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SEPARATION OF THE COHERENT AND INCOHERENT SCATTERING OF C₂Cl₆ BY POLARIZATION ANALYSIS

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Résumé. - Dans la diffusion des neutrons par C₂Cl₆, la contribution incohérente à la largeur quasiélastique donne des informations sur le mouvement réorientationnel des molécules à partir de la diffusion des atomes individuels. La diffusion cohérente contient par contre des effets d'interférence. L'analyse de polarisation permet de distinguer entre la diffusion cohérente et la diffusion de spin incohérente. S'il est possible de moduler la polarisation du faisceau incident, on peut effectuer une expérience en temps de vol à haute résolution, avec analyse de polarisation. Sur le diffractomètre D7, nous avons utilisé huit détecteurs avec analyseur à supermiroir de section efficace de 10 x 5 cm² et un polariseur à supermiroir. Un flipper de Mezei fonctionnant avec une séquence de pulses pseudostatistiques et un second polariseur, génère le faisceau modulé de neutrons dans un état de spin donné. Les résultats montrent qu'il est possible de séparer les contributions cohérente et incohérente. Une analyse des résultats de temps de vol permet de déterminer les temps de relaxation des mouvements moléculaires.

Abstract. - In neutron scattering by C₂Cl₆ the incoherent contribution to the quasi-elastic linewidth gives information on the reorientational motion of the molecules by scattering on individual atoms. The coherent scattering contains in contrast also interference effects.

With polarisation analysis it is possible to distinguish between coherent and spin incoherent scattering. If it is possible to modulate the polarisation of the incoming beam one can perform high resolution TOF experiments simultaneously with polarisation analysis.

On the diffractometer D7 we use 8 detectors with supermirror analysers of 10 x 5 cm² cross-section and a supermirror polariser. A Mezei flipper supplied with a pseudostatistical pulse sequence and a second polariser generate the modulated neutron beam of one spin state.

The results show that it is possible to separate the coherent and incoherent scattering peak. An analysis of the TOF data gives the relaxation times of the molecular motion.

Introduction - The diffuse scattering of neutrons provides important information on the dynamics of condensed matter. In the plastic phase of molecular crystals a large contribution to this scattering is elastic and quasielastic. This is due to the reorientational motion of individual molecules and to molecule-molecule correlations.

Under the assumption of rigid molecules it is the incoherent scattering which is easier accessible to an analysis because we can deduce the density distribution (at t → ∞) of an individual incoherently scattering atom in the molecule from the elastic incoherent part. The quasielastic incoherent contribution on the other hand gives hints to the kind of motion of the molecule. The energy integrated incoherent scattering - the scattering by a single atom at t = 0 - is independent of the momentum transfer Q. The integration over elastic and quasielastic incoherent scattering only gives an intensity proportional to the Debye Waller factor. Coherent elastic contributions enter completely into the Bragg scattering as long as the orientational disorder is of a dynamical origin. From the coherent quasielastic part one gets again details of the molecular motion. The modulation in Q-space, however, is in general due to the interference terms and therefore different from
that of the incoherent scattering. The quasielastic coherent line widths may differ from the quasielastic incoherent widths also. Most importantly the energy integrated quasielastic coherent scattering can be compared with the diffuse X-ray scattering.

During the last few years a large amount of hydrogenous organic molecules have been examined \[1\]. Due to the very large incoherent cross section of hydrogen, one gets in these substances only the incoherent scattering part. Coherent scattering in a plastic crystal has been studied only once until now on a purely coherent scatterer CBr\(_4\) \[2\]. In the present work we investigate the diffuse scattering in an orientationally disordered crystal for which the incoherent cross section is comparable to the coherent one. A theoretical model must describe the coherent and incoherent parts simultaneously. Because it is difficult to fit such a complex formalism to the experimental data, it appeared desirable to separate the different scattering parts by an experiment.

**Experimental Aspects** - The behaviour of the neutron spin in coherent and incoherent scattering processes is well known and was demonstrated for the first time experimentally by Moon, Riste and Koehler \[3\]. In Table 1 are summarized the main features for spin flip (++) and non-spin flip scattering (++).

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<th>incoherent</th>
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<td>spin</td>
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<td>l(_2)</td>
</tr>
<tr>
<td>isotope</td>
<td>l(_3)</td>
<td>l(_3)</td>
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<td>1/3</td>
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<tr>
<td>(\downarrow\downarrow)</td>
<td>0</td>
<td>2/3</td>
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**Table 1**

It is evident that the spin flip scattering is only incoherent and therefore polarization analysis is a good method for the separation of coherent and incoherent parts. In the C\(_2\)Cl\(_6\) molecule the carbon atoms are in a good approximation only coherent scatterers \(\sigma_{\text{inc}} < 0.02\)b; \(\sigma_{\text{coh}} = 5.6\)b, while the chlorine atoms scatter as well coherently \(\sigma_{\text{coh}} = 11.59\)b as incoherently \(\sigma_{\text{inc}} = 5.47\)b. 70% of the incoherent cross section is due to nuclear spin incoherence \[4\]. To perform polarization analysis one needs a setup to determine the polarization of the neutrons before and after scattering. Such an installation is now available on the instrument D7 for diffuse scattering at the ILL for 8 detectors \[5\] (fig.1). The arrangement for spin analysis consists in a polariser using supermirrors \[6\] a spin flipper and in front of each of the 8 detectors a supermirror analyser. The polariser has a cross section of 5cm x 6.5cm. It consists of mirrors on top of 0.1mm thick boron glasses (cover glass for microscopy). The supermirror action \[7\] is effected by 80 layers of titanium and cobalt as a polarising layer with a gradient of the lattice constant so that the angle of total reflection of the cobalt alone is increased by a factor of two. Between the boron-glass and the supermirror is an anti-reflecting layer of gadolinium and titanium with an index of refraction one which excludes total reflection of the wrong spin by the glass substrate. The anti-reflecting layer reduces the reflected beam to 1%. The total thickness of gadolinium is 700Å, distributed in the layer with increasing content of titanium in the upper part of the layer. The correct concentration of the alloy is reached by evaporating thin layers of the material to alloy. In the polari-
ser these super mirrors are curved with a radius of curvature of 10m. They are ar-
ranged as in a Soller collimator with a distance of 1mm. In this way, one has a neu-
tron guide effect and all neutrons getting through this polariser are reflected at
least once.

Fig. 1 - TOF apparatus with polarisation analysis
1. neutron beam coming from the graphite monochromator
2. super mirror polariser
3. statistical spin flipper
4. super mirror analyser
5. spin flipper
6. furnace and sample
7. 8 detectors with polarisation analysis
Dotted areas represent a magnetic guide field.

In this way all transmitted neutrons must be polarised. This argument also applies
for the analysers, which have a larger cross section of 5cm x 10cm. The spectral
behaviour of transmission of the polarisers is given in fig. 2.
Fig. 2 - "Spin-up" spectrum in front of the polariser (a) and spectrum transmitted by the polariser (b) (52% transmission).

The 8 analysers in front of the detectors are mounted on a bench in the distance of 1.5m from the sample. This bench can be moved automatically by a step motor to make a scan like fig. 3. The flipper behind the polariser is a Mezei flipper [8]. Magnetic guide fields are installed between the flipper and the sample, and between sample and analyser.

In the time-of-flight measurement we use a pseudo-statistical flipper [9] with a shift register sequence. The sequence has a length of 127 units, and two memory channels are used per unit. An exact rectangular pulse shape is generated by varying the voltage across the flipper coil: 70 V are used during the rise time of the pulse and a lower value after the designed flipping field has been reached. The only action of the flipper (3 in fig. 1) is to invert the spin direction. A super mirror (4) following the flipper (3) generates the pseudo-statistical sequence of neutrons with an up spin. This sequence is used for measuring non-spin-flip scattering. Since the analysers (2) accept only up spins, one has, for spin-flip scattering, to invert the polarisation of the beam, before it passes the samples. This is accomplished by another flipper (5).

The powder sample (2.36g; 0.02 mole) is contained in an aluminium tube with 1 mm wall. This tube is kept in the vacuum and heated from the top and the bottom by bifilar heaters. All measurements were done at 393(±1)K.

Results and Discussion - The experimental set up was used in two different modifications: as a conventional diffractometer, and as a TOF spectrometer, in both cases with polarisation analysis.

Figure 3 shows a diffraction spectrum measured with $\lambda=4.85 \, \text{Å}$ in a Q-range from 0.1$\text{Å}^{-1}$ up to 2.2$\text{Å}^{-1}$. The data points represent the spin flip scattering, which is purely incoherent, and therefore the energy integrated intensity shows no modulation with Q. The dependence on the Debye Waller factor is small compared to experimental errors. The non spin flip scattering (solid line) contains coherent scattering, isotope incoherent scattering and 1/3 of the spin incoherent scattering. In the special case of the chlorine atom this incoherent part, which contributes to the non spin flip scattering, has approximately the same value as the spin flip scattering. That means that the difference between the two curves in fig.3 is the coherent scattering, which is constant up to a Q-value of about 1.5 $\text{Å}^{-1}$ but shows a remarkable increase at the end of the spectrum. This observation fits well with the diffuse intensity distribution observed earlier in a diffraction experiment without polarisation analysis over a more extended Q-scale (fig.4). The Q-dependent modulation of the intensity due to the coherent scattering part can be explained by scattering from individual orientationally disordered molecules:

$$I_{\text{coh}}(Q) = \langle r^2 \rangle - \langle r \rangle^2$$ (solid line in fig.4).
Fig. 3 - Powder diffraction pattern taken in the plastic phase of $\text{C}_2\text{Cl}_6$ (393K) with polarisation analysis. Data points - spin flip scattering, solid line - non-spin flip scattering.

Fig. 4 - The diffuse intensity distribution obtained in our experiment without polarisation analysis. The arrows indicate the Q-positions of the Bragg reflections seen in fig. 3.

Here $F$ is the molecular structure factor and the brackets denote an average over the different possible orientations of the molecule, which were calculated by a Monte Carlo method [10]. The incoherent part was calculated with the value given for the scattering amplitude of chlorine and taken as a flat background (broken line in fig. 4).

Fig. 5 - The spin flip scattering as a function of $E$ and $Q$. 

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We started the time dependent analysis by examining the spin flip scattering. Fig. 5 shows typical time of flight spectra transferred to an energy scale at five different Q-values. Due to the pure incoherent character of these data one can separate the incoherent elastic structure factor (EISF). The EISF is the elastic fraction of the energy integrated intensity at a fixed Q-value. The elastic part was determined by fitting the instrumental resolution function to the energy spectra. The resolution function was obtained in a measurement of a vanadium standard (FWHM : 0.20 meV). In general the Q-dependence of the EISF allows one to distinguish between rotational disorder models, which have a different density distribution for the incoherent scatterer at infinite time. Fig. 6 shows the calculated curves for the EISF for a rotational jump model where the molecule performs jumps between a finite number of preferred orientations given by the crystal symmetry. This model will be discussed analytically in the following. Curve is the EISF for a continuous rotation, where in the limit of t → ∞ the molecule is allowed to have any orientation. The EISF is given by the Fourier transform of the sphere on which the chlorine atoms move. The experimental data, measured in a Q-region where both models show a significant difference, exclude clearly the isotropic model. This is in good agreement with the anisotropic averaged scattering length density found earlier from coherent Bragg intensity data [11]. The Q and dependence of the elastic and quasielastic part of the mentioned jump model can be calculated by two alternative methods. First by solving a set of coupled differential equations for the time-dependent probability function of the incoherent scatterer [1]. A second more elegant way is provided by group theory [12]. Both methods lead under the assumption of regular octahedra for the C2C16 molecule to the scattering function:

\[ S_{\text{inc}}(Q,\omega) = \frac{1}{6} \sigma_{\text{inc}} \cdot e^{-\Delta W} \left\{ (1 + 4A + B) \delta(\omega) + 2(1 - 2A + B) \mathcal{L} (\omega, \Gamma_E) + 3(1 - B) \mathcal{L} (\omega, \Gamma_{T1}) \right\} \]  

with \( A = \frac{\sin(\sqrt{2}QD)}{\sqrt{2}QD} \) and \( B = \frac{\sin 2QD}{2QD} \) where D is the diameter of the octahedron. The first term in the sum represents the EISF. The second and third terms vanish for \( Q = 0 \).

The quasielastic part is described by two Lorentzians \( \mathcal{L} \) of the form:

\[ \mathcal{L} (\omega, \Gamma) = \frac{1}{\pi} \frac{\Gamma}{\omega^2 + \Gamma^2} \]  

The integral over the energy of eq. (1) gives an incoherent intensity which varies in Q only via the Debye Waller factor \( e^{-\Delta W} \).

The characteristic widths for the irreducible representations E and T1 can be given in terms of the individual jump rates \( \lambda \) referring to 90°, 180° and 120° jumps:

\[ \Gamma_E = 6(\lambda_{90} + \lambda_{180} + 2\lambda_{120}) \]  
\[ \Gamma_{T1} = 4(\lambda_{90} + 2\lambda_{180} + 2\lambda_{120}) \]  

The equation shows that in this high symmetrical case we can only obtain two different rates, where the first is a linear combination of the elementary jump around the four and three-fold axes. The widths were obtained by fitting the scattering function \( S_{\text{inc}}(Q,\omega) \), see eq.(1), simultaneously to the energy spectra of the different values for Q.

In this preliminary state the data allow to determine one relaxation time:

\[ \tau = \frac{1}{\lambda_{90} + 2\lambda_{120}} = 5.0 \pm 0.8 \times 10^{-12} \text{s} \]
The value obtained for $1/\gamma_{180}$ had a statistical uncertainty comparable to its calculated magnitude, and is therefore not given here. A further experiment with better counting statistics as well as the data treatment of the coherent scattering is under way.

Fig. 6 - Elastic incoherent structure factors (EISF)

1. Rotational jump model
2. Rotational diffusion model

References