Very High Energy Neutron Scattering from Hydrogen

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Abstract

The neutron scattering from hydrogen in polythene has been measured with the direct time-of-flight spectrometer, MARI, at the ISIS facility of the Rutherford Appleton Laboratory with incident neutron energies between 0.5 eV and 600 eV. The results of experiments using the spectrometer, VESUVIO, have given intensities from hydrogen containing materials that were about 60\% of the intensity expected from hydrogen. Since VESUVIO is the only instrument in the world that routinely operates with incident neutron energies in the eV range we have chosen to measure the scattering from hydrogen at high incident neutron energies with a different type of instrument. The MARI, direct time-of-flight, instrument was chosen for the experiment and we have studied the scattering for several different incident neutron energies. We have learnt how to subtract the gamma ray background, how to calibrate the incident energy and how to convert the spectra to an energy plot. The intensity of the hydrogen scattering was independent of the scattering angle for scattering angles from about 5 degrees up to 70 degrees for at least 3 different incident neutron energies between 20 eV and 100 eV. When the data was put on an absolute scale, by measuring the scattering from 5 metal foils with known thicknesses under the same conditions we found that the absolute intensity of the scattering from the hydrogen was in agreement with that expected to an accuracy of $\pm 5.0\%$ over a wide range of wave-vector transfers between 1 and 250 Å\textsuperscript{\text{-1}}. These measurements show that it is possible to measure the neutron scattering with incident neutron energies up to at least 100 eV with a direct geometry time-of-flight spectrometer and that the results are in agreement with conventional scattering theory.

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but no aspect has been identified that is clearly responsible for the observed decrease in the intensity of the scattering from hydrogen.

This paper describes a different approach. We have used a conventional direct time-of-flight instrument, MARI, at the ISIS pulsed source. Before this experiment the instrument has been used with incident neutron energies of 1 eV but this is too small to enable the scattering to be measured at the same energies as those employed on VESUVIO. We have therefore investigated if MARI could be used with higher incident neutron energies. After some difficulties we have successfully operated the spectrometer at a range of energies between 0.5 and 600 eV. This necessitated removal of the nimonic chopper that normally reduces the background on MARI and so developing a way of measuring and subtracting this background reliably from the data. We also had to rewrite the spectrometer software so that it correctly enabled us to operate at higher incident neutron energies.

Our most recent set of experiments used samples held at room temperature and supported so that there was no other material in the neutron beam at the sample position. We studied three samples of polythene, low density at two thicknesses and high density at one thickness and to obtain an accurate absolute normalisation we used five metal foils of Pb, Ni, Cu, Nb and V. The samples were all chosen to be of a thickness such that less than 5% of the incident neutron beam was scattered and the counting times were typical a few hours for each sample. We shall describe the results in detail for three different incident neutron energies of 20, 40 and 100 eV.

The scattering expected in the impulse approximation from hydrogen from a direct time-of-flight spectrometer shows a peak in the energy transfer on the energy loss side equal to the kinetic energy of the recoiling hydrogen atom. As the angle increases the recoil energy increases until it becomes close to the incident neutron energy for a scattering angle of 90°. The scattering then cuts off abruptly due to the kinematics of the scattering. In the recoil approximation the width of the scattering is expected to increase linearly with the energy transfer while the intensity is expected to be independent of the scattering angle. The Waller-Froman J factor [6] is unity for hydrogen recoil.

The scattering was binned into 5 degree segments, the background subtracted, the changing efficiency of the detectors with energy allowed for, and the time-of-flight spectrum converted to an energy spectrum. The results are shown in fig 1 for several different scattering angles. At high scattered energies, low angles, the background subtraction has worked well and the result is flat on the high energy side of the peak. At small scattered energies the statistics become less satisfactory because of the increasing effect of the time to energy conversion. Because of this data with a scattering angle greater than 70° was not used in the analysis. The data for each spectrum was fitted to two Gaussians. One arises from the hydrogen recoil and the other from the carbon recoil. The parameters for the carbon peak were obtained from the data taken at high scattering angles 100° to 130° and extrapolated to lower angles to give the starting parameters (this data is not easily visible in the plots in fig. 1). The intensities obtained from some of these analyses are shown in fig. 2. It is clear that the intensity for each set of measurements is constant within error over the accessible range.

The absolute normalisation of the data was performed using the scattering from the metal foils. The data is similar to that for polythene except that the recoil energies are much smaller. The data was analysed in the same way and the average results are shown in fig. 3. For each incident neutron energy the intensity of the scattering was calculated from the known cross-section and thickness of the foil and compared with the measured scattering. The scatter about each straight line gives a measure of the accuracy of the absolute intensity measurements. These results were then used to calculate the absolute intensity of the hydrogen scattering from the polythene and the two are compared in fig. 4 as a function of the intensity of the scattering.
Figure 1. The scattering observed from polythene for one sample and at three angles as an energy scan.

Figure 2. The intensity of the scattering observed for each three samples and incident energies of 20, 40, 100 eV.

Figure 3. The scattering from the metal foils for each of the 3 incident energies plotted against the calculated scattering.

Figure 4. A summary of all the data for the hydrogen scattering plotted as absolute intensities against the calculated scattering. The straight line gives the scattering for each hydrogen atom as 80±4 barns.
The results shown in fig.2 were normalised to the scattering when the wave-vector became small. The results clearly show for all the results that within error the scattering is independent of the wave-vector transfer. This is different from the results found for polythene with VESUVIO because in that case the polythene scattering decreased in intensity as the wave-vector transfer increased. Our second result is illustrated in fig.4 where the results for the different samples of polythene are shown plotted against the calculated cross-section so that the slope of the line gives the scattering cross-section for two hydrogen atoms. This result of averaging to obtain the best estimate of the cross-section for hydrogen gives a result that is within 2% of the nominal value and we estimate the error to be ± 4%. These results clearly show that the scattering from hydrogen does not vary with the wave-vector transfer and that it has a value that is agreement with other measurements of that cross-section. A large part of the error in these measurements, particularly from the metal foils, arises from uncertainty in the scattering cross-sections particularly in the eV energy range. The measurements in this part of the energy range often differ by a few percent points and this makes a substantial contribution to the error.

Nevertheless we have shown with a conventional direct geometry chopper that quantitative experiments can be performed with 100 eV incident neutrons. The energy resolution is increasing rapidly at these energies and better measurements would require a redesign of the choppers available for experiments using MARI. This and also possibly improvements to shorten the time bins measuring the time-of-flight would make a larger improvement to the resolution and experiments would still be possible because the neutron flux was very large. Some experiments using the MARI spectrometer to measure electronic transitions are in progress to exploit this new range of possible experiments.

The conclusion about our results is that experiments using MARI have some considerable advantages over the earlier measurements on VESUVIO. This is largely because it is technically possible to perform measurements with different incident energies so as to have the same wave-vector transfer at differing scattering angles. Because VESUVIO has very few absorption foils the wave-vector transfer can usually only be obtained with one set of instrumental parameters. The detector coverage is also larger on MARI. Nevertheless these features do not provide an explanation of the discrepancy between the VESUVIO and MARI results. We were surprised in our experiments how strong the gamma ray flux was at high energies. Possibly this has not been taken account of properly in the VESUVIO experiments at least in part because it was not possible to obtain the same energy transfer with different scattering energies. Needless to say we consider that since we obtain the expected theoretical results our conclusion is that the MARI experiment is to be preferred. Since completing these measurements we have learnt of the measurements using an indirect time-of-flight instrument similar to VESUVIO but with an electron linac used to produce its neutrons. This experiment showed that the recoil from hydrogen in mixtures of deuterated water agrees with the conventional theory [7]. Taken together the experiments on the electron linac and ours suggest conclusively that the scattering from hydrogen at high energies agrees with conventional impulse-approximation theory.

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