Neutron Compton scattering — critical analysis of some basic theoretical assumptions

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Abstract. A detailed overview of the conventional (van Hove) neutron scattering theory and its specification to the Impulse Approximation (IA) are given. The IA constitutes the basis of all interpretations of results from neutron scattering experiments in the Compton regime, in which is widely believed that single-particle properties are probed. Here the validity of this approximation is carefully scrutinized and it is shown that there are several different steps in the derivation, whose validity can be questioned: (1) the scattering system is a closed system (in the quantum mechanical sense); (2) all entanglement in the scattering system is absent (N-body operators are replaced by single particle operators); (3) consequently the one-body momentum distribution \( n(p) \) is introduced ad hoc; (4) the \( \delta \)-function in energy assumed in the derivation is not valid when the interaction times underlying the IA are so short that the uncertainty relation allows a noticeable spread in the energy balance. Additionally, (5) the entanglement due to the direct interaction of the neutron with a nucleus is completely absent in the general formalism. The concrete experimental context of recent neutron Compton scattering (NCS) experiments at the ISIS neutron spallation source is considered and emphasis is put on the ultrafast (attosecond) scattering time for the neutron-proton scattering. Recent modern theoretical developments are shortly mentioned in the discussion, which take into account the neutron’s coherence length being larger than the de Broglie wavelength, or decoherence phenomena during the short but finite scattering time.

1. Neutrons, NCS and the eVs/Vesuvio instrument
In this paper the standard theory of neutron scattering [1, 2, 3] is presented in considerable detail as well as its specification to neutron Compton scattering (NCS), also called deep inelastic neutron scattering (DINS) [4]. This technique involves epithermal neutrons. All interpretations of results from neutron scattering in the Compton regime are based on the so-called impulse approximation (IA) [4]. The short scattering times as well as the intuitively simple theoretical "one-body picture" provided by the IA are commonly taken to represent the essential physical features of NCS. In more illustrative terms, the main contents of this article may be considered to refer to an 'ideal machine' where single nuclei are probed.

In this paper the validity of the standard (or conventional) theory and in particular the IA is carefully scrutinized. Modern developments, e.g. questions concerning the neutron’s coherence length \( \lambda_{coh} \) (see [5]) are shortly considered in section 6.

The VESUVIO spectrometer of the ISIS neutron spallation source (Rutherford Appleton Laboratory, U.K.) provides neutrons in the epithermal regime; see [5]. The resulting momentum
and energy transfers from the probing neutron to the scattering system lie in the range of

$$\Delta E_{\text{NCS}} \approx 1 - 50 \text{ eV}, \quad |q| \sim 20 - 150 \text{ Å}^{-1}. \quad (1)$$

The measured peaks in the TOF-spectra (time-of-flight) are well-resolved due to the high momentum transfers, cf. fig. 1. Each recoil peak $X$ corresponds to scattering from one kind of nuclei, each having the mass $M_X$. The neutron flux provided by the Vesuvio instrument is low and therefore scattering of two or more neutrons from one nucleus at the same time is practically impossible.

![Diagram of time-of-flight spectrometer](image)

**Figure 1.** (a) Schematic representation of time-of-flight (TOF) spectrometer eVS/Vesuvio of ISIS. (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. The peak area of atom $X$ (i.e., H, D, etc.) gives the neutron-$X$ scattering intensity. All figures adapted from [6].

In sections 2 and 3 the conventional formalism [1, 2] is derived as well as its specification to NCS - the IA [4]. Section 4 gives an insight into the characteristic scattering time $\tau_{\text{sc}}$ of the neutron Compton scattering regime [7, 4, 5]. In sections 5 and 6 several particularly significant steps in the derivation of sections 2 and 3 are closely examined:

(i) that the scattering system is assumed to be closed (in the quantum mechanical sense);
(ii) the physical justification of the closure relation for specific experimental prerequisites;
(iii) that the $\delta$-function assuring strict energy conservation, as assumed in the derivation, is not valid when the interaction times are so short that the uncertainty relation allows a noticeable spread in the energy balance;
(iv) that all particle-particle entanglement of the scattering system in the IA is assumed to be "broken" (N-body wavefunctions and operators are replaced by single-particle wavefunctions and operators) and the one-body momentum distribution $n(p)$ is introduced ad hoc.

All these points are discussed qualitatively. A quantitative evaluation would require a large amount of numerical calculations that are beyond the scope of this paper. An additional estimate of the characteristic 'scattering time' is presented which implies that the systems involved in NCS are open quantum systems [8, 9].

In the context of the discussions of section 6 it will furthermore become obvious that the coherence length $\lambda_{\text{coh}}$ of the neutron appears to be related with new phenomena (see latest
developments [5]). Moreover, it will be mentioned that the phenomena of quantum entanglement and decoherence between the particles of the scattering system may play a significant role in the NCS dynamical process [10, 11].

2. Conventional neutron scattering theory
The general neutron scattering theory is given by the fundamental van Hove formalism [3], which is based on first order perturbation theory. The latter corresponds, for scattering processes, to the first Born approximation [12].

One considers a closed N-body system described by the Hamiltonian $H_c$ (the subscript "c" indicating "closed" and/or "complete"). The system is perturbed by the impinging neutron.

The starting point in the theory [1, 3, 2] is Fermi’s golden rule [12] which is derived within first order perturbation theory, assuming that the perturbation is sufficiently weak. In the domain of neutron scattering, however, this is not justified in a strict mathematical sense as the perturbation is very strong and being represented by Fermi’s pseudo-potential well localized in space (see Eq. (6) below). Still, according to Squires ([1] p. 16), the golden rule and the pseudo-potential together hold as a good approximation:

"The justification for the use of the golden rule in these circumstances is that, when combined with the pseudopotential, it gives the required result of isotropic scattering for a single fixed nucleus."

In the first Born approximation the incident and scattered wave functions of the neutron are assumed to be plane waves with wave vectors $k_0$ and $k_1$, respectively. (This is an approximation, since s-wave scattering means that the two-particle wave-function has spherical symmetry in the center-of-mass system.)

To assure that the total energy is conserved and to obtain an expression for the double differential cross section, a corresponding δ-function

$$\delta(E_{\nu'} - E_{\nu} + E_0 - E_1)$$

is added to the time-independent expression for the partial differential scattering cross section [1]

$$\frac{d\sigma}{d\Omega}_{\nu \rightarrow \nu'} = \frac{k_1}{k_0} \left( \frac{m_n}{2\pi h^2} \right)^2 |\langle \nu' k_1 | V(r) | \nu k_0 \rangle|^2$$

Thus Eq. (2), denoting that the overall system “neutron & N-body scattering system” is closed, joined together with Eq. (3) give the basic equation of neutron scattering (see [1] Eq. 2.15)

$$\frac{d^2\sigma}{d\Omega d\omega}_{\nu \rightarrow \nu'} = \frac{k_1}{k_0} \left( \frac{m_n}{2\pi h^2} \right)^2 |\langle \nu' k_1 | V(r) | \nu k_0 \rangle|^2 \delta(E_{\nu'} - E_{\nu} + h\omega) ,$$

where $d\Omega$ is the small solid angle in which the scattered neutron with wave-vector $k_1$ is detected and the final state of the neutron together with the system $|\nu' k_1\rangle$ is assumed to fulfil $|\nu'\rangle \otimes |k_1\rangle$. Initial and final eigenstates of the unperturbed and closed Hamiltonian $H_c$ (with energies $E_{\nu}$ and $E_{\nu'}$, respectively) refer to the collective states $|\nu\rangle$ and $|\nu'\rangle$ of the overall N-body scattering system and correspond to the single transition $|\nu\rangle \rightarrow |\nu'\rangle$. $\hbar q$ and $\hbar \omega$ denote the momentum and energy transfers from the neutron to the probe, respectively; i.e.

$$\hbar q = \hbar k_0 - \hbar k_1 , \quad \hbar \omega = E_0 - E_1 .$$

1 To quote Squires in full ([1] p. 16): "The justification for the use of the golden rule in these circumstances is that, when combined with the pseudopotential, it gives the required result of isotropic scattering for a single fixed nucleus. It may be noted that the pseudopotential does not correspond even approximately to the actual potential"
The subscript '0' labels the initial, the subscript '1' the final quantities of the neutron involved in the scattering process.

Introducing Fermi’s pseudo-potential

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

(6)

where \( b \) denotes the bound scattering length of the nucleus and \( m_n \) denotes the mass of the neutron, into the basic equation (Eq. (4)) and taking the sum over all scattering centres (denoted by the subscript 'j') one obtains straightforwardly (see [1] Eq. 2.40)

\[
\left( \frac{d^2\sigma}{d\Omega d\omega} \right)_{\nu \rightarrow \nu'} = \frac{k_1}{k_0} \sum_j b_j \langle \nu' | \exp(i\mathbf{q} \cdot \mathbf{r}_j) | \nu \rangle \frac{2}{\hbar} \delta(E_\nu - E_{\nu'} + \hbar\omega) .
\]

(7)

Please note that in Eq. (7) the variables representing the neutron are c-numbers \( k_0, k_1 \) as well as the vector momentum transfer \( \mathbf{q} = \mathbf{k}_0 - \mathbf{k}_1 \). As mentioned above, the states \( |\nu'\rangle \otimes |\mathbf{k}_1\rangle \) which practically means that the quantum dynamical variables of the neutron are not taken into account. In other words, possible entanglement of the neutron with the system under consideration is presumed to be absent.

Following van Hove [3], one substitutes the \( \delta \)-function in Eq. (7) by its spectral decomposition

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

(8)

and one obtains

\[
\left( \frac{d^2\sigma}{d\Omega d\omega} \right)_{\nu \rightarrow \nu'} = \frac{k_1}{k_0} \frac{1}{2\pi\hbar} \sum_{j,j'} \int_{-\infty}^{\infty} \exp(-i\omega t) \langle \nu | \exp(-i\mathbf{q} \cdot \mathbf{r}_j) | \nu' \rangle \exp(-iE_{\nu'}t/\hbar) \exp(i\mathbf{q} \cdot \mathbf{r}_{j'}) | \nu' \rangle dt .
\]

(9)

In Eq. (10) the time-independent matrix element originating from the golden rule (cf. [1], Eq. (2.2)) is put inside the time integral of the spectral decomposition, Eq. (8), and the time-depending energy terms \( \exp(-iE_{\nu'}t/\hbar) \) and \( \exp(-iE_\nu t/\hbar) \) of the spectral decomposition Eq. (8) are inserted into the second matrix element.

The second, now time-depending matrix element of Eq. (10) is transformed by using time unitarity

\[
\langle \nu' | \exp(i\mathbf{E}_{\nu'}t/\hbar) \exp(i\mathbf{q} \cdot \mathbf{r}_j) \exp(-i\mathbf{E}_\nu t/\hbar) | \nu \rangle
\]

\[ = \langle \nu' | \exp(i\mathbf{H}_c t/\hbar) \exp(i\mathbf{q} \cdot \mathbf{r}_j) \exp(-i\mathbf{H}_c t/\hbar) | \nu \rangle \]

\[ = \langle \nu' | U(t)^\dagger \exp(i\mathbf{q} \cdot \mathbf{r}_j) U(t) | \nu \rangle , \]

(10)

where

\[ U(t) | \nu \rangle = \exp(-i\mathbf{E}_\nu t/\hbar) | \nu \rangle \]

(11)

with

\[ U(t) = \exp(-\frac{i}{\hbar} \mathbf{H}_c t) \]

(12)
denoting the unitary time evolution operator. After this transition one yields
\[
\left( \frac{d^2 \sigma}{d\Omega d\omega} \right)_{\nu \to \nu'} = \frac{k_1}{k_0} \frac{1}{2\pi\hbar} \sum_{j,j'} b_j b_{j'} \int_{-\infty}^{\infty} \exp(-i\omega t) \times \langle \nu | \exp (-i \mathbf{q} \cdot \mathbf{r}_j (0)) | \nu' \rangle \langle \nu' | \exp (i \mathbf{q} \cdot \mathbf{r}_j (t)) | \nu \rangle \, dt ,
\]
where Eq. (14) refers to the Heisenberg representation of the position operators
\[
\mathbf{r}_j (t) \equiv U^\dagger (t) \mathbf{r}_j U (t) .
\]

In order to obtain a measurable expression for the double differential cross-section, Eq. (13) (which still includes all initial and final states of the system), as the first step one sums over all final states \( \nu' \), keeping the initial state \( \nu \) fixed, and then, as the second step, averages over all \( \nu \) [1]. The elimination of the final states is done through the mathematical procedure of closure\(^2\)
\[
\sum_{\nu'} | \nu' \rangle \langle \nu' | = \mathbf{1} ,
\]
or for a pair of operators \( \mathbf{A} \) and \( \mathbf{B} \)
\[
\sum_{\nu'} \langle \nu | \mathbf{A} | \nu' \rangle \langle \nu' | \mathbf{B} | \nu \rangle = \langle \nu | \mathbf{A} \mathbf{B} | \nu \rangle .
\]

In order to take the initial states \( \nu \) into account probabilities \( W_\nu \) of the \( N \)-body scattering system are introduced and
\[
\sum_\nu W_\nu \langle \nu | \mathbf{O} | \nu \rangle = \langle \mathbf{O} \rangle
\]
(\( \mathbf{O} \): arbitrary operator) denotes a thermodynamic average over the initial states, which for thermal neutron scattering belong to thermal equilibrium. \( W_\nu = \frac{1}{Z} \exp (-E_\nu \beta) \) is given by the Boltzmann distribution, \( Z = \sum_\nu \exp (-E_\nu \beta) \) is known as the partition function which ensures \( \sum_\nu W_\nu = 1 \), and \( \beta = \frac{1}{k_B T} \).

These operations lead to the result
\[
\frac{d^2 \sigma}{d\Omega d\omega} = \frac{b^2}{k_0} \frac{k_1}{2\pi} \int_{-\infty}^{\infty} \exp (-i\omega t) \sum_{j,j'} \langle \exp (-i \mathbf{q} \cdot \mathbf{r}_j (0)) \exp (i \mathbf{q} \cdot \mathbf{r}_j (t)) \rangle ,
\]
where, for simplicity, all \( b_j \) are assumed to be equal. The intermediate correlation function [1, 2]
\[
F(q, t) = \frac{1}{N} \sum_{j,j'} (\exp (-i \mathbf{q} \cdot \mathbf{r}_j (0)) \exp (i \mathbf{q} \cdot \mathbf{r}_j (t)))
\]
and the dynamical structure factor
\[
S(q, \omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \exp (-i\omega t) F(q, t) \, dt
\]
give the well-known, final result [1, 2]
\[
\frac{d^2 \sigma}{d\Omega d\omega} = Nb^2 \frac{k_1}{k_0} S(q, \omega) .
\]

\(^2\) From a mathematical viewpoint using the closure relation is a distinct and useful procedure. But are there any physical limitations to this mathematical proceeding in experimental set-ups? (see discussion below)
It should be noted that the quantity we measure in an experiment is not the cross-section itself but the number of neutrons \( I_{sc} \) with fixed initial energy \( E_0 \), scattered per second into a small solid angle \( d\Omega \) (in a given direction) with final energy between \( E_1 \) and \( E_1 + dE_1 \) (and thus \( \hbar d\omega = dE_1 \)). It is proportional to the partial differential cross-section, i.e.

\[
I_{sc}(\Omega, \omega) \propto \frac{d^2\sigma}{d\Omega d\omega}.
\] (22)

3. Neutron Compton Scattering (NCS)

The wavevector transfer in NCS is much larger in extend than the inverse of the nearest-neighbour distance of the nuclei \( q = |q| \gg \frac{2\pi}{a} \). As a consequence we will not detect details of the coherent superposition of scattering amplitudes from different nuclei as it will diminish over a small solid angle — the detector opening and becomes effectively incoherent. According to this common view, the intermediate correlation function in the incoherent approximation \([4, 7, 13]\) takes a much simpler form

\[
F(q, t) = \langle \exp(-i \mathbf{q} \cdot \mathbf{r}(0)) \exp(+i \mathbf{q} \cdot \mathbf{r}(t)) \rangle,
\] (24)

where the index \( j \) of the \( j^{th} \) particle can be obviously dropped and \( \mathbf{r}(t) \) denotes the Heisenberg position operators.

The intermediate correlation function \( F(q, t) \) belongs to the class of stationary time correlation functions which are time translation invariant

\[
C(t_{ini}, t_{end}) = C(t_{end} - t_{ini})
\]
and therefore fulfil

\[
C(t_{ini}, t_{end}) = C(t_{ini} + s, t_{end} + s),
\]

where \( C \) is an arbitrary stationary time correlation function describing thermal equilibrium, cf. Eq. (19), \( s \) is an arbitrary translation in time and \( t_{ini} \) and \( t_{end} \) are two time points with \( t_{ini} \leq t_{end} \). The time points '0' and 't' of Eq. (24) correspond to \( t_{ini} \) and \( t_{end} \), respectively; i.e. \( F(q, t) \) depends on the time interval \( t_{end} - t_{ini} = t \) only and not on any individual time points '0' and 't'. The argument \( t \) in the Heisenberg position operator \( \mathbf{r}(t) \), therefore, ought to be regarded as a time interval \( t_{end} - t_{ini} \). The same conclusion also holds for \( \mathbf{r}(0) \), since '0' corresponds to \( t_{ini} \).

Eq. (24) is often misinterpreted to "prove" that only one-body dynamics is effective in NCS. Since the Heisenberg position operators \( \mathbf{r}(t) \) are N-body position operators (and do not merely account for "the single particle \( j \)") several particles of the system under consideration can become entangled.

The pioneers of NCS Hohenberg and Platzmann [14] proposed the possibility of obtaining information about the single-particle momentum distribution of helium with neutrons of sufficiently high energies (i.e., for helium atoms in the range of 1 eV). Their original idea can be described along the following lines: The neutron scattering acts as a microscopic probe [14] (NCS \( \sim 0.1 \) Å) so the scattering consists of two-body collisions. The 'time passage' of the neutron through the sample must be short compared to the characteristic time scales of the

\[ 3 \] In other terms, for large enough wave vector transfers (as e.g. in NCS) the many contributions with \( j \neq j' \) in the intermediate correlation function, Eq. (19), correspond to uncorrelated phase differences causing fine-oscillating (i.e. constructive and destructive) interference patterns which tend to average to zero, and therefore the scattering becomes effectively incoherent.
dynamic response of the system (see also [2], p. 73). In terms of the uncertainty relation the 'time passage' or the 'time window' happens to be short if the recoil energy of the particle is much greater than the characteristic energy of its bond (e.g. for atoms of a molecule) or kinetic energy (e.g. for helium atoms in the normal or superfluid state). Under these circumstances, one conventionally believes that the dynamic structure factor contains information about the single-particle or single-nucleus momentum distribution.

This interpretation of NCS in terms of the impulse approximation (IA) is not entirely correct, because as described above particle-particle entanglement contained in the Heisenberg position operators \( r(t) \) of Eq. (24) is not included, and by the assumption of a mere single-particle or single-nucleus momentum distribution broken 'by hand'. Therefore also particle-particle decoherence phenomena are left out entirely.

Let us describe the further derivation based on the IA in some detail. For sufficiently short times the Heisenberg position operators are expanded and take the form [4, 13]

\[
\mathbf{r}(t) = \mathbf{r} + \frac{t}{M} \mathbf{p},
\]

(25)

where \( M \) denotes the \( j \)th nucleus mass and \( \mathbf{p} \) the momentum conjugate to \( \mathbf{r} \). As noted above, the operator entity \( \mathbf{r}(t) \) (which may be written as \( \mathbf{r}(t_{ini}, t_{end}) \)) on the left-hand side of Eq. (25) is a N-body operator, as well as the operators on the right-hand side \( \mathbf{r} \) and \( \mathbf{p} \) (which correspond to \( \mathbf{r}(t_{ini}), \mathbf{p}(t_{ini}) \)). This interpretation directly follows from Eq. (24) as \( F(q, t) \) is a stationary time correlation function which has the property of time translation. In other words, neither time \( t' \) nor time \( t \) in the expansion of the Heisenberg position operator is a preferred instant in time. Only the time window \( t_{end} - t_{ini} = t \) is of importance for the evaluation of \( F(q, t) \). Accordingly, we write

\[
\mathbf{r}(t_{ini}, t_{end}) = \mathbf{r}(t_{ini}) + \frac{t_{end} - t_{ini}}{M} \mathbf{p}(t_{ini}).
\]

Note also that the linear expansion of \( \mathbf{r}(t) \) is only possible for velocity independent interatomic forces (cf. [7], p.45).

Sears introduces the impulse approximation in a very elegant way [7], which is as follows. For simplicity we may assume that the scattering system consists of identical particles (atoms). In a very short but finite time interval, the quantum velocity operators \( \mathbf{v}(t) = \mathbf{p}(t)/M \) of a particle with mass \( M \) may not commute for different times. Therefore the intermediate correlation function involves the time-ordering operator \( T \)

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

(26)

where

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

(27)

is the recoil energy of a stationary nucleus in the collision with the neutron. For sufficiently large \( q \), formally \( q \rightarrow \infty \), the right-hand side of Eq. (26) is appreciably different from zero only if \( t \rightarrow 0 \), and thus \( \mathbf{v}(t') \approx \mathbf{v}(0) \equiv \mathbf{v} \). Thus, in the limit of \( q \rightarrow \infty \) one gets

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

(28)

and going to \( \omega \) space by Fourier transform we obtain an expression for the dynamic structure factor in the short-time limit or, equivalently, in the limit of high energies and high impulse transfers

\[
S(q, \omega) \rightarrow \langle \delta(\hbar \omega - \hbar \omega_r + \hbar \mathbf{q} \cdot \mathbf{p}/M) \rangle \equiv S_{IA}(q, \omega)
\]

(29)
with \( v = \frac{p}{M} \). The \( \delta \)-function expresses conservation of energy and momentum for the collision of an epithermal neutron with an atom of initial velocity \( v \). Eq. (29) represents the IA and can be written in the following equivalent form that explicitly expresses the scattering from single nuclei [4, 7, 13]

\[
S_{IA}(q, \omega) = \int n(p) \delta \left( \hbar \omega + \frac{\hbar^2 p^2}{2M} - \frac{\hbar^2 (p + q)^2}{2M} \right) dp \\
= \int n(p) \delta(\hbar \omega - \hbar \omega_r - \hbar q \cdot \frac{p}{M}) dp.
\] (30)

where \( n(p) \) is the (one-body) momentum distribution of the scattering nucleus before collision, i.e. that of the unperturbed system. The well-known partial cross section in the IA writes [1, 3, 2]

\[
\frac{d^2 \sigma}{d \Omega d \omega} = N b^2 k_1 k_0 S_{IA}(q, \omega).
\] (31)

The short scattering times as well as the intuitively simple theoretical "one-body picture" provided by the IA, as described above, are regarded to represent the essential physical features of NCS [4, 7, 13].

While for sufficiently short times the N-body position operators \( r(t) \), according to the derivations above, become one-body operators the N-body wavefunction of the system is not said to factorize into single-particle wavefunctions. Nevertheless, in Eq. (30) the possible entanglement by this N-body wavefunction is tacitly assumed to be negligible entanglement, or in other terms, plays no role by definition.

4. Scattering and collision time in NCS

Exploring the magnitude of the scattering time window in NCS of the elementary neutron-atom (especially, neutron-proton) scattering process a result is obtained that is found in the literature as the outcome of a couple of different estimates (a good overview can be found in [6] with references therein). Recollecting that the momentum and energy transfers in NCS are large (see Eq. (1)) the characteristic time scale — often termed "scattering time" \( \tau_{sc} \) [4] — of the neutron-proton scattering process lies in the attosecond time range [7, 4, 14].

\[
\tau_{sc} \sim 100 - 1000 \text{ as}
\] (32)

(as: attosecond). The estimate of the scattering time corresponds to \( \hbar/\Delta E \), where \( \Delta E \) is the width of the recoil peaks (more precisely, as measured at constant \( q \)). It also can be taken from the theoretical result valid in the IA [7, 15, 16]

\[
\tau_{sc} |q| v_0 \approx 1,
\] (33)

where \( v_0 = \frac{\Delta p}{M} \) (\( \Delta p \): width of the momentum distribution) denotes the root-mean-square (rms) velocity of the nucleus and \( \hbar q \) is the momentum transfer. This formula, introduced by Sears [7] represents the decay time of the intermediate correlation function \( F(q, t) \) and is connected to the dynamic structure factor \( S(q, \omega) \), Eq. 20, by Fourier transform. The intuitively expected "actual duration" of one single scattering event (neutron with proton) may be even much shorter [6]. A neutron with a kinetic energy of \( E_0 \sim 10 \text{ eV} \) (and velocity \( v_0 \sim 44 \text{ km/s} \)) will go over a distance equal to the range of the strong interaction, \( R_{\text{strong}} \sim 10^{-5} \text{ Å} \), in a time about three orders of magnitude shorter than \( \tau_{sc} \), i.e.

\[
t_{\text{strong}} = R_{\text{strong}}/v_0 \sim 2 \times 10^{-20} \text{ s}.
\] (34)

The comparison of these two characteristic times leads us to the following clarification [17](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.

For further considerations of these two characteristic times Eq. (32) and Eq. (34) see section 6 below.

5. Critical Analysis

Some specific points of the derivation of sections 2 and 3 are reconsidered/examined in more detail.

5.1. Applicability of the closure relation

The application of closure, Eq. (15), is the crucial step for the transition from Eq. (13) to the final expression for the double differential cross section Eq. (18) of the general neutron scattering theory. Needless to say, the application of closure is mathematically justified. However, the justification for applying it to an experiment in some cases lacks physical prerequisites as is shown in the following.

E.g., let us think of a standard (direct geometry) instrument where the initial energy of the probing particle $E_n^0$ is fixed. Then $E_n^0$ gives a natural upper bound for the available energy transfers, which, in terms of section 2 is expressed as

$$E_{\nu'} - E_{\nu} \leq E_n^0.$$  

Thus, for any initial energy of the system $E_{\nu}$ there exists a maximal accessible final energy which we denote by $E_{\nu'}^{\text{max}}$.

Is $E_n^0$ large enough, so that any possible state of the N-body system can in principle be excited, then the applicability of the closure relation is also physically justified. If instead $E_n^0$ is smaller than a set of energy transfers $\{E_{\nu'} - E_{\nu}\}$, then the closure does not correspond to the physical situation under consideration as the more energetic final states cannot be excited. In this sense, let us put

$$\hat{\mathbf{I}} = \sum_{0}^{\nu_{\text{max}}'} |\nu'\rangle\langle \nu'| + \sum_{\nu_{\text{max}}'}^{\infty} |\nu'\rangle\langle \nu'|.$$  

(35)

Formally, the introduction of the closure Eq. (35) into Eq. (13), by using the linearity of the integral, yields

$$\int_{-\infty}^{\infty} \exp(-i\omega t) \langle \nu | \hat{\mathbf{A}} \hat{\mathbf{I}} \hat{\mathbf{B}} | \nu \rangle \, dt = \int_{-\infty}^{\infty} \exp(-i\omega t) \sum_{0}^{\nu_{\text{max}}'} \langle \nu | \hat{\mathbf{A}} | \nu' \rangle \langle \nu' | \hat{\mathbf{B}} | \nu \rangle \, dt$$

$$+ \int_{-\infty}^{\infty} \exp(-i\omega t) \sum_{\nu_{\text{max}}'}^{\infty} \langle \nu | \hat{\mathbf{A}} | \nu' \rangle \langle \nu' | \hat{\mathbf{B}} | \nu \rangle \, dt.$$  

(36)

The operators $\hat{\mathbf{A}}$ and $\hat{\mathbf{B}}$ are defined as $\hat{\mathbf{A}} = \exp(-i\mathbf{q} \cdot \mathbf{r}_j(0))$ and $\hat{\mathbf{B}} = \exp(i\mathbf{q} \cdot \mathbf{r}_j(t))$, the sum over the $j$ and $j'$ terms in Eq. (13) as well as the constants are left out for simplicity.

Note that it is erroneous to believe that the first term on the right-hand side of Eq. (36) contains only the states $\{\nu' \leq \nu_{\text{max}}'\}$. All the states $\nu'$ are intrinsically incorporated in the Heisenberg operator, namely $\hat{\mathbf{B}}$, and therefore all states $\nu'$ exist in the first term of the right-hand side.

Correspondingly, the second term on the right-hand side of Eq. (36) contains all states $\nu'$ and not only $\{\nu' \geq \nu_{\text{max}}'\}$.
5.2. Physical constraints of the $\delta$-function

As pointed out in section 2, the $\delta$-function appearing in Eq. (4) has been introduced “by hand” in order to guarantee strict energy conservation (see [1], p.14). In the conventional derivation this introduction is justified in the limit $t \to \infty$ of the expression Eq. (37), see below, and [18], Eq. 42.4. Therefore, the question arises whether this introduction of the delta function is physically justified for the short scattering times in the neutron Compton scattering regime.

Let us proceed according to Landau and Lifshitz [18]. Essentially, we follow the presentation of §41 to §44: According to §44 [18] the transition rate for the system under the influence of a time independent perturbation from a state of energy $E$ to a state of energy $E'$ is proportional to

$$\frac{\sin^2 \frac{E'-E}{2\hbar}}{\pi(E'-E)^2 t}. \quad (37)$$

In the following let us consider a system of two interacting particles [18]. The energies $E$ and $\varepsilon$ of these two particles at a particular point of time, in general, deviate from the energies of the system $E'$ and $\varepsilon'$ measured after an interval of time $\Delta t$. The most probable value of the difference $E'-E$ is of the order $\hbar/\Delta t$. For the system under consideration an estimate is obtained

$$|E + \varepsilon - E' - \varepsilon'| \Delta t \sim \hbar, \quad (38)$$

which does not depend on the magnitude of the perturbation (cf. Eq. (44.1), [18]).

In the favourable case that $\varepsilon$ and $\varepsilon'$ can be exactly determined ($\Delta \varepsilon = \Delta \varepsilon' = 0$) one obtains

$$\Delta(E' - E) \sim \frac{\hbar}{\Delta t}. \quad (39)$$

In the context of NCS the uncertainty in energy $\Delta(E' - E)$ corresponds to the width of the dynamic structure factor $S(q, \omega)$. The mean energy transfer $\Delta E_{NCS}$ (usually called recoil energy $E_{rec}$) happens to be of similar magnitude as the width of the structure factor. (This is an experimental fact.) Thus:

$$\Delta E_{NCS} \sim \Delta(E' - E).$$

Therefore Eq. (39) gives us an estimate of the time interval $\Delta t$ through the energy transfer $\Delta E_{NCS}$ for neutron-proton scattering

$$\Delta t \sim \frac{\hbar}{\Delta E_{NCS}} \leq 1 \text{ fs}.$$ 

In NCS, the scattering time fulfills the relation $\tau_{sc} \sim \frac{\hbar}{\Delta(E' - E)}$. Therefore we have

$$\tau_{sc} \sim \Delta t.$$ 

Recall that the scattering process is accomplished during a time interval of the order $\tau_{sc}$ and therefore large times, $t \to \infty$, play no role in the dynamics of the neutron-proton Compton scattering process.

However, as is well-known and pointed out above, the transition of Eq. (37) into the delta function $\delta(E' - E)$ is possible only for $t \gg \frac{\hbar}{|E'-E|}$, or formally, in the limit of $\Delta t \to \infty$ (see [18], Eq. 42.4).

Recalling that the intermediate correlation function Eq. (19) is based upon the basic formula, Eq. (4), where the delta distribution is supposed to assure energy conservation, we conclude that the approximation of short times done in the IA is in blatant contradiction to these basic assumptions of the conventional formalism.
5.3. The transition from the N-body to the effective one-body picture in NCS

The scattering in the Compton regime, as pointed out in section 3

\[ q = |q| \gg \frac{2\pi}{d} \]

is incoherent and the intermediate correlation function becomes

\[ F(q, t) = \langle \exp(-i q \cdot r(0)) \exp(i q \cdot r(t)) \rangle, \quad (40) \]

as the index \( j \) of the \( j^{th} \) particle can be dropped. But still, Eq. (40) corresponds to the whole N-body system (cf. section 2) as is the N-body Heisenberg operator \( r(t) \equiv U^\dagger(t) r U(t) \).

In order to arrive at the final result of the IA, Eq. (28), the following key approximation (i.e. a short-time expansion) is introduced (see: section 2, Eq. (25); [4], Eq. (6); [13], Eq. (7))

\[ r(t) = r + \frac{t}{M} p. \quad (41) \]

At this point, it should be emphasized that whereas \( r(t) \) is an N-body operator, all operators on the right-hand side, and especially \( p \), are one-body operators. In other words, this key approximation claims to represent a mathematically strict transition from N-body to one-body operators in Eq. (41), yielding the intermediate correlation function of the IA \( F_{IA}(q, t) \equiv \langle \exp[i(\omega_n + q \cdot v)t] \rangle \), Eq. (28). Yet, the physical context requires a different conclusion; see section 6(B). Possible entanglement and decoherence phenomena between different particles are therefore manually excluded from having any significance in Eq. (28).

Summing up, the system under consideration in the above approximation is a single nucleus in an effective Born-Oppenheimer potential. Together, they define an effective single particle Hamiltonian. Therefore, the momentum distribution \( n(p) \) of Eq. (30) describes a single particle in a Born-Oppenheimer potential. This also implies that the effective wave-functions appearing in the main formula Eq. (28) are one-body quantities (see [4], p. 5957).

6. Discussion

In the preceding sections several steps of the conventional theory of neutron scattering were analysed in some detail with particular focus on the Compton regime of the theory. Here we summarize the main results and provide several additional comments and explanations.

(A) The standard, conventional theory as found in the textbooks [1, 2], also called the van Hove formalism [3], is based on the theoretical framework of first order perturbation theory, specifically, on so-called Fermi’s golden rule. Within this theoretical framework, all dynamical quantities (operators) of the impinging particle, i.e. the neutron, are missing in the main formula of the theory, see e.g. Eq. (19). This presumes that the neutron-nucleus entanglement plays no role in the derivation and final results although, in general, the entanglement is caused by the same interaction of the two particles that results in the scattering. E.g., note that the final Eq. for \( F(q, t) \) comprises only position operators of the scattering nuclei, but not of the neutron. The same holds for the wavefunctions included in the thermodynamic average \( \langle ... \rangle \) of Eq. (19). Quantities referring to the neutron appear exclusively in the form of mere c-numbers — the momentum transfer \( \hbar q = \hbar k_0 - \hbar k_1 \) and the magnitudes of the wavevectors of incident and scattered neutron \( k_0 \) and \( k_1 \).

(B) The transition from the N-body Heisenberg position operators \( r(t) \) to single-particle operators \( r \) and \( p \) in Eq. (25) is achieved ”by hand”, and not by a mathematical proof. The expansion of Eq. (25) for small \( t \), i.e. in the limit \( t \to 0 \), seems to clearly show that the operators on the rhs ought to be Schrödinger one-body operators. Yet, this is erroneous, since the quantity \( r(t) \) appears in the equilibrium correlation function \( F(q, t) \), which is by definition \( t \)-translation
invariant, and thus $r$ of the rhs of Eq. (25) must represent a position operator at any time point $t$ (and not only at $t = 0$). This, in consequence, means that $r$ ought to be the N-body Heisenberg-operator of Eq. (14) at time $t_{ini}$. (For $t_{ini}$ see section 3; Eq. (25) of section 3 refers to Eq. (41) in section 5.)

(C) A further consequence of (B) is that the introduction of the momentum distribution $n(p)$ (cf. Eq. (30)), which is assumed to be a single-particle quantity, is ad hoc. A similar conclusion holds for the wavefunctions appearing in Eq. (28) as they are just assumed to be single-particle quantities. This is a tacit assumption, claiming that the many-body wavefunctions factorize into effective single-particle wavefunctions, which is uncertain for condensed matter systems or liquids frequently studied in NCS. Summing up, entanglement and decoherence phenomena between two or more different particles in the systems studied is presumed to be absent and manually excluded.

(D) Due to the ultrashort scattering time $\tau_{sc}$ in NCS one has to consider a significant spread in energy and therefore the $\delta$-function is a questionable oversimplification. It cannot be overemphasized that if the $\delta$-function cannot be introduced into the formalism, Eq. (8), there is no way to arrive at the Heisenberg position operators. If there are no Heisenberg position operators the closure procedure leading from Eq. (13) to Eq. (18) cannot be carried out [1]. As Squires puts it (see [1], pp. 20-21. I replaced the $\lambda'$ terms in the quotation by $\nu'$ terms as in the derivations above, and also properly renumbered the equations):

We can see at this stage the purpose of expressing the $\delta$-function for conservation of energy as an integral with respect to time in Eq. (8). The $E_{\nu'}$ term in the argument of the $\delta$-function prevents us from using Eq. (15) to sum over $\nu'$. By means of Eq. (8) we are able to bring the $E_{\nu'}$ term inside one of the matrix elements, where it reappears as an operator depending on the Hamiltonian. There is now no term in $\nu'$ outside the two matrix elements, and the sum over $\nu'$ can be carried out immediately.

Summing up, if the $\delta$-function cannot be introduced there is no possibility to arrive at the main result of the conventional theory Eq. (19) and consequently the main result of the IA Eq. (28) cannot be obtained.

(E) In the light of Relativity Theory, the results of section 4 show further crucial consequences in the Compton regime of neutron scattering. For illustration, let us present a numerical example in the following. Let us consider a typical epithermal neutron with a mean root velocity $v_0 \sim 44$ km/s corresponding to $E_0 \sim 10$ eV. For a scattering system where single nuclei can be resolved, a semiclassical estimate is carried out regarding each scattering event to be independent of the others. In this semiclassical picture it takes $\tau_{col} \sim 2 \times 10^{-20}$ s for the neutron to go past the range of the nucleus given by the strong force $10^{-5}$ Å.

The effective scattering system (e. g. a proton and parts of its surroundings) therefore must have a linear dimension smaller than $[6]$

$$\Delta s_{caus} = c \cdot \Delta t_{strong} \sim 0.1 \, \text{Å}$$

where $c$ is the velocity of light, since the neutron-nucleus scattering dynamics during $\Delta t_{strong}$ cannot be causally influenced by other particles being more than $\Delta s_{caus}$ apart from the colliding nucleus. The dimension of a typical covalent bond H-X (with X= C, O, N, etc.) is roughly 1 Å and exceeds the event-horizon defined by $\Delta s_{caus}$. For the H$_2$ molecule with an internuclear distance of approximately 0.75 Å, consequently, only one proton and a part of the electrons’ degrees-of-freedom causally participate in the collision process. In other terms, the usual view of the H$_2$ molecule to represent a closed system appears to contradict the principle of causality. In consequence, the notion of a closed “effective one-body” scattering system has no physical significance in this example. Therefore, the above line of argumentation implies that the systems
studied in NCS experiments ought to be treated — for fundamental reasons — as open quantum systems.

(F) Last but not least, some modern theoretical developments may be mentioned here. In the conventional theory [1, 4] the neutron’s coherence length plays no role. In contrast, it is of great significance in the theoretical model of Ref. [5], in which the observed ”anomalous” NCS intensity effect from protons [20] is explained as a result of interference when the scattering particle (i.e. neutron) interacts with more than one hydrogen nucleus. The coherence volume of the actual setup, which limits the number of interfering particles, is therefore an important parameter. In particular, the author of [5] provides a quantitative description of the neutron’s coherence volume for the actual VESUVIO spectrometer and of the probability for the scattering neutron to interact coherently with two or more protons or deuterons.

Another theoretical model for the interpretation of the aforementioned ”anomalous” intensity effect [20, 15, 21] is proposed in Ref. [22]. Central to this qualitative model is the decoherence process of the scattering proton and its adjacent particles during the time-window of scattering time $\tau_{sc}$.

Note that quantum coherence and entanglement is preserved over the sub-fs scattering times relevant for the NCS process, even in the condensed systems studied [5, 20, 15, 21]. Compton scattering opens, therefore, a time-window for studies of coherence and decoherence not easily accessible by other methods.

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References
[1] Squires G L 1996 Introduction to the Theory of Thermal Neutron Scattering (Mineola: Dover)
[2] Lovesey S W 1984 Theory of Thermal Neutron Scattering from Condensed Matter (Oxford: Oxford University Press)
[3] van Hove L 1954 Phys. Rev. 95 249
[4] Watson G I 1996 J. Phys.: Condens. Matter 8 5955
[5] Karlsson E B 2012 Int. J. Quantum Chemistry 112 587-602
[6] Chatzidimitriou-Dreismann C A and Tietje I C 2010 J. Phys.: Conf. Ser. 237 012010
[7] Sears V F 1984 Phys. Rev. B 30 44
[8] Joos E, Zeh H D, Kiefer C, Kalini D J W, Kupsch J, Stamatescu I-O 1996 2003 Decoherence and the Appearance of a Classical World in Quantum Theory (Springer-Verlag Berlin Heidelberg New York)
[9] Breuer H P and Petruccione F 2002 The Theory of Open Quantum Systems (Oxford: Oxford University Press)
[10] Schulman L S and Gaveau B 2006 Phys. Rev. Lett. 97 240405
[11] Chatzidimitriou-Dreismann C A, Gray E MacA, and Blach T P 2011 AIP Advances 1 022118
[12] Ballentine L E 1998 Quantum Mechanics – A Modern Development (Singapore: World Scientific)
[13] Andreani C, Colognesi D, Mayers J, Reiter G F and Senesi R 2005 Adv. Phys. 54 377
[14] Hohenberg P C and Platzman P M 1966 Phys. Rev. 152 198
[15] Chatzidimitriou-Dreismann C A, Vos M, Kleiner C and Abdul-Redah T 2003 Phys. Rev. Lett. 91 057403
[16] Karlsson E B, Chatzidimitriou-Dreismann C A, Abdul-Redah T, Streffer R M, Hjörvarsson B, Ohrlmalm J and Mayers J Europhys. Lett. 1999 46 617.
[17] Gidopoulos N I (a) Phys. Rev. B 2005 71 054106; (b) Non-adiabatic electronic excitations in neutron Compton scattering, ISIS Science Highlights, 2004.
[18] Landau L D, Lifshitz E M (a) 1958 course of theoretical physics 3: quantum mechanics (Pergamon Press London - Paris) (b) 1979 Lehrbuch der theoretischen Physik III: Quantenmechanik (Akademie-Verlag Berlin)
[19] Marshall W and Lovesey S W 1971 *Theory of Thermal Neutron Scattering*, (Oxford: Oxford University Press)

[20] Chatzidimitriou-Dreismann C A, Abdul-Redah T, Streffer R M F and Mayers J 1997 *Phys. Rev. Lett.* **79** 2839

[21] Cooper G, Hitchcock A P and Chatzidimitriou-Dreismann C A 2008 *Phys. Rev. Lett.* **100** 043204

[22] Chatzidimitriou-Dreismann C A and Stenholm S, in: V M Akulin, A Sarfati, G Kurizki, and S Pellegrin, *Decoherence, Entanglement and Information Protection in Complex Quantum Systems*, NATO Science Series II – Vol. **189** (Springer, Dordrecht, 2005); pp. 555-562; also available at arXiv:quant-ph/0702038v1.

[23] Merzbacher E 1961 *Quantum mechanics* (John Wiley & Sons, Inc.)