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Quantum coherence and decoherence of protons and muons in condensed matter

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Abstract. The coherence in quantum superposition states of protons (and chemically similar particles, the positive muons) has been studied in some condensed matter environments. It is shown that if the proton systems and the experimental techniques used to study them are carefully selected, it is possible to observe quantum delocalization states of single particles and to understand the mechanisms for their loss of coherence. Quantum correlated two- and multiparticle states of protons lose coherence very fast when coupled to condensed matter environments, but new sub-femtosecond techniques have made them accessible to experimental studies. The degree of decoherence can be measured as function of time and the decoherence mechanisms can, at least in certain cases, be identified. Although less clean than in corresponding studies of quantum optical systems, these studies can be seen as a first step towards understanding the conditions for preservation of quantum correlation and entanglement in massive systems. Some consequences and some suggestions for future work are discussed.

PACS. 03.65.-w Quantum mechanics – 61.12.-q Neutron diffraction and scattering

1 Introduction

The great majority of all experiments and discussions on the specific aspects connected with the superposition principle of quantum mechanics originate from the field of quantum optics; one reason being that photons interact sufficiently weakly with their environments to keep quantum coherence for times long enough to observe interferences and other characteristic effects of quantum superposition and entanglement. Photons are also easy to handle, manipulate and detect with modern techniques.

Entangled states of atoms and photons have been kept alive for times of the order of microseconds [1], but only when the atoms are extremely well isolated in cavities and with a limited number of photons present. In double-slit experiments on atoms and molecules [2], interaction with the environment must also be kept at a minimum. As soon as atoms are brought in closer contact, particularly in different forms of condensed matter, quantum coherence between them is lost within a very short time and superposition effects are washed out.

It is however, appropriate to remember that a change of phase relations within a certain subset of a multi-particle system actually takes a finite time (longer if the subsystem considered is small and weakly coupled to the rest of the system, as expressed for instance by the Joos-Zeh relations [3] for quantum decoherence of particles of different sizes embedded in environments of different strengths). But, except in the specific cases of BE-condensates and superfluidity, one has to use probes of extremely short "exposure time" in order to observe effects of the superposition principle in condensed matter systems. The present work deals with a couple of such methods and some consequences that can be drawn from such experiments about the indivisibility of complex systems.

The studies discussed here are limited to hydrogen atoms placed in different environments. Hydrogen isotopes (by which we will here also include the positive muon, which chemically acts as a lightweight proton) are likely to show the strongest quantum effects owing to their low masses; one aspect of this being their relatively large thermal de Broglie-wavelengths, of the order of Angstroms at room temperature.

First, single particles will be considered, where a proton or a positive muon is delocalized over two or more sites in a tunneling state. The conditions for survival of this kind of superposition states will be discussed. Secondly we will consider two-particle states where neighbouring pairs of protons or deuterons are correlated due to quantum exchange. Examples show how interaction with the local environment phases out this kind of multi-particle superposition states within a few femtoseconds.

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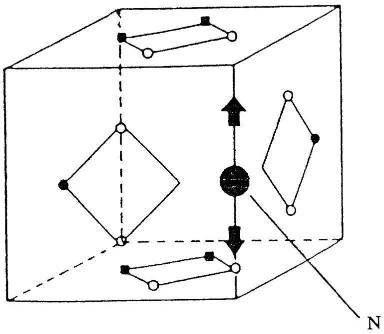

Fig. 1. H-sites near a N-impurity in bcc Nb. H-sites shown by the same symbols have identical energies.

2 Protons and positive muons in tunneling states

In certain kinds of crystals, an implanted proton or positive muon can be found in a tunneling state, where it occupies simultaneously two or more identical interstitial sites between the host atoms. One example is a Nb-crystal weakly doped with N-atoms that substitute for Nb at certain positions in the bcc Nb-lattice (see Fig. 1). If a H-atom is also introduced, it will be trapped near an N-atom, where there are pairs of interstitial H-sites which are energetically equivalent and separated by a relatively weak potential barrier. The result is that the proton can tunnel between the two sites 1 and 2, a situation equivalent to the well-known quantum double well problem, and the proton finds itself in a quantum superposition state

$$\Psi_s = \left(1/\sqrt{2}\right)\{\Psi(1)+\Psi(2)\};$$
$$\Psi_a = \left(1/\sqrt{2}\right)\{\Psi(1)-\Psi(2)\}\cdot \qquad (1)$$

This kind of state was first studied by Wipf *et al.* [4] using inelastic neutron scattering. Its character is governed by a tunneling matrix element J for the proton. Since in the practical case the tunneling of a proton "carries with it" a deformation of the crystal lattice and (in metallic systems) a screening electron cloud, phononic and electronic properties are also strongly involved. This give rise to a reduction of the "bare" matrix element, ($J \rightarrow J_{\rm eff}$), but also to strong mechanisms for quantum decoherence. The energy difference between the two "tunnel-split" states in equation (1), which is equivalent to $2J_{\rm eff}$, was determined from the inelastic neutron scattering to be 0.23 meV. When there are transitions between the two states the inelastic neutron scattering peak is lifetime broadened. Its width Γ gives a measure of the "lifetime" of the tunneling states ($\Gamma = \hbar/\tau_{\rm coh}$, where $\tau_{\rm coh}$ is the characteristic time over which quantum coherence in Eq. (1) is preserved).

Wipf *et al.* first followed the temperature dependence of $\Gamma = \hbar/\tau_{\rm coh}$ at low temperatures in normal conducting Nb and found it to be a linear function of T (Fig. 2a). Such a linearity is expected, according to Kondo [5], if the temperature is low enough for the decoherence mechanism to be mainly caused by the interaction with the surrounding conduction electrons. At higher temperatures phonons are excited, which are expected to give a lifetime broadening Γ proportional to a higher power of T. The explicit expression for the conduction induced decoherence rate $1/\tau_{\rm coh}$, as given by Kondo, is

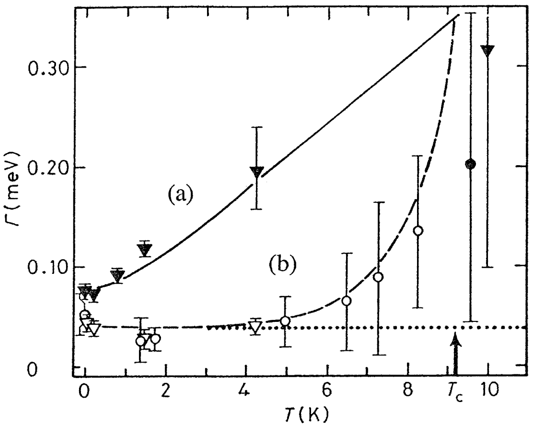

Fig. 2. The linewidth $\Gamma = \hbar/\tau_{\rm coh}$ measured in neutron scattering on H in Nb-metal, showing (a) the linearity with T in the normal conducting state (with $\tau_{\rm coh}$ of the order of 10^{-13} s) and (b) the strong reduction of Γ (*i.e.* increase of $\tau_{\rm coh}$) in the superconducting state. The vertical arrow marks the instrumental linewidth. From reference [8].

$$1/\tau_{\rm coh} = \pi K k_{\rm B} T/\hbar \qquad (2)$$

where K is a strength parameter for conduction electrons scattering on protons (it can be expressed in terms of phase shifts and a scattering potential [6]). For a temperature of 1 K, decoherence rates of the order of 10^{11} s^{-1} are expected.

The disappearance of the tunneling state as a result of interaction with the environment can be illustrated by plotting the probability for the particle to be at one of the two sites (compare the text-book example of the N-tunneling in the NH$_3$ molecule in Ref. [7]) as function of time. The tunneling frequency is $\omega = J_{\rm eff}/\hbar$. The perturbation of the tunneling state is connected with a spread σ_ω in ω caused by energy changes in the two wells (in the actual case by the conduction electron interaction whose fluctuation is proportional to kT in the metallic state). The spread in ω makes the oscillation decay with a Gaussian envelope function

$$f(t) = \exp[-\sigma_\omega^2 t^2] \cos\omega t. \qquad (3)$$

If $t > 1/\sigma_\omega$ this oscillation will be strongly damped (see Fig. 3) and the particle will soon be localized in one of the sites. The tunnel-split state can exist only at low temperatures, where $\tau_{\rm coh}$ is long.

Particle states in this kind of environment provide some of the best understood examples of mechanisms for quantum decoherence. In further experiments it has been shown [8] that if the Nb-metal is in the superconducting state (it is possible to switch between the superconducting and the normal state by applying a magnetic field) the decoherence rate is drastically reduced because of presence

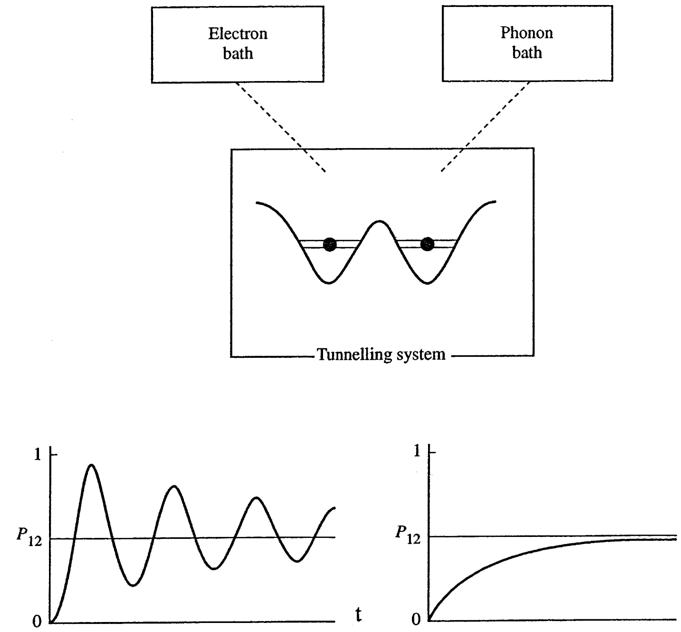

Fig. 3. The probability for finding a proton in potential well 2 after starting in well 1, when damped by decoherence induced by interaction with phonon or conduction electron baths (sketched above). The right hand figure represents overdamping.

of the energy gap $\Delta_{\rm sc}$ for the conduction electrons. The density of states for electrons at the Fermi level, which are the "active" ones in the interaction with the protons, is then changed such that

$$1/\tau_{\rm coh} = \pi K k_{\rm B} T \Big/ \hbar\{1 + \exp[\Delta_{\rm sc}(T)/k_{\rm B}T]\} \tag{4}$$

which means that not only the phonon bath but also the electron bath is effectively decoupled at $T \ll T_{\rm sc}$ in Nb-metal (see Fig. 2b).

Similar experiments have been made on positive muons implanted in metallic host crystals. The effective tunneling matrix elements $J_{\rm eff}$ for muons are essentially larger than for protons in the same potential wells (their mass is about 10 times lower) and positive muons can be delocalized over several interstitial sites if the coherence conditions allow it. The mobility and localization of positive muons was first followed by Hartmann *et al.* [9] from high to low temperatures, 100 K → 0.03 K in a normal conducting metal, through the phonon-dominated down to the electron dominated range. In the latter, the muons can tunnel from site to site, but they are in a quantum diffusion regime. This is not a proper tunneling state of the type described by equation (1), since coherence is lost between each tunneling transition ($\tau_{\rm coh} < 1/\omega$), but for muons in Al, it was later shown by Karlsson *et al.* [10,11] that there is a dramatic difference when the metal becomes superconducting below 1.2 K. Equation (4) then allows the muons to be delocalized over times as long as 10^{-6} s in quantum superposition states. These involve several crystallographic sites at the lowest temperatures. Well below $T_{\rm sc}$ the muons can be said to be in propagating states, similar to those for conduction electrons in a metal.

In μSR (muon spin rotation) one measures the spin depolarization rate $P_z(t)$ of the muons. The signature of a completely delocalized state is a time-independent function $P_z(t)$. In Figure 4 (up and left), this state does not exist because the 75 ppm Li-impurities destort the Al-lattice and destroy the similarity between the muon sites (and therefore the tunneling possibility); in Figure 4 (down and left, up and right) an increasing fraction of the implanted muons become delocalized and when impurities are practically absent, Figure 4 (down and right), delocalization is complete. Muons in propagating states can be scattered elastically by impurity atoms present in the metals [11], but elastic collisions do not lead to loss of coherence.

3 Proton and deuteron pairs in quantum superpositions

So far, only the coherence in delocalized one-particle states has been treated. Two or more particles can exist in quantum superposition states, not describable by a simple product of wavefunctions for the separate particles but only by a superposition of such products. The simplest example is the wavefunction for two indistinguishable particles α and β, situated at two positions 1 and 2. If their spin is I which can be coupled to a total spin of $\mathbf{J} = \mathbf{I} + \mathbf{I}$, the antisymmetry rules forces them to have a wavefunction of the form

$$|i\rangle = \left(1/\sqrt{2}\right)\{\phi_1(\mathbf{R}_\alpha)\phi_2(\mathbf{R}_\beta) + \zeta\phi_1(\mathbf{R}_\beta)\phi_2(\mathbf{R}_\alpha)\}\chi^J_M(\alpha,\beta) \tag{5}$$

where the spatial coherence is described by the phase factor $\zeta = (-1)^J = \exp(\pm i\pi/2)$ and χ^J_M is the coupled spin function. For electrons, the exchange correlation is a basic ingredient in all quantum calculations, but it is usually neglected in the description of the nuclear part of the wavefunctions in molecules and solids. Only in certain specific situations has it been necessary to take into account exchange correlations for pairs of nuclei (hydrogen) in order to explain experimental data; the classical example being the thermal neutron scattering on hydrogen molecules [12]. The cross-sections for $J = 0$ and $J = 1$ are dramatically different, effects that can be traced back to the indistinguishability of the two scattering particles.

In principle, non-separability may not only be the result of coupling spatial nuclear wavefunctions to a common total spin (as in the exchange correlation above), but also to other shared degrees of freedom such as phononic or electronic excitations, in analogy to the atom-photon-atom entanglement often studied in quantum optics. However, the phase factors in such coupled states will not have the simple form that is valid for the exchange interaction in equation (see Eq. (5)), but must be evaluated from an explicit model for the particular entanglement mechanism.

Not unexpectedly, the quantum superposition states of two- and more atoms or nuclei are strongly perturbed

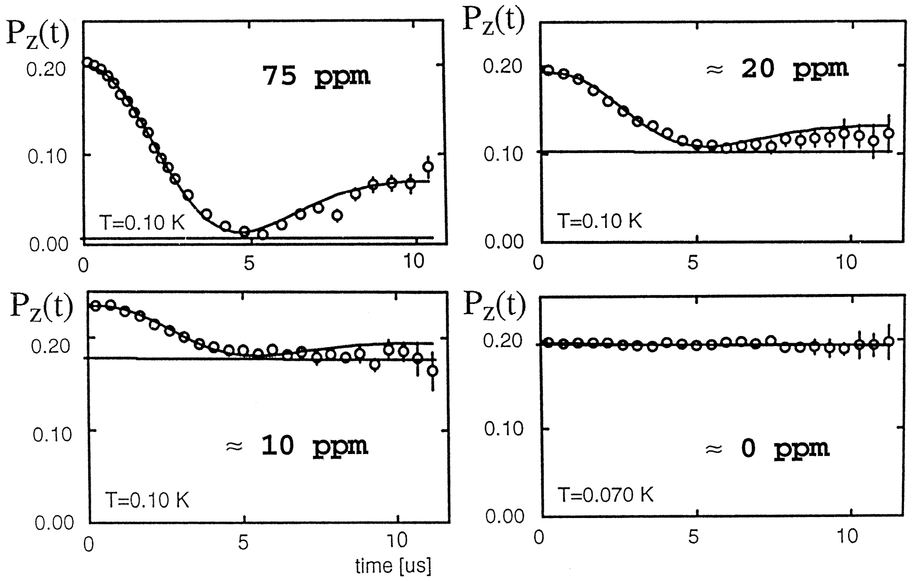

Fig. 4. Muon depolarization in Al for different sample conditions (see text). In down and right panel, depolarization is absent, which means that the muons stay in a coherent, strongly delocalized state in the purest aluminium over their whole lifetime (2×10^{-6} s), when T is well below the superconducting transition. From reference [10].

by environmental interactions. The quantum phase relations that exist at one particular moment are easily lost, particularly in condensed matter systems. Normally, coherence times may not exceed a few femtoseconds even for molecular-sized quantum systems, and for macroscopic ones (like the grains of dust considered in simple decoherence models [3,13]) overall coherence has been estimated to be preserved only for 10^{-20} s or less. Experimental methods that allow the study of local few-proton correlations (or entanglement) must therefore take "snapshots" with time windows of 10^{-15} s or less. Up to now, two methods with characteristic time windows in this range, Compton scattering with neutrons and Raman scattering [14], have been used to look for proton and deuteron quantum correlations in condensed matter.

In neutron Compton scattering (NCS), neutrons of much higher energy, 10–100 eV, than those normally employed in thermal scattering (≈ 0.02 eV), scatter on the nuclei in the sample. This leads to a recoil of the nuclei and an outgoing neutron with reduced energy and momentum. Scattered neutrons of one particular energy are selected by a resonance method [15] and the kinetics is such that, at each neutron scattering angle θ, the corresponding energy for the incoming neutrons (and therefore the total time-of-flight from the production target) is characteristic for each type of scattering nuclei. In the time-of-flight spectrum, the intensity for scattering on H, D, C, etc., can therefore be obtained separately. Furthermore, each particular scattering angle θ for each scattering mass M is associated with a characteristic scattering time [16]

$$\tau_{\mathrm{sc}}(\theta) \approx M/(k(\theta))\sqrt{\langle p^2 \rangle} \tag{6}$$

which can be calculated from known parameters ($k(\theta)$ is a trigonometric function). The times τ_{sc} fall in the range $\approx 10^{-16}$–10^{-15} s, depending on the mass of the scattering particles and the average momentum $\sqrt{\langle p^2 \rangle}$ that the particle has in its thermal motion when hit by the neutron. This does not only provide a suitable time window for observation of the correlation effects discussed here, but also a possibility for a kind of time differential measurement of the neutron scattering cross-sections: the scattering intensities $I(\theta)$ can be transformed to functions of scattering time, $I(\tau_{\mathrm{sc}})$; and directly from the spectra it is possible to derive cross-section ratios $\sigma_{\mathrm{H}}(\tau_{\mathrm{sc}})/\sigma_{\mathrm{M}}(\tau_{\mathrm{sc}})$, where M represents some heavier nucleus in the sample. It has been observed in several measurements on condensed systems: water [17] and different polymers [18,19] that the H-cross-sections are lower by 20−30% than expected for individually scattering protons as given in standard tables. For scattering on protons and deuterons in metallic hydrides like NbH$_x$ and PdH$_x$ [20,21] it has also been observed how the anomalous cross-section ratios $\sigma_{\mathrm{H}}/\sigma_{\mathrm{M}}$ measured at shorter times τ_{sc} approach the tabulated values when τ_{sc} reaches times of the order of 10^{-15} s (see Fig. 5). The latter is a clear indication that the anomalies have to do with a quantum state that decays very fast as coherence is lost in the proton subsystem.

The theoretical cross-sections for pairs of exchange correlated nuclei under Compton scattering conditions have been derived by Karlsson and Lovesey [22,23]. The starting point is equation (5) which represents the initial state, and the final state was assumed to have the form

$$\left(1/\sqrt{2}\right)\{\exp(\mathrm{i}\mathbf{p}' \cdot \mathbf{R}_\alpha)\Psi(\mathbf{R}_\beta) + \zeta' \exp(\mathrm{i}\mathbf{p}' \cdot \mathbf{R}_\beta)\Psi(\mathbf{R}_\alpha)\}\chi^{J'}_{M'}(\alpha,\beta) \tag{7}$$

with the recoiling particle represented by a plane wave. It should be noted that in this expression, where Ψ stands for the state of the non-recoiling member of the pair, the quantum correlations are not yet broken and α and β are not yet distinguishable; this occurs only later, through a collapse process in the interaction with the condensed matter environment. With this Ansatz, and the usual scattering operator $V = b_\alpha \exp(\mathrm{i}\mathbf{k} \cdot \mathbf{R}_\alpha) + b_\beta \exp(i\mathbf{k} \cdot \mathbf{R}_\beta)$ the matrix elements $\langle f|V|i\rangle$ are evaluated as

$$\langle f|V|i\rangle = \left(1/\sqrt{2}\right)\left\{\chi^{J'}_{M'}(\alpha,\beta)[b_\alpha + \zeta\zeta' b_\beta]\chi^{J}_{M}(\alpha,\beta)\right\} \times K(\mathbf{p})[T_2 + \zeta\exp(-\mathrm{i}\mathbf{p}\cdot\mathbf{d})T_1]. \quad (8)$$

Here, $K(\mathbf{p})$ describes the so-called Compton profile (shape of the momentum distribution in the outgoing neutron spectrum) and T_i are overlap integrals $\int \mathrm{d}\mathbf{R}\Psi^*(\mathbf{R})\phi_i(\mathbf{R})$, which to a good approximation have the values $1/\sqrt{2}$. The cross-sections are dependent on how the momentum of the local vibration $\mathbf{p}$ is related to the vector $\mathbf{d} = \mathbf{R}_1 - \mathbf{R}_2$ connecting the two nuclear sites. For half-integer values of particle spins ($I = 1/2$ for protons), one has, per particle,

$$\sigma/\sigma_{\mathrm{sp}} = (1/2)(2I+1)^{-1}\{I|T_1 + \exp(\mathrm{i}\mathbf{p}\cdot\mathbf{d})T_2|^2 + (I+1)|T_1 - \exp(\mathrm{i}\mathbf{p}\cdot\mathbf{d})T_2|^2\} \quad (9)$$

if no $J' \neq J$ selection rule is imposed [23]. The maximum cross-section reduction for protons occurs if $\mathbf{p}$ is perpendicular to $\mathbf{d}$ (1/4 of the standard value σ_{sp}), whereas for isotropic vibrations a reduction to 1/3 is expected. For deuteron pairs, a formula analogous to equation (9) also predicts reductions, but of lower magnitude. If $|f\rangle$ is not a superposition, but a simple product state of particles α and β, the result of the calculation is the standard cross-section, as expected.

In Compton scattering, the decoherence rate is strongly influenced by the after-effects of the recoil process itself. For protons in the metal hydrides mentioned above, where the decoherence time has been measured to be 0.6×10^{-15} s (*cf.* Fig. 5), the spatial coherence of the protons is lost primarily by the shake-up when the recoiling proton leaves the scattering sites (it takes about one femtosecond for the proton to move a distance of 0.5 Å). The coherence time expected when the proton subsystem is perturbed only by the natural processes is expected to be essentially longer, but still shorter than the 10^{-13}–10^{-12} s typical of thermal neutron scattering (for which no H cross-section anomalies are observed in the metal hydrides).

The situation in water is particularly well suited for analysis of the short-time quantum correlations and their disappearance by use of the explicit theoretical model [23], valid for a pair of correlated nuclei. The interaction of the protons with their environment in water is also well studied, particularly by infrared and Raman spectroscopy [24]. The H-vibrations in water have such values for $\sqrt{\langle p^2\rangle}$ that the time range for observation according to equation (6) lies as low as at 5×10^{-17}–5×10^{-16} s. Over this interval, the correlations in equations (1, 2) are still well developed and the cross-section reductions observed by Dreismann *et al.* [17] could be explained quantitatively [25] by the Karlsson-Lovesey theory assuming fully correlated protons in H_2O.

The decoherence of the proton-proton correlation in water is caused by surrounding water molecules which perturb the vibrations in the H_2O molecule. It is only when the vibrations are perturbed randomly by the hydrogen bonds to neighbouring molecules, and many environmental modes are mixed in, that the quantum phase relations are lost. Each hydrogen bond represents a potential well with a characteristic bond frequency around the value ω_σ (*cf.* Fig. 6). This broadens the stretching vibration ω_s to a band with frequency $\omega_s \pm \omega_\sigma$ (where ω_σ is about one tenth of ω_s). On an energy scale, the band width is known from vibrational spectroscopy [26] to be about 0.03 eV.

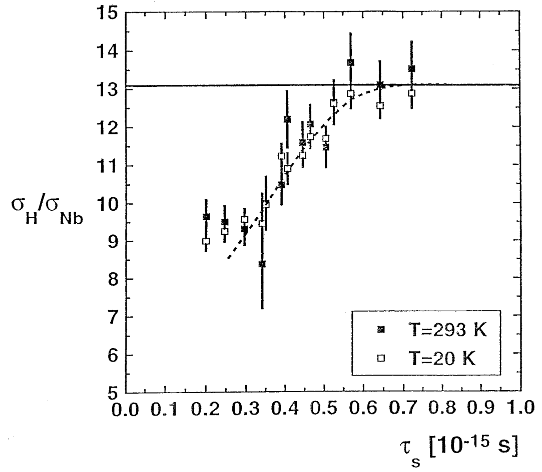

Fig. 5. The observed cross-section ratio $\sigma_{\mathrm{H}}/\sigma_{\mathrm{Nb}}$ in the metal hydride NbH as function of scattering time. From reference [20].

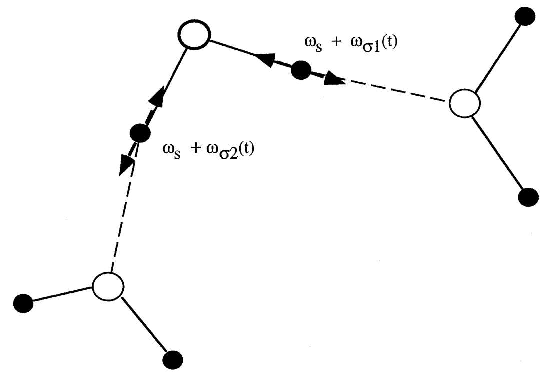

Fig. 6. Water molecule with coupling to a fluctuating environment through hydrogen bonds.

The spatial part of the final state in equation (7) can be written as a superposition of states with particle α starting from $\mathbf{R}_\alpha = \mathbf{R}_1$ (with β staying at site $\mathbf{R}_\beta = \mathbf{R}_2$) and α starting from $\mathbf{R}_\alpha = \mathbf{R}_2$ (with β remaining at site $\mathbf{R}_\beta = \mathbf{R}_1$), and since the Ψ's in equation (7) can be expressed as $\Psi = T_1\phi_1 + T_2\phi_2$,

$$|f'\rangle = \left(1/\sqrt{2}\right)\{\exp[\mathrm{i}\mathbf{p}'\cdot(\mathbf{R}_\alpha - \mathbf{R}_1)]T_2\phi_2(\mathbf{R}_\beta) + \zeta'\exp[\mathrm{i}\mathbf{p}'\cdot(\mathbf{R}_\alpha - \mathbf{R}_2)]T_1\phi_1(\mathbf{R}_\beta)\}\cdot \quad (10)$$

This is one of two possible final state (without environmental interaction). The other one, $|f''\rangle$, is obtained by interchanging the indices.

When the water proton at site 1 is coupled to a stretching vibration with angular frequency $\omega_1 = \omega_s + \omega_{\sigma_1}$

and the one at site 2 to a vibration with $\omega_2 = \omega_s + \omega_{\sigma 2}$, the coupled wavefunction is obtained by multiplying each spatial single particle wavefunction by oscillating terms, $\exp(\mathrm{i}\omega_1 t)$ and $\exp(\mathrm{i}\omega_2 t)$. From equation (10) one obtains the form (extracting a common phase factor $\exp[\mathrm{i}\omega_1 t]\exp[-\mathrm{i}\mathbf{p}'\cdot\mathbf{R}_1]$),

$$\begin{aligned}(1/\sqrt{2})\{&\exp[\mathrm{i}\mathbf{p}'\cdot\mathbf{R}_\alpha]T_2\phi_2(\mathbf{R}_\beta)\\ &+\zeta'\exp[\mathrm{i}(\omega_{\sigma 2}-\omega_{\sigma 1})t]\exp[\mathrm{i}\mathbf{p}'\cdot\mathbf{d}]\\ &\times\exp[\mathrm{i}\mathbf{p}'\cdot\mathbf{R}_\alpha]T_1\phi_1(\mathbf{R}_\beta)\}\cdot \qquad (11)\end{aligned}$$

If now, in the last line, $\exp[\mathrm{i}\phi]$ is introduced for $\zeta' = (-1)^{J'}$, where $\phi = \pi$ for J' odd and $\phi = 0$ for J' even, equation (11) displays how the phase factor in the spatial part of the two-proton wavefunction disappears and reappears with the factor $\exp[\mathrm{i}\{\phi + (\omega_{\sigma 2} - \omega_{\sigma 1})t]$. But in the fluctuating network of hydrogen bonds in liquid water, the factor $(\omega_{\sigma 2} - \omega_{\sigma 1})$ charges randomly, with an r.m.s.-value of σ on a time-scale which is of the same order of magnitude as the vibrational period itself. The energy spread of 0.03 eV mentioned above corresponds to $\sigma \approx 0.5 \times 10^{14}$ s^{-1}. If the distribution in $\omega \approx \omega_s$ can be represented by a Gaussian function one obtains therefore for the mean value of the phase factor,

$$\int \mathrm{d}\omega' \exp[(\omega' - \omega)^2/4\sigma]\exp(\mathrm{i}\omega' t) = \exp[-\sigma^2 t^2]\exp(\mathrm{i}\omega t) \qquad (12)$$

where the decay factor $\exp[-\sigma^2 t^2]$ measures the decoherence (like the damping factor $\Delta\omega$ in the oscillating amplitude of the tunnel-split state in Eq. (3)). From the known value of σ the characteristic decoherence time for the proton-proton correlation water, as seen by the neutron, can therefore be estimated to be $\tau_{\mathrm{coh}} = 2 \times 10^{-14}$ s. After a complete loss of phase memory the end result is one of the two simple product states

$$\begin{aligned}|f'\rangle &= (1/\sqrt{2})\exp[\mathrm{i}\mathbf{p}'\cdot\mathbf{R}_\alpha]T_2\phi_2(\mathbf{R}_\beta)\\ \text{or } |f''\rangle &= (1/\sqrt{2})\exp[\mathrm{i}\mathbf{p}'\cdot\mathbf{R}_\beta]T_1\phi_1(\mathbf{R}_\alpha) \qquad (13)\end{aligned}$$

where either particle α *or* particle β has been identified as the one leaving the molecule in the recoil process in the Compton scattering

As mentioned briefly already, the Compton scattering of neutrons (NCS) has the disadvantage that it actively contributes to the decoherence process through the violent shake-up following the recoil. Actually, a softer scattering process should be needed to follow the natural decoherence process in water. It is hoped that data from inelastic neutron scattering on water could be helpful to fill in data for the decoherence in the 10^{-14} s range. It works in the transferred momentum range $Q = 5$–20 Å^{-1}. Below $Q \approx 15$ Å^{-1} the whole H_2O molecule recoils with an energy less than 0.3 eV (much lower than the 50 eV typical for the liberated protons in NCS), From the neutron coherence length $\lambda_{\mathrm{coh}} = \lambda^2/2\Delta\lambda$ estimated from the wavelength resolution $\Delta\lambda$ and the velocity v of the neutrons, it can be concluded that the scattering time (and therefore the time window for observation) under these conditions should be in the range 2–100 fs. A decrease of the intensities of neutrons scattered on protons, when moving from the long observation times at low Q, to the shorter ones at higher Q (where decoherence processes have not had time to operate) should be expected. From the empirical evidence gained from reference [20] the corresponding decrease with Q is expected to be much less evident (or absent) for scattering on D_2O. It should be mentioned here that another neutron scattering experiment (on protons in $KHCO_3$) by Ikeda and Fillaux [27] has shown exchange-related interferences at $Q \approx 5$ Å^{-1} for a scattering time that also falls in the 100 fs range. Deuterons in $KDCO_3$ showed no such effect.

If measurement conditions are chosen properly, it might be possible to observe effects of quantum correlated proton states even with the use of thermal neutrons, not only in the H_2 molecules mentioned, but also in other molecules. Returning to the correlated two-particle model, the final state will have the form, for "soft" encounters, where there is no recoil effect or shake-up (only the possibility of a spin flip which changes J to J' and ζ to $\zeta' = (-1)^{J'}$),

$$\begin{aligned}|f\rangle = |\gamma', JM\rangle = (1/\sqrt{2})\{&\phi_1(\mathbf{R}_\alpha)\phi_2(\mathbf{R}_\beta)\\ &+\zeta'\phi_1(\mathbf{R}_\beta)\phi_2(\mathbf{R}_\alpha)\}\chi^{J'}_{M'}(\alpha,\beta) \qquad (14)\end{aligned}$$

as long as there is no decoherence. The same procedure as the one giving equation (8) for the Compton scattering will now give the following expression for the matrix elements,

$$\begin{aligned}\langle f|V|i\rangle = (1/2)\{&\langle\gamma'|\exp(\mathrm{i}\mathbf{q}\cdot\mathbf{R}_\alpha)|\gamma\rangle\langle J'M'|b_\alpha|JM\rangle\\ &+\langle\gamma'|\exp(\mathrm{i}\mathbf{q}\cdot\mathbf{R}_\beta)|\gamma\rangle\langle J'M'|b_\beta|JM\rangle\\ &+\zeta\zeta'\langle\gamma'|\exp(\mathrm{i}\mathbf{q}\cdot\mathbf{R}_\alpha)|\gamma\rangle\langle J'M'|b_\alpha|JM\rangle\\ &+\zeta\zeta'\langle\gamma'|\exp(\mathrm{i}\mathbf{q}\cdot\mathbf{R}_\beta)|\gamma\rangle\langle J'M'|b_\beta|JM\rangle\cdot \qquad (15)\end{aligned}$$

Here, $\langle J'M'|b_\alpha|JM\rangle = \chi^{J'}_{M'}(\alpha,\beta)b_\beta\chi^{J}_{M}(\alpha,\beta)$, etc., and $\mathbf{q}$ is the momentum transfer. Noting that $\langle J'M'|b_\beta|JM\rangle = \zeta\zeta'\langle J'M'|b_\alpha|JM\rangle$ (see Ref. [22], Eq. (4.1)) and using the usual expression for the scattering amplitude operators, $b_i = A(i) + (1/2)B(i)\mathbf{s}\cdot\mathbf{I}$, $i = (\alpha, \beta)$, one recovers the standard expressions for the thermal neutron cross-section of the exchange correlated protons in the hydrogen molecules (in their ground vibrational and rotational states and for $\mathbf{R}_\alpha + \mathbf{R}_\beta = 0$ and $\sin[(1/2)\mathbf{q}\cdot(\mathbf{R}_\alpha - \mathbf{R}_\beta)] \ll 1$, see Ref. [28], Chap. 1.7):

$$\sigma(J=0) = 4\pi A^2;\ \sigma(J=1) = 4\pi\left[A^2 + (1/8)B^2\right].$$

It is more interesting, however, to consider what would happen in an experiment with neutron reflection, where quantum interference enters in a different way. It is textbook knowledge that only the A-terms in $b_i = A(i) + (1/2)B(i)\mathbf{s}\cdot\mathbf{I}$ contribute to the coherent scattering, and therefore to the reflected intensity. A commonly used explanation for the absence of the B-term is (see, *e.g.*, Segre [29], p. 570) that, as soon as a spin-flip has occurred the scattering site for the incoming neutron has been identified, and there is no interference left to build up a reflected wave.

In the exchange correlated two-particle model, the following matrix elements will enter the quantities $Q(1)$ and $Q(2)$, whose correlation describes the reflected intensity,

$$Q(1) = (1/2)[b_\alpha(1) + \zeta\zeta' b_\beta(1)]$$
$$Q(2) = (1/2)[b_\alpha(2) + \zeta\zeta' b_\beta(2)]. \qquad (16)$$

In the formulation of the coherent cross-section, there is then a contribution also from the spin-flip B-terms to the correlation $\overline{Q(1)} * \overline{Q(2)}$, which does not occur for scattering on uncorrelated particles. For protons, the B-terms are exceptionally large compared to the A-terms ($A = -0.38 \times 10^{-12}$ cm, $B = 5.78 \times 10^{-12}$ cm) and even if only a small fraction of the coherence remains over times characteristic for thermal scattering, the effect of the B-terms might be detectable. For deuterons, where the A-term is dominating, no such effects are expected to be visible.

Reflectivity experiments on water surfaces have actually been performed by Streffer *et al.* [30], with an indication that the reflectivity is increased by about 10% compared to what is expected from standard theory. In the light of what has been said above about the decoherence time for exchange correlated protons in H_2O, such a result is not unreasonable; if the decoherence time is 2×10^{-14} s and the scattering time is 10 times longer, the effect of the B-terms would have diminished about two orders of magnitude, but since it is very large initially it could still contribute in a measurable way. It would be interesting to have this observation confirmed by other measurements, since it means that even for scattering times of the order of 10^{-13} s there would be reminiscences of the relative quantum phase that the protons had in water at the beginning of the neutron encounter. The effect itself is a subtle feature of quantum mechanics that can described in words by saying that "a spin-flip can occur without identifying a specific scattering site for a particle, by which 'incoherent B-terms' become coherent".

4 Discussion

The topics treated here may give rise to a couple of speculations of more general nature. One interesting point in the treatment of the water molecule is that, during one femtosecond (and when unperturbed by a measurement, probably for several femtoseconds), the two protons must be looked at as indistinguishable particles. When the molecule is attached to two other neighbouring water molecules through hydrogen bonds, there is actually a bridge through the two correlated protons similar to that of an EPR pair of photons in quantum optics. This is a coupling of a specific quantum character that might have an influence on the hydrogen bonding. In particular, these very ephemeric couplings may be of importance in the first preparatory stages of chemical reactions involving protons, perhaps also in reactions in biomolecular systems.

Another topic that deserves some attention is the fact that the times within which two- and multiparticle states stay coherent is generally shorter than the times it takes to establish thermal equilibrium in condensed systems. This important difference between the quantum dephasing time and the thermal relaxation times is also stressed in the works by Joos and Zeh [3] and by Zurek [31]. The results of the neutron Compton scattering experiments, with their time window below 10^{-15} s, have actually turned out to be independent of temperature (from 300 K down to 20 K for NbH_x [20]). This is expected since these times are clearly "subthermal" with respect to the times of the order of 10^{-13} s needed for thermal equilibrium.

Decoherence also plays a key role in the thermodynamic definition of time. The loss of phase memory through decoherence introduces a time asymmetry and makes retrodiction impossible. From the point of view of the local system, unitarity seems to be lost (although it is maintained when the whole chain of couplings to further and further external systems is included). It is interesting to note that, in this respect, the flow of time is more discontinuous in the local proton subsystem, where time symmetry is preserved over steps of the order of femtoseconds, than it is for the time as seen from the point of view of a macroscopic system.

The above examples have shown that the processes by which coherence is lost can be measured and understood, at least if the quantum systems are simple and the environment is well defined. Lastly, therefore, it may be remarked that the examples mentioned here may help to throw light on the quantum measurement problem, in particular at what level the "measuring apparatus" selects a particular value from the possibilities inherent in a quantum superposition. It is evident that even for a quantum object exceeding a few atomic distances in size, in contact with a condensed matter environment, this is a very fast process. For any other, reasonably sized object in contact with a measuring apparatus, it is practically instantaneous.

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