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A neutron spin filter based on transmission through nuclear-spin-polarized ³He gas has been applied to polarization analysis of small angle neutron scattering (SANS). Such spin filters, which are based on the large spin dependence of the absorption of neutrons by ³He, make SANS polarization analysis possible because of their large angular acceptance. In the present experiment, a ³He-based analyzer was employed to separate nuclear scattering into its coherent and spin-incoherent components. Polarized ³He analyzers were prepared by two different optical pumping methods and installed on the NG3 SANS instrument at the NIST Center for Neutron Research (NCNR). Measurements were taken on cellophane tape and silica gel, for which the scattering is almost completely incoherent and coherent, respectively, and on a combined sample. For the combined sample, separation of the coherent part from the incoherent part was successfully demonstrated using polarization analysis.

Introduction

Polarization analysis is a powerful technique in neutron scattering experiments, since it has the capability to unambiguously distinguish coherent nuclear scattering from spin-incoherent nuclear scattering, or to identify magnetic (neutron-electron) scattering from nuclear scattering (Moon *et al.*, 1969). A standard technique for polarizing thermal neutrons, and for analyzing the polarization state of scattered thermal neutrons, is to use magnetic Bragg reflection from a suitable crystal, while for longer wavelength neutrons a multilayer or supermirror can be employed (Williams, 1988). However, for small angle neutron scattering (SANS) instruments the angular acceptance of these devices is inadequate for the scattered beam, which can have a divergence of 20º. The analyzers employed in the present work rely on the large spin dependence of the transmission of neutrons by ³He. ³He-based polarization analyzers are more practical for SANS instruments because of their broad angular acceptance, which is limited only by the solid angle subtended by the ³He cell. For the relatively small diameter beams typically employed on SANS instruments the complete pattern of scattered neutrons can be readily analyzed. In addition, the glass ³He cell itself contributes little scattering, unlike mirror polarization devices.

The use of polarization analysis to separate coherent nuclear scattering from spin-incoherent nuclear scattering has been applied to a variety of materials (Gabrys *et al.*, 1999), such as polymers (Gabrys

et al., 1993), the plastic crystal C_2Cl_6 (Gerlach *et al.*, 1982), and heavy water (Dore *et al.*, 1976). The goal of the present experiment was to extend this technique to the divergent beams encountered in SANS using a ³He analyzer and known scattering samples. Measurements were taken on two samples: six layers of cellophane tape, for which the scattering is dominated by the spin-incoherent scattering of hydrogen, and a silica gel, for which the scattering is primarily coherent nuclear scattering. A known mix of coherent and spin-incoherent cross sections was then achieved by placing both of these samples at the sample position. As shown in Fig. 1, the key elements of the apparatus are a supermirror polarizer, spin flipper, sample, ³He analyzer, solenoid, and SANS detector.
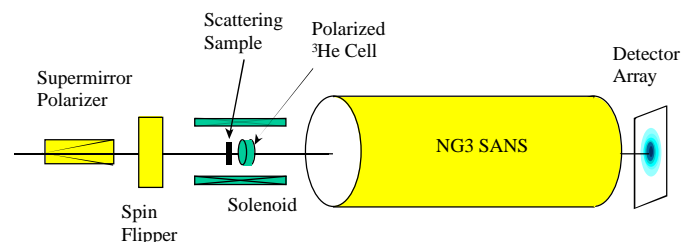

Figure 1
Schematic diagram of the apparatus.

Polarization analysis

Polarization analysis makes it possible to extract the amounts of coherent and incoherent scattering in a given sample. This capability is rooted in the non-zero probability of a neutron spin-flip for spin-incoherent nuclear scattering in contrast with zero probability of a spin-flip for coherent nuclear scattering. Hence we assume that the total scattering, N_T, falls into two components: N_1, for which the scattering has some known probability of spin-flip scattering (incoherent), and N_2 (where $N_T = N_1 + N_2$), for which the scattering is completely non-spin-flip (coherent). This approach yields results that can also be applied to the separation of magnetic from nuclear scattering. The analysis sequence is to 1) correct the raw data for an imperfect spin flipper, 2) correct for imperfections in the polarizer and analyzer so as to extract the amounts of non-spin-flip and spin-flip scattering (labelled N_{NSF} and N_{SF} respectively), and 3) determine the components N_1 and N_2.

In the first step, data obtained with the spin flipper energized is corrected for the imperfect flipping efficiency. The corrected number of counts when the spin flipper is energized is given by

$$N_{on}^{corr} = \frac{1}{e}\left[N_{on} - (1-e)N_{off} \right] \qquad (1)$$

where N_{on} and N_{off} refer to the number of neutrons detected (either scattered or transmitted) with the spin flipper on (energized) and off, respectively, and e is the fraction of neutrons actually flipped by the spin flipper. Raw data are corrected using Eq. (1) before use in the analysis below.

The second step is correcting for imperfection in the polarizer and analyzer. The polarization of the beam incident on the sample is given by $P=(N_+ - N_-)/N$, where N_+ (N_-) is the number of incident neutrons with spin parallel (antiparallel) to the ³He, and

$N = N_+ + N_-$. Similarly, the quality of the ^{3}He analyzer is described by the analyzer transmission asymmetry, A_a, given by $A_a = (T_+ - T_-)/(T_+ + T_-)$, where T_+ (T_-) is the transmission of the ^{3}He analyzer for neutron spin parallel (antiparallel) to the ^{3}He polarization. The analyzer flipping ratio, given by $F = T_+ / T_-$, can be obtained from A_a using the relationship $F = (1 + A_a)/(1 - A_a)$. We define the observed number of neutrons scattered by the sample for the incident neutron beam polarization parallel (antiparallel) to the ^{3}He polarization to be N_p (N_a), and the observed asymmetry in the neutrons scattered by the sample, A_s, to be

$$A_s = \frac{N_p - N_a}{N_p + N_a}. \qquad (2)$$

(Note that depending on the relative orientation of the ^{3}He polarization and the neutron polarization exiting the polarizer, either N_p or N_a is corrected using Eq. (1).) The number of neutrons scattered by the sample with no spin-flip is given by N_{NSF} and the number scattered with a spin-flip is given by N_{SF}, where $N = N_{NSF} + N_{SF}$. N_p and N_a are related to N_{NSF} and N_{SF} by

$$N_p = \frac{N_+}{N}(N_{NSF}T_+ + N_{SF}T_-) + \frac{N_-}{N}(N_{NSF}T_- + N_{SF}T_+) \qquad (3a)$$

and

$$N_a = \frac{N_+}{N}(N_{NSF}T_- + N_{SF}T_+) + \frac{N_-}{N}(N_{NSF}T_+ + N_{SF}T_-). \qquad (3b)$$

Solving for N_{NSF} and N_{SF} yields

$$N_{NSF} = N_T \frac{A_0 + A_s}{2A_0} \qquad (4a)$$

and

$$N_{SF} = N_T \frac{A_0 - A_s}{2A_0}, \qquad (4b)$$

where $A_0 = P A_a$ is the overall analyzing power of the polarizer-analyzer combination. Hence N_{NSF} and N_{SF} can be determined by measuring A_s and A_0. By applying Eq. (3) to transmission ($N_{NSF} = N$ and $N_{SF} = 0$) we find that A_0 is given by

$$A_0 = \frac{(N_+T_+ + N_-T_-) - (N_+T_- + N_-T_+)}{(N_+T_+ + N_-T_-) + (N_+T_- + N_-T_+)} = \frac{N_p^0 - N_a^0}{N_p^0 + N_a^0}. \qquad (5)$$

where N_p^0 (N_a^0) is the sample-out neutron transmission for the incident neutron beam polarization parallel (antiparallel) to the ^{3}He polarization. Small differences in the effective analyzing power of the system for scattering as compared to transmission are discussed further below.

The amount of scattering in the two groups can now be determined. If we assume that the probability of a spin-flip for neutrons in the component N_1 is given by p, we find

$$N_1 = \frac{N_{SF}}{p} \qquad (6)$$

For the special case of spin-incoherent scattering for the N_1 component, $\frac{1}{3}$ of the scattering is non-spin-flip and $\frac{2}{3}$ is spin-flip (Moon *et al.*, 1969; Williams, 1988). Hence $p = \frac{2}{3}$, which yields $N_I = N_1 = \frac{3}{2} N_{SF}$ and $N_C = N_2 = N_{NSF} - \frac{1}{2} N_{SF}$, where N_I and N_C are the incoherent and coherent components of the total scattering, respectively. For a sample that exhibits multiple scattering, the value of p will deviate from $\frac{2}{3}$, but can be calculated if the attenuation of the beam due to the spin-incoherent component of the scattering is known. Based on the measured value of 0.833 ± 0.005 for the transmission of the tape sample, we used $p = 0.630$ in our analysis. Note that for tape, pure incoherent scattering is assumed for the calculation of the value of p.

Apparatus

The principles and apparatus for the polarized ^{3}He analyzers that we employed are described elsewhere (Jones *et al.*, 1999). Here we discuss only the salient features that are relevant to this experiment. Separate experiments were performed with each of two ^{3}He analyzers, one prepared by spin-exchange optical pumping and the other by metastability-exchange optical pumping. Throughout this paper we present the data obtained using the spin-exchange analyzer; similar results were obtained with the metastability-exchange analyzer. The analyzers were prepared using apparatus at the NCNR and transported using a portable solenoid to the NG3 SANS apparatus. As shown in Fig. 1, an end-compensated solenoid, 30 cm diameter and 60 cm long, immersed the sample and the ^{3}He cell in an axial holding field of 5 mT (50 G). The solenoid provided a uniform magnetic field so that the ^{3}He would not relax due to magnetic field gradients. The ^{3}He polarization underwent a slow exponential decay during the experiment due to relaxation on the cell walls, producing a time-varying for A_0. The spin-exchange (metastability-exchange) cell exhibited a relaxation time of 120 (16) hours. For the strong scatterers employed in this work the complete set of data for each sample was acquired in one hour, hence an average value for A_0 was adequate for a given set of data.

The value of A_0 is also dependent on the path length of any given scattered neutron through the polarized ^{3}He gas, which in turn is dependent on the cell geometry and the scattering angle. Because the spin-exchange method is most efficient at ^{3}He pressures of a few bar or higher, spin-exchange cells must be constructed with rounded features for structural stability. For this work, the 5.6-cm ID spin-exchange cell was a cylinder with rounded corners and an average length of 1.4 cm, and was filled to a pressure of 3.5 bar. Because the sample was immediately in front of this short cell the scattered neutron beam sampled only the central 2 cm of the cylinder, over which the path length is not expected to vary by more than 10%. For the metastability-exchange method high ^{3}He pressures are not generally required, hence flat-windowed cells can be used. In this case, the path length for any given scattering angle deviates slightly (1.5% for 10º) from that of transmitted neutrons. In this work, the metastability-exchange cell was a long right circular cylinder, 4.4 cm ID and 10 cm long, and was filled with a mixture of 25% ^{3}He and 75% ^{4}He at a total pressure of 2 bar (Jones *et al.*, 1999).

For the spin-exchange cell the value of A_0 was 0.78 for the data shown here. Although it is not necessary to know P and A_a individually, we note that these values were $P = 0.88$ and $A_a = 0.89$ ($F = 17$). For reference, the transmissions through the ^{3}He analyzer were $T_+ = 0.17$ and $T_- = 0.01$, and the ^{3}He polarization was 46%. If the analyzer had instead been operated as a polarizer, it would have produced 89% neutron polarization with 9% absolute transmission of an incident unpolarized beam.

The NG3 SANS apparatus is equipped with a supermirror to polarize the well-collimated incident neutron beam perpendicular to the beam direction. A precession coil spin-flipper is located after the polarizer to invert this polarization when desired. The magnitude of the neutron polarization was preserved in an adiabatic rotation from vertical to axial at the entrance to the ^{3}He analyzer. An average neutron wavelength of 0.80 nm (8.0 Å) was employed, with wavelength spread of 15% full width at half-maximum. Each analyzer cell was located at the center of the solenoid, about 1 cm behind the sample. The 64 cm by 64 cm NG3 detector was located at the closest possible position, 1.88 m from the sample.

The spin flipper efficiency was determined to be $e = 0.96$ using a second supermirror to analyze the beam, but we are not confident of this value for two reasons: 1) this measurement was not done until several months after the experiment, and 2) extracting e requires more knowledge of the second supermirror than is presently available. Since e is expected to be 0.99 for the wavelength spread of 15% (Hayter, 1978), we used an intermediate value of $e = 0.98$, and estimate the uncertainty to be 0.02.
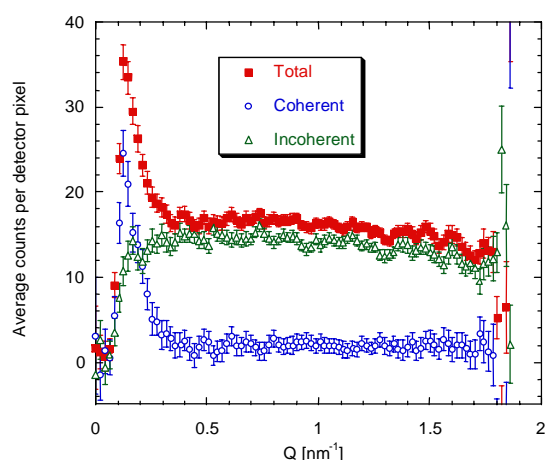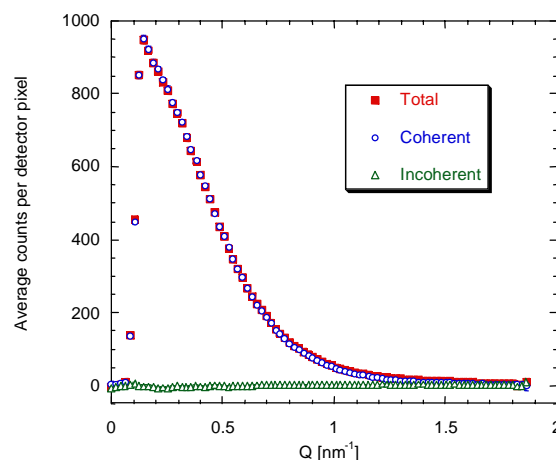

Figure 3
Total scattering from the silica sample (filled squares), and for the coherent (open circles) and spin-incoherent (open triangles) scattering determined from polarization analysis.

Results and Discussion

Figs. 2 and 3 show the azimuthally-averaged results for the total background-corrected scattering and the coherent and spin-incoherent components determined from the analysis described in Sec. 2. The tape data are clearly dominated by spin-incoherent scattering, whose intensity should be Q ndependent until the scattering drops behind the beam stop at small Q. The silica data, on the other hand, show almost all coherent scattering. The rise in the coherent scattering for the tape sample at low Q is not fully understood, but may be due to scattering from the aluminum sample holder. The error bars reflect the uncertainty due to counting statistics.

Scattering from the combined sample should reveal a small component of incoherent scattering equal to that observed for the tape sample. More precisely, the extracted components for the combined sample should correspond to the transmission-weighted sum of the extracted components for the individual silica and tape samples. We define N_C^T, N_C^S, N_C^{S+T} to be the extracted coherent components for the tape, silica, and combined samples, respectively, and N_I^T, N_I^S, N_I^{S+T} to be the extracted spin-incoherent components. In Fig. 4 we compare N_I^{S+T} to $N_I^T T_S + N_I^S T_T$, and N_C^{S+T} to $N_C^T T_S + N_C^S T_T$, where $T_T = 0.833 \pm 0.005$ and $T_S = 0.885 \pm 0.006$ are the transmission values for the tape and silica samples, respectively. Because of the large scattering from the silica sample, Fig. 4 is an expanded plot that highlights the extracted incoherent scattering, which is in reasonably good agreement with the weighted sums. The effect of uncertainties in the measured values of A_0 and e affects both the extracted incoherent component for the combined sample and the summed incoherent data in the same way, with the maximum effect occurring at the lowest Q values (e.g. ± 15% at $Q \approx 0.3$).

Figure 2
Total scattering from the tape sample (filled squares), and the coherent (open circles) and spin-incoherent (open triangles) scattering determined from polarization analysis.
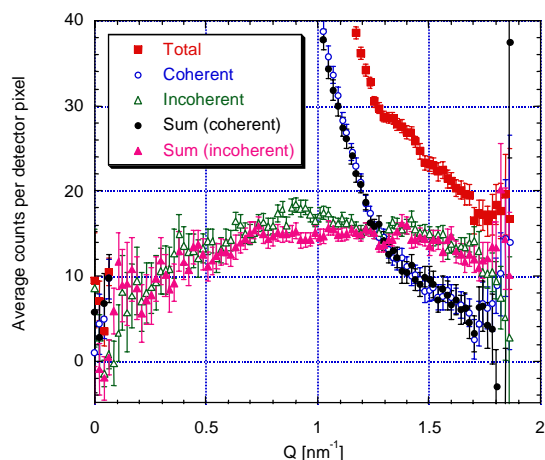
Figure 4
Total scattering from the combined (silica+tape) sample (filled squares), and the coherent (open circles) and spin-incoherent (open triangles) scattering determined from polarization analysis. For comparison, the transmission-weighted sums of the coherent and incoherent components for the individual silica and tape samples are shown by filled circles and filled triangles, respectively.

Recent measurements have shown that there is substantial inelastic scattering for cold neutrons incident upon room temperature samples of water or polymers (Ghosh & Rennie, 1999; Ghosh & Rennie, 1990). Neutrons that are inelastically scattered to shorter wavelengths experience a higher analyzer transmission asymmetry. In addition, they experience higher transmission through the analyzer, but also a lower probability of detection by the NG3 ^{3}He detectors (Glinka *et al.*, 1998). Fortuitously, the extracted component of incoherent scattering for the tape sample is relatively insensitive to the presence of inelastic scattering. We estimate that inelastic effects in the tape sample could raise the observed average value of 0.88 for the fraction of incoherent scattering (Fig. 2) by 5% - 10%.

In summary, we have employed a ^{3}He spin filter to perform polarization analysis on a SANS apparatus. Using this method, we have separated coherent scattering from spin-incoherent scattering in a combined sample of cellophane tape and silica. The success of this experiment opens up the possibility of separating magnetic from nuclear scattering in SANS experiments, which will be of great utility in the study of magnetic materials. The need to cool and/or apply a strong magnetic field to the samples for such experiments will introduce two additional technical issues in the application of neutron spin filters. First, the analyzer cell will need to be separated from the sample by larger distances, which will require larger diameter ^{3}He cells. We are developing larger cells for both optical pumping methods, and in a recent SANS experiment we successfully employed a 9.5-cm ID metastability-exchange cell with 34% ^{3}He polarization and a relaxation time of 32 h. Second, the analyzer solenoid will need to be shielded from the applied magnetic field (Heil *et al.*, 1999). We will be developing shielded ^{3}He analyzers for such applications.

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