

Attosecond Neutron Compton Scattering from Protons Entangled with Adjacent Electrons

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Abstract. The effect of “anomalous” scattering of neutrons and electrons from protons in the electron-volt energy-transfer range is considered, and related experimental results are mentioned. A recent independent confirmation of this effect with a new data analysis procedure is presented. Due to the very short characteristic scattering time, there is no well defined separation of time scales of electronic and protonic motions. An outline of a proposed theoretical interpretation is presented, which is based on the fact that scattering protons represent *open* quantum systems, thus being subject to decoherence.

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1. Introduction

Several neutron Compton scattering (NCS) experiments on liquid and solid samples containing protons or deuterons show a striking anomaly, which is a shortfall in the intensity of epithermal neutrons scattered by the protons and deuterons. For example, neutrons colliding with water for just 100–500 attoseconds ($1 \text{ as} = 10^{-18} \text{ s}$) will see a ratio of hydrogen to oxygen of roughly 1.5 to 1, instead of 2 to 1 corresponding to the chemical formula H_2O , cf. [1, 2]. The experiments were done at the ISIS neutron spallation facility, Rutherford Appleton Laboratory, UK. Due to the large energy and momentum transfers applied, the duration of a neutron–proton scattering event is a fraction of a femtosecond which is extremely short compared to condensed-matter relaxation times.

This new effect has been confirmed using an independent method, electron–proton Compton scattering (ECS), at the Australian National University [3, 4]. ECS experiments from a solid polymer showed the exact same shortfall in scattered

electrons (with energy about 20–35 keV, scattering angle 45°) from hydrogen nuclei, comparable to the shortfall of scattered neutrons in accompanying NCS experiments on the same polymer. The similarity of the results is striking because the two projectiles interact with protons via fundamentally different forces — electromagnetic and strong [2–4].

Due to its novelty and far-reaching consequences, however, this effect has been the focus of various criticisms, cf. [5, 6]. Therefore, considerable work to identify possible sources of experimental and data-analysis errors was made during the last five years, which succeeded to demonstrate the excellent working conditions of the spectrometer Vesuvio at ISIS [7]. Moreover, extending these investigations, the complete “exact formalism” of data analysis [5] was applied to NCS-data by Senesi et al. [8], for the first time; analysis of time-of-flight (TOF) spectra from solid HCl revealed the existence of a strong “anomalous” decrease of the scattering intensity from H (up to 34%). Additionally, this result was found to be in excellent agreement with the corresponding outcome of the standard data analysis procedure applied at ISIS [8].

Recently, scattering of neutrons in the 24–150 keV incident energy range from H_2O relative to that of D_2O was investigated [9]. In clear contrast to the NCS and ECS results, it was claimed that the measured neutron scattering intensity ratios

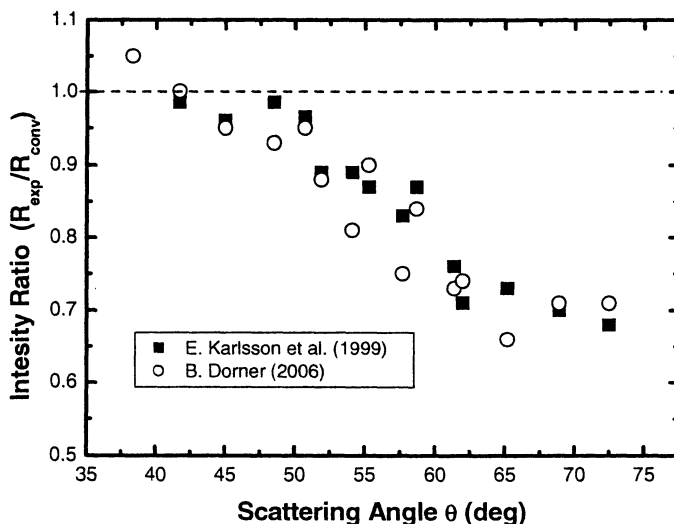


Fig. 1. “Anomalous” scattering from the solid metallic hydride $\text{NbH}_{0.8}$ [12]. Shown is the measured ratio $R_{\text{exp}} = \sigma_{\text{H}}/\sigma_{\text{Nb}}$ of scattering cross-sections of H and Nb normalized with their expected (tabulated) ratio R_{conv} . Broken line: conventional expectation; “full” symbols: results taken from the original publications [12]; “open” symbols: results of the model-independent data analysis procedure invented by Dorner [11](a)

exhibit no anomalous behavior. However, an improved analysis [10] of the keV data within the frame of standard theory showed that the considered scattering anomaly is present at both 5–100 eV and the keV ranges of neutron energies.

Very recently, the mentioned standard NCS-data analysis method [7] was successfully compared with a newly proposed (by B. Dorner, ILL) model-free data-analysis procedure, the latter being independent of the form of the momentum distribution and the resolution function [11]. In this work, the original results from the metallic hydride $\text{NbH}_{0.8}$ [12] were analyzed. The comparison of results obtained with the mentioned two independent methods underline the importance of the effect under consideration, see Fig. 1.

2. On Scattering Time

In the context of NCS, as provided by the Vesuvio setup, the Impulse Approximation (IA) is valid [14, 15] and the characteristic time scale — often termed “scattering time”, τ_{sc} — of the neutron–proton scattering process is very short [3, 7, 12, 13],

$$\tau_{sc} \sim 100\text{--}1000 \text{ as} \quad (1)$$

(as: attosecond). This is a consequence of the large energy (up to 100 eV) and momentum transfers attained with the Vesuvio instrument [7] and follows from the theoretical result valid in the IA [14, 15]

$$\tau_{sc} q v_0 \approx 1, \quad (2)$$

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

$$S(q, \omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \exp(-i\omega t) F(q, t) dt. \quad (3)$$

It is interesting to note that the “actual duration” τ_{act} of a neutron–proton interaction should be even shorter, as a classical estimate indicates. For example, a neutron with kinetic energy $E_0 \approx 10$ eV will pass a distance of 10^{-5} Å (i.e. the range of the strong interaction) in a much shorter time, $\tau_{act} \approx 10^{-19}$ s. In the light of Relativity Theory, this has a crucial consequence: the “actual” (or “effective”) scattering system — i.e. a proton and its adjacent electrons — has a linear dimension not larger than

$$\Delta s_{\max} = c \cdot \tau_{act} \lesssim 0.3 \text{ \AA}$$

(c : velocity of light), since the neutron–nucleus scattering dynamics during τ_{act} cannot be causally influenced by other particles being more than Δs_{\max} apart from the nucleus. Consequently, the scattering system must necessarily be of “microscopic”

dimensions: it contains the scattering nucleus and a part of the adjacent electron density.

However, this is not in conflict with the above estimate, for the following reason. As standard theory shows [15], $S(q, \omega)$ is peaked around the nuclear recoil energy $E_q = \hbar^2 q^2 / 2m$. The scattering time τ_{sc} is also given by the inverse of the width ΔE 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 $\hbar q$. That is, $\tau_{sc} \approx \hbar / \Delta E$. (Interestingly, as Gidopoulos [16] showed, τ_{sc} is also about the inverse of the energy spread of the proton wavefunction after collision.) For a typical value $\Delta E \approx 10$ eV, one gets $\tau_{sc} \approx 10^{-16} - 10^{-17}$ s. In other words, the scattering time τ_{sc} gives a *statistical measure of the length of the time interval during which an actual neutron-proton collision may occur* — in the same way that the spatial extent of a particle wavefunction gives a statistical measure of the extent of the region in which the particle may be found.

To shed more light upon the issue of “relevant scattering time”, one may also refer to the celebrated Margolus–Levitin theorem [17]. Let us consider the neutron-proton system during the collisional process. Obviously, the initial and final states of it are very different and so they can safely be assumed to be orthogonal. This theorem asserts that it 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 [17]. Note that in NCS one has $E_s \approx \hbar\omega$ with $\hbar\omega$ taken at the peak center, and thus E_s is of similar order as the aforementioned energy spread ΔE . Thus it is revealing that also this time T_{\perp} is very similar to the aforementioned scattering time τ_{sc} .

3. Theoretical Model — NCS from Open Quantum Systems

In the following we present an outline of a recently proposed theoretical interpretation of the considered effect, which is based on the general theory of scattering from open quantum systems [18]. As explained above, the scattering system must necessarily be of “microscopic” dimensions (i.e. it is of the order of one Å or less), and since it is embedded in condensed matter it represents an “open” quantum system [21]. This point is crucial, since standard neutron scattering theory always assumes a condensed matter scattering system to be closed, see e.g. [19, 20].

3.1. Scattering from closed systems

First let us consider neutron scattering from a *closed* 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

$$V(\mathbf{r}) = \lambda n(\mathbf{r}), \quad \text{with} \quad \lambda = \frac{2\pi\hbar^2}{m} b. \quad (4)$$

m is the neutron mass, $n(\mathbf{r})$ is the particle density operator

$$n(\mathbf{r}) = \frac{1}{V} \sum_{j=1}^N \delta(\mathbf{r} - \mathbf{R}_j), \quad (5)$$

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

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

$$\Psi(t) = \Psi(0) - i\lambda \int_0^t n(\mathbf{r}, t') dt' \Psi(0). \quad (6)$$

We write the transition probability $W(t)$ between initial states ψ_i (with probability P_i) and final states ψ_f of the scattering system to be given by

$$W(t) = \sum_{i,f} |\langle \psi_f | \lambda \int_0^t n(\mathbf{r}, t') dt' | \psi_i \rangle|^2 P_i. \quad (7)$$

It should be noted that ψ_i and ψ_f are eigenstates of the N -body Hamiltonian H_0 omitting the probe system [19, 20]. The transition probability is then given in the form

$$W(t) = \lambda^2 \int_0^t dt' \int_0^t dt'' \sum_f \langle \psi_f | n(\mathbf{r}, t') \rho n(\mathbf{r}, t'') | \psi_f \rangle, \quad (8)$$

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

In an actual scattering experiment from condensed matter, we do not measure the cross-section for a process in which the scattering system goes from a specific initial state ψ_i to another state ψ_f , both being unobserved states of the many-body system. Therefore, one takes an appropriate average over all these states [19, 20], as done in Eq. (7).

Given the initial (\mathbf{k}_0) and final (\mathbf{k}_1) momenta of an impinging neutron and 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

$$n(\mathbf{r}, t) = \frac{1}{(2\pi)^3} \int d\mathbf{q} n(\mathbf{q}, t) \exp(i\mathbf{q} \cdot \mathbf{r}), \quad (9)$$

where, in the case of neutron scattering, cf. Eq. (5),

$$n(\mathbf{q}, t) = \sum_j \exp[-i\mathbf{q} \cdot \mathbf{R}_j(t)]. \quad (10)$$

Since $n(\mathbf{r}, t)$ is Hermitian, we have $n^\dagger(\mathbf{q}, t) = n(-\mathbf{q}, t)$ and one obtains from Eq. (7)

$$W(t) = \lambda^2 \int_0^t dt' \int_0^t dt'' \sum_f \langle \psi_f | n(\mathbf{q}, t') \rho n(-\mathbf{q}, t'') | \psi_f \rangle. \quad (11)$$

Assuming as usual that $\sum_f |\psi_f\rangle\langle\psi_f| = \mathbf{1}$ [19, 20] we get

$$\sum_f \langle\psi_f|n(\mathbf{q}, t')\rho n(-\mathbf{q}, t'')|\psi_f\rangle = \text{Tr} [n(\mathbf{q}, t')\rho n(-\mathbf{q}, t'')]. \quad (12)$$

If the integration in Eq. (11) is extended over all times (i.e. $t \rightarrow \infty$), this ensues over-all energy conservation. This reproduces the well-known result of standard neutron scattering theory, cf. [19, 20]. Here, however, it is important to retain the finite duration of the scattering time, $t < \tau_{sc}$. This introduces an additional freedom into the theory, because we may be able to observe the influence of the decoherence on the scattering yield; see below. The result will be expressed in terms of the correlation function

$$C(\mathbf{q}, \tau) = \text{Tr} [n(\mathbf{q}, t)\rho n(-\mathbf{q}, t + \tau)] = \text{Tr} [n(\mathbf{q}, 0)\rho n(-\mathbf{q}, \tau)], \quad (13)$$

where we have utilized the fact that the scattering system is stationary. By introducing the so-called *scattering time* τ_{sc} , representing the time interval in which the scattering process may happen, we find

$$W(\tau_{sc}) = \lambda^2 \int_0^{\tau_{sc}} dt' \int_0^{\tau_{sc}} dt'' C(q, t'' - t') = \lambda^2 \int_0^{\tau_{sc}} dt' \int_0^{t'} d\eta [C(q, \eta) + C(q, -\eta)]. \quad (14)$$

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

$$W(\tau_{sc}) \approx 2\lambda^2 \tau_{sc} \int_0^{\tau_{sc}} d\eta C(q, \eta). \quad (15)$$

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

$$\dot{W} \equiv \frac{W(\tau_{sc})}{\tau_{sc}} = 2\lambda^2 \int_0^{\tau_{sc}} d\eta C(q, \eta). \quad (16)$$

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.

3.2. Dynamics of open systems and scattering

We now consider the scattering system to be open and strongly interacting with its environment. We introduce a set of preferred coordinates $\{|\xi\rangle\}$, cf. [21–23], representing the relevant system's degrees of freedom coupled to the neutron probe. The density matrix ρ in (13) is then the *reduced* one in the space spanned by these

states, and it is obtained by tracing out the degrees of freedom of the “environment” [21–23]. For simplicity, throughout this section, let us denote this reduced density matrix by ρ , too.

In the subspace spanned by the preferred coordinates (also termed “pointer basis”), let us assume a Lindblad-type equation [24](a) of the form

$$\partial_t \rho = -i[H, \rho] + \mathcal{R}\rho \equiv \mathcal{L}\rho \quad (17)$$

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

$$\langle A(t) \rangle \equiv \text{Tr}(\rho(t)A) = \text{Tr}(e^{\mathcal{L}t}\rho(0)A) = \text{Tr}(\rho(0)e^{\mathcal{L}^\dagger t}A), \quad (18)$$

where \mathcal{L}^\dagger is defined by $\text{Tr}((\mathcal{L}X)Y) = \text{Tr}(X(\mathcal{L}^\dagger Y))$. Thus it holds $\partial_t \langle A(t) \rangle = \mathcal{L}^\dagger \langle A(t) \rangle$, cf. [24](a). Here is assumed that \mathcal{L} is time independent.

For the correlation functions like the one in Eq. (13) it then holds

$$\langle A(t)B \rangle = \text{Tr}[\rho(0)(e^{\mathcal{L}^\dagger t}A)B] = \text{Tr}[Ae^{\mathcal{L}t}(B\rho(0))] \equiv \text{Tr}(A\rho_B(t)), \quad (19)$$

where $\rho_B(t)$, as defined in Eqs. (19), obeys the equation $\partial_t \rho_B(t) = \mathcal{L}\rho_B(t)$ with 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, Eq. (17).

For simplicity of the further derivations, let us assume here a simple Lindblad-type ansatz for the master equation having only one Lindblad variable X . (In the real system we would have a multitude of such variables.) Thus

$$\partial_t \rho = -i[H, \rho] - K[X, [X, \rho]] = \mathcal{L}\rho, \quad (20)$$

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

$$H|\xi\rangle = \mathcal{E}_\xi|\xi\rangle, \quad X|\xi\rangle = \xi|\xi\rangle. \quad (21)$$

This time evolution is now introduced into the correlation function $C(\mathbf{q}, \tau)$, Eq. (13). A short straightforward calculation (see Ref. [18] for full details) yields for the transition rate the result

$$\begin{aligned} \dot{W} = 2\lambda^2 \int_0^{\tau_{sc}} \sum_{\xi, \xi'} \exp[-i(\mathcal{E}_{\xi'} - \mathcal{E}_\xi)\tau] \exp[-K(\xi' - \xi)^2\tau] \\ \times \langle \xi | n(-\mathbf{q}, 0) | \xi' \rangle \langle \xi' | n(\mathbf{q}, 0) \rho(0) | \xi \rangle d\tau. \end{aligned} \quad (22)$$

The decoherence-free limit of this result (i.e. $K = 0$) corresponds to the conventional result of scattering theory. The oscillatory terms $\exp[-i(\mathcal{E}_{\xi'} - \mathcal{E}_\xi)\tau]$ are

due to the unitary dynamics caused by the commutator part $-i[H, \rho]$ of the master equation (20) for ρ . These factors have the absolute value 1. If decoherence is present ($K > 0$), and especially if $K^{-1} \sim \tau_{sc}$, the additional contractive factors $\exp(-K(\xi' - \xi)^2 \tau) \leq 1$ can be seen to cause a decrease of the transition rate and thus of the associated cross-section. This can be illustrated as follows.

Let us first assume that the reduced density operator $\rho(0)$ can be chosen to be *diagonal* in the preferred ξ -representation (which corresponds to the usual random phase approximation). Then each term of Eq. (22) contains the factor

$$\langle \xi | n(-\mathbf{q}, 0) | \xi' \rangle \langle \xi' | n(\mathbf{q}, 0) \rho(0) | \xi \rangle = |\langle \xi | n(-\mathbf{q}, 0) | \xi' \rangle|^2 \langle \xi | \rho(0) | \xi \rangle \geq 0. \quad (23)$$

In the more general case with $\rho(0)$ being not diagonal in the ξ -representation, one may proceed as follows. The decoherence factors $\exp(-K(\xi' - \xi)^2 \tau)$ 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. (22) should be positive, too. The further terms with ξ being much different from ξ' 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. [22].

The main conclusion from the preceding considerations is that the time average over τ_{sc} in Eq. (22) always *decreases* the value of $\dot{W} \equiv W(\tau_{sc})/\tau_{sc}$, due to the presence of the contractive factors $\exp(-K(\xi' - \xi)^2 \tau) \leq 1$. In other words, the effect of decoherence (and/or dephasing) during τ_{sc} plays a crucial role and may lead to an “anomalous” decrease of the transition rate and the associated scattering intensity. This result is in line with that of Ref. [25], which investigated the standard expression of the double differential cross-section of neutron Compton scattering theory [14, 15] by assuming decoherence of final and initial states of the scattering system.

For further illustration let us consider the following two specific limiting cases: (A) For “vanishing” decoherence, $K \rightarrow 0$, the above contractive factors go to 1 and thus the anomalous scattering effect disappears. That is, the preceding result (22) agrees with the conventional theoretical results [15, 20]. (B) In the opposite case, $K \rightarrow \infty$, only the “diagonal” terms with $\xi = \xi'$ survive in Eq. (22) and the related contractive factors go to 1; additionally, the oscillating factors become equal to 1. Consequently, the time-integration in Eq. (22) has no effect and the rhs of this equation goes over to the standard expression Eq. (16). Also this result is in line with conventional expectations.

4. Conclusions

Theoretical considerations suggest the presence of attosecond quantum entanglement of the scattering protons and the surrounding electrons [1, 3, 12, 13, 25]. Furthermore the usual Born–Oppenheimer approximation is not applicable [13, 16, 25, 26]. Moreover, recent NCS results from liquid HD and the equimolar H_2 – D_2 mixture showing identical anomalous scattering, provided strong evidence that quantum ex-

change correlations between proton pairs cannot be the main physical reason for the effect [27]. For further theoretical discussions, see [25].

In contrast to standard scattering theory [19, 20] of thermal and/or cold neutrons, in which decoherence plays no role at all, our theoretical treatment of NCS is based on the physical fact that micro- and/or mesoscopic scattering systems in condensed matter are open quantum systems. This was shown to follow from the ultrashort scattering time of NCS and ECS. The revealed “anomalous” effect, which has no interpretation in the frame of standard theory [19, 20], indicates that attosecond entanglement (and its decoherence) involving protons are quantum phenomena of broader significance and relevance than realized so far.

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