NEUTRON CROSS SECTIONS OF Pr, Yb, Lu, Er, Ho and Tm

R. L. ZIMMERMAN, L. Q. AMARAL, R. FULFARO, M. C. MATTOS, M. ABREU and R. STASIULEVICIUS

PUBLICAÇÃO IEA N.º 149

Setembro — 1967

INSTITUTO DE ENERGIA ATÔMICA
Caixa Postal 11049 (Pinheiros)
CIDADE UNIVERSITÁRIA "ARMANDO DE SALLES OLIVEIRA"
SÃO PAULO — BRASIL

NEUTRON CROSS SECTIONS OF Pr, Yb, Lu, Er, Ho and Tm

R.L. Zimmerman, L.Q. Amaral, R. Fulfaro, M.C. Mattos,
M. Abreu and R. Stasiulevicius

Divisão de Física Nuclear
Instituto de Energia Atômica
São Paulo - Brasil

Publicação IEA nº 149
Setembro - 1967

Comissão Nacional de Energia Nuclear

Presidente: Prof. Uriel da Costa Ribeiro

Universidade de São Paulo

Reitor: Prof.Dr. Luiz Antonio da Gama e Silva

Instituto de Energia Atômica

Diretor: Prof. Rômulo Ribeiro Pieroni

Conselho Técnico-Científico do IEA

Prof.Dr. José Moura Gonçalves } pela USP
Prof.Dr. José Augusto Martins }

Prof.Dr. Rui Ribeiro Franco } pela CNEN
Prof.Dr. Theodoreto H.I. de Arruda Souto }

Divisões Didático-Científicas

Divisão de Física Nuclear -
Chefe: Prof.Dr. Marcello D.S. Santos

Divisão de Radioquímica -
Chefe: Prof.Dr. Fausto Walter de Lima

Divisão de Radiobiologia -
Chefe: Prof.Dr. Rômulo Ribeiro Pieroni

Divisão de Metalurgia Nuclear -
Chefe: Prof.Dr. Tharcísio D.S. Santos

Divisão de Engenharia Química -
Chefe: Lic. Alcídio Abrão

Divisão de Engenharia Nuclear -
Chefe: Engº Pedro Bento de Camargo

Divisão de Operação e Manutenção de Reatores -
Chefe: Engº Azor Camargo Penteado Filho

Divisão de Física de Reatores -
Chefe: Prof. Paulo Saraiva de Toledo

Divisão de Ensino e Formação -

NEUTRON* CROSS SECTIONS OF Pr, Yb, Lu, Er, Ho and Tm

R.L. Zimmerman, L.Q. Amaral, R. Fulfaro, M.C. Mattos,
M. Abreu and R. Stasiulevicius

RESUMO

Foram medidas por transmissão as secções de choque totais do praseodímio, itérbio, lutécio, érbio, hôlmio e túlio, para neutrons, no intervalo de energia 0,001 a 1,0 eV, dando-se uma atenção tôda especial quanto à obtenção de valôres da energia térmica 0,025 eV. Usou-se, no reator de pesquisa tipo piscina do Instituto de Energia Atômica, um espectrômetro de cristal acoplado a um monocromador mecânico e um chopper lento. Foram-nos fornecidas amostras dos óxidos, na forma de pó, em alta pureza, tendo-se tido grande cuidado em eliminar a contaminação por terras raras que têm alta secção de choque para neutrons. Os dados foram analisados para se determinar as secções de choque de absorção e de espalhamento nucleares. Foram considerados os efeitos das ressonâncias nucleares e do espalhamento paramagnético atômico. No caso do lutécio, apresentamos os parâmetros da primeira ressonância. Nos casos do túlio e hôlmio, os valôres experimentais das secções de choque de espalhamento paramagnético são comparados com aquêles esperados usando-se as funções de onda eletrônicas calculadas.

RÉSUME

On a mesuré par transmission la section efficace totale du praseodyme, ytterbium, lutetium, erbium, holmium et thulium, pour les neutrons dans l'intervalle d'énergie 0,001 à 1,0 eV, en faisant spéciale attention quant à obtenier des valeurs à l'énergie thermique... 0,025 eV. On a employé, au réacteur de recherche type piscine de l'Instituto de Energia Atômica, un spectromètre à cristal acouplé à un monochromateur mécanique et un chopper lent.

---

* Publicado em Nuclear Physics A95 (1967) 683-693; (C) North-Holland Publishing Co., Amsterdam

Des échantillons en poudre des oxides nous ont été fournis avec haute pureté, en faisant spécialement attention pour éliminer la contamination par des terres rares de haute section efficace pour les neutrons. Les données ont été analysées pour déterminer les sections efficaces d'absorption et de diffusion nucléaires. On a considéré les effets des résonances nucléaires et de la diffusion paramagnétique atomique. Dans le cas du lutetium, on présente les paramètres de la première résonance. Dans les cas du thulium et holmium, les valeurs expérimentales de la section efficace de diffusion paramagnétique sont comparées avec celles espérées en utilizant les fonctions d'onde électroniques calculées.

NEUTRON CROSS SECTIONS OF Pr, Yb, Lu, Er, Ho and Tm

R. L. ZIMMERMAN, L. Q. AMARAL, R. FULFARO, M. C. MATTOS, M. ABREU and R. STASIULEVICIUS

Instituto de Energia Atômica, São Paulo, Brasil

Received 18 November 1966

Abstract: The total neutron cross sections of praseodymium, ytterbium, lutetium, erbium, holmium and thulium have been measured by transmission within the neutron energy range 0.001 to 1.0 eV, with special attention given to obtain values at the thermal energy 0.025 eV. A crystal spectrometer coupled with a mechanical monochromator and a slow chopper were used at the swimming pool research reactor of the Instituto de Energia Atômica. Powder samples of the oxides were supplied in high purity with particular care to eliminate contamination by the rare earths which have high neutron cross section. The data were analysed to determine the nuclear absorption and the nuclear scattering cross sections. Effects of nuclear resonances and of the atomic paramagnetic scattering were considered. In the case of lutetium, the parameters of the first resonance are presented. In the cases of thulium and holmium, experimental values of the paramagnetic scattering cross sections are compared with those expected from calculated electronic wave functions.

E | NUCLEAR REACTIONS Pr, Yb, Lu, Er, Ho, Tm (n, n), $E = .001$ to 1.0 eV; measured $\sigma_{nT}(E)$; deduced σ_{nA}, σ_{nS}; Ho, Tm deduced paramagnetic scattering, Lu deduced Γ, Γ_n^0. Natural targets.

1. Introduction

The interpretation of the total interaction between slow neutrons and rare-earth atoms involves both the high density of nuclear resonances and the magnetic forces between the neutrons and unpaired orbital electrons. Previous measurements of rare-earth neutron cross sections have often not been over a neutron energy range appropriate for the analysis of these interactions.

This work describes an effort to measure and to analyse the total cross sections of Pr, Yb, Lu, Er, Ho and Tm over an energy interval (0.001 to 1.0 eV) such that it is possible to determine the nuclear interactions and the paramagnetic interaction owing to their different energy dependences. Some of the results in this work have been published previously [1-3].

High purity powder oxides were supplied by the IEA Chemistry Division. For the samples which are particularly sensitive to contamination by the rare earths which have high neutron cross section, cross-section measurements were made before and after a final purification to insure that an otherwise undetectable quantity of gadolinium was not affecting the results. A crystal spectrometer coupled with a mechanical

velocity selector that eliminates order contamination [4,5]) was used for most measurements; part of the lutetium results were obtained with a slow chopper time-of-flight equipment [6]).

2. Data analysis

The total cross sections calculated from transmission measurements are shown in figs. 1–6. The oxygen cross section was subtracted incoherently at all energies.

For isolated rare-earth atoms at ordinary temperatures, the total neutron cross section may be expressed as the sum of three partial cross sections

$$\sigma = \sigma_s + \sigma_a + \sigma_{pm},$$

where σ_a is the nuclear absorption cross section, σ_s the nuclear scattering cross section and σ_{pm} the paramagnetic scattering cross section, which results from the electromagnetic interaction between the magnetic moments of the neutron and that of the atom. A transmission measurement, such as we used, while yielding a relatively precise value of the total cross section, gives information about the partial cross sections only through their known different dependences on neutron energy.

The nuclear scattering σ_s although generally consisting of resonance scattering, given by the Breit-Wigner formula, potential scattering and sometimes coherent interference between the two, is constant near thermal energy unless the total cross section gives evidence that a nuclear resonance falls in this region. In those cases, its behaviour was calculated from measurements of the resonance parameters. Coherent effects in the oxide crystals occur at lower energies, where the nuclear absorption usually dominates, and were neglected.

The absorption cross section σ_a varies with energy according to the Breit-Wigner formula whose parameters must be determined experimentally. In general, nuclear resonances far removed from the thermal neutron energy, as well as bound states, must be considered to calculate the departure from a $1/v$ dependence near thermal energy.

The behaviour of the paramagnetic scattering cross section is given by the average magnetic form factor of the unpaired atomic electrons $\overline{f^2}$ in the formula

$$\sigma_{pm} = \tfrac{2}{3}\pi \left(\frac{e^2}{mc^2}\right)^2 \gamma^2 \mu^2 \overline{f^2},$$

where μ and γ are the magnetic moments of the atom and of the neutron, respectively, and are expressed in Bohr magnetons, and e, m and c have their usual meaning. The asymptotic value at neutron wavelengths greater than 10 Å for which the form factor approaches unity depends only on the magnetic moments. In every case we analysed our data using the asymptotic value calculated from the known atomic magnetic moments [7]). In two cases, holmium and thulium, the paramagnetic cross section is a large fraction of the total cross section such that information about the form factor

may be derived from our measurements and compared with form factors calculated [8]) from the 4f wave functions of Blume, Freeman and Watson [9]).

Qualitatively, our analysis procedure may be described as follows: At very low energies, nuclear absorption dominates and may be well determined by subtracting the nearly asymptotic value of σ_{pm} and an estimated value of σ_s. The important value of nuclear absorption at thermal energy is then obtained by extrapolation from the low-energy value, calculating the departure from $1/v$ dependence caused by the known resonances and by the most probably bound states. The bound state parameters are fortunately restricted by the knowledge of their contribution to low-energy absorption; i.e., the difference between the observed absorption and that from all the positive resonances.

At high energies, the paramagnetic cross section is approaching zero, and the nuclear absorption has sometimes become small enough to allow determination of the scattering cross section.

At some intermediate energy, in principle, it is possible to determine the paramagnetic scattering cross section and, therefore, the magnetic form factor. The relative magnitudes of the cross sections were such that this was possible only for the cases of holmium and thulium, as shown in figs. 8 and 9, and discussed below.

3. Discussion of results

3.1. THE THULIUM CROSS SECTION

Thulium is a typical example of the means of analysing the experimental data, and fig. 1 shows its total cross section versus energy. The absorption cross section dominates at very low energy, where it has a $1/v$ behaviour. Over this region the nuclear scattering cross section gives a comparatively small contribution of little influence, and the asymptotic value of the paramagnetic cross section is well known; the value 35 b is given by the interaction between the magnetic moment of the neutron and the one from the ion of thulium [7]). In this region of energy, the absorption cross section was determined and afterwards extrapolated to higher energies, taking into account the deviation from $1/v$, in the case of thulium, which is almost entirely due to the first resonance at 3.92 eV. At 0.025 eV thermal energy, the total cross section was observed to be 134 ± 2 b, and we determined the absorption cross section as 106 ± 3 b.

The contribution to thermal absorption by all positive resonances was calculated using resonance parameters known within certain experimental errors [10]). This contribution is 73 ± 10 b; 33 ± 10 b is attributed to bound states or energy levels below the binding energy of the neutron.

The curve of the absorption cross section that appears in this figure is composed of the contribution both of positive resonances and of bound states, the latter gives a small deviation from $1/v$ for higher energies, that depends on the parameters assumed for the bound state.

The value and the limits of variation of the scattering cross section necessary to explain the observed values near 1 eV, where the influence of the paramagnetic cross section is known to be of little influence, were determined analysing the contributions from positive resonances and from bound states and their uncertainties. A value of 12 ± 2 b for the nuclear scattering cross section was taken as constant over the considered energy range. The error quoted here includes a contribution from the uncertainties in the published resonance parameters, as well as the uncertainty in the bound state parameters which are, of course, not completely determined.

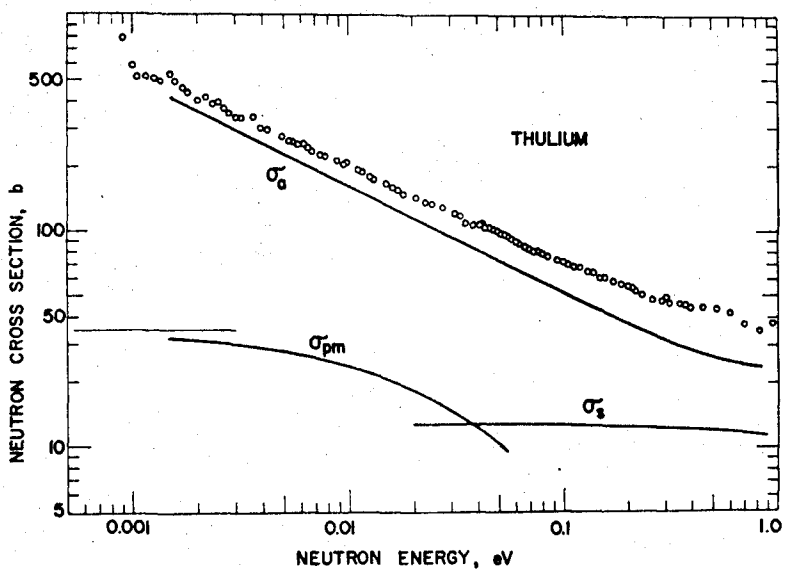

Fig. 1. Total cross section of thulium. The observed data shown as open circles were analysed as a sum of the nuclear absorption, paramagnetic scattering and nuclear scattering, σ_a, σ_{pm} and σ_s, respectively, shown by the solid curves. The functional form σ_{pm} is shown for reference only and, except for the asymptotic value shown here at 35 b, is not used to determine the nuclear partial cross sections.

The curve of the paramagnetic cross section appearing in fig. 1 was calculated [8]) using the paramagnetic form factors calculated with Hartree-Fock wave functions for isolated ions of rare earths tabulated by Blume, Freeman and Watson [9]). The nuclear partial cross sections were determined without the use of this calculated curve, except for low energies very near its experimentally known asymptotic value. Therefore, the experimental neutron cross sections may be used at other energies to determine the paramagnetic scattering cross section. Fig. 8 shows the paramagnetic cross section expressed as a ratio to the asymptotic value and the average magnetic form factor calculated from the theory of ref. [9]). Although there is some indication that the form factor falls more slowly at short wavelengths than expected theoretically, indicating more diffuseness in the 4f orbits, the discrepancies are not serious.

3.2. THE HOLMIUM CROSS SECTION

Fig. 2 shows the results obtained for holmium. The analysis of the data used the asymptotic paramagnetic cross section of 68 b. At thermal energy, an absorption

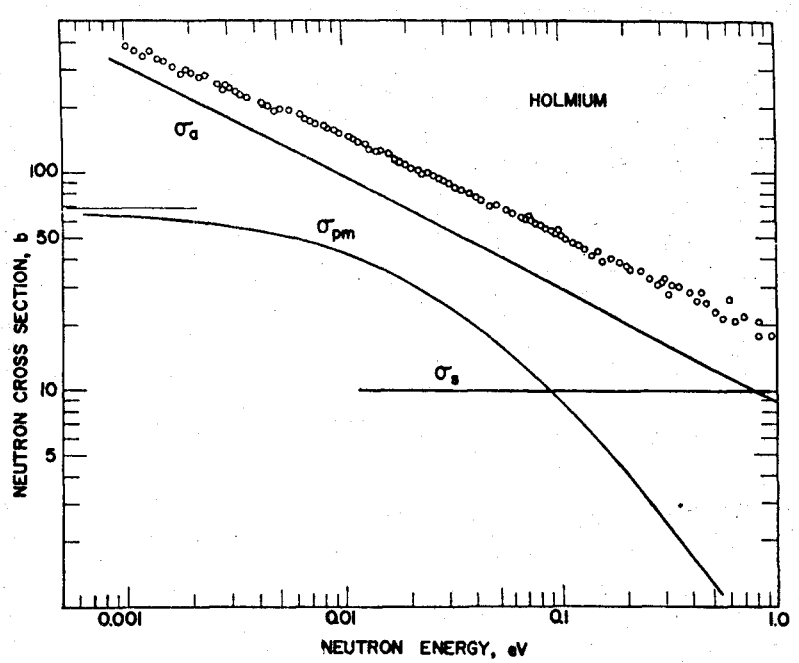

Fig. 2. Total cross section of holmium. The observed data were analysed similarly to those of thulium.

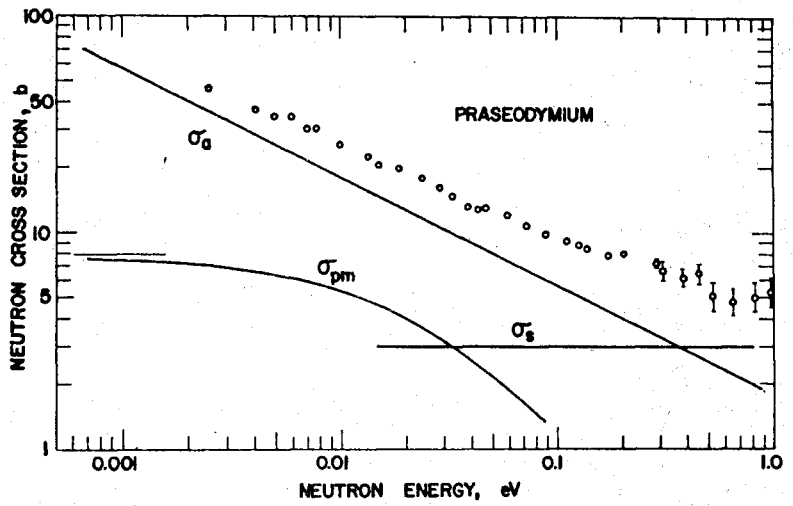

Fig. 3. Total cross section of praseodymium.

cross section of 60 ± 2 b was determined, the total cross section being 98 ± 2 b. The contribution of positive resonances has been calculated as 23 ± 2 b, which leaves

37±2 b to be attributed to contributions from bound states; they cause a small deviation from $1/v$ in the region near 1 eV. The limits of this deviation and its most probable value were calculated to determine the value and probable error of the scattering cross section which is determined in this energy region. A value of 10±2 b for the nuclear scattering cross section was found.

Fig. 9 shows the holmium results for the paramagnetic cross section, obtained in the same way as for thulium. Again there is no serious disagreement between the experiments and the calculated form factors.

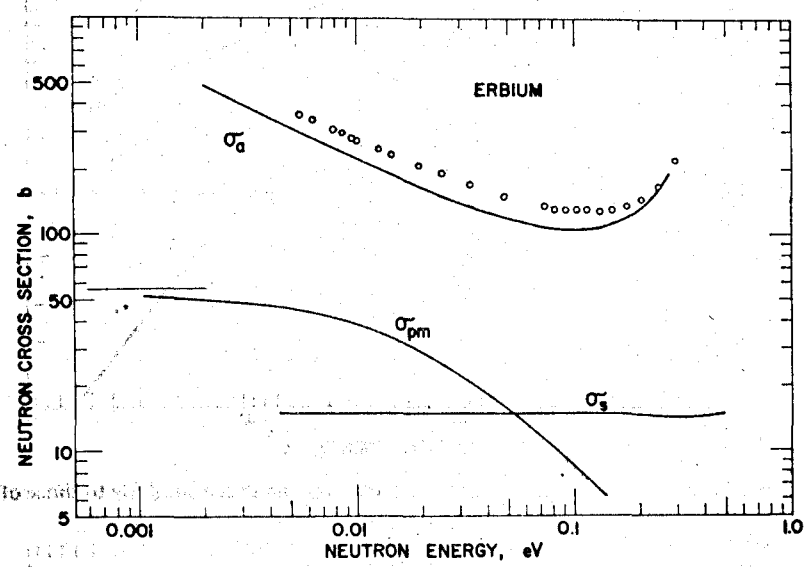

Fig. 4. Total cross section of erbium.

3.3. THE PRASEODYMIUM CROSS SECTION

Fig. 3 shows the results obtained for praseodymium. The total cross section at thermal energy is 17.6±0.5 b, and the thermal absorption is 11.5±1.0 b from the asymptotic paramagnetic cross section of 7.8 b. Although about 10 b of the thermal absorption are due to bound states, the absorption in this case is probably $1/v$ because of the large level spacing. Near 1 eV the total cross section was used to determine the nuclear scattering as 3±1 b. Because the magnetic moment of praseodymium is small, no analysis was made for the paramagnetic cross section.

3.4. THE ERBIUM CROSS SECTION

Fig. 4 shows the total cross section of erbium. The total cross section at 0.025 eV is 192±6 b, and the absorption value determined at this energy was 150±8 b when using the asymptotic paramagnetic cross section of 56 b. Practically all absorption comes from the first resonances at 0.46 and 0.574 eV. Within the experimental errors of the resonance parameters published, no contribution due to bound states can be

detected. A nuclear scattering of 15 ± 5 b was used in our analysis. Because the absorption is dominant over the whole energy region considered, our data no not well determine the nuclear scattering or the paramagnetic scattering cross section.

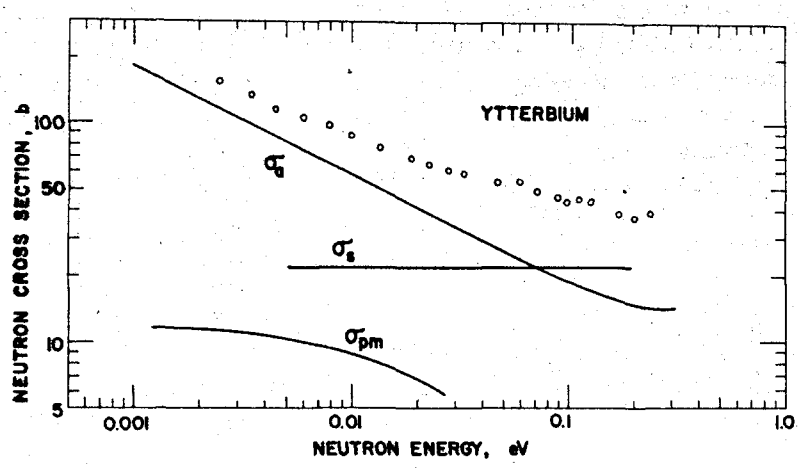

Fig. 5. Total cross section of ytterbium.

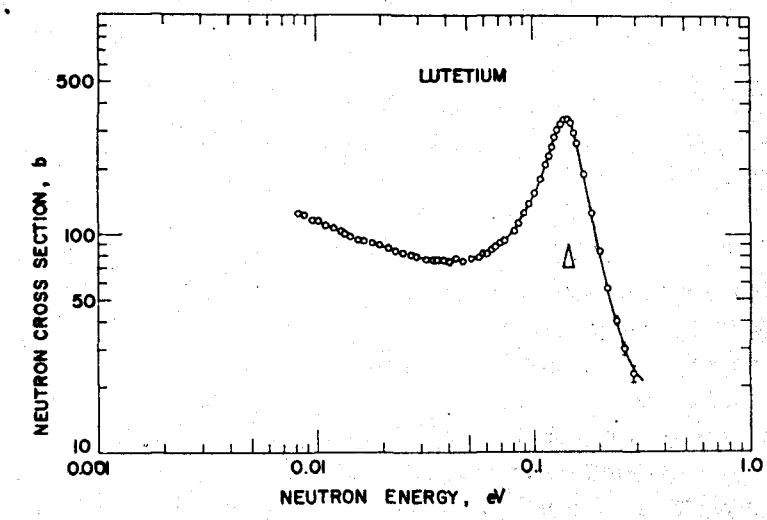

Fig. 6. Total cross section of lutetium. The experimental points below 0.04 eV were taken with a slow chopper. The rest were taken with a crystal spectrometer as with all the other elements. The solid curve shows the Breit-Wigner fit.

3.5. THE YTTERBIUM CROSS SECTION

Fig. 5 shows the ytterbium results. The total cross section at thermal energy is 64 ± 2 b with thermal absorption 37 ± 5 b and nuclear scattering 22 ± 5 b, when using the asymptotic value of the paramagnetic cross section 12.3 b. To this thermal absorption, only about 5 b are contributed by the first resonance at 0.597 eV. No

other parameters have been published, but the level spacing appears [10]) to be large enough so that the rest of the absorption should have a $1/v$ dependence.

3.6. THE LUTETIUM CROSS SECTION

The experimental results shown in fig. 6 are the lutetium total cross section versus neutron energy and were obtained with the crystal spectrometer for energies higher than 0.04 eV and with the slow chopper for energies below this value. In this case there is no paramagnetic cross section, because the magnetic moment of the Lu ion is zero. In the energy region measured the isotope ^{176}Lu presents a resonance due mostly to absorption.

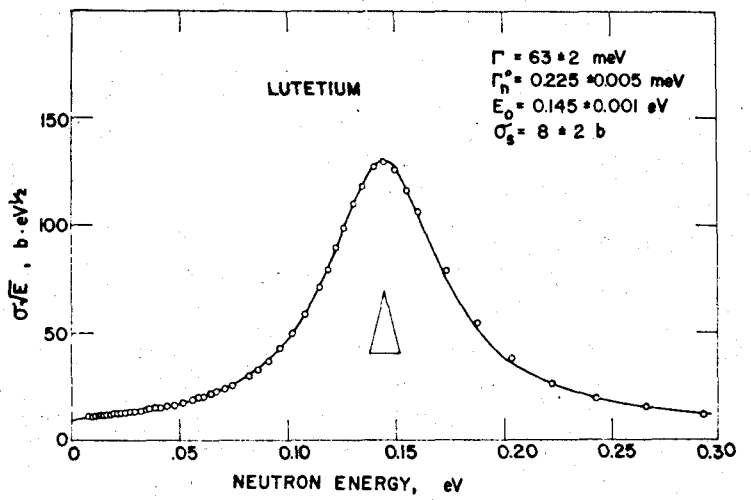

Fig. 7. Plot of $\sigma\sqrt{E}$ for lutetium. The experimental points and the Breit-Wigner fit are the same as in fig. 6. The triangle shows the resolution of the crystal spectrometer.

The total cross section was studied in detail, and the Breit-Wigner parameters were determined by adjusting the theoretical curve to the experimental results. The small effects of finite resolution and Doppler effect have been considered.

In fig. 7 a plot of $\sigma\sqrt{E}$ versus the neutron energy with the curve obtained is shown. In this energy region there is predominance of the S-wave interaction, and the usual Breit-Wigner formula can be used for the resonance cross section. The effect of interference between potential and resonance scattering is negligible. The nuclear scattering is considered constant over the considered energy region. The influence of absorption peaks at other energies gives a $1/v$ contribution. Small deviations at the higher energies occur because there is a contribution from bound states.

The following resonance parameters have been obtained:

$$E_0 = 0.145 \pm 0.001 \text{ eV},$$
$$\Gamma = 63 \pm 2 \text{ meV},$$
$$\Gamma_n^0 = 0.225 \pm 0.005 \text{ meV}.$$

From our analysis, it was possible also to separate the absorption and scattering partial cross sections at thermal energy, the total cross section being 82 ± 3 b. A value 8 ± 2 b was obtained for the nuclear scattering, leaving a thermal absorption of 74 ± 3 b, of which about 11 b are contributed by bound states.

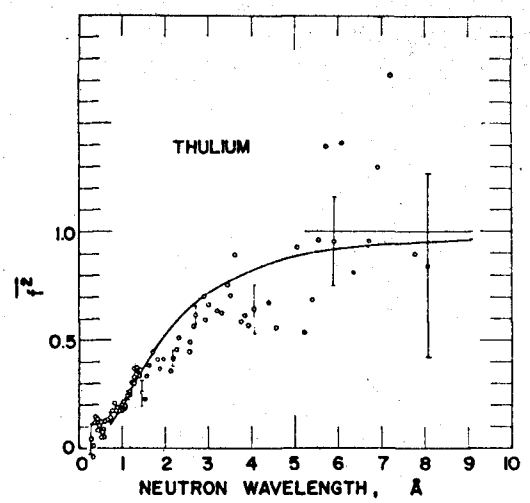

Fig. 8. The average magnetic form factor of thulium obtained from the observed paramagnetic cross section divided by its asymptotic value of 35 b. Typical statistical errors are shown. The solid curve is calculated from the wave functions of ref. [9]).

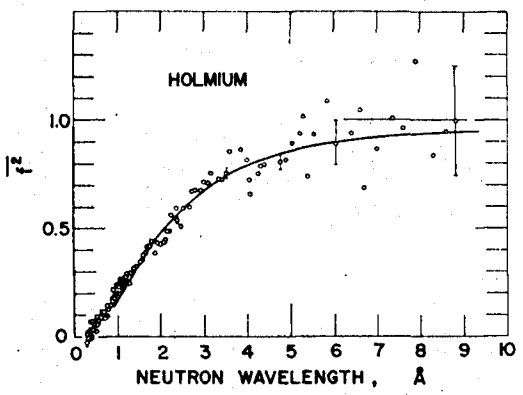

Fig. 9. The average magnetic form factor of holmium. The statistical errors are relatively smaller than those shown for thulium in fig. 8 because the nuclear absorption is greater in the latter case.

4. Conclusion

Table 1 shows a summary of the observed total cross sections and the derived partial cross sections at thermal energy for the six elements. The partial cross sections have greater errors due to uncertainties in the separation of the partial cross sections. The nuclear scattering cross sections listed here are not to be interpreted necessarily as the potential scattering. Interference with resonance scattering may give scat-

tering at thermal energy larger or smaller than the potential scattering by an amount dependent on the parameters of resonances and bound states, which are now insufficiently well known.

Our paramagnetic cross section of holmium may be compared with a similar analysis by Bernstein *et al.* [11]). Their paramagnetic cross section differs considerably from ours although their total cross sections agree with ours. Their use of 13 b for nuclear scattering, 64 b for thermal absorption, and their analysis before the discovery of the 3.92 eV resonance all contributed to a prediction of the paramagnetic cross section that is lower than ours.

TABLE 1

Thermal cross sections in b

	σ_{total}	$\sigma_{absorption}$	$\sigma_{scattering}$
Pr	17.6±0.5	11.5±1.0	3±1
Yb	64 ±2	37 ±5	22±5
Lu	82 ±3	74 ±3	8±2
Er	192 ±6	150 ±8	(15±5)
Ho	98 ±2	60 ±2	10±2
Tm	134 ±2	106 ±3	12±2

Our observation of the average holmium form factor similarly disagrees with a calculation used as a comparison in the experimental work by Koehler *et al.* [12]), who used the theory of Trammell [13]) with hydrogen-like wave functions.

Our results agree with the form factor calculated from the wave functions of Blume, Freeman and Watson [9]), who also mention the discrepancy with Trammell.

The authors want to express their gratitude to the many colleagues who helped with the experiments, especially to the reactor staff under the supervision of Azor Camargo Penteado and to Dr. Brill, Dr. Alcidio Abrão and Dr. Ludmilla Federgrün, who purified and analysed the samples. The authors are grateful to Professor Marcello Damy de Souza Santos for his interest and support during the course of the experiments.

References

1) R. L. Zimmerman, M. Abreu, O. Martins and M. C. Mattos, Proc. 4th Inter-American Symp. on the peaceful application of nuclear energy, Vol. I Mexico City (Pan American Union, Washington, 1962) p. 55

2) L. Q. Amaral, M. Abreu, F. G. Bianchini and M. C. Mattos, Proc. Study Group Meeting on the utilization of research reactors, Vol. II, São Paulo (IAEA, Vienna, 1965) p. 133
3) R. Fulfaro, M. C. Mattos and R. Stasiulevicius, Tech. Report IEA 124 (São Paulo, Brasil, July, 1966)
4) R. Brenner and R. L. Zimmerman, Proc. of the Study Group Meeting on the utilization of research reactors, Vol. II, São Paulo (IAEA, Vienna, 1965) p. 123
5) F. G. Bianchini, M. Abreu, L. Q. Amaral and O. W. Martins, *ibid* p. 107
6) S. B. Herdade, L. Q. Amaral, L. A. Vinhas and C. Rodriguez, Ciência e Cultura (Brasil) **18** (1966) 69
7) J. A. Gibson and G. S. Harvey, Technical report AFML-TR-65-430 (January 1966)
8) M. C. Mattos, Technical report IEA 99, São Paulo, Brasil (August 1965); presented at the 5th rare-earth research conf. Ames, Iowa, (1965)
9) M. Blume, A. J. Freeman and R. E. Watson, J. Chem. Phys. **37** (1962) 1245
10) D. J. Hughes and R. B. Schwarts, BNL 325 (1958)
11) S. Bernstein *et al.*, Phys. Rev. **87** (1952) 487
12) W. C. Koehler, E. O. Wollan and M. K. Wilkinson, Phys. Rev. **110** (1958) 37
13) G. T. Trammell, Phys. Rev. **92** (1953) 1387