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PHYSICAL REVIEW C

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Microscopic ¹³⁰Ba (n, γ) Cross Section and the Origin of ¹³¹Xe on the Moon^{*†}

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The ¹³⁰Ba (n, γ) cross section has been measured from 0.5 to 5000 eV with an energy resolution of 0.4%. The large epithermal resonances found in this experiment explain the anomalously high concentrations of ¹³¹Xe found in lunar rocks, and the measured resonance parameters provide one with a quantitative tool for correlating burial depth with exposure age for lunar samples.

I. INTRODUCTION

Studies of lunar rocks¹⁻⁶ have revealed anomalously high concentrations of ¹³¹Xe, which apparently are associated with the cosmic-ray flux at the lunar surface, the depth (or shielding) history of the lunar samples involved, and the abundance of barium in these samples. Kaiser and Berman⁷ have proposed that the explanation of this anomaly is the existence of a large, nonthermal-neutron-capture cross section for ¹³⁰Ba, most likely in the resonance-energy region. They performed an integral measurement of this cross section by irradiating samples of natural barium with a linac-produced neutron source, whose energy spectrum was a good approximation to the neutron spectrum in the lunar regolith, and then measuring the neutron-produced ¹³¹Xe with a mass spectrometer. They concluded that the large average ¹³⁰Ba (n, γ) cross section they obtained indeed could account for the "¹³¹Xe anomaly," but pointed out the need for a detailed microscopic crosssection measurement. This paper reports such a measurement.

II. EXPERIMENT

A schematic diagram of the experimental apparatus is shown in Fig. 1. A pulsed 140-MeV electron beam from the Livermore linac (beam-pulse width 15 ns, repetition rate 720 pulses/s, peak

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current 5 A) was directed upon a water-cooled tantalum neutron-producing target. The partially moderated neutrons, which passed through an evacuated and collimated 14.5-m flight tube, struck the (n, γ) sample, which was contained in a thin-walled aluminum can and positioned between a pair of 12.5-cm-diam by 7.5-cm-thick deuterated-benzene liquid-scintillator photon detectors, viewed by fast phototubes. Slabs of graphite, 2.5 cm thick, were placed between the sample and the detectors in order to make possible linear pulse-height weighting of the capture γ -ray signals (see below). The detectors subtended a total solid angle at the sample position of $0.4 \times 4\pi$ sr.

Figure 2 shows a block diagram of the electron-

ics and data-collection system. The discriminator bias-level settings were adjusted, with the aid of various γ -ray sources, to a level corresponding to Compton electrons from 300-keV γ rays. Both the neutron time of flight and the linear pulse height for each event were stored on a magnetic drum in a two-dimensional array consisting of 20 000 time-of-flight channels by 64 pulse-height channels. A linear weighting function was applied to the pulse-height information in order to assure the spectral independence of the capture- γ -ray detector efficiency,⁸⁻¹⁰ and hence of the neutroncapture cross section obtained. We estimate that the uncertainty from this source remaining in the measured capture width for an individual reson-



FIG. 1. Schematic diagram of the experiment. Neutrons produced when the pulsed electron beam from the linac strikes the tantalum target pass through the collimated flight tube and strike the (n,γ) sample, which is viewed, through graphite slabs, by a pair of liquid-scintillator photon detectors.



FIG. 2. Block diagram of the electronics and datacollection apparatus. Both neutron-time-of-flight and capture- γ -pulse-height signals were stored in a twodimensional array on the magnetic drum.

ance is at most 10%; for the integral cross section it is considerably less.

Although the nominal timing resolution was 1.5 ns/m, the experimental energy resolution was determined over a large range by the size of the neutron-producing target. The beam-burst width was so short that the uncertainty in the neutron time of flight does not contribute appreciably to the energy resolution below several keV, and the effects of the sample thickness and detector time resolution are negligible. The measured reso-



FIG. 3. Measured resonance widths plotted as a function of neutron energy. Contributions from Doppler broadening $[2(E_nkTm/M)^{1/2}]$, flight-path uncertainty $(2E\Delta l/l)$, and flight-time uncertainty $(2E\Delta t/t)$ are also shown. Including these effects we find five resolved resonances [shown as triangles $\langle \nabla \rangle$] while all others are unresolved [shown as dots (•)].

nance widths are plotted as a function of neutron energy in Fig. 3 along with the contributions to the widths from flight-path uncertainty, flighttime uncertainty, and Doppler broadening. From this plot, it can be seen that the resonances below about 200 eV are resolved.

Experimental runs were made with samples of enriched ^{130}Ba and natural barium, both in the form of $Ba(NO_3)_2$ powder. The masses of the samples were adjusted so that the number of ^{135}Ba atoms in each sample was the same. The relative

Isotope	Enriched ¹³⁰ Ba s [2.37 g Ba(N	sample 100) ol	Natural barium sample	
	Atomic percentage	(g/cm^2)	Atomic percentage	(g/cm ²)
130	37.05	0.064	0.101	0.00017
132	0.88	0.0015	0.097	0.00016
134	3.52	0.006	2.42	0.0041
135	6.34	0.010	6.59	0.011
136	5.55	0.0095	7.81	0.013
137	6.75	0.012	11.32	0.019
138	39.91	0.069	71.66	0.12

TABLE I. Relative isotopic abundances and sample thicknesses.

isotopic abundances and the thicknesses of the samples are given in Table I.

The energy dependence of the neutron flux was measured by using ¹⁰B as an experimental sample and making use of the $E^{-1/2}$ dependence of the ${}^{10}B(n, \alpha\gamma)$ cross section. For this run, a pulseheight window was set on the discriminators, in effect transforming them into single-channel analyzers, bracketing the 477-keV ^{10}B reaction γ ray, and thus greatly reducing backgrounds from higher-energy γ rays. All the experimental runs were made with a $650 - mg/cm^2$ -thick cadmium filter in the neutron flight path. Use was made of the fact that the cadmium resonance at 89.6 eV, shown in Fig. 4, was completely black in the ¹⁰B run to show that backgrounds resulting from scattered neutrons, faulty collimation, ambient neutrons and γ rays, and the like, were small. The neutron energy scale was set with reference to the known resonances in ¹³⁵Ba, cadmium, and aluminum, as well as a number of well-known resonances throughout the energy range of the experiment observed in a separate experimental run taken with a tantalum sample.



FIG. 4. A portion of the 10 B neutron-spectrum run, showing the transmission dip at 89.6 eV resulting from the presence of the cadmium filter in the neutron flight path. Since no backgrounds have been subtracted from these data, the count rate at the transmission minimum is a measure of the background at that neutron energy.

III. RESULTS

The present ¹³⁰Ba (n, γ) cross-section results are shown in Fig. 5. Well-known¹¹ resonances in ¹³⁵Ba at 24.4, 82, 88, 283, and 405 eV were used to normalize the ¹³⁰Ba cross section. Resonances up to 2.5 keV were assigned to the various barium isotopes by comparing the areas under the resonances for both the enriched ¹³⁰Ba and the natural barium samples. These isotopic assignments are listed in Table II.

There were 39 additional resonances observed between 2.5 and 5.0 keV. Although definite isotopic assignments were not made for these resonances, the majority must be either ¹³⁰Ba or ¹³²Ba resonances. The exception to this is a resonance at 4712 eV which was assigned to ¹³⁶Ba [see Fig. 5(c)]. The values for $g\Gamma_n\Gamma_\gamma/\Gamma$ listed in Table II were obtained for the ¹³⁰Ba resonances from the area A under these resonances using the relation

$$A = 2\pi^2 \lambda^2 g \Gamma_n \Gamma_{\nu} / \Gamma, \qquad (3.1)$$

where χ is the neutron wavelength and g is the statistical factor (2J+1)/[2(2I+1)] (J is the compound-state spin, I is the initial-state spin).

As shown in Fig. 3, there were five resolved resonances assigned to ¹³⁰Ba in these data. For these resonances a total width, Γ , can be extracted by taking into account the effects of Doppler broadening, flight-path uncertainty, and flight-time uncertainty. If we assume that these five resonances are all s wave (l=0, g=1), then we can derive values for Γ_n and Γ_γ from Γ and the resonance area. Values derived for Γ_n and Γ_γ by this method are listed in Table III.

However, the other 35 resonances assigned to ¹³⁰Ba in Table II were not resolved, so that values of Γ cannot be extracted directly from the data. Since the variation of Γ_{γ} from resonance to resonance should be small in this mass region, we can assume that Γ_γ is constant in order to extract values for Γ_n using Eq. (3.1). We assume that $\overline{\Gamma}_{\gamma} = 100$ meV for ¹³⁰Ba, similar to the measured average values of Γ_{γ} for other nuclei in this mass region.¹² (This is also similar to the value of $\overline{\Gamma}_{\gamma}$ = 95 meV derived from our data in Table III.) We also assume that all resonances are s wave. The values of Γ_n thus extracted for all the resonances in the ¹³⁰Ba (n, γ) cross section up to 2775 eV are listed in Table IV. It can be seen from both Tables III and IV that the uncertainties in the derived parameters are large, so that these widths probably should be used only to obtain average resonance parameters.

Although no previous microscopic neutron-capture measurement on ¹³⁰Ba has been reported, there have been a few neutron total-cross-section



FIG. 5. 130 Ba (n, γ) cross section, plotted as a function of incident laboratory neutron energy E: (a) for the energy region $E \le 200$ eV (resolved-resonance region); (b) for 200 eV $\le E \le 1300$ eV (resonances not resolved, except for the one at E = 380 eV; (c) for 1300 eV $\le E \le 9000$ eV (the Al resonance at 5.906 keV is due to the sample container). The cross-section ordinate refers only to 130 Ba resonances; resonances assigned to other barium isotopes are indicated.



FIG. 5 (Continued)

measurements performed on barium isotopes,¹³⁻¹⁵ one of them¹³ with a sample somewhat enriched (14.4%) in ¹³⁰Ba. Some results of these experiments can be compared with the resonance parameters presented here. For the 46.4-eV resonance, Vertebnyi *et al.*¹³ report $E_n = 46.6$ eV and $\Gamma_n = 27 \text{ meV}$; Van de Vijver and Pattenden¹⁵ report $E_n = 48.6$ eV and $\Gamma_n = 30$ meV, but prefer an assignment to ¹³²Ba; and Alves *et al.*¹⁴ report $E_n = 46.2$ eV, but give no isotopic assignment. For the 58.0-eV resonance, Ref. 13 gives E_n = 58.2 eV and Γ_n = 144 meV; Ref. 14 gives E_n = 57.2 eV, $\Gamma_n = 190$ meV, and $\Gamma_{\gamma} = 90$ meV, but assigns it to ¹³²Ba; and Ref. 15 gives $E_n = 57.9$ eV and Γ_n =164 meV. Vertebnyi et al.¹³ also saw resonances at 137 and 186 eV, corresponding to the present values for E_n of 136 and 185 eV, and give them a probable assignment to ¹³⁰Ba. Alves et al.¹⁴ assign six other resonances to 130 Ba between 379 and 896 eV and give Γ_n values for them; we identify 10 resonances for ¹³⁰Ba between 379 and 900 eV, but only two of these definitely correspond to ones reported in Ref. 14. Finally, both Refs. 14 and 15

assign to other barium isotopes a number of other resonances which correspond in energy to ¹³⁰Ba resonances seen in this experiment; no doubt some of these have been misassigned. A comparison of the present results with these various data is given in Table II.

IV. DISCUSSION

The total area under the 40 resonances in the 130 Ba (n, γ) cross section between 46.4 eV and 2.8 keV is approximately 2.0×10^4 beV. From the resonance areas we can obtain a resonance absorption integral (R), where it is defined as

$$R = \int_{E_1}^{E_2} \left[\sigma_{\gamma}(E) / E \right] dE, \qquad (4.1)$$

where the limits of integration refer to the energy range of this experiment. We can approximate this integral by a sum over all the resonances as

$$R \approx \sum_{i} (1/E_{i}) \int \sigma_{\gamma}(E) dE = \sum_{i} (A_{i}/E_{i}), \qquad (4.2)$$

since the energy, E, is essentially constant over

TABLE II. Isotopic assignments of resonance	es up to 2775 eV listed below were obtained from a comparison of en-
riched ¹³⁰ Ba and natural-barium-sample data.	Values for $g\Gamma_n\Gamma_\gamma/\Gamma$ for these resonances were obtained from resonance
areas as discussed in the text.	, .

Present results		Previous assignments ^a			
E_0	Assignment	$g \Gamma_n \Gamma_\gamma / \Gamma^{\text{D}}$	Dof 15	Dof 14	D of 19
(ev)	Assignment	(mev)	Rel, 15	Rei, 14	Ref. 13
24.4	135				
46.4	130	$29.0^{+8}_{-5:8}$	(130, 132)		130
58.0	130	76.9 ± 15.4	(130, 132)	132	130
80.9	135		,		
86.2	135				
102.3	134				
104.2	132		135	135	
125.9	132		137		
136.2	130	$62.5_{-12.5}^{+18.8}$			130
184.5	130	82.9 ± 16.6			130
213	130	0.8 ± 0.2			
219	135				
224	135				
242 ^c	132		135		
283	135				
287	130	26.5 ± 5.3			
316	135				
336	130	50.3 ± 10.0			
358	130	4.0 ± 0.8			
377	(135)				
380	130	42.4 ± 8.5		130	
406	135				
420	(136, 137)		137	137	
425	132				
434	135				
445	130	42.1 ± 8.4	136	130	
466	135				
500	132		134	134	
511	136				
558	130	8.2 ± 1.6			
578	137				
084 651	130	37.2 ± 7.4		105	
651	(136)			135	
000	(135)		105	135	
608	132	54 0 11 0	135		
713	130	34.3 ± 11.0			
723	(134)	23.8 ± 0.0	195	195	
744	130	60 3 + 12 1	199	100	
832	d	00.0 - 14.1	135		
837	130	65.8 ± 13.2	100		
850	130	61.6 ± 12.3			
881	134		135	135	
893	132			130	
903	130	45.5 ± 9.1			
942	(134, 135)			135	
948	132				
973	134				
1029	135				
1048	137				
10 §4	132				
1098	130	74.2 ± 14.8	137		
1121 ^c	(134)				
1149	132				
1156	d		135		
1172	130	77.7 ± 15.5			
1199	d		135		

	Present results		 All About an arts 		Previous assignments ^a	
E_0		$g\Gamma_n\Gamma_\gamma/\Gamma^b$				
(eV)	Assignment	(meV)		Ref. 14	Ref. 13	Ref. 12
1205	132					
1205	(134)			134		
1250	130	4.6 ± 0.9		101		
1266	130	37.3 ± 7.5				
1200	195	01.0 ± 1.0				
13/8	197					
1950	134					
1971	139					
1071	132					
1406	155 d					
1400	u 190	66 1 + 19 9				
1410	120	54.1 ± 10.8				
1400	120	$J_{4.1} \pm 10.0$				
1404	190	91 0 ± 16 9				
1004	130	81.0±10.2				
1570	130	83.2 ± 10.0				
1099 1001 C	(124 125)	04.4 + 10.9				
1641	(104, 100)			136		
1672	d			100		
1685	130	807+161				
1715	d	00,1 = 10,1				
1721	130	90.9 ± 18.2				
1750	132					
1784	130	89.8 ± 18.0				
1824	130	76.2 ± 15.2				
1871	132			135		
1890	(134, 135)			134		
1934	130	102.1 ± 20.4				
1952	(136, 137)					
1977	132					
1998	(134, 135)			135		
2029	132					
2054	132					
2095	130	64.3 ± 12.9				
2121	130	83.3 ± 16.7				
2155	130	58.2 ± 11.6				
2227	(130)					
2256	135					
2387	(130)	85.0 ± 17.0				
2456	(130)	75.4 ± 15.1				
2486	(130)	67.2 ± 13.4				
2775	(130)	61.3 ± 12.3				

TABLE II (Continued)

^a Assignments are listed only where they disagree with present results or where relevant comparisons can be made. ^b Experimental error includes uncertainties in pulse-height weighting, cross-section normalization, background sub-

traction, and flux determination.

^c Possible doublet,

^d Could not be assigned from the present data alone.

the narrow resonances in the ¹³⁰Ba (n, γ) cross section. For the above energy range, we therefore obtain a value for R equal to 180^{+60}_{-40} b. The total uncertainty in this number includes the uncertainties resulting from pulse-height weighting, cross-section normalization, and background subtraction.

To explain the excess 131 Xe found in Apollo-12

lunar rocks, Schwaller *et al.*¹⁶ estimated that the resonance integral for the ¹³⁰Ba (n, γ) cross section must be at least 150 b. This result was derived from cosmic-ray fluxes and the neutron slowing-down density of lunar material. Considering the complexity of understanding the lunar history of a sample, the agreement between our results and

TABLE III. Neutron and total radiation widths for resolved ¹³⁰Ba resonances. These widths were obtained from resonance areas under the assumption that $l = 0_{\text{TV}}$ (g=1) for these resonances. Uncertainties in Γ_n and Γ_γ are correlated as can be seen from Eq. (3.1).

	Pres	ent results	Previous	
<i>E</i> ₀ (eV)	Γ_{γ} (meV) ^a	Γ_n (meV)	$(\text{meV})^{b}$	measurements $\Gamma_n \pmod{(\text{meV})}$
	100.07			
46.4	122 ± 65	38 ± 20	5.6	270
58.0	110 ∓ 38	250 ± 85	32.8	144 ^c
136.2	80 ∓ 26	300 ± 100	25.7	
184.5	130 ∓ 107	230 ± 190	16.9	
380	45 ± 5	1475 ± 140	75.7	1390 ^d

 a The average value of $\overline{\Gamma}_\gamma$ calculated from the first four resonances is 95±20 meV.

^b Reduced neutron width, $\Gamma_n^0 = \Gamma_n (E_n)^{-1/2}$.

^c Reference 13.

^dReference 14.

the above estimate is very good. It is clear that the quantitative results of the present measurement, along with the earlier and more qualitative measurements of Kaiser and Berman,⁷ show that the ¹³⁰Ba(n, γ) reaction indeed is almost surely the source of the excess ¹³¹Xe found in the lunar samples. This knowledge of the ¹³⁰Ba capture cross section therefore provides the key link for correlating burial depth with exposure age for lunar samples.

E ₀ (eV)	Γ _n (meV)	Γ_n^0 (meV) ^a	$\begin{array}{cc} E_0 & \Gamma_n \\ (eV) & (meV) \end{array}$	Γ ⁰ (meV)
46.4 58.0 136.2 184.5 213 287 336 358 380 445 558 558 582 698 713 744	$\begin{array}{c} (\text{me V}) \\ 41^{+15}_{-11} \\ 330 \pm 300 \\ 170^{+140}_{-90} \\ 480 \pm 480 \\ 1 \pm 0.3 \\ 36 \pm 10 \\ 105 \pm 40 \\ 4 \pm 1 \\ 75 \pm 25 \\ 105 \pm 25 \\ 10 \pm 2 \\ 60 \pm 20 \\ 125 \pm 55 \\ 45 \pm 15 \\ 155 \pm 80 \end{array}$	5.9 21.0 42.4 14.7 0.1 2.1 5.7 0.2 3.9 3.6 0.4 2.5 4.7 1.7 5.7	$\begin{array}{c} (ev) & (mev) \\ 1250 & 5\pm 1 \\ 1266 & 60\pm 20 \\ 1416 & 195\pm 115 \\ 1468 & 120\pm 50 \\ 1482 & 75\pm 25 \\ 1534 & 425\pm 425 \\ 1576 & 500\pm 500 \\ 1599 & 540\pm 540 \\ 1685 & 420\pm 420 \\ 1721 & 1000\pm 1000 \\ 1784 & 880\pm 880 \\ 1824 & 320\pm 270 \\ 1934 & c \\ 2095 & 180\pm 100 \\ 2121 & 500\pm 500 \end{array}$	(mev) 0.2 1.7 5.2 3.1 2.0 10.9 12.6 13.5 10.2 24.1 20.8 7.5 3.9 10.9
837 850 903 1098 1172	135 ± 80 195 ± 115 160 ± 80 85 ± 30 290 ± 225 350 ± 310	5.7 6.7 5.5 2.8 8.8 10.2	$\begin{array}{cccccccccccccccccccccccccccccccccccc$	$ 10.9 \\ 3.0 \\ 11.7 \\ 6.2 \\ 4.1 \\ 3.0 $

TABLE IV. Neutron widths obtained for ¹³⁰Ba resonances under the assumptions that $\Gamma_{\gamma} = \text{constant}$ (100 meV) and that all resonances are s wave (g = 1).

^a Reduced neutron width, $\Gamma_n^0 = \Gamma_n (E_n)^{-1/2}$.

^b Notice that assumption of $\Gamma_{\gamma} = 100$ meV leads to large discrepancy for Γ_n between Tables III and IV.

^c Could not be determined by this method.

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How Good Are the Theoretical Internal-Conversion Coefficients?*

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We have accurately measured the total-conversion coefficient for the 156.0-keV, M4 transition in ¹¹⁷Sn as $\alpha_T = 46.40 \pm 0.25$. The Hager and Seltzer theoretical value is 47.8. A comparison between experimental and theoretical α_K and α_T values for 15 E3 and M4 transitions shows that the theoretical values are systematically 2-3% higher.

I. INTRODUCTION

It is generally believed that the theory of internal conversion is in broad agreement with experiment. There exist several tabulations of calculated internal-conversion coefficients. Basically, these calculations require a knowledge of the bound-state and continuum electron wave functions. In the tables of Hager and Seltzer,¹ the wave functions are relativistic Hartree-Fock-Slater solutions to the Dirac equation.

To estimate the uncertainties in any calculation is difficult. It is difficult enough to assess the influence of physically reasonable variations in the assumptions that go into any calculation (what is the correct screening function or nuclear charge distribution? - is a central potential or nonrelativistic treatment adequate? - etc.), not to speak of several "one-percent effects" that are altogether omitted (higher-order terms in the fine-structure constant, static nuclear multipole moments, penetration effects, chemical shifts, electron correlations, etc.). What, in particular, is the combined effect of all these effects? To answer these questions indirectly, we have resorted to the philosophy, "The test of all knowledge is experiment," expressed succinctly by Feynman, Leighton, and Sands.²

II. EXPERIMENTAL PROCEDURE AND RESULTS

To test the theory to say 1%, we of course need a conversion-coefficient (α) measurement that

we can trust to the same accuracy. When an isomer deexcites via two transitions in cascade, it is possible to deduce the α_T ratio by measuring photon intensities only³—a measurement inherently capable of better than 1% accuracy. Consider the case of 14-day ¹¹⁷Sn^m decay shown in Fig. 1. The 156.0-keV (γ_1) transition is known to be M4,⁴ and the 158.6-keV (γ_2) transition M1 + <0.05% E2.⁵ We can write

¹⁶H. Schwaller, P. Eberhardt, J. Geiss, H. Graf, and N.

Grögler, Earth Planet. Sci. Lett. 12, 167 (1971).

$$I_{\gamma_1}(1 + \alpha_{1T}) = I_{\gamma_2}(1 + \alpha_{2T}), \quad \alpha_T = I_{ce}/I_{\gamma}.$$

If the photon-intensity ratio $I_{\gamma_2}/I_{\gamma_1}$ is measured and if α_{2T} is small (say <0.2) and set equal to the experimental or theoretical value, α_{1T} can be readily obtained. The crucial point is that any percentage uncertainty in α_{2T} is reduced by a factor $\alpha_{2T}/(1+\alpha_{2T})$ in the determination of α_{1T} .

The 156.0-158.6-keV γ -ray doublet, well resolved with a 1.0-cm³ Ge(Li) x-ray detector, is shown in Fig. 1. The ¹¹⁷Sn^m sources were produced by the ¹¹⁸Sn(n, 2n) reaction with 14-MeV neutrons. 13 spectra were recorded with two different detectors, two analyzers, and two irradiated ¹¹⁸Sn foils (0.51 and 1.52 mm thick).

Bearing in mind that we are interested only in the relative photopeak areas, an iterative selfconsistent analysis of each spectrum was carried out with the same shape functions (a smoothedstep function to represent the background continuum + a Gaussian term + a tailing term) employed to represent both peaks. Apart from statistical and curve-fitting uncertainties (typically 0.2-0.3%