TOTAL NEUTRON CROSS SECTIONS OF PRASEODYMIUM, YTTERBIUM, LUTETIUM AND ERBIUM

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RESUMO

A secção de choque total foi medida para os óxidos das terras raras praseodímio, itérbio, lutécio e érbio, pelo método da transmissão, no intervalo de energia 0,0005 a 1 electron volt, com especial atenção para conseguir valores na energia térmica 0,025 ev. As amostras dos óxidos foram fornecidas por uma indústria local, com alto grau de pureza. Para essas medidas, no reator de pesquisa tipo piscina do Instituto de Energia Atômica, usou-se como monocromador um espectrômetro de cristal e um seletor mecânico de velocidade. Foi estudada uma ressonância nuclear do Lu^{176} que se apresenta no intervalo de energia considerado; os parâmetros encontrados concordam com os valores publicados. O comportamento, previsto teòricamente, da secção de choque em baixa energia permitiu-nos a determinação das secções de choque parciais na energia térmica, entretanto com precisão não muito grande.

RESUMÉ

La section efficace totale des oxides des terres rares praseodymium, ytterbium, lutetium et erbium a été mesurée dans l'intervalle d'énergie de neutrons 0,0005 à 1 électron volt, en faisant spéciale attention quant à obtenir des valeurs

à l'énergie thermique 0,025 ev. Les échantillons des oxides ont été fournis par une industrie local, avec haut dégré de pureté. Pour ces mesures, au réacteur type piscine de l'IEA, on a utilisé comme monochromateur un spectromètre à cristal et un sélecteur mécanique de vitesse. On a étudié une résonance nucléaire du Lu^{176} qui se présente dans l'intervalle considéré ; les paramètres trouvés sont d'accord avec les valeurs publiées. Le comportement prévu théoriquement de la section efficace à basse énergie nous a permis la détermination des sections efficaces partielles à l'énergie thermique, cependant pas avec grande précision.

ABSTRACT

The total neutron cross section of oxides of the rare earths praseodymium, ytterbium, lutetium and erbium has been measured by transmission within the neutron energy range 0.0005 to 1 electron volt, very special attention having been given to obtain values at the thermal energy 0.025 ev. The oxide samples were supplied by a local industry at a high degree of purification. For these measurements, a crystal spectrometer and a mechanical velocity selector were used as monochromators at the swimming pool research reactor of the Instituto de Energia Atômica. A nuclear resonance presented in the considered energy range in Lu^{176} has been studied; the parameters found agree with the published values. The theoretical expected behaviour of the cross section at low energy has enabled us to determine the partial cross sections at thermal energy, however not too accurately.

INTRODUCTION

The cross sections of the rare earths for neutron absorption have been given very special attention. Because these rare

earths are present in the fragments of nuclear fission, their cross sections play an important part in the calculations of reactor physicists who must predict the effects of neutron absorbers in a nuclear reactor. In addition, the rare earth nuclei are among those which have the highest absorption cross sections below 1 Mev[1].

While these reasons have prompted experimental physicists to collect a large amount of data[2], a few elements still exist whose cross sections have never been measured in the thermal neutron energy. Although the measurements themselves are in principle very simple, the materials are often very rare and difficult to purify sufficiently.

We are fortunate in Brazil to have a large source of rare earth materials in the mineral monazite, as also a local industry well equipped to separate and purify the individual rare earths. One of these industries has isolated some of these materials in large quantities and in a high degree of purification.

This paper is presented as a complement to "Neutron Cross Sections of Praseodymium, Ytterbium and Lutetium"[3], by Zimmerman et al. Many of the values already published have been retaken, in order to furnish a more complete table.

EXPERIMENTAL APPARATUS

The neutrons for this work were produced in the Instituto de Energia Atômica reactor, a Babcock Wilcox swimming pool research reactor designed to operate at a maximum power of 5 Mw. For part of this experiment the reactor was operated eight hours each week at 2 Mw, the schedule having since been

increased to sixteen hours a week, at the same power.

The thermal neutron flux measured with gold foils near the core of the reactor is 1.2×10^9 nv/kw.

A crystal spectrometer and a mechanical velocity selector constructed at the shops of the Instituto de Energia Atômica were used as monochromators.

The crystal spectrometer is very efficient when used in the range of 0.025 to 1 ev, due to its good resolution. However, a crystal does not provide efficient energy selection for neutrons of sub-thermal energies (i.e. below the peak of the Maxwellian distribution) because of the interference of reflected neutrons in second and higher orders. This contamination may be eliminated from the beam when we use a crystal together with a mechanical velocity selector; this instrument may be used alone in the sub-thermal region if only a monochromator of relatively low resolution is required.

In a general way, the spectrometer is similar to what has been described in the literature [4,5,6,7]. Angles may be read on a vernier scale with a precision of 0.01 degree. The supporting base of the crystal has six degrees of freedom and may be adjusted manually to optimize the alignment. The measurements were taken with a natural crystal of calcite and one of hematite, their respective interplanary distances being 3.03 Å and 4.77 Å; the latter was used for the sub-thermal measurements.

The energy resolution of a neutron crystal monochromator is determined to some extent by the initial collimation, the collimation on the arm of the spectrometer, and by the mosaic structure of the crystal. It is seldom determined wholly by one or two of these

as may be observed in the literature. The collimators were designed so as to allow the resolution to be determined almost entirely by the crystal structure. With calcite crystal the resolution is 18 minutes.

The IEAR-1 mechanical velocity selector is similar to the one described by Dash and Sommers[8]. The selector design and construction supervision were by R.L.Zimmerman, a scientist on leave from the Case Institute of Technology, who spent one year at the I.E.A. It consists of 30 cadmium covered steel disks, each one having on its periphery 36 equally spaced slots; these disks are mounted along an axis, in such a way that a cylinder of 74 cm is formed, with 36 helical channels on its surface. When the selector turns at a given rotation velocity, only neutrons of velocity within a given range can go through the helical channel. The selected energy and the resolution depend on the rotation velocity which is electronically controlled, as also on the helical channel inclination. When we use the selector as a filter for eliminating higher order contamination in the crystal, we can work with poor resolution, thus increasing the intensity. Having this aim in mind, we used a helical channel inclination which provides a resolution of 50% in wavelength. For measurements using the selector as monochromator the channel inclination has been increased, giving a resolution of 25% in wavelength.

Neutrons entering the mechanical velocity selector came from one of the reactor beam-hole through ports. The crystal spectrometer was first located close to the reactor in a horizontal experimental port, as shown in figure 1; later it has been used assembled to the selector as shown in figure 2.

Outside the beam-holes we have a thermal flux of 2×10^3 nv/kw in the horizontal port and of 7×10^4 nv/kw in the through port.

Commercial boron tri-fluoride detectors, enriched in the isotope B^{10}, were used for neutron detection. A good part of the electronics, including essential components, was manufactured by local industry from advanced modern designs.

The samples were placed in aluminium holders and introduced into the beam in 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; a background was subtracted from each counting.

To avoid the influence on the transmission of the variations of the reactor power and the instability of the electronics, 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; in this way, the stability of the systems can be tested continuously by the relation between the two counting rates.

SAMPLE PREPARATION

The oxides of praseodymium, erbium, lutetium and ytterbium were placed in containers designed to give the best transmission[9].

The samples used were supplied by the Orquima S.A. Lab; the method employed, using ionic resins, assured us of the degree of purification necessary for these experiments. Only lutetium oxide presented 8.5% of ytterbium oxide.

The effects of sample contamination by water were noticed in the first measurements by the increase of the measured cross

section with time. To avoid this contamination, the samples were calcined for one hour at 900°C and stored in a dessicator during the measurement interval.

The absence of gadolinium in a quantity that could affect the value of the measured cross section was checked by measuring the cross section before and after a purification process which would reduce the gadolinium concentration by more than a factor of 100; no variations of the measured cross sections larger than the experimental error have been noticed.

The stoichiometric formulae used for the oxides were[10]:

Lu_2O_3, Yb_2O_3, Er_2O_3 and Pr_6O_{11}.

On the three first oxides small differences from these formulae do not affect the measurements because lutetium, ytterbium and erbium have cross sections much higher than oxygen. For the praseodymium oxide the assumed formula was Pr_6O_{11}; this formula has been determined after one-hour calcination at 900°C, by dissolving the oxide in hydrochloric acid, in the presence of potassium iodide. The free iodine corresponds to the active oxygen, not stoichiometric, existing in the Pr_2O_3. This iodine is titrated with sodium thiosulfate[3].

EXPERIMENTAL RESULTS

Through transmission measurement, total cross section has been calculated; the conventional formula has been used for the error[8]. Some of the calculations were made manually, others by an IBM-1620 computor. The corrections due to oxygen were made; for lutetium, the correction due to ytterbium oxide present in the sample was made.

The sub-thermal region has been covered by measurements made with the selector used as a filter for the crystal and as a monochromator, except for lutetium; all lutetium data were obtained with the crystal spectrometer. The second order correction was made by using as standard sample gold, whose microscopic cross section is known with precision.

In the tables of figures 3,4,5 and 6, the results obtained are shown; the error quoted includes the systematic error in determining the sample thickness as well as the statistical errors for the individual measurements. All results referring to lutetium appeared in a previously published paper[3], for which the crystal spectrometer has been used alone as a monochromator. Ytterbium and praseodymium values in the region from 0.46 to 0.025 ev have also been retaken.

The experimental results were compared to the theoretical cross section. In the region where we work we have predominance of interactions of neutrons l = 0 with nuclei. In this case, the scattering and capture cross sections, in the neighbourhood of a resonance energy E_o for the neutrons, are given by the Breit-Wigner formulae:

$$\sigma_{scat}(E) = \pi \lambda^2 f_g \left| \frac{i\,\Gamma_n}{(E - E_o) + \frac{i}{2}\Gamma} + \frac{4\pi R}{\lambda} \right|^2 + 4\pi R^2 (1 - f_g)$$

and

$$\sigma_{cap}(E) = \pi \lambda^2 f_g \frac{\Gamma_n \Gamma_\gamma}{(E - E_o)^2 + \left(\frac{\Gamma}{2}\right)^2} .$$

where $\Gamma = \Gamma_n + \Gamma_\gamma$ and $g = \frac{1}{2}\left(\frac{2J + 1}{2I + 1}\right)$

with $J = I \pm \frac{1}{2}$

In these formulae I is the spin of the target nucleus, of radius R; J is the spin of the compound nucleus and f is the abundance of the isotope presenting resonance. The width of the level Γ_γ changes slightly with energy, and can be considered as a constant, and we have $\Gamma_n = \Gamma_n^0 \sqrt{E}$, with Γ_n^0 constant.

For the total cross section we should consider the influence of the absorption peaks in higher energies, which contribute with an addend proportional to $\frac{1}{\sqrt{E}}$ in lower energies. For the considered elements, the existing nearby resonances are due chiefly to absorption, i.e. $\Gamma_\gamma \gg \Gamma_n$; we can thus neglect the term due to the interference between the amplitude of potential scattering and that from these nearby resonances.

So we have for the total cross section.

$$\sigma_t = fg\pi\lambda^2 \frac{\Gamma_n \Gamma}{(E - E_0)^2 + (\frac{\Gamma}{2})^2} + \sigma_s + \frac{A}{\sqrt{E}} ,$$

where σ_s is the cross section due to potential scattering together with a slowly varying scattering cross section arising from the interference between potential scattering and scattering from distant resonances; A is a constant that can be estimated for the far-away resonances.

For lutetium, we were able to determine the parameters of the resonance presented by the Lu^{176} isotope in the studied interval, by adjusting the theoretical curve to the experimental one, after having made the correction due to the finite resolution. With our triangular resolution function, the cross section in the peak has had a correction of 2% and the other parameters correction was of the same order. No corrections were made for the Doppler effect.

It is known that lutetium has a very high spin, equal to 9 or 10, and so g is very close to 0.5. The value σ_s has been assumed through values of nearby nuclei.

With $f = 0.0264$, $g = 0.5$ and $\sigma_s = 10$ barns the adjustment of the curves gave the following values:

$$E_o = 0.141 \pm 0.002 \text{ ev},$$
$$\Gamma_\gamma = 0.0584 \pm 0.003 \text{ ev},$$
$$\Gamma_n^o = 0.000201 \pm 0.0000004 \text{ ev and}$$
$$A = 10 \text{ barns.ev}^{1/2},$$

that are in accordance with the published values[2]. The theoretical curve, affected by the resolution, is seen as well as the experimental points in figures 7 and 8.

For ytterbium, we did not measure experimentally the cross section near the closest resonance peak, which we know is due to the Yb^{168} isotope, of spin zero, but we worked in a region where its influence can be felt. We assumed the published values[11,2]

$$E_o = 0.597 \text{ ev}$$
$$\Gamma = 0.073 \text{ ev and}$$
$$\sigma_o \Gamma^2 = 1.28 \text{ barns.ev}^2,$$

where σ_o is the cross section value at energy E_o. The cross section may be written as a function of these parameters and we have

$$\sigma_t = \frac{\sigma_o \Gamma^2 \sqrt{\frac{E_o}{E}}}{4(E - E_o)^2 + \Gamma^2} + \sigma_s + \frac{A}{\sqrt{E}},$$

where Γ is considered as a constant.

The adjustment of the theoretical curve to the experimental one led to the value $\sigma_s = 20$ barns and $A = 6.0$ barns. $\text{ev}^{1/2}$.

The theoretical curve and the experimental points can be seen in figure 9.

For erbium, we have two close resonances, at 0.047 and 0.058 ev, due to the Er^{167} isotope. Also for this element, we did not measure the cross section near the resonance peaks, but in a region where their influence can be felt. As there are no parameters of these resonances in the literature, we made an adjustment only valid in the region where they contribute to the total cross section with an addend proportional to $\frac{1}{\sqrt{E}}$. Thus

$$\sigma_t = \sigma_s + \frac{A}{\sqrt{E}}$$

has been adjusted to the experimental points at low energy, and we obtained σ_s = 10 barns and A = 26.2 barns. $ev^{1/2}$. The experimental points may be compared to this approximation in figure 10.

The measured cross sections of praseodymium are relative to energies much smaller than the one of the closest resonance peak, that is at E_o = 85.5 ev. In the region where we work, the same formula used for erbium is valid. The curves adjustment has been obtained assuming σ_s = 3 barns and A = 2.2 barns. $ev^{1/2}$ and the theoretical curve, as well as the experimental points, are seen in figure 11.

We determined, experimentally, the total cross section of the four elements at the thermal energy 0.025 ev. A comparison with values already published could be made only for partial cross sections, because there are no previous measurements of the total cross section at the thermal energy. Through the

theoretically expected behaviour of the cross section, we were able to determine the partial scattering and absorption cross sections, though not too accurately. In our partial cross sections, a relatively large experimental error has had to be taken into account because of the uncertainty in making the separation of the total cross section into two components. The result of this analysis is shown in figure 12, as well as the previously published values[2,12,13]. It can be seen that there is coherence between these values.

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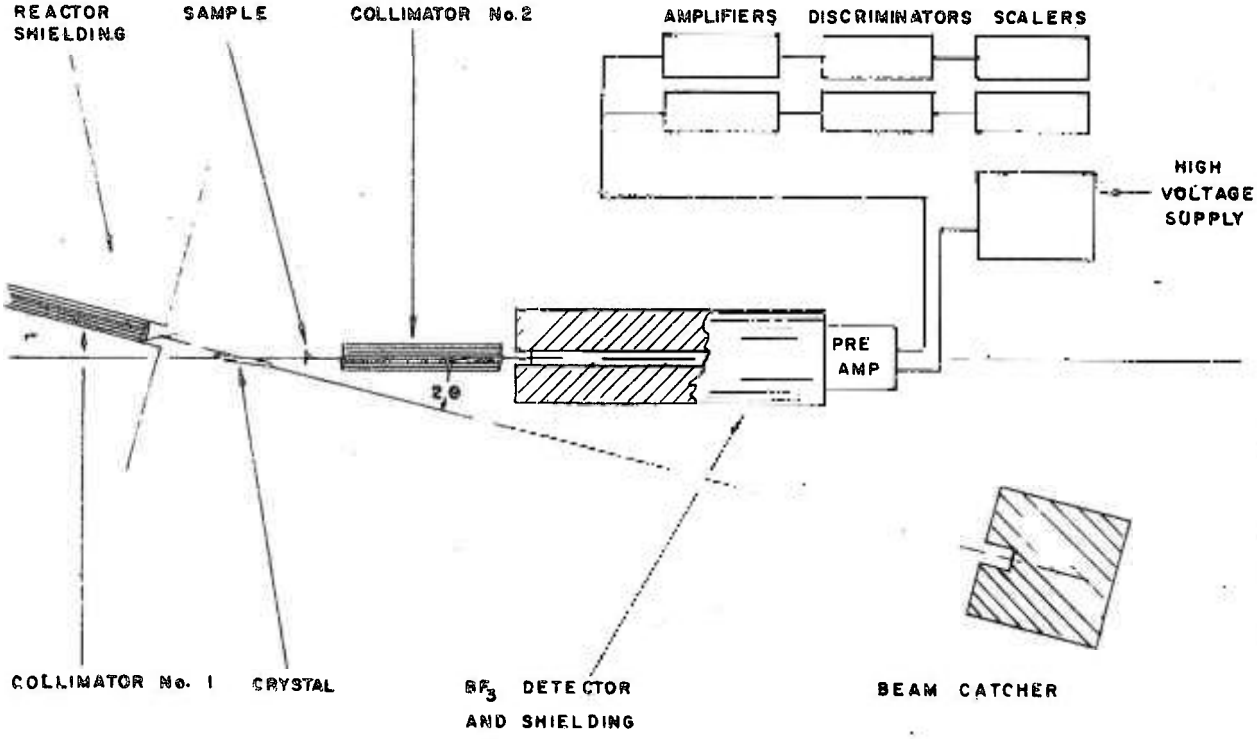

Figure 1

Schematic diagram of the spectrometer and electronics

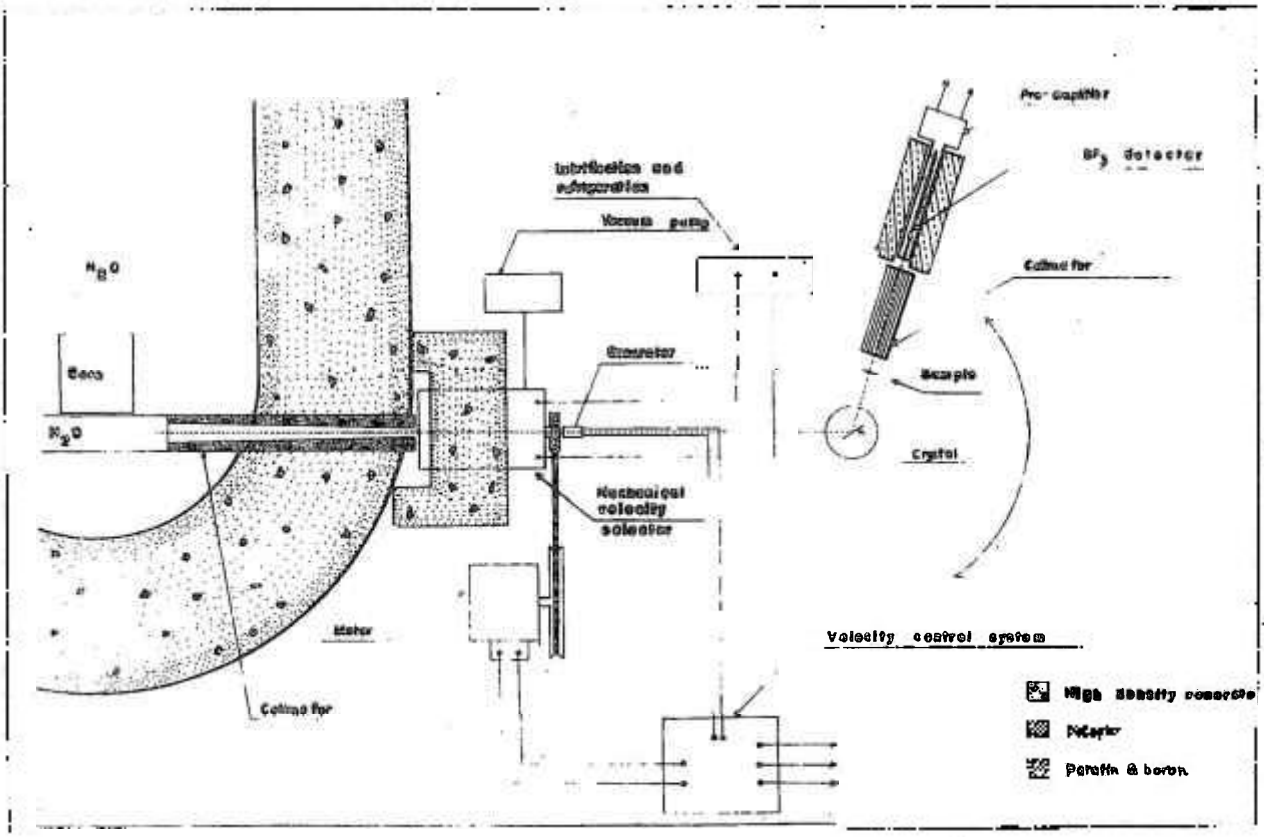

Figure 2

Schematic diagram of the experimental apparatus mounted in the beam hole through port.

Energy (ev)	σ_{Lu} (barns)
0.203	79 ± 1
0.185	121 ± 3
0.172	187 ± 2
0.159	266 ± 2
0.149	305 ± 1
0.139	331 ± 2
0.136	342 ± 1
0.131	329 ± 2
0.127	310 ± 1
0.118	268 ± 1
0.112	222 ± 1
0.0994	168 ± 1
0.0888	136 ± 1
0.0724	113.6 ± 0.9
0.0595	102.9 ± 0.7
0.0470	102.5 ± 0.4
0.0390	108 ± 1
0.0378	114 ± 2
0.0328	110 ± 2
0.0290	118 ± 2
0.0259	118 ± 4
0.0244	116 ± 2
0.0232	128 ± 3
0.0210	126 ± 3
0.0185	131 ± 2

Figure 3

Total neutron cross section of Lutetium

Energy (ev)	σ_{Yb} (barns)
0.240	39 ± 1
0.203	37 ± 1
0.172	39 ± 2
0.127	44 ± 2
0.112	45 ± 2
0.0994	44 ± 1
0.0888	46 ± 1
0.0724	49 ± 1
0.0595	54 ± 1
0.0470	54 ± 1
0.0328	59 ± 1
0.0280	61 ± 1
0.0230	65 ± 1
0.0188	69 ± 1
0.0135	77 ± 2
0.0100	88 ± 2
0.00785	98 ± 2
0.0060	105 ± 2
0.0045	115 ± 2
0.0035	134 ± 2
0.0025	156 ± 3
0.00175	189 ± 4

Figure 4

Total neutron cross section of Ytterbium

Energy (ev)	σ_{Pr} (barns)
0.97	5.3 ± 0.8
0.82	5.0 ± 0.8
0.65	4.8 ± 0.7
0.524	5.1 ± 0.8
0.456	6.5 ± 0.7
0.385	6.2 ± 0.6
0.310	6.7 ± 0.7
0.290	7.3 ± 0.4
0.203	8.1 ± 0.2
0.172	7.9 ± 0.4
0.149	8.5 ± 0.5
0.127	8.8 ± 0.2
0.112	9.2 ± 0.2
0.0888	10.0 ± 0.2
0.0724	10.8 ± 0.2
0.0595	12.1 ± 0.2
0.0470	13.1 ± 0.1
0.0434	12.8 ± 0.3
0.0390	13.2 ± 0.3
0.0330	14.8 ± 0.3
0.0290	16.2 ± 0.3
0.0240	18.1 ± 0.4
0.0186	20.0 ± 0.4
0.0150	20.8 ± 0.4
0.0135	22.6 ± 0.5
0.0099	25.9 ± 0.5
0.0078	30.8 ± 0.6
0.0071	30.4 ± 0.6
0.0060	34.5 ± 0.7
0.0050	34.6 ± 0.7
0.0041	37.5 ± 0.8
0.00255	46.9 ± 0.9
0.00175	61 ± 1

Figure 5

Total neutron cross section of Praseodymium

Energy (ev)	σ_{Er} (barns)
0.295	223 ± 4
0.244	167 ± 3
0.205	144 ± 3
0.175	134 ± 3
0.151	130 ± 3
0.132	127 ± 3
0.116	130 ± 3
0.103	130 ± 3
0.092	131 ± 3
0.082	130 ± 3
0.074	134 ± 3
0.048	150 ± 3
0.0335	170 ± 3
0.0248	192 ± 4
0.0194	208 ± 4
0.0145	236 ± 5
0.0127	250 ± 5
0.0100	273 ± 5
0.0095	277 ± 5
0.00856	295 ± 6
0.00776	307 ± 6
0.00647	340 ± 7
0.00549	360 ± 7
0.00275	505 ± 10
0.00230	553 ± 11
0.00205	593 ± 12
0.00180	634 ± 13
0.00150	688 ± 14
0.00125	751 ± 15
0.00104	840 ± 17
0.0085	958 ± 19
0.00067	1077 ± 22

Figure 6

Total neutron cross section of Erbium

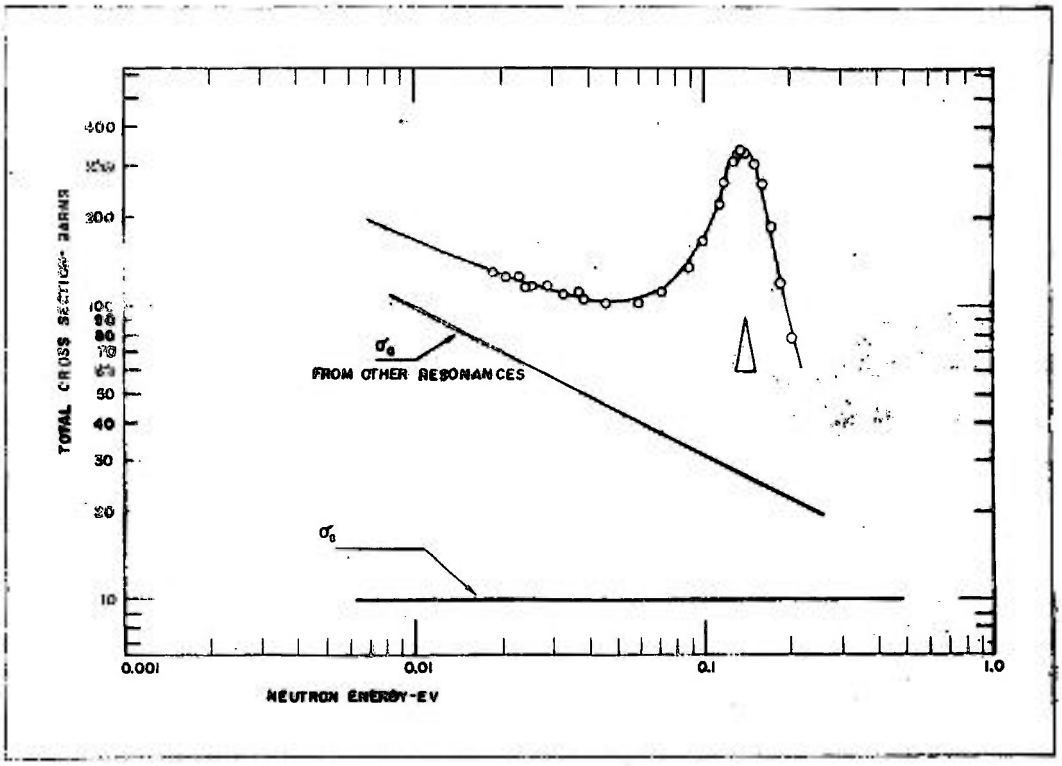

Figure 7

Total neutron cross section of Lutetium. The solid curve, adjusted for the experimental points, is already affected by the resolution, represented schematically by the triangle.

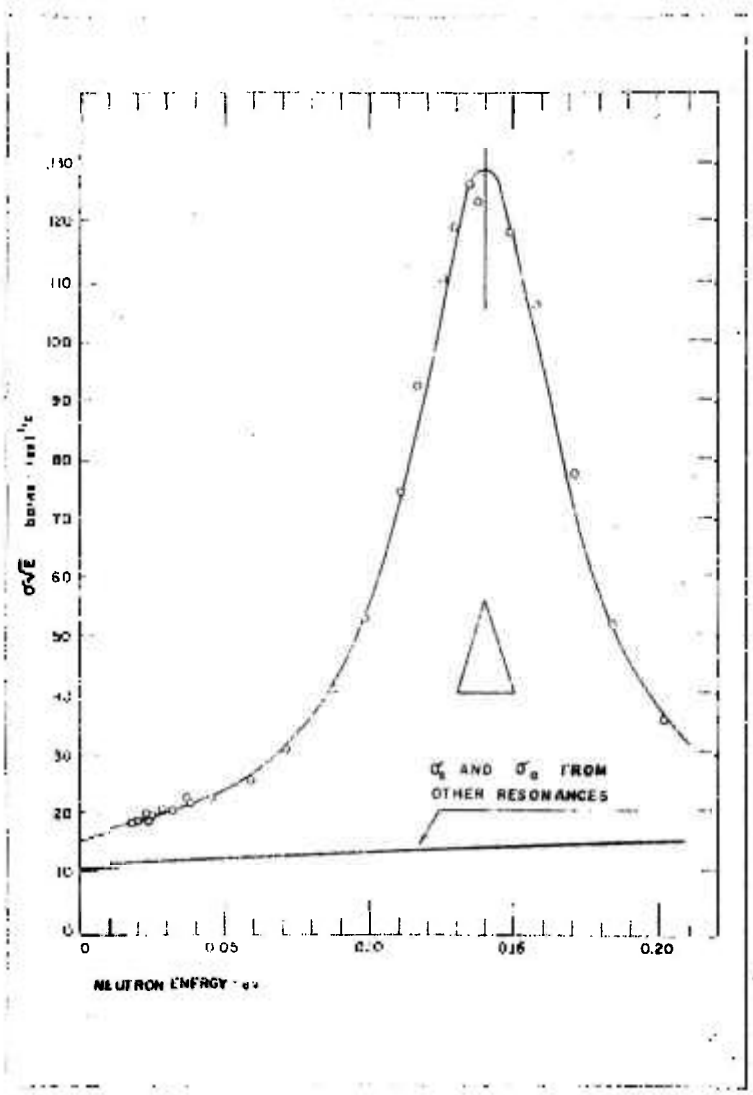

Figure 8

Experimental results of the total neutron cross section measurements of Lutetium plotted as $\sigma\sqrt{E}$ versus E. The solid curve, adjusted for the experimental points, is already affected by the triangle. The curve symmetry, except for $\sigma_s\sqrt{E}$, indicates that the term due to interference is negligible.

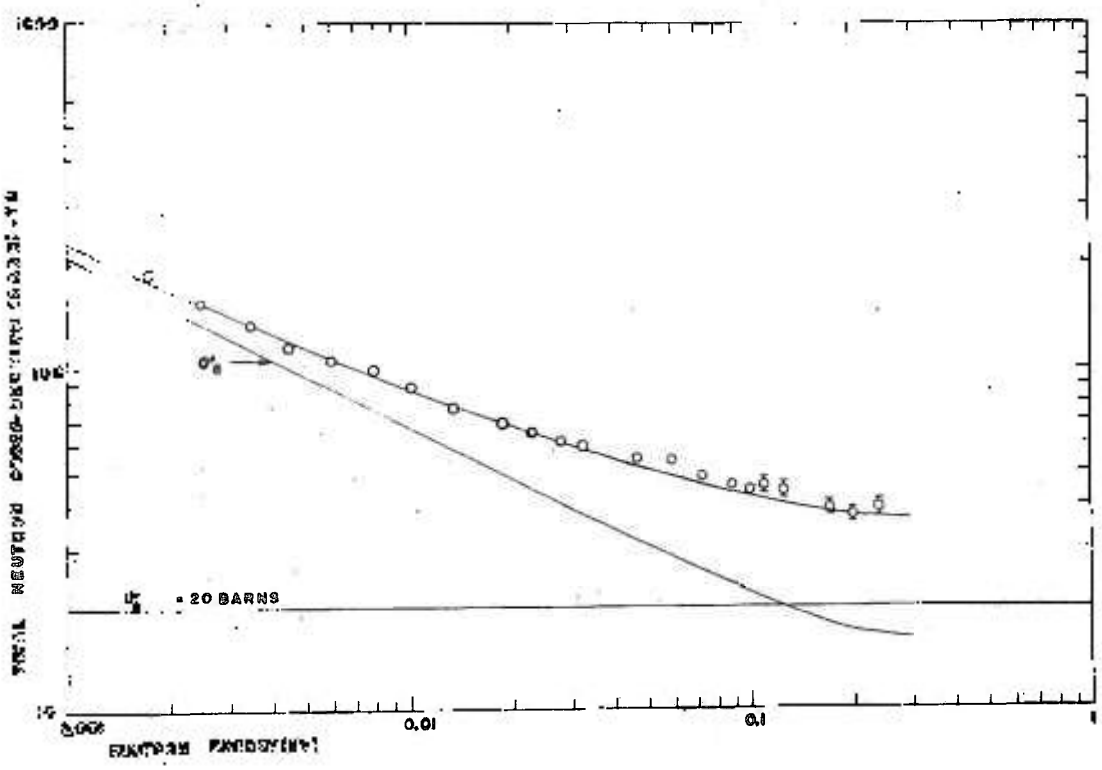

Figure 9

Total neutron cross section of Ytterbium. The solid curve has been adjusted to the experimental points.

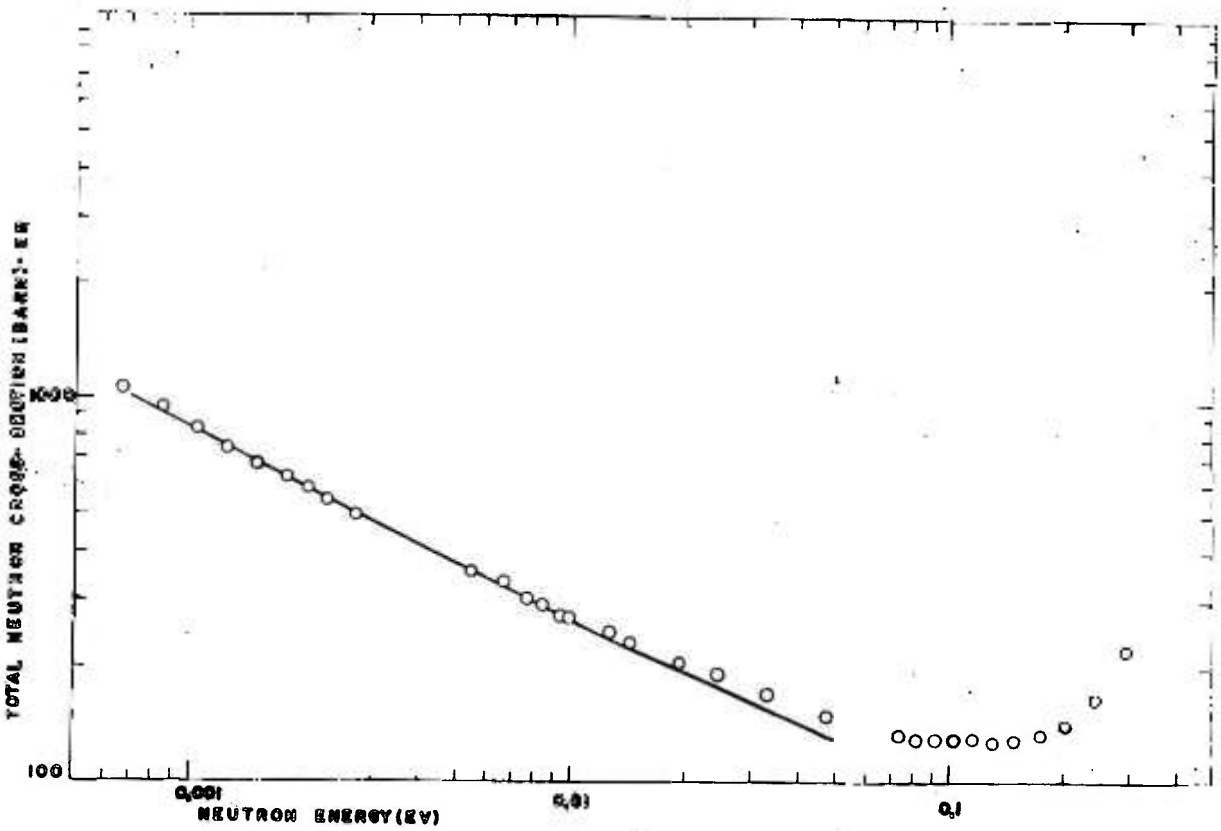

Figure 10

Total neutron cross section of Erbium. The solid curve has been adjusted for lower energies, when the absorption term is proportional to $\frac{1}{\sqrt{E}}$.

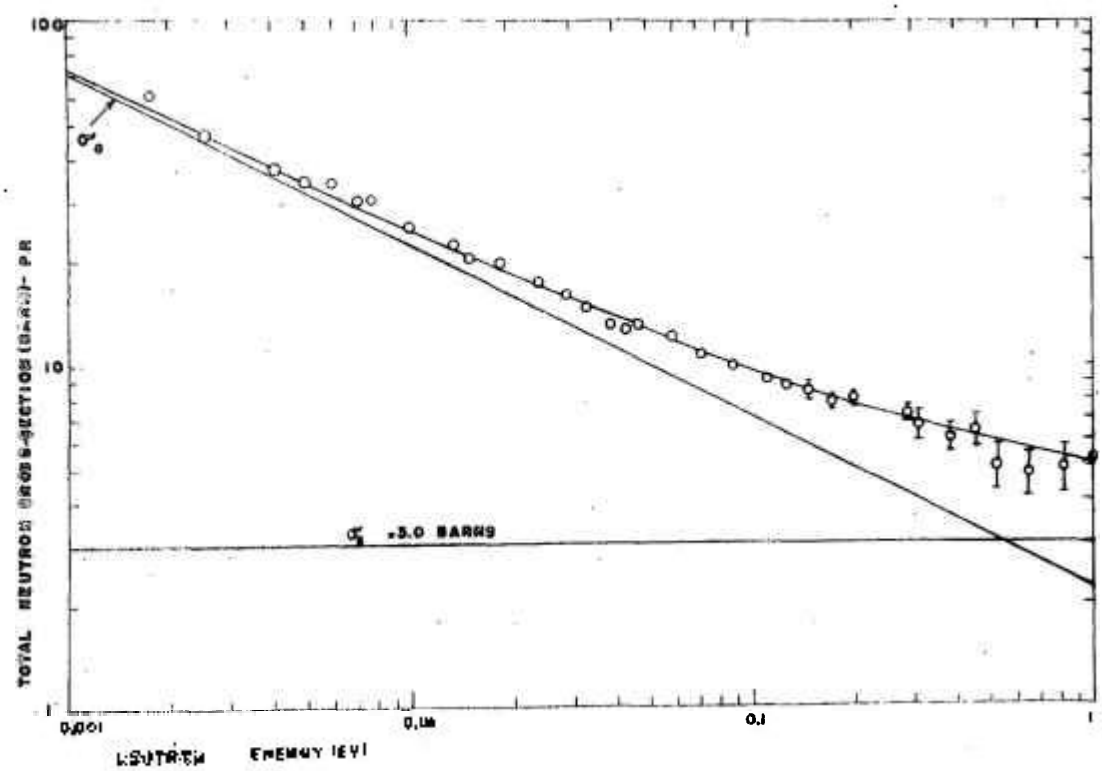

Figure 11

Total neutron cross section of Praseodymium. The solid curve has been adjusted to the experimental points.

	Results obtained by us			Published values [2,12,13]	
	σ_{total}	σ_a	σ_s	σ_a	σ_s
Pr	17.0 ± 0.3	14 ± 1	3 ± 1	11.2 ± 0.6	4 ± 1
Er	190 ± 4	180 ± 5	10 ± 5	166 ± 17	15 ± 4
Yb	63 ± 1	43 ± 3	20 ± 3	36 ± 4	12 ± 5
Lu	118 ± 1	108 ± 5	10 ± 5	108 ± 5	-

Figure 12

Neutron cross section at the thermal energy 0.025 ev, in barns.