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Recent Measurements of the Scattering Laws of Some Heterogeneous  
Moderators

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Recent measurements of the Scattering Laws of Some Hydrogeneous  
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## I. Introduction

The study of the scattering of low energy neutrons by many-particle systems yields valuable information on the dynamical behavior of such systems. The most detailed quantity that can be measured experimentally is the double differential scattering cross section  $\frac{d^2\sigma}{d\Omega dE}$  which consists in the Fermi Pseudopotential approximation of two factors. One depends only on the properties of the neutron and the other, the so called scattering law, depends only on the dynamical properties of the scatterer.

Often the scatterer is isotropic with respect to the incident neutron beam (polycrystalline solids, liquids and gases), in this case the scattering law can be written:

$$S(\alpha, \beta) = \frac{4 k_B T}{\sigma_b} \left(\frac{E_0}{E}\right)^{1/2} e^{\beta/2} \frac{d^2\sigma}{d\Omega dE} \quad (1)$$

where  $E_0$  is the incident,  $E$  the scattered energy,  $\sigma_b$  the bound atom cross section of the scattering atom,  $k_B T$  is the temperature in energy units,  $d\Omega$  is the element of solid angle into which the neutrons are scattered.

$\alpha$  and  $\beta$  have the following meaning

$$\alpha = \frac{\sqrt{E + E_0 - \sqrt{EE_0} \cos \theta}}{A k_B T} \quad (2)$$

$A$  = ratio of the mass of the scattering nucleus to the mass of the neutron

$$\beta = \frac{E - E_0}{k_B T} \quad (3)$$

Measurement of  $S(\alpha, \beta)$  of moderator and reflector materials for all possible  $\alpha$  and  $\beta$  values is desirable for a better description of the neutron thermalization process in reactors. The fine details of  $S(\alpha, \beta)$  are not so important for thermalization studies, however, accurate measurements of  $S(\alpha, \beta)$  also yield valuable information of more fundamental interest. Although the main objective of the present work was of the first kind we have tried to extract from the measured scattering laws physically interesting features.

In section II of this paper a short description of the experimental arrangement and of the data processing is given, in section III the results on water vapour, benzene and zirconium hydride are described.

## II. Experimental Procedure and Methods of Data Processing

### 1. Apparatus

The Karlsruhe rotating-crystal time-of-flight spectrometer described elsewhere [1] was used to produce the monoenergetic pulsed beams needed for the measurements to be described. The incident neutron energy was between 18 and 80 meV. The pulse width at half height of the incident beam at the scattering sample was 20  $\mu$ sec, time-of-flight resolution 20  $\mu$ sec/m, and primary energy resolution about 5 % at 18 meV.

Nine detectors at a distance of 2 meters from the sample arranged between 20 and 140° detected simultaneously the scattered neutrons. During the experiment the output of the detectors was handled by MIDAS (a multiple input data acquisition system using a Control Data 160-A computer) and written on magnetic tape [2].

For the benzene measurements an arrangement similar to that described, installed at the reactor DIORIT, having only six detectors and a conventional multichannel analyzer was used.

### 2. Scattering Samples

To get a reasonable amount of scattering from a small volume of H<sub>2</sub>O vapour a temperature of 241°C and a pressure of 25 atmospheres were chosen. The container was a cylinder, 5 cms in diameter with an active scattering zone 5 cm in length, made of aluminium. The wall thickness of the container was 0.18 cm. Under the given conditions the density of vapour was 12 mg/cm<sup>3</sup> and the transmission about 90 % for an incident energy of 18.2 meV. For the surface reaction of the aluminium with the water vapour producing Al(OH)<sub>3</sub> a correction of the order of 2 % has been made for the number of scattering molecules.

In the case of benzene samples of 5 x 12 cm<sup>2</sup> size and 0.06 cm thickness were used. The containers used 0.05 cm thick aluminium

windows.

In the early zirconium hydride measurements powder of  $ZrH_{1.8}$  was used. Although a few runs at room temperature were made most were performed at  $210^{\circ}C$ . In the later experiments the sample was a hydrogenated plate of zirconium metal 0.05 cm thick  $4.5 \times 7 \text{ cm}^2$  in size with the composition  $ZrH_{1.1}$  which was easier to handle in heating.

For all the investigated substances the normal procedure was to make a run of several days for each incident energy and to do a similar background run where the sample was omitted. The sample-out run delivered all the data necessary for background corrections.

### 3. Data Processing

The data processing cycle starts with totalizing the information written on magnetic tape, using the CDC-160 A computer. Further processing was on an IBM-7070 where after determining of detector sensitivity the "scattering-law program" was used to evaluate  $\alpha$ ,  $\beta$ ,  $S(\alpha, \beta)$  and  $S(\alpha, \beta)/\alpha$  as well as the statistical errors due to the number of collected counts. In this program also smoothing of the raw data can be done if the conditions make this advisable. Smoothing has been done mainly for background runs and the measurements at lowest energy.

In these calculations no general corrections for energy resolution and multiple scattering are done. However, in the cases where the extrapolation method to be described now is used a resolution correction for the elastic peak has been carried out.

To get the generalized frequency distribution  $p(\beta)$  the  $S/\alpha$  values were plotted against  $\alpha$  on a semi-logarithmic scale and extrapolated to  $\alpha = 0$ . With the  $p(\beta)$  (eq. 4) guessed in this way a LEAP [3]

$$p(\beta) = \beta^2 \lim_{\alpha \rightarrow 0} S/\alpha \quad (4)$$

calculation was done to determine  $S/\alpha$  again on the base of a harmonic approximation for the motion of the scattering nuclei. In the range covered by the experiments essentially a phonon ex-

pansion is used, varied in the case of non-solids by folding with a simple diffusion model.

### III. Experimental Results

#### 1. Water Vapour

Runs with incident energies of 0.018 and 0.032 eV were made. The evaluated energy and momentum transfer ranges were  $0 < \beta < 5$  and  $0 < \alpha < 5$  ( $T = 514^\circ\text{K}$ ). The density of the vapour ( $12 \text{ mg/cm}^3$ ) should be small enough to suppress significant interaction between molecules.

A typical scattering curve is shown in Fig. 1. In this diagram the counting rates in 5 adjacent channels are smoothed and the background is subtracted. This smoothing process corresponds to the experimental resolution and therefore should not introduce serious errors. There is a slight structure in the curve. At the points indicated by the arrows are bands of energy transfer known from rotation spectra of  $\text{H}_2\text{O}$ . From the experimental data scattering-law values  $S(\alpha, \beta)$  have been derived. The values for one scattering angle obtained at different incident energies are plotted in Fig. 2. The lines are calculated with Krieger-Nelkin theory [4] using an effective mass of  $\bar{m}_r = 1.9$  determined from the Sachs-Teller mass tensor of the  $\text{H}_2\text{O}$  molecule. The assumptions of this theory, i.e. that the incident energy is large compared with the rotational level spacing and small compared with the vibrational levels of the molecule, are satisfied by the experiment. However, only for great momentum transfer values the theory agrees with experiment. The overall agreement can be improved by using an effective mass of the order of 4 to 5 which also has been observed by Hughes et al. [5] with cold neutrons. A good description of the experimental results was obtained also by approximating the  $\text{H}_2\text{O}$  molecule as a symmetric top with an effective moment of inertia  $I = 1.96 \cdot 10^{-40} \text{ gcm}^2$  which is the average of the principal moments of inertia of the  $\text{H}_2\text{O}$  molecule. The squares in Fig. 2 were calculated in this way considering only incoherent scattering from the protons. There have been attempts to treat the rotation of asymmetric tops in neutron scattering theory by quantum mechanics [6]. Unfortunately no numerical results for the double differential scattering cross section of  $\text{H}_2\text{O}$  molecules are

available so far from these calculations.

A set of  $S(\alpha, \beta)$  curves for constant  $\beta$  as parameter is shown in Fig. 3.

## 2. Benzene

With the benzene samples described in section II. measurements with the incident energies 0.025, .032 and 0.059 eV have been made. The range of  $\alpha$  and  $\beta$  values covered was:  $0 < \alpha < 12$ ,  $0 < \beta < 3$  ( $T = 293^\circ K$ ). A few examples of  $S(\alpha, \beta)$  values extracted from the measurements are plotted in Fig. 4. The shown solid lines are results from LEAP calculations. The effect of most of the high energy vibration not direct accessible in the present measurement is corrected for by an additional ADDELT calculation using the LEAP output and the known vibration frequencies. This contributes mainly to the Debye-Waller factor. The dashed lines are from a "Benzene gas" model described by Boffi et al. [7]. In the curves only two terms in the phonon expansion are taken in consideration. Typical extrapolation curves  $S/\alpha$  vs.  $\alpha$  for determining  $(S/\alpha)_{\alpha=0}$  are shown in Fig. 5 and the resultant generalized frequency distribution  $p(\beta)$  is given in Fig. 6. The indicated errors are the possible uncertainties in the extrapolation process. The  $p(\beta)$  is in good agreement with results of infrared and Raman measurements. The high energy part of  $p(\beta)$  ( $\beta > 1$ ) is determined by the properties of the benzene ring, the lowest normal vibration of which is predicted to be at  $\beta = 1.99$ . This vibration, however, is inactive in infrared and Raman measurements. The broad peak of the distribution near  $\beta = 2$  may be attributed to this vibration.

For lower energies the interaction between molecules contributes to the frequency spectrum. From Raman measurements in solid benzene [8] frequencies at about  $\beta = 0.32$  and  $\beta = 0.52$  are known. The broad peak centered at about  $\beta = 0.48$  can be attributed to this motion which from theoretical analysis are very probably torsional vibrations of the benzene molecule in the field of its neighbours. For studying these motions in more detail a higher resolution than that used in the present measurements is necessary.

### 3. Zirconium Hydride

Fig. 7 shows a typical scattering distribution obtained with the  $ZrH_{1.1}$  plate at  $210^{\circ}C$ . Although the high energy peak contributes the most important part to inelastic scattering, the main aim of the present work was to determine the contribution of the low energy acoustical modes. The high energy line has been carefully studied by Mc Reynolds et al. [10] using 4 A neutrons with higher accuracy than this work yields. Therefore we used the peak half width of these authors in our data evaluation. As is expected from theory the low energy modes take only a few percent of the frequency distribution. We obtained the best fit with the  $S/\alpha$  values using the distribution  $p(\beta)$  shown in Fig. 8.

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### References

- [1] Carvalho, F. et al.: Study Group Meeting on Research Reactor Experimental Techniques, Bucharest, October 1964.
- [2] Krüger, G. and Dimmler, G.: EANDC Conf. on Automatic Acquisition and Reduction of Nuclear Data, Karlsruhe, July 1964 p.49.
- [3] Egelstaff, P. A. and Schofield, P.: Nucl. Sci. Engng. 12 (1962) 260.
- [4] Krieger, T. J., Nelkin, M. S.: Phys. Rev. 106 (1957) 200.
- [5] Hughes, D. J., et al.: Phys. Rev. 119 (1960) 872.
- [6] Volkin, H. C.: Phys. Rev. 113 (1959) 866.
- [7] Boffi, V. C. et al.: "Inelastic Scattering of Neutrons in Solids and Liquids", Vol. I, IAEA, Vienna (1963) 285.
- [8] Fruhling, A.: Ann. de Phys. 6 (1951) 401.
- [9] Gläser, W. and Beckurts, K. H.: Nucl. Sci. Eng. 20 (1964) 235.
- [10] McReynolds, A. W. et al.: GA-471, Chapter 3 (1958).



Legends to figures:

- Fig. 1: Typical scattering distribution from  $\text{H}_2\text{O}$  vapour at  $241^\circ\text{C}$  and 25 atmospheres
- Fig. 2:  $S(\alpha, \beta)$  for the detector at  $43.9^\circ$  at two incident energies
- Fig. 3: A plot of  $S(\alpha, \beta)$  using  $\beta$  as a parameter.  $S(\alpha, \beta)$  is per molecule.
- Fig. 4: A few examples of  $S(\alpha, \beta)$  vs.  $\alpha$  for liquid benzene.
- Fig. 5: Examples of extrapolation of benzene  $S/\alpha$  values for  $\alpha = 0$ .
- Fig. 6: The frequency distribution  $p(\beta)$  derived from the measurements.
- Fig. 7: Typical scattering distribution obtained with  $\text{ZrH}_{1.1}$  at  $210^\circ\text{C}$ . Incident energy was 0.033 eV, scattering angle  $77.1^\circ$ .
- Fig. 8: Fitted low energy distribution  $p(\beta)$  for zirconium hydride.

FIG. 1

$E_0 = 18.2 \text{ meV}$

$\theta = 43.9^\circ$

COUNTS

300

200

100

70

90

110

130

150

170

190

210

230

CHANNEL

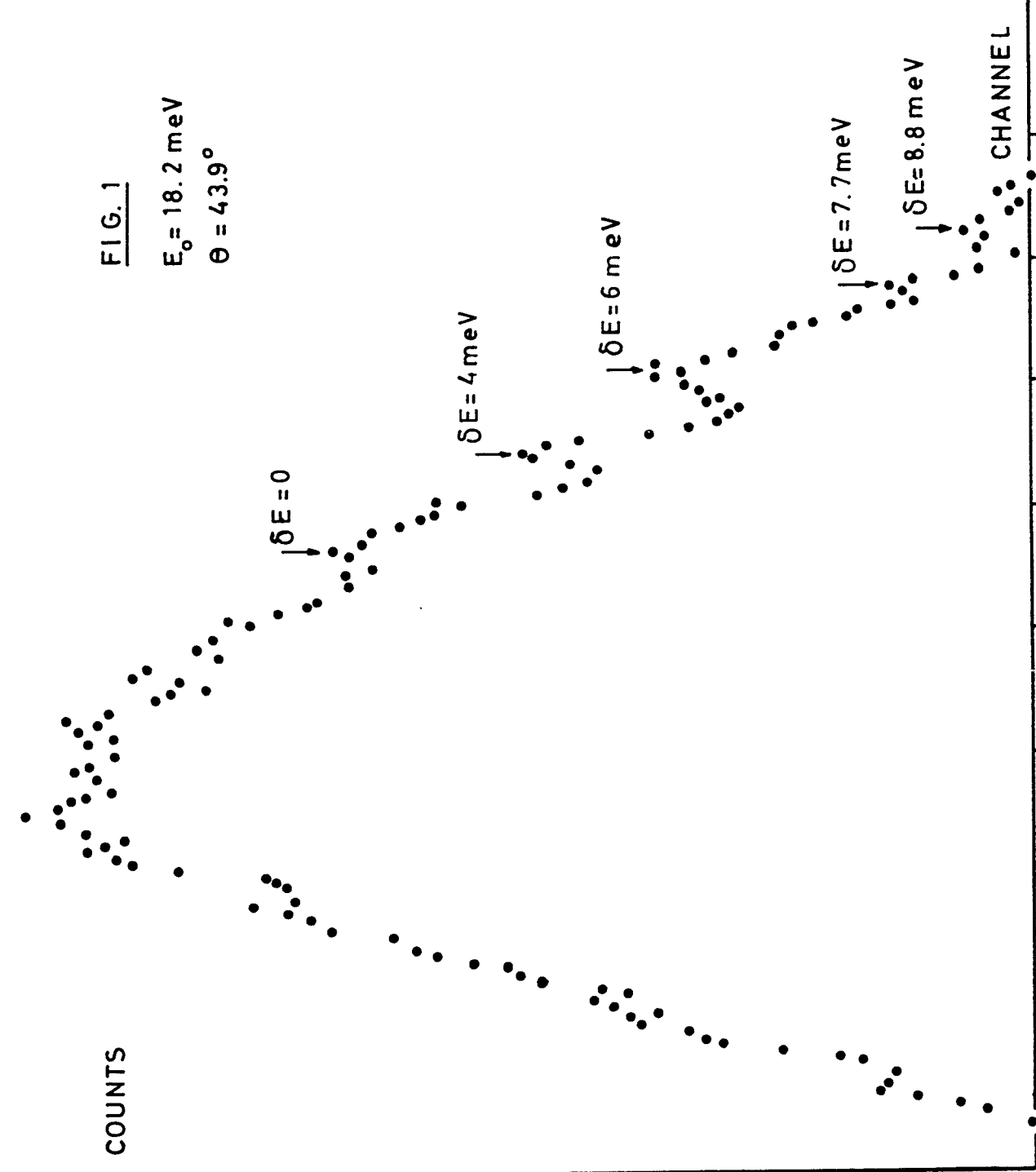
$\delta E = 0$

$\delta E = 4 \text{ meV}$

$\delta E = 6 \text{ meV}$

$\delta E = 7.7 \text{ meV}$

$\delta E = 8.8 \text{ meV}$



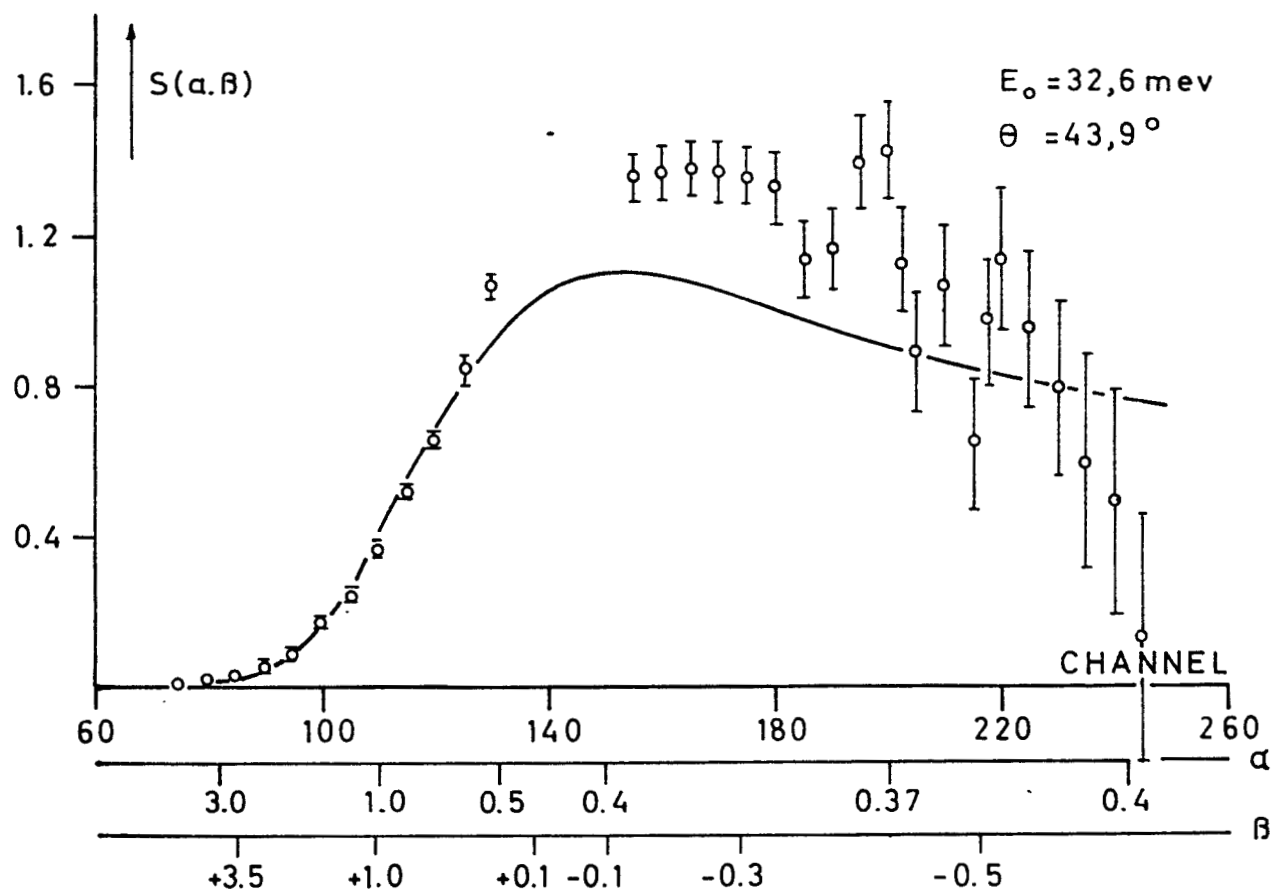
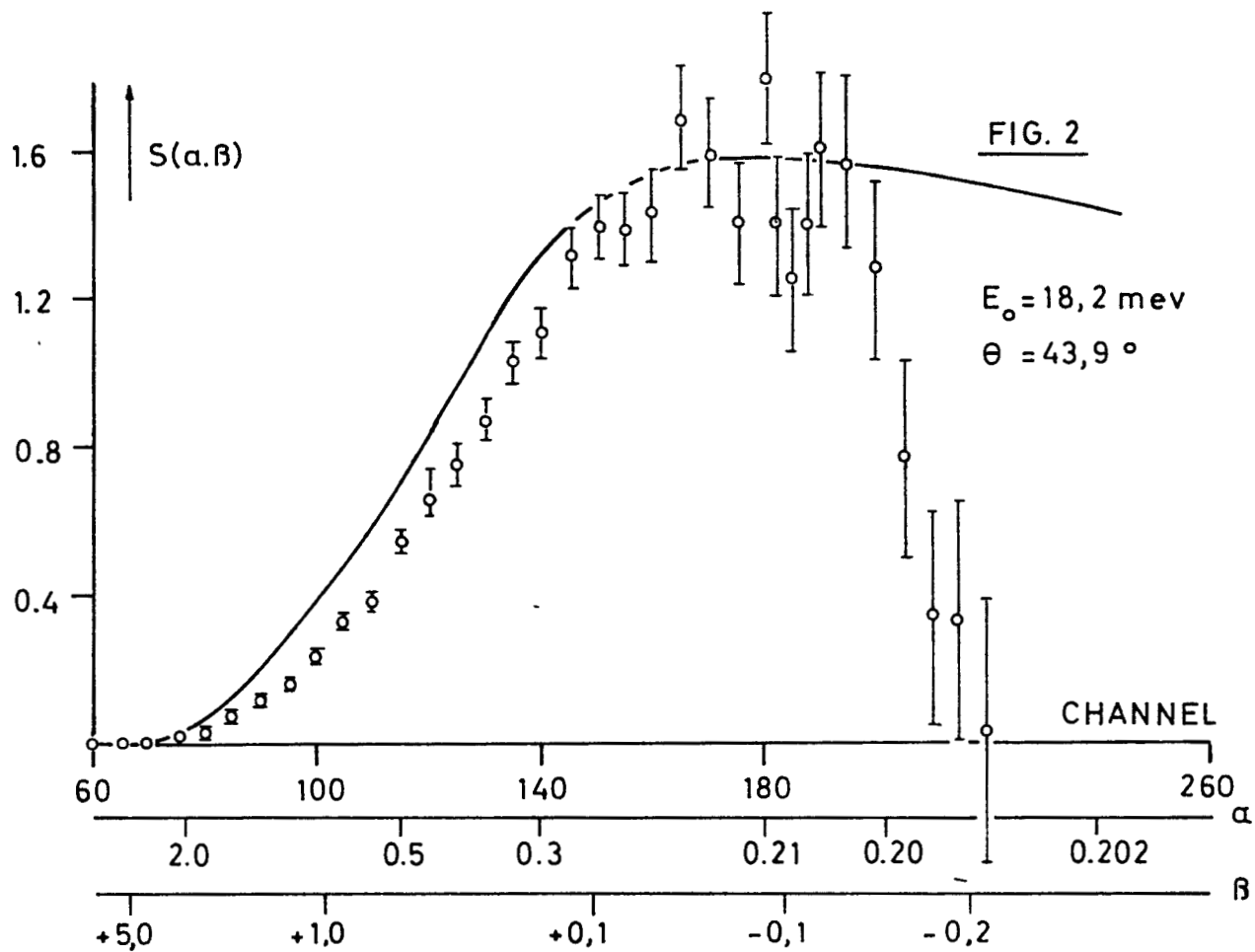
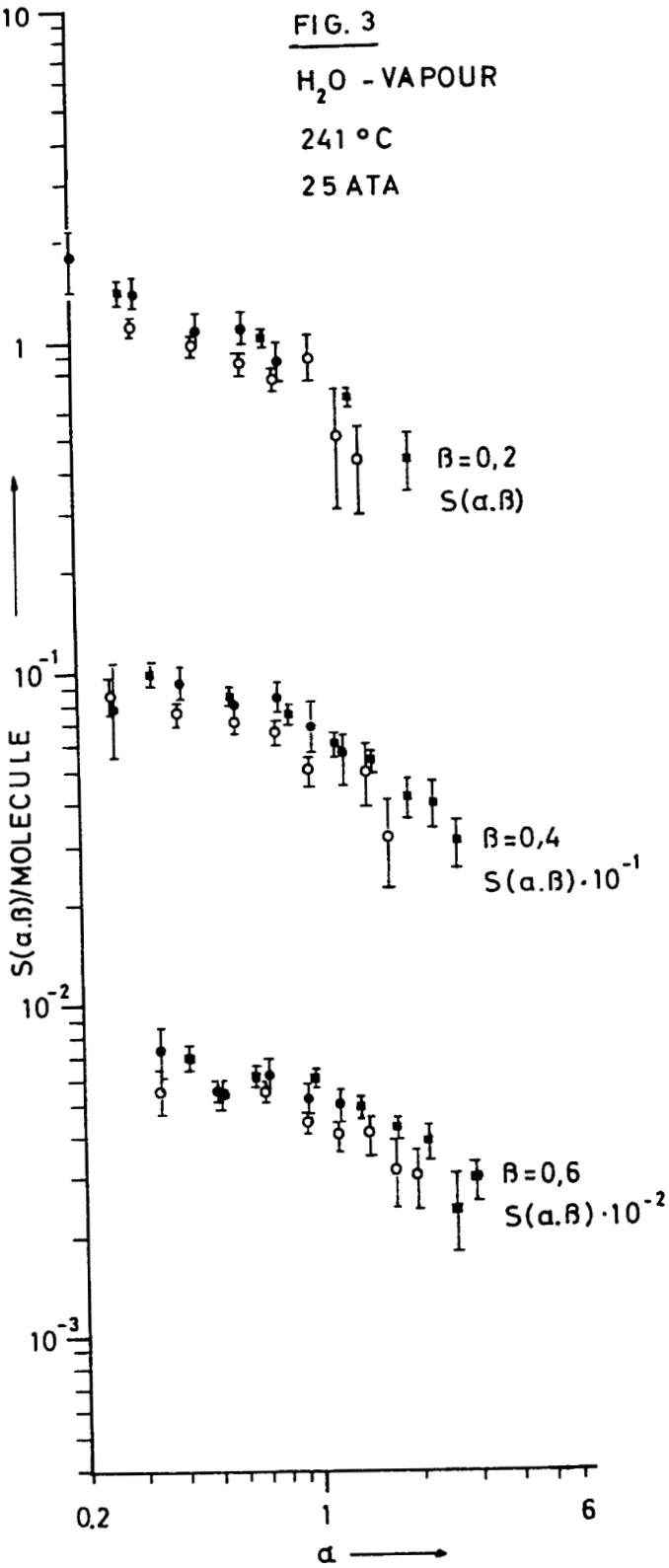


FIG. 3

H<sub>2</sub>O - VAPOUR

241 °C

25 ATA



18,2 mev - two runs (o, ●)

32,6 mev - one run (■)

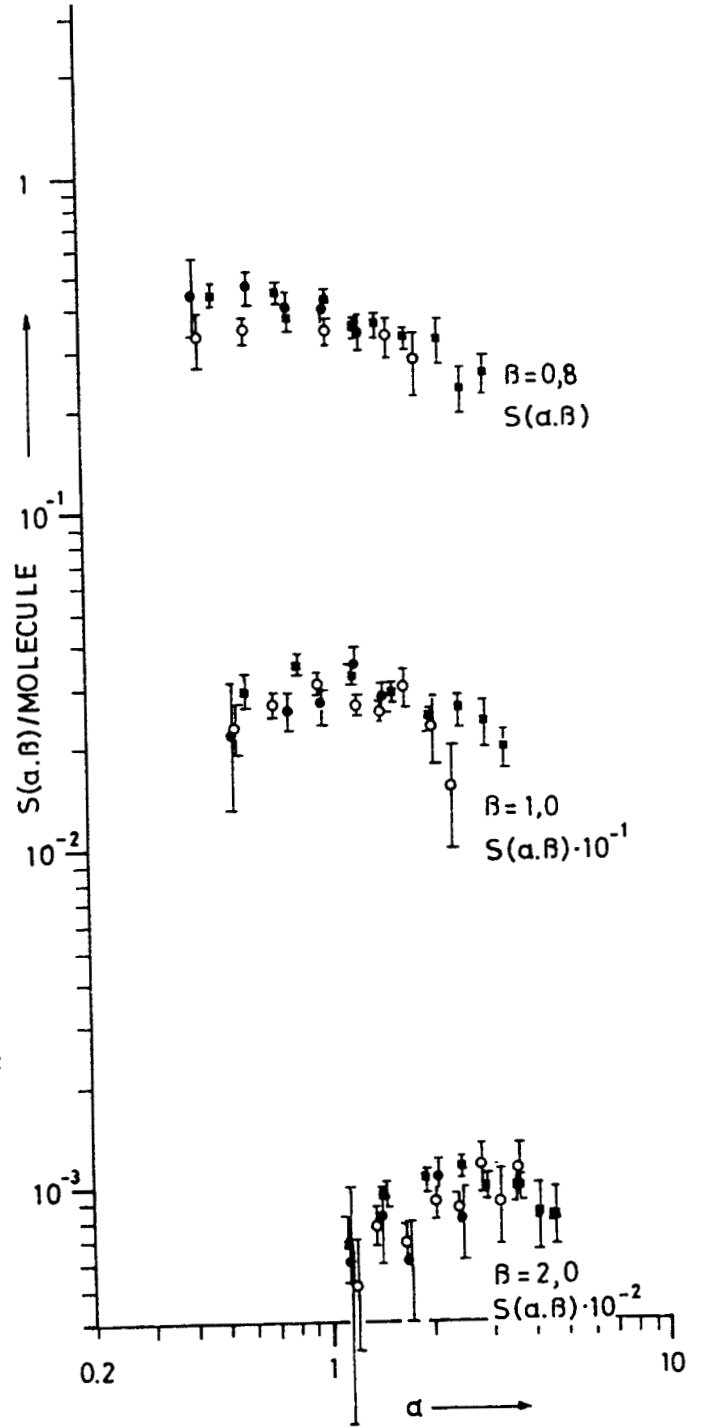


FIG. 5

- ▲ = 25.1 meV
- ▼ = 31.5 meV
- = 59.2 meV
- 

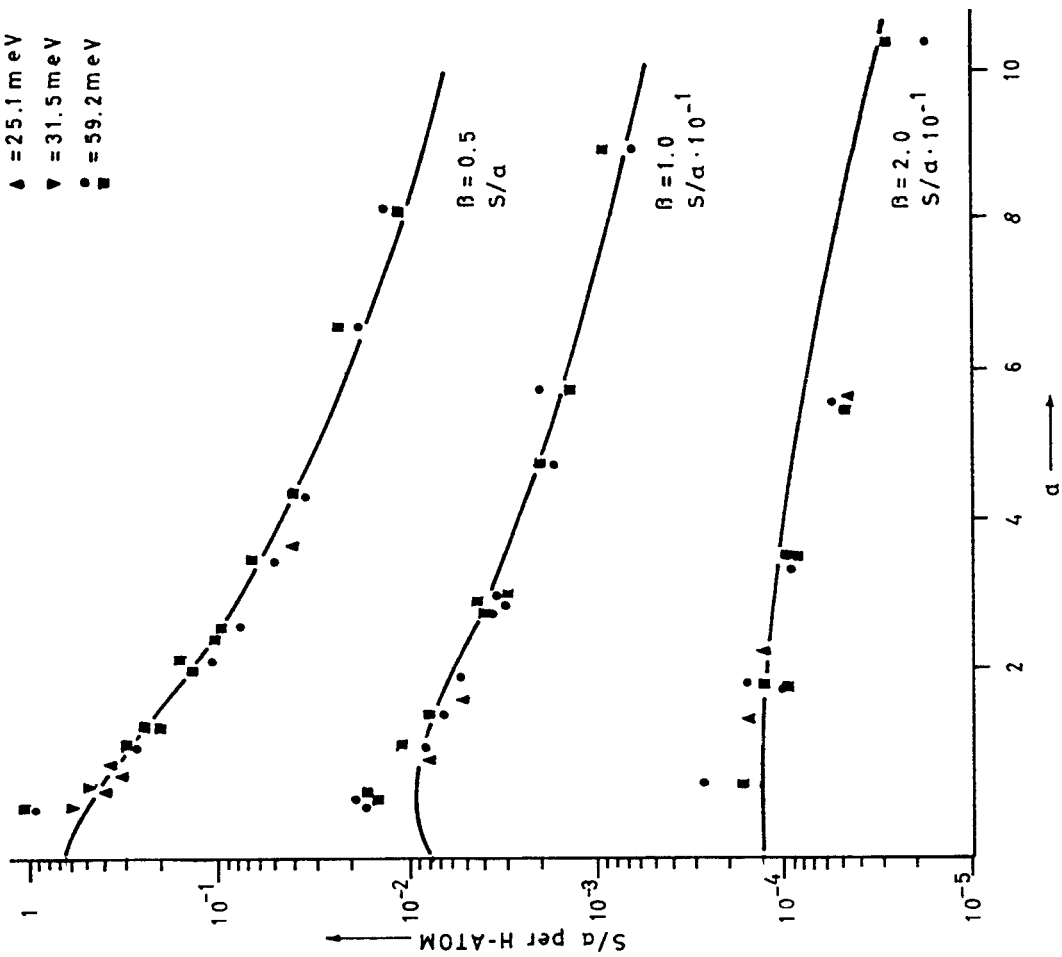


FIG. 4

- ▲ = 25.1 meV
- ▼ = 31.5 meV
- = 59.2 meV
- 

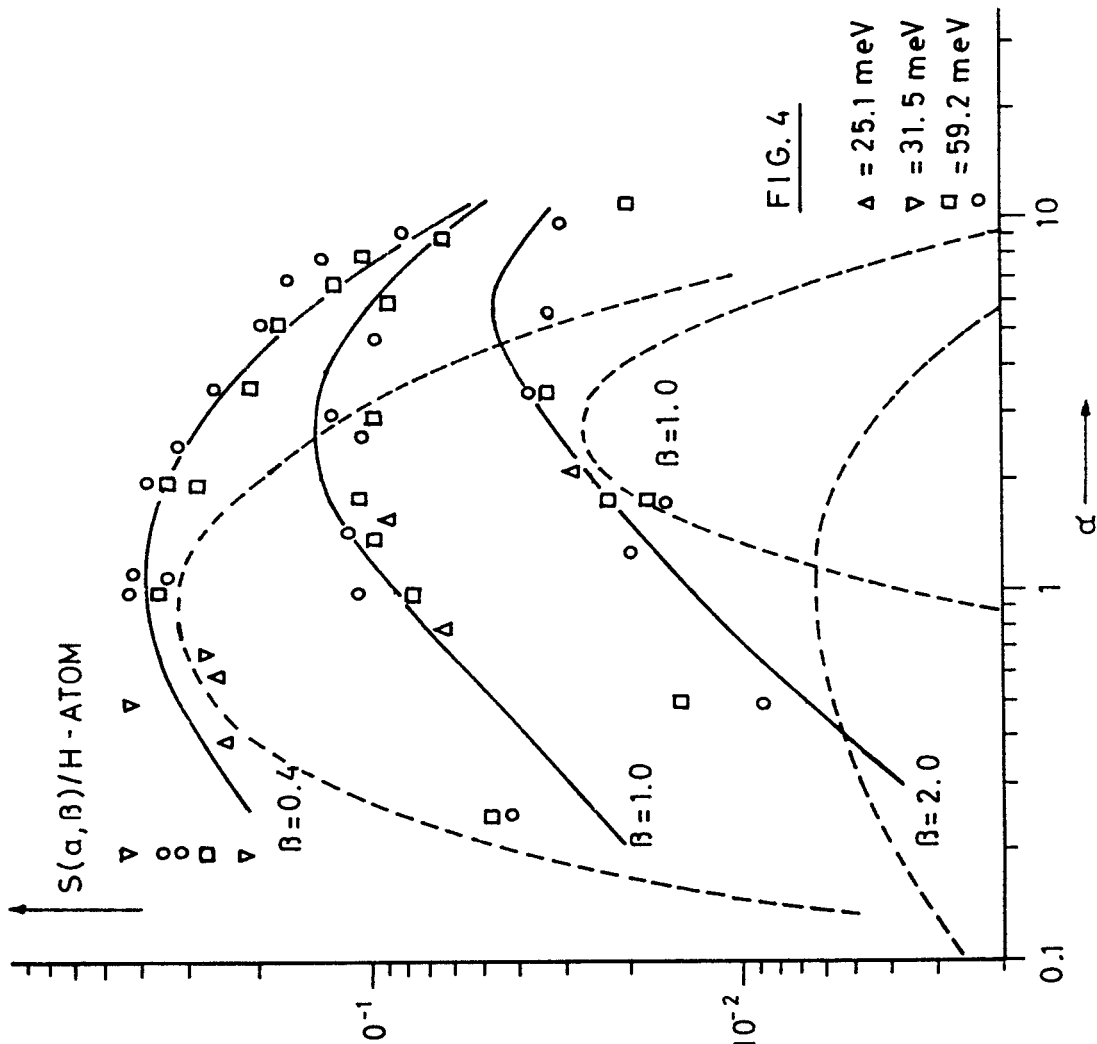


FIG. 8

ZrH  
T 210°C

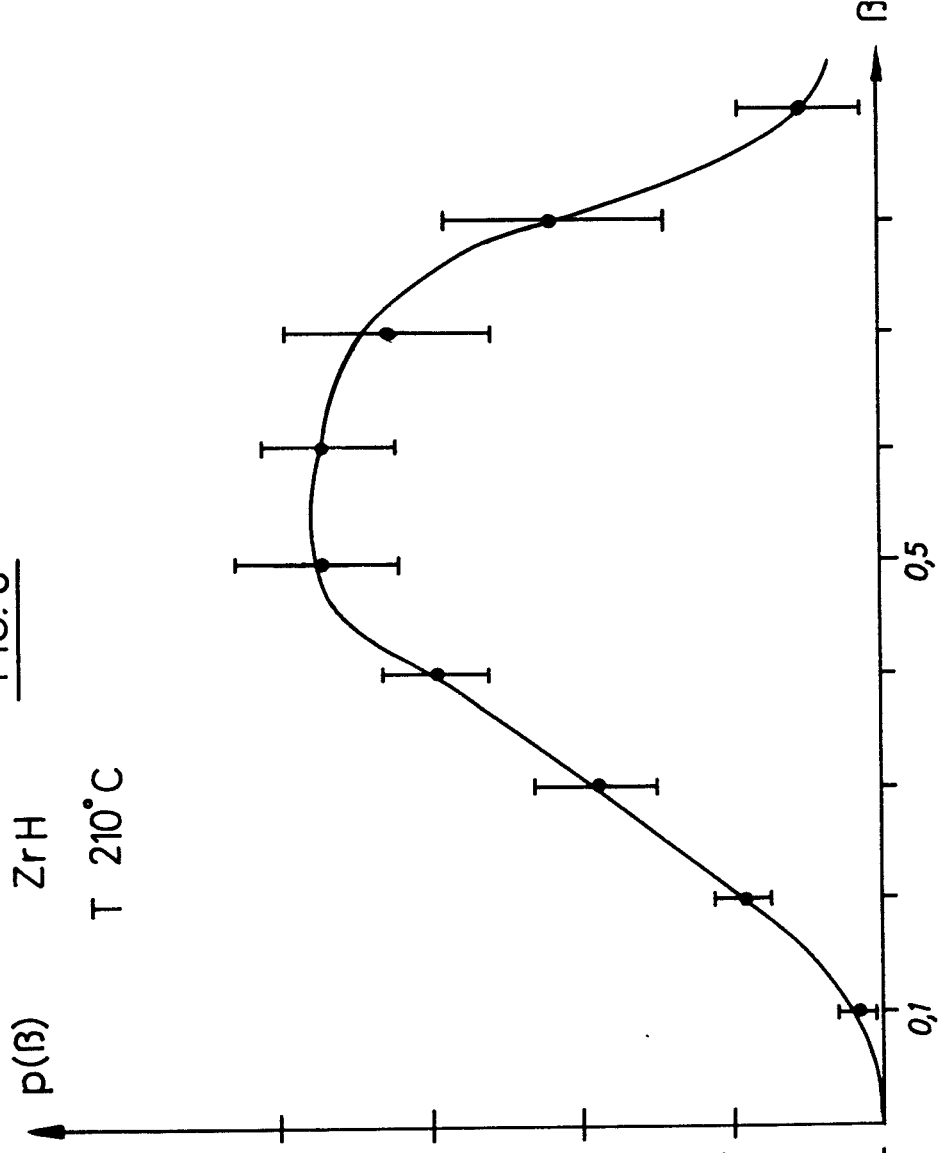


FIG. 6  
BENZENE  
20°C

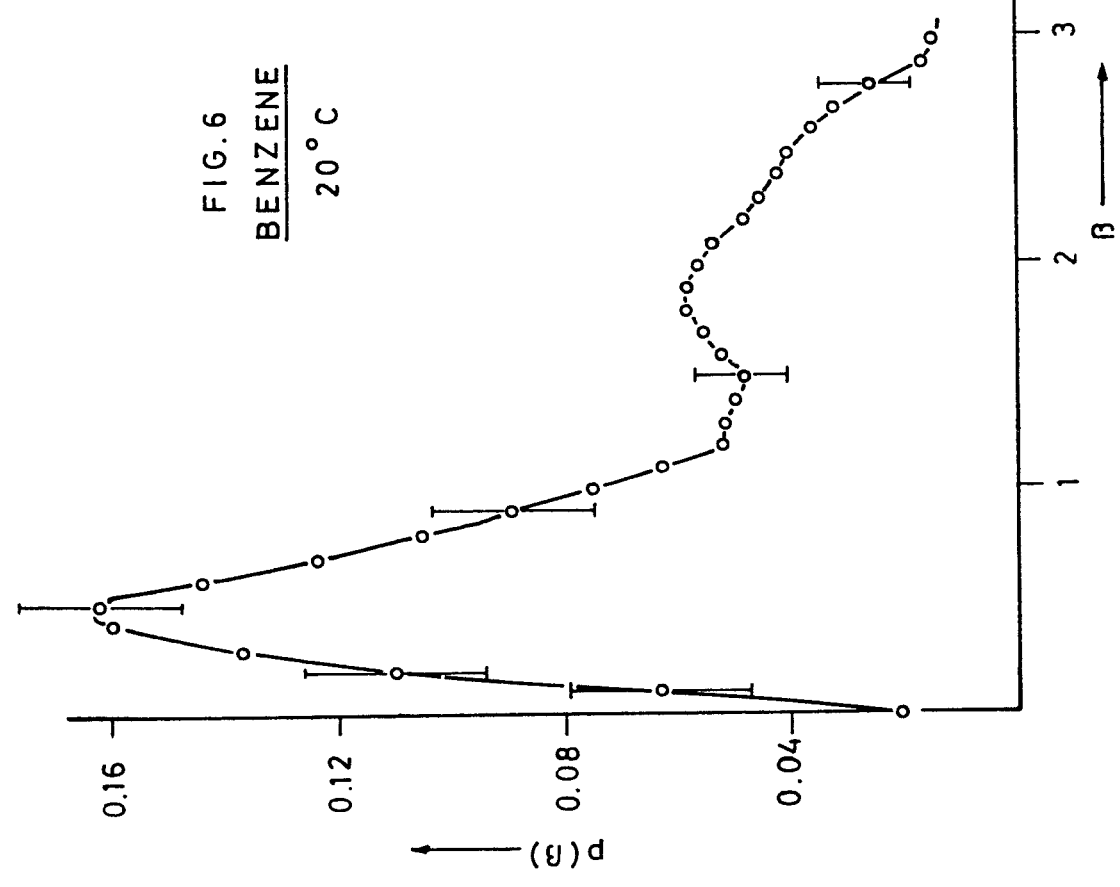


FIG. 7

ZrH<sub>1.1</sub>

210°C

E<sub>0</sub> = 32.6 meV

θ = 77.1°

