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On the multiple scattering corrections in an inelastic neutron scattering experiment

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A phenomenological model for multiple scattering calculation in inelastic neutron scattering experiments is applied to the experimental data taken from a coherent scatterer (polycrystalline D_2O) and compared with the results of a Monte Carlo evaluation on the same sample.

1. Introduction

In a slow neutron inelastic scattering experiment a proper computation and subtraction of the multiple scattering contribution is essential; in particular, this is a crucial step in any experiment aiming at the extraction of the hydrogen-projected vibrational density of states in hydrogenous samples.

In a previous paper [1], referred to as P1 hereafter, we addressed this problem in the case of a polycrystalline sample of H_2SO_4. In P1 we proposed a phenomenological model based on the Impulse Approximation (IA) to account for the Multiple Scattering Contribution (MSC); so doing, we obtained a satisfactory result in extracting the vibrational density of states in the case of a purely incoherent scatterer. However, in that case we could not perform a Monte Carlo calculation of the MSC for an H_2SO_4 polycrystalline sample and our results were compared only qualitatively with those coming from a Monte Carlo calculation on liquid water. This did not allow us to draw definitive conclusions about the validity of our method.

In this paper we present new experimental data of inelastic neutron scattering from a sample of polycrystalline D_2O which is, on the contrary, a strongly coherent scatterer. For such a sample the density of states can be properly extracted only in the region of the intramolecular modes ($\hbar\omega > 200$ meV).

The aim of the present work is to compare the prediction of our phenomenological approach to the MSC with the results of an appropriate Monte Carlo (MC) evaluation on the same sample. In this context two points will be discussed in some detail, namely:

(1) The angular dependence of the MSC, which in our model is completely neglected;
(2) The possible existence of structures in the MSC at fixed angle which may show up in a proper MC simulation.

We want to stress that, being D_2O, a strongly coherent scatterer, the agreement between our approach and the MC calculation is not obvious even in the intramolecular vibrational frequency region.

2. Experimental results and evaluation of the multiple scattering contribution

Neutron scattering data were taken from a D_2O polycrystalline sample on the HRMECS spectrometer installed at the Intense Pulsed Neutron Source (Argonne National Laboratory, USA). This is a direct geometry spectrometer in which a Fermi chopper is used to produce monochromatic neutrons incident on the sample [2]. The D_2O sample at $T = 22$ K was contained in a flat aluminum 100×100 mm^2 and 3 mm thick cell placed at 90° to the incident beam; the beam energy

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was $E_0 = 697$ meV. The energy and momentum of scattered neutrons were recorded in time of flight using rings of detectors placed in the angular range $-19° \leq 2\theta \leq 19°$ and $85° \leq 2\theta \leq 140°$, where 2θ is the scattering angle. Further details on the experimental setup and the complete data analysis can be found in a companion paper [3].

We turn now to a description of the two modes of evaluation of the MSC. The basic ingredients of our model applied to a slab geometry are (for details see P1):

(1) the multiple scattering is essentially due to double scattering events and is almost isotropic;
(2) for high incident neutron energy and at low scattering angles, the relevant MSC comes mainly from two scattering events 90° apart.

In our experimental conditions ($E_0 = 697$ meV), the momentum transfer Q is of the order of 20 Å^{-1}, for $\hbar\omega \sim 300$ meV and $2\theta = 90°$; in this condition the scattering law $S(Q, \omega)$ approaches the IA. In the IA, at large Q where pronounced interference effects in the coherent response are negligible, the coherent contributions are evaluated in the incoherent approximation [4]. The total differential cross section then becomes proportional to an $S(Q, \omega)$ Gaussian in form, with a weight given by the total (coherent and incoherent) cross section. As a consequence, one can measure $S_{90°}(Q, \omega)$ (i.e. $2\theta = 90°$) and the MSC can be estimated as a convolution of two identical Gaussians of parameters (E_0, σ_0^2). The result is a Gaussian of variance $\sigma_M^2 = 2\sigma_0^2$ peaked at $E_M = 2E_0$. The MSC estimated in this way must then be put an on absolute scale.

Using the above prescription, a Gaussian has been fitted to our data recorded at $2\theta = 87°$ and $\sigma_0 = (192 \pm 2)$ meV, $\sigma_M = (272 \pm 3)$ meV were found. Indeed, due to the experimental arrangement, data at $2\theta = 90°$ cannot be taken. The Gaussian shaped MSC of parameters (E_M, σ_M^2) have then been normalized to the overall multiple scattering fraction, Δ, evaluated following Sears [5]. In our experiment Δ was 17% of the total scattered intensity. It is worth noting that the double scattering contribution (using [5] again) was estimated to be 16% of the total; on these grounds it is reasonable to assume that the MSC is entirely due to double scattering events. As an example the final result of the calculation is shown in fig. 1, where the MSC is plotted together with $S(Q, \omega)$ prior to subtraction, at $2\theta = 12°$.

On the other hand, in the approach based on the MC calculation the MSC can be evaluated from the density of states and from the experimental parameters in three steps: calculate the symmetrized $S_{SYM}(Q, \omega)$, determine the multiple scattering component with Copley's MSCAT code [6] and then compare it with the experimental data. The symmetrized $S_{SYM}(Q, \omega)$ is calculated by means of a modified version of Copley's SABMAK routine [6]; the calculation of $S_{SYM}(Q, \omega)$ assumes the incoherent, monoatomic, cubic, harmonic approximation for one- and two-phonon terms, while third and higher order phonon terms are calculated according to a modification of Sjolander's theory [7]. In our case the density of states $g(\omega)$ for D_2O was obtained from H_2O data [8] scaling the frequency ω by a factor $\sqrt{2}$ to account for the isotopic effect. The total MSC as obtained with the MSCAT code using the above $S_{SYM}(Q, \omega)$ together with the appropriate experimental setup are shown in fig. 2 for four selected scattering angles 2θ out of a calculated total of eight. It is immediately apparent that the MSC in the region of interest to our experiment, i.e. above 200 meV [3], is a

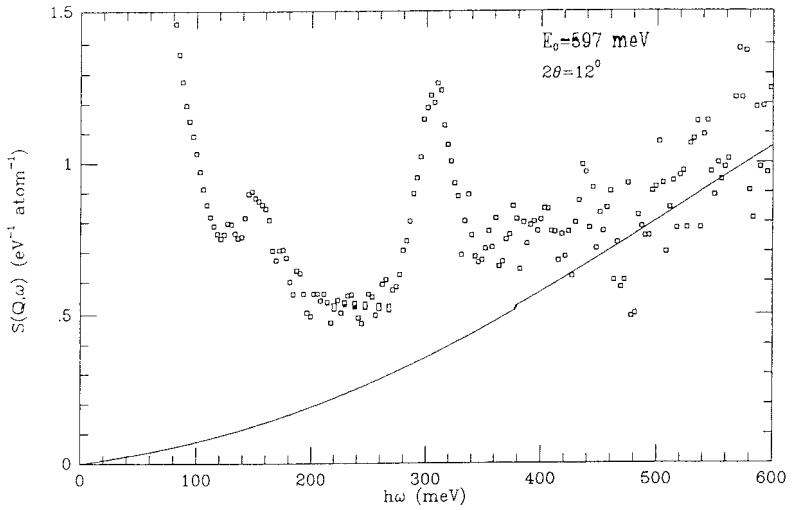

Fig. 1. Experimental scattering law for deuterium atoms $S(Q, \omega)$ (squares) of the D_2O polycrystalline sample before multiple scattering correction compared with the MSC according to our phenomenological model (continuous line).

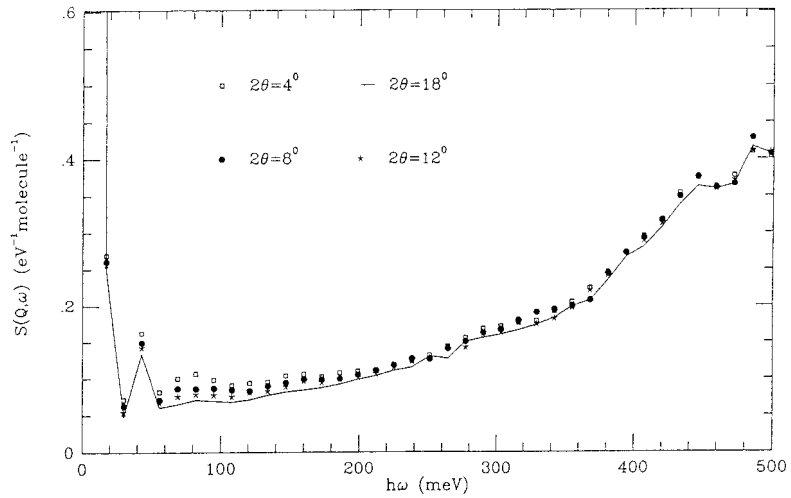

Fig. 2. Behaviour of the MSC as a function of energy at four selected scattering angles according to the Monte Carlo evaluation.

monotonically increasing function of frequency and no clear evidence of structures can be found. Moreover, in the above energy region for the whole experimental 2θ range the angular dependence of the MSC is almost absent and in any case within the estimated statistical accuracy of the MC calculation.

3. Discussion and conclusions

The results from the two different calculations are presented in fig. 3, where the data are plotted on the same scale as a function of frequency. From fig. 3 one can see that the agreement between the results of the Copley's code and our phenomenological approach is quite good for energy values higher than 200 meV, although the first one calculated the total MSC and the latter estimates the double scattering contribution alone. This finding confirms, that, in a slab geometry, the double scattering is indeed the relevant contribution to the total multiple scattering. The systematic deviation observed in the low energy region was however expected: indeed, the IA works only in the region above 200 meV, and therefore our approach does not properly

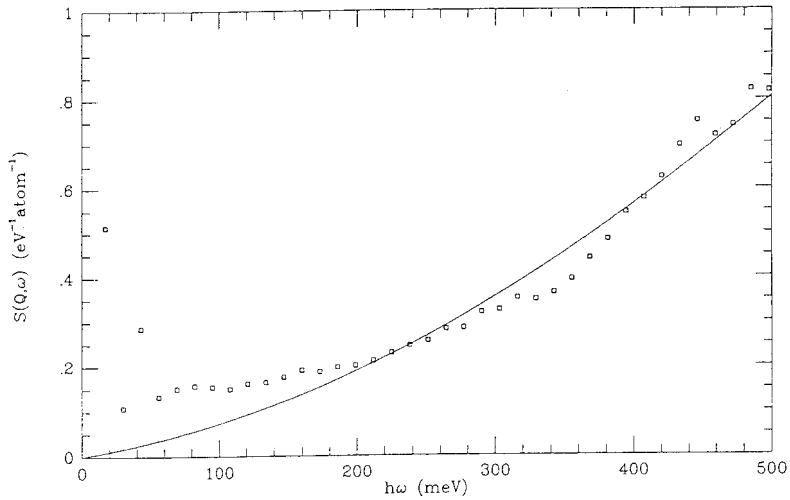

Fig. 3. Comparison of the two evaluations of the multiple scattering fraction at $2\theta = 12°$ for deuterium atoms. The line corresponds to our model, while the squares refer to the MC computation.

account for the scattering elsewhere. On the other hand the MC calculation accounts also for the low frequency contributions to the scattering, although this is still done within the incoherent approximation. The comparison shown in fig. 3 together with the result of fig. 2, implying that no appreciable dependence of the MSC on the scattering angle exists, establishes the validity of our method also in the case of a partially coherent scatterer. This has two main consequences. The first one is that the MSC can be estimated at a fair level of accuracy with no computational effort. The second one is of a more fundamental character; in fact one needs an a priori knowledge of the density of states $g(\omega)$ in order to estimate the MSC with the MC method; this is somewhat in contrast with $g(\omega)$ being the quantity to be measured in the experiment. On the other hand, our model does not suffer from this weakness, since it is based on the IA which is (asymptotically) valid whatever the form of the interatomic potential and hence independent of the particular form of $g(\omega)$.

Incidentally, we want to stress again that the method works for both incoherent and coherent scatters; however this is probably inherent to the nature of inelastic scattering itself, since coherent scattering effects is more likely to show up in the elastic and quasi-elastic regions.

References

[1] C. Andreani, V. Merlo and M.A. Ricci, Nucl. Instr. and Meth. B36 (1989) 216.
[2] C.K. Loong, S. Ikeda and J.M. Carpenter, Nucl. Instr. and Meth. 260 (1987) 381.
[3] C. Andreani, J. Mayers, P. Postorino and M.A. Ricci, to be published in Molecular Physics.
[4] S. Lovesey, Theory of Neutron Scattering from Condensed Matter, vol. 1, Intern. Series of Monographs on Physics 72 (Clarendon, Press, Oxford, 1984).
[5] V.F. Sears, Adv. Phys. 24 (1975) 1.
[6] J.R.D. Copley, Comput. Phys. Commun. 7 (1974) 289.
[7] A. Sjolander, Ark. Fys. 14 (1958) 315.
[8] C. Andreani, P. Bosi, C.K. Loong and F. Sacchetti, J. Chem. Phys. 83 (1985) 750.