

Search for Anomalous Scattering of keV Neutrons from H₂O-D₂O Mixtures

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We measured the neutron scattering intensities from pure liquid H₂O relative to that of pure D₂O and also relative to H₂O-D₂O mixtures, at room temperature. This study is relevant to the problem of quantum entanglement. The neutrons were generated from an electron Linac and the final energy of the scattered neutrons was fixed at 24.3 keV using a 20 cm thick pure iron filter. The scattering intensity ratios were found to agree with expected values deduced from the tabulated total cross sections within an accuracy of 3%. Thus no anomaly was observed.

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Recently, there was a flurry of activity in relation to the neutron Compton scattering (NCS) from a H₂O-D₂O mixture [1] and from H-containing compounds and polymers [2–5]. It was found that, at epithermal energies in the 10–200 eV range, the neutron scattering cross section from H, at high momentum transfers, decreases by around 40% compared to that of other elements. Similar effects were reported when some or all of the H atoms were replaced by D, in polystyrene (C₈H₈), in benzene (C₆H₆) [2], in Nb hydride and Pd hydride [3], and also in formvar (C₈H₁₄O₂) [5]. This result seemed to disagree with any logical expectation. In the case of H₂O, this anomaly was attributed to a quantum entanglement effect between the scattered neutron and the two protons of the H₂O molecule. When one of the protons of a H₂O molecule is replaced by a deuteron (by adding D₂O to light water), this quantum effect was found to decrease gradually with increasing the relative number of D's in the H₂O-D₂O mixture. It was also noted that in metal hydrides this anomaly becomes more pronounced at higher momentum transfers, which correspond to higher incident neutron energies. This in turn is related to short interaction times (in the subfemtosecond range) between the neutron and the scattering proton.

A theoretical support for this anomalous effect was proposed [6–8], based on a model in which short-lived ($\tau < 10^{-15}$ s) correlations in the spatial and spin degrees of freedom of the hydrogen isotopes are assumed. According to this model, the correlations cause an entanglement of degrees of freedom in the two protons of H₂O. This induces a reduction of the cross section per proton below the cross section for a single isolated proton. According to [7], even though the neutron wavelengths are much smaller than the distance between protons, nevertheless they are viewed by the neutron as nonindividual scatterers. Quantum entanglement will then decrease the scattering cross section.

The above reported reduction in scattering intensity from protons in H₂O induced some authors [9] to propose that, within the time scale of around 10^{-16} s, the correct chemical formula of water is H_{1.5}O and not H₂O. It may be

noted, however, that a recent experimental search for an anomaly in the *total* neutron cross section in H₂O-D₂O mixtures, in the 1–100 eV range, did not find such an effect [10] within an accuracy of 0.3%. Moreover, a recent theoretical study by Cowley [11] questioned the existence of such an anomalous effect because it is inconsistent with the first moment sum rule for neutron scattering which is related to the total neutron cross section in H and D. For the same reason, it was suggested that the theory given in Refs. [7,8] is incorrect. In Ref. [11], some experimental concerns were also raised questioning the validity of the method of data analysis used [1–5] in deducing the area of the scattering signal. However, in a more recent and detailed paper [12], an attempt was made to answer the experimental reservations raised in Refs. [10,11] and tried to show that, in effect, the reported anomaly in the H₂O-D₂O cross section may be smaller in magnitude than was reported in Refs. [1–5], but claimed that the anomaly exists.

Because of the great importance of this problem and its possible relevance to quantum entanglement, we report a new search for an anomalous behavior of the neutron scattering intensities at higher energies in the 30 keV range. Such energies are in the region of much shorter interaction times ($\sim 10^{-17}$ s), where the entanglement phenomenon is claimed to exist [7,8]. To do so, we employed a new method in which the ratios of the scattering intensity from a pure H₂O sample and H₂O-D₂O mixtures were compared. A careful analysis of our results did not find any anomaly in the scattering cross section ratios. This work constitutes an independent experimental test of the above effect at higher energy.

Experimentally, the scattering measurements were carried out using the Gaertner electron linear accelerator at Rensselaer Polytechnic Institute (RPI), operated at ~ 50 MeV, a pulse repetition rate of 225 Hz with an electron pulse width of 1 to 2 μ s and an average e current of 50 μ A. 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

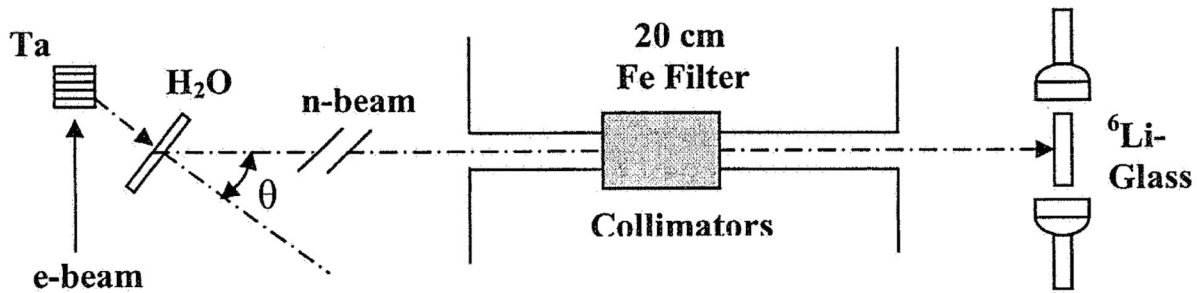


FIG. 1. Schematic view of the layout of the experiment. The distance between the Ta target and the ⁶Li-glass *n* detector is 25.3 m.

scattered by a thin water sample, set at a distance of 15 cm from the Ta target. In this experiment, the sample contained either pure H₂O, H₂O-D₂O mixture, or pure D₂O, acting as the source of scattered neutrons. Such neutrons are then passed through a 20 cm thick block of pure iron before hitting a ⁶Li-glass detector, placed at a flight distance of 25.3 m from the *n* source. Because of scattering by pure iron, nearly all neutrons are expected to be filtered out of the beam except those at 24.3 keV, 81 keV, and other discrete higher energies, corresponding to the deep minima in the iron total neutron cross section [13,14]. These minima are created by the destructive interference between the nuclear resonance scattering of neutrons and potential scattering. In effect, the iron filter converts a white neutron spectrum to a neutron beam with discrete energies. In the present work, we concentrate on the 24.3 keV scattered neutron group, because the higher energy neutrons were not as easily separable from the other neutron “lines.” Thus, 24.3 keV is also the fixed final energy of the scattered neutrons. Note that the 24.3 keV line, created by the 20 cm iron filter, has an asymmetric shape with a full width at half maximum of 2.1 keV. This iron filter, placed at a distance of 12 m from the water scatterer, had an extra advantage serving as an additional shield to the *n* detector from the intense gamma flash created when the electrons strike the Ta photoneutron target of the Linac.

The neutron detector consisted of a single 1.27 cm thick ⁶Li-glass scintillator of 12.6 cm diameter, viewed by two 12.6 cm diameter photomultipliers placed on opposite sides of the glass scintillator. The neutron beam was collimated to a cross sectional diameter of 12.0 cm at the detector position. A typical time-of-flight (TOF) spectrum of the filtered beam is shown in Fig. 2, revealing the 24.3 keV neutron group, well isolated from the other high energy “lines” and from the gamma flash.

It may be noted that the tantalum target was placed so as not to be directly visible to the *n* detector, to avoid the effects of the gamma flash. The water sample (10.0 g in the case of pure H₂O) is contained inside an aluminum 10 cm × 6 cm rectangular can 1.8 mm thick, covered by thin anodized aluminum foils (0.076 mm thick), and were prepared using pure distilled H₂O and D₂O (99.9% atom

D). The molar fractions of D₂O in the mixtures were $X_D = 0.29, 0.48, 0.70, \text{ and } 0.84$. In practice, all samples contained the same number of water molecules. All measurements were carried out relative to pure H₂O samples of identical geometry. The background was determined using identical blank aluminum containers with no water. In each set of measurements, two samples were used, one containing 10.0 g of pure H₂O and the second an H₂O-D₂O mixture. These were mounted on a sample changer and alternately interposed into the neutron beam for 10 and 30 min (depending on the scattering intensity of the sample), while the TOF spectrum was recorded and accumulated. The thickness and the geometry of the water inside the Al can are important and were inspected using an x-ray camera. The height of the water level in each pair of samples was compared and found to be usually the same to within 2 mm. Such a relatively small difference was found to have no measurable effect on the scattering intensity ratios. An independent fission detector, placed inside a separate

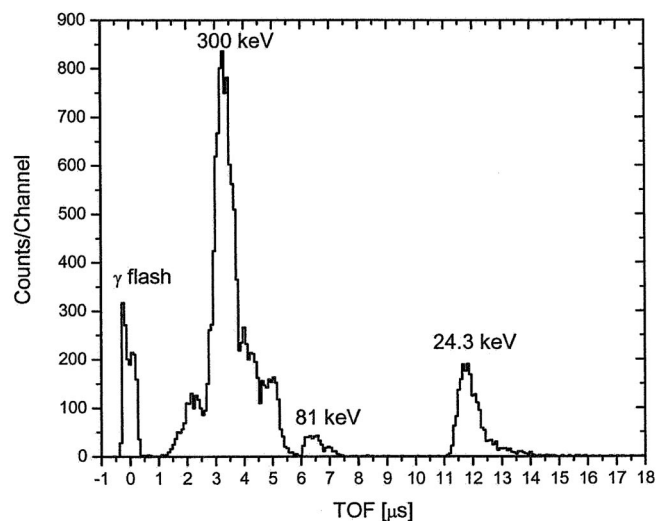


FIG. 2. Time-of-flight spectrum of the scattered neutrons from a H₂O sample detected after passing through a 20 cm Fe filter. The γ flash is depressed by the effect of the 20 cm iron filter. The 300 keV group is the average energy of neutron lines weighted by the ⁶Li (*n*, α) cross section of the detector.

beam tube, was employed as a neutron flux monitor and served to normalize the TOF spectra.

Since in the present arrangement the final energy of all scattered neutrons from H, D, and O is the same, namely, 24.3 keV, hence the n -counting efficiency is also the same. Because of kinematics, the incident n energies and hence the energy transfers depend on mass. At 45° , the corresponding incident energies for H, D, and O are 48.6, 32.9, and 25.2 keV, hence the energy transfers are 24.3, 8.6, and 0.9 keV, respectively. Note that the *total* n cross sections at the above incident energies are 15.7, 3.3, and 3.8 b, respectively. At 45° , the *laboratory* scattering cross sections of neutrons from H, D, and O are 3.5, 0.47, and 0.33 b/sr. All energy transfers are far higher than the binding energies of the atoms in a water molecule, hence each neutron scatters from a single nucleus and the impulse approximation can easily be assumed to be exact. Note that the momentum and energy transfers in the present case are about 10^3 higher than that of the epithermal region used in Refs. [1–5].

As already noted, the final energy of the scattered neutrons is the same, irrespective of mass. Hence the neutron TOF from the different masses is practically the same. The small differences arise from the fact that it takes different time intervals for the neutron to traverse the 15 cm path between the moderator and scatterer. Thus, the intensity of the scattered 24.3 keV TOF line is the sum of the scattering signals from H, D, and O. Each sample subtended scattering angles between 25° and 65° . The corresponding incident energies in the case of n - p scattering are 29.6 and 136 keV. Since the n scattering cross section in the laboratory system from H is much higher than from D and O, the scattering signal will be dominated by the H signal. Hence the scattering intensity from the H_2O molecule is also expected to be much higher than that of D_2O . Thus any anomalous decrease of the scattering intensity from H, due to quantum entanglement, must show up as a drop in the intensity of the 24.3 keV TOF scattering peak from the entire molecule. This decrease in the scattering intensity is reduced, however, due to the additional unchanged contribution of the O atoms in both H_2O and D_2O . Assuming that the anomalous drop in the scattering intensity from an H atom is about 40% (as reported for epithermal energies in Refs. [1,3–5]), the inclusion of the O contribution in the H_2O molecule reduces this anomalous drop from 40% to 35% relative to that of a D_2O molecule.

Note that the scattering intensity is represented by the area of the 24.3 keV line in the TOF spectra. The measured scattering intensity ratios from the pure H_2O sample relative to that of the H_2O - D_2O mixture (in the laboratory system) as a function of the D_2O concentration, X_D , are given in Fig. 3. The figure also shows the calculated scattering intensity ratios (solid line) obtained by using the conventional laboratory scattering cross sections of H, D, and O. These were deduced using the ENDF tabu-

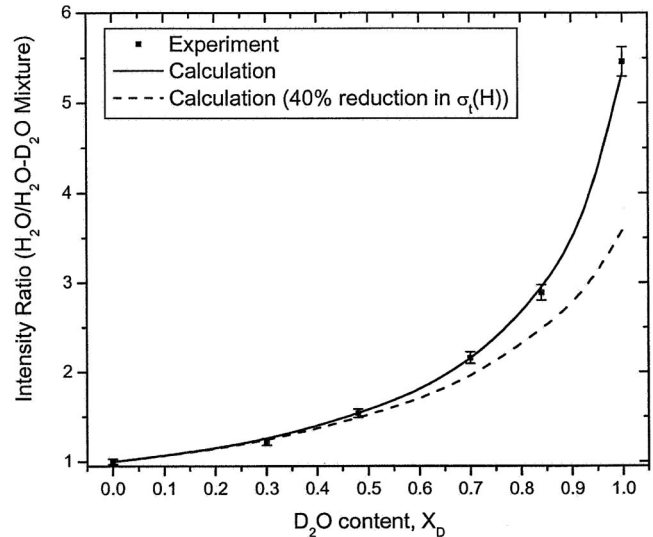


FIG. 3. Measured (squares) and calculated (solid line) scattered intensity ratios versus X_D , the D_2O concentration in the H_2O - D_2O mixture sample. The dashed line represents calculated ratios obtained by assuming a 40% drop in the n - p scattering cross section; it is shown to illustrate the extent of deviation from the measured data that would be caused by such a drop in cross section.

lated data [15] of the *total* neutron cross sections. Here we accounted for the scattering of neutrons in the sample in both the incident and scattered channels, using the exact geometry of the water sample with respect to the neutron beam (Fig. 1). The dotted line of Fig. 3 represents the same scattering cross section ratios obtained by artificially reducing the n - p scattering cross sections by 40%. A remarkable agreement was obtained between the measured and the conventionally calculated scattering intensity ratios for all X_D values of D_2O molar fractions. Note that, because a *ratio* between scattering cross sections is being considered, the difference between the solid and dotted lines is small for mixtures containing less than 50% D_2O . It is only for mixtures containing 70% D_2O and higher that a big divergence between the two calculated ratios occurs. The overall errors of the measurements are estimated to be about 3% while the actual statistical errors were about 2%. The calculated results are averages over scattering angles between 25° and 65° . Similar agreement with conventional calculations was also obtained when larger water scatterers (10 cm wide) were used subtending a wider range of scattering angles.

A few remarks concerning the above calculation are in order. In calculating the intensity ratios in the TOF spectra, one has to account for $N(E_i)dE_i$, the variation of the neutron flux $N(E_i)$ with the incident laboratory energy E_i which, for this energy region, has been taken as $\alpha/E^{0.65}$, where α is a constant. In TOF space, this should be written as a time distribution, $N(t_i) = N(E_i)dE_i/dt_i$, where the factor dE_i/dt_i may be obtained from kinematics. The

energy variation of the neutron flux emitted by the water-cooled Ta target is important and was tested experimentally in the 24–150 keV region, of our interest in the present work. A good agreement with the $1/E^{0.65}$ assumption was obtained. Note, however, that the $1/E^{0.65}$ shape is slightly distorted because of the effect of Al (1.6 mm thick) containing the Ta target. Aluminum has two resonances at 35.2 and 87.3 keV, which create two dips in the shape of the neutron flux and could affect the scattering signals from both D and H. A Monte Carlo calculation showed that those dips have a negligible effect on the ratio, $I(\text{H}_2\text{O})/I(\text{D}_2\text{O})$. The multiple scattering effects of the neutrons were calculated for a sample thickness (1.8 mm) and geometry of Fig. 1, using the Monte Carlo code, MCNP5 [16]. The effect on the *ratio* of scattering intensities from the two samples is $<1\%$ and was neglected. Note, however, that the calculated ratio of second to first order scattering of neutrons is $\sim 9\%$ for both the H_2O and D_2O samples; thus the effect on the ratio is very small.

We hereby deal with the n - p interaction time involved in the present experiment. To do so, we adopt the literature definition [7,17,18] of the interaction time τ_{sc} of the n - p scattering process as given by $\tau_{sc}(\theta) \sim M\hbar/[k(\theta)\langle p^2 \rangle^{1/2}]$, where θ is the neutron scattering angle, M is the mass of the scattering nucleus, $k(\theta)$ is 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 water. Thus, the neutron interaction time at the high momentum transfers occurring in the present experiment is of the order of 7×10^{-18} s and is shorter by a factor 70 compared to that of the NCS work [1–5]. From a consideration of the NCS results of Ref. [4], it may be seen that a strong shortfall of the neutron scattering from H would be expected at scattering times $\tau_{sc} < 4 \times 10^{-15}$ s. A similar result also follows from the theoretical work of Karlsson and Lovesey [4,6] and of Karlsson [7]. In fact, according to the theory of Karlsson [7], a short interaction time of $\sim 10^{-17}$ s should cause a strong exchange correlation effect between the two protons of H_2O (due to the absence of decoherence). This is expected to result in a strong decrease of the neutron scattering intensity from H_2O , as compared to that from HDO or D_2O . In the present work, however, no such effect was detected at such high energies. The fact that there is a very good agreement between the measured and calculated ratios shows that there is no anomaly in the scattering cross sections from H_2O at ~ 40 keV to within an accuracy of 3%. The present results are in agreement with the sum rule criterion mentioned by Cowley [11] because our scattering cross sections are based on the tabulated values [15] of the neutron total cross sections from H, D, and O.

This experiment was carried out to search for a decrease in the n -scattering intensity at interaction times in the subfemtosecond regime, 10^{-17} s. These times are shorter than those covered in Refs. [1–5], which reported the anomalous scattering effect in H_2O - D_2O at room tempera-

ture, and were attributed to quantum entanglement. The present method also uses neutron inelastic scattering but is different from that used in [1–5], as explained above. We conclude that within a statistical accuracy of 3% there is no evidence for any deviation from that conventionally calculated on the basis of the tabulated total neutron cross sections. Using the terminology of Ref. [9], we may emphasize another conclusion of the present experiment by stating that, within the n - p interaction times of $\sim 10^{-17}$ s, the water molecule chemical formula is H_2O and *not* $\text{H}_{1.5}\text{O}$ as has been proposed.

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