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PUBLICAÇÃO IEA N.º 124

Julho — 1966

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CIDADE UNIVERSITÁRIA "ARMANDO DE SALLES OLIVEIRA"
SÃO PAULO — BRASIL

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RESUMO

Foi medida a secção de choque total do hôlmio e túlio, para nêutrons de energia entre 0,001 e 1 eletron volt. Foi usado o espectrômetro de cristal do IEA acoplado a um seletor mecânico de velocidade para eliminar reflexões de ordem superior do cristal. Amostras de Ho_2O_3 e Tm_2O_3 em pó foram preparadas em alto gráu de pureza, tomando-se cuidado especial em eliminar contaminação por terras raras de alta secção de choque, para nêutrons. Pode-se obter informações definidas sôbre o raio da órbita eletrônica 4f através de medidas precisas da secção de choque total num grande intervalo de comprimentos de onda conhecidos. Nossas medidas, levando em conta o espalhamento paramagnético, permitiram-nos a determinação das secções de choque parciais, algumas delas com melhor precisão do que havia sido publicado anteriormente.

RESUMÉ

La section efficace de l'holmium et du thulium a été mesurée pour les neutrons d'energie entre 0,001 et 1 electron volt. Le spectromètre à cristal de l'IEA a été employé acouplé à un sélecteur mécanique de vitesse pour éliminer les réflexions d'ordre supérieur du cristal. Des echantillons de Ho_2O_3 et Tm_2O_3 en poudre ont été preparés avec haute pureté, en faisant spécialement attention pour éliminer la contamination par des terres rares de haute section efficace pour les neutrons. Des informations définies sur le rayon de l'orbite electronique 4f peuvent être obtenues par des mesures précises de la section efficace totale dans un large intervalle de longueurs d'onde connues. Nos mesures, en prennant compte de la diffusion paramagnétique, nous ont permis la determination des sections efficaces partielles, quelques unes avec meilleur precision qu'il avait été publié precedémment.

ABSTRACT

The total neutron cross section for holmium and thulium was measured by transmission for neutron energies between 0.001 and 1 electron volt. The IEA crystal spectrometer was used together with a mechanical velocity selector to eliminate higher order reflections from the crystal. Powder samples of Ho_2O_3 and Tm_2O_3 were prepared in high purity, taking particular care to eliminate contamination by the rare earths of high neutron cross section. Definite information on the radius of the 4f electron orbits can be given by accurate total cross section measurements for a wide range of known wavelengths. Our measurements, taking into account the paramagnetic scattering, have enabled us to determine the partial cross sections, some with better accuracy than those previously published.

I - INTRODUCTION

In a program of total cross section measurements of the rare earth elements at this Institute, holmium and thulium were given special attention, because of the interesting interaction between the neutrons and the atomic electrons, and because of their high absorption cross section for thermal neutrons. Being holmium and thulium present in the fragments of nuclear fission, their cross sections play an important part in reactor physics calculations which must predict the effects of neutron absorbers in a nuclear reactor.

Thulium, in a (n, γ) reaction becomes Tm^{170} that has a half-life of 129 days and emits a 84 KeV gamma ray, being for this reason used as a low energy gamma source in industry and medicine.

II - EXPERIMENTAL METHOD

The source of neutrons for this work was the Instituto de Energia Atômica swimming pool research reactor operated at 2 MW. The thermal neutron flux measured with gold foils near the core of the reactor is 2.4×10^{12} neutrons/cm^2.sec.

A crystal spectrometer and a mechanical velocity selector, constructed at the Instituto de Energia Atômica's workshop, were used as monochromators. Order contamination was eliminated from the beam by using the crystal together with the mechanical velocity selector.

The crystal spectrometer was first located close to the reactor in a radial beam-hole, as shown in Figure 1. Later, the crystal and the selector were located at one of the tangential beam-holes, as shown in Figure 2. The thermal flux outside the radial port was found to be 4×10^6 neutrons/cm^2/sec. and 1.4×10^8 neutrons/cm^2/sec. outside the tangential port. The crystal spectrometer angles can be reproduced within a precision of 0.01 degree. The measurements were made with crystals of calcite,

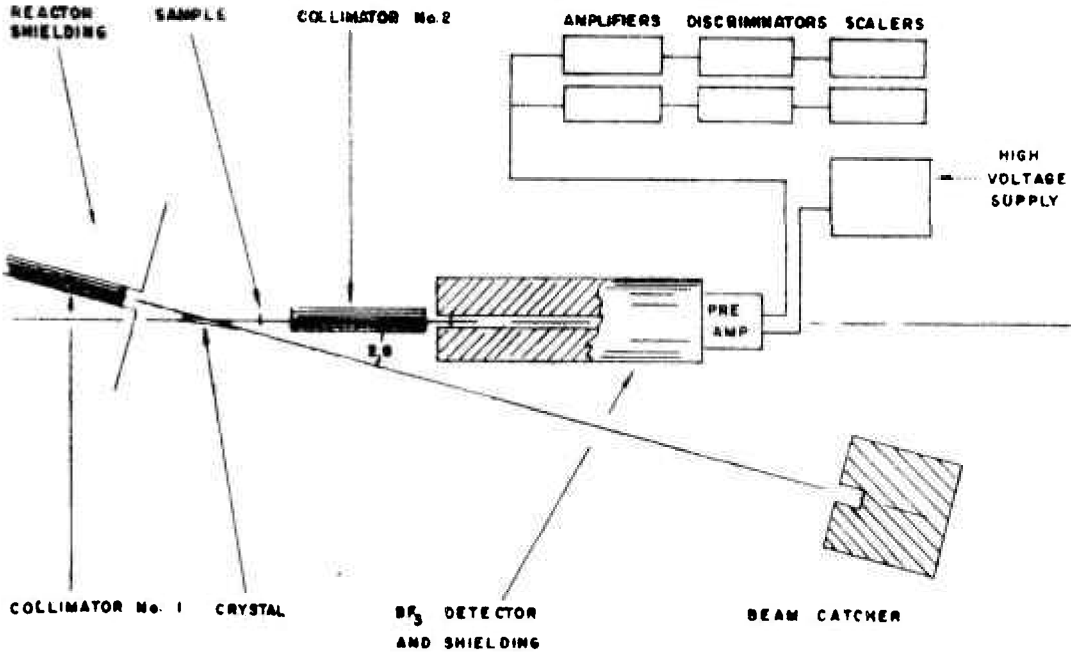

Figure 1 - Schematic diagram of the crystal spectrometer

aluminium and mica, having interplanar distances of 3.03, 1.21 and 9.84 Angstroms, respectively. With the calcite crystal the angular resolution, determined mostly by the initial collimation, was 10 minutes and at thermal energy the resolution in energy was 2.0%.

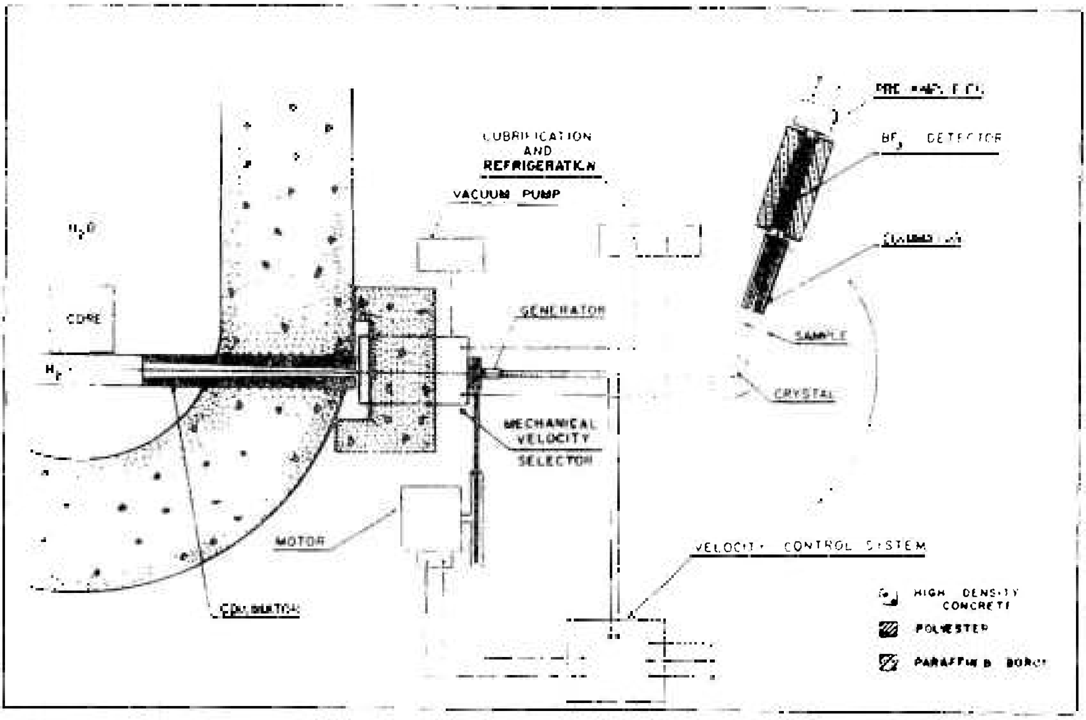

Figure 2 - Schematic diagram of the mechanical velocity selector together with the crystal spectrometer

The mechanical velocity selector(1) gives a neutron energy with a resolution which depends on the rotation velocity, and on the helical channel inclination. The resolution was of about 50% in wavelength. The final resolution was determined by the crystal spectrometer and order contamination was completely eliminated by the mechanical velocity selector.

Commercial boron tri-fluoride detectors, enriched in the isotope B^{10}, were used for neutron detection.

The samples of holmium and thulium oxides were supplied by the Chemical Engineering Division of this Institute. The separation method employed, using ion exchange resins, assured us the degree of purification required for this experiment (99,7% for holmium and 99,9% for thulium).

The samples were placed in aluminium containers and introduced into the beam in a reproducible position. The transmission through the sample was obtained by measuring the counting rate with the sample in the beam, and the rate obtained with an identical empty sample holder in the beam. The background was subtracted from each counting. The containers were designed to give a transmission which minimized the time required to reduce the statistical errors(2).

To avoid the influence on the transmission of the fluctuations of the reactor power, the transmission measurements were repeated several times in cycles, according to a routine designed to cancel linear drifts. The detector pulses were amplified, analyzed and counted by two independent electronic systems.

The total cross sections were calculated from the transmission measurements. The conventional formula(2) has been used for the errors. The calculations were made by the IEA IBM-1620 computer. The correction due to oxygen was made simply by subtracting the free atom oxygen cross section of 3.8 barns per atom.

Figures 3 and 4 show the results obtained for holmium and thulium, respectively.

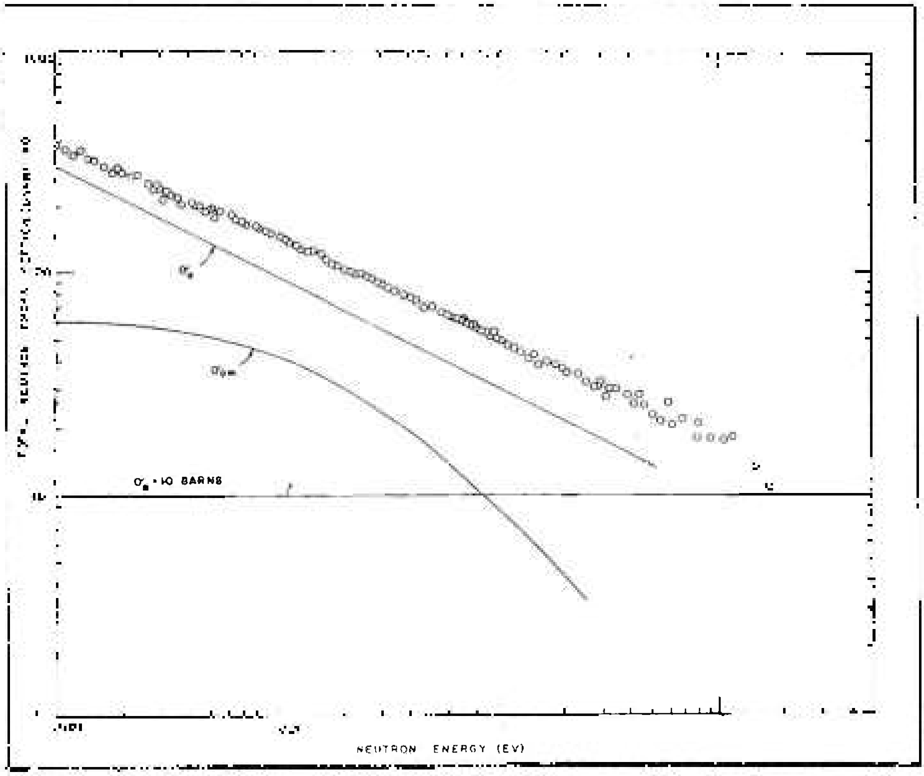

Figure 3 - Total neutron cross section of holium as a function of neutron energy. The absorption cross section, σ_a, which is dependent on $1/\sqrt{E}$, and the calculatet paramagnetic cross section, σ_{pm}, are shown.

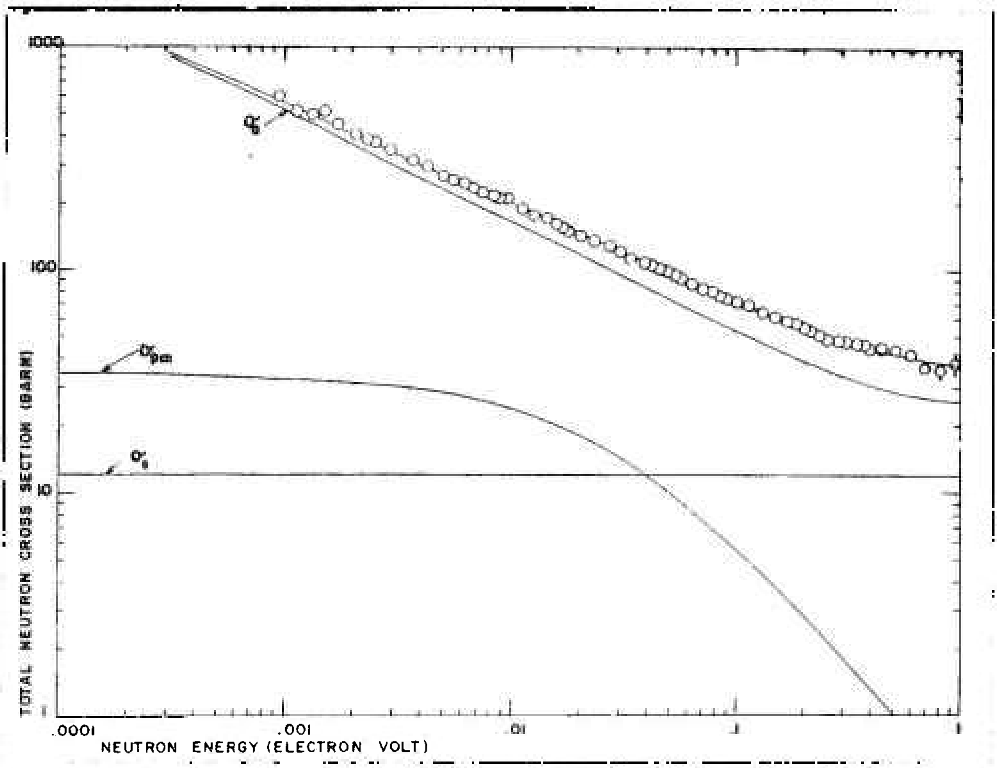

Figure 4 - Total neutron cross section of thulium as a function of neutron energy. The absorption cross section, σ_a, which is dependent on $1/\sqrt{E}$, and the calculated paramagnetic cross section, σ_{pm}, are shown.

III - ANALYSIS OF DATA

The total cross section of holmium and thulium, σ_T, consists of three completely independent partial cross sections. Then,

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

where σ_s and σ_a are the nuclear scattering and absorption, respectively, and σ_{pm} is the paramagnetic cross section, due to the interaction between the magnetic moment of the neutron and that of the atom.

The paramagnetic scattering component to the neutron scattering has been calculated by using the paramagnetic form factors calculated(3) with Hartree-Fock wave functions for isolated ions of rare earths and using the values tabulated by Blume , Freeman and Watson(4).

Nuclear scattering was assumed to be independent of energy in the interval measured and the absorption cross section was assumed to vary as $1/\sqrt{E}$ over almost all the range of this experiment.

For holmium and thulium, the nuclear resonance spacing is about 6 electron volts(5). The first resonance at 3.92 electron volts, for both of them, has no effect on the assumptions concerning the dependence on energy of the nuclear absorption cross sections, except for the higher energies. Nuclear scattering contributes little to our experimental data, except for the high energy limit of our experiments, where its effect is noticeable.

The value of the nuclear scattering was determined in the region of higher energies, where it dominates, and where there is no influence from paramagnetic scattering. The determined nuclear scattering for holmium is

$$\sigma_s = 10 \text{ barns},$$

and for thulium,

$$\sigma_s = 12 \text{ barns.}$$

Nuclear absorption dominates at low energies and may be quite well determined by subtracting the asymptotic value of the paramagnetic scattering,

$$\sigma_{pm} = 65.2 \pm 1.3 \text{ barns, for holmium}$$

or

$$\sigma_{pm} = 35.4 \pm .7 \text{ barns, for thulium,}$$

calculated(3) from the holmium or thulium magnetic moment(6). The nuclear absorption cross section thus determined from our data, for holmium, is

$$\sigma_a = 61 \text{ barns,}$$

and, for thulium, is

$$\sigma_a = 105.7 \text{ barns,}$$

reduced to their values at thermal neutron energy .025 electron volts. Our extrapolation to thermal energy correctly accounted for the deviation of σ_a from the $1/\sqrt{E}$ curve.

With the aim of obtaining more information about the thermal absorption cross section of thulium, we calculated the contribution of all the known resonances, σ_{res}, and this added up to 73 barns. Since, however, $\sigma_a = 105.7$ barns, there were still about 33 barns left to be explained. Remembering that, when nuclei are bombarded by slow neutrons, effects due to levels below the neutron binding energy in the compound nuclei are generally observed(7), the values for the absorption cross section and for the contribution of all the resonances indicated that the 33 barns had to be due to those levels.

One of our aims in analyzing σ_a, for thulium, was the establishment of limiting values for our experimental value of σ_s. We verified that the contribution due to bound states does not influence our determination of the nuclear scattering. However, σ_{res} has been calculated basing on the tabulated resonance parameters, that have certain errors. Thus we determined $\sigma_{res} = 73 \pm 10$ barns. Having analyzed the variations in σ_{res} and having adjusted σ_s for each case, in such a way that the experimental points could be explained, the maximum and minimum variation in σ_s was calculated from the experimental data. Thus we determined, for thulium,

$$\sigma_s = 1\text{[illegible]} \pm \text{[illegible]} \text{ barns.}$$

By compounding the deviations in σ_T and σ_s, the standard deviation in σ_a can be evaluated:

$$\sigma_a = 105.7 \pm 3.0 \text{ barns.}$$

Besides this, the experimental value of the thermal paramagnetic cross section was determined by subtracting σ_a and σ_s from the thermal total cross section. Thus, for thulium, a value,

$$\sigma_{pm} = 16.8 \pm 3.5 \text{ barns,}$$

was obtained which agrees with the calculated(3) value for the thermal energy, that is

$$\sigma_{pm} = 16.5 \text{ barns.}$$

The curve for the total cross section of thulium, described by the experimental points, was formed by the curves of the calculated σ_a, σ_s and σ_{pm}.

A similar analysis was made for the holmium data, for obtaining the errors in σ_s and σ_a. We obtained, for holmium,

$$\sigma_s = 10 \pm 1.5 \text{ barns}$$

and

$$\sigma_a = 61 \pm 3 \text{ barns,}$$

at thermal energy.

IV - CONCLUSIONS

Figure 5 shows, for holmium, our experimental points after having subtracted the contributions due to nuclear scattering and absorption.

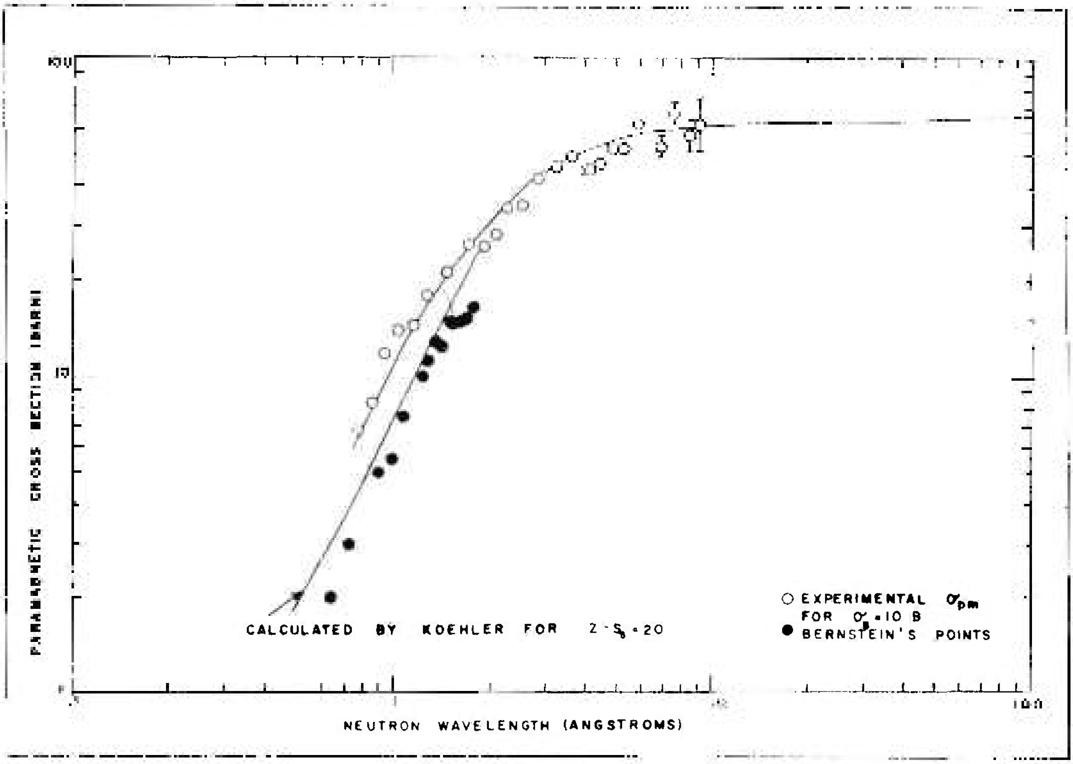

Figure 5 - Comparison between the experimental paramagnetic cross section of holmium, obtained after having subtracted the contribution of nuclear scattering and absorption, and the theoretical paramagnetic cross section, calculated from the form factors of Blume, Freeman and Watson(4) . The older theory quoted by Koehler *et al.*(9) is shown. The apparently discrepant points due to Bernstein et al.(8) are explained in the text.

Some information unavailable from the theory can be obtained by comparing experimentally determined paramagnetic scattering cross section with that expected from the Hartree-Fock wave function calculations, using the values tabulated by Blume, Freeman and Watson(4). One sees that, for our holmium results, the disagreement is not serious. However, a real test of the theory depends on an accurate measurement of the nuclear scattering cross section, which is not available.

The determined values of σ_{pm}, for holmium, by Bernstein et al.[8] are different from ours although Figure 3 shows that the total cross sections agree in the region where their statistical errors were comparable with ours. Their paramagnetic cross sections differ from our values which were estimated using more recently available information. Their results were corrected using a nuclear scattering cross section of 13 barns, based on a total cross section of 28 barns at .5 eV. From the compilation of all total cross section values it seems that 28 barns is at least 3 barns too high, as it can be seen in Figure 3. In addition, the effects of the then unknown 3.92 eV resonance on the capture cross section at .5 eV (about 1 barn) were not taken into account by them, and the paramagnetic scattering cross section at .5 eV was understimated by about 1 barn. Taken together, their estimate of 13 barns for nuclear scattering is about 3 barns too high. On our lower energy range, the nuclear scattering is relatively unimportant; in the absence of direct measurements, a value of 10 ± 1.5 barns was used based on the analysis already described. Finally, analysis with our new value of the thermal absorption cross section of 61 ± 3 barns instead of 64 assumed by Bernstein et al. accounts for the remaining small discrepancy between the analysis of our results and that of their careful measurements.

Figure 5, relative to holmium, also shows the disagreement between the older calculation quoted in the experimental work of Koehler et al.(9), who used the theory by Trammell(10) with hydrogen -

like wave functions, and the calculation based on the publication of Blume, Freeman and Watson(4), who used the same theory with Hartree-Fock wave functions. Blume, Freeman and Watson noticed this disagreement. The interpretation of our results, according to Trammell's theory, favours the smaller 4f shell implied by the more recent calculations of Blume, Freeman and Watson.

Measurements of total cross section of thulium have previously been made by Joki and Evans(11), in a smaller interval of energy, from .038 to 1 electron volt; σ_T at .025 electron volts was not measured by them, but an extrapolated value is given for the thermal energy,

$$\sigma_T = 134 \pm 3 \text{ barns},$$

and for the partial cross sections,

$$\sigma_s = 6.5 \pm 3.0 \text{ barns, and}$$

$$\sigma_a = 127 \pm 4 \text{ barns.}$$

When one compares our results, for thulium, in the region where they measured, a good agreement is observed. It should be noticed, nevertheless, that in their analysis of data the paramagnetic interaction was not taken into account and that, if it had been, they would have obtained

$$\sigma_a = 110.5 \pm 4 \text{ barns},$$

that agrees with the result obtained by us

$$\sigma_a = 105.7 \pm 3.0 \text{ barns.}$$

These two values for σ_a can be compared with

$$\sigma_a = 106 \pm 20 \text{ barns},$$

obtained by activation by Seren, Friedlander and Turkel(12). Our value, however, does not agree with the pile oscillator value of

$$\sigma_a = 118 \pm 6 \text{ barns},$$

given by Pomerance(13).

Table I gives our results and a comparison with previously published measurements.

TABLE I

Thermal Cross Section

Element	The Present Experiment				Previously Published Results				Reference
	σ_T	σ_a	σ_s	σ_{pm}	σ_T	σ_a	σ_s	σ_{pm}	
Ho	99.7 ± .2	61 ± 3	10 ± 1.5	27.9	97 ± 3			16.5	8
								24.5	9
						59.6 ± 11.0			12
						64 ± 3			13
Tm	134.5 ± 2.0	105.7 ± 3.0	12 ± 2	16.5	134 ± 3	127 ± 4	6.5 ± 3.0		11
						106 ± 20			12
						118 ± 6			13

V - ACKNOWLEDGMENTS

The interest and encouragement of Prof. M.D. de Souza Santos are greatly acknowledged here. The authors wish to thank Dr. R. L. Zimmerman for valuable discussions and S.B. Herdade for his assistance in carrying out the experiments.

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