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Slow Neutron Resonances in Holmium, Thulium, and Lutetium*

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The total cross sections of holmium, thulium, and lutetium have been measured over the neutron energy range from 0.1 to ~30 ev. Resonances were found in Ho^{165} at 3.96, 12.8, 19, 22, and 39 ev; in Tm^{169} at 3.92 14.8, and 17.6 ev. Of the many resonances found in lutetium, the following tentative isotopic assignments have been made: Lu^{175}—resonances at 5.30, 11.4, 14.4, 20.6, 24, and 31 ev; Lu^{176}—resonances at 0.142, 1.57 2.62, and 4.80. It is probable that many additional resonances are present at higher energies in Lu^{176} but were not observed. The average spacing between resonances in Ho^{165}, Tm^{169}, and Lu^{175} (odd-even isotopes) is estimated to be of the same order of magnitude; i.e., respectively 10, 10, and 6 ev; while the spacing in Lu^{176} (odd-odd) appears to be much smaller, of the order of 1 to 2 ev. Parameters for several of the resonances have been obtained.

INTRODUCTION

THE rare earth elements occupy more than half of that region of the nuclear periodic table lying between the closed shells of 82 and 126 neutrons. Neutron cross-section data for these interesting elements are incomplete, largely because of the difficulty in obtaining specimens of adequate quantity and purity.

The three elements reported in this paper are the three heaviest odd-Z elements of the rare earth group. Preliminary results have been reported previously.[1] Other cross-section measurements on these elements have been limited to the thermal cross sections,[2,3] and to a measurement of the holmium cross section from 0.025 to 0.5 ev by Stephenson.[4,5] Holmium and thulium each occur as single isotopes,[6] Ho^{165} and Tm^{169} (both odd-even); while lutetium occurs as two isotopes,[7] Lu^{175} (odd-even) and Lu^{176} (odd-odd). Isotopic abundances, spins, and thermal absorption cross sections are summarized in Table I.

The thermal cross sections of all four isotopes concerned are relatively large, indicating the existence of resonances at neutron energies of a few electron volts. Level spacings obtained from fast neutron activation cross sections[8] furnish information pertaining to expected differences between the resonance structure in the odd-even and the odd-odd isotopes. Spacing between resonances in Lu^{176} would be expected to be significantly smaller according to those measurements.

EXPERIMENTAL DETAILS

Specimens

As previously indicated, the greatest difficulty encountered in these measurements is the problem of obtaining adequate specimens. These measurements were made possible by Dr. F. H. Spedding and his colleagues at the U. S. Atomic Energy Commission's Ames Laboratory, who generously made available excellent specimens of the sesquioxides of these elements. The purity of the samples exceeded 99.9 percent. Thick samples were made by mounting the oxides in precision quartz cells. The thickness of each sample was determined from the weight of the material and the dimensions of the cell. An additional thin sample of thulium was prepared from a solution of accurately measured concentration of $Tm(NO_3)_3$ in D_2O. In measuring the transmissions, comparisons were made between the samples and empty identical quartz cells, and in the case of the solution, with an identical cell filled with D_2O. In general, the samples were not thick enough to obtain reliable measurements in regions where the cross section was less than 20 barns.

Spectrometer

The measurements were made with the BNL crystal spectrometer which has previously been described in detail.[9] Monochromatic neutrons were obtained from the $(12\bar{3}1)$ planes of a single crystal of Be, except below 0.3 ev where the (200) planes of NaCl were used.

RESULTS

Holmium

The total cross section observed for holmium in the energy range from 0.3 to 50 ev is shown in Fig. 1. Resonances were observed at 3.96, 12.8, 19, 22, and 39 ev. An additional resonance observed at 8.2 ev is

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[1] Foote, Sailor, and Landon, Phys. Rev. **90**, 362 (1953).
[2] H. Pomerance, Phys. Rev. **83**, 641 (1951).
[3] H. Bomke and H. Reddemann, Z. Physik **120**, 56 (1943); W. Riezler, Ann. Phys. **41**, 476 (1942); Atterling, Bohr, and Sigurgeirsson, Arkiv Mat. Astron. Fysik **32A**, No. 2 (1946); W. Bothe, Z. Naturforsch **1**, 179 (1946); Seren, Friedlander, and Turkel, Phys. Rev. **72**, 888 (1947); H. Pomerance, Oak Ridge National Laboratory Reports ORNL-577, 33, 1949 and ORNL-366, 47, 1949 (unpublished).
[4] T. E. Stephenson, Oak Ridge National Laboratory Report ORNL-1068, 1951 (unpublished).
[5] Bernstein, Borst, Stanford, Stephenson, and Dial, Phys. Rev. **87**, 487 (1952).
[6] F. W. Aston, Proc. Roy. Soc. (London) **A146**, 46 (1934); W. T. Leland, Phys. Rev. **77**, 634 (1950).
[7] J. Mattauch and H. Lichtblau, Z. Physik **111**, 514 (1939).
[8] Hughes, Garth, and Levin, Phys. Rev. **91**, 1423 (1953).
[9] L. B. Borst and V. L. Sailor, Rev. Sci. Instr. **24**, 141 (1953).

believed due to Sm impurity. The possibility that the 3.96-ev resonance found in this specimen is due to thulium impurity (see next section) has been considered but discounted for the following reasons: The resonant energy for the two cases is significantly different; the other thulium resonances could not be detected in this specimen; and a spectroscopic analysis of the material showed no measurable trace of thulium.[10]

A rough estimate of the level spacing[11] in the compound nucleus at an excitation energy equal to the binding energy of the last neutron can be made from the number of resonances observed in the 50-ev interval. The spacing is seen to be of the order of 10 ev.

A detailed analysis of the observed resonances in order to obtain the Breit-Wigner parameters has not yet been completed; however, some preliminary estimates of the "strength," $\sigma_0\Gamma^2$ of the 3.96- and the 12.8-ev resonances have been obtained. A least-squares fit to the wings[12] of the 3.96-ev resonance gave $\sigma_0\Gamma^2=77\pm10\times10^{-24}$, and an area analysis[13] of the 12.8-ev resonance gave $\sigma_0\Gamma^2=300\pm100\times10^{-24}$ cm^2 ev^2.

It is interesting to examine the relationship of the 3.96- and 12.8-ev resonances to the thermal absorption cross section. In particlular, one would like to see what evidence this information would furnish in regard to the possible occurrence of a resonance at slightly negative neutron energy, i.e., a level in the compound nucleus at an excitation slightly below the binding energy. Such a level was suggested by Wigner as a possible explanation of the anomalously large thermal scattering cross section in Ho found by Stephenson.[4]

There are two possible ways for combining the two resonances to obtain the resultant contribution to the thermal cross section. The first of these methods of

TABLE I. Nuclear properties of Ho, Tm, and Lu isotopes.

Isotope	Abundance (percent)	Spin	Thermal absorption cross section (barns)
Ho^{165}	100[a]	7/2[b]	64[c]
Tm^{169}	100[d]	1/2[e]	118[c]
Lu (element)			108[c]
Lu^{175}	97.4[f]	7/2[g]	16[h]
Lu^{176}	2.6[f]	7[i]	3640[h]

[a] Reference 6.
[b] H. Schüler and T. Schmidt, Naturwiss. 23, 69 (1935).
[c] Reference 2.
[d] F. W. Aston, reference 6, and C. R. Lagergren and M. E. Kettner, Phys. Rev. 80, 102 (1950).
[e] H. Schüler and T. Schmidt, Naturwiss. 22, 838 (1934).
[f] Hayden, Hess, and Inghram, Phys. Rev. 77, 299 (1950); see also reference 7.
[g] H. Schüler and T. Schmidt, Z. Physik 95, 265 (1935).
[h] Seren, Friedlander, and Turkel, Phys. Rev. 72, 888 (1947); see also reference 3.
[i] H. Schüler and H. Gollnow, Z. Physik 113, 1 (1939).

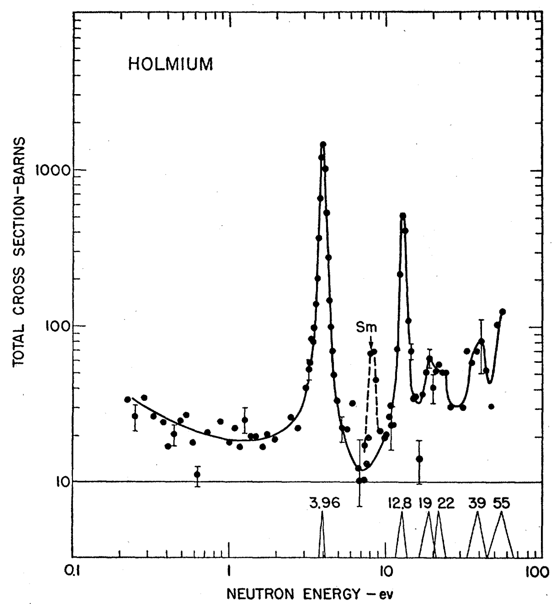

FIG. 1. The observed total cross section of holmium. The resonance at 8.2 ev is at the same energy as an unusually strong resonance in Sm^{152}. The observed peak could be accounted for by about 0.1 percent Sm impurity in the specimen. The statistical error is smaller than the diameter of the points for cross sections greater than 100 barns.

combination would be valid if the levels were of different total angular momentum, in which case the cross section contributions to thermal would be additive. From the single-level dispersion formula one obtains a contribution of 15.5±2.0 barns from the 3.96-ev resonance and 10.3±3.0 barns from the 12.8-ev resonance; both effectively are entirely absorption. Thus, the total of the two would give a thermal absorption cross section of 26±5 barns. Additive cross section contributions from the higher resonances would amount at most to 1 or 2 barns. Thus, on the above assumption, it is seen that the observed resonances do not fully account for the thermal cross section of 64 barns found by Pomerance.[2]

The second method of combination probably must be used when the two levels have the same total angular momentum. For this case, according to the Wigner many-level dispersion formula,[14] it is not valid to add the cross-section contributions of the two resonances, but instead one must add the "absorption amplitudes" to account for interference effects between the two resonances. As far as is presently known, these amplitudes can be both of the same sign or of opposite sign. The two possible combinations of signs give, respectively, 50 barns or 4 barns for the resultant thermal contribution. These numbers serve merely to illustrate the possible importance of interference effects and, of course, could be in serious error because the effects of other resonances cannot be taken properly into account.

[10] F. H. Spedding (private communication).
[11] The level spacing here refers to only those levels which can be excited by slow neutrons, i.e., levels having the same parity as the ground state of the target nucleus and having total angular momentum of $I+\frac{1}{2}$, or $I-\frac{1}{2}$, where I is the spin of the target nucleus.
[12] For method, see V. L. Sailor, Phys. Rev. 91, 53 (1953).
[13] E. Melkonian, Phys. Rev. 90, 362 (1953).
[14] E. P. Wigner, Phys. Rev. 70, 606 (1946).

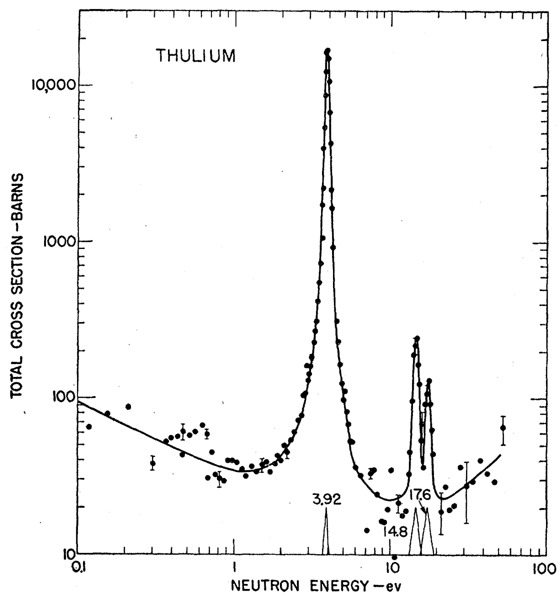

FIG. 2. The observed total cross section of thulium. The statistical errors are smaller than the diameter of the points for cross sections greater than 100 barns.

From the above discussion, it is seen that the question of a negative energy resonance in Ho close to zero energy remains unsettled. It is possible that the shape of the total cross-section curve in the thermal region could give a conclusive answer provided that a sample (e.g., a solution) could be used which eliminated the crystalline coherent scattering effects observed by Stephenson.

Thulium

The total cross section observed for thulium is presented in Fig. 2. Resonances are present at 3.92, 14.8, and 17.6 ev. Data above 30 ev are inconclusive and cannot be considered. The level spacing for this compound nucleus is of the order of 10 ev.

The 3.92-ev resonance has a "strength" $\sigma_0\Gamma^2=380 \pm 40\times 10^{-24}$ cm^2 ev^2, obtained by fitting the wings of the resonance. It should be noted that this resonance is about five times as strong as the corresponding resonance in holmium. Asymmetry in the wings of the thulium resonance indicates that the scattering component is relatively large, although more detailed analysis will be required before a quantitative estimate of $g\Gamma_n$ can be made. This resonance by itself would give a thermal absorption cross section of 78±7 barns, compared with the observed 118 barns.[2]

Lutetium

The total cross section of lutetium over the energy range from 0.3 to 35 ev is presented in Fig. 3. The peak observed at 0.035 ev is the second order of the 0.142-ev resonance. For this spectrometer, second-order contamination becomes serious below 0.08 ev. The resonances observed in lutetium are listed in Table II. The values of $\sigma_0\Gamma^2$ were obtained by the area analysis method.[13] A more detailed analysis was made of the 0.142-ev resonance by fitting the data to the single-level formula. At such a low energy Doppler broadening and instrument resolution could be neglected. The parameters obtained for the resonance were $E_0=0.142 \pm 0.002$ ev, $\sigma_0=350\pm 20\times 10^{-24}$ cm^2, and $\Gamma=0.063 \pm 0.005$ ev. If we assume that this resonance is in Lu^{176} (see discussion below), then several additional combinations of the parameters can be computed.[11] For the isotope, $\sigma_0=13\,500\times 10^{-24}$ cm^2; $\sigma_0\Gamma^2=54\pm 12\times 10^{-24}$ cm^2 ev^2; $g\Gamma_n=(0.925\pm 0.090)\times 10^{-4}$ ev; and $\sum_a =(9400\pm 660)\times 10^{-24}$ cm^2. Since the spin of Lu^{176} is $\geqslant 7$, g can have the values $0.533\geqslant g\geqslant 0.467$, or $g\approx \frac{1}{2}$. Thus Γ_n can be estimated as 1.8×10^{-4} ev.

Tentative isotopic assignments of several of the resonances can be made from the observed values of $\sigma_0\Gamma^2$. Normally within this energy range, values of $\sigma_0\Gamma^2$ (corrected for isotopic abundance) lie within the limits[15] from 50 to 1000×10^{-24} cm^2 ev^2. Examination of Table II shows that the first three resonances are unusually weak and that assignment to Lu^{176} gives more normal values of $\sigma_0\Gamma^2$. The thermal activation cross section of Lu^{176} (Table I) adds further support for the assignment of the 0.142-ev resonance to this isotope. The 4.80-ev resonance is more difficult to assign since the strength would be larger than normal if assigned to 176 and smaller than normal if assigned to 175; however, the weight of the evidence would favor assignment to 176. The 5.30- and the 11.4-ev resonances appear to belong to Lu^{175}. The shape, the observed width, and the very large $\sigma_0\Gamma^2$ of the 14.4-ev resonance strongly

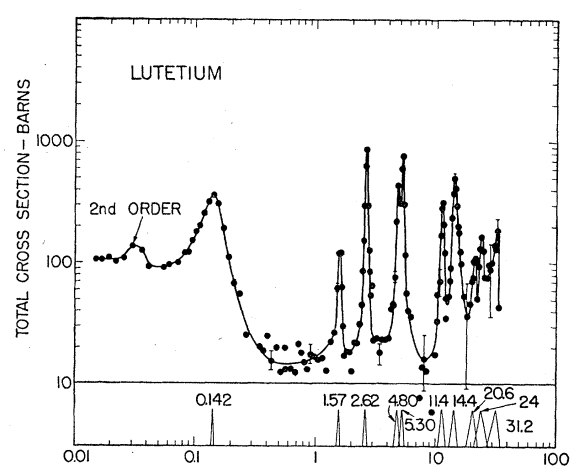

FIG. 3. The observed total cross section of lutetium. The peak at 0.035 ev is the second order of the 0.142-ev resonance. Second-order effects become negligible (<1 percent) above 0.1 ev. Statistical errors are smaller than the diameter of the points for cross sections greater than 100 barns except at very high energies.

[15] W. W. Havens, Jr. and T. I. Taylor, Nucleonics 6, No. 2, 66 (1950) (see Fig. 2).

indicate that this is two or more unresolved resonances. At least one of these belongs to Lu^{175}.

The spacing between the observed resonances must be carefully considered before attempting to estimate the level spacing in the two compound nuclei. Assuming the isotopic assignments to be correct, we see that four resonances occur in Lu^{176} below 5 ev, but none above 5 ev. Calculations show that resonances having a strength of the order of 1 to 5×10^{-24} cm^2 ev^2 would be difficult to observe above 4 or 5 ev with the resolution of this spectrometer. This would be particularly true if other stronger resonances were present to mask the weak ones. It is reasonable to assume, therefore that more resonances were present in Lu^{176} but were unobservable above 5 ev. The only hope of observing Lu^{176} resonances above 5 ev would be to use a specimen enriched in this isotope. If we consider only the range 0 to 5 ev, then the spacing of the Lu^{176} resonances is of the order of 1.2 ev. If all the remaining resonances are in Lu^{175}, the spacing for this target isotope would be of the order of 5 or 6 ev. The 0.142-ev resonance does not fully account for the thermal activation cross section of Lu^{176}; however, the 5.30-ev resonance, if the isotopic assignment is correct, will account for most of the Lu^{175} thermal activation cross section.

TABLE II. Summary of lutetium resonances. The strengths $\sigma_0\Gamma^2$ were obtained by method of area analysis, except for the 0.142-ev resonance, which was obtained by fitting the data to a Breit-Wigner curve. The isotopic strength is obtained by dividing $\sigma_0\Gamma^2$ by the abundance, F.

E_0 (ev)	$\sigma_0\Gamma^2$ (barn ev^2)	Tentative isotopic assignment	Isotopic strength $\sigma_0\Gamma^2/F$
0.142	1.4± 0.3	176	54±12
1.57	0.9± 0.3	176	35±12
2.62	5.5± 2.0	176	210±77
4.80	21. ± 7.	(176?)	810±270
5.30	45. ±15.	175	46±15
11.4	58. ±20.	175	60±20
14.4	(560.)	(175)[a]	...
20.6	...[b]	(175?)	...
24.	...[b]	(175?)	...
31.	...[b]	(175?)	...

[a] This is apparently two or more unresolved resonances, one of which belongs to Lu^{175}.
[b] Data inadequate for analysis.

CONCLUSIONS

The isotopes Ho^{165} and Tm^{169} have slow neutron resonances spaced at intervals of about 10 ev; while the spacing in Lu^{175} is approximately 6 ev, and in Lu^{176} 1.2 ev. The smaller spacing in Lu^{176} is consistent with qualitative expectations. At the present, it is not possible to account unambiguously for the thermal absorption cross sections of these isotopes except possibly Lu^{175}. There is need for further study at higher energies of all these isotopes, and need for a study over the entire energy range with enriched samples of Lu^{176}.

The authors are grateful to Dr. F. H. Spedding for furnishing the excellent rare earth specimens which made these measurements possible. It is a pleasure to acknowledge the valuable assistance of Mr. Ronald Smith in the operation and maintenance of the spectrometer.