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Anomalous neutron scattering from hydrogen

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Abstract

The intensity of the high-energy neutron scattering from hydrogen suggests that the cross-section is smaller than expected from conventional scattering theory. There have been several suggestions for the origin of this discrepancy including quantum entanglement, the breakdown of the Born–Oppenheimer approximation and experimental error but the situation is still not resolved. In this paper we re-examine the analysis of the experimental data and show that it can only be performed if we assume the validity of the impulse approximation and conventional scattering theory. This is because the integral over energy of the scattering function along the constant scattering angle trajectory diverges, in principle, for all scattering angles. This result formally invalidates the procedure normally used for extracting the experimental intensity and calculating the zeroth and first moments of $S(Q, \omega)$. We propose that a less assumption dependent way of obtaining the intensity is to combine the data from several individual detectors to produce a map of $S(Q, \omega)$ and then to numerically integrate this along lines of constant wavevector to obtain the moments, which can then be compared directly with scattering theory without the use of the impulse approximation. We also consider two approximate analysis methods that use the impulse approximation. The one that assumes the validity of the y -scaling impulse approximation is the more satisfactory. We apply both methods to analyse some recent experimental data on polythene, and the results show that the hydrogen scattering is 20% less than expected. We then finally show that experiment and theory can be reconciled if the monitor detector efficiency is energy dependent and suggest that this or possibly a breakdown of the Born–Oppenheimer approximation coupled with a careful analysis, as described above, could account for the discrepancy between theory and experiment.

(Some figures in this article are in colour only in the electronic version)

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1. Introduction

There has been considerable interest in the origin of the anomalous decrease in the scattered intensity of neutrons from hydrogen ever since the experiments of Chatzidimitriou-Dreismann *et al* in 1997 on mixtures of water and heavy water [1]. These experiments were performed with the neutron high-energy inverse time-of-flight spectrometer, VESUVIO, at the ISIS facility of CCLRC, for which the incident neutron energy is large, several eV, and the results were analysed as deep inelastic neutron scattering, DINS. They found that the intensity of the scattering from water compared with that from heavy water could be as much as 40% lower than expected for angles of scattering from 35° to 70°. Since then similar experiments have been repeated with the same unique instrument on metal hydrides [2], polystyrene and benzene [3] and Formvar [4], and although the experiments differ in detail they all show significantly lower scattering from the hydrogen than predicted from conventional neutron scattering theory. The authors initially explained this decrease as arising from quantum entanglement effects occurring on a short timescale, and this suggestion was supported by theoretical work of Karlsson and Lovesey [5]. One of us [6] showed that the observed scattering in both the experiment and the theory of Karlsson and Lovesey was inconsistent with the first moment sum rule and furthermore that quantum entanglement should not influence the first moment sum rule at high energies. This result has been supported by the theoretical work of Sugimoto *et al* [7]. The conclusion was that the experiments were flawed. The experiments were re-examined in detail and a paper published by one of us suggesting that the experimental results did show the anomaly [8]. As a result it was then proposed that the results might arise if there was a failure of the Born–Oppenheimer approximation because of an interaction between the hydrogen motion and an electronic mode [9, 10]. As yet the experiments have not shown direct evidence of scattering from an electronic mode.

All of these experiments were performed on one instrument, VESUVIO, which is an inverse time-of-flight instrument, meaning that the time-of-flight technique is used to determine the incident neutron energy and the scattered neutron energy is determined by the energy of absorption in a gold or uranium foil. Other experiments have now been performed using high energy total scattering that showed no effect on the intensity [11], with high energy electrons that showed an effect similar to that observed with neutrons [4] and with keV neutrons using a very high energy instrument, similar in principle to VESUVIO. In this last case the original authors reported that the results showed no effect [12] but a reanalysis of their data [13] showed that there was a decrease in the intensity similar to that reported with the VESUVIO spectrometer. This brief history of the anomalous decrease in the scattered intensity from hydrogen shows that an effect is observed which has not yet been explained in terms of current theoretical models and for which as yet there has been no satisfactory explanation of why the experimental results are intrinsically incorrect or have been analysed incorrectly.

In this paper we mostly re-examine the analysis of the data. Initially, in section 2, the zeroth and first moments of the scattering are discussed using the impulse approximation to describe the scattering. The experiments are performed using time-of-flight techniques with the detectors positioned at constant scattering angles. Because hydrogen has almost the same mass as the neutron, however, we shall show that the zeroth and hence the first moment of the Van Hove scattering function $S(Q, \omega)$ are always divergent when the integral is performed for scans made along the neutron trajectory with the scattering angle held constant. This divergence of the moment at large incident neutron energies has not been observed in detail, but it demonstrates that if the impulse approximation fails, then the necessary corrections to obtain the scattering cannot be made satisfactorily to compare with calculations. In section 2

a proposal is put forward about the way in which we should proceed so that the results can be compared with the theoretical calculations.

The alternative approach is to assume the validity of the impulse approximation. The data can then be corrected to compare with theory by applying the conversion from a time-of-flight spectrum to an energy spectrum. This is a well known geometrical transformation. It must also be corrected for the variation of the incident neutron flux with energy. Finally, corrections for the different slopes of the hydrogen scattering and the neutron trajectory must be made by using the impulse approximation for the scattering and then either using the Waller–Froman, J , factor [6], or fitting the parameters in the impulse approximation directly to the experimental data [8]. In the former approach the correction factors are calculated at the peak of the impulse approximation and then held constant when applied to the experimental data such as the width and intensity of the scattering. This approach is discussed in section 3 and we shall call it the conventional approach. The latter approach uses y -scaling and fits the parameters of the impulse approximation directly to the data. It is more accurate, especially when the width of the scattering becomes wide, and we shall call it the y -scaling technique. It is discussed in section 4. In these sections we illustrate the approaches to analysing the measurements of the scattering using data from various polythene and lead foils. These data have not been published before and were taken because both materials can be obtained as foils and so a container is not needed, which would also scatter neutrons when the sample was inserted into VESUVIO to perform the measurements. The experiments were performed using the Au foils to filter the scattered neutrons and the sample was kept at room temperature while the other parameters of the experiment were similar to those employed in earlier experiments using VESUVIO [7]. The results are discussed and summarized in a final section, where it is suggested that agreement between experiment and conventional theory can only be obtained if there is a systematic error in the experiments, which, we suggest, may arise from an unexpected energy dependence of the monitor detectors.

2. Scattering at large incident energies

Neutron scattering is governed by the equations of conservation of energy and momentum, which can be written in a conventional notation for an isotropic system as

$$\hbar\omega = \frac{\hbar^2}{2m}(k_0^2 - k_1^2) = E_0 - E_1 \quad (1)$$

and

$$Q^2 = k_0^2 + k_1^2 - 2k_0k_1 \cos \phi \quad (2)$$

where ϕ is the scattering angle between the incident and scattered beams, subscript 0 refers to the incident neutrons and 1 to the scattered neutrons. The standard expression for the count rate in each time channel of width dt for an inverse time-of-flight instrument is for a monatomic system with a neutron scattering length of b given by [14]

$$C(t) dt = AE_0 I(E_0) N |b|^2 S(Q, \omega) dt \quad (3)$$

where A is a constant, $I(E_0)$ is the incident flux, that varies with the incident neutron energy, N is the number of scattering centres and $S(Q, \omega)$ is the Van Hove scattering function. For a deep inelastic scattering experiment from an isotropic monatomic material, assuming that the impulse approximation is valid, the maximum in the scattering occurs when

$$\hbar\omega = \frac{\hbar^2 Q^2}{2M}. \quad (4)$$

Substituting this expression into equations (1) and (2) and putting the mass, M , for hydrogen equal to the neutron mass, m , shows that the maximum in the DINS scattering will occur when

$$k_1 = k_0 \cos \phi. \quad (5)$$

This result implies that for scattering at 90° the incident neutron energy and the energy transfer become infinite. This property is unique for hydrogen and plays an important part in our development. In order to discuss the scattering at high energies we have used the impulse approximation [15]. A Gaussian form was used for the response function, and making use of y -scaling gives the response function in the elegant form

$$S(Q, \omega) = \frac{M}{Q\sqrt{2\pi w^2}} \exp(-y^2/2w^2) \quad (6)$$

where

$$y = \frac{M}{Q} \left(\hbar\omega - \frac{\hbar^2 Q^2}{2M} \right) \quad (7)$$

and w determines the scaled energy-width divided by the wavevector transfer. The scattering at large incident neutron energies will be considered using this approximation and the trajectory in $Q - \omega$ space given by a constant scattering angle experiment. The scattering function is then given by equations (6) and (7) and at large incident neutron energies y becomes

$$y = \frac{\hbar M^{1/2} (E_0 - E_1 - E_0 - E_1 + 2\sqrt{E_0 E_1} \cos \phi)}{(2(E_0 + E_1 - 2\sqrt{E_0 E_1} \cos \phi))^{1/2}}. \quad (8)$$

For large $E_0 \gg E_1$ the argument can be rewritten in a power series of E_1/E_0 and the leading term is

$$y^2 = \frac{4\hbar^2 M E_0 E_1 \cos^2 \phi}{2E_0} = 2\hbar^2 M E_1 \cos^2 \phi. \quad (9)$$

The importance of this result is that it is independent of the incident neutron energy, E_0 , so that the argument of the exponential does not change as the neutron energy increases. This is in contrast to the behaviour of a constant Q trajectory, where this factor increases with increasing energy and the scattering function decreases exponentially with the square of the incident energy transfer. Essentially at constant scattering angle the incident energy increases and both the energy transfer and the recoil energy increase, but in such a way that the argument of the exponential remains a constant. The scattering function has the form $S(Q, \omega) \approx C/\omega^{1/2}$ as $\omega \rightarrow \infty$, which, when integrated over all energy transfers, is divergent, at least in principle, for all scattering angles, and the first moment diverges more rapidly.

Although we have derived the divergence using the impulse approximation it does not depend on the details of that approximation and very general arguments can be used to suggest that $S(Q, \omega)$ must be given by the expressions given above at least approximately. The magnitude of the divergence depends on the square of the cosine of the angle of scattering so that the divergence becomes most significant as the scattering angle approaches 90° , on the scattered energy E_1 so that the divergence is larger for small scattered energies and on the width of the impulse approximation so that as w becomes larger the size of the divergence increases.

We have performed calculations using the impulse approximation to illustrate the effect of the divergence on $S(Q, \omega)$. The width of the impulse approximation was chosen to be appropriate for describing hydrogen in polythene and the other parameters chosen to simulate the conditions found in our experiments. The results of the calculations are plotted in figure 1 as a function of the time of flight of the incident neutrons

$$J(t) dt = E_0^{3/2} S(Q, \omega) dt. \quad (10)$$

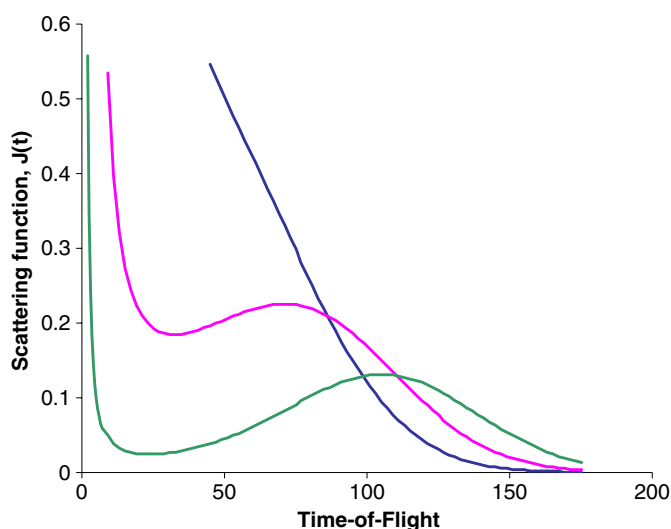


Figure 1. The spectra obtained as a function of time-of-flight $J(t)$ for constant scattering angles calculated using the impulse approximation. The three angles of scattering are 70° , 74° and 78° .

In a time-of-flight scan the divergence occurs at $t = 0$ rather than at infinite energy and figure 1 shows the divergence more clearly than if $S(Q, \omega)$ was plotted against an energy scale. The integral does not, however, depend on whether the integral is performed over all t or all E_0 . The results, in figure 1, show $J(t)$ for three different scattering angles. At a scattering angle of 70° and for lower scattering angles there is a peak at finite energy transfer which corresponds at least approximately to the impulse approximation peak and the divergence occurs only at very high incident neutron energies. The scattering can then be separated into two components. At a scattering angle of 74° the two peaks overlap and it becomes difficult to separate the scattering into two peaks. At still larger angles of 78° the divergence has overwhelmed the impulse approximation peak and there is only one peak in the spectrum.

In figure 2 we show for comparison the intensity that would be obtained in an experiment, $C(t)$, equation (3). This does not have a divergence at short times and there is a peak at finite times. This is because at short times $C(t)$ differs from $J(t)$ by $E_0^{-1/2} I(E_0)$, which decreases as $(E_0)^{-1.4}$ compared with $J(t)$. We conclude that the divergence in $J(t)$ is not immediately observable in experiments. It will require careful experiments at high scattering angles to clearly resolve the existence of the divergence and to make reliable and useful measurements of the scattering at high energies so that the moments of $S(Q, \omega)$ can be obtained from the experiment.

We have also performed calculations including the small differences in the mass of the neutron and the proton. These corrections alter the scattering function at very high incident energies and, in principle, remove the divergence. These effects only occur at exceedingly high incident energies and so do not change the main point of this section.

The above discussion shows that it is very difficult to obtain the moments of $S(Q, \omega)$ from the measurements at high energies. The correct approach to obtain these moments is to collect the scattering at a large number of different scattering angles and then to obtain a two-dimensional plot of $S(Q, \omega)$ by using a programme similar to MSLICE for powders. The resulting scattering function can then be integrated along a constant Q trajectory to obtain the integrated scattering function and that integral could be compared directly with the theory. To

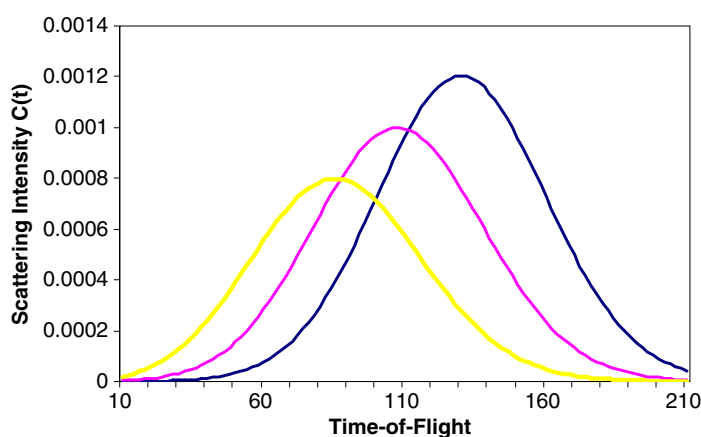


Figure 2. The calculated spectra $C(t)$ observed in an experiment at constant scattering angle using the impulse approximation. The three angles of scattering are 70° , 74° and 78° .

our knowledge this procedure has not as yet been attempted for any of the high energy transfer experiments on VESUVIO, and most likely requires improvements of the instrument to obtain satisfactory results.

3. The conventional data analysis

There are different ways of analysing the data from VESUVIO but they all give similar results and we shall divide them into two methods; the conventional approach and the y -scaling approach. The conventional approach proceeds by fitting the inelastic neutron scattering, $C(t)$, for a particular angle of scattering, by a Gaussian form in the time-of-flight scan for each different mass of the nucleus. The position, width and integrated intensity of each peak is obtained by a least squares fit. In order to compare these results with the theoretical estimates for the scattering function, $S(Q, \omega)$, whose intensities and widths are given by constant Q scans, two corrections must be applied [6]. Firstly the results should be converted from a time-of-flight distribution to an energy distribution. This requires multiplying the area by $(E_0)^{3/2}$ which is proportional to $\cos^{-3} \phi$. The other factor is the Waller–Froman Jacobian, which accounts for the change from scanning along the line of constant scattering angle to a line of constant Q . Calculation shows that at the centre of the scan this correction factor is $J = \cos^2 \phi$ [6] so that the conversion of the width in a time-of-flight scan to a width in a constant Q scan as a function of energy is proportional to $\cos^2 \phi / \cos^3 \phi = \cos^{-1} \phi$, while for the intensity a similar argument combined with a division by $I(E_0) \cos \phi$ gives the integrated scattering for a constant Q scan.

The scattering from hydrogen in polythene was measured using VESUVIO with gold filters and a very similar configuration to that employed in earlier experiments. The incident flux was determined from the measured monitor counts by assuming the monitor detectors had an efficiency that is inversely proportional to the incident neutron velocity. The result was that the incident flux varied as $I(E_0) = E_0^{-0.9 \pm 0.05}$, in agreement with the results from earlier measurements [8]. The sample was made from sheets of 1 mm of lead and 0.15 mm of polythene held at room temperature. In figure 3 we show the results deduced for the integral over the scattering function in a constant Q scan as a function of scattering angle when divided by the scattering from the Pb foil. From the thickness of both sheets and a measurement of the density of the polythene, the impulse approximation leads to the expectation that the ratio of

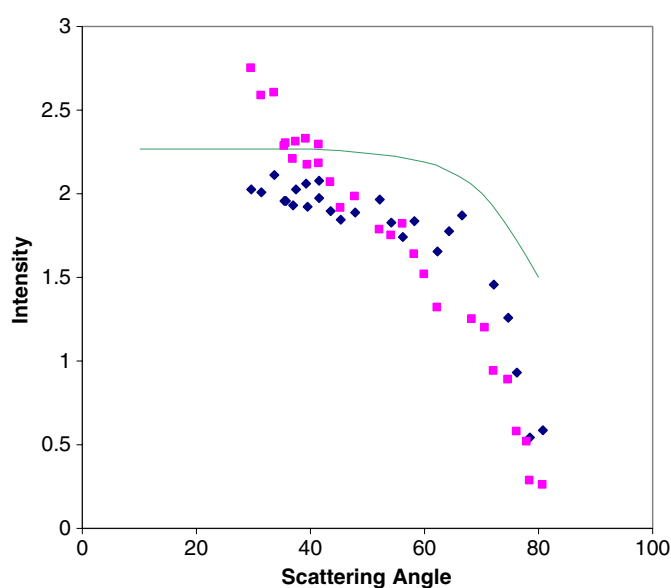


Figure 3. The ratio of the intensity of the scattering from the H and the Pb peaks obtained using the conventional analysis. The calculations were made assuming two Gaussian peaks (squares) and one Gaussian and one Lorentzian (diamonds). The continuous curve is obtained from simulated data by analysis using two Gaussian curves.

the intensity of the hydrogen peak to the lead and carbon peak should be 2.27 ± 0.05 , where the error arises largely from uncertainty in the density. In figure 3 we show two sets of results; one was calculated exactly as described above while for the other set the Pb peak was taken as having a Lorentzian form rather than a Gaussian form. The errors can be assessed mostly easily from the scatter of the points in figure 3. The latter model gave a better fit to the data and arises because the width of the Pb peak is caused largely by the experimental resolution rather than the intrinsic Pb width. The results in figure 3 show that the experimental results decrease with increasing angle of scattering with a particularly rapid decrease occurring above a scattering angle of 60° . It is not surprising that at small angles the first method gave the larger value of the intensity because the H and Pb peaks then overlap and the change in the assumed line shape will tend to increase the intensity assigned to the H peak. Similar results were obtained for the width of the scattering, which when divided by the wavevector transfer was not constant but also decreased with increasing scattering angle.

We then simulated the scattered experimental intensity by using the impulse approximation, as described above, and used the conventional data analysis techniques to analyse the low energy peak in the simulated data. The results are shown in figure 3 by the continuous line and are constant up to a scattering angle of 55° but then show a marked decrease. We should note that the simulation and the data cannot be expected to give exactly the same results, because the simulated data have no background and the results are certainly dependent on the exact assumptions made about the background subtraction in the analysis procedure. Similar results were also obtained for the width of the scattering as those obtained for the intensities.

The decrease in the intensity at large scattering angles arises because the neutron scattering trajectory in ω - Q space and the energy of the scattering from hydrogen are very similar as shown by equation (4). The neutron scattering then occurs over a wide range of energies

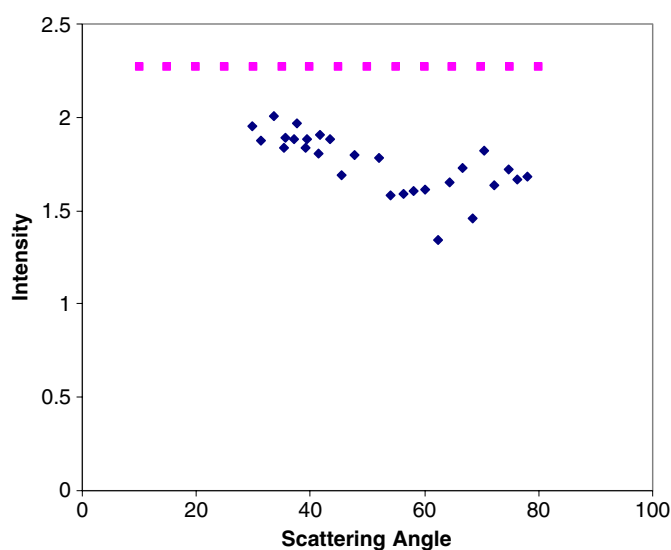


Figure 4. The y -scaling analysis of the intensity from the H peak divided by the intensity of the Pb peak as a function of the scattering angle. Squares correspond to the simulation and diamonds to the experimental data.

and a very much wider range of energies than would be obtained in a constant Q scan. The corresponding increase in the width and the intensity is large and cannot be adequately treated by the Waller–Froman correction calculated as a single number at the peak of the scattering. Both the experimental results and the simulations show that the decrease in the intensity of the scattering occurs for sufficiently large angles if the data is analysed with this form of the Waller–Froman correction.

4. y -scaling analysis

An alternative approach is to use the impulse approximation. The experimental spectra are fitted by adjusting the width parameter w and the overall intensity to provide a good description of each spectrum [8]. This procedure then obtains better correction factors than the single number Waller–Froman approach because it takes account of the non-linear behaviour of this correction as the scattering angle increases. The results of this procedure for the observed intensity ratios are shown in figure 4. The ratio of the intensities of the hydrogen peak to the lead peak decreases steadily as the scattering angle increases and is approximately 20% less than the behaviour expected from the impulse approximation.

The impulse approximation has also been used to calculate the scattering from hydrogen using the y -scaling approach of equations (6) and (7) with the width parameter chosen to describe the width determined experimentally for hydrogen in polythene. The simulated data was then analysed using the y -scaling technique. The results are shown in figure 4 as the scattering angle increases up to an angle of 75° . Not surprisingly the intensity is almost constant and shows that the simulations and this analysis procedure are consistent with one another.

5. Conclusions

The inelastic neutron scattering from hydrogen is anomalous because the energy and wavevector of the scattering both become infinite when the scattering angle is 90° . This has the effect that the zeroth and first moments of $S(Q, \omega)$, when obtained for a constant

scattering angle, are divergent for all angles. Although this conclusion has been proven by using the impulse approximation, it can also be obtained without recourse to the details of that approximation. The consequence is that the experimental results can at present only be analysed by assuming that the impulse approximation is valid. Then many of the experiments on hydrogen containing materials give intensities that are between 20% and 50% of those predicted; this suggests that the impulse approximation may be invalid for reasons that are not at present theoretically understood. In this paper we have suggested that the way to make progress is to measure the scattering for many closely spaced detectors and then to calculate the scattering function $S(Q, \omega)$ directly. Integration over this function would then give the moments of the scattering function without making assumptions about the form of the cross-section and assuming the validity of the impulse approximation.

If the scattering is described by the impulse approximation, we have shown that the conventional method of analysis using the conversion of a time-of-flight spectrum to an energy spectrum and then using the Waller–Froman J factor to convert from a constant angle to a constant Q spectrum by evaluating the J factor only at the peak of the scattering is applicable only for scattering angles below 50° . The experiments are also difficult at small angles of scattering because the spectra from both light and heavy atoms then overlap with one another. At higher angles the width of the scattering is sufficiently large that higher order terms in the Waller–Froman correction become appreciable. The approach of fitting the data with the impulse approximation using the y -scaling form of the cross-section overcomes the problems with the conventional approach and enables the data to be analysed up to a scattering angle of at least 65° . At higher angles the hydrogen peak appears at very short times of flight where current data are not reliable. Hence, at present, there is little information about the detailed form of $S(Q, \omega)$ in this energy range.

We have shown that the analysis of the experimental data from hydrogen is complex and requires considerable care. However, even with the y -scaling approach the experimental results for the hydrogen peak give a smaller intensity than expected theoretically by about 20%, and the decrease below the theory is becoming larger as the scattering angle increases. This decrease in the intensity is somewhat larger than the systematic errors, although there are improvements to the instrument which could reduce these errors further. At present we must either invoke a breakdown in conventional theory or systematic error in the experimental results.

The most likely cause of systematic error in the experiment is in the determination of the incident flux because the monitor detectors have not been independently calibrated at these energies. In order to investigate the decrease in the scattered intensity further we show results that would be obtained if the incident flux varied as $I(E_0) = D/(E_0)^{1.1}$ instead of the measured power law of 0.9. The results shown in figures 5 and 6 are now in very reasonable agreement with the theoretical values for scattering angles below 60° . However, at larger angles there are deviations in both figures 5 and 6. These are possibly consistent with a lower value of the exponent in the power law but may also result from difficulties in the analysis for large scattering angles. We have no reason to question our experimental determination of the exponent for the incident flux dependence unless the monitor detectors deviate from the $1/\sqrt{E_0}$ efficiency. The detectors have not to our knowledge been calibrated at the high energies used for experiments on hydrogen. Only much lower energies are accessed for experiments on heavier materials. This effect would not then significantly alter the results for heavier materials than hydrogen. We therefore consider this calibration should be done as soon as possible. In conclusion we are not yet certain whether for our polythene and lead samples or for other hydrogen containing samples the results are consistent with the expected theoretical values.

In conclusion, at present the analysis of the data can correctly be performed only by using the impulse approximation and suggests that there is an unexplained discrepancy if theory and

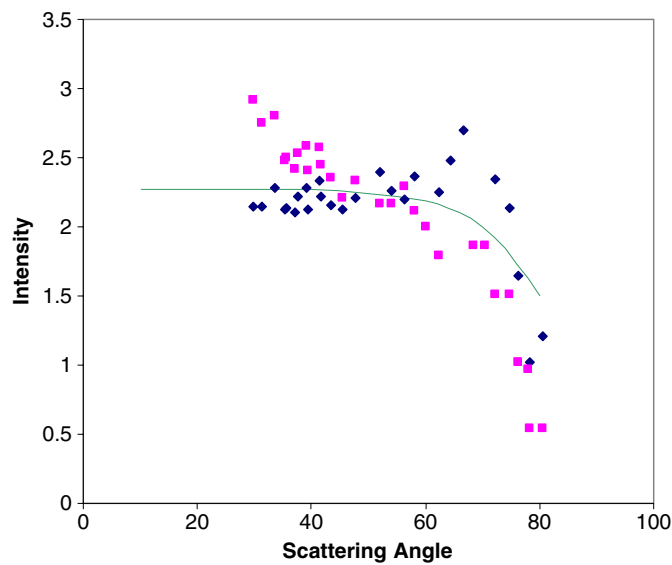


Figure 5. The same as figure 3 but with the incident flux arbitrarily changed as described in the text.

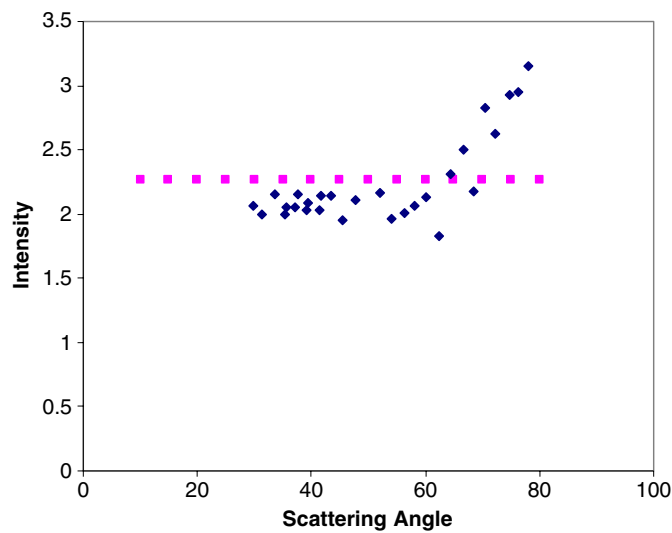


Figure 6. The same as figure 4 but with the changed incident flux as described in the text.

experiment are to agree. It has been pointed out that an analysis with fewer assumptions could be made if the scattering could be measured at a large number of different scattering angles and interpolated to obtain constant Q scans. This would allow a determination of whether the anomaly is due to a breakdown of the impulse approximation or a breakdown of conventional neutron scattering theory. An unexpected energy dependence of the efficiency of the monitor detectors could also account for the anomalies and further calibrations of the incident beam monitor are highly desirable.

Acknowledgments

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