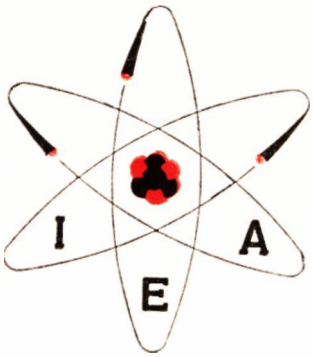

PARAMAGNETIC STUDIES OF HOLMIUM BY NEUTRON TOTAL CROSS SECTION MEASUREMENTS

by

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RESUMO

Foi medida a secção de choque total do hôlmio, para neutrons de comprimentos de onda entre 0,20 e 9,00 Angstroms . 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 em pó foram preparadas em alto grau de pureza, tomando-se cuidado especial em eliminar contaminação por terras raras de alta secção de choque, para neutrons. Ainda que os fatores de forma paramagnética sejam usualmente determinados por medidas de secção de choque diferencial para neutrons de um único comprimento de onda, medidas precisas da secção de choque total num grande intervalo de comprimentos de onda conhecidos dão informações definidas sôbre o raio da órbita eletrônica 4f. Nossas medidas indicam um raio diferente daquele usado por Blume, Freeman e Watson, que calcularam o fator de forma assumindo funções de onda hidrogênicas. Será discutida a possível escolha de diferentes constantes de screening.

RESUME

La section efficace totale de l'holmium a été mesurée pour les neutrons de longueur d'onde entre 0,20 et 9,00 Angstroms. 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 échantillons de Ho_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. Bien que les facteurs de forme soient ordinairement déterminés par des mesures de section efficace différentielle à une seule longueur d'onde du neutron, des mesures précises de la section efficace totale dans un large intervalle de longueurs d'onde connues donnent des informations définies sur le rayon de l'orbite electronique 4f. Nos mesures indiquent un rayon différent de celui employé par Blume , Freeman et Watson, qui ont calculé le facteur de forme en assumant des fonctions d'onde hydrogéniques. Il sera discuté le choix possible de différentes constantes de screening.

ABSTRACT

The total neutron cross section for holmium was measured for neutron wavelengths between 0.20 and 9.00 Angstroms. 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 were prepared in high purity, taking particular care to eliminate contamination by the rare earths of high neutron cross section. Although paramagnetic form factors are usually determined by differential cross section measurements with one neutron wavelength, accurate total cross section measurements for a wide range of known wavelengths give definite information on the radius of the 4f electron orbits . Our measurements indicate a radius different from the one used by Blume, Freeman and Watson, who calculated the form factor assuming hydrogenic wave functions. The possible choise of different screening constant will be discussed.

I. INTRODUCTION

During a program of total cross section measurements of the rare earth elements at this Institute, holmium was given special attention because of the interesting interaction between the neutrons and the atomic electrons. This paramagnetic scattering has been noticed before in total neutron cross sections by Bernstein et al.[1] and in neutron diffraction work by Koehler, Wollan and Wilkinson[2].

The total neutron cross section for scattering of neutrons by an atom in the presence only of an interaction between the magnetic moment of the neutron, γ, and that of the atom, μ, is

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

where $\frac{e^2}{mc^2}$ is the classical electron radius, and f is the neutron scattering form factor for those unpaired electrons which contribute to the magnetic moment of the holmium ion.

The magnetic moment of the holmium ion has been measured as being 10.34 ± .10 Bohr magnetons by Strandburg, Legvold and Spedding[3]. The paramagnetic scattering form factor has been calculated by Blume, Freeman and Watson[4], using Hartree - Fock wave functions for isolated ions. These form factors, together with known values of the neutron and ionic magnetic moments, may be used to predict the paramagnetic scattering component to the neutron scattering.

However, it is essential to make accurate measurements of the neutron cross section to supply information unavailable from the theory. In particular, the ionic form factor for the ion in the solid state may be expected to be different from the one for

a free ion. In addition, measurements over a wide range of neutron energies are required to determine the variations of the nuclear cross sections with neutron energy.

Ordinarily, scattering form factors are best determined by differential scattering experiments. Holmium has been meàsured in this way by Koehler, Wollan and Wilkinson[2]. In the present work, it is shown that careful total cross section measurements, besides giving the nuclear cross sections, give information about the 4f electron orbits.

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 in the shops of the Instituto de Energia Atômica , 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. We had a thermal flux of 4×10^6 neutrons/cm^2/sec. outside the radial port and of 1.4×10^8 neutrons/cm^2/sec. outside the tangential port. The crystal spectrometer angles were read on a vernier scale with a precision of 0.01 degree. The measurements were made with crystals of calcite, aluminium and mica. With the calcite

crystal the resolution was 18 minutes.

The mechanical velocity selector[5] gives a neutron energy with a resolution which depends on the rotation velocity, and on the helical channel inclination. We used a resolution of 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.

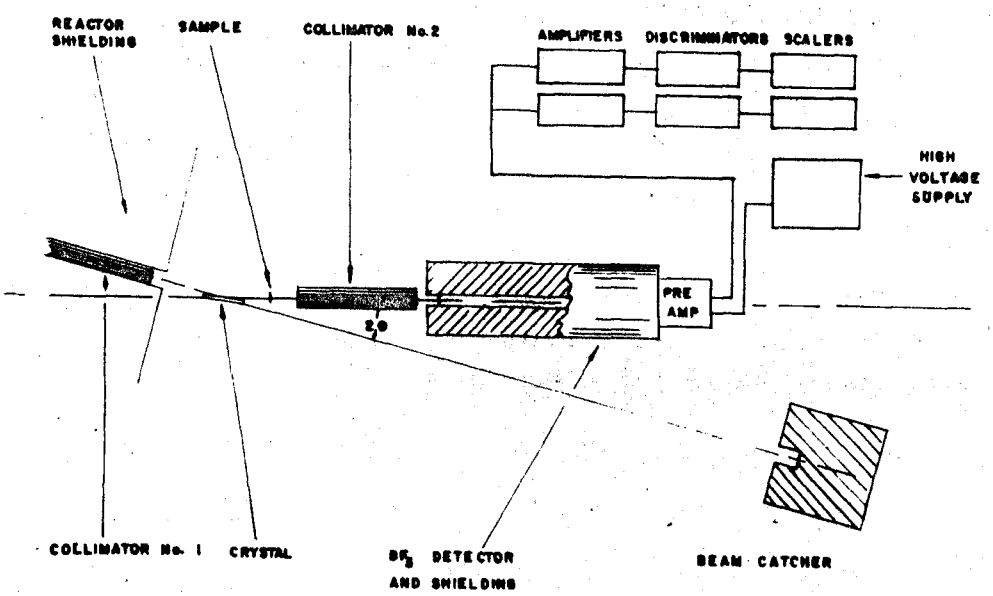

Figure 1 - Schematic diagram of the crystal spectrometer

The samples of holmium oxide were supplied by the Chemical Engineering Division of this Institute. The separation method employed, using ion exchange resins, assured us of the degree

of purification necessary for this experiment.

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. A background was subtracted from each counting. The containers were designed to give a transmission which minimized the time required to reduce the statistical errors[6].

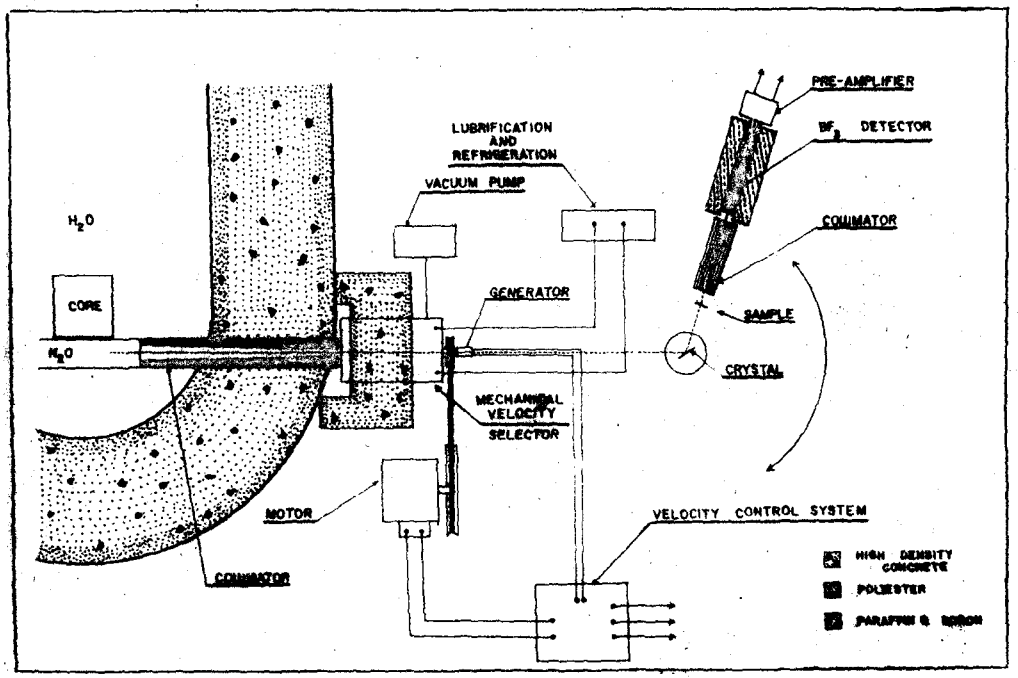

Figure 2 - Schematic diagram of the mechanical velocity selector together with the crystal spectrometer.

To avoid the influence on the transmission of the fluctuations of the reactor power and the instability of the

electronic circuits, 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 has been used for the error. The calculations were made by an 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. Figure 3 shows the results in the usual way with total cross section versus neutron energy.

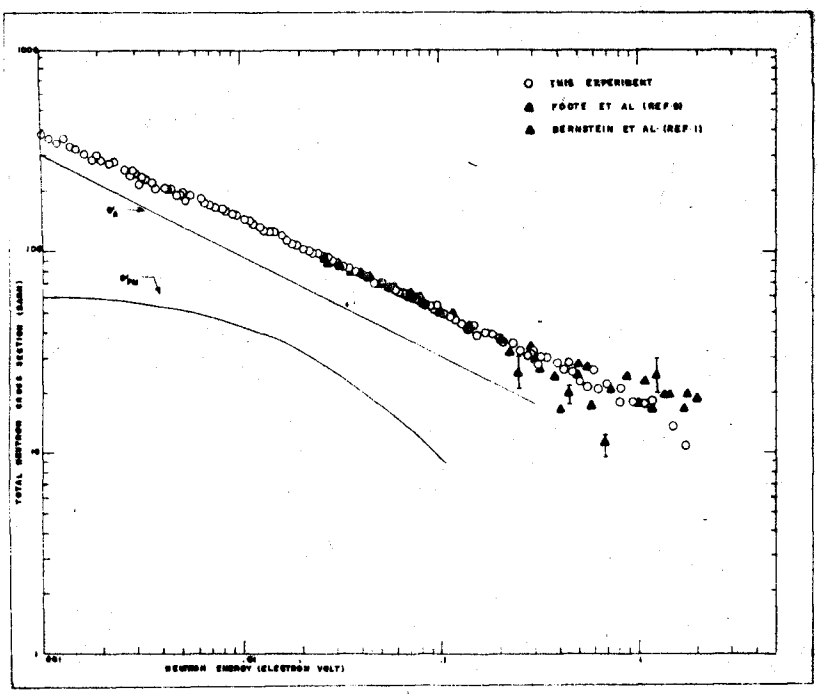

Figure 3 - Total neutron cross section of holmium as a function of neutron energy. The absorption cross section, σ_a, wich is dependent on $1/\sqrt{E}$, and the calculated paramagnetic cross section, σ_{pm}, are shown.

Figure 4, with total cross section *versus* neutron wavelength, shows the same data, together with the paramagnetic scattering calculated from the form factors of Blume, Freeman and Watson[4].

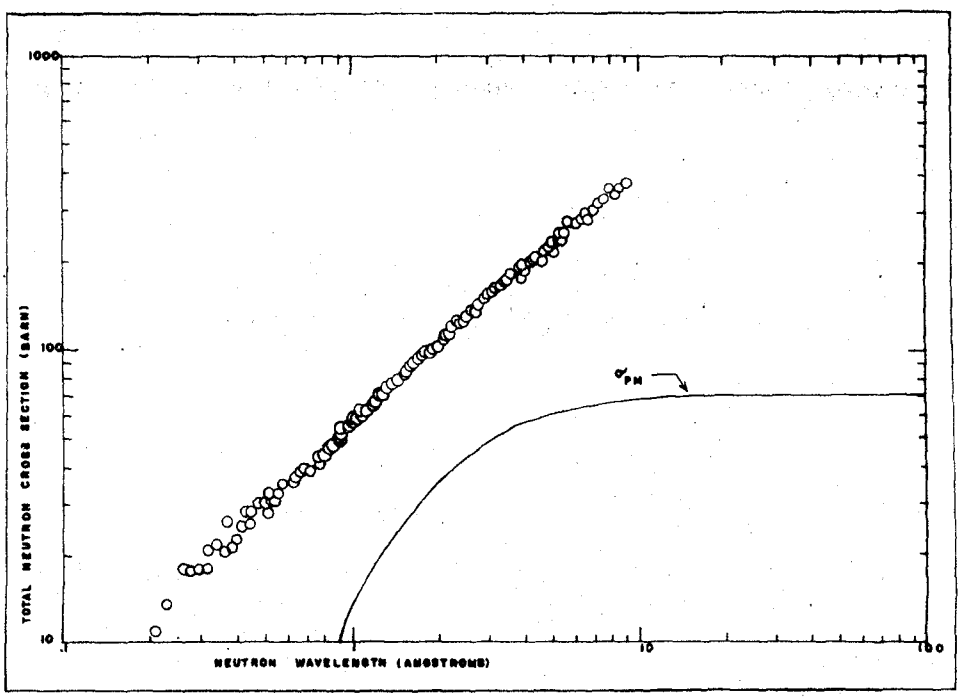

Figure 4 - Total neutron cross section of holmium as a function of neutron wavelength. The calculated paramagnetic cross section, σ_{pm}, is shown.

III. ANALYSIS OF DATA

The total cross section of holmium, σ_T, consists of three completely independent partial cross sections, one of which is the paramagnetic scattering, σ_{pm}. Then,

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

where σ_s and σ_a are the nuclear scattering and absorption, respectively. σ_s was assumed to be independent of energy and σ_a was assumed to vary as $1/\sqrt{E}$ over the range of this experiment. The nuclear resonance spacing is about 6 electron volts[7]. The first resonance at 3.92 electron volts has no effect on these assumptions. It is unlikely that a bound state would influence the dependence on energy of the nuclear cross sections.

Nuclear scattering contributes least to our experimental data. For the purpose of analysis, σ_s was taken as 7 barns by comparing with neighbouring nuclei[7]. Nuclear absorption dominates at very low energies and may be quite well determined by using the asymptotic value of $\sigma_{pm} = 65.2 \pm 1.3$ barns, calculated from the holmium ion magnetic moment.

The nuclear absorption cross section thus determined from our data is $\sigma_a = 61 \pm 3$ barns, reduced to its value at thermal neutron energy .025 electron volts.

IV. CONCLUSIONS

Figure 5 shows our experimental points after having subtracted the contributions of nuclear scattering and absorption.

Comparing our experimentally determined paramagnetic scattering cross section with that expected from the Hartree--Fock calculations of Blume, Freeman and Watson[4] one sees that the disagreement is not serious. A 4f shell radius smaller by 10%, or a screening constant 5% less, would minimize the discrepancy between their theory and our results. However, an agreement within experimental errors cannot be obtained only by changing the radius, but rather by admitting the possibility of

a slightly more diffuse wave function, such as proposed by Judd and Lindgren[8]. Unfortunately, no paramagnetic neutron scattering form factors are available using their wave functions.

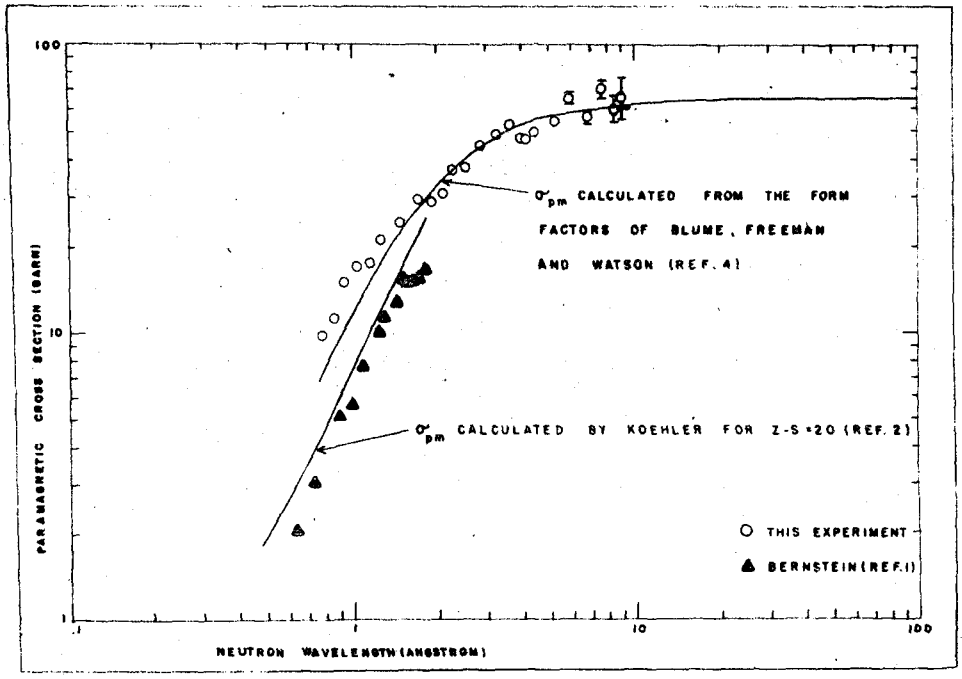

Figure 5 - Comparison between the experimental paramagnetic cross section, obtained after having subtracted the contribution of nuclear scattering and absorption, and the theoretical paramagnetic cross section, calculated from the form factors of Blume, Freeman and Watson[4]. The older theory quoted by Koehler et al[2] is shown. The apparently discrepant points due to Bernstein et al[1] are explained in the text.

The determined values of σ_{pm} of Bernstein et al.[1] 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 differs

from our values which have taken advantage of more recent 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 .05 eV. It now appears from the compilation of all total cross section values that 28 barns is at least 3 barns too high, as it can be seen in Figure 3. In addition, they neglected the effects of the then unknown 3.92 eV resonance on the capture cross section at .05 eV (about 1 barn), and they underestimated (by about 1 barn) the paramagnetic scattering cross section at .05 eV. Taken together, their estimate of 13 barns for nuclear scattering is about 5 barns too high. In our lower energy range, the nuclear scattering is relatively unimportant; in the absence of direct measurements, we used 7 barns based on measured values of nuclear radii of neighbouring nuclei[10]. 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 also shows the disagreement between the older calculation quoted in the experimental work of Koehler et al.[2], who used the theory by Trammell[11] with hydrogen-like wave functions, and the calculation of Blume, Freeman and Watson[4] who used the same theory with Hartree-Fock wave functions. Blume, Freeman and Watson noted 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.

We plan to continue our work with metallic samples to eliminate small uncertainties in subtracting the scattering by oxigen. In addition, it is planned to make an independent measurement of the nuclear absorption cross section with

facilities existing in this Institute.

V. ACKNOWLEDGMENTS

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