Breaking time-inversion invariance through decoherence — Energetic consequences for attosecond neutron scattering

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Abstract.

Nuclei and electrons in condensed matter and/or molecules are usually entangled, due to the prevailing (mainly electromagnetic) interactions. However, the "environment" of a microscopic scattering system (e.g. a proton) causes ultrafast decoherence, thus making atomic and/or nuclear entanglement effects not directly accessible to experiments. However, our neutron Compton scattering experiments from protons (H-atoms) in condensed systems and molecules have a characteristic collisional time about 100—1000 attoseconds. The quantum dynamics of an atom in this ultrashort, but finite, time window is governed by non-unitary time evolution due to the aforementioned decoherence. Unexpectedly, recent theoretical investigations have shown that decoherence can also have the following energetic consequences. Disentangling two subsystems A and B of a quantum system AB is tantamount to erasure of quantum phase relations between A and B. This erasure is widely believed to be an innocuous process, which e.g. does not affect the energies of A and B. However, two independent groups proved recently that disentangling two systems, within a sufficiently short time interval, causes increase of their energies. This is also derivable by the simplest Lindblad-type master equation of one particle being subject to pure decoherence.

Our neutron-proton scattering experiments with H2 molecules provide for the first time experimental evidence of this effect. Our results reveal that the neutron-proton collision, leading to the cleavage of the H-H bond in the attosecond timescale, is accompanied by larger energy transfer (by about 2—3%) than conventional theory predicts. Preliminary results from current investigations show qualitatively the same effect in the neutron-deuteron Compton scattering from D2 molecules. We interpret the experimental findings by treating the neutron-proton (or neutron-deuteron) collisional system as an entangled open quantum system being subject to fast decoherence caused by its "environment" (i.e., two electrons plus second nucleus of H2 or D2). The presented results seem to be of generic nature, and may have considerable consequences for various processes in condensed matter and molecules, e.g. in elementary chemical reactions.

1. Introduction

Quantum entanglement (QE) has emerged as the most emblematic feature of quantum mechanics. It provides novel insights into the fundamental structure of physical reality [1, 2, 3, 4], and has raised widespread interest in various branches of science, and recently also in quantum information technology [5].
The environment surrounding a quantum system interacts with it and can, in effect, monitor (or ”measure”) some of the system’s observables. As a result, the phenomenon of environment-induced decoherence [6, 7, 8, 9] emerges, and QE is destroyed. Other, supposedly more general models of decoherence, e.g. those appearing in fundamental modifications of the Schrödinger time evolution, play no role in the present paper. A deeper understanding of decoherence is also expected to lead to new insights into quantum fundamentals, particularly quantum measurement and the quantum-classical transition.

Owing to the prevailing electromagnetic interactions, nuclei and electrons in condensed matter and/or molecules are also in principle entangled. Under ambient conditions, the ”environment” of microscopic systems (e.g. a neutron colliding with a proton) causes ultrafast decoherence, thus making atomic and/or nuclear entanglement effects not directly accessible to experiments. In this context, it may also be noted that decoherence is recognized as one of the main obstacles to realizing quantum computing and quantum information processing [5].

Decoherence (and, in particular, disentanglement of quantum-entangled systems) is an omnipresent phenomenon of dynamics of quantum open systems. It is usually an extremely fast dynamical process; for various examples, see e.g. [10] and the textbooks cited above. However, neutron Compton scattering (NCS, also called ”deep inelastic neutron scattering”, DINS) [11, 12] is an ultrafast scattering technique that can probe quantum dynamics of nuclei in the attosecond time window, and so it may open up a way for investigating short-lived QE even in condensed matter at ambient conditions. NCS was first suggested in the pioneering work of Hohenberg and Platzman [11] as a means of measuring nuclear momentum distributions in condensed matter. Conceptually based on the well-known Compton effect [13] the NCS method is analogous to the measurement of electronic momentum distributions in atoms and solids by Compton scattering of high-energy photons from electrons, or measurement of nucleon momenta by quasi-elastic scattering of high-energy electrons from nuclei.

For neutron—proton scattering, and for the experimental setup reported below, the effectuated momentum and energy transfers are large,

\[ \Delta E \sim 1 - 30 \text{ eV}, \quad |q| \sim 20 - 120 \text{ Å}^{-1}, \]

and the ”collisional time window” or ”scattering time” \( \tau_{sc} \) is very short,

\[ \tau_{sc} < 10^{-15} \text{ s} \]

[12, 14]. This is of the order of the timescale of ”electronic motion”, e.g. the duration of electronic rearrangements accompanying the formation or the breaking of chemical bonds. Thus one may expect that NCS can reveal unknown features of quantum dynamics of atoms participating in chemical bonds [15]. Such features are revealed by the ”anomalous” decrease of NCS-intensity from protons (in short, H-intensity deficit) of several materials; cf. [16, 17, 18, 19]. Furthermore, this attosecond effect was confirmed by Cooper et al. applying an independent experimental method, i.e. electron—atom Compton scattering [17, 20]; see also the detailed theoretical analysis by Bonham et al. in the frame of the first Born and Born-Oppenheimer approximations [21].

However, this paper does not deal with the usual objectives of NCS, i.e. widths of Compton profiles, or anomalous scattering intensities. Instead, here we consider only the simplest property of the NCS collisional process which is directly accessible to the experiment: i.e. the mean energy transfer from the neutron to the struck nucleus, called ”recoil energy” \( E_{rec} \). This quantity is completely determined by the position of the recoil peak-maximum (or center-of gravity) in the measured time-of-flight (TOF) interval.

Our experimental results presented below demonstrate experimentally that decoherence in neutron—proton Compton scattering from isolated \( \text{H}_2 \) molecules can also have unexpected
energetic consequences. The theoretical basis of new phenomenon under consideration may be described as follows. Disentangling two subsystems $A$ and $B$ of a quantum system $AB$ is tantamount to erasure of quantum phase relations (i.e. decoherence) between $A$ and $B$. It is widely believed that this erasure is an innocuous process, which e.g. does not affect the energies of $A$ and $B$. Surprisingly, recent theoretical investigations by two independent groups showed that disentangling two systems, within a sufficiently short time interval, can cause increase of their energies. Moreover, this intrinsic energy increase is also shown to be derivable by the simplest Lindblad-type master equation, which describes one particle being subject to pure decoherence (in the Markovian limit).

Our neutron—proton scattering experiments with $H_2$ molecules provide for the first time experimental evidence of this effect [22]. The presented results reveal that the neutron-proton collision leading to the cleavage of the $H—H$ bond on an attosecond timescale is accompanied by a larger energy transfer (by about 2—3%) than conventional theory predicts. Moreover, preliminary results from current investigations reveal qualitatively the same effect in the neutron—deuteron Compton scattering from $D_2$ molecules. We interpret the experimental findings by treating the neutron—proton (or neutron—deuteron) collisional system as an entangled open quantum system being subject to fast decoherence caused by its “environment” (i.e., two electrons plus second nucleus of $H_2$ or $D_2$).

The outline of the paper is as follows. After some short introductory remarks on quantum entanglement and nonlocality in Sec. 2, we discuss in Sec. 3 the recent theoretical models exhibiting the energetic consequences of decoherence. In addition, we show here the presence of the same effect in the well-known formalism of Lindblad-type master equations describing pure decoherence. Sec. 4 contains the experimental method and the main experimental results. The proposed theoretical interpretation of the observations in Sec. 5, which is only qualitative, appears to confirm the new decoherence effect. In Sec. 6, among additional remarks and conclusions, we present current (preliminary) results of scattering from molecular $D_2$ which give further confirmation of the phenomenon under consideration. For readers interested in experimental details, the Appendices provide a concise (but rather self-contained and fully referenced) description of the experimental method and its specific features, of data analysis procedure, and of associated technical and/or instrumental details.

2. Remarks on quantum entanglement

The notion of entanglement (in short QE) was introduced by Schrödinger. It describes the physical consequences of the mathematical fact that the tensor product $H_A \otimes H_B$ of two vector spaces $H_A$ and $H_B$ is larger than their Cartesian product $H_A \times H_B$ [23]. Namely, not every element of the tensor product can be factorized as $\psi_A \otimes \psi_B$, with $\psi_i \in H_i$ ($i = A, B$).

The crucial physical consequence of this fact becomes obvious by taking $H_A$ and $H_B$ to be the state spaces of two quantum objects $A$ and $B$. A pure state $|\Psi\rangle \in H_A \otimes H_B$ of the combined quantum system is called separable, or not entangled, if it can be factorized as $|\Psi\rangle = |\psi_A\rangle|\psi_B\rangle$. But due to the superposition principle of quantum mechanics there are also states of the form

$$|\Psi\rangle = c_1|\psi_A\rangle|\psi_B\rangle + c_2|\phi_A\rangle|\phi_B\rangle + \ldots,$$

($\psi_i, \phi_i \ldots \in H_i$, $i = A, B$). which are not separable in general. In that case, the individual quantum objects cannot be assigned independent pure states, and one says that the two objects are entangled with each other, or simply entangled.

A well known example of an entangled quantum state is the so-called singlet state

$$|\Psi^-\rangle = \frac{1}{\sqrt{2}} (|0\rangle_A|1\rangle_B - |1\rangle_A|0\rangle_B)$$

(2)
of two (distinguishable) qubits (or two spin-1/2 particles, etc.). In a singlet each individual particle $A$ and $B$ has no definite value 0 or 1 (or: spin direction). For example, the state of the particle $A$ is not a pure state but the mixed state

$$\rho_A = \text{Tr}_B |\Psi^-(\Psi^-)| = \frac{1}{2} (|0\rangle_A \langle 0|_A + |1\rangle_A \langle 1|_A).$$

(3)

QE has various counter-intuitive consequences, one of them being nonlocality. Einstein, Podolsky and Rosen (EPR) argued that spatially separated objects must have separable physical descriptions and thus concluded that the quantum mechanical description of objects in an entangle state must be incomplete, i.e. so-called "hidden variables" must exist [1]. This constitutes the main point of the famous EPR argument.

Three decades later, Bell, in looking more thoroughly at the measurements that can be made on two entangled but spatially well-separated, objects, showed that the probabilistic predictions of quantum mechanics can be incompatible with the existence of such hidden variables [2]. In particular, Bell showed this for specific entangled states. Twenty-five years later, Gisin succeeded in proving that this is true for all entangled states [3]. Thus [23]

$$\text{Entanglement} \Rightarrow \text{Nonlocality}$$

(4)

Today one can say that entanglement and nonlocality are fundamental properties of physical reality.

The context in which the notion of entanglement was first introduced and discussed was purely theoretical and/or philosophical (see e.g. [4]). Only in the last 15—20 years was it recognized that entanglement and decoherence should have practical applications: quantum computing, quantum coding, quantum cryptography, etc. [5].

2.1. Dynamical creation of entanglement.

Quantum entanglement (QE) in molecules and/or condensed matter is the consequence of many-body interactions. Let $\Psi_A$ and $\Psi_B$ be the state vectors of two systems $A$ and $B$. The two systems, with "free" Hamiltonians $H_A$ and $H_B$, may interact, the corresponding Hamiltonian being $V_{AB}$. Let the state of the composite system 'A and B', in short $AB$, at time $t = 0$ be separable, $\Psi_{AB}(0) = \Psi_A \cdot \Psi_B$. With the exception of pathological cases, $V_{AB}$ does not commute with $H_A$ and $H_B$. Then it holds for $t > 0$ that the complete evolution operator of the composite system $AB$,

$$U_{AB}(t) = e^{-\frac{i}{\hbar}(H_A + H_B + V_{AB})t}$$

(5)

doest not factorize into a product of two "free" evolution operators, $U_A$ and $U_B$. Consequently, the wave function $\Psi_{AB}(t)$ at time $t > 0$, $\Psi_{AB}(t) = U_{AB}(t)\Psi_{AB}(0)$, does not factorize into a product of two wave functions (say, $\Psi'_A$ and $\Psi'_B$), each of them representing the state of one of the two systems $A$ and $B$; i.e.

$$\Psi_{AB}(t) = U_{AB}(t)\Psi_{AB}(0) \neq \Psi'_A(t) \cdot \Psi'_B(t).$$

(6)

That is, $\Psi_{AB}(t)$ is an entangled state being caused by $V_{AB}$ which couples the specific degrees of freedom appearing into it. For example, Coulombic interactions, which depend on the spatial degrees of freedom of the particles (i.e., nuclei and electrons) lead to spatial entanglement; magnetic interactions between spins lead to spin entanglement being of central importance for the emerging quantum technology of "spintronics" [5].

For closed quantum systems the time evolution is $t$-inversion invariant, and the process of Eq. (6) represents a unitary transformation, because the operator $U_{AB}$ is unitary, i.e. $U_{AB}(t)U_{AB}^\dagger(t) = 1$. 

4
3. Energetic consequences of decoherence

Decoherence is the typical quantum dynamical phenomenon which destroys quantum state superpositions (quantum interference, quantum phases), thus leading to the "appearance of a classical world in quantum theory"; cf. the textbooks and review articles [6, 7, 8, 9].

Decoherence, similarly to many other irreversible mechanisms, is a time-oriented process, i.e. it breaks the well-known time-inversion invariance of the Schrödinger equation. A considerable number of models of decoherence, based on a variety of physical motivations, have been proposed and investigated in the scientific literature; cf. [6, 7, 8, 9]. A particularly illuminating comparison of some models and their motivations has been provided by Diósi, who also demonstrated that a possible intrinsic time uncertainty of quantum mechanics can cause decoherence [24].

Quite unexpectedly, it turns out that "pure" decoherence, which is known to be much faster than friction [6, 7, 8], may have a specific peculiar consequence. Namely, as shown below in the frame of three well-known theoretical models, pure decoherence turns out to be intrinsically connected with an unavoidable increase of mean energy of the system — in the absence of any direct interaction with other systems. This is certainly unexpected, since erasure of quantum phases is usually not acknowledged as a source of energy.

3.1. Lindblad equation and energy increase

To describe the dynamics of open quantum systems and decoherence, various generalizations of the Schrödinger equation have been proposed; see the textbooks [6, 7, 8]. Among these theoretical approaches, the Born-Markov master equation plays an enormously important role. Master equations of the so-called Lindblad form refer to a particular (albeit quite general) class of Markovian master equations, which ensure positivity of the reduced density operator $\rho(t)$ describing the system, i.e. $\langle\rho(t)\rangle \geq 0$, for any pure state $|\psi\rangle$ of the system and for all $t$. The most general mathematical form of such equations was derived by Gorini, Kossakowski and Sudarshan [25] and Lindblad [26].

Here we consider the simplest Lindblad-type ansatz for the master equation for the statistical operator $\rho$ of an open quantum system, which includes only one Lindblad operator $R$; in a real system we would have a multitude of such dynamical variables. We set

$$\frac{\partial \rho}{\partial t} = -\frac{i}{\hbar} [H, \rho] - \Gamma [R, [R, \rho]]$$

(7)

where $\Gamma > 0$ is a positive constant and $H$ is the Hamiltonian. The first term on the right-hand side (rhs) describes the usual unitary time evolution of the state. The double commutator term describes decoherence (and/or dephasing). It may be noted that this equation describes "pure" decoherence, i.e. it does not contain any term describing friction, the latter being included in Caldeira-Leggett-type equations [27].

Considering Eq. (7) in the $R$-representation, the double commutator term takes the form

$$-\Gamma (r - r')^2 \langle r | \rho | r' \rangle$$

which, for positive $\Gamma$, leads to an exponential decay of the nondiagonal elements of the density matrix:

$$\langle r | \rho(t) | r' \rangle = \langle r | \rho(0) | r' \rangle \exp(-\Gamma (r - r')^2 t) \quad \text{for} \quad 0 \leq t$$

(8)

where $t = 0$ is the initial time [6, 7, 8]. Obviously, this result does not hold for $t \leq 0$. In other words, coherent superpositions of different states $|r\rangle$ and $|r'\rangle$ will be suppressed over time. The most well-known specialization of Eq. (7) is the case of an one-particle system that interacts with an external (thermal) environment, in which case $R$ is taken to be the position operator, $q$. This corresponds to the Joos-Zeh master equation [28].
It is well known that Eq. (7) preserves the normalization $Tr\rho = 1$, which is very satisfactory. However, as Ballentine [29] pointed out, the expectation value of the energy, $\langle H \rangle = Tr(H\rho)$ is in general not conserved, as the following short calculation proves:

$$\frac{d\langle H \rangle}{dt} = \frac{d}{dt} Tr(H\rho) = Tr \left( H \frac{\partial \rho}{\partial t} \right) = -\Gamma Tr([H, R][R, \rho]) \quad (9)$$

Here the operator identity $Tr(A[B, C]) = Tr([A, B]C)$ was used. Obviously, if the Hamiltonian and the Lindblad operator $R$ do not commute, $\langle H \rangle$ is not conserved for every state $\rho$.

Of particular importance for illustration, as well as for the following discussions, is the special case of a "free" particle in one dimension with momentum operator $p$ and Hamiltonian $H = p^2/2m$. The decoherence in position owing to interactions with the "environment" is described by taking $R$ to be the particle’s position operator $q$ [28]. Then, since $[p, q] = h/i$ and $[p^2, q] = p^2q - qp^2 = (p^2q - pqp) + (qpq - qp^2) = p[p, q] + [p, q]p = \frac{2h}{i}p$

one obtains from the last equation

$$\frac{d\langle H \rangle}{dt} = -\Gamma Tr([p^2/2m, q][q, \rho]) = -\frac{\Gamma}{2m} \frac{2h}{i} Tr(p[q, \rho]) = -\frac{\Gamma}{2m} \frac{2h}{i} Tr([p, q]\rho) \quad (10)$$

$$= + \frac{\Gamma h^2}{m} > 0 \quad (11)$$

This surprising result holds also in three dimensions, and for inclusion of scalar and/or vector potentials. It also remains valid for more general cases, e.g. even in cases with the decoherence factor $\Gamma$ becoming a function of the "distance" $|r - r'|$; see [29, 30].

This result is quite disturbing, as it contradicts conventional expectations about what the consequences of decoherence should be (besides the destruction of coherent superpositions of quantum states). Namely, theorem (11), which follows from the Lindblad equation (7) for a "free" particle, seems to represent a serious weakness of the theory, because it implies that the system’s mean energy should always increase; or, in other terms, the environment would act as an inexhaustible energy source. As discussed by Ballentine, this continuous gain of energy is clearly incompatible with the system’s attainment of equilibrium [29]. For a more thorough discussion of these difficulties, see Refs. [29, 30].

3.2. The theoretical model by Schulman and Gaveau

Surprisingly, a recent independent and more general theoretical analysis by Schulman and Gaveau [31, 32] has derived essentially the same result. We begin with a short description of the general model.

A quantum system $A$, e.g. a quantum oscillator, with free Hamiltonian $H_A$ makes an elastic collision with a second system $B$ with free Hamiltonian $H_B$. Let the interaction Hamiltonian (potential) be $V_{AB}$. The total Hamiltonian is then

$$H = H_A + H_B + V_{AB} \quad (12)$$

Initially, i.e. before the collision, the two particles are not entangled and so one takes the complete density matrix $\rho(0)$ to be a product, $\rho(0) = \rho_A(0) \otimes \rho_B(0)$. Subsequent to their collision they become entangled and the exact density operator

$$\rho(t) = U(t) \rho(0) U^\dagger(t)$$
(\(U(t)\): time evolution operator) is not representable by the product of the individual density operators \(\rho_A(t) = \text{Tr}_B \rho(t)\) and \(\rho_B(t) = \text{Tr}_A \rho(t)\). \((\text{Tr}_i\) denotes the partial trace over the variables of system \(i\).

However, it is widely believed that once the particles are separated the quantum correlations associated with the entanglement can be dropped (provided one does not perform an experiment of Einstein-Podolsky-Rosen type [1]), simply because measurements of physical quantities of each of the two particles cannot depend on their QE. Thus the replacement

\[
\rho(t) \rightarrow \rho_A(t) \otimes \rho_B(t)
\]

i.e. the erasure of entanglement, is assumed to be innocuous, as it should e.g. not affect the energies of the systems.

The striking result of Schulman and Gaveau is that this does not hold in general [31]. Putting

\[
\Delta \rho(t) = \rho_A(t) \otimes \rho_B(t) - \rho(t)
\]

and for a particular form of the interaction Hamiltonian, they show that for sufficiently short times the following relation holds:

\[
\Delta E \equiv \text{Tr}(\Delta \rho(t)H) = \text{Tr}(\Delta \rho(t)V_{AB}) > 0.
\]

In other words, the replacement of the entangled \(\rho(t)\) by the non-entangled state \(\rho_A(t) \otimes \rho_B(t)\), i.e. the decoherence of the \(AB\) system, necessarily increases the system’s energy, for sufficiently short times [31, 32]. This appears highly paradoxical since, as Schulman and Gaveau put it: ”...losing quantum correlations should not heat the gas. You do not burn your finger because of a partial trace over a density matrix” [31].

A detailed derivation of this inequality is presented in [32]. Moreover, this result was shown to be valid for a large class of potentials, e.g. for two-body interactions, although it does not hold universally [32]. This is an interesting point, and therefore the following example should be mentioned. The spin-boson model Hamiltonian

\[
H = H_A + H_B + V_{SB} = \omega_a a^{\dagger}a + \omega_b b^{\dagger}b + g(a^{\dagger} + a)(b^{\dagger} + b)
\]

was shown to exhibit the considered effect, in clear contrast to the related Jaynes-Cummings model

\[
H = H_A + H_B + V_{JC} = \omega_a a^{\dagger}a + \omega_b b^{\dagger}b + g(a^{\dagger}b + b^{\dagger}a),
\]

in which the effect is absent [31].

One may object that the above result, Eq. (14), is unphysical since it violates energy conservation. This, however, is not the case, as the detailed discussions of Ref. [31] showed. It was stressed that, in the situation contemplated, the coupling Hamiltonian must be considered as time dependent, because the physical approach and separation of the particles leads to a time-dependent coupling coefficient. Thus, energy conservation need not apply. Moreover, it was pointed out that the translational degrees of freedom of \(A\) and \(B\) supply this "additional" energy \(\Delta E > 0\) [31].

To make qualitative contact with our NCS experiment under consideration, the impinging neutron \(n\) may be associated with system \(A\), and the struck proton \(p\) together with its effective Born-Oppenheimer (BO) potential with system \(B\). The neutron-proton interaction is associated with \(V_{AB}\); see also below. In this context it should be noted that the phenomena of entanglement and decoherence play no role in conventional neutron scattering theory [12, 39, 40, 41]. However, the existence of quantum entanglement follows from first principles within quantum theory, and is essentially a consequence of the non-commutativity of the interaction Hamiltonian with the free Hamiltonian.
3.2.1. Digression: On van Kampen’s "repeated randomness assumption". In [32] it was mentioned that some physical topics like the Boltzmann H-theorem and Kampen’s criticism of Green-Kubo formulas may be related to the aforementioned erasure of quantum correlations and its effects. Hence it may be interesting to consider in some detail the possible conceptual interrelation and/or similarity between

(a) the Schulman-Gaveau approach of applying the decoherence assumption (13) before each new collision of system $A$ with $B$, and

(b) van Kampen’s "repeated randomness assumption", which was shown to underly the fundamental basis of statistical mechanics of irreversible processes [33, 34, 35, 36].

The mentioned "randomness assumption" refers to erasure (or coarse graining, averaging out) of quantum phases being represented with the non-diagonal elements of the exact density operator $\rho(t)$. This corresponds to decoherence or dephasing, in present-day nomenclature. Scrutinizing linear response theory and the validity of the well-known Green-Kubo formulas, van Kampen concluded the following: the assumption of the Kubo theory [37, 38] that linearity of the macroscopic response to an external force $F$ means that the equation of motion for $\rho(t)$ should also be solved to first order in $F$, during a short time interval $\Delta t$, constitutes a "fatal flaw" (see [36], p. 280). Moreover, this microscopic linearity was shown to be equivalent to a tacitly assumed randomness (i.e. decoherence) not only at the initial time $t = 0$, as the Kubo theory claims, but also continuously during the whole $\Delta t$ [36].

For illustration, let us consider the case of electric conductivity in a metal. Van Kampen’s result implies here that the well-known appearance of resistivity, and associated energy transfer (dissipation) from the electrons to the lattice, can only be proven within Kubo’s theory by assuming that decoherence takes place continuously during the whole $\Delta t$, thus keeping the density operator at equilibrium (and hence diagonal in the energy representation), $\rho(t) = \rho_{\text{eq}}$, during $\Delta t$. Or, in simpler terms: if one assumes $\rho(0) = \rho_{\text{eq}}$ only, as standardly done in theoretical treatments, and if linearization of $\rho(t)$ owing to a small disturbing electric field $F(t)$ is carried out correctly and without applying further assumptions, then one obtains zero resistance and zero energy dissipation!

The analogy to the Schulman-Gaveau results of calculations presented in [31] is given by the repeated disentanglement $\rho(t) \rightarrow \rho_A(t) \otimes \rho_B(t)$ after each collision and before the next one takes place. Without erasing quantum correlations there is no heating up of the systems $A$ and $B$.

3.3. The model by Erez et al. and Gordon et al.

It may be noted that the model of Ref. [31] assumes the appearance of decoherence, as the replacement $\rho(t) \rightarrow \rho_A(t) \otimes \rho_B(t)$ shows, but does not describe the disentanglement process explicitly. Thus, the environment and the couplings of the two systems to it do not appear explicitly in the model.

Very recently, theoretical investigations that address these issues have been presented by Erez et al. [42] and Gordon et al. [43], which explore explicitly the aforementioned decoherence process in a different physical context. The focus here is the cooling of two-level model systems (quantum bits) on ultrashort time scales. It is shown that these anomalous non-Markov cooling processes stem from the hitherto unfamiliar coherent quantum dynamics of the system-bath interaction well within the bath memory time. This is an interesting point, since the significance of the bath’s dynamics becomes apparent.

In the course of their analysis the authors derive a result analogous to Eq. (14), first for brief quantum non-demolition (QND) measurements [42] and later for the general case of any abrupt disturbances, e.g. phase changes [43].

In Ref. [42] the "measurement" (or interaction) of a quantum system $S$ by a "detector" $D$ (i.e. a second quantum system) is considered, represented by the interaction $H_{SD}(t)$. The authors
study the total Hamiltonian

\[ H_{\text{tot}} = H_S + H_B + H_{SB} \]  

(15)

of the system (with Hamiltonian \( H_S \)) that interacts with the bath (with Hamiltonian \( H_B \)) and is intermittently perturbed by the coupling of the system to the detector:

\[ H(t) = H_{\text{tot}} + H_{SD}(t) \].  

(16)

It is essential that the interaction Hamiltonians \( H_{SB} \) and \( H_{SD} \) do not invoke the rotating-wave approximation; i.e. energy conservation between the system and the bath or the detector is not imposed, on the ultrashort time scales considered [42].

It was proved [42] that a nearly impulsive (projective) disturbance of the system always produces an energy increase (called "heating") of the equilibrium state immediately thereafter. This general conclusion was supported by numerical results, for a disturbance of finite, albeit brief, duration. For an ultrashort time interval \( 0 \leq t \leq \tau \), the authors show the important results

\[ \langle H_{SB}(0) \rangle \leq 0 \rightarrow \langle H_{SB}(\tau) \rangle = 0 \]  

(17)

where \( \langle H_D \rangle \) remains unchanged. This transfer of energy, associated with a change in the entanglement between the system and the bath, triggers the quantum dynamics that redistributes their mean energy (and entropy).

At time \( t = \tau + \Delta t \) after the measurement (because \( H_{SD}(t \geq \tau) = 0 \)), and during sufficiently short time intervals \( \Delta t \), the rotating wave approximation breaks down; only \( \langle H_{\text{tot}} \rangle \) is conserved, until the next measurement. But now \( \langle H_{SB} \rangle \) decreases because the correction to the ground-state energy of \( H_{\text{tot}} \) should be negative for a weakly perturbing interaction \( H_{SB} \). As a result, one obtains the relations

\[ \frac{d}{dt} \langle (H_S + H_B) \rangle_{\tau+\Delta t} > 0 \]  

\[ \frac{d}{dt} \langle H_{SB} \rangle_{\tau+\Delta t} < 0 \]  

(18)

which implies a post-disturbance "heating" of the system and the bath combined [42]. These results were recently extended to the general case of any abrupt disturbance [43].

To make a qualitative contact with the basic features of our NCS experiment under consideration, one may associate the proton \( p \) with the quantum system \( S \), the proton’s environment (denoted by \( \mathcal{E} \) below) with the bath \( B \), and the neutron with \( D \). The aforementioned "additional" energy \( \Delta E \) immediately after the disturbance —in Ref. [43] denoted by \( \delta \langle H_{\text{tot}} \rangle^M \) for projective measurements and by \( \delta \langle H_{\text{tot}} \rangle^\phi \) for an abrupt phase shift— was proved to be positive, \( \Delta E > 0 \); i.e., a "heating" of the equilibrium state appears immediately after the external abrupt disturbance [43]. Recall that the NCS process is not a QND process, and thus one may expect that \( \Delta E \) can contain a contribution from the translation energy of the impinging neutron.

However, the "environment" of the struck proton hardly resembles a thermal bath and the coupling \( H_{SB} \) is very strong. Therefore the above theoretical results cannot be directly related with our experimental findings on \( \text{H}_2 \) presented below. However, a relation to NCS experimental results from liquid or solid \( ^4\text{He} \), in the normal or superfluid state, could be appropriate, because the struck \( \text{He} \) (i.e. the system) interacts weakly with the atoms of its environment.
4. Experimental results
We now proceed to our new experimental results [22] and their physical interpretation in the theoretical context of the models of the preceding section. It will be shown that, although one usually thinks of the destruction of quantum correlations as purely an information issue, here it appears to have measurable energetic consequences.

We reiterate that issues regarding the "anomalous" scattering intensity effect [16, 17, 18, 19], or the widths of Compton profiles [12], are not addressed in the present paper.

4.1. NCS measurements
In this section we present very recent experimental NCS results from gaseous molecular H$_2$ and D$_2$ and associated current discussions [22, 44, 45], which provide for the first time evidence for the novel decoherence effect predicted by the theoretical models of the preceding section.

To facilitate the presentation, we first outline some essential elements of data recording and analysis; full details are given in the Appendices and in [22].

![Figure 1. Examples of measured TOF spectra. The data are from liquid benzene (C$_6$H$_6$) in an Nb cell, at the specified scattering angles [48]. Solid lines are fits to the data. The H recoil peak is well separated from those of C and Nb. As expected, there is no H recoil peak in the backward scattering spectrum. In backward scattering regime, the C- and Nb-recoil peaks are resolved owing to the larger energy transfers involved.](image)

From the measured time-of-flight (TOF) spectra (see Figure 1 for two examples), momentum (ℏq) and energy (ℏω) transfers from the neutron to the stuck particle are determined by standard methods; see Appendix B. The main object of interest is here the position of the recoil peak of a struck nucleus (proton or deuteron) in TOF space. According to conventional theory, this immediately translates to the energy transfer from the neutron to the nucleus in an elementary collisional two-body process, and therefore it is considered to be well understood.

Within conventional NCS theory which assumes validity of the impulse approximation (IA) [12, 46, 47], the elastic collision of a neutron with an approximately free atom with mass $M$ and initial momentum $p$ results in the neutron’s lost energy $ℏω$ being transferred to the struck atom.
\[
h_\omega = \left( \frac{h \mathbf{q} + \mathbf{p}}{2M} \right)^2 - \frac{p^2}{2M} = \frac{(h \mathbf{q})^2}{2M} + h \mathbf{q} \cdot \mathbf{p} \quad \frac{M}{2M}
\]

This equation represents energy conservation. The recoil energy \( h_\omega_{\text{rec}} = \frac{h^2 q^2}{2M} \) represents the energy of recoil of a stationary nucleus owing to the collision. Thus scattering from a gaseous sample of such atoms leads to a recoil peak centered at \( h_\omega_{\text{rec}} \), exhibiting a peak width caused by the term \( h \mathbf{q} \cdot \mathbf{p} / M \) which represents Doppler broadening. The above energy balance holds exactly in the IA, which however is not completely fulfilled at moderate momentum and energy transfers. In such cases, so-called final state effects (FSE) become apparent. They are caused by environmental forces on the struck particle. It was shown that FSE cause a shift of the maximum of the recoil peak to lower energy than the recoil energy [12, 46, 22].

In the context under consideration, this point is crucial. In the framework of conventional NCS theory, a shift to higher energy transfers than \( h_\omega_{\text{rec}} \) would imply that there is a higher energy loss in the scattering process than the IA predicts. Physically this means that the struck atom would be “more free” than even an isolated atom floating in space, which of course is meaningless. Hence, deviations from the IA must give peak shifts to less than \( h_\omega_{\text{rec}} \), since they are always caused by the atom not being free, owing to its interaction with other atoms. Thus there is an additional resistance to motion of the struck atom, which manifests as a slightly lower energy transfer than the IA predicts. Summarizing, a peak-maximum shift to higher energies than \( h_\omega_{\text{rec}} \) is impossible within conventional NCS theory.

**Figure 2.** Compton profiles of ortho-D\(_2\) at 20 K, measured at ISIS (parallel shifted for visibility). Adapted from Fig. 26 of [50]. Points are experimental data (after data processing). The lines represent various theoretical models discussed in that reference.

The measured TOF spectra may be straightforwardly transformed to obtain the associated
experimental Compton profiles [12, 46, 47] of the recoiling atoms, commonly denoted $J(y)$; see Appendix C. This function represents the distribution of the one-dimensional projection $h y$ of atomic momentum $\mathbf{p}$ (before collision) along the $\mathbf{q}$ direction represented by the unit vector $\mathbf{e}_q$. According to the IA, [12, 46, 47]

$$J(y) = \int n(p) \delta(p \cdot e_q - h y) \, dp.$$  

(20)

Here, $n(p)$ is the momentum distribution of the struck particle with mass $M$ before collision, which is the quantity of interest for theoretical investigations. For isotropic systems, the specific direction $\mathbf{e}_q$ becomes immaterial.

In the IA, $J(y)$ must be centered at $y = 0$ [12, 44, 46, 47]; for further details, see Appendix C. The aforementioned conclusion about peak shifts implies that a shift of the peak maximum (or center of gravity) of $J(y)$ to $y_{\text{max}} > 0$ values is impossible within conventional theory, since deviations from the IA must displace $J(y)$ to $y_{\text{max}} < 0$. This theoretical expectation is nicely confirmed with the experimental NCS results from earlier liquid ortho-D$_2$ investigations reproduced in Fig. 2. For the low scattering angle shown, the Compton profile $J_D(y)$ is clearly shifted to $y_{\text{max}} < 0$, whereas the peak is centered at $y_{\text{max}} = 0$ for the highest scattering angle, for which the IA appears to be valid, as expected by conventional theory.

4.2. Main results in forward scattering

Figs. 3-4 show the Compton profiles $J(y)$ of D and H obtained from measurements of gaseous H$_2$ and D$_2$ on the Vesuvio (formerly eVS) NCS spectrometer at the ISIS neutron spallation source. The data are from detectors positioned at scattering angles $\theta < 90^\circ$, i.e. in the "forward" scattering regime.

These experimental data were obtained by transforming to $y$ space the TOF spectra measured as described above and in Appendix B. For our data analysis we used calibration parameters measured directly with steel rules and a protractor [58] by the ISIS Metrology Group; cf. Appendix C.

We have analyzed the sensitivity [49] of the peak positions of $J(y)$ in $y$-space to calibration uncertainties and found that the results in $y$-space are reliable to better than 0.3 Å$^{-1}$, for each single detector. Of course, this value improves for an average $J(y)$ obtained from several detectors. This estimate is supported by the small scatter in the positions of the individual-detector $J(y)$ curves shown in all four Figs. 3(a,b) and 4(a,b). The most important findings are:

(A) The peak maxima (or the centers-of-gravity) of the D Compton profiles are found to be at $y_{\text{max}} \leq 0$ as conventionally expected, for all momentum transfers investigated (Fig. 3a,b). The IA appears to be well obeyed for high momentum transfers. In these experiments, the D—D bond is not broken (see below). The Compton profiles (a) and (b) are in full agreement with the well-established results of Andreani et al. [51]. Therefore they also represent a confirmation of both the correct working of the present configuration of the eVS/Vesuvio spectrometer and our data analysis, and a baseline for comparison with our results for H, measured and analyzed in the same way. (Data measured with backward scattering detectors, during the same time, are reported in subsection 6.1. below.)

(B) The H Compton profiles obtained at low scattering angles are centered at $y_{\text{max}} \leq 0$ as conventionally expected and experimentally observed earlier [51, 52]; see Fig. 4a. In contrast, the H Compton profiles measured at $\theta_{\text{high}}$ are centered at momentum values $y_{\text{max}} > 0$, thus contradicting conventional theory; see Fig. 4b. Equivalently, the actual energy transferred in the collision is larger (by about 2-3%) than the IA predicts.

The magnitude of this effect may be estimated as follows: From Eq. (20), the excess energy
**Figure 3.** Experimental (not normalized, with no corrections included) Compton profiles $J_D(y)$ of D in $D_2$ gas at 44 K and 10 bar in an Al cell. This figure is identical with Fig. 1 of Ref. [22].

- **(a)** $J_D(y)$ measured with one block of 8 detectors, at the lowest ($\theta_{\text{low}} \sim 33^\circ - 36^\circ$) scattering angles available. The signal contribution on the left is due to scattering from the Al cell. The results clearly exhibit conventional FSE for low momentum transfers.
- **(b)** $J_D(y)$ measured with one block of 8 detectors, at the highest ($\theta_{\text{high}} \sim 61^\circ - 66^\circ$) scattering angles available presently. These Compton profiles are in agreement with earlier well-established results [51, 52], like e.g. those in Fig. 2.

The transfer is

$$\delta E = \hbar^2 q y_{max}/M$$  \hspace{1cm} (21)

and thus the ratio $R$ of $\delta E$ to the recoil energy is $R = \delta E/\hbar \omega_{\text{rec}} = 2y_{max}/q$. For H, $m \approx M$ and $q \approx k_1 \tan \theta$, where $k_1$ is the wavevector of the scattered neutron. From the observed shift of $J_H(y)$ at high $q$ we estimate $y_{max} \approx 1.5 \text{Å}^{-1}$, which amounts to an excess energy $\delta E \approx 590$
Figure 4. Experimental (not normalized, with no corrections included) Compton profiles \( J_H(y) \) of H in \( \text{H}_2 \) gas at 41 K and 10 bar. This figure is identical with Fig. 2 of Ref. [22]. The instrumental setup was the same as for the experiment with \( \text{D}_2 \); see the text and Fig. 1. The results (a) at \( \theta_{\text{low}} \) are in agreement with the corresponding results of Andreani et al. [51], in which conventional FSE are visible in the experimental Compton profiles \( J_H(y) \). The H–H bonds are not broken after scattering. In contrast, the results (b) at \( \theta_{\text{high}} \) (with \( q \sim 95 \text{Å}^{-1} \) and \( \hbar\omega \sim 19 \text{eV} \)) are centered at momentum values \( y_{\text{max}} > 0 \). Scattering brakes the H-H bonds. We note that results from detectors at \( \theta > 50^\circ \) were not reported in Refs. [51, 52].

meV. Therefore one obtains \( R \approx (2 \cdot 1.5)/(48.6 \cdot \tan 63^\circ) = 0.03 \).

This is interpreted to represent the quantity \( \Delta E \) of Eq. (14), i.e. \( \Delta E = \delta E \).

Some additional comments related to the working conditions of the spectrometer and/or reliability of the above main results are in order here. The agreement between the D results and conventional theory necessarily means that H exhibits an "anomalous" behavior. Namely, the H recoil peak is shifted to positive \( y \) values at high energy transfer, which is impossible within any conventional theory. However, by artificially changing the instrument’s calibration parameters,
the position of the H recoil peak in y space could be "shifted back" to become centered at $y = 0$ and in agreement with conventional theory. But then the D recoil peak would be also shifted to negative y values, thus contradicting numerous literature reports [51, 52, 50] that D obeys conventional theory well as to the position of its recoil peak and thereby refuting those reports [44]. See also Appendix C.

5. Discussion

Observation (B) stands in strict contrast to conventional NCS theory for the reasons described above. It may, however, find a plausible explanation within the framework of open quantum system dynamics, as we propose in the following.

Before collision, the proton is assumed to be bound in an effective BO potential which, in the usual theoretical picture [12], is related to the "environment" $E$ (the two electrons and the second proton of H$_2$); the three quantum systems $n$, $p$ and $E$ are not entangled. The $n-p$ collision creates QE between the two particles, indicated by $\rho_{np}(t)$, cf. Sect. 2. Furthermore, within the time window $\tau_{sc}$, the struck proton collides and/or strongly interacts with its environment $E$, causing a complex many-body quantum dynamical process. These interactions create new entanglement between the involved charged quantum systems $p$ and $E$, which then naturally leads to decoherence of the neutron-proton system under investigation. Thus one may write

$$\rho_n(0) \otimes \rho_p(0) \otimes \rho_E(0) \rightarrow \rho_{np}(t) \otimes \rho_E(t) \rightarrow \rho_n(t') \otimes \rho_{pe}(t') \rightarrow \ldots$$

(22)

for $0 \leq t < t' \leq \tau_{sc}$. The third step of this dynamical chain of events is due to the "measurement" of $p$ by the environment $E$ [6, 7, 8]. (Later on, at time $t''$, and after equilibration of the highly excited system "$p$ plus $E"$, a non-entangled state $\rho_p(t'') \otimes \rho_E(t'')$ may occur again.)

For our purposes, it is important to clarify the physical cause of decoherence in the actual experimental context. To do so, we note that the $n-p$ scattering time related to our NCS experiment turns out to be of the same order as the characteristic time of electronic motion and/or charge redistribution, $\tau_{el.motion}$, following the violent excitation and/or electronic "shake-up" of $E$, both lying in the attosecond time range [53]

$$\tau_{sc} \sim \tau_{el.motion}.$$  

(23)

These excitations are caused by the sudden movement of the struck proton which, in our experiments, has a mean kinetic energy and an energy spread of several electron volts after the collision, which is of the order of the separation of the Born-Oppenheimer molecular electronic levels. Under such conditions, neither the adiabatic nor the sudden approximation [54] can be applied, and thus the "environment" $E$ appears to play an active role in the quantum dynamics of the $p-n$ system. Additionally, neither can $E$ be considered here as a memory-less (Markov) bath, nor is it completely "frozen". Using common illustrative terms, one can say that $E$ continuously monitors, observes, or measures the $p-n$ system. This leads to disentanglement and decoherence, which may be expected to have a sub-femtosecond characteristic time [53].

Obviously, the relation (23) holds only for a restricted range of neutron energies (e.g. those of our experiment), and thus is certainly not valid in conventional thermal and/or cold neutron scattering.

That $\tau_{el.motion}$ lies in the sub-femtosecond time regime has been confirmed by attosecond laser spectroscopy. For example, Goulielmakis et al. [55] reported the real-time observation of valence electron motion in Kr atoms. After ionization, the electron-ion pair was demonstrated to constitute an entangled system and its attosecond decoherence was probed.

As mentioned above, this physical picture is not in line with conventional NCS (and general neutron scattering) theory, in which the entanglement between the neutron and the struck
particle is ignored. However, as shown in Sect. 2, denying this entanglement contradicts the principles of quantum mechanics.

Furthermore, conventional NCS theory assumes that single-particle properties are measured, that the environment has no dynamical degrees of freedom (since it is represented by an effective Born-Oppenheimer potential), and that stationary scattering states are fully sufficient for the theoretical description; see also below.

That the effect discussed here is observed for H$_2$ but not for D$_2$ (at forward scattering angles) indicates that molecular dissociation might play an important role here. At the recoil energy, H$_2$ dissociation occurs at approximately $q > 68$ Å$^{-1}$, a condition readily met at $\theta_{high}$ where $q_{rec} \sim 95$ Å$^{-1}$ (and corresponding $\hbar \omega_{rec} \sim 19$ eV). In contrast, dissociation in D$_2$ would occur at about $q > 96$ Å$^{-1}$, which is much higher than the available $q_{rec} \sim 60 - 65$ Å$^{-1}$ at $\theta_{high}$ [52]. Consequently, at $\theta_{high}$, the neutron—H scattering probes final unbound states in the continuum, whereas neutron—D scattering takes place in the physical space of bound states only. The irreversible electronic shake-up caused by H—H bond breaking should exhibit quite different dynamics from the excitation of bound states of H$_2$ or D$_2$, thus leading to potentially different decoherence dynamics. Thus it is tempting to assume that the specific decoherence mechanisms in these two cases be the cause for the qualitatively different scattering behavior of H$_2$ and D$_2$.

Needless to say, the complex five-body dynamical problem (i.e. neutron plus two nuclei plus two electrons) has no analytic solution, and a proper quantitative theoretical model of it is not available thus far. However, first theoretical-numerical investigations of the proton momentum distribution $n_H(p)$ in condensed systems, in relation with NCS processes, have been recently initiated [56].

6. Additional remarks and conclusions

6.1. Evidence of decoherence effect in D$_2$ in backward scattering

![Figure 5](https://example.com/figure5.png)

**Figure 5.** Compton profiles of gaseous D$_2$ at 41 K, preliminary results. Backward scattering, $J_D(y)$ obtained from 87 detectors. The data were taken during the same experiment as that reported above; see Fig. 3.

The above results obtained from H$_2$, together with the proposed theoretical interpretation,
suggest that NCS from D$_2$ at *backward* scattering angles, $\theta_{\text{back}} > 90^\circ$, may also show the decoherence effect under consideration. This is because the energy and momentum transfers on D are, in this regime, considerably higher than in the "forward" scattering regime. The preliminary findings of our current investigations are as follows.

Fig. 5 shows the accumulated D Compton profile (not normalized, as measured) measured by 87 detectors positioned in the "backward" scattering regime of Vesuvio at the specified angular range. The shift of $J_D(y)$ to positive $y$-values is again clearly visible in the data, even without any data processing or corrections. We emphasize again that this stands in blatant contradiction of conventional theory. Qualitatively the same shift was observed in various groupings containing 10 detectors each. Therefore we can confidently state that the effect under consideration is reproducible, significant and, within experimental error, constant over the available backward angular range of $132^\circ - 163^\circ$.

For the determination of this $J_D(y)$ we used the "standard" instrument calibration data (i.e., the associated file IP0002) provided to the users of Vesuvio, since direct calibration data are not available for the backward-scattering detectors. The standard procedure of the instrumental calibration was very recently published [58]. Since there is no H recoil peak in backward scattering to compare with, as done in the analysis of measurements in the forward scattering regime, we determined the position of the Al and Pb recoil peaks originating from the cell, and found that they are centered at the conventionally expected value $y = 0$.

6.2. On conventional theory of neutron scattering and decoherence

It is sometimes claimed that the phenomena of entanglement and decoherence are irrelevant and/or nonexistent in the physical context of general neutron scattering and NCS [59]. Therefore the following explanations and clarifications should be helpful.

Conventional NCS theory [12], as well as general neutron scattering theory [39, 40, 41], are based on quantum theory of systems with well defined Hamiltonians, which implies that these are *closed* quantum systems, and the associated *unitary* time evolution. The basic formalism is that of time-dependent first-order perturbation theory, and in particular Fermi’s Golden Rule; cf. [40], Eq. (2.15). The right-hand side of this formula contains Fermi’s Golden Rule [40, 41] with the delta-function explicitly included in order to guarantee strict energy conservation. $V$ is the Fermi pseudopotential; $b$ is the bound scattering length of atom; $|\nu_i\rangle$, with $i = 0, 1$ are stationary states (with energies $E_{\nu_i}$) of the complete closed system before and after scattering, respectively; the remaining symbols are defined above.

The assumption that $|\nu_i\rangle$ are stationary states of the assumed closed system, i.e. eigenfunctions of the complete Hamiltonian $H_c$ of the N-body system, is essential. This is because the further derivations leading to the well known van Hove results, e.g. the intermediate correlation function

$$F(\mathbf{q}, t) = \frac{1}{N} \sum_{j,k} \langle \exp(-i\mathbf{q} \cdot \mathbf{r}_j(0)) \exp(i\mathbf{q} \cdot \mathbf{r}_k(t)) \rangle$$

and the dynamic structure factor

$$S(\mathbf{q}, \omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \exp(-i\omega t) F(\mathbf{q}, t) \, dt$$

would be invalid. For further details the interested reader is referred to the above references.
are based on the introduction of the Heisenberg operators

\[ r(t) = U(t)^\dagger r U(t) \]  \hspace{1cm} (27)

where \( U(t) \) the unitary time evolution operator

\[ U(t) = \exp(-\frac{i}{\hbar} H_c t) \] \hspace{1cm} (28)

i.e., \( U(t)^\dagger = U(t)^{-1} \) holds. Recall that \( H_c \) is the Hamiltonian of the closed \( N \)-body system — not only of the single nucleus in the two-body neutron—nucleus collision [40, 41]. Note also that neutron–nucleus entanglement, caused by their interaction during the scattering process, plays no role in this formalism.

As an approximation in conventional NCS theory, the complete Hamiltonian is replaced "by hand" with a Hamiltonian of one particle in some effective Born-Oppenheimer potential [12, 50]. In this theoretical framework, of course, any non-unitary decoherence processes in the system play no role "by definition".

However, the scattering nucleus in the aforementioned experiments represents an open quantum system strongly interacting with particles in its environment. Hence the assumed \( N \)-body stationary states of the complete Hamiltonian are a means to perform calculations and lack direct physical significance.

The aforementioned impossibility to incorporate decoherence in the standard formalism of neutron scattering, which is governed by \( t \)-invariant evolution, leads sometimes to bizarre assumptions. For example, the criticism of Ref. [59] against the theoretical interpretation [53] of the "anomalous" intensity decrease effect [16, 17, 18, 19] is based on the unphysical form of a decohering density matrix introduced into the calculations by

\[ \rho_{i,j}(r - r') \rightarrow \rho_{i,j}(r - r') \exp(-\Lambda(r - r')^2|t|) \] \hspace{1cm} (29)

see [59], Eq. (36). Note the incorrect assumption represented by the absolute value \( |t| \). This makes the functional form of the assumed density matrix \( t \)-inversion invariant, thus standing in blatant contrast to the spirit and physical nature of decoherence.

### 6.3. Further remarks

(A) Since the observed new effect appears to have a fundamental theoretical basis given by three independent models (see Section 3), we believe our results are of generic nature.

(B) Generally speaking, the results presented above suggest that the relevant theoretical framework of "fast" collisional processes is that of non-unitary dynamics of open quantum systems [6, 7, 8, 9]. Moreover, decoherence seems to play a more prominent role here which has not been recognized so far. The quantitative theoretical analysis of the new results remains a challenge to modern theory.

The decoherence appearing in the complex dynamics summarized in relations (22) merits further comments. Decoherence is the focus of very active research on the foundations of quantum theory. Since 1970, various pioneering theoretical investigations of related processes showed that complete information about a particle (here: struck proton) is carried away into the "environment" by interacting microscopic objects and/or a thermal bath. The corresponding information transfer should be considered irreversible, at least in the sense that the "lost" information cannot practically be retrieved. The reader interested in the historical development of the fundamental theory of decoherence may also refer to the original references [60, 61, 62, 27, 28].

(C) The above NCS results and the suggested qualitative interpretation may lead to the following proposition: Not a wavefunction (state vector) \( \Psi \), but a (reduced) density operator
$\rho$ is the physical entity of basic importance. In other terms, $\rho$ is not a mere "approximation" related to "missing knowledge" of an "exact state" $\Psi$, but rather the opposite holds true: $\Psi$ ought to be an approximation for the description of real physical states.

This viewpoint is in line with the theoretical work of Beretta [63]. This work deals with model evolution equations (applicable not only to open systems but also to closed isolated systems) capable of describing – simultaneously and in competition with the usual Hamiltonian unitary evolution – the natural tendency of any initial non-equilibrium state to relax towards thermodynamic equilibrium. The author also makes the following interesting point:

"The central conceptual difference between the proposed approach, and the approaches based on attempting to derive the KSGL [Kossakowski-Sudarshan-Gorini-Lindblad] equation, is that this approach regards a non-pure density matrix as representing a real ontological object, the actual state of the world, and is not understood as just an epistemic ignorance of which particular pure state the world is 'really' in." (See [63], p. 142.)

Furthermore, the characteristic time parameter $\tau_D$ in Beretta's theory (see [63], Eq. (35)) called "intrinsic dissipative time", seems to be conceptually connected with the characteristic decoherence time of the scattering process under consideration.

Moreover, Beretta's geometrically motivated construction effectively suppresses the term corresponding to

$$R^\dagger \rho R$$

in the Lindblad equation (7). This difference between the two theoretical models can be subjected to experimental test. Namely, in view of the novel experimental NCS findings presented above, it would be very interesting to investigate the predictions of Beretta's theory with respect to the experimental observations, and compare them with those of the Lindblad equation. This may shed new light on the fundamentals of decoherence, microscopic irreversibility, and other non-unitary dynamical processes.

(D) In this context it should be pointed out that the description of open quantum systems is possible not only in terms of a reduced density matrix, but also in terms of wave functions, but these are then usually solutions of effective nonlinear Schrödinger equations (see, e.g. [64]).

(E) One may wonder why the dynamics of open quantum systems should be relevant for the system "neutron plus proton of H$_2$", although "neutron plus H$_2$ molecule" (in gas phase) is a closed system (in the theoretical frame of non-relativistic quantum mechanics.) Here it is instructive to refer to the aforementioned investigation of krypton atoms by Goulielmakis et al. [55], applying attosecond laser-spectroscopic techniques. As a result, even this monoatomic system was found to exhibit short-lived entanglement and decoherence, which are typical phenomena of open quantum systems (see above).

(F) Owing to the fact that the timescale of the considered scattering process and of electronic motion are of the same order, both being in the attosecond regime, we anticipate the above phenomenon to be a starting point for more sophisticated time-dependent studies in atoms (like e.g. ion–electron decoherence accompanying electron extraction from an atom with an attosecond laser pulse) and in molecules (e.g. chemical bond cleavage caused by collisions of other probe particles with molecules).

(G) A few remarks on the "anomalous" intensity effect [16, 17, 20, 53, 19] in molecular hydrogen are in order. Previous NCS experiments on liquid H$_2$ and D$_2$ [57] performed in 2003/04 with the earlier instrumental setup eVS showed that the "anomalous" ratio of the scattering intensities $I_H/I_D$ is independent of $\theta$ in the whole scattering angle range $30^\circ < \theta < 70^\circ$. However, the instrumental energy resolution at that time was significantly inferior than that of the present setup Vesuvio (see [58] for details) and thus possible peak-overlap effects might have masked an angular dependence in the measured ratio $I_H/I_D$. Additionally, the full details of the associated calibration parameters (cf. Appendix C) are not available to us, and
thus a possible positive y-shift of the H-recoil peak in the earlier data [57] cannot be tested reliably. (Note that the detectors of eVS were freely movable and their positions were often changed after each experiment.) Decoherence has been proposed to be the common theoretical basis (i) of the ”anomalous” intensity [53, 18] and (ii) of the y-shift of Compton profile [22], but a quantitative treatment is still the subject of current theoretical explorations. Further experimental investigations on H$_2$–D$_2$ mixtures and HD are planned.

(H) The general character of the decoherence effect of Refs. [31, 32, 42, 43] leads one to speculate that the new findings presented in this paper might be of relevance for quite different scattering processes (e.g., electron—molecule scattering, proton—nucleus scattering in high energy physics, etc.).

Additionally, as decoherence represents the main barrier to the realization of a quantum computer [5], the above results may also be of relevance for the performance of read-write processes and/or quantum-gate operations in quantum information devices. Undoubtedly a deeper understanding of the relation between decoherence and disentanglement, and the underlying non-unitary quantum dynamics, will be of importance for both the foundation of quantum mechanics and practical quantum information applications.

6.4. Note added
Very recently (March 2012), the preliminary results of Fig. 5, section 6.1, have been confirmed and presented in detail in Ref. [80].

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Appendix A. Neutron Compton scattering
This appendix aims at presenting a complete, although concise, account of all essential details of the applied experimental NCS method, instrumental details, and data analysis leading to the results presented in the main text.

We begin with a short description of the basics of NCS and its characteristic time window, following Watson [12].

NCS differs from other neutron scattering methods by the high energy and momentum transfers involved, typically in excess of several eV and 20 Å$^{-1}$, respectively. Neutron scattering is renowned as a sensitive probe of collective properties in condensed matter, but in this extreme range of energies and momenta – the so-called impulse limit – it is widely believed that single-particle properties are probed. The physical picture advocated is that the neutron-nucleus (or neutron-atom) scattering process occurs so rapidly, compared with the time-scales of atomic motion in the sample, that the measured response is rather simply related to the equilibrium momentum distribution of the atoms.

In the impulse limit, the scattered intensity, as a function of energy measured by a detector at a fixed scattering angle, consists of a peak centred at the ”recoil energy”

$$E_{\text{rec}} = \hbar \omega_{\text{rec}} = \hbar^2 q^2 / 2M$$

which represents the energy of recoil of a stationary nucleus with mass M owing to the collision. The dependence of the peak position on the mass M of the struck nucleus (atom) implies that the scattering from different atoms appears at different energies. This ability to
separate contributions from different atomic species (and isotopes) is a very useful feature of the NCS technique. Due to the smallness of the nucleus compared with the neutron’s de Broglie wavelength (of the order of 0.1 Å), the scattering is isotropic (i.e., s-wave scattering) in the centre-of-mass system.

Doppler broadening results in a recoil peak whose width is proportional to the mean kinetic energy of the atoms, and whose detailed shape depends on the distribution of atomic momenta. The principal tool in interpreting experiments is the so-called impulse approximation, IA [12, 46]. The IA treats the scattering essentially as a billiard-like collision of two mass points and predicts a precise relationship between the scattering data, in the form of the Compton profile (denoted as \( J(y) \), see below), and the atomic momentum distribution. This makes it possible to compare results of experiments with predictions of theoretical models of microscopic properties, e.g. effective Born-Oppenheimer potentials of the struck atoms as being calculated by quantum-chemistry methods.

NCS is closely related to techniques of other fields of physics [65]. Compton scattering using x-rays, gamma rays, or electrons in the 10—100 keV regime provides information on electron momentum distributions [66, 67, 68]. In nuclear physics, one probes the momentum distribution of nucleons in nuclei with deep inelastic scattering (DIS) [69] of protons or electrons with energies of 100—1000 MeV. At still higher energies (e.g., 10 GeV and above), DIS of electrons, muons or neutrinos [70, 71, 72] probes the internal structure of nucleons and has been a key experiment in the confirmation of the existence of quarks.

Appendix A.1. Scattering time

We now consider the characteristic timescale of an elementary scattering process. As mentioned in the Introduction, this ”scattering time” is very short; in our NCS experiments, for the neutron-proton collision it is of the order \( \tau_{sc} \sim 10^{-16} - 10^{-15} \) s.

The quantity \( \tau_{sc} \) was first introduced by Sears [46, 12] as

\[
\tau_{sc} \approx \frac{M}{q \sqrt{\langle p_q^2 \rangle}},
\]

(A.1)

where \( p_q \) is the projection of particle’s momentum \( \mathbf{p} \) along \( \mathbf{q} \) and the expectation value \( \langle ... \rangle \) is taken over the particle’s ground state. It is also found that \( \tau_{sc} \approx \hbar/D_{eq} \), where \( D_{eq} \) is the width of the dynamic structure factor at \( q = \text{const} \). A similar estimate of \( \tau_{sc} \) can be obtained with the relation

\[
\Delta_E \tau_{sc} \geq \frac{\pi \hbar}{2}
\]

(A.2)

derived by Ballentine [73], where \( \tau_{sc} \) may be interpreted as the shortest time required for the state vector \( |\psi(t)\rangle \) to become orthogonal to the initial state \( |\psi(0)\rangle \), and \( \Delta_E \) represents a standard measure of the statistical spread of energy in the state.

The terminology ”scattering time” can be misleading in some circumstances. The physical meaning of \( \tau_{sc} \) is as follows. First, the time interval during which the neutron and the nucleus are in physical proximity is much shorter, as the following estimate shows. For example, a neutron with kinetic energy \( E_0 \approx 10 \) eV will pass a distance of \( 10^{-5} \) Å, which is about the range of the strong nuclear force, in a much shorter time, i.e. \( 10^{-19} - 10^{-20} \) s. However, this is not in conflict with the above estimate, for the following important reason: \( \tau_{sc} \) gives a measure of the length of the time interval during which the neutron-proton collision may occur [22]. Therefore, in view of the ”attosecond double-slit” experiment by Paulus and co-workers [74], the scattering process under consideration may be viewed as a quantum interference process over the time interval \( \tau_{sc} \). This conceptual viewpoint (i.e., interference-in-time) is also in line with the results obtained from a temporally diffracted beam of slow Cesium atoms, using a Young-slit-type interferometer, in
the time domain [75]. The first theoretical study of diffraction by a time slit was presented by Moshinski [76].

Appendix B. Experimental procedure

Our experiments applying the NCS method were done at the electron volt spectrometer (presently called "Vesuvio") or the neutron spallation source ISIS, Rutherford Appleton Laboratory, UK. As originally proposed by Hohenberg and Platzman, neutron scattering at high energy and momentum transfers (say, $\hbar \omega > 1$ eV, $q > 20$ Å$^{-1}$) can be used to directly measure the distribution of momentum $\hbar \mathbf{p}$ of light atoms in condensed matter systems [11]. Here it is assumed that the scattering is essentially incoherent and the so-called Impulse Approximation (IA) applies, and thus single-particle properties are probed [11, 46, 12].

The Vesuvio spectrometer applies the standard time-of-flight (TOF) technique to determine the energy and momentum transfers to the neutron during each scattering event. The TOF for each detected neutron is given by

$$ t = \frac{L_0}{v_0} + \frac{L_1}{v_1} + t_0 \quad (B.1) $$

Here $L_0$ (equal to 11.005 m [58]) is the distance from the source to the sample, $L_1$ (typically 0.50 – 0.65 m) that from sample to detector. $v_0$ and $v_1$ are the velocities of the incident and scattered neutron, respectively. The detector is at scattering angle $\theta$ (about 30° – 67° for forward scattering). $t_0$ is a small time offset due largely to electronic delays. For the data analysis of our experiment [22, 44] we used values for $L_1$ and $\theta$ for each individual detector measured directly with steel rules and a purpose-built protractor [58].

Vesuvio is a so-called "inverse geometry" instrument meaning that the final velocity $v_1$ of the neutrons contributing to the TOF spectra is fixed, whereas $v_0$ varies; for details see e.g. [77]. The final neutron energy is presently fixed at $E_1 = 4906$ meV (the resonance energy of Au-197 used as analyzer foil [22]), corresponding to velocity $v_1 = 3.063 \times 10^4$ m/s and wavevector of scattered neutrons $k_1 = 48.66$ Å$^{-1}$.

It follows from Eq. (B.1) that to each $t$ corresponds to an initial velocity $v_0$ and thus to an energy transfer

$$ \hbar \omega = \frac{(\hbar k_0)^2}{2m} - \frac{(\hbar k_1)^2}{2m} \quad (B.2) $$

For the corresponding momentum transfer $\hbar q$ from the neutron to the struck atom, $\hbar q = \hbar k_0 - \hbar k_1$,

$$ q = \sqrt{k_0^2 + k_1^2 - 2k_0k_1 \cos \theta} \quad (B.3) $$

Hence, for each value of $t$ in an experimental TOF spectrum recorded at a particular detector, the associated momentum ($\hbar q$) and energy ($\hbar \omega$) transfers from the neutron to the struck particle are uniquely determined. For scattering from H, $q$ and $\omega$ vary considerably over the range of the recoil peak, especially for high scattering angles.

From the measured time-of-flight (TOF) spectra, momentum ($\hbar q$) and energy ($\hbar \omega$) transfers from the neutron to the struck particle are determined by standard methods. Within the IA, the elastic collision of a neutron with an approximately free atom with mass $M$ and initial momentum $\mathbf{p}$ results in the neutron’s lost energy $\hbar \omega$ being transferred to the struck atom:

$$ \hbar \omega = \frac{(\hbar q + \mathbf{p})^2}{2M} - \frac{p^2}{2M} = \hbar \omega_{\text{rec}} + \hbar q \cdot \mathbf{p}/M \quad (B.4) $$

The recoil energy $\hbar \omega_{\text{rec}} = \hbar^2 q^2/2M$ represents the energy of recoil of a stationary nucleus owing to the collision. ($\hbar q$: momentum transfer from neutron to the struck nucleus/atom.)
Thus scattering from a gaseous sample of such atoms leads to a recoil peak centered at $\hbar \omega_{\text{rec}}$, exhibiting a peak width caused by the term $\hbar \mathbf{q} \cdot \mathbf{p} / M$ which represents Doppler broadening.

The above energy balance holds exactly in the IA If however final state effects (FSE) become apparent, as in the above experiments at lower scattering angles, the maximum of the recoil peak is shifted to lower energy than the recoil energy. This shift is also analytically proven for the three-dimensional quantum harmonic oscillator.

As discussed in the main text, a peak-maximum shift to higher energies than $\hbar \omega_{\text{rec}}$ is impossible in conventional NCS theory.

For scattering from free atoms with zero initial momentum, $p = 0$, conservation of kinetic energy and momentum in an elastic neutron-atom collision yield the kinematic relation

$$\frac{v_1}{v_0} = \frac{k_1}{k_0} = \frac{\cos \theta + \sqrt{(M/m)^2 - \sin^2 \theta}}{M/m + 1} \quad (B.5)$$

which holds in both quantum and classical mechanics. Here $m$ and $M$ are the masses of the neutron and struck nucleus, respectively. This physically corresponds to neutrons detected at the centre of the measured recoil peak.

**Appendix C. Compton profiles and their measurement**

From the measured time-of-flight (TOF) spectra one determines the dynamic structure factor [40], which in the case of the IA takes the simple form [12]

$$S(q, \omega) = \int n(p) \delta (\hbar \omega - \hbar \omega_{\text{rec}} - \hbar \mathbf{q} \cdot \mathbf{p} / M) \, dp \ . \quad (C.1)$$

$n(p)$ is the momentum distribution of the struck particle before collision. The delta function incorporates energy conservation.

To compare results obtained with different detectors (and/or to improve counting statistics), one usually applies the so-called West scaling (or $y$ scaling) [46, 47], which, for isotropic samples, amounts to the following. In the IA can be shown that, for each kind of struck particles of mass $M$, the two variables $\omega$ and $q$ are uniquely coupled through a new scaling variable with the physical dimensions of momentum, commonly denoted by $\hbar y$ (or simply $y$) and defined by

$$\hbar y = \frac{M}{\hbar q} (\hbar \omega - \hbar \omega_{\text{rec}}) \ , \quad (C.2)$$

which simplifies the dynamic structure factor to [12]

$$S(q, \omega) = \frac{M}{\hbar q} J(y), \quad \text{with} \quad J(y) = \int n(p) \delta (p \cdot e_q - \hbar y) \, dp \quad (C.3)$$

$J(y)$ is the Compton profile, which gives the distribution of the one-dimensional projection $\hbar y$ of atomic momentum $p$ (before collision) along the $q$ direction represented by $e_q$, i.e. $|e_q| = 1$. This scaling property of the IA implies that all detectors (at various scattering angles) should yield the same Compton profile $J(y)$ [12, 46, 47]. This quantity, and the associated $n(p)$, are widely believed to depend on the effective Born-Oppenheimer potential $V(r)$ of the struck particle [12]. For a harmonic $V(r)$ the associated $J(y)$ is Gaussian.

The Compton profiles shown in Figs. 3-5 were obtained by transforming to $y$ space the TOF spectra measured as described above.

We have analyzed the sensitivity of the peak positions of $J(y)$ in $y$ space to calibration uncertainties and found that the results in $y$ space are reliable to better than 0.3 Å$^{-1}$ for each...
single detector [49]. This estimate is supported by the small scattering of the positions of the individual-detector $J(y)$ curves shown in all four Figs. 3a,b and 4a,b.

Our measurements on $H_2$ and $D_2$ gas were made using an all-aluminium pressure cell with parallel internal surfaces perpendicular to the incoming beam. The mounting of the cell within the cryostat ensured reproducibility of the sample position between experiments to better than about 2 mm, with negligible effect on the comparison of results between $H_2$ and $D_2$ [49]. The spectrometer configuration was identical in the two experiments. Exactly the same analysis was applied to each data set.

As mentioned above and detailed in our papers [22, 44], we used calibration parameters measured directly with steel rules and a protractor [58] by the ISIS Metrology Group, for our data analysis. In contrast, the standardly available Vesuvio calibration parameters [45] (listed in instrument parameter files) are obtained by an iterative sequential procedure involving inelastic and diffraction measurements [58]. Using the standard calibration parameters, we demonstrated [44] that, if the experimental results for $H$ in gaseous $H_2$ are in agreement with conventional theory (as claimed in [45]), then those for $D$ in gaseous $D_2$ obtained in the same way cannot be, and vice-versa. Crucial to this alternative, however, the standardly available calibration parameters may be "adjusted" to ensure that the position of the $H$ peak conforms to conventional theory; see Ref. [10] cited in [44].
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