added to the original barium-containing solution and a hydroxide precipitation was performed. No significant activity was ever found in this precipitate.

About 300 mg lanthanum nitrate carrier was again added to the solution, which had been acidified. The solution was allowed to stand until it was time for another La^{140} separation.

The La^{140} samples were mounted on aluminum trays, 7.1 cm^2 area, and the activity measured with a previously calibrated Geiger-Müller counter having a mica end-window of mass 2.8 mg per cm^2.

The decay curve of a lanthanum sample was followed for some time. This was corrected for the residual background due to barium contamination, which amounted to about one percent of initial La^{140} activity. The counting rate of the La^{140} at the time of precipitation was obtained by extrapolation. The number of disintegrations per minute was estimated from the counting rate by applying the following corrections:*** self-absorption of β-rays in the sample ($\sim$5 percent), external absorption by the air gap and counter window ($\sim$3 percent), back-scattering by the aluminum tray ($\sim$20 percent), counter efficiency, and percentage chemical recovery. The counter efficiency was found to be 7.5 percent using a standard Co^{60} source.

IV. RESULTS

The β-activity was identified as La^{140} by measurement of half-period and absorption of the β-rays in aluminum. Figure 1 shows a typical decay curve, which has a half-period of 41 hours when corrected for barium contamination. Figure 2 shows absorption measurements on the β-activities of the lanthanum separated from the irradiated barium and of La^{140} obtained by irradiating spectroscopically pure lanthanum in the pile.

The experimental results are shown in Tables I and II. Four samples of barium carbonate were irradiated at three different times and at three different positions in the pile.

The flux of thermal neutrons was determined by separate activations at the position for sample *D*. Five samples of lanthanum yielded a flux of 3.5×10^{12} neutrons/cm^2/sec. based on a capture cross section of 8.4 barns[1] for La^{139}. The variations from the mean were less than 3 percent. Two samples of barium carbonate yielded 1.67×10^{-12} and 1.75×10^{-12} neutron/sec./atom Ba^{138} for $\rho v\sigma_0$. Assuming $\sigma_0=0.511$ barn[1] for Ba^{138} the values of ρv are 3.3×10^{12} and 3.4×10^{12} neutrons/cm^2/sec., in good agreement with the measurements using lanthanum. It should be noted that σ_1 of Ba^{139} depends on the product of ρv and $\rho v\sigma_0$. The uncertainty in σ_1 arising from the error in $(\rho v)^2\sigma_0$ is somewhat reduced by the direct measurement of $\rho v\sigma_0$.

The fluxes of thermal neutrons for samples *E*, *F*, and *G* could not be conveniently measured by the activation method. The fluxes were therefore estimated from the measured heat output of the pile when operating, known pile constants, and the known spatial distribution of neutron density.

The irradiations of the samples were interrupted a number of times by the pile being shut down. The amounts of Ba^{139} and Ba^{140} built up have been accurately determined by a step by step application of Eqs. (1) and (2) over the times of operation (ρv tabulated) and shutdown ($\rho v=0$).

The final value of the capture cross section of Ba^{139} is 4.7 barns. The internal agreement of the various measurements is very satisfactory. Actually cadmium differences were not taken, but some rough experiments using cadmium to shield the samples indicate that the contribution of epicadmium neutrons to the activation is small. The measured cross section is therefore assumed to refer to neutrons of speed 2200 m/sec. for which the fluxes are quoted.

It is difficult to assess the error that should be associated with the measured cross section. Seren, Friedlander, and Turkel[1] assign a probable error of 20 percent to their measured cross sections of Ba^{138} and La^{139}, which were used here. We feel that 1.2 barns is sufficient for the probable error in our measured value of 4.7 barns.

Since this work was done we have learned that Katcoff[6] obtained 3.8 barns for the capture cross section of Ba^{139} in a previous measurement. He separated and measured La^{140} three times from a sample of irradiated barium carbonate. The agreement between the two independent measurements is satisfactory. Katcoff's result also depends on the accuracy of 0.511 barn assumed for the cross section of Ba^{138}.

We wish to thank Miss A. R. Rutledge for assistance in computing.

*** These were carefully determined in subsidiary experiments. The values in brackets are given only to indicate their magnitudes.

[6] S. Katcoff, Report CC-2908 of the Manhattan Project (1945).