Scattering of 64 eV to 3 keV Neutrons from Polyethylene and Graphite and the Coherence Length Problem

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We measured the neutron scattering intensity ratios from polyethylene (CH_2) relative to graphite (C) at several discrete final energies, of narrow widths (~3 eV) between 64 eV and 3 keV. The final energies were selected using a ^{238}U filter. This experiment was carried out to search for any anomaly in the *n*-*p* scattered intensities from CH_2 caused by the neutron coherence length. The scattered intensity ratios were found to conform to conventional expectations and no anomaly was observed.

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A long series of papers was published since 1997 concerning an anomalous decrease of the scattering intensity of neutrons from protons of hydrogen-containing compounds. In the first Letter reporting such *n*-*p* scattering anomalies [1], a H_2O sample was used, and the *n*-scattering intensity was compared to that of a D_2O/H_2O mixture. It was reported that at epithermal energies in the 10–200 eV range, the neutron scattering cross section from H, at high momentum transfers, drops by about 40% compared to conventional values. The same technique was used to study other H-containing samples, Nb, Yb, and Pd hydrides [2], acetone (C_3OH_6) [3], polymers such as polystyrene and formvar ($C_8H_{14}O_2$) [4], and other cases mentioned in Refs. [2–4]. In those cases, which were mostly studied at room temperature, the anomalous effect was found to be dependent on the scattering angle, dropping more with higher momentum transfer. The results of those experiments, which are not in line with conventional expectations, were considered to provide a strong and direct evidence of quantum entanglement of protons, for very short durations of the order of $\sim 10^{-16}$ sec, in condensed matter. In all the above studies, the neutron Compton scattering (NCS) technique was used in conjunction with the VESUVIO spectrometer (formerly known as EVS) at Rutherford Laboratory, UK.

However, in a very recent Letter [5], using a different experimental system, and employing a higher neutron final energy of 24.3 keV, the ratio of the neutron scattering intensities from H_2O and D_2O was measured, but no anomalous drop in the *n*-*p* scattering intensity was observed and the scattering cross section was found to be normal. Following this recent work, three main questions were raised claiming that the results of Ref. [5] are not necessarily in contradiction to the aforementioned anomaly, first because the final neutron energy, 24.3 keV of [5] was too high, corresponding to a much too short scattering time 10^{-17}–10^{-18} sec, where the *n*-*p* scattering intensities would reveal no anomaly as explained in [6]. Second, it was suggested that the *n*-*p* scattering cross section should have been compared not to *n*-D in D_2O (which could by itself reveal an anomaly) but to a higher nuclear mass where the *n* scattering anomaly is less likely to occur [6–8]. Third, in Ref. [5], the neutron coherence length l_c of the scattered neutrons was very small, $l_c \sim 0.02$ A, and hence not conforming to the main condition set in Refs. [7,9] for observing the *n*-*p* scattering anomaly. This condition requires that $l_c = d_{HH}$, where $l_c = \lambda E/\Delta E$ (here, λ, the wavelength of the scattered neutrons of final energy E, detected with an energy resolution ΔE) with $d_{HH} \sim 1.5$ Å, the distance between two close protons in H_2O. This last condition was the basis for explaining the *n*-*p* scattering anomaly [7,9] in NCS experiments. In Refs. [7,9], the scattered neutrons were detected after passing through a narrow energy filter (e.g., ^{197}Au foil, 0.00125 cm thick, of final energy, $E = 4.9$ eV, with $\Delta E \sim 0.28$ eV resulting in $l_c = 2.3$ Å, which is larger than d_{HH}). It was claimed that the sharp energy selection makes the coherence length of the neutron wave packet long enough to cause a drop in the *n*-*p* scattering intensity via *n*-wave interference from two exchange-correlated protons. This condition [9] was also used to explain how a drop in the *n*-*p* scattering intensity could coexist with a normal value [10] of the *total* *n*-scattering cross section at 10 to 100 eV. The interference process involved is thought to be similar to that [11] of thermal *n* scattering from ortho- and para-H_2 at ~10 K.

In the present work, we addressed the above three concerns by carrying out an experiment similar to that of Ref. [5], but replacing the iron filter with a ^{238}U filter and using a CH_2 sample where the *n*-*p* scattered intensity is compared to that from a large nuclear mass, namely, ^{12}C in graphite. The U filter creates ~20 sharp energy windows ($\Delta E \sim 3$ eV) with final energies between 64 and 2600 eV (see Table I); the 64 eV line is low enough to overlap the high energy end of the NCS work. By measuring the *n*-scattering intensity ratios from CH_2 and C, using the *n* energies of Table I, we were able to test the validity of the above coherence length criterion at several final *n* energies. A remarkable agreement was obtained between the mea-

TABLE I. Final energies, full-width-at-half-maximum (ΔE), and coherence lengths of the ^{238}U-filtered neutron lines used in the present work.

Peak Energy (eV)	FWHM (eV)	$l_c = \lambda E/dE$ (Å)
64.4	1.3	1.8
100.3	2.7	1.1
186.2	3.8	1.0
346.2	1.6	3.4
593.5	1.5	4.7
659.5	1.7	4.3
935.2	2.0	4.4
956.6	2.3	3.9
988.6	3.1	2.9
1391.6	2.4	4.5
1520.4	2.2	5.1
1660.4	1.9	6.2
1778.6	3.0	4.0
1962.9	7.0	1.8
2183.0	3.0	4.5
2543.4	5.7	2.5

sured and the conventionally calculated scattering intensity ratios at all energies and no anomaly was observed.

Experimentally, the scattering measurements were carried out using the Gaerttner electron linear accelerator at RPI, operated at ~50 MeV, a pulse repetition rate of 400 Hz with an electron pulse width of 120 ns and an average e current of 40 μA. The use of a narrow pulse width enabled us to measure the sharp n lines with good resolution. The experimental arrangement is described in Fig. 1. The e beam strikes a water-cooled tantalum target producing a white source of neutrons which are then scattered by polyethylene (CH_2) or graphite samples, set at a distance of 14.5 cm from the Ta target. The neutrons are then passed through a 7.6 cm depleted ^{238}U (containing 0.2% ^{235}U) filter before reaching the ^{6}Li glass detector placed at a distance of 25.5 m from the Ta target. The peaks of the U-filtered lines correspond to deep minima in the ^{238}U total neutron cross section [12]. These minima are created by the destructive interference between the nuclear resonance scattering of neutrons and potential scattering. In effect, the U filter converts a white n spectrum into a multiline spectrum. Table I lists some of the stronger final n energies E of the ^{238}U filter, together with the calculated values of l_c which are of the order or larger than 1.8 Å, the

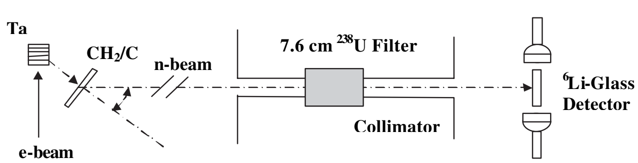

FIG. 1. Schematic view of the layout of the scattering experiment. The ^{238}U filter is 7.6 cm thick. The Ta target to n-detector distance is 25.5 m.

distance between 2 protons in CH_2. Rectangular samples (100 mm × 62 mm) were used, of thicknesses and weights CH_2 : 1.2 mm, 7.39 g; graphite: 3.94 mm, 42.55 g, each subtending scattering angles between 28° and 64° centering at 44°. The samples were held on thin Al strips, to minimize the background. The background was determined using a blank holder. The use of a solid sample (CH_2 and C) is preferable over a liquid (H_2O) used in [5], as it is self-supporting and does not undergo shape deformation during the run. Further, no container is necessary, thus reducing the background and improving the signal to noise ratio. The choice of CH_2 has an advantage as it involves C-H bonds where the anomalous effect at high momentum transfers was reported to be large (~50% in formvar [4] and ~70% in acetone [3]). The n flux was monitored both by integrating the electron charge striking the Ta target and independently by using a fission detector placed inside a separate beam hole. The energies of the scattered neutrons were determined by measuring the time of flight (TOF) to reach the detector at a distance of 25.5 m from the Ta neutron source. The final energies of neutrons scattered from CH_2 and C are the same and predetermined by the energy windows of the ^{238}U filter. Hence the TOF of neutrons from the two different masses, ^{12}C and ^{1}H is nearly the same. The small differences are due to the different time intervals for the neutron to traverse the 14.5 cm path between the Ta source and scatterer. This difference shows up clearly at low energies, revealing a shift of ~6 channels between the TOF spectra of CH_2 and C, at 64 eV, where each channel equals 64 ns. The shift reduces to ~1 channel at 2000 eV. A single 1.27 cm thick ^{6}Li-glass scintillator of 12.7 cm diameter was used as the neutron detector. The neutron beam was collimated to a cross sectional diameter of 12 cm at the detector position. Figure 2 shows a typical TOF spectrum of scattered neutrons from the CH_2 sample. Note the excellent resolution

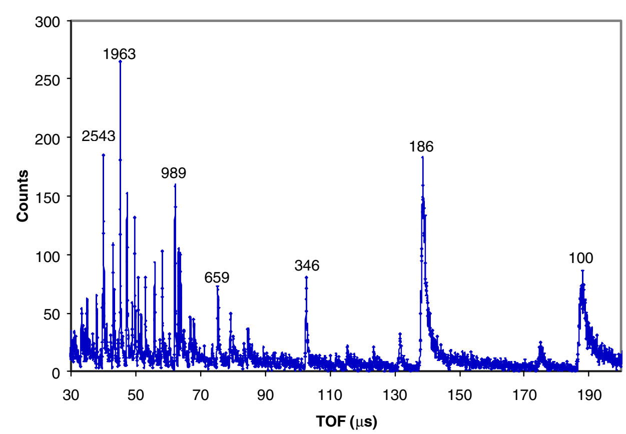

FIG. 2 (color online). Time of flight spectrum of scattered neutrons from a 1.2 mm thick CH_2 sample, filtered by a 7.6 cm thick ^{238}U plate. A ^{6}Li-Glass n-detector was used. The length of the flight path is 25.5 m. The peaks are labeled in energy units of eV.

of the peaks which enabled us to obtain the scattering intensity ratios for each n line separately. It may be noted that the Ta target was set in such a way as not to be directly visible to the n detector. The U filter served also as an effective shield, reducing the intensity of the gamma flash reaching the n detector.

The intensity of each of the scattered TOF lines from CH_2 is the sum of the scattering signals from H and C nuclei. Because of the kinematics, however, the corresponding neutron incident energies are different in the laboratory system. For a final energy E_f, the incident n energy depends on both the scattering mass and scattering angle θ. At $\theta = 45°$, the incident n energies on H and C are: $E_i = 2E_f$ and $1.05E_f$, respectively. In calculating the n-scattering intensities, it is necessary to know the variation of the neutron flux with the incident laboratory energy. This was determined experimentally to be $\sim 1/E^{0.65}$ and is the same as in the previous work [5].

Since the n-scattering cross section in the laboratory system from H is much higher than that of C, the scattering signal from CH_2 is dominated by the H signal. At 45° and $E_f = 1000$ eV, the scattering cross section ratio $\sigma_s(n-p)/\sigma_s(n-C)$ is 10.8. In fact, accounting for the sample thicknesses, the calculated scattering intensity ratio at 45° is 3.9. Thus any anomalous drop in the n-p scattering intensity must show up as a drop in the intensity of the signal from CH_2. Assuming that the anomalous decrease of $\sigma_s(n\text{-}p)$ from a C-H scatterer is $\sim$40% (see Refs. [3,4]), the overall calculated scattering intensity ratio from the actual CH_2/C samples at 45° becomes 2.6 at 1000 eV which is much lower than the conventional ratio (Fig. 3). Note that because the n lines are very sharp, the corresponding number of counts in the TOF spectrum is usually low. To gain statistics and reduce the statistical error in the measured ratio of some weak scattering signals, such as the three neighboring peaks at 935, 957, and 989 eV, we summed up their separate signals and determined their combined weighted energy and scattering intensity ratio. A similar procedure was used for the n lines at 346, 591, and 659 eV and for the two lines at 2183 and 2543 eV. The error introduced by this procedure is very small due to the weak variation of the calculated ratios [including multiple scattering (MS)] within the above energy ranges. In Fig. 3, we plotted the final calculated and measured ratios for CH_2 and C versus energy after averaging over all lab scattering angles between 28° and 64°. The dashed line shows the calculated *single* neutron scattering intensity ratios, while the solid line shows the calculated ratios obtained after including MS; both calculations were carried out using the general purpose Monte Carlo code MCNP5 [13]. The single n-scattering calculations were tested analytically and an excellent agreement was obtained with the MCNP results. The lower dotted line represents the single scattering ratios obtained by artificially reducing the n-p cross sections by 40%. Conventional laboratory scattering cross sections of H and C were deduced from the *total* neutron cross sections, taken from the ENDF tabulated data [12]. In all calculations, the variation of the scattering cross section with energy as well as the exact geometry and the thickness of the samples were accounted for. Note that the number of atoms of the C sample is 3.4 times larger than that of H atoms in CH_2. The choice of relatively thick samples was dictated by the beam intensity, the limited Linac running time and the background. In fact the relative thicknesses of CH_2 and C were selected in such a way as to make the MS contribution nearly the same, at $\sim$1000 eV, for both samples, so that the *total* scattering intensity ratio is nearly the same as *single* scattering intensity ratio at $\sim$1000 eV (Fig. 3). Note that the E dependence of MS on H is different than that on C. The MS amounted to $\sim$21% of the single scattering. The statistical error of the measured ratios is around 6%, becoming smaller for the more intense lines. From Fig. 3, it is clear that there is a good overall agreement between the measured and calculated scattering intensity ratios in the 64 to 2600 eV energy range. All the measured ratios are contained within less than 2 standard deviations of the conventionally calculated values and there seem to be no effect of the relative magnitude of the coherence length relative to d_{HH} in CH_2. In effect, no anomaly was observed. Note that the n-p interaction time of all n energies of Table I is shorter than $\sim 2 \times 10^{-16}$ sec. Hence no decoherence of the phase relations between assumed entangled proton pairs can occur. The n-interaction time is obtained [14,15] from the relation $\tau_{sc}(\theta) \sim M/[k(\theta)\langle p^2\rangle^{1/2}]$, where θ, the neutron scattering angle, M the mass of the scattering nucleus, $k(\theta)$ the momentum transfer, and $\langle p^2\rangle^{1/2}$ is related to the square root of the mean vibrational kinetic energy of the H atom in CH_2. At 2600 eV, $\tau_{sc}(\theta) \sim 2.2 \times 10^{-17}$ sec at 45°.

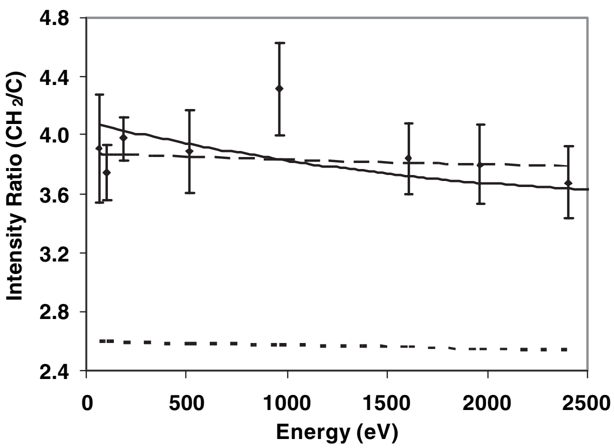

FIG. 3. Scattering intensity ratios from CH_2 and C. The upper two lines are conventionally calculated ratios, showing the single (dashed line) and total, including MS (solid line). The bottom dotted line is the single scattering result where the n-p cross section is reduced by 40%.

Theoretically, a breakdown of the Born-Oppenheimer approximation (BOA) was suggested [16,17] to explain the drop in the *n*-*p* scattering intensity at high momentum transfers in NCS measurements. This involves a nonadiabatic excitation of the electronic degrees of freedom. Such atomic excitations are supposed to diminish the energy of some scattered neutrons, hence decreasing the neutron intensity detected in the peak. However, this same process was considered by Colognesi [18], for the specific case of *n* scattering from H_2 where the atomic excitation could be easily evaluated; it was concluded the BOA has a negligible effect on the measured cross sections.

In a most recent Letter [19], the effect of entanglement on the *n*-scattering intensity from a proton pair in solids was calculated using a different approach. The result is that such an effect is expected to be undetectable in NCS experiments because of the high energy transfer and the broad energy resolution. This result disagrees with the previous theoretical models of Refs. [6–9]. It appears therefore that up to now, there is no theoretical consensus as to the origin of the claimed measured shortfall of *n*-*p* scattering intensity in NCS experiments.

In conclusion, the main goal of the present experiment was to find out if a drop of the *n*-*p* scattering intensity could be observed if the two conditions set in Refs. [7,9] are satisfied. The *n* energies used were much lower than that of Ref. [5] and overlapped the high energy tail of the NCS experiments where the scattering times are less than $\sim 2 \times 10^{-16}$ sec. The scattered neutrons are detected using sharp energy windows (created by the 7.6 cm thick ^{238}U filter) ensuring that the neutron coherence lengths are of around the same distance d_{HH} in CH_2. Assuming that *n* scattering from ^{12}C has no anomaly, in the 64 to 2600 eV range, the results of the present experiment show no anomaly in *n* scattering from a pair of exchange-correlated protons in CH_2. Hence, the conditions brought forward to explain the anomalous drop in *n*-*p* scattering of the NCS work [7,9], cannot be applied to the higher neutron energies used in the present work. In this respect, it would be very interesting to carry out a NCS measurement [2] using a higher energy resonance such as the 60.3 eV of a gold filter or the 20.9 and 36.7 eV resonances of a uranium filter. It will thus be possible to find out whether the *n*-*p* anomalous drop in scattering intensities will disappear at such higher energies as observed in the present work.

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