Search for Anomalous Effects in H_2O/D_2O Mixtures by Neutron Total Cross Section Measurements

J. J. Blostein,[1,2] J. Dawidowski,[1,2,*] S. A. Ibáñez,[2] and J. R. Granada[1,2]

[1]*Consejo Nacional de Investigaciones Científicas y Técnicas (CONICET), Bariloche, Argentina*

[2]*Centro Atómico Bariloche and Instituto Balseiro, Comisión Nacional de Energía Atómica, Universidad Nacional de Cuyo, (8400) Bariloche, Argentina*

(Received 4 September 2002; published 14 March 2003)

We investigate anomalies ascribed to quantum entanglement phenomena recently reported in deep inelastc neutron scattering (DINS) measurements by an independent neutron technique on H_2O/D_2O mixtures. We performed transmission experiments to study several liquid H_2O/D_2O mixtures at room temperature in the epithermal energy range. We obtain the total cross sections of the mixtures, which are in agreement with the expected results according to the tabulated values within a 0.3% relative error. We observe no anomalies and stress the limitations of the validity of the data-processing procedures employed in the DINS experiments where the anomalies were reported.

DOI: 10.1103/PhysRevLett.90.105302 PACS numbers: 67.20.+k, 61.12.Ex, 61.25.Em

The neutron cross section of hydrogen has been a very well-known magnitude since the early days of neutron scattering [1]. Several decades ago, neutron total cross sections of light [2] and heavy [3] water were exhaustively studied driven by the interest in the development of reactor physics. In recent years, anomalous values of the parameters that characterize the interaction between neutrons and protons composing hydrogenated liquids at room temperature were reported [4,5]. In particular, light water/heavy water mixtures were examined, and a dependence of the bound-atom cross section of hydrogen with the molar fraction of deuterium in the mixture was found, based on experimental data obtained from the deep inelastc neutron scattering (DINS) technique for incident neutron energies in the range of some tens of electron volts. These results, which go counter to the well-established theory of neutron scattering, were interpreted in terms of the existence of quantum entanglement between neighboring hydrogen atoms, produced in times of the range of 10^{-16} s. Theoretical support for such an assumption was provided in Ref. [6], based on a model of coupling between pairs of atoms. An experimental test of such anomalies was performed through precision interferometric measurements on such liquid mixtures which showed no evidence of the purported phenomenon [7]. However, the experiment was performed at a low incident neutron energy (11.2 meV), and for this reason it was not in the range of interaction times where this phenomenon was claimed to exist [8]. From a different point of view, the data-processing method employed in those DINS experiments presented some questionable points. In fact, as was shown in Ref. [9], the commonly employed approximation to analyze neutron Compton profiles (based on a convolution not directly deducible from the exact expressions) induced deviations in the observed peak intensities in the same sense as the observed anomalies. Furthermore, the procedure to obtain peak intensities in the DINS technique had not been used before to study total cross sections, and it was not considered as the most suitable neutronic tool for that purpose, as it was pointed out in that reference. Instead, transmission experiments were employed since the very beginnings of neutronics to obtain such magnitudes, and in fact it has been one of the main techniques employed to build the existent reference tables of neutron scattering lengths. In order to elucidate the existence of this phenomenon by means of a standard technique capable to yield unequivocal results, we performed neutron transmission measurements on different light water/heavy water mixtures. In this Letter we present results of neutron total cross sections on such mixtures, employing the standard transmission technique, in the range from 1 to 100 eV (i.e., a range compatible with the above mentioned reports), and we analyze and compare our results with those obtained with the DINS technique. After a careful analysis of the neutron total cross sections we did not find any evidence of an anomalous effect.

The transmission experiments were performed at the Bariloche electron linear accelerator (LINAC) facility (Argentina). The accelerator operated at a frequency of 100 Hz (with electron pulse widths of 1 μs) and a 25-μA mean electron current. Neutrons were moderated in a 40-mm thick polyethylene slab, after which a cadmium sheet was placed to let only epithermal neutrons emerge from the source. Measurements were carried out at room temperature, employing the "sample in-sample out" technique [10] every 10 min. The detector bank consisted of seven ^{3}He proportional counters (10 atm filling pressure, 6" active length, and 1" diameter) placed on the incident beam path at 827 cm from the neutron source. The neutron beam was collimated, so its cross-sectional diameter was 13.5 cm at the detection position. The time-of-flight spectra were recorded in 4096 channels, 2 μs width each. An independent detector placed near the neutron source

was employed as a monitor, which served to normalize the spectra. The samples were cycled into the beam employing a rotative sample changer, located 3 m from the neutron source. The background contribution was carefully determined with a polyethylene beam stop covered with cadmium at the sample position and was also checked by means of resonant filters of indium, gold, cobalt, and uranium of thicknesses sufficient to saturate the nuclear absorption at the resonance energies. The samples were prepared in a dry atmosphere starting from reactor-quality D_2O (0.9987 M) and nanopure H_2O. The molar concentrations of deuterium in the mixtures were $x_D = 0.15$, 0.3, 0.4, 0.5 and 0.9; pure H_2O and D_2O were also employed as samples. Two aluminum cans (of 7- and 10-mm thickness, respectively) were employed as sample containers, and an empty can of the same material was employed on the open beam ("sample out") position. A control lead sample (whose transmission was previously known [11]) was employed, and the stability of the different cycles of the measurement was verified.

The recorded spectra were corrected by dead-time effects [12] and the energy scale was corrected by the mean-emission-time effect of the moderator [13] and electronic delay. Contributions by inscattering (single and multiple) from the sample to the detectors were analyzed and proved to be at least 2 orders of magnitude less than the intensity of the transmitted beam.

Transmissions are obtained from the ratio

$$T(E) = \frac{S(E) - B(E)}{F(E) - B(E)}, \tag{1}$$

where $S(E)$, $F(E)$, and $B(E)$ stand for the (monitor normalized) sample-transmitted ("sample-in"), free-beam ("sample-out") and background recorded counts at the energy E. This quantity can be written as

$$T(E) = e^{-\Sigma_{\mathrm{tot}}(E)d}, \tag{2}$$

where d is the sample thickness and $\Sigma_{\mathrm{tot}}(E)$ the macroscopic total cross section. The macroscopic total cross section is related to the microscopic cross sections of the different molecules present in the sample through

$$\Sigma_{\mathrm{tot}}(E) = n_{x_D}[f_{H_2O}\sigma_{H_2O}(E) + f_{HDO}\sigma_{HDO}(E) + f_{D_2O}\sigma_{D_2O}(E)], \tag{3}$$

where n_{x_D} is the number of molecules (either of H_2O, D_2O, or HDO) per unit volume in the mixture, and f_{H_2O,HDO,D_2O} are the fractions of each molecular species in the mixture. These fractions can be calculated from the equilibrium constant κ of the reaction $H_2O + D_2O \leftrightarrow 2HDO$, where $\kappa = 3.75$ at room temperature [14]. n_{x_D} was calculated from the mass density of pure H_2O and D_2O [15] and was corrected by excess volume effects [16]. In the epithermal region where the free-atom regime is an accurate description of the system dynamics [17], Eq. (3) must tend to the asymptotic value

$$\Sigma_{\mathrm{free}} = n_{x_D}[2x_D\sigma_D^{\mathrm{free}} + 2(1 - x_D)\sigma_H^{\mathrm{free}} + \sigma_O^{\mathrm{free}}], \tag{4}$$

independently of the formation of HDO.

In Fig. 1 we show the measured macroscopic total cross sections Σ_{tot}. Absorption cross sections of hydrogen, deuterium, and oxygen were subtracted, according to the "$1/v$" absorption law [18], which is valid in the thermal energy range and was also verified up to the keV region [19] in the case of hydrogen. The thermal absorption cross section data were taken from Ref. [20]. The scattering cross sections thus obtained for each mixture were employed to obtain the free-atom cross section. For this purpose the cross sections were fitted with the asymptotic expression [21]

$$\Sigma_{\mathrm{scatt}}(E) = \Sigma_{\mathrm{free}}\left(1 + \frac{C}{E}\right), \tag{5}$$

where the constant C is related to the effective temperature of the vibrating atoms. The obtained values of Σ_{free} for each mixture are shown in Fig. 2. In the upper frame we show an average of the results obtained for the 7- and 10-mm thick samples compared with the values obtained by the calculation with the tabulated values of the scattering cross sections [20], in Eq. (4).

We observe that there is an excellent agreement between the measured values and the calculation. The errors of the measurements are within the symbol size. The lower frame shows the percent relative difference with the calculated value for both samples, showing that the agreement is better than 0.3% for the present measurements. The error bars, indicated only for the 10-mm width

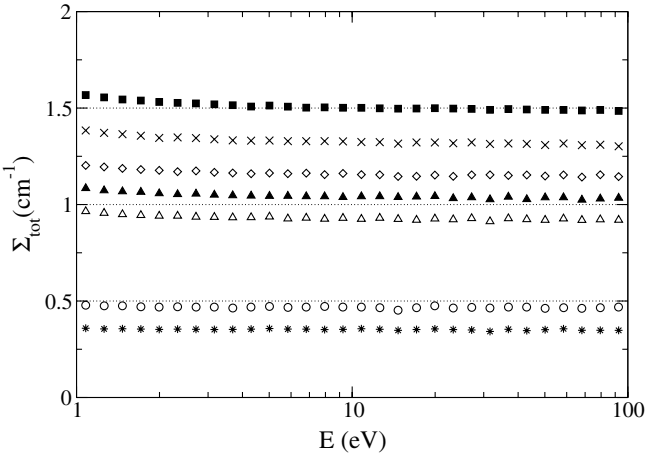

FIG. 1. Measured macroscopic total cross sections of pure H_2O (black squares) and the mixtures $x_D = 0.15$ (crosses), $x_D = 0.3$ (white diamonds), $x_D = 0.4$ (black triangles), $x_D = 0.5$ (white triangles), $x_D = 0.9$ (white circles), and pure D_2O (asterisks). The results from 7- and 10-mm thick samples coincide within the symbol size.

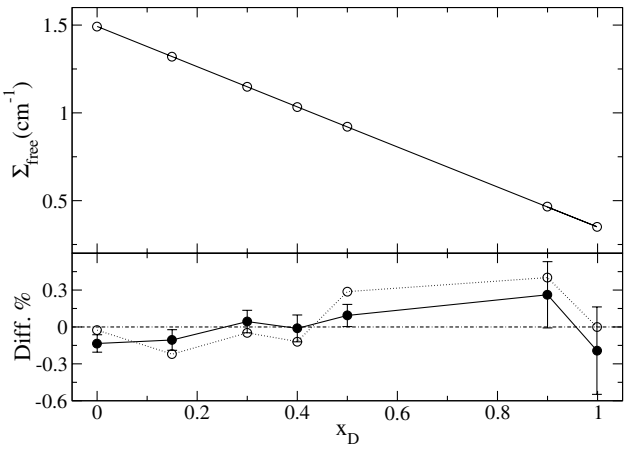

FIG. 2. Upper frame: macroscopic free atom scattering cross sections for the measured H_2O/D_2O mixtures as a function of the molar fraction of deuterium x_D (symbols), compared with the values predicted from the data taken from tables (line) [20]. The symbol size is larger than the error bars. Lower frame: percent relative discrepancy with those values for the 10-mm thick (black circles), and the 7-mm thick samples (white circles). Error bars, indicated only for the first ones, are similar in both cases. Connecting lines are included as an eye guide.

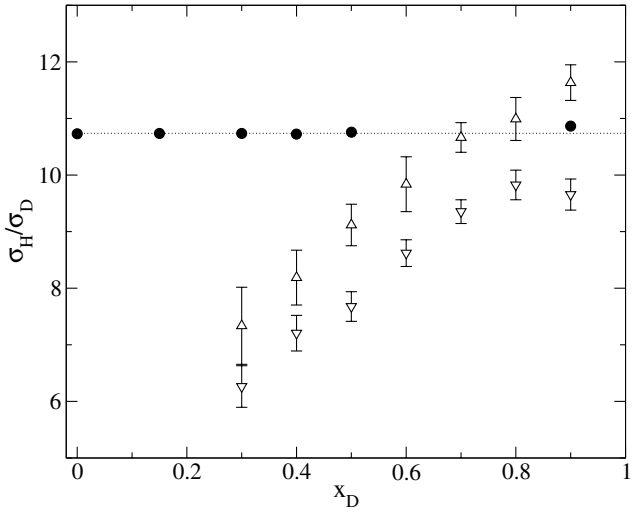

FIG. 3. Ratio of the bound-atom scattering cross sections σ_H/σ_D obtained from the present work (black circles), where σ_H is obtained from Eq. (4), and σ_D and σ_O from the tabulated values. The horizontal dotted line indicates the ratio of the tabulated values (10.737). The error bars are within the symbol size. Upside and downside triangles show the data sets presented in Ref. [4].

sample, are similar for both samples. It is worth mentioning that the C values obtained for pure light and heavy water are in excellent agreement with the well-known reference values presented in [22,23], while in the mixtures they are in full agreement with those obtained from the linear combination of the different molecular species (H_2O, D_2O, and HDO) in equilibrium. It should also be stressed that similar values of Σ_{free} are obtained if no model is employed to fit the scattering cross section, and instead a direct average of the total cross sections over the range from 10 to 100 eV is performed.

We now analyze the significance of the present results in the light of the presumed existence of the quantum entanglement phenomenon responsible for large variations in the hydrogen cross section present in the H_2O/D_2O mixtures. We restrict ourselves to the conclusions drawn in Ref. [4], where variations as large as 35% were observed in the ratio σ_H/σ_D through DINS experiments. These variations were subsequently ascribed to a variation in the hydrogen cross section, since additional reports showed no significant variation in the deuterium parameter with respect to the tabulated values [5]. To compare our results with those of Ref. [4] we performed the ratio of the bound-atom scattering cross sections σ_H/σ_D, which are related with the free-atom parameters above referred through the factor $(A + 1)^2/A^2$, where A is the mass of the scattering atom in neutron mass units. The parameter σ_H was extracted from our experimental data through the use of Eq. (4), and the values σ_D and σ_O taken from Ref. [20]. In Fig. 3 we show this ratio compared with the ratio of the tabulated values (10.737). We observe that the values obtained from our experiment are in excellent agreement with the previously known value, and no dependence on x_D is found. As a matter of fact, from our data we found $\sigma_H/\sigma_D = 10.735 \pm 0.007$, in good agreement with the ratio of the tabulated values. We also show in Fig. 3 the points presented in Ref. [4] for the observed values of σ_H/σ_D in a DINS experiment, which do not agree with our present result or with the tabulated values.

It is worth discussing the equivalence of the present experiments and those presented in Refs. [4,5]. The energy range of the incident neutrons presented here (1 to 100 eV) widely comprises that presented in the mentioned references, where the largest operative energy for a uranium filter and a scattering angle of 70° is about 75 eV. On the other hand, the measured magnitude (viz. the scattering cross section) is the nuclear parameter that characterizes the interaction between neutrons and the nuclei. This is the same magnitude observed in a DINS experiment governing the observed peak intensities. To further make explicit the comparison of the measured quantities, it is illustrative to mention that either technique (the total cross section or DINS) observes different integrals of the basic scattering law $S(Q, \omega)$, which is solely a property of the sample. On one hand, in the DINS technique the recorded spectrum at a time of flight t (for each atomic species) is [9]

$$c(t) = \Delta\Omega \frac{\sigma_b}{4\pi} \int_{\substack{E_{\inf} \\ t=\text{const}}}^{\infty} dE_0 \Phi(E_0) \sqrt{\frac{E}{E_0}} S(Q, \omega) \varepsilon(E) \times [1 - e^{-nT\sigma_F(E)}] \left| \frac{\partial E}{\partial t} \right|, \quad (6)$$

where $\Phi(E_0)$ is the energy spectrum on the incident neutrons, $\varepsilon(E)$ is the detector efficiency, and $\Delta\Omega$ is the solid angle subtended by the detectors. The resonant filter characteristics are contained in the term between square brackets, where n is the number density, T is its thickness, and $\sigma_F(E)$is its total cross section. The integral is calculated at a constant time of flight t, and the integration limits are defined by the allowed kinematic range. On the other hand, the total cross section is the integral [24]

$$\sigma(E_0) = \frac{\sigma_b}{2k_0^2}\int_0^\infty QdQ \int_{\omega_{\min}}^{\omega_{\max}} S(Q, \omega)d\omega, \qquad (7)$$

where in the same way as in Eq. (6) the integration limits are those of the allowed kinematic range. An inspection of Eqs. (6) and (7) leads us to conclude that any anomaly detected in the behavior of the constant σ_b, or in a global change of intensity of the function $S(Q, \omega)$ detectable in a DINS experiment, must also be observed in a transmission experiment. In fact, anomalies of the order of those mentioned in Ref. [4] would be manifested in a severe departure from linearity of the results presented in the upper frame of Fig. 2.

An insight in the causes of the anomalies reported in Refs. [4,5] can be found in the data analysis procedures there employed, an issue that was thoroughly examined in Ref. [9]. In the cited paper, we showed that the approximations employed in the standard DINS data treatment produce anomalous deviations in the same sense as reported in Refs. [4,5]. However, from the conclusions of Ref. [9], we could deduce only that the magnitude of the anomalies (if any) would be lower than in Refs. [4,5]. From the results of the present Letter we do not observe any anomaly in the total cross section of H_2O/D_2O mixtures, and the results here obtained are in full agreement with the neutron scattering theory known so far.

Finally, we must stress the need to make a critical revision of the different results of the same kind reported in several systems employing DINS experiments. Furthermore, we express that it would be most profitable for the DINS users technique to employ a new data treatment procedure in line with the formalism introduced in Ref. [9].

We must thank Dr. R. E. Mayer for helpful discussions on the experimental setup. We acknowledge L. Capararo for the technical support and M. Schneebeli and P. D'Avanzo for the LINAC operation. This work was supported by ANPCyT (Argentina) under Project PICT No. 03-4122.

*Electronic address: javier@cab.cnea.gov.ar

[1] J. M. Blatt and V. F. Weisskopff, *Theoretical Nuclear Physics* (John Wiley and Sons, New York, 1952), p. 48.
[2] E. Melkonian, Phys. Rev. **76**, 1750 (1949).
[3] H. C. Honeck, Trans. Am. Nucl. Soc. **5**, 47 (1962).
[4] C. A. Chatzidimitriou-Dreismann, T. Abdul-Redah, R. M. F. Streffer, and J. Mayers, Phys. Rev. Lett. **79**, 2839 (1997).
[5] C. A. Chatzidimitriou-Dreismann, T. Abdul-Redah, and J. Mayers, Physica B (Amsterdam) (to be published).
[6] E. B. Karlsson and S.W. Lovesey, Phys. Rev. A **61**, 062714 (2000).
[7] A.Iofe, M. Arif, D. L. Jacobson, and F. Mezei, Phys. Rev. Lett. **82**, 2322 (1999).
[8] C. A. Chatzidimitriou-Dreismann, T. Abdul-Redah, R. M. F. Streffer, and B. Hessmo, Phys. Rev. Lett. **84**, 2036 (2000).
[9] J.J. Blostein, J. Dawidowski, and J. R. Granada, Physica (Amsterdam) **304B**, 357 (2001).
[10] F. Kropff, J. R. Granada, L. A. Remez, and A. Vasile, Ann. Nucl. Energy **3**, 55 (1976).
[11] J. R. Granada, C. Bonetto, S. A. Hayashi, K. Kobayashi, S. Yamamoto, Y. Fujita, and I. Kimura, Physica (Amsterdam) **190B**, 259 (1993).
[12] G. J. Cuello, P. J. Prado, and J. Dawidowski, Nucl. Instrum. Methods Phys. Res., Sect. A **325**, 309 (1993).
[13] R. E. Mayer, P. C. Florido, J. R. Granada, J. Dawidowski, and V. H. Gillette, Physica (Amsterdam) **180B & 181B**, 944 (1992).
[14] I. Kirshebaum, *Physical Properties and Analysis of Heavy Water* (McGraw-Hill, New York, 1951).
[15] *Handbook of Chemistry and Physics*, edited by D. R. Lide (CRC Press, Boca Raton, FL, 1992), 73rd ed.
[16] M. K. Dutta-Choudhury and W. Alexander Van Hook, J. Phys. Chem. **84**, 2735 (1980).
[17] S.W. Lovesey, *Theory of Neutron Scattering from Condensed Matter* (Oxford University Press, Oxford, 1987).
[18] S. F. Mughabghab, M. Divadeenam, and N. E. Holden, *Neutron Cross Sections* (Academic Press, New York, 1981), Vol. 1, Pt. B, p. 79-1.
[19] K. Senoo, Y. Nagai, T. Shima, and T. Ohsaki, Nucl. Instrum. Methods Phys. Res., Sect. A **339**, 556 (1994).
[20] L. Koester, H. Rauch, and E. Seymann, At. Data Nucl. Data Tables **49**, 65 (1991).
[21] D. E. Parks, M. S. Nelkin, J. R. Beyster, and N. F. Wikner, *Slow Neutron Scattering and Thermalization* (W. A. Benjamin, New York, 1970), p. 152.
[22] J. M. Neill, J. L. Russell, Jr., and J. R. Brown, Nucl. Sci. Eng. **33**, 265 (1968).
[23] A. T. D. Butland and C. T. Chudley, J. Br. Nucl. Energy Soc. **13**, 99 (1974).
[24] J. Dawidowski, J. R. Santisteban, and J. R. Granada, Physica (Amsterdam) **271B**, 212 (1999).