Attosecond neutron scattering from open quantum systems: entanglement and decoherence phenomena of protons in condensed matter and molecules

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Abstract. In general, nuclei and electrons in condensed matter and/or molecules are entangled, due to the prevailing (electromagnetic) interactions. As a matter of fact, the "environment" interacting with a microscopic system of interest (e.g. a proton) causes the destruction of the entanglement. This process, called decoherence, has until now prevented experimenters from directly accessing atomic and/or nuclear entanglement effects in real experiments. However, our neutron and electron Compton scattering experiments from protons (H-atoms) in several condensed systems and molecules at ambient conditions demonstrated a new striking effect, i.e. an "anomalous" decrease of scattering intensity from protons, which seem to become partially "invisible" to the neutrons. This effect, which has no interpretation within conventional neutron scattering theory, is proposed to be caused by the non-unitary time evolution (due to decoherence) during the ultrashort, but finite, time-window of the neutron-proton scattering process. Due to the large energy (several eV) and momentum (20-200 Å⁻¹) transfers of these experiments the collisional time between the probe particle and a struck proton is 100–1000 attoseconds long. It is shown that, due to this short timescale, the scattering process must be theoretically treated within quantum dynamics of open quantum systems. New experimental neutron Compton scattering results from a single crystal KHCO₃ are presented. They provide the first direct evidence for a connection between the momentum distribution and the "anomalous" scattering intensity of H. Additionally, recent experiments with electron-atom Compton scattering at momentum transfers are mentioned, which reveal the presence of this effect even in scattering from single H₂ molecules in the gas phase. Theoretical discussions "from first principles" are presented, also in relation to the quantum Zeno effect, which underline the crucial role of decoherence in the considered experiments. The experimental results and their qualitative interpretation show that epithermal neutrons being available at spallation sources (e.g. ISIS/U.K., SNS/U.S.A. or ESS/Sweden), and electron spectrometers with large scattering angles, provide tools for investigating new physical and chemical phenomena in the sub-femtosecond timescale.

1. Introduction
The counter-intuitive phenomenon of quantum entanglement (QE) — also called Einstein-Podolsky-Rosen correlations or Schrödinger cat states — between two or more quantum systems has emerged as the most emblematic feature of quantum mechanics [1]. QE is at the heart of the intriguing complexity of describing quantum many-body systems in physics, chemistry and biology.

Nuclei and electrons in condensed matter and/or molecules are usually entangled, due to the prevailing (electromagnetic) interactions. Usually, the "environment" of a microscopic scattering system (e.g. a proton) causes an ultrafast decoherence (see, e.g. [2]), thus making atomic and/or nuclear entanglement effects not directly accessible to experiments. It may be noted that the phenomenon of decoherence...
represents the main obstacle preventing the construction of a quantum computer [3]. In this article, the term entanglement may refer to various dynamical variables (e.g. position, momentum and/or spin degrees of freedom).

However, as shown below, neutron Compton scattering (NCS) is an ultrafast scattering technique and thus it may open up a way for investigation of short-lived quantum entanglement even in condensed matter at ambient conditions. In this respect it is important to emphasize that, under the physical conditions of NCS, each impinging neutron collides with one nucleus (e.g. one proton of H\textsubscript{2}) only, and not with the "entire molecule". This is clearly demonstrated with the aid of the measured time-of-flight (TOF) spectra: E.g., the position of the measured H-recoil peak of molecular hydrogen, H\textsubscript{2}, lies always in the TOF-position corresponding to the collision with a particle of one atomic mass unit (i.e., the proton), and not two atomic mass units (i.e., the whole hydrogen molecule).

This fact also shows that the NCS process represents scattering from open quantum systems [2], e.g. one proton of H\textsubscript{2} molecules. Consequently, the remaining adjacent particles (e.g., the second proton and the two electrons of H\textsubscript{2}) represent the environment which strongly interacts with the scattering system under investigation (i.e., a single proton, in the considered example). These strong interactions are the cause of ultrafast decoherence. The conceptual connection and/or interrelation with the physical field of quantum optics is obvious.

Starting in 1995, our NCS experiments from protons (H-atoms) in several condensed systems and molecules [4] demonstrated a new striking effect, i.e. an "anomalous" decrease of scattering intensity from protons, which seem to become partially "invisible" to the neutrons [4, 5]. Several experiments were done at ambient conditions. Due to the large energy (several eV) and momentum transfers of these experiments, the collisional time between the probe particle and a struck proton is about 100–1000 attoseconds long (see below).

Many NCS experiments in various systems (molecular hydrogen, metallic hydrides, polymers, inorganic crystals, gases, etc.) have been carried out successfully and demonstrated this attosecond quantum effect; see e.g. [4, 5] and references cited therein.

This effect has no interpretation within conventional scattering theory of NCS (also called deep-inelastic neutron scattering, DINS) [6, 7]. According to our theoretical predictions and/or investigation, it is caused by the non-unitary time evolution (due to decoherence) during the ultrashort, but finite, time-window of the neutron-proton scattering process; see theoretical discussions below.

For clarity, and to be specific, in the following we constrain our considerations to the physical context of NCS only. The following sections present:

(A) a short description of the NCS technique and a novel experimental result;
(B) a discussion of the characteristic ultashort (attosecond) timescale of NCS from protons (H-atoms), and the necessity to apply the theory of open quantum systems to the scattering process;
(C) a qualitative theoretical understanding "from first principles" of the new effect of "anomalous" decrease of scattering intensity, which is based on the quantum Zeno effect [8, 9, 10] and/or a fundamental theoretical result by Schulman and Gaveau [11, 12];
(D) a theoretical model of scattering which "extends" conventional theory through the inclusion of entanglement and decoherence [13];
(E) a "first principles" theoretical model of scattering from open quantum systems subject to a Lindblad-type dynamics [14]. In particular we consider the case that the duration of the scattering process is of similar order as the decoherence time of the scatterer.

The theoretical models (C)-(E), which have a common conceptual basis, show that a reduction of the system's transition rate due to scattering is derived. This is tantamount to a shortfall of scattering intensity.

The results and their qualitative interpretation show that epithermal (say, 1–100 eV) neutrons being available at spallation sources (presently at ISIS/U.K.), and electron spectrometers with large scattering
angles and associated large momentum transfers, provide novel tools for investigation of new attosecond physical and chemical phenomena in condensed matter and molecules.

2. Neutron Compton scattering

The experimental method utilized here is neutron Compton scattering (NCS), also called deep inelastic neutron scattering (DINS), provided by the electron-volt spectrometer Vesuvio (formerly eVS) at the ISIS neutron spallation source, Rutherford Appleton Laboratory, UK. This is an inverse-geometry spectrometer, meaning that the final neutron energy is fixed, see e.g. [7, 15].

Neutron scattering at high energy transfers ($\hbar \omega \gtrsim 1\text{eV}$) and momentum transfers ($\hbar q$, with $q \gtrsim 20\text{\AA}^{-1}$) can be used to directly measure the distribution of momentum $\hbar p$ of light atoms in condensed matter systems[16]. This experimental method (NCS) is analogous to the measurement of the electronic momentum distribution through Compton scattering of X- and $\gamma$-rays from electrons [17], or the nucleon momentum distribution via quasi-elastic electron scattering from nuclei. Here it is assumed that the scattering is essentially incoherent and the so-called impulse approximation (IA) applies [16, 18], and thus single-particle properties are probed [6, 7].

A well founded brief review of the theory of NCS was given by Sears [18], which is as follows. In a time-of-flight (TOF) scattering experiment, cf. figure 1a, one determines from the measured spectra the partial (or: double) differential cross section and the associated dynamic structure factor $S(q, \omega)$.

For the scattering of a neutron by a system of $N$ identical atoms it holds

$$\frac{d^2\sigma}{d\omega d\Omega} = N\beta^2 \frac{k_1}{k_0} S(q, \omega)$$

(b: bound scattering length of atom; $k_0, k_1$: absolute values of wavevectors of incident and scattered neutron; $d\Omega$: small solid angle subtended by the neutron detector in direction of $k_1$ at the target). $\hbar q$ and $\hbar \omega$ are the momentum and energy transfers from the neutron to a scattering nucleus, respectively.; i.e, $\hbar q = \hbar k_0 - \hbar k_1$, $\hbar \omega = E_0 - E_1$. The subscripts "0" and "1" refer to neutron quantities before and after collision, respectively.

Figure 1. (a) Schematic representation of time-of-flight (TOF) spectrometer eVS/Vesuvio of ISIS; adapted from [15]. (b) A TOF-spectrum of a 20:80 H$_2$O-D$_2$O mixture in an Al can together with the corresponding fit (full line). The H and D recoil peaks are well separated from each other and from the joint O/Al peak; adapted from [4]. The peak area of atom X (i.e., H, D, etc.) gives the neutron-X scattering intensity.
The introduced potential of the neutron-nucleus interaction effectuating the neutron scattering is known as Fermi’s pseudo-potential [19, 20, 21]

\[ V(r) = \frac{2\pi\hbar^2}{m} b \delta(r) \]  

(\(m\): neutron mass) which approximates the short-range strong interaction. According to the basic van Hove theory [19] \(S(q, \omega)\) is given by

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

where

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

is the so-called intermediate correlation function. \(\langle ... \rangle\) denotes a thermodynamics average, and \(\mathbf{r}_j(t)\) denotes the position operator of atom \(j\) at time \(t\) in the Heisenberg representation.

Here it should be pointed out that the whole derivations of the above formulas are based on Fermi’s golden rule, which, for scattering processes, is equivalent to the first Born approximation [19, 20, 21]. Recall also that both these approximations are essentially equivalent to first-order perturbation theory. However, this theory does not hold for a singular potential like Eq. (2). The justification for the use of Fermi’s golden rule in these circumstances is that, in combination with Eq. (2), it gives the required result of isotropic (s-wave) scattering for a single fixed nucleus; cf. [20, 21] and references therein.

In NCS \(q = |\mathbf{q}| \gg 2\pi/d\), where \(d\) is the nearest-neighbor distance, and therefore one may neglect the \(j \neq k\) terms in the sum and obtains

\[ F(q, t) = \langle \exp(-i\mathbf{q} \cdot \mathbf{r}(0)) \exp(+i\mathbf{q} \cdot \mathbf{r}(t)) \rangle \]

where \(\mathbf{r}(t)\) denotes the position operator of a scattering atom at time \(t\) in the Heisenberg representation. However, it should be pointed out that \(\mathbf{r}(t)\) is a Heisenberg-operator, and thus it is an \(\tilde{N}\)-body operator, despite the (probably misleading) notation of ”position operator of a scattering atom”.

The last equation is referred to as the incoherent approximation. Here one neglects coherent scattering contributions, because the condition \(q \gg 2\pi/d\) implies that the coherent superposition of scattering amplitudes from different atoms averages to zero over a small solid angle \(d\Omega\) associated with the detector (neutron counter).

To introduce the impulse approximation, IA, one may proceed as follows. In classical mechanics one has \(\mathbf{r}(t) - \mathbf{r}(0) = \int_0^t \mathbf{v}(t')dt'\), where \(\mathbf{v}\) is the velocity of the scattering atom. In quantum mechanics, however, the Heisenberg operators \(\mathbf{v}(t')\) at different times do not commute. Nevertheless, as shown by Sears, it holds

\[ F(q, t) = \exp(i\omega_r t) \left\langle T \exp \left[ i\mathbf{q} \cdot \int_0^t \mathbf{v}(t')dt' \right] \right\rangle \]

where \(T\) is the time-ordering operator and

\[ \hbar\omega_r = \hbar^2 q^2 / 2M \]

represents the energy of recoil of a stationary nucleus with mass \(M\) due to the collision. For sufficiently large \(q\), formally \(q \to \infty\), the right-hand-side (rhs) of Eq. (4) is appreciably different from zero only if \(t \to 0\), and thus \(\mathbf{v}(t') \approx \mathbf{v}(0) \equiv \mathbf{v}\). Thus, for \(q \to \infty\) one obtains

\[ F(q, t) \to \langle \exp[i(\omega_r + \mathbf{q} \cdot \mathbf{v})t] \rangle = F_{IA}(q, t) \]

(8)
and therefore

\[ S(q, \omega) \rightarrow \langle \delta(\omega - \omega_r + q \cdot v) \rangle \equiv S_{IA}(q, \omega). \]  

(9)

The delta function expresses conservation of energy and momentum in the collision of a neutron with an atom having initial velocity \(v\), and follows from the assumed validity of Fermi’s golden rule [20]. This formula represents the impulse approximation (IA). As pointed out by Sears, this approximation neglects interatomic forces in the final state, but not in the initial state, since the thermodynamic average \(\langle \cdots \rangle\) refers to the interacting system.

\(\text{Figure 2. Time-of-flight (TOF) spectra of YH}_3\text{ at various scattering angles } \theta. \text{ (For clarity, the spectra are parallel shifted along the ordinate.) The peak separation between H and the heavier atom (here: Y and Al) increases with } \theta. \text{ The position of the H peak shows that the mass of the recoiling particle is 1 a.m.u.}\)

It should be emphasized that the measured TOF-positions of the recoil peaks correspond to scattering from single nuclei. E.g., the peaks marked with "H" in figure 2 correspond to a recoiling particle of 1.0 atomic mass unit (a.m.u.), that is, to a single proton. The same holds also for NCS from molecular H\(_2\) [22], that is, the scattering particle is a "single proton", and not a H\(_2\) molecule "as a whole" (since the latter has the mass of 2 a.m.u.).

To simplify notations, and in accordance with standard notation used in the literature, the subscript \(\text{IA}\) is dropped in the following.

The dynamic structure factor in the IA can be expressed equivalently as

\[ S(q, \omega) = \int n(p) \delta \left( \frac{\hbar^2 p^2}{2M} - \frac{\hbar^2 (p + q)^2}{2M} \right) dp \]

\[ = \int n(p) \delta(\hbar\omega - \hbar\omega_r - \hbar q \cdot p / M) \ dp . \]  

(10)

\(M\) and \(n(p)\) are the mass and momentum distribution of the scattering atom (nucleus) before collision, respectively. A useful feature of NCS is that the recoil peaks of light atoms (e.g. H, D, He) are well resolved from those of heavier atoms in the TOF-spectra (compare figure 1). In a typical NCS experiment, the characteristic time \(\tau_q\) of a neutron-nucleus collision, termed "scattering time", is very short. For the neutron-proton collision \(\tau_q\) is of the order \(10^{-16} - 10^{-15}\); see below.

As the term \(q \cdot p\) in Eq. (1) shows, the energy distribution of the scattered neutrons are directly related to the distribution of particle momenta \(\hbar p\) (before collision) parallel to the wavevector transfer \(q\). In fact, the nucleus is in a quantum state having a distribution of initial momenta, and the recoil peak will therefore be Doppler broadened.

In the IA can be shown [23] that, for each kind of struck particles of mass \(M\), the two variables \(\omega\) and \(q = |q|\) are explicitly coupled through the scaling variable \(y\) defined as[18, 23]

\[ y = \frac{M}{\hbar^2 q} (\hbar\omega - \hbar\omega_r) , \]  

(11)
which reduces Eq. (10) to

\[ S(q, \omega) = \frac{M}{\hbar q} J(y, \hat{q}), \]  

(12)

with

\[ J(y, \hat{q}) = \int n(p) \delta(p \cdot \hat{q} - \hbar y) \, dp \]  

(13)

where the neutron Compton profile (also termed "longitudinal momentum distribution"), \( J(y, \hat{q}) \), is the distribution of the one-dimensional projection \( \hbar y \) of atomic momentum \( p \) along the \( \hat{q} \)-direction indicated by the dimensionless unit vector \( \hat{q} \) or, equivalently, by the scattering angle \( \theta \). This property is known as \( y \)- or West-scaling[18, 7, 23] and implies that all detectors yield the same \( J(y) \), as pointed out above.

\[ J_M(y) = \begin{cases} M = 1.0078 \text{ a.m.u.} \end{cases} \]

**Figure 3.** A typical Compton profile \( J(y) \) of \( H \) at scattering angle \( \approx 45^\circ \). The broken line represents the experimental resolution function using the Au-foil analyzer, see [15].

For isotropic systems, mostly studied by NCS, the direction \( \hat{q} \) becomes immaterial and one writes \( J(y) \). Thus, for large momentum transfers, the Compton profiles obtained from all detectors (which are set at different scattering angles \( \theta \)) are expected to "collapse" onto one \( \hat{q} \)-independent Compton profile \( J(y) \). According to standard theory, this quantity, and the associated \( n(p) \), depend on the effective Born-Oppenheimer (BO) potential \( V(r) \) of the struck particle. For a harmonic \( V(r) \), the associated \( J(y) \) is Gaussian [6, 7], i.e.,

\[ J(y) \propto \exp(-y^2/2\sigma^2), \]  

(14)

where \( \sigma \) is the width of the longitudinal momentum distribution.

The standard theoretical results described above all presuppose that \( J(y) \) can be described by a single-particle in a potential, that is, that the BO is valid [6, 7]. It is believed that NCS is the only technique capable of measuring momentum distributions and BO potentials directly. The explicit relations between a Compton profile \( J(y) \), a momentum distribution \( n(p) \) and the associated effective BO potential \( V(r) \) are well known in standard NCS theory [6, 7].

2.1. The "anomalous" scattering effect

The first direct experimental evidence of short-lived (that is, attosecond) QE involving protons in condensed matter was provided by means of the novel neutron Compton scattering (NCS) method. Starting in 1995, our experiments on liquid water and H\(_2\)O-D\(_2\)O mixtures [4] have revealed the following striking effect: The intensity of neutrons scattered from protons exhibits a considerable "anomalous" shortfall which can be as much as 30%. This finding unequivocally contradicts conventional theory [4]. Further NCS experiments confirmed the existence of this effect in various condensed matter systems, e.g., metallic hydrides [24, 25], polymers [26, 27], "soft" condensed matter [28], and liquid benzene.
Experimental evidence of the effect under consideration has been provided even in liquid H$_2$ and HD [22, 26].

We now shortly discuss the striking decrease of NCS-intensity from H, which is represented by the violation of the basic equation $R_{\text{exp}} = R_{\text{conv}}$, see below. As an example, let us consider the scattering results [4] from H$_2$O-D$_2$O mixtures. From a measured TOF spectrum, the data analysis procedure [15] determines the partial differential cross section $d^2\sigma/d\omega d\Omega$, Eq. (1). The relevant peak areas $A_X$ (with X=H, D, O) are then extracted from $d^2\sigma/d\omega d\Omega$. Thus one can determine the ratio

$$R_{\text{exp}} \equiv A_H/A_X. \quad (15)$$

According to standard NCS theory [6, 18] the conventionally expected value $R_{\text{conv}}$ of this ratio is calculated with (cf. [4, 15])

$$R_{\text{conv}} = \frac{N_H b_H^2}{N_X b_X^2}. \quad (16)$$

$N_X$ is the number density of atom X. Instead of the conventionally expected equality $R_{\text{exp}} = R_{\text{conv}}$, however, the experimental results presented below (and various others reported in the literature) show that this equation is strongly violated, i.e.,

$$R_{\text{exp}} < R_{\text{conv}}. \quad (17)$$

This surprising effect has no interpretation in the frame of conventional theory [6, 7].

**Figure 4.** TOF spectra from liquid C$_6$H$_6$ in an Nb can, at T=295 K. The error bars are due to counting statistics only. The full lines denote the fitted TOF spectra. The positions of the peaks are not fitting parameters. For the scattering angle $\theta = 65^\circ$ the C- and Nb-recoil peaks overlap. For $\theta = 132^\circ$, however, the maxima of these two peaks are well resolved, thus facilitating the reliable determination of the peak intensities.

**Figure 5.** The experimentally determined ratios of neutron cross-sections of H and C, from the NCS spectra from liquid C$_6$H$_6$, for different scattering angles (positions of the detectors). The vertical line at 14.7 denotes the ratio of cross-sections $\sigma_H/\sigma_C = b_H^2/b_C^2$ according to conventional theory and tabulated values of these quantities.
2.2. NCS results from liquid benzene

For illustration of this “anomalous” intensity effect, earlier results obtained with the eVS-spectrometer from liquid benzene [26] are presented; see figures 4 and 5.

The expected ratio of scattering intensities from H and C, \(R_{\text{conv}}\), is equal to the tabulated values of corresponding cross-sections \(\sigma_X = 4\pi b_X^2\) (with \(X = H, C\)). The measured ratio \(R_{\text{exp}}\) is about 25% smaller than \(R_{\text{conv}}\).

3. NCS from a single crystal KHCO\(_3\): momentum distributions and intensities

We present a striking new result of our current analysis of NCS data [29] obtained from a single crystal of KHCO\(_3\) at room temperature with the eVS/Vesuvio spectrometer. KHCO\(_3\) is a prototypical system for proton transfer dynamics. In a single crystal, all H atoms belong to OH bonds and all these OH bonds are virtually parallel to each other throughout the crystal. Therefore, with a single crystal properly oriented with respect to the neutron beam it is possible to probe the proton dynamics specifically along each degree of freedom of H vibrational motion. Figure 6 shows results of current investigations, which compare for the first time:

(a) the measured kinetic energy of H (which is proportional to \(\sigma_H^2\), where \(\sigma_H\) is the width of H-Compton profile \(J_H(y)\)) at a given scattering angle, which is determined from the measured Compton profile \(J_H(y)\) by standard theory [6, 7], with

(b) the associated measured intensity of the H-recoil peak at given scattering angle (normalized by the sum of the scattering intensities of all heavier peaks, i.e. C, O and K).

\[ \text{Figure 6. NCS from a single crystal KHCO}_3\text{. Measured NCS intensities from H (normalized with those of all heavier peaks) and measured kinetic energies of H, at various scattering angles. Negative (positive) angles: left (right) to beam direction. The H recoil peaks are well separated from the “common” recoil peak of C, O and K. Experimental data from [29].} \]

These new results are quite remarkable for several reasons:

(c) The scattering angle (and thus momentum transfer \(q\)) dependences of both quantities are very strong. The higher kinetic energies at small (positive and negative) scattering angles are due to the simple fact that the momentum transfer \(q\) — which varies over the TOF-H-peak — is chosen to be roughly parallel to the OH bonds, which have a “stronger” local BO potential (and thus also higher vibrational energy, which is well known from conventional vibrational spectroscopy). At larger (positive and negative) scattering angles, \(q\) and the OH-direction build a large angle, and thus \(q\) probes another (and much weaker) local BO potential. Thus the strong scattering-angle dependence of H-kinetic energy is qualitatively well understood.

(d) However, the strong scattering-angle dependence of the H-peak intensity \(I_H\) contradicts the standard NCS-theory [6, 7], according to which \(I_H\) must remain constant for all angles. This result represents the novel “anomalous” scattering intensity effect under consideration.
Moreover, and most surprisingly, the comparison of the two quantities shown in figure 2 shows a clearly visible, strong correlation between them, which remained unnoticed until now. Qualitatively speaking, the "anomalous" intensity deficit from H seems to become stronger for "broader" BO potentials and the associated more delocalized protons. In other terms, it obviously exists a physical connection between H-Compton profile \( (J(y)) \) and H-intensity \( (I_H) \). This finding appears to be in blatant contrast to the associated statement of Ref. [7], see p. 463.

4. Remarks on electron-atom Compton scattering experiments

The "anomalous" scattering effect under consideration has also been validated by electron-atom Compton scattering (ECS) at large momentum transfers. The experiments have been taking place in Australia [5, 30, 31], and most recently in Canada [32]. The experiment [32] may be of particular interest to theoreticians: \( \text{H}_2 \) is the best theoretically investigated molecule, and the scattering protons are those of isolated \( \text{H}_2 \) molecules in the gas phase. A short description of the results is as follows.

ECS from gaseous \( \text{H}_2, \text{D}_2, \text{a} \) 50:50 mixture of \( \text{H}_2 \) and \( \text{D}_2, \) and HD was investigated with 2.25 keV impact energy and 100° scattering angle, corresponding to a momentum transfer with ca. 40 Å\(^{-1}\). In contrast to the typical conditions of NCS, the energy transfers applied here were less than the dissociation energies of the molecules. The spectral positions of the H and D recoil peaks follow standard Rutherford scattering theory. However, the scattering intensities contradict standard theory. Namely, in the spectrum of the 50:50 \( \text{H}_2\text{-D}_2 \) mixture, the integrated intensity of the H peak was about 30% lower (as compared to that of D) than predicted by Rutherford scattering, despite equal screening of nuclear charges by the electrons in all molecules. In contrast, the ratio of scattering intensities from HD was found to agree with Rutherford scattering. (This represents an interesting difference to the NCS experiment [22], and is currently under theoretical investigation.) Recently, a detailed theoretical analysis by Bonham et al. demonstrated that this striking effect cannot be explained by scattering theory calculations based on the first Born and Born-Oppenheimer approximations [33].

Very recently, these novel experimental findings have been criticized to be instrumental artifacts, and it was speculated that the anomaly is caused by differences in the molecular speeds of \( \text{H}_2 \) and \( \text{D}_2 \) [34]. However, these criticisms have been refuted by further experimental investigations [35]. Especially, the measured decrease of H-intensity of about 30% could be expected if the sample were an effusive gas jet which allowed free escape of the sample gas molecules. This is not the case in the ECS experiments since a gas cell was used [32, 35]. Moreover, if such a molecular speed effect were the cause of the observed effect, then the simultaneously performed \( (e, e + \text{ion}) \) mass spectra would show the same anomaly since the \( (e, e + \text{ion}) \) coincidence experiment involves the same gas cell collision region, the same electron transfer and focusing lenses, and the same electron detection hardware as the energy loss function of the spectrometer [35]. Based on these facts, the conclusion of [35] was that the arguments advanced in [34] to "explain" the experimental quasi-elastic \( \text{H}_2\text{-D}_2 \) relative intensity anomaly under consideration, are erroneous.

5. Criticisms and their experimental tests

Considerable efforts to identify possible sources of NCS-experimental errors have been made during the last 10 years; for an account in detail of data analysis and experimental procedures and tests, see [15].

Senesi et al. [36] investigated all the criticisms raised in [37] concerning the effects of instrumental resolution and filter absorption profile, by applying for the first time the so-called "exact method" of data analysis proposed [37]. Data obtained from solid HCl at \( T = 4 \) K were analyzed. The results demonstrated unequivocally the presence of the same anomaly in the scattering from protons, in agreement with the results of the standard data analysis procedure of ISIS [36].

The standard method of NCS-data processing [7, 15] was recently compared with a newly proposed (by B. Dorner, ILL) model-free data-analysis procedure. This procedure is independent of the form of the momentum distribution and the resolution function [38]. In [38], the original results from the
metallic hydride NbH$_{0.8}$ [24] were analyzed. The comparison of results obtained with the mentioned two qualitatively independent methods underline the importance of the effect under consideration.

The differential cross-section $d\sigma/d\Omega$ of neutrons in the 24–150 keV incident energy range scattered from H$_2$O relative to that of D$_2$O and H$_2$O–D$_2$O mixtures was reported recently in Ref. [39], claiming that the measured neutron scattering intensity ratios in the keV range exhibit no anomalous behavior. It should be stressed that this experiment in not equivalent to the standard NCS experiments which measure the partial differential cross-section. Despite this fact, however, a detailed scrutiny of [39], for single [40] and multiple [41] neutron-nucleus scattering events, demonstrated that the data analysis procedure in [39] was erroneous: We find that an anomalous reduction of H$_2$O scattering intensity (relative to that of D$_2$O) of about 20% is still present in the original experimental data.

Most recently, Stock et al. [42] presented a different approach to our "anomalous" scattering effect by investigating the hydrogen cross section in polyethylene (PE) using the direct geometry time-of-flight spectrometer MARI of ISIS with the incident energy fixed at a series of values ranging from $E_0 = 0.5$ eV to 100 eV. They claim that their measurements do not show any anomaly and thus contradict the results obtained at eVS/Vesuvio. However, the experimental method and data processing of [42] were scrutinized in great detail by Mayers et al. [43], who showed that, contrary to the claims of the authors of Ref. [42], the MARI energy resolution at eV energies is greatly inferior (i.e., by 1-2 orders of magnitude) to that on eVS/Vesuvio. This crude resolution of MARI in the aforementioned energy range invalidates the claim in [42] to have a unique demonstration that the "anomalous" intensity effect (or: intensity deficit) in the Compton profile measured on eVS/Vesuvio is an experimental artifact.

6. Collisional time window: attosecond scattering time

In the context of NCS, in which the effectuated momentum and energy transfers are large,

$$\Delta E \approx 1 - 100 \text{ eV}, \quad |q| \sim 20 - 200 \text{ Å}^{-1},$$

(18)

the Impulse Approximation (IA) is valid [18, 6] and the characteristic time scale — often termed "scattering time", $\tau_{sc}$ — of the neutron-proton scattering process is very short [5, 24],

$$\tau_{sc} \sim 100 - 1000 \text{ as}$$

(19)

(as: attosecond). These sub-femtosecond scattering times are a consequence of the large energy and momentum transfers attained with the Vesuvio instrument, Eqs. (18), and they follow from the theoretical result valid in the IA [18, 6]

$$\tau_{sc} |q| v_0 \approx 1,$$

(20)

where $v_0$ is the root-mean-square (rms) velocity of the nucleus and $\hbar q$ is the momentum transfer from the neutron to the proton. The time $\tau_{sc}$ is given by the $t$-width of the intermediate correlation function $F(q, t)$, which is connected to the dynamic structure factor $S(q, \omega)$ by Fourier transform [18]

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

(21)

Additionally, as shown by Gidopoulos within first-order time-dependent perturbation theory[46], another physical meaning for $\tau_{\eta}$ is the inverse of the energy spread $\Delta E_{\Psi_q}$ of the nuclear wavepacket $\Psi_q$ after collision, i.e. $\tau_{\eta} = \hbar / \Delta E_{\Psi_q}$. Also this physical view provides a similar scattering time (19).

To shed more light upon the issue of "relevant scattering time", one may also refer to the Margolus-Levitin theorem [44]. Let us consider the neutron-proton system during the collisional process. Obviously, the initial and final states of it are very different and thus orthogonal to each other. The theorem asserts that in takes at least a time $T_\perp \geq (\pi \hbar) / (2E_s)$ for the system to evolve from its initial to any orthogonal final state. $E_s$ is the system’s average energy minus its ground state energy. $T_\perp$ provides
a strict bound for the considered dynamical process [44]. Note that in NCS one has $E_s \approx E_0$ and thus $E_s$ is larger than, but of similar order as, the aforementioned energy spread $\Delta E$. Thus it is revealing that also this characteristic time $T_{\perp}$ is very similar to the aforementioned scattering time, i.e. $T_{\perp} \sim \tau_{sc}$.

A numerically similar estimate can be obtained with the relation

$$\Delta E \tau_0 \geq \pi \hbar/2 \quad (22)$$

by Ballentine [45]. $\tau_0$ denotes the shortest time required for the state vector $|\psi(t)\rangle$ to become orthogonal to the initial state, and $\Delta E$ is a standard measure of the statistical spread of energy in the state (e.g. the square root of the variance). Thus we have $\tau_{sc} \approx \tau_0$, which is satisfactory.

It is interesting to note that the intuitively expected “actual duration” of a neutron-proton interaction via the strong interaction may be even shorter, as a classical estimate indicates. E.g. a neutron with kinetic energy $E_0 \approx 10$ eV (and associated velocity $v_0$) will pass a distance $R_{\text{strong}} \approx 10^{-5} \text{ Å}$ (i.e. the range of the strong interaction) in a much shorter time, i.e.

$$\Delta t_{\text{strong}} = R_{\text{strong}}/v_0 \approx 10^{-19} - 10^{-20} \text{ s}.$$

However, this is not in conflict with the above estimate, for the following reason. As standard theory shows [6], $S(q, \omega)$ is peaked around the nuclear recoil energy $E_q = \hbar^2 q^2/2m$. The scattering time $\tau_{sc}$ is the inverse of the width $\Delta E$ of $S(q, \omega)$, and $S(q, \omega)$ plays the role of the probability density distribution for transferring energy $\hbar \omega$ from the neutron to the proton, when the momentum transfer is $\hbar q$. It then holds $\tau_{sc} \approx \hbar/\Delta E$. For a typical value $\Delta E \approx 10$ eV, one gets $\tau_{sc} \approx 10^{-16} - 10^{-17} \text{ s}$. In other words [46](b):

The scattering time $\tau_{sc}$ gives a statistical measure of the length of the time interval during which an elementary neutron-proton collision may occur, in the same way that the spatial extent of a particle wavefunction (or wavepacket) gives a statistical measure of the extent of the region in which the particle may be found.

### 7. Causality: open quantum systems
In the light of Relativity Theory, this has a crucial consequence: The effective scattering system — i.e., a recoiling proton and some of the adjacent electrons — must have a linear dimension smaller than

$$\Delta s_{\text{caus}} = c \cdot \Delta t_{\text{strong}} \lesssim 0.3 \text{ Å}$$

(for the above mentioned example with $E_0 = 10$ eV), where $c$ is the velocity of light, since the neutron-nucleus scattering dynamics during $\Delta t_{\text{strong}}$ cannot be causally influenced by other particles being more than $R_{\text{strong}}$ (i.e. the range of the strong force) apart from the colliding nucleus. Consequently, the scattering system must necessarily be of “microscopic” dimensions; it contains the scattering nucleus and a part of the adjacent electron density.

In other terms, causality implies that any cause of a single neutron-proton Compton scattering event can not be situated farther than $\Delta s_{\text{caus}}$ from the struck nucleus. In this context it should be noted that a typical covalent bond H-X (with X= C, O, N, etc.) is about 1 Å and therefore the atom X is beyond the causality horizon defined by $\Delta s_{\text{caus}}$, i.e. X-nucleus has no influence on the quantum dynamics of the neutron-proton Compton scattering event.

For example, the internuclear distance of the H$_2$ molecule is about 0.75 Å. Consequently, it is only one proton and a part of the electrons’ degrees-of-freedom that is/are causally participating in the collision process; it is not the whole molecule that is interacting with the neutron. This thus implies that the usual approximation of a H$_2$ molecule to be a closed system interacting with a neutron contradicts the principle of causality.

Summarizing, the systems we examine in NCS experiments must be treated — for fundamental reasons — as open quantum systems.
8. Origins of entanglement: theoretical remarks

Since conventional theory cannot explain the observed anomalies, it has been suggested to extend the existing theory to allow for a treatment of neutron scattering by quantum entangled particles [4]. However, the theoretical models existing until now appear to differ in important theoretical aspects:

(i) the relevance of quantum exchange correlations ([47] and earlier references cited therein), and
(ii) the relevance of decoherence of quantum entangled states during the scattering process [13, 14].
(iii) In this context one should also mention the suggested interpretation based on excitation of electronic degrees of freedom [46, 49, 50].

For details about the possibility of excitation of electronic degrees of freedom in a neutron-nuclear collision due to non-adiabatic effects, i.e. the breakdown in the Born-Oppenheimer (BO) approximation, see [46, 49]. Obviously, in this case it would be impossible to determine one-particle effective BO potentials, as standard NCS theory claims [6, 7]. It should also be noted that the predicted additional peaks due to these excitations have not been observed in the experimental TOF spectra of any sample investigated thus far.

Quantum exchange correlations, mentioned in (i), represent a well known effect of standard quantum mechanics, which applies to two (or more) identical particles and leads to quantum entanglement between spatial and spin degrees of freedom, as e.g. in the well-known case of ortho- and para-H₂. These two states of molecular hydrogen have different total neutron scattering cross-sections for slow (thermal and/or cold) neutrons [20]. It was suggested by Karlsson [47, 48] that exchange correlations between proton pairs – even the two H atoms belong to different molecules, e.g. in liquid HD – can be the cause of the "anomalous" scattering effect under consideration, under the condition that they fall within the coherence volume of the neutron wavepacket; see [47] and papers cited therein for details.

However, the variety of physical conditions characterizing protons in the investigated condensed systems indicate that exchange correlations cannot play the dominant role in the context of NCS. E.g., the considered effect appears, with roughly similar magnitude, even in systems where the protons occupy non-equivalent (and thus distinguishable) sites, like polystyrene [26] and amphiphiles [28]. Furthermore, NCS investigations of the heteronuclear hydrogen molecule HD (with no exchange correlations) and of H₂ (which exhibits such correlations) reveal that the considered QE-effect does appear in both samples even with the same magnitude [22].

Moreover, our current analysis of NCS-data from a single-crystal sample of Rb₃H(SO₄)₂ [work in progress] reveals a strong "anomalous" intensity effect, although the minimal distance between two H atoms in the crystal is 5.07 Å. This distance is larger than the coherence length of the neutron wavepacket (estimated to be about 2.5 Å in the applied instrumental setup, see [48]).

Alternatively, QE in condensed matter may be caused by many-body interactions. This follows from first principles of quantum mechanics. Let Ψ_A and Ψ_B be the state vectors of two systems A and B, which may be assumed either as indistinguishable (thus exhibiting exchange correlations) or not. The two systems with Hamiltonians H_A and H_B shall interact either directly or indirectly (via a third subsystem, e.g. environmental electronic charges), the corresponding Hamiltonian being V_AB. As usually assumed, let the state of the composite system ‘A and B’, in short AB, at time t = 0 be not entangled, i.e. Ψ_{AB}(0) = Ψ_A · Ψ_B. In most 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,

\[ U_{AB}(t) = e^{-i\frac{1}{\hbar}(H_A+H_B+V_{AB})t} \]  (23)

does not factorize into a product of two ‘individual’ evolution operators, say U_A and U_B, representing the dynamics of the individual (isolated) systems A and B. Consequently, the wave function Ψ_{AB}(t) at time t, Ψ_{AB}(t) = U_{AB}(t)Ψ_{AB}(0), does not factorize into a product of two wave functions (say, Ψ_A and Ψ_B), each of them representing the state of one of the two systems A and B; i.e.

\[ Ψ_{AB}(t) = U_{AB}(t)Ψ_{AB}(0) \neq Ψ_A(t) · Ψ_B(t) . \]  (24)
In other words, $\Psi_{AB}(t)$ represents an entangled state. This indicates that spatial QE is of particular interest, because the strongest interactions in the systems under investigation are given by Coulombic interactions, which depend on the spatial degrees of freedom of the participating particles (i.e., nuclei and electrons).

8.1. Theoretical models
We consider this “dynamical origin” of quantum entanglement to play the dominant role in the context of our NCS scattering experiments. It should be noted that this type of (creation, control and/or manipulation of) QE is also used and/or addressed in all experimental and theoretical fields related with quantum computing and/or quantum information [3].

The following sections of the paper deal with the theoretical treatment and interpretation of the striking attosecond effect under consideration. Various theoretical approaches are presented, which however are based on a common physical context, i.e. entanglement and its fast decoherence in open quantum systems (like molecules and/or condensed matter systems) at ambient experimental conditions.

9. Quantum Zeno effect and reduced scattering intensity
The so-called “Zeno paradox”, or quantum Zeno effect, is a well known quantum mechanical phenomenon, theoretically introduced by Misra and Sudarshan [8, 9, 10]. A short outline is as follows: Let $|\phi\rangle$ be the state vector of a quantum system at time $t = 0$ and $H$ the relevant Hamiltonian. Putting $\hbar = 1$, the time evolution of the state vector, $|\phi(t)\rangle$ is given by $\exp(-iHt)|\phi\rangle$, and the survival probability $S(t)$ for the system to “remain” in the initial state is then

$$S(t) = |\langle\phi|\exp(-iHt)|\phi\rangle|^2$$

If now one considers small times $t$, a power series expansion is possible:

$$\exp(-iHt) \approx 1 - iHt - \frac{1}{2}H^2t^2 + ...$$

so that the survival probability becomes

$$S(t) = |\langle\phi|\exp(-iHt)|\phi\rangle|^2 \approx 1 - (\Delta H)^2t^2$$

where

$$(\Delta H)^2 \equiv \langle\phi|H^2|\phi\rangle - \langle\phi|H|\phi\rangle^2.$$ 

That is, $S(t) \to 1$ for sufficiently small times $t \to 0$.

This leads to the following considerations. The time evolution leads from $|\phi(t)\rangle$ to a linear superposition of available quantum states. For example, an excited atomic state may evolve into a superposition of itself and the (stable) unexcited states, emitting a photon.) A measurement to determine whether the initial state survives can be formally described as an instantaneous projection of the superposition back onto the initial state $|\phi(t)\rangle$. (This is usually termed projective or von Neumann-type measurement.) Therefore, in this process, quantum phase relations between the involved states are destroyed, i.e. decoherence takes place. From these considerations follows that, if such projective measurements are done repeatedly at times $0, \Delta t, 2\Delta t, 3\Delta t, ... n\Delta t, ...$, then the initial quantum state $|\phi\rangle$ is hindered and evolves slower, and in the limit $\Delta t \to 0$ the time evolution becomes completely interrupted.

There exists a vast literature on the quantum Zeno effect, in which various scenarios of the mentioned projective measurements are proposed and/or analyzed. In the present NCS experimental context, of particular interest are continuous measurements which may also lead to the Zeno effect [9, 10]. Here, the time evolution of the neutron-proton collision process is continuously “observed”, or continuously “measured” by the adjacent particles (electrons and nuclei), i.e. the “environment”. 
**Figure 7.** Illustrative description of the Zeno effect. Closed system, left panel A): Fermi’s golden rule illustrated as a jump from initial ($\Psi_i$) to final $\Psi_f$ state of the impinging neutron and the recoiling atom. Closed system, right panel A): The dynamical process of the transition creates quantum phases between $\Psi_i$ and $\Psi_f$ which develop in the course of time. Open system, panel B): The system’s interaction with the environment destroys (or hinders) the dynamical development of quantum phase relations between $\Psi_i$ and $\Psi_f$, which is tantamount to a reduced transition rate (and associated scattering neutron intensity).

Moreover, the continuous observation can be incorporated in the Hamiltonian, and thus one dispenses with instantaneous quantum jumps [9]. E.g., a neutron-proton scattering process from H$_2$ becomes continuously observed by the "environment", i.e. the two electrons and the second proton which interact with the struck proton during the whole collisional process. In qualitative terms, this leads to the aforementioned destruction of quantum phases and the associated hindered transition to the final scattering state(s). Obviously, this is tantamount with a reduced neutron-proton scattering intensity and/or cross-section.

This physical picture also shows that, in the considered physical context, the “time window” $\tau_{sc}$ of scattering must be sufficiently short, of the order of the fast evolving quantum phase relations (or: coherences) of the collisional system "neutron plus proton plus environment".

The cartoon of figure 7 may help to illustrate this process. The "environment" of the neutron-proton collisional system is represented as a "cloud" interacting with the system.

**10. Schulman–Gaveau effect and reduced scattering intensity**

Further insight into the quantum-phase destruction mechanism described above can be provided with the aid of the Schulman–Gaveau effect [11, 12]. This work permits a first principles interpretation of, and provides new physical insight into, the reduced scattering intensity phenomenon [4, 5], which may be
outlined as follows.

A probe particle (neutron, with free Hamiltonian $H_n$) of sufficiently high energy impinges on a molecule (condensed system, etc., with Hamiltonian $H_{Sys}$). We shall consider scattering from a proton $p$ (with free Hamiltonian $H_p$) of this molecule (system) and thus write its Hamiltonian as $H_{Sys} = H_p + H_{Rest}$. Let the neutron-proton interaction of this scattering be $gV_{pn}$, $g$ being a c-number representing the coupling strength. The full Hamiltonian is then

$$H = H_n + H_p + H_{Rest} + gV_{pn}.$$  \hspace{1cm} (25)

[In the notation of [11] we have: $H_A = H_n$ and $H_B = H_p + H_{Rest}$. To make contact with [11], the coupling Hamiltonian can be formally considered to be of the form $V_{pn} = V_n \otimes V_p$, with $V_n$ and $V_p$ some operators of the neutron and the proton, respectively. In specific cases, the coupling strength $g$ may be explicitly time-dependent. To make contact with [12], Sect. V, Eqs. (70,74), the coupling Hamiltonian may be a function of the $p-n$ distance $|r_p - r_n|$, i.e. $V_{pn} = W(|r_p - r_n|); r_X$: position of particle $X$.]

As discussed above, the scattering (interaction, collisional) time, $\tau_{sc}$, of the neutron-proton interaction in the experiments [4, 5] lies in the sub-femtosecond time scale. This supports the present physical assumption (or, in other words, the working hypothesis) that the effect of Schulman and Gaveau [11] can be applicable here.

The following remarks may be helpful. The standard (i.e. well established and widely applied) neutron scattering theory, which is based on the first Born approximation, is known as the van Hove theory; see e.g. [20]. This is the only theory applied to thermal and cold neutron scattering, which are "slow" scattering processes as compared to NCS. As first-order perturbation theory is used, here there is no notion at all of entanglement between the neutron and the scatterer (proton, system) after the collision. Thus this general theory corresponds to the case of a factorizing final-state density operator, i.e.

$$\rho(t) = \rho_n(t) \otimes \rho_{Sys}(t) \quad \text{for} \quad t \geq \tau_{sc}. \hspace{1cm} (26)$$

Let us assume here that in the sub-femtosecond (NCS) experiments [4, 5] the above assumed factorization is physically incorrect. This is due to the fact that even the electronic rearrangements and/or relaxation, the fastest relaxation processes in condensed and/or molecular matter, lie in the sub-femtosecond timescale too. That is, we assume here that the exact total density operator $\rho(t)$ is entangled and denote the difference as

$$\Delta \rho(t) \equiv \rho_n(t) \otimes \rho_{Sys}(t) - \rho(t) \hspace{1cm} (27)$$

where the partial trace $\rho_n(t) = Tr'\rho(t)$ is introduced, with $Tr'$ the trace over all other variables up to $n$; and correspondingly $\rho_p(t) = Tr'\rho(t)$.

It is shown in [11, 12] that

$$\Delta H = Tr[\Delta \rho(t)H] = Tr[\Delta \rho(t)(gV_{pn})] > 0, \hspace{1cm} (28)$$

under rather general conditions for the interaction Hamiltonian. That is, if one would evaluate the energy using the exact density operator $\rho(t)$ it would be unchanged from its time-0 value. However, if $\rho(t)$ is replaced by the uncorrelated density operator $\rho_n(t) \otimes \rho_{Sys}(t)$, then the energy could change, and for short times it in fact increases. This result is shown [12] to be more general than was quoted in the original paper [11]. These results demonstrate that operations that seem quite innocent, in particular the erasure of certain entanglement, can have significant effects.

Proposed connection with NCS experiments. — This result is the crucial point in the following comparison between results from conventional "slow" neutron scattering experiments (i.e. thermal and cold neutron scattering) and from the new "fast" NCS experiments. The "anomalous" NCS effect can be understood by noting that the scattering intensity $I_H$ from protons (and the associated cross-section $\sigma_H$)
may be assumed to be a monotonous function of the expectation value of the interaction Hamiltonian squared:

$$I_H \sim |\langle g \hat{V}_{pn} \rangle|^2,$$

or the time average of this quantity over the duration of the scattering process, for time-dependent $g(t)$ and/or time-dependent density operator.

In the conventional "slow" scattering experiments the density operator is assumed to be not entangled, $\rho_n(t) \otimes \rho_{Sys}(t)$. This should be a good approximation, because decoherence is an extremely fast process in strongly interacting systems (like condensed matter or molecules).

In the "fast" NCS experiments we may assume that this decoherence mechanism does not destroy the quantum entanglement (QE) completely, and therefore let us denote the density operator by $\rho_{QE}$.

Thus using the Schulman–Gaveau result we conclude that

$$\text{Tr}[\rho_n(t) \otimes \rho_{Sys}(t) g \hat{V}_{pn}] > \text{Tr}[\rho_{QE} g \hat{V}_{pn}]$$

and equivalently

$$I_H(\text{slow scattering}) > I_H(\text{NCS}).$$

This difference is a natural consequence of the decoherence, as revealed by the Schulman–Gaveau effect, and does not depend on any specific property of the interaction Hamiltonian. This point was already noted in [5], in which results of neutron and electron scattering from protons were compared. Obviously, if one denies the existence of decoherence in the NCS physical frame [51], the last inequality becomes an equality and, therefore, the "anomalous" scattering effect should "disappear". However, this difference can be considerable; cf. the novel experimental results presented in figure 6.

11. Introducing entanglement and decoherence into conventional formalism

In this section, we "extend" the standard expression Eq. (1) of the partial differential cross-section of conventional scattering theory [19, 20, 6, 7] by introducing ultrafast entanglement and decoherence of final and initial states of the scattering system. Recall that decoherence is an unknown phenomenon in the frame of conventional theory. Here we follow the presentation of [13].

To simplify notations, let us temporarily assume that the condensed system consists of $N$ atoms of the same kind only, say, hydrogen atoms. The starting point is the expression for the partial differential cross section, Eq. (1), which we shall consider here in more detail.

The number of neutrons ($I_{nc}$) with fixed initial energy $E_0$, scattered per second into a small solid angle $d\Omega$ (in some given direction) with final energy between $E_1$ and $E_1 + dE_1$ (and thus $dE_1 = d\omega$),

$$I_{nc}(E_1, \Omega) \propto \frac{d^2 \sigma}{d\Omega dE_1},$$

i.e., it is proportional to the partial differential cross section [20]

$$\frac{d^2 \sigma}{d\Omega dE_1} = \frac{k_1}{k_0} \sum_{\nu' \nu} W_{\nu'} \left| \sum_j b_j \langle \nu' | \exp (i\mathbf{q} \cdot \mathbf{r}_j) | \nu \rangle \right|^2 \delta(E_{\nu'} - E_{\nu} + E_0 - E_1).$$

$b_j$ is the bound scattering length of atom $j$ ($= 1, ..., N$). $\nu$ and $\nu'$ refer to the initial and final stationary states of the whole $N$-body scattering system (with energies $E_{\nu}$ and $E_{\nu'}$, respectively), corresponding to the transition $|\nu\rangle \rightarrow |\nu'\rangle$.

Recall that this formula is derived from Fermi’s golden rule, after introducing the additional delta-function, which represents strict energy conservation [20, 21]. (See also additional remarks in the Discussion section below.)
The sum $\sum_j$ contains $N$ terms. So the square of its absolute value, $|\sum_j|^2$, is the sum of $N^2$ terms of which a typical member is

$$M_{j,j'}(\nu\nu') = b_j b_{j'} \langle \nu' | \exp (i \mathbf{q} \cdot \mathbf{r}_j) | \nu \rangle \exp (-i \mathbf{q} \cdot \mathbf{r}_{j'}) | \nu' \rangle |
u\rangle$$

$$= b_j b_{j'} \text{Tr} \left[ \rho_{\nu'} \{ \exp (i \mathbf{q} \cdot \mathbf{r}_j) | \nu \rangle \exp (-i \mathbf{q} \cdot \mathbf{r}_{j'}) \rangle \} \right]$$

$$= b_j b_{j'} \text{Tr} \left[ \rho_{\nu'} \exp (i \mathbf{q} \cdot \mathbf{r}_j) \rho_{\nu} \exp (-i \mathbf{q} \cdot \mathbf{r}_{j'}) \right]$$

$$= b_j b_{j'} \text{Tr} \left[ \rho_{\nu'} V_{j} \rho_{\nu} V_{j'} \right]$$

(34)

where $V_j \equiv \exp (i \mathbf{q} \cdot \mathbf{r}_j)$, $V_j^\dagger \equiv \exp (-i \mathbf{q} \cdot \mathbf{r}_j)$ and $\rho_{\nu'} \equiv |\nu'\rangle \langle \nu'|$, $\rho_{\nu} \equiv |\nu\rangle \langle \nu|$. $\text{Tr} [...]$ denotes the trace operation.

In the physical context of NCS one has $q \gg 2\pi/d$, where $d$ is the nearest-neighbor distance of two scattering nuclei. As a consequence, the spatial scale of the scattering event, represented by $1/q$ (where $q = |\mathbf{q}|$), is too small for one to detect interference effects due to scattering from pairs of different nuclei, $j \neq j'$. This is an important point, because sometimes one confuses the limiting case of large momentum transfers — which, unfortunately, is commonly called "incoherent approximation" [6, 18]— with the well-known "incoherent scattering component" (which constitutes only one part of the total scattering intensity) appearing in scattering of thermal [20] or cold neutrons.

Thus all terms with $j \neq j'$ do not contribute to the measured scattering intensity, and therefore one obtains from Eq. (34)

$$M_{jj'(\nu\nu')} = b_j^2 \text{Tr} \left[ \rho_{\nu'} V_j \rho_{\nu} V_j^\dagger \right] \quad \text{or} \quad M_{(\nu\nu')} = b^2 \text{Tr} \left[ \rho_{\nu'} V \rho_{\nu} V^\dagger \right].$$

(35)

The index $j$ may be dropped, since all particles are by assumption equivalent. Since the quantity $V \rho_{\nu} V^\dagger = |\nu\rangle \langle \nu| V \,angle \langle V|$ is a projector (thus having eigenvalues 0 or 1) and $\rho_{\nu'}$ is a density operator (thus having non-negative eigenvalues), the product $(\rho_{\nu'} V \rho_{\nu} V^\dagger)$ must be hermitian and positive semidefinite. Consequently, $\text{Tr} \left[ \rho_{\nu'} V \rho_{\nu} V^\dagger \right] \geq 0$ and $M_{(\nu\nu')} \geq 0$. Thus, in the conventional theoretical expression given by Eq. (33), and for sufficiently large momentum transfers (so that $j = j'$), occur only non-negative terms.

The initial and final states, $\nu$ and $\nu'$, refer to the whole (closed) N-body system and are assumed to be eigenstates of the system’s complete Hamiltonian $H_\tau$ [20]. Each individual NCS process, however, is expected to involve only a much smaller number of “relevant” degrees of freedom, i.e. those belonging to the relevant microscopic open quantum system; see above. The latter can be described by reduced density operators $\tilde{\rho}_{\nu'}$ and $\tilde{\rho}_{\nu}$ obtained from $\rho_{\nu'}$ and $\rho_{\nu}$ through partial trace $\text{Tr}_{\text{env}}$ over the huge number of the remaining degrees of freedom of the N-body system (the “environment”):

$$\tilde{\rho}_{\nu'} = \text{Tr}_{\text{env}} [\rho_{\nu'}], \quad \tilde{\rho}_{\nu} = \text{Tr}_{\text{env}} [\rho_{\nu}].$$

(36)

Thus one easily obtains the relation

$$0 \leq M_{(\nu\nu')} = b^2 \text{Tr} \left[ \rho_{\nu'} V \rho_{\nu} V^\dagger \right] = b^2 \text{Tr}_{\text{rel}} \left[ \tilde{\rho}_{\nu'} V \tilde{\rho}_{\nu} V^\dagger \right],$$

(37)

where $\text{Tr}_{\text{rel}}$ denotes the trace over the degrees of freedom of the relevant system.

In the following we investigate possible implications of QE and its decoherence for the scattering process. The physical context is associated with the relation

$$\tau_{\text{dec}} \sim \tau_{\text{sc}}.$$ (38)

Let $\{|\xi\rangle\}$ be the preferred representation [52] being selected by the decoherence accompanying the dynamics of the relevant scattering system. Using as an explicit example a Lindblad-type decoherence model [2], we have

$$\langle \xi | \tilde{\rho}_{\nu'}(t) | \xi' \rangle = \langle \xi | \tilde{\rho}_{\nu'}(0) | \xi' \rangle e^{-\lambda |\xi - \xi'|^2 t}. $$

(39)
In general, the positive constant \( \Lambda \) should strongly depend on the state \( \nu' \), but we drop a related subscript for simplicity of notations. In order to simplify the following derivations, we may temporarily assume \( \tilde{\rho}_\nu \) to be time-independent.

Performing now the trace in Eq. (37) with respect to the basis \( \{ |\xi\rangle \} \) and noting the closure relation \( \int d\xi' |\xi'\rangle \langle \xi'| = 1 \), Eq. (37) yields

\[
M_{(\nu\nu')}(t) = b^2 \int d\xi d\xi' \langle \xi | \tilde{\rho}_\nu(t) | \xi' \rangle \langle \xi' | V \tilde{\rho}_\nu V^\dagger | \xi \rangle = b^2 \int d\xi d\xi' \langle \xi | \tilde{\rho}_\nu(0) | \xi' \rangle \langle \xi' | V \tilde{\rho}_\nu V^\dagger | \xi \rangle e^{-\Lambda |\xi-\xi'|^2 t} .
\]  

This quantity contains a \( t \)-dependence due to decoherence, which concerns the non-diagonal elements of the reduced (or: relevant) density operator \( \tilde{\rho}_\nu(t) \). Clearly, for \( \Lambda = 0 \) this equation is identical with Eq. (37), that is: \( M_{(\nu\nu')}(t) = M_{(\nu\nu')}(0) = M_{(\nu\nu')}(t) \), for all \( t \).

Since time-resolved details of this very fast \( t \)-dependence are not accessible experimentally, comparison with measurements should be made after taking the time average over the duration of the scattering process, given by the scattering time \( \tau_{sc} \). Thus, the associated experimentally relevant quantity is

\[
\overline{M}_{(\nu\nu')} = \frac{1}{\tau_{sc}} \int_0^{\tau_{sc}} dt \, M_{(\nu\nu')}(t) .
\]  

As expected, the last formula includes also the cases in which decoherence is absent, i.e. \( \Lambda = 0 \), since then it trivially holds \( \overline{M}_{(\nu\nu')} = M_{(\nu\nu')}(0) \). But for the general case \( \Lambda > 0 \), all (absolute values of the) contributions to the rhs of Eq. (40) are reduced due to the exponentials \( e^{-\Lambda |\xi-\xi'|^2 t} \).

Based on physical considerations, one can conclude that this causes a decrease of the quantity \( \overline{M}_{\nu\nu'} \). For this purpose, note first that the "diagonal" terms with \( \xi = \xi' \) in Eq. (40) are \( t \)-independent and positive, as easily seen:

\[
\langle \xi | \tilde{\rho}_\nu(0) | \xi \rangle \langle \xi | V \tilde{\rho}_\nu V^\dagger | \xi \rangle = \langle \xi | \tilde{\rho}_\nu(0) | \xi \rangle \langle \xi V^\dagger | \tilde{\rho}_\nu | V^\dagger \xi \rangle \geq 0 .
\]  

By continuity, all associated terms with \( \xi \approx \xi' \) should be positive, too, and they contribute most significantly to the process. This point has been emphasized by Mensky [2](b). Thus, we finally obtain the desired result \( \overline{M}_{(\nu\nu')} < M_{(\nu\nu')}(0) \), which is tantamount to a decrease of scattering intensity.

Now we can drop the preceding assumption of \( \tilde{\rho}_\nu \) being decoherence-free. Then the above procedure of Eqs. (39)–(42) can be straightforwardly extended. The localization rates \( \Lambda \) will then depend on both states \( \nu \) and \( \nu' \). The main conclusion remains unchanged.

Moreover, investigations of the limiting cases (i) \( \tau_{dec} \gg \tau_{sc} \), and (ii) \( \tau_{dec} \ll \tau_{sc} \) (applicable to all other neutron scattering techniques than NCS), showed that the associated theoretical results are completely in line with the predictions of conventional theory [19, 20, 6, 7] — simply because in these cases the decoherence dynamics becomes irrelevant.

In summary, it has been shown that the exponentially decaying factors due to decoherence (in a microscopic open quantum system) may lead to a reduction of the time averages \( \overline{M}_{(\nu\nu')} \) and of the scattering intensity, Eq. (32), thus providing a qualitative theoretical interpretation of the new NCS effect under consideration.

12. A "first principles" model of fast decoherence in scattering dynamics

Here we present a simple theoretical model [14] for the theoretical interpretation of the considered NCS phenomenon, which is more explicit that the aforementioned qualitative considerations. The basic physical idea remains however the same, i.e. the crucial role that quantum entanglement and ultrafast decoherence play in the physical context under consideration; for more details see [14].
Let us consider neutron scattering from a system consisting of $N$ particles with the same scattering length $b$, and the N-body Hamiltonian $H_{\text{total}} = H_0 + V$ with the interaction

$$V(r) = \lambda n(r), \quad \lambda = \frac{2\pi \hbar^2}{m} b. \quad \text{(43)}$$

$m$ is the neutron mass, $n(r)$ is the particle density operator

$$n(r) = \frac{1}{V} \sum_{j=1}^{N} \delta(r - \mathbf{R}_j) \quad \text{(44)}$$

where $V$ is the volume, and $\mathbf{R}_j$ is the spatial position of the $j$-th particle; cf. the textbook [20].

In the interaction picture, the Schrödinger equation is now (setting for simplicity $\hbar = 1$) $i\partial_t \Psi = \lambda n(r,t) \Psi$, with the perturbative solution

$$\Psi(t) = \Psi(0) - i\lambda \int_{0}^{t} n(r,t') dt' \Psi(0). \quad \text{(45)}$$

We write the transition probability $W(t)$ between initial states $\psi_i$ (with probability $P_i$ that the scattering system is in the state $\psi_i$) and final states $\psi_f$ of the scattering system to be given by

$$W(t) = \sum_{i,f} | \langle \psi_f | \lambda \int_{0}^{t} n(r,t') dt' | \psi_i \rangle |^2 P_i. \quad \text{(46)}$$

It should be noted that $\psi_i$ and $\psi_f$ are eigenstates of the N-body Hamiltonian $H_0$ omitting the probe system [20]. This allows us to write the transition probability in the form

$$W(t) = \lambda^2 \int_{0}^{t} dt' \int_{0}^{t} dt'' \sum_{f} \langle \psi_f | n(\mathbf{q},t') \rho n(\mathbf{q},t'') | \psi_f \rangle, \quad \text{(47)}$$

with $\rho = \sum_{i} | \psi_i \rangle P_i \langle \psi_i |$, where we have noted that $n^\dagger(\mathbf{r},t) = n(\mathbf{r},t)$.

To describe an actual scattering experiment from condensed matter, one takes an appropriate average over all these states [20], as done in Eq. (46).

Furthermore, the initial ($\mathbf{k}_0$) and final ($\mathbf{k}_1$) momenta of an impinging probe particle (neutron) may be assumed to be well defined [20]. Introducing the momentum transfer $\mathbf{q} = \mathbf{k}_0 - \mathbf{k}_1$ from the probe particle to the scattering system, the Fourier transform of the particle density reads

$$n(r,t) = \frac{1}{(2\pi)^3} \int d\mathbf{q} n(\mathbf{q},t) \exp(i \mathbf{q} \cdot \mathbf{r}) \quad \text{(48)}$$

where, in the case of neutron scattering, cf. Eq.(44),

$$n(\mathbf{q},t) = \sum_{j} \exp[-i\mathbf{q} \cdot \mathbf{R}_j(t)]. \quad \text{(49)}$$

Since $n(r,t)$ is Hermitian, we have $n^\dagger(\mathbf{q},t) = n(-\mathbf{q},t)$ and one obtains from Eq. (46)

$$W(t) = \lambda^2 \int_{0}^{t} dt' \int_{0}^{t} dt'' \sum_{f} \langle \psi_f | n(\mathbf{q},t') \rho n(-\mathbf{q},t'') | \psi_f \rangle. \quad \text{(50)}$$
The sum over \( \psi_f \) runs over all possible eigenstates of \( H_0 \) which constitute a complete set, i.e. 
\[ \sum_f \langle \psi_f | n(q, t') \rho n(-q, t'') | \psi_f \rangle = \text{Tr} \left[ n(q, t') \rho n(-q, t'') \right] \]  
(51)

where \( \text{Tr}[...] \) denotes the trace operation. As done in standard theory, in Eq. (51) one sums over all final states, keeping the initial state \( \psi_i \) fixed, and then averages over all \( \psi_i \) (see e.g. [20], p. 19). The right-hand-side of Eq. (51) contains the density operator \( \rho \) of the system before collision. Also, if the integration in Eq. (50) is extended over all times (i.e., \( t \to \infty \)), there ensues over-all energy conservation. This reproduces the well known result of standard neutron scattering theory.

Here, however, it is important to retain the finite duration of the scattering time. This introduces an additional freedom into the theory, because we may be able to observe the influence of the decoherence on the scattering yield. The result will be expressed in terms of the correlation function

\[ C(q, \tau) = \text{Tr}[n(q, t) \rho n(-q, t + \tau)] = \text{Tr}[n(q, 0) \rho n(-q, \tau)], \]  
(52)

where we have utilized the fact that the scattering system is stationary.

By introducing a measurement time (the so-called scattering time) \( \tau_{sc} \), representing the time interval within the scattering process may happen, we find

\[ W(\tau_{sc}) = \lambda^2 \int_0^{\tau_{sc}} dt' \int_0^{\tau_{sc}} dt'' C(q, t'' - t') = \lambda^2 \int_0^{\tau_{sc}} dt' \int_0^{t'} d\eta [C(q, \eta) + C(q, -\eta)]. \]  
(53)

Here we use the stationarity of the correlation function [20]. If we assume this function to be real, and that \( C(q, \eta) \approx 0 \) for \( \eta \gtrsim \tau_{sc} \), we obtain the result

\[ W(\tau_{sc}) \approx 2\lambda^2 \tau_{sc} \int_0^{\tau_{sc}} d\eta C(q, \eta). \]  
(54)

Now we can introduce the transition rate, \( \dot{W} \) say, which is defined as

\[ \dot{W} \equiv \frac{W(\tau_{sc})}{\tau_{sc}} = 2\lambda^2 \int_0^{\tau_{sc}} d\eta C(q, \eta). \]  
(55)

Here the correlation function is analogous to the so-called intermediate function of neutron scattering theory [20]. This result for the scattering yield is analogous to that of standard theory.

We now introduce a set of preferred coordinates \{ \( \xi \) \}, cf. [2, 52]. These represent the relevant degrees of freedom coupled to the neutron probe, thus representing and/or defining an open quantum system. The density matrix needed in (52) is then the reduced one in the space spanned by these states, and it is obtained by tracing out the (huge number of the) remaining degrees of freedom belonging to the “environment”. To simplify notations, we denote this reduced density matrix by \( \rho \) too.

In the subspace spanned by the preferred coordinates (also termed “pointer basis”), we may assume the relevant density matrix to obey a Lindblad-type equation of the form [2]

\[ \partial_t \rho = -i [H, \rho] + \mathcal{R} \rho \equiv \mathcal{L} \rho \]  
(56)

with the formal solution: \( \rho(t) = e^{\mathcal{L}t} \rho(0) \). Let us look at a time-dependent expectation value

\[ \langle A(t) \rangle \equiv \text{Tr}(\rho(t)A) = \text{Tr} \left( e^{\mathcal{L}t} \rho(0)A \right) = \text{Tr} \left( \rho(0) e^{\mathcal{L}^\dagger t} A \right), \]  
(57)
where we define $\mathcal{L}^\dagger$ by setting $Tr ((\mathcal{L}X)Y) = Tr (X (\mathcal{L}^\dagger Y))$. Thus we obtain a Lindblad time evolution for the operators too by writing

$$\partial_t A(t) = \mathcal{L}^\dagger A(t).$$

(58)

This form was actually the original Lindblad result as introduced in the quantum algebraic formulation of the theory of semigroups. Note that this assumes that $\mathcal{L}$ does not depend on time.

Now we find that we may use this formalism to calculate correlation functions like the one in (52). We write

$$\langle A(t)B \rangle = Tr \left[ \rho(0) \left(e^{\mathcal{L}^\dagger t} A \right) B \right] = Tr \left[ A e^{\mathcal{L}t} (B \rho(0)) \right] \equiv Tr (A \rho_B(t)),$$

(59)

where $\rho_B(t)$, as defined in Eqs. (59), obeys the equation

$$\partial_t \rho_B(t) = \mathcal{L} \rho_B(t)$$

(60)

and the initial condition $\rho_B(0) = B \rho(0)$. Thus, except for the initial condition, we have to solve the same equation of motion as for the density matrix (56).

We here assume a simple Lindblad-type ansatz for the master equation in the relevant subspace. In order to show the effect of dephasing, we simply assume one Lindblad variable $X$; in the real system we would have a multitude of such variables. We set

$$\partial_t \rho = -i [H, \rho] - K [X, [X, \rho]] = \mathcal{L} \rho,$$

(61)

where $K > 0$ and $H$ is the reduced (or relevant Hamiltonian) of a microscopic or mesoscopic scattering system and the double commutator term describes dephasing (and/or decoherence). For simplicity of the further calculations, we here assume that we can take the preferred coordinates to commute with the total Hamiltonian

$$H | \xi \rangle = \mathcal{E}_\xi | \xi \rangle, \quad X | \xi \rangle = \xi | \xi \rangle.$$

(62)

This time evolution is now introduced into the correlation function $C(q, \tau)$, Eq. (52). A short straightforward calculation (see Ref. [14] for full details) yields for the transition rate the result

$$W = 2\lambda^2 \int_{0}^{\tau_{dec}} \sum_{\xi, \xi'} \exp \left[ -i \left( \mathcal{E}_{\xi'} - \mathcal{E}_\xi \right) \tau \right] \exp \left[ -K \left( \xi' - \xi \right)^2 \tau \right] \times$$

$$\times \langle \xi | n(-q, 0) | \xi' \rangle \langle \xi' | n(q, 0) \rho(0) | \xi \rangle d\tau.$$

(63)

Obviously, the decoherence-free limit of this result, i.e. with $K = 0$, corresponds to the conventional result of scattering theory. The oscillating factors $\exp \left[ -i \left( \mathcal{E}_{\xi'} - \mathcal{E}_\xi \right) \tau \right]$ are characteristic for the “unitary-type” dynamics caused by the commutator part $-i [H, \rho]$ of the master equation (61) for the reduced (or relevant) density matrix $\rho$. These factors have the absolute value 1 and are well known from standard theory [20]. On the other hand, decoherence (with a characteristic time $\tau_{dec}$) is present in the case $\tau_{dec} = K^{-1} \sim \tau_{res}$, leading to the additional restrictive factors $\exp(-K \left( \xi' - \xi \right)^2 \tau) \leq 1$, which can be seen to cause a decrease of the transition rate and thus of the associated cross-section. This can be illustrated in physical terms as follows.

Let us first assume that the reduced density operator $\rho(0)$ can be chosen to be diagonal in the preferred $\xi-$representation (which corresponds to the usual random phase approximation at $t = 0$). Then each term of Eq. (63) contains the factor

$$\langle \xi | n(-q, 0) | \xi' \rangle \langle \xi' | n(q, 0) \rho(0) | \xi \rangle = |\langle \xi | n(-q, 0) | \xi' \rangle|^2 |\langle \xi | \rho(0) | \xi \rangle| \geq 0.$$

(64)

If the assumption of diagonal form for $\rho(0)$ would be considered as being "too strong", one may note the following: The decoherence factors $\exp(-K \left( \xi' - \xi \right)^2 \tau)$ imply that only terms with $\xi \approx \xi'$
contribute significantly to the transition rate. Thus we may conclude that, by continuity, all associated terms with \( \xi \approx \xi' \) in Eq. (63) should be positive, too. The further terms with \( \xi \) being much different from \( \xi' \) can be positive or negative. But they may be approximately neglected, since they decay very fast and thus contribute less significantly to \( \dot{W} \); cf. [2](b).

The main conclusion from the preceding considerations is that the time average in Eq. (63) always decreases the value of \( W \equiv W(\tau_{sc})/\tau_{sc} \), due to the presence of the exponential factors \( \exp(-K(\xi'-\xi)^2 \tau) \leq 1 \). In other words, the effect of decoherence (and/or dephasing) during \( \tau_{sc} \) plays a crucial role and may lead to an "anomalous" decrease of the transition rate and the associated scattering intensity.

Interestingly, in the limit of "very slow" decoherence, \( K \to 0 \), this "anomaly" disappears, i.e. the scattering results are predicted to agree with conventional theoretical expectations.

In the opposite limiting case of "very fast" decoherence, \( K \to \infty \), one immediately sees that only the "diagonal" terms with \( \xi = \xi' \) survive in Eq. (63) — which is due to the action of the factors \( \exp(-K(\xi'-\xi)^2 \tau) \leq 1 \). At the same time, for all oscillating factors holds: \( \exp[-i(\xi_2' - \xi_2) \tau] \to 1 \). Consequently, Eq. (63) tends to the standard expression Eq. (55) in the limit of short scattering times. Also this result is according to conventional expectations.

Additionally, the above theoretical remarks and derivations clearly show that Fermis' golden rule (GR) cannot be strictly valid for the quantum dynamical description of a microscopic open quantum system in the ultrashort timescale of NCS; see also section 13(B-C) below. E.g., for an open quantum system, the assumed stationary states for the derivation of GR are totally fictitious. Confusion related with this fact can lead one to conclude that the decoherence phenomenon is irrelevant and/or nonexistent in the physical context of NCS [51].

13. Additional remarks and discussion
(A) Common in all the theoretical considerations and models of the theoretical sections 9–12 is the physical insight that short-lived quantum entanglement and decoherence between a struck quantum system (say, proton) and its adjacent particles play a central role in the dynamical process of scattering. The phenomenon of decoherence is unknown in conventional neutron scattering theory [20, 21, 6, 7]. The discussion of the NCS physical parameters showed that the characteristic timescale of NCS lies in the attosecond range. A similar conclusion holds also for electron-atom Compton scattering.

(B) Considering scattering from single molecules in the gas phase, the following question may arise. E.g., for the case of H\(_2\) and D\(_2\) scattering experiments, the "environment" consists of only 3 other particles (the second proton/deuteron and two electrons). Such a "mini" environment seems to offer the unique chance to investigate the possibility to go beyond the aforementioned decoherence-type theoretical approaches. That is, instead of treating the impinging neutron and the scattering nucleus (proton) as an open system, one could as well consider the closed system "neutron plus H\(_2\)" as a few body system. Scattering of one electron or neutron from a fully entangled four body system — that is, considering a five body closed system — seems to be in reach of present days theory. Consequently, the experimental results could potentially be explained by a five body theory (for the H\(_2\) case) without talking about open quantum systems.

Although a definitive answer to this question is presently not known, we believe that the arguments presented in section 7 deny the suggested possibility. Namely, the ultrafast attosecond neutron-proton collisional time was shown to imply that the second proton of H\(_2\) has no causal influence on the dynamics of the neutron-proton collisional system.

(C) General neutron scattering theory (cf. [20, 21]), as well as the particular NCS theory [6, 7] are based on the quantum theory of systems with well defined Hamiltonians and the associated unitary time evolutions (in the frame of first-order perturbation theory). This is the case for closed quantum systems. In this theoretical framework, of course, the phenomenon of decoherence plays no role "by definition". Therefore it should be stressed that the physical conditions of the real NCS process are consistent with the theoretical frame of scattering from open quantum systems. E.g. for NCS from molecular hydrogen
(gas phase) the relevant quantum system consists of an impinging neutron and one proton of \( \text{H}_2 \) only — as the H-peak position in the TOF spectrum proves beyond doubt — probably being “dressed” with some electronic degrees of freedom of the electrons of the chemical bond. The remaining adjacent particles and/or degrees of freedom (i.e., the second proton and a part of the two electrons of \( \text{H}_2 \)) represent the “environment”. The latter strongly interacts with the scattering system (dressed proton) through electromagnetic forces, thus causing ultrafast decoherence. The conceptual connection with the physical fields of “quantum optics” and/or “quantum computing” should be obvious.

In this context it cannot be overemphasized that in the general theory \([20, 21]\), as well as in the more specific standard NCS theory \([6, 7]\), the time evolution is assumed to be unitary. Let us discuss this in some detail. The transition from Eq. (33),

\[
\frac{d^2 \sigma}{dk_1 dE_1} = \frac{k_1}{k_0} \sum_{\nu \nu'} W_\nu \sum_j b_j \langle \nu | \exp(iq \cdot r_j) | \nu \rangle \right| \sum_{j} \left| \langle \nu | \exp(iq \cdot r_j) | \nu \rangle \right|^2 \delta(E_\nu - E_{\nu'} + E_0 - E_1), \tag{65}
\]

— which represents Fermi’s golden rule \([20, 21]\) with the additional inclusion of the shown delta-function (in order to guarantee strict energy conservation) — to

\[
\frac{d^2 \sigma}{d\omega d\Omega} = b^2 \frac{k_1}{k_0} \frac{1}{2\pi} \int_{-\infty}^{\infty} dt \exp((-i\omega t) \sum_{j,k} \langle \exp(-i\mathbf{q} \cdot \mathbf{r}_j(0)) \exp(-i\mathbf{q} \cdot \mathbf{r}_k(t)) \rangle \right|, \tag{66}
\]

— that is, Eqs. (1) and (4) — is effectuated by the spectral decomposition of the delta function

\[
\delta(E_\nu - E_{\nu'} + E_0 - E_1) = \frac{1}{2\pi \hbar} \int_{-\infty}^{\infty} dt \exp[i(E_\nu - E_{\nu'} + E_0 - E_1)/\hbar] \exp(-i\omega t), \tag{67}
\]

and the introduction of the unitary time evolution operator \( U(t) \) through the relation

\[
U(t) |\nu\rangle = \exp(-iHc t) |\nu\rangle = \exp(-iE_{\nu} t) |\nu\rangle \tag{68}
\]

because \(|\nu\rangle \) and \(|\nu'\rangle \) are assumed to be eigenfunctions of the complete Hamiltonian \( H_c \) of the \( N \)-body system. Furthermore, application of the (assumed validity of the) closure relation over the final states

\[
\sum_{\nu'} |\nu'\rangle \langle \nu'| = \hat{1}
\]

leads to the introduction of the Heisenberg representation of the position operators

\[
\mathbf{r}_j(t) = U^\dagger(t) \mathbf{r}_j U(t)
\]

in Eq. (4), i.e.

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

(see e.g. the textbooks \([20, 21]\) for a detailed derivation). These results are due to the assumption that the closed \( N \)-body scattering system is considered to have a well defined Hamiltonian, \( H_c \), which determines the complete quantum dynamics with the aid of \( U(t) \).

Of course, this is not true for open quantum systems which must exhibit non-unitary time evolutions and decoherence \([2]\). In the context of NCS, rejection of these facts leads to confusion, in which Fermi’s golden rule is claimed to be strictly valid and/or non-stationary states of the scattering system are claimed to be totally fictitious.
(D) Concerning the time range of validity of first-order time-dependent perturbation theory, and thus also of validity of Fermi’s golden rule, one should keep in mind that one necessary condition by the derivation is the assumed ”long time”,

\[ t \gg \frac{\hbar}{\Delta E} \]

with \( \Delta E \) being the energy difference of final and initial states; see textbooks of quantum mechanics. The same holds for the delta-function in Eq. (65), which is justified only if \( t \gg \frac{\hbar}{\Delta E} \). This condition, however, is in conflict with the estimated characteristic ultrashort collisional time of NCS; see Section 6.

(E) As discussed in Section 7 — and illustrated with the example of NCS from a single \( H_2 \) molecule — the physical reason which requires the scattering particle (e.g. a proton) to be an open quantum system is the causality principle and the finite velocity (\( \leq c \)) of any physical signal. Thus, in the physical context of NCS, this has a far-reaching consequence: 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 with ”missing knowledge” of an ”exact state” \( \Psi \), but rather the opposite holds true: \( \Psi \) ought to be a ”bad” approximation, because it is not consistent with causality (in the NCS case under consideration).

This conclusion appears to be consistent with a theoretical result by Beretta [53], which should be mentioned here. The theoretical work [53] 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 nonequilibrium 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 [53], p. 4.)

Furthermore, the characteristic time parameter \( \tau_D \) in Beretta’s theory (see [53], Eq. (35)) called ”intrinsic dissipative time” seems to be conceptually connected with the characteristic decoherence time \( \tau_{\text{dec}} \) in the preceding theoretical models of Sections 11 and 12.

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. [54]).

(F) The novel NCS-experimental findings from a single crystal (presented in Section 3) provide strong evidence for the following effect: For protons, variations of the widths of Compton profiles \( J_H(y) \) are strongly correlated with the variations of NCS-intensity \( I_H \), and therefore also with the ”anomalous” decrease of scattering intensity. This possible correlation and/or connection is novel and, until now, it has been categorically denied (see e.g. last paragraph on p. 463 of [7]). Moreover, current high-precision NCS-experiments on \( H_2 \) (gas phase, various temperatures) provide further evidence for this possible connection [E. Gray et al., work in progress].

(G) As mentioned in Section 4, the ”anomalous” scattering intensity effect has been independently confirmed using electron-proton Compton scattering (ECS) [5, 31, 32, 35]. ECS-investigations from a solid polymer showed exactly the same shortfall in scattered electrons (with energy about 15-30 keV) from hydrogen nuclei, comparable to the shortfall of scattered neutrons in accompanying NCS experiments on the same polymer. Thus this effect appears to be independent of the two fundamental interactions involved (i.e., the electromagnetic and strong interactions) [5, 55].

(H) It may be interesting to note that the theoretical approaches to explain a reduced scattering signal (see the preceding sections) are sensitive to H/D isotope effects as they are seen in some NCS experiments. One reason for this is that the scattering time (\( \tau_{\text{sc}} \), see section 6) varies with the momentum transfer, which strongly depends on the mass of the struck nucleus.
(I) The experimental results discussed and/or presented above and their qualitative theoretical interpretation show that "fast" (say, with 1–100 eV kinetic energy) neutrons provided at spallation sources (e.g., ISIS/U.K., SNS/U.S.A., ESS/Sweden), and electron spectrometers with large scattering angles and associated large momentum transfers, represent novel tools for investigation of attosecond physical and chemical phenomena in condensed matter and molecules, even at ambient conditions. The experimental results represent a challenge to conventional (time-independent) theories of scattering and molecular spectroscopy, which may need to be supplemented by time-dependent theories (also beyond the Born–Oppenheimer approximation) to describe phenomena of Attosecond Physics.

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