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INELASTIC SCATTERING OF THERMAL NEUTRONS FROM DOWTHERM "A"

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Abstract — Résumé — Аннотация — Resumen

INELASTIC SCATTERING OF THERMAL NEUTRONS FROM DOWTHERM "A". The angular and energy distributions of neutrons scattered from room temperature samples of Dowtherm "A" have been measured using a few different initial neutron energies in the range from 10^{-2} to 10^{-1} eV.

The experiment has been performed with a rotating crystal spectrometer as pulsed monoenergetic beam source and a 16×256 - channel time analyser for timing of the scattered neutrons. With this arrangement data for several angles have been taken simultaneously.

Results are represented in the form of differential cross-sections and also in the "Scattering Law" form. Physical interpretation with the aid of the space-time correlation function of Van Hove is discussed.

DIFFUSION INÉLASTIQUE DE NEUTRONS THERMIQUES PAR LE DOWTHERM "A". L'auteur a mesuré la distribution angulaire et le spectre d'énergie de neutrons diffusés par des échantillons de Dowtherm "A" à la température ambiante, en employant des neutrons dont l'énergie incidente variait de 10^{-2} à 10^{-1} eV.

L'expérience a été faite à l'aide d'un spectromètre à cristal tournant, employé comme source monoénergétique pulsée, et d'un analyseur en temps de 16 × 256 canaux. Ce dispositif a permis de recueillir simultanément des données pour différents angles.

Les résultats sont présentés sous la forme de sections efficaces différentielles et aussi sous la forme d'une loi de diffusion. L'auteur discute l'interprétation physique de ces résultats à l'aide de la fonction de corrélation spatiotemporelle de Van Hove.

НЕУПРУГОЕ РАССЕЯНИЕ ТЕПЛОВЫХ НЕЙТРОНОВ ДОУТЭРМОМ "А". Угловые и энергетические распределения нейтронов, рассеянных образцами доутэрма "А" при комнатной температуре, были измерены при различных исходных энергиях нейтронов в пределах от 10^{-2} до 10^{-1} эв.

Опыт приводился на спектрометре с вращающимся кристаллом, который представляет собой источник пульсирующего моноэнергетического пучка и временной многоканальный анализатор 16 x 256 для определения времени пролета рассеянных нейтронов. Одновременно с этим были получены данные для нескольких углов рассеяния.

Результаты представлены в виде дифференциальных сечений, а также в виде "закона рассеяния". Обсуждается их физическая сущность и согласование с пространственной временной функцией Ван Гофа.

DISPERSIÓN INELÁSTICA DE NEUTRONES TÉRMICOS POR EL DOWTHERM "A". El autor ha medido la distribución angular y energética de neutrones dispersados por muestras de Dowtherm "A" a temperatura ambiente, para una serie de energías iniciales de los neutrones comprendidas entre 10^{-2} y 10^{-1} eV.

El experimento se ha realizado con un espectrómetro de cristal rotativo, que se ha utilizado como fuente pulsada de haz monoenergético, y un analizador de tiempo de 16 × 256 canales para el cronometraje de los neutrones dispersados. Con este dispositivo, se han obtenido simultáneamente datos correspondientes a diferentes ángulos.

El autor expone los resultados en forma de secciones eficaces diferenciales y también en forma de "ley de dispersión". Estudia la interpretación física del fenómeno con ayuda de la función de correlación espacio-tiempo de Van Hove.

I. INTRODUCTION

In the past few years considerable effort has been spent on measuring the influence of chemical binding on the scattering of slow neutrons. In this

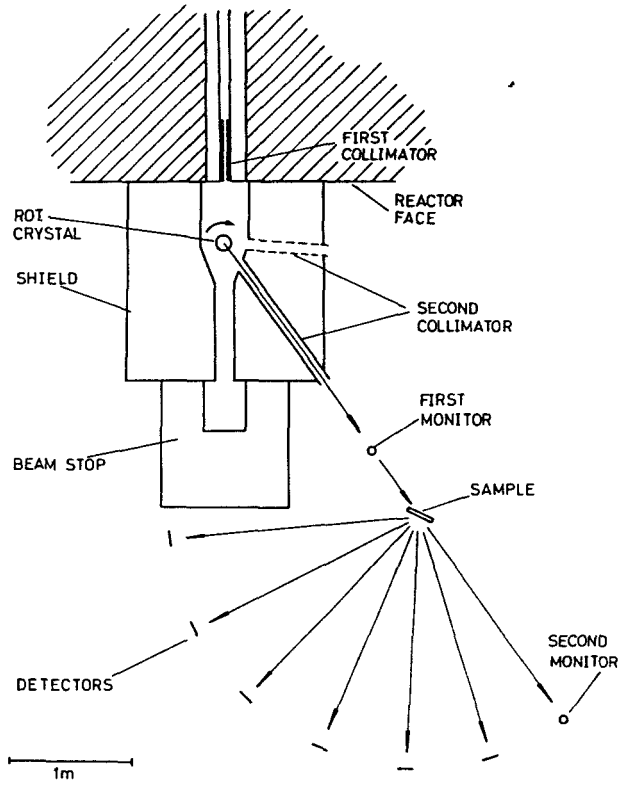

Fig. 1

Schematic diagram of the rotating crystal spectrometer at the reactor Diorit, Würenlingen, Switzerland.

In scattering experiments the sample position was 2.65 m behind the crystal. For a primary energy of 0.032 eV the pulse width at this position was 22 μs and the intensity about $1.5 \cdot 10^3$ n/cm^2 s. The performance of the machine was tested continuously with two BF_3 monitors in the monochromatic beam. One was a short distance in front of the sample and the other 2 m behind it. With these monitors energy, resolution and intensity of the incoming neutrons could also be measured.

The scattered neutrons are detected with a set of 5 in diam. scintillation counters positioned at a distance of 2 m from the sample and at angles of 20°, 40°, 60°, 70°, 80°, 100° and 120° with respect to the monoenergetic beam. The signals of each detector were fed into a group of 256 channels of a 4096-channel magnetic core time sorter, using 8 μs channel width. The detector efficiencies have been determined by scattering from vanadium.

Scattering samples

For liquid Dowtherm "A" sample containers consisting of an aluminum frame 0.06 cm thick covered with 0.03 cm thick windows of aluminum have

been made. The surface of the sample was 5 cm broad and 15 cm high, so that the full beam cross-section could be used. The transmissions of our samples varied between 90 and 75% for different incident energies and different horizontal inclinations to the beam. Diphenyl which is solid at room temperature was heated slightly above melting point and cast in a similar frame.

III. MEASUREMENTS AND RESULTS

Before scattering runs were started, a flux map at the sample position was taken and the detector efficiencies were determined. Usually 20-h runs, 10 h for sample in and 10 h for background, were made. Fig. 2 shows the results of a typical sample and background run.

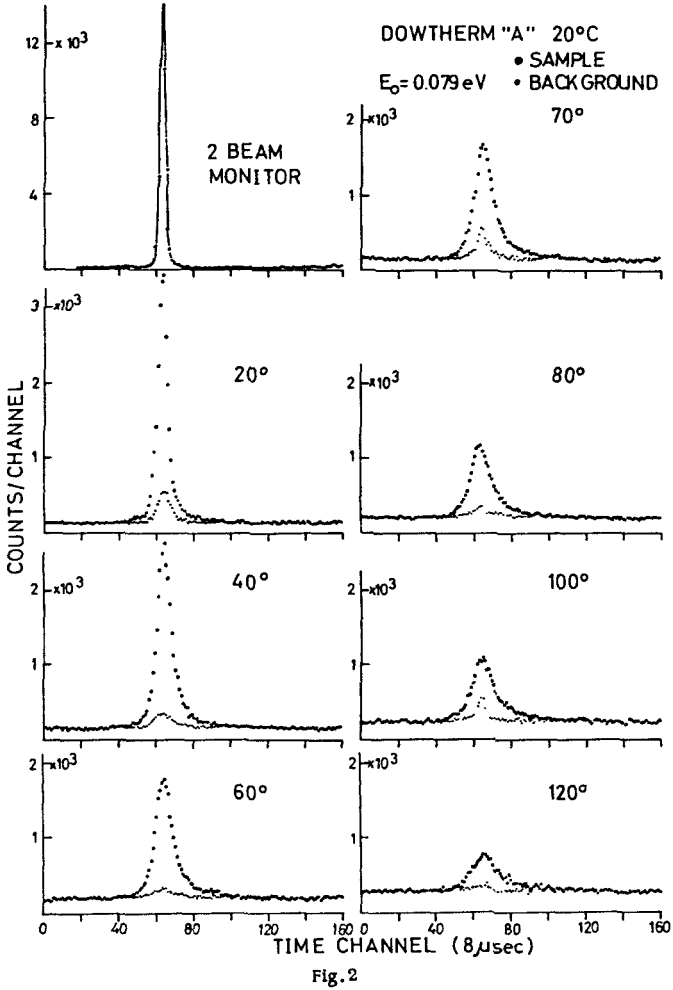

Fig. 2

Raw scattering curves for an incident energy of 0.079 eV and seven scattering angles. These data were obtained in 20 h, 10 h for sample in and 1 h for background.

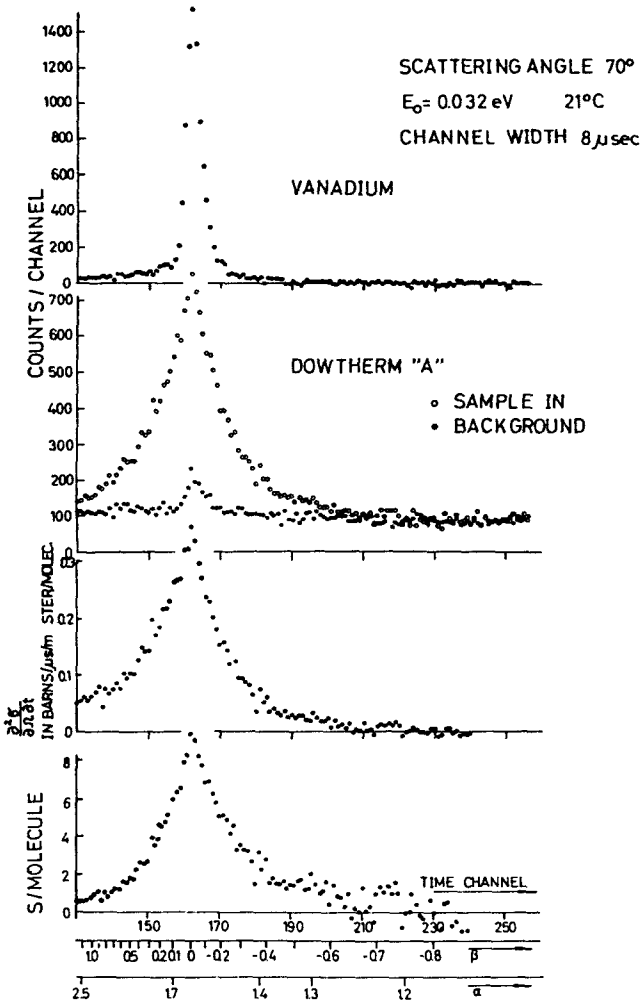

Fig. 3

The principal steps in conversion of the experimental data.

Memory content of the time analyser was punched on paper tape. This tape was fed directly into a computer. A computer programme has been written with which the data were normalized for equal initial intensity and, if necessary, also for equal background at small and large neutron velocities. Then data were converted to differential scattering cross-sections and finally to the scattering law form $S(\alpha, \beta)$. These steps are demonstrated in Fig. 3 for the results at an angle of 70° and an energy of 0.032 eV. No smoothing of the statistics and no corrections for the finite energy resolution have been done. The S-values as far as now available are shown in Fig. 4 and 5. In these diagrams the results of two incident energies and seven different scattering angles are plotted for fixed β-values as parameters. This kind of plot is a good check of the reliability of the data.

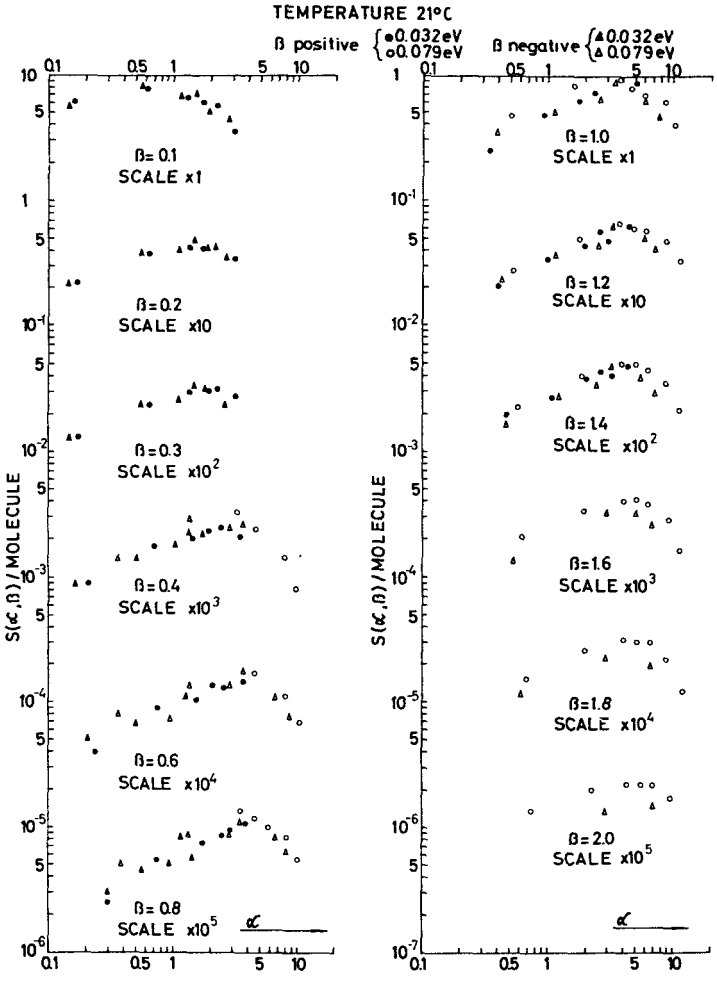

Fig. 4

Scattering law for Dowtherm "A" at room temperature represented per molecule.

IV. DISCUSSION AND CONCLUSIONS

No significant differences between the room temperature scattering data for Dowtherm "A" and diphenyl have been observed. The diphenyl cross-sections are somewhat higher for $\beta = 0$ and decrease more rapidly with increasing β-s for small β-s. Also there is no clear indication of resonance bands in the data.

The exact way to determine the space-time self-correlation function $G_s(r, t)$ of Van Hove is to make double Fourier transformations of the measured differential scattering cross-sections. Because we have measured only a limited range of energy and momentum transfers the results of such transformations would be doubtful. Instead we have tried the method of Egelstaff

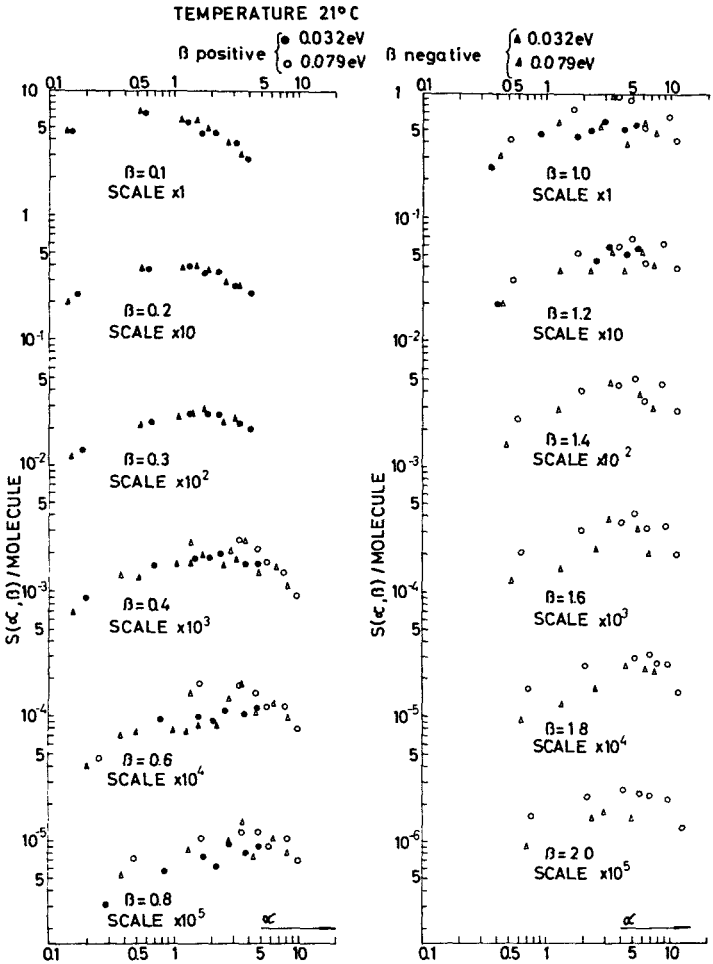

Fig. 5

Scattering law for diphenyl at room temperature.

to extrapolate S/α to $\alpha = 0$ for different fixed β. Multiplication of the limit $[S/\alpha]_{\alpha=0}$ with β^2 yields a generalized frequency distribution $p(\beta)$ which is directly related to the velocity correlation function of the system. There is a fair agreement between solid and liquid data. The curves also show two peaks. In the region of the last peak several infra-red absorption bands have been found. But at present the uncertainties of the most points are too large as to draw more serious conclusions.

Measurements at other initial energies are in progress and measurements with higher temperatures are planned.

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DISCUSSION

W. KLEY: I do not quite understand your technique of obtaining background data by removing the scattering sample from the impinging beam. We have found that if the sample is taken out of the beam the fast neutron background is also removed and therefore cannot be detected.

W. GLÄSER: I can only say that this is the usual technique for measuring background in other time-of-flight experiments as well. In background measurements account must of course be taken of the empty sample container in the beam. The monochromatic beam is a very pure one, so there is only a very small fast neutron background. We have made corrections for differences in sample background in the beam and outside it.

B.N. BROCKHOUSE: One of the attractive features of the rotating crystal spectrometer used by Dr. Gläser is that the fast neutron background is time-independent, so that a variety of ways are available for dealing with it.

W. KLEY: Yes, but with the technique described by Dr. Gläser you not only produce thermal-neutron pulsed beams but you also get a continuous background of fast neutrons. When you put the sample in the pulsed beams, you have the continuous background of fast neutrons which is then seen by the detector as long as the sample is in the beam. When you take the sample out, you remove the fast neutron background.

W. GLÄSER: That is true, but there will be a difference between these two sorts of background and since the background is not time-dependent you can make a very good correction for these effects. The results would be somewhat better than with a fast-chopper system for example.

P.K. IYENGAR: I think Dr. Kley's point is that you could have cut the beam, say with a cadmium shield, and then have taken the background, which would include the fast neutron effects.

B.N. BROCKHOUSE: I think it would be appropriate at this point to present a brief report on work which has been done by my colleague L.N. Becka on rotational transition in cyclohexane.

A number of "globular" organic compounds*, which undergo phase changes that are believed to be of rotational nature, have been studied using the rotating crystal spectrometer** at the NRU reactor at Chalk River. The

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* TIMMERMANS, J. phys. and chem. Solids, 18, (1961) 1.

** BROCKHOUSE, B.N. "Inelastic Scattering of Neutrons in Solids and Liquids", IAEA, Vienna (1961)113.

way much information about many-particle systems such as solids, liquids and gases has become available.

The information about scattering behaviour is of primary interest for reactor physics because the binding effects determine the slowing down of neutrons in the energy region where the spectrum shape is most important. Data of moderators like H_2O, D_2O, graphite, beryllium and terphenyl have been measured with the phased-rotor systems at Chalk River and Idaho Falls [1, 2] .

Of similar interest for organic moderated and cooled reactors are the chemical compounds diphenyl, diphenyloxyd and the eutectic mixture of both called Dowtherm "A".

In continuing the series of experiments with moderators several samples of Dowtherm "A" and diphenyl have been studied using a rotating crystal time-of-flight spectrometer. The selected materials have the advantage of easy handling and of being primarily incoherent scatterers.

A few incident neutron energies, 0.019, 0.032 and 0.079 eV have been used. For each energy the scattered intensities were taken simultaneously at several angles. Usually the seven detector angles 20°, 40°, 60°, 70°, 80°, 100° and 120° were realized. Examples of results are presented as raw data and in the converted form as differential scattering cross-section per solid angle and time-of-flight. Results as far as now available are also represented in the well known scattering law form $S(\alpha, \beta)$. $S(\alpha, \beta)$ [3] is different from the function $S(\kappa, \omega)$ defined by VAN HOVE [4]. Energy change is measured in units of $K_B T$, $\beta = \hbar\omega / K_B T$, where $\hbar\omega$ is the energy change and $K_B T$ is the temperature of the sample in energy units. Momentum transfer is represented by $\alpha = \hbar^2 \kappa^2 / 2M K_B T$, where $\kappa = \kappa - \kappa_0$ is the difference of initial and final wave vector of the neutron and M is the mass of the principal scattering nucleus. Furthermore the detailed balance factor is extracted with the consequence that $S(\alpha, \beta)$ is even in β.

The determination of the generalized frequency distribution $p(\beta)$ proposed by EGELSTAFF [5] was tried.

II. EXPERIMENTAL ARRANGEMENT

Apparatus

Bursts of monoenergetic neutrons have been producted with a rotating crystal using a neutron beam from the reactor Diorit at Würenlingen. A preliminary description of the spectrometer has been given earlier [6] . The general layout is shown in Fig. 1. The rotating single crystal fulfills two tasks. Firstly, due to the Bragg reflection at the planes of the crystal it produces monoenergetic neutrons in a selected direction and secondly, due to its spinning, it chops the beam and delivers short pulses for the time-of-flight measurements. The present measurements are done with a copper crystal usually spinning at 14 200 rpm, using two (111) reflections per revolution. The advantages of this device are its simplicity, the fact that there is almost no time-dependent background and the possibility of using several incident energies simultaneously. Principally the last feature allows to take all information needed for scattering law calculations in one single run.

results for cyclohexane are shown in the figure below. At 115°K, below the transition at 186°K, the patterns show strong elastic peaks and at least two inelastic peaks (at 6.2 and 10 meV). The fact that the elastic peaks for the

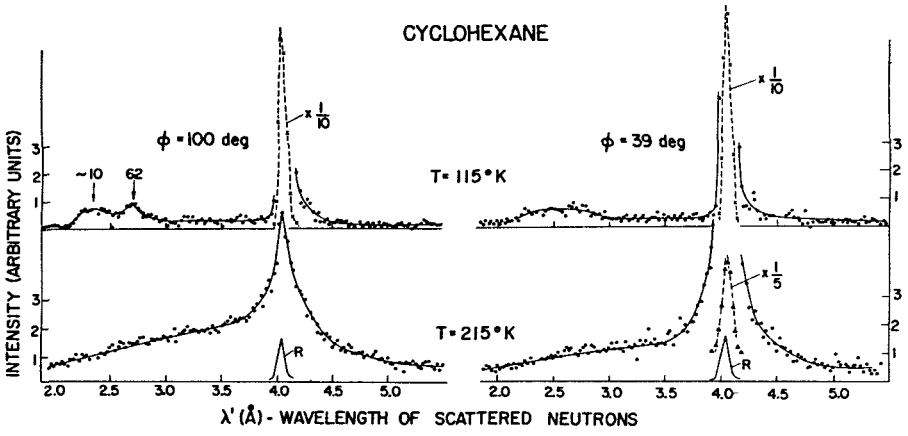

Fig. 1

Wavelength distributions for cyclohexane at two angles and at temperatures above and below the transition.

two angles are not greatly different shows that the Debye-Waller factor is normal, and therefore that the molecule is not in rotation. At 215°K the Debye-Waller factor is greatly reduced, especially at the larger angle. Thus the proton motion is very great and the molecule is presumably in rotation. Since the two peaks also disappear, they must represent rotational levels.

Other substances studied were 2-2-dimethylbutane and 1-4-diazo-bicyclo-(2.2.2)-octane, with differing but interesting results. A full report is being submitted to J. chem. Phys.