Anomalous Deep Inelastic Neutron Scattering from Liquid H₂O-D₂O: Evidence of Nuclear Quantum Entanglement

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A deep-inelastic (Compton) neutron scattering experiment on H_2O-D_2O mixtures at $T = 293$ K has been carried out. Well-resolved H and D neutron recoil peaks in the time-of-flight spectra permit the direct determination of the ratio $Q = \sigma_H/\sigma_D$ of the H and D total cross sections. In contrast to every conventional expectation, Q has been observed to vary strongly with the H-D composition. This striking effect provides, for the first time, direct evidence for short-lived entanglement of adjacent protons (deuterons) in condensed matter at room temperature.   [S0031-9007(97)04226-9]

Dynamics of proton transfer and H bonds in condensed matter is of considerable interest to several fields of physics, chemistry, and biology [1–3]. In particular, quantum aspects of protonic motion (in various systems like, e.g., water, biomolecules, and metallic hydrides) are currently subject to intensive theoretical and experimental investigations (cf. [1,4,5]). These aspects are mainly attributed to the well-known ability of protons to exhibit enhanced quantum tunneling through potential barriers, due to their low mass. Besides quantum aspects of single particles, however, there exist also various quantum correlation (or entanglement, interference, nonseparability, etc.) effects between two or more particles. The latter effects, belonging to fundamental quantum theory, have been studied very intensively during the last two decades (cf. [6–8]).

Our neutron scattering experiment reported in this Letter has been motivated by qualitative theoretical investigations [9] concerning the possibility of short-lived quantum entanglement of adjacent protons in condensed matter at ambient conditions, say $T = 293$ K. Under such conditions, the "lifetime" of the entanglement, i.e., the *decoherence* time τ_{dec} [7], is believed to be much shorter than the present-day time-resolution techniques can resolve, and therefore it is commonly expected to have no experimental significance. In contrast to that expectation, it was suggested [9] that scattering from entangled particles may cause an "anomalous"—according to conventional theory—component in the (light or neutron) scattered field. Intuitively, one may appreciate the possible existence of this new effect, provided that the characteristic "scattering time"—i.e., in simple terms, the time interval during which an incident particle may interact with the scattering center—is not much longer than τ_{dec}. A recent Raman light-scattering experiment [5] on liquid H_2O-D_2O mixtures succeeded to measure the expected anomalous component in the scattered light, thus providing experimental evidence for the quantum entanglement of the fermionic OH- (and bosonic OD-) vibrational states. However, since the light field interacts with the electronic charges of the molecules, this finding is only indirectly related to nuclear quantum entanglement. In contrast, the neutron experiment presented in the following provides for the first time direct evidence for quantum entanglement of adjacent H (and D) nuclei. It should also be mentioned that the intuitive picture of necessarily "overlapping wave functions" of single particles in order to exhibit quantum entanglement is not correct [10].

Let us begin with a description of some essential features of the present experiment and its main result. The used (unpolarized, epithermal) neutrons have energies in the range of several eV. The associated de Broglie wavelength λ_{dB} is of the order of 0.1 Å. The time-of-flight (TOF) spectra of the scattered neutrons from liquid H_2O-D_2O mixtures with a well-defined H-D isotopic fraction N_H/N_D (N_S: particle number density of species S) were measured. The neutrons are scattered from the nuclei only. The very weak neutron-electron interaction can be safely neglected in the present context. The neutron-nucleus scattering time previously mentioned may be given by the time interval $\Delta t = \hbar/\Delta E$ associated with the transition of the nucleus from its initial to its post-collisional state, where ΔE is the energy transferred from the neutron to the nucleus. For example, for a typical value of, say, $\Delta E = 10$ eV one has $\Delta t = 6.6 \times 10^{-17}$ s. The scattering of epithermal neutrons from H and D is inelastic, i.e., kinetic energy is transferred to the recoiling nuclei from the incident neutrons. Furthermore, due to the fact that the λ_{dB} of the neutrons is much smaller than the mean internuclear distances, the scattering looks virtually identical to the scattering that would be observed if it were completely incoherent. The recorded TOF spectra (cf. Fig. 1) yield well-resolved peaks for H and D, due to the large recoil energies (see below for details). The areas A_S ($S = H, D$) under the H and D peaks (recorded by each of the detectors) are then determined. Multiple scattering and/or self-shielding is negligible here (see below). Because of the prevalent scattering conditions, the equations
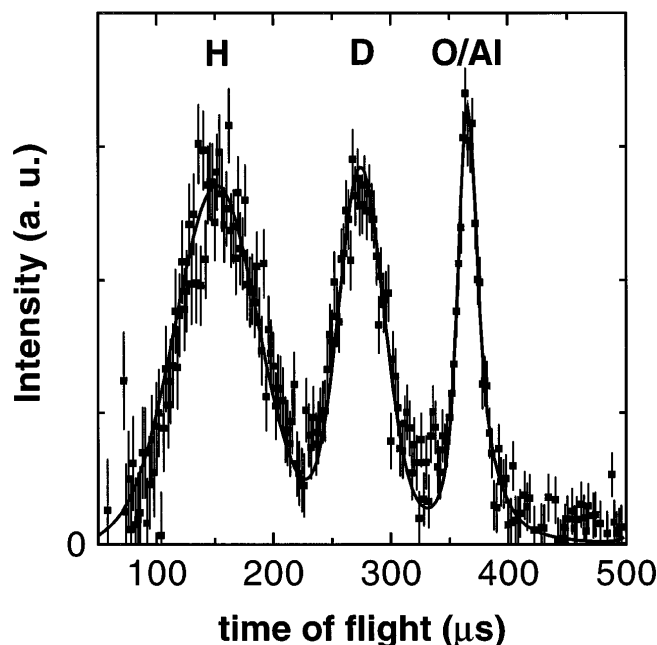
$$A_S = CI_0 N_S \sigma_S \tag{1}$$

FIG. 1. A typical TOF spectrum (full squares) of a H_2O-D_2O mixture with $X_D = 0.8$ as well as the associated fit (full line). The spectrum was recorded during the first experimental period at a scattering angle of $69°$ using a Au-foil analyzer. With increasing time of flight the peaks are due to scattering at H, D, and O/Al nuclei. The O and Al peaks are not resolved. The time of flight is measured between the neutron moderator and the detector. Note that the peak positions are not fitting parameters, but are completely determined by the energy and momentum conservation laws. The distances moderator sample and sample detector were 11.055 and 0.69 m respectively, corresponding to an infinite energy channel of 26.8 μs and an elastic channel of 387.6 μs.

are expected to hold strictly; σ_S ($S = $ H, D) represent the total cross sections of H and D, I_0 the number of incident neutrons, and C, the constant depending on the geometry of the experimental setup. Therefore one obtains

$$A_H/A_D = N_H\sigma_H/N_D\sigma_D \equiv QN_H/N_D, \qquad (2)$$

with $Q = \sigma_H/\sigma_D$. Since the H-D molar fractions of the samples are known with high precision, the numerical value of Q follows immediately from Eq. (2). The most striking feature of our results is that the numerical value of $Q = \sigma_H/\sigma_D$ is not constant for all samples, but appears to depend strongly on the H-D composition of the liquid (cf. Fig. 2). This effect is in clear contrast to every conventional expectation.

We now proceed to the presentation of relevant experimental details. The measurements have been performed at the ISIS pulsed neutron source on the electron volt spectrometer (eVS). The reproducibility of the results has been firmly established through three different experimental series (during March and July 1995 [11], and September 1996; seven days each). This instrument applies a well-known filter difference technique to determine the TOF spectra of the scattered neutrons [12]. From these spectra energy and momentum transfers are deduced, using standard techniques [13]. The first two series of mea-
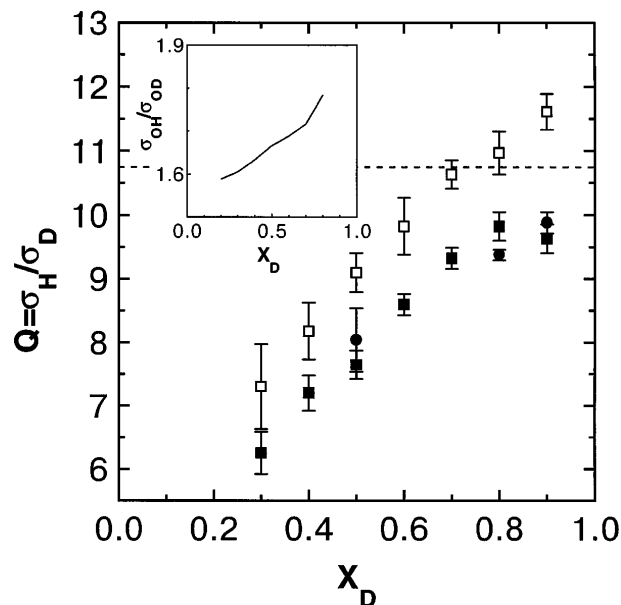

FIG. 2. Dependence of the ratio Q on the deuteron content (X_D) of the mixture. Full (open) symbols represent measurements with Au (U) analyzer (full circles: March 1995; full squares: July 1995; open squares: September 1996). Depicted are the means of 16 detectors and the associated standard deviations as error bars. The dashed line represents the conventionally expected value of Q (= 10.7). The inset shows the associated $Q = \sigma_{OH}/\sigma_{OD}$ values from the Raman light scattering experiment (Ref. [5]).

surements were done using a gold foil analyzer (having a Lorentzian resolution function centered at 4.908 eV and a half width at half maximum, HWHM, of 0.138 eV [14]). The third series was performed using a uranium foil analyzer (with a Gaussian resolution function centered at 6.771 eV and HWHM about 0.062 eV [14]). The U-foil analyzer has a better resolution, but also the disadvantage of producing much weaker signal intensity, being about one-tenth that of the Au analyzer. During the scattering, energy transfers are in the 5–20 eV region, where the so-called *impulse approximation* (IA) [15,16] is known to be a very good approximation in systems containing H and D atoms [17]. The IA is applicable in those cases where the energy transferred from the neutron to the target particle is significantly larger than its binding energy, so that the particle can be considered as quasifree. (The IA is also widely applied in the usual photon Compton scattering experiments.) The well-resolved peaks of H and D in the measured TOF spectra, (cf. Fig. 1) were analyzed using the standard techniques applied at ISIS (cf. [14,16,17]). Here it should be pointed out that the positions of the peak maxima are not fitting parameters, but completely determined by the energy and momentum conservation laws underlying the scattering process; see also point (E).

In the experiments, 16 detectors (being positioned at different scattering angles) and three different liquid sample geometries (provided by cans set perpendicular to the incident neutron beam) were used. In the first experiment we used plane aluminum cans (wall thickness 0.15 mm) of

65 mm height and 65 mm width, providing a sample thickness of ca. 0.2 mm. During the second experiment we used plane vanadium cans (wall thickness ca. 0.15 mm) of 50 mm height, 5 mm width, and a sample thickness of about 0.4 mm. In these two experiments a Au-foil analyzer was used and the detectors were set in the angular range 50° to 75°. In the third experimental period we used plane vanadium cans (wall thickness ca. 0.15 mm) of 70 mm height, 20 mm width, and a sample thickness of about 0.3 mm. In this experiment, the detectors were set in the angular range 30° to 60° and a U-foil analyzer was used. During the last experimental period we also tested the cans used before, showing the results to be independent of the used sample geometry. In all experiments the scattering power was kept at 10% or less, to maintain a sufficiently low level of multiple scattering; see also point (C). As an example, Fig. 1 shows a measured TOF spectrum belonging to the first experiment.

Figure 2 shows the experimental results of $Q = \sigma_H/\sigma_D$ [cf. Eq. (2)] for the different samples. The Q values shown are the mean values taken from the 16 detectors, with standard deviations shown as error bars. The isotopic compositions of the samples, represented by the molar fraction X_D of D of the samples, were determined with accuracy better than 0.3% (and reexamined with densitometry after the completion of the measurements). It should be stressed that the value of Q, as determined by each detector, appears to be *independent* of the scattering angle (i.e., the position of each detector), despite the wide variation in peak separations and associated overlappings. Moreover, the fitted values of the widths of the H and D peaks in individual spectra are also *independent* of the molar composition X_D. As mentioned in connection with Eqs. (1) and (2), the variation of Q with X_D represents the new effect being inexplicable by standard theory, the latter predicting the constancy of Q (with $Q = 10.7$) for every value of X_D.

After close examinations and/or various tests, the following possible errors have been definitely ruled out:

(A) The assumed validity of the IA represents no problem, since deviations from the IA are small and well understood [15,17]. Performed corrections due to the IA changed the values of Q only by a few percent.

(B) The resolution function R of the instrument is incorporated as a convolution, which is accurate only if R does not vary over the width of each peak. For measurements with a Au foil (with Lorentzian R), numerical simulations showed that the Q values may be slightly obscured, since the numerical procedure transfers some of the fitted area of the H to the D peak. Also this effect is small and cannot account for the observations. To test this conclusion experimentally, in the third series of measurements the U-foil analyzer was used (see above), for which these possible artifacts are negligible. Although the corresponding Q data appear to be slightly shifted from the previous results, the crucial feature is that the slope of the whole Q curve (see Fig. 2) remained unchanged.

(C) Monte Carlo simulations incorporating the sample geometries described previously showed that multiple scattering cannot cause any significant change of the Q values. More quantitatively, for a sample with $X_D = 0.5$ and the sample geometry used in the first experimental series, inclusion of multiple scattered neutrons in the simulated data tends to decrease the conventionally expected value of Q only by about 2%. We also made associated test measurements using foils of different thicknesses (about 0.2–0.6 mm) of solid polyethene, showing that multiple scattering effects are not significant. Recall also that the scattering power was always kept about 10% or less.

(D) A correction for the intensity of the incident neutrons, which varies as a function of TOF must also be made. This can be accurately measured by independent calibration measurements [18]. Furthermore the H and D peak positions in the TOF spectra and hence the range of incident energies sampled in the measurements vary strongly with scattering angle. However no variation in the Q values with scattering angle is observed, showing that the incident spectrum shape is accurately described in the fitting programs.

(E) If the H- and D-peak widths, as used in the analysis of the data of all mixtures, were fixed at the values obtained for pure H_2O and pure D_2O, so that each spectrum was fitted only with the heights of the H, D, and O/metal peaks as parameters, the Q values obtained from this procedure were not significantly different than the previous ones. This suggests that correlations between the fitting parameters do not have any significant effect on the results obtained, thus providing further evidence for the validity of the applied data analysis procedure.

(F) Moreover, various tests with other samples (polyethene foils, Pb/Be, and Sn/Be pairs of slabs, with widely differing geometries) gave always ratios of cross sections as conventionally expected. All these tests provided also further evidence that multiple scattering is insignificant in the context under consideration. Very recently, some additional measurements on mixtures of powders of H and D polystyrene were performed, which also gave values of σ_H/σ_D as conventionally expected.

Summarizing, the observed dependence of $Q = \sigma_H/\sigma_D$ on the molar H-D composition of the liquid H_2O-D_2O mixtures represents a striking new effect. This effect is obviously in clear contrast to every conventional expectation, since it is not evident how and/or why Eqs. (1) and (2)—which imply the constancy of Q—could be violated.

In this context, the recent Raman light-scattering experiments on the same mixtures [5] should be mentioned, which revealed a similar anomalous variation of the ratio σ_{OH}/σ_{OD} of the cross sections of the OH and OD vibrational modes (see Fig. 2 inset). The fact that the relevant interactions in these two scattering experiments are completely different—i.e., electromagnetic versus strong interaction—corroborates the conclusion that the considered effect may be caused by *quantum entanglement* (QE),

rather than by quantum interactions. (Recall that QE may exist even in the absence of interactions [10].)

Let us here shortly outline our qualitative theoretical considerations which led us to the performance of this experiment. As a working hypothesis, the possibility of a short-lived and spatially restricted QE between adjacent protons (deuterons) in the liquids may be assumed [5,9]. The QE of the nuclear degrees of freedom can be mediated through the electromagnetic interactions between the electronic charges surrounding the nuclei. Despite the possibly very short decoherence time of such QE's at $T = 293$ K (see the introductory paragraphs), the incident neutrons can "experience" entangled scattering centers, because the duration of the scattering process is also very short. In this case, the scattering process is associated with the value of the matrix element

$$M = \langle \Phi^i_{1,2} | H_I | \phi^f_1 \phi^f_2 \rangle, \qquad (3)$$

where H_I is the interaction Hamiltonian, $\Phi^i_{1,2}$ is the initial entangled state of two H's or D's, and ϕ^f are the final single-particle states [5]. Furthermore, the possibility of QE in the H-D mixtures is certainly restricted due to the spin superselection rule. Proceeding along the theoretical lines of Ref. [5], one can show that M vanishes in certain cases (depending on the intrinsic symmetries of the ground state $\Phi^i_{1,2}$), which then leads to the description of the "anomalous changes" of cross sections, as observed [19,20]. Qualitatively speaking, in those cases where $M = 0$ the incident neutrons cannot interact "properly" with the nuclei, which is tantamount with a change of the apparent cross section [5]. In this context, it should be emphasized that the scattering experiment determines directly the (ratio of the) products $N_S \sigma_S$ ($S = $ H, D), rather than σ_H / σ_D [cf. Eqs. (1) and (2)]. N_H / N_D is known through the sample preparation. Thus the presented experimental results can be alternatively viewed to be related with "effective changes" of N_H and N_D being caused by attenuated (enhanced) QE of protons (deuterons) in the H_2O-D_2O mixtures [20]. For an introduction to multiparticle QE, see [21].

Further work, as well as related neutron reflectivity and interferometry experiments, are presently in progress. The effect reported in this Letter may have considerable implications in various physical, chemical, and biological subfields.

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