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TOTAL CROSS SECTION MEASUREMENTS OF H₂O and ZrH₂ WITH VERY COLD NEUTRONS

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ABSTRACT

There is a growing interest in the development of very cold neutron moderators for future neutron sources since neutrons in this energy range (1 meV and lower) are expected to have advantages over conventional sources in a number of areas (ranging from fundamental physics studies to soft matter dynamics and applications requiring focusing optics). Unfortunately, existing models for interactions of neutrons with materials within this energy range are often inaccurate and consequently accurate modeling of source performance is presently not possible. We have initiated a research program whose goal is to measure total neutron cross section of various materials that may be considered for use in VCN moderator assemblies in order to provide some initial benchmarks against which developing VCN kernels may be compared. As a first step in this program, we have used the SANS beamline at the LENS facility at Indiana University to measure the total cross section of water and zirconium-hydride down to energies on the order of 100 μ eV. We will report on the experimental details of these measurements and discuss the anticipated capabilities of this instrument for this class of measurement.

1. Introduction

1.1. Very Cold Neutrons

Over the last several years, there has been an increasing interest developing in the production and utilization of so-called Very Cold Neutrons [1]. This regime has been loosely identified as neutrons with wavelengths of 2 nm or longer (energies on the order of 200 μ eV or less). It has been known for some time that for instruments capable of utilizing

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the full phase space of a cold neutron beam, the statistical precision of the measurement increases with phase space density and phase space density increases with decreasing spectral temperature. Interest in this regime is also motivated by the realization that focussing optics may become more effective as the wavelength increases. No source has yet been constructed around this idea, but there is a beam line covering this energy range available at the ILL, and the LENS facility is also capable of studying neutrons within this energy range. Utilizing such neutrons in the investigation of materials will require a number of developments in the design of sources and instruments as well as an increased understanding of how such neutrons interact with matter.

1.2. The need for data in the VCN regime

Contemporary design of neutron sources relies heavily on Monte Carlo modeling of neutron transport, but in trying to apply these techniques within the VCN regime one quickly must confront inadequacies in the information available in the relevant cross section libraries. In a recent review of ENDF and ACE library data for water and ZrH_x , significant deviations of the library models from available total cross-section data for even these comparatively well-studied materials at neutron energies below 1 meV [2]. In the case of H_2O , available models overestimate the total cross section by a factor of almost 2 for energies near 100meV, with even larger deviations seen at lower energies. Design of optimized sources is impossible if the available models are unable even to accurately describe the total cross section of the relevant materials in the energy range of interest.

In an effort to provide support to future work on VCN utilization and source design, we have initiated a program devoted to measuring total neutron cross sections for a number of candidate materials down to energies of 100 μ eV or lower using the unique capabilities of the LENS facility. In this report we provide data on the early measurements we have made on light water and zirconium hydride as we benchmark the capabilities of this facility for this sort of measurement. We discuss some of the aspects of the source that must be accounted for in the measurement and provide some indication of the range we expect to be able to cover in total cross section measurements with slight modifications to the source and procedures used in this initial test.

2. Experimental Details

We performed our measurements on the SANS beamline at the Low Energy Neutron Source (LENS) at Indiana University [3]. Unlike the vast majority of pulsed neutron sources, LENS has the unusual characteristic that its pulse parameters may be varied to the demands of a particular experiment. In our case, we reduced the pulse frequency from its normal 20Hz to 10 Hz to gain access to neutron energies below 0.16 meV by extending the frame overlap out to 39 μ eV. We also reduced the pulse width to 150 μ s to allow simultaneous access to energies as high as several eV in the same measurement. The proton beam had a peak current of 25mA and the protons were accelerated to 13MeV (total beam power roughly 500W). For these measurements, we removed the Be filter that is normally used to reduce the fast neutron flux at the sample position.

The sample was positioned at a distance of 8 m from the moderator and the transmitted neutrons were detected with a scintillation detector (GS-20 Li-loaded glass coupled to a 6236-01 Hamamatsu PMT) positioned roughly 50 cm from the sample. The detector was positioned with the transmitted beam tangent to the edge of the scintillator, so

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that the 2-D detector of the SANS instrument would detect any neutrons scattered at small angles in one direction. The signal in the 2-D detector was then used to estimate the small-angle scattering contribution to the signal in the primary detector.

Another feature of the LENS source is that it produces considerably fewer gamma and hard x-ray photons than typical spallation or reactor sources. Nevertheless, we discovered that, when viewing the source directly, the signal in our scintillation detector was dominated by photons (primarily decay photons from ^{28}Al) for times of flight greater than 56 ms (corresponding to neutron energies less than 0.12 meV). We estimate that with this configuration we had roughly 4 gammas reaching the sample for every neutron with an energy below 1eV. With an estimated relative gamma sensitivity of roughly 5×10^{-4} this produced an unacceptable background at the low energy end of our measurement. In order to extend our measurements into the desired energy range (on the order of 0.1 meV), we reduced this background by inserting a room-temperature lead filter (5cm thick) into the flight path near the source. This reduced the background at long times of flight by roughly an order of magnitude and had the added advantage of reducing the peak neutron count rate by roughly a factor of three (thereby reducing the deadtime correction needed for neutron energies near 50meV to less than 6%). Unfortunately, it also further reduced the flux available at the lowest neutron energies, and future measurements would benefit from using a detector with less gamma sensitivity and/or a lead filter held at liquid nitrogen temperature. In these measurements, the low energy limit is still determined by this gamma background even with the presence of the Pb filter.

We had attempted to reduce the spectral temperature of the standard LENS moderator by doping it with a small amount of Ar to suppress the methane phase transition at 20K. Unfortunately, we were not able to get enough Ar into the methane on this run and there were also problems loading the moderator (it ended up being only about 70% full). Consequently, the intensity below 1 meV was roughly a factor of three lower for these measurements than is typically available from the LENS moderator. The impact of both this faulty fill and the presence of the Pb filter on the neutron spectrum used for these measurements is shown below in figure 1. Overall, we anticipate that with an improved detector/filter combination and standard LENS moderator we could extend the range of total cross-section measurements on this instrument to close to 50 μeV with measurement times of less than 10 hours.

The ZrH_2 samples were 325 mesh powder samples loaded into the cells and shaken gently until there were no visible cracks or gaps in the filling of the circular cells. The packing fraction was determined by weighing the powder in the cells after the measurement (and confirming this weight by weighing the cells themselves before and after the removal of the powder; these two results were mutually consistent to within 1.5%) and dividing this by the known dimensions of the cell. The powders were loaded (and removed) in an inert-atmosphere glove box. The powder was obtained from ALFA AESAR and had a nominal purity of 99.8%. For the water samples we used water obtained from a commercial lab purification system (Millipore Direct-Q system) and this was loaded directly into the sample cell with a pipet.

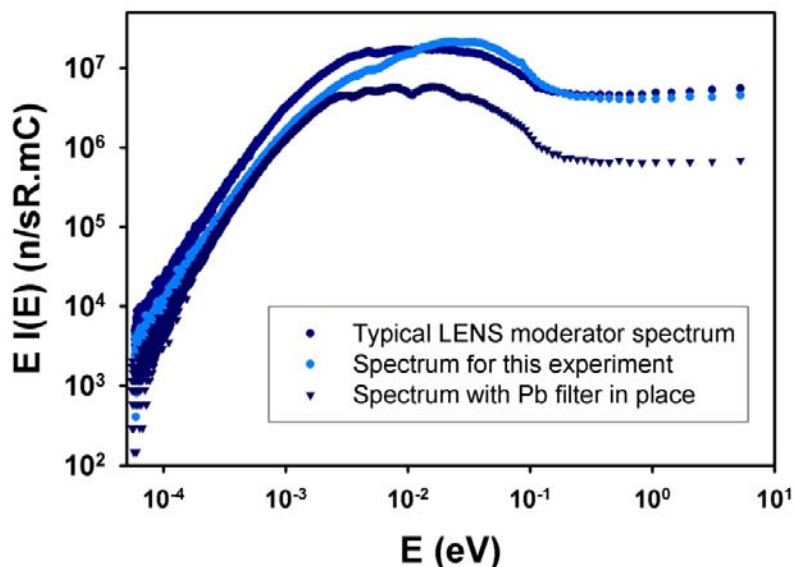


Fig. 1 Spectral intensity from the LENS moderator under various circumstances, including the spectrum used for these measurements, with a 5 cm lead filter.

3. Results

3.1 Water Total Cross-section

We first consider our measurements of normal water. Two different thicknesses were measured as a check on our ability to adequately account for sample thickness and to correct for background and other systematic errors. The samples were held in quartz banjo cells of the type that are typically used for SANS measurements, and the beam was defined by a 15 mm wide pin hole cut into a 1.3 cm thick piece of BN held just in front of the sample. The alignment of both the sample and the detector with the beam was confirmed using a laser illumination device integrated into the final stages of the SANS instrument collimation column. The results of these measurements (using samples of 1 and 5 mm thickness) are shown below. The sample-in measurements were made in 2 hours and 4.5 hours respectively for the 1 mm and 5 mm thicknesses, with a total of 8 hours taken for the empty cell and background measurements.

The measurements on the two different thicknesses of water are mutually consistent, except for discrepancies slightly larger than the statistical error bars for energies above 1 eV. At energies below 200 μ eV, the statistical uncertainty becomes significantly larger for the thicker sample. These measurements are also in good agreement with previous measurements on this material [2,4], suggesting that for homogeneous liquid samples our experimental arrangement is adequate. We also note that these data suggest that measurements of total cross section of hydrogenous materials down to 100 μ eV may be obtained with roughly 10% statistical accuracy in several hours with this apparatus. By using longer pulse widths (sacrificing simultaneous access to energies above several hundred meV) and increasing the frequency to 15Hz, the data collection time, or the size of the statistical uncertainties could be reduced while still reaching below 100 μ eV.

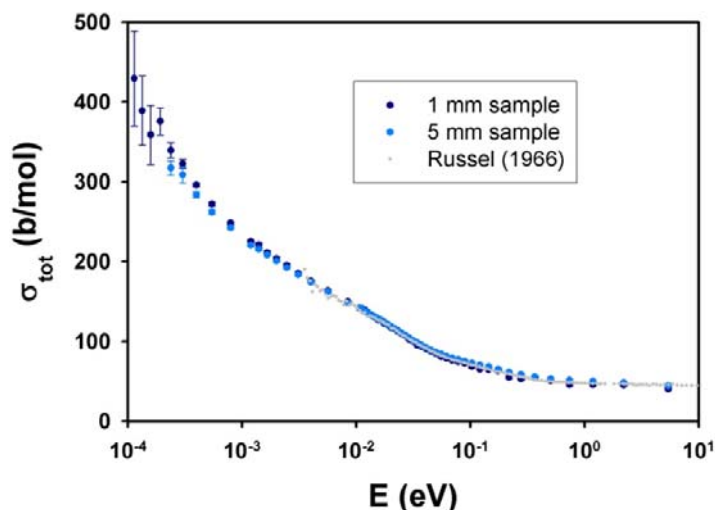


Fig. 2 Total cross section of water measured on samples of two different thicknesses. The two measurement agree with each other within their mutual errorbars except for four measurement above 100 meV.

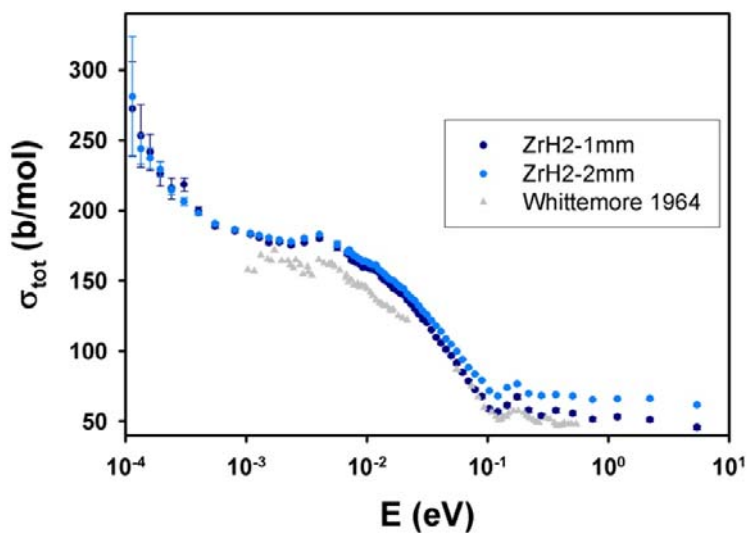


Fig. 3 Total cross section of ZrH_2 measured on samples of two different thicknesses, and compared to results published earlier by Whitmore. The LENS data from the 1mm sample could be brought into reasonable agreement with Whitmore's data by assuming a packing fraction or sample thickness roughly 10% greater than the measured values. The discrepancies seen between the two thicknesses could not be resolved by any reasonable variation in the parameters of the measurement, and suggest some systematic error (also on the order of 10%) at energies above 30 meV for this sample that remains unresolved.

3.1 ZrH_2 Total Cross-section

The ZrH_2 samples present a slightly different picture. Although with this material we also see that measurements over the full range of energy (100 μ eV to over 1eV) may be made with uncertainties on the order of 10 to 15 %, in this case we see clear evidence for systematic errors in comparing the measurements at the two different thickness to each other and when compared to earlier measurements of others [5]. Although it is certainly true that this material exhibits much more small angle scattering than water, the measurements in the SANS detector suggest that this contribution only approaches a few percent of the direct signal at the lowest energies. It seems most likely that the source of the systematic error is some sort of sample-dependent background at relatively short times. Unfortunately, the most obvious candidate (gamma radiation short lived isotopes in the source) would seem to be excluded by the presence of the thick Pb filter and the relatively small thickness of the samples.

4. Conclusions

We have demonstrated the ability to measure neutron total cross sections on the SANS instrument at LENS over an energy range from roughly 100 μ eV to over 1.0 eV in a single setup of the accelerator and instrument. We have demonstrated this ability with measurements on two materials that could play a role in the design of future neutron sources H_2O and ZrH_2 (although deuterated versions of these materials would be more likely to be useful in future VCN sources). In the case of the powder ZrH_2 , we see systematic errors in our measurements on the order of 10%, particularly for energies above 100 meV. Inhomogeneities in the H concentration produced significant small angle scattering in this material, and this may have something to do with these systematic errors, although our best estimates for the contribution from this background suggest that it is not large enough to account for the observations. With modest modifications to the experimental apparatus, and the source characteristics we believe that it should be possible to extend total cross-section measurements on these and other materials of interest to VCN source development to roughly energies as low as 50 μ eV in the future.

5. Acknowledgements

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5. References

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