Neutron Compton scattering by proton and deuteron systems with entangled spatial and spin degrees of freedom

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Several recent experiments on liquid and solid samples containing protons or deuterons have shown an interesting anomaly, which is apparently absent when the hydrogen isotopes are replaced by heavier particles. The anomaly is a shortfall in the intensity of energetic neutrons scattered by the samples; specifically, the intensity per hydrogen isotope in bulk samples is smaller than the intensity for total scattering by an isolated hydrogen isotope. Short-lived correlations in the spatial and spin degrees of freedom of the hydrogen isotopes have been proposed as an explanation of the anomaly. The correlations involve entanglements of the degrees of freedom created by the requirements of quantum mechanics applied to identical particles. By using energetic neutrons to perform Compton scattering experiments on the hydrogen isotopes, the time scale of the experimental probe covers the region of 10^{-16}–10^{-15} s where entangled states might still be expected to survive. The proposed explanation is pursued here by reporting the cross section for Compton scattering, also known as deep inelastic scattering, by two identical nuclei occupying nonequivalent states. The model reproduces the observed dependence of the cross section on energy transfer, in which intensity accumulates at the recoil energy of a single nucleus. Several features of the model demand that the intensity at the recoil energy is indeed less than the intensity for total scattering by an isolated nucleus. Although the pair approximation used here is unquestionably a first approximation to real many-body entanglements, it is a compelling explanation of all the observations, including the restoration of the normal cross section at longer observation times achieved by moving to longer scattering times.

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I. INTRODUCTION

Several very carefully controlled neutron scattering experiments [1–4] have shown a shortfall in the intensity recorded for scattering by hydrogen isotopes. In the experiments, primary neutrons with energies of a few eV are utilized to perform Compton scattering (also known as deep inelastic scattering), which is described by the impulse approximation, to a good approximation [5]. The shortfall in intensity is as much as 40% of the normally expected cross sections. In two [2,3] of the recent papers it has been demonstrated that the shortfall exists only during the first 10^{-15} s for protons loaded in metals, whereas in other H-containing materials [1,4] the shortfall is similar in size but has a longer lifetime.

No complete theoretical explanation of the anomalous findings has yet been given. It has been proposed [1] that the findings in question, and similar results obtained from Raman scattering by mixtures of protons and deuterons in water [6], might be caused by entanglement of the spatial degrees of freedom of the hydrogen isotopes probed in scattering. For identical particles, with spin I, entanglement of the spatial and spin degrees of freedom is created by the requirement of quantum mechanics that on interchange of two particles the complete wave function acquires a phase factor $(-1)^{2I}$. Such an entanglement of atoms is known from quantum optics experiments, where it may exist for times up to 10^{-5} s in very well shielded environments [7], but it has never been directly observed in condensed matter where, if it exists, it would have a very much shorter lifetime. However, some unexplained proton correlations in metal hydrides, e.g., strongly isotope-dependent diffusion of hydrogen isotopes on a metal surface [8] and diffusion anomalies of protons in the presence of positive muons [9(a)], can be viewed as indirect evidence for short-lived quantum correlations in the isotopically pure systems, enhancing the diffusion. Similarly, nonlinearities in the ionic conductivity of water as functions of the deuterium-hydrogen content [10] have been taken as evidence for quantum correlations that are broken in the mixed H-D systems. (The possibility of ''coherent dissipative systems,'' including short-lived and spatially strongly restricted entanglement between atoms in condensed matter, has been discussed in several papers [11–13].)

Following a preamble in the next section on the interpretation of entanglement in quantum systems and known limitations of our model, based on scattering by a unit consisting of two identical particles, Secs. III and IV describe the calculation we have done. Section V contains the application of our theoretical findings to the interpretation of the experiments in question. Conclusions are gathered in Sec. VI.

II. PREAMBLE

As concerns neutron scattering, it is tempting to try to trace the shortfall in intensity back to a mixing of scattering amplitudes for total spin $I-\frac{1}{2}$ and $I+\frac{1}{2}$, since these amplitudes are strongly different, in particular for scattering by protons. The assumption is then that the eV neutrons, which have such short wavelengths that each of them can ‘‘see’’ only particles at one of the hydrogen sites involved, scatter on a quantum state that is a superposition of components with different spin projections. Such superpositions are a characteristic of quantum entanglement of the spin degrees of freedom of two or more particles. Spin entanglement is necessarily coupled to a spatial entanglement (and vice versa) for identical particles. This means that the complete wave function for the system is also a superposition of components where particle labels have changed places between the hydrogen sites involved. For the moment, we leave the question open about how such entanglement can possibly be created and carry out a calculation of the consequences for neutron Compton scattering should entanglement indeed exist at the encounter of the neutron with the proton (or deuteron) system.

A starting point is the case of two protons, labeled α and β. Such a pure system (which nobody has so far been able to study experimentally) was recently considered theoretically [14]. If the two protons are produced by separating them from each other (and from the bound electrons) in a hydrogen molecule, they will exhibit a quantum entanglement expressed (for a spin singlet state $J=0$ for the pair) through the wave function,

$$\Psi(J=0) \propto [P(\mathbf{R}_\alpha)Q(\mathbf{R}_\beta)+P(\mathbf{R}_\beta)Q(\mathbf{R}_\alpha)][\uparrow(s_\alpha)\downarrow(s_\beta) - \uparrow(s_\beta)\downarrow(s_\alpha)], \tag{2.1}$$

where P (right-hand channel, say) and Q (left-hand channel, say) are distinct one-particle orbitals. The spin part is, similarly, expressed through a superposition of spin-up $\uparrow(s_\alpha)$ and spin-down $\downarrow(s_\beta)$ functions for the protons α and β. Evidently, this wave function would lead to an equal probability for finding the two particles in the (separated) product states,

$$P(\mathbf{R}_\alpha)\uparrow(s_\alpha)Q(\mathbf{R}_\beta)\downarrow(s_\beta), \quad P(\mathbf{R}_\alpha)\downarrow(s_\alpha)Q(\mathbf{R}_\beta)\uparrow(s_\beta),$$
$$P(\mathbf{R}_\beta)\uparrow(s_\beta)Q(\mathbf{R}_\alpha)\downarrow(s_\alpha), \quad P(\mathbf{R}_\beta)\downarrow(s_\beta)Q(\mathbf{R}_\alpha)\uparrow(s_\alpha). \tag{2.2}$$

One of these four states would also be the result if all entanglement (in spin and coordinate space) were broken by a measurement process. Equation (2.1) represents a so-called maximally entangled state, enforced by the Pauli rules for fermions. Similar expressions are well known from the photon-photon pairs studied extensively in Einstein-Podolski-Rosen-type experiments (where, however, the spatial part of the two-particle wave function is usually not written out). At the right-hand side (and similarly for measuring at the left-hand side) it is therefore not known, *a priori*, whether the particle observed will be α or β, nor whether this particle will have spin up or spin down.

Of course, the real situation for protons (or deuterons) in a condensed matter system is very different from that of isolated, entangled proton pairs assumed so far. First of all, the particles are strongly interacting with their environment, which limits the survival of spatial entanglement drastically (decoherence times in a metallic hydride are estimated to be of the order of 10^{-16} s for correlated objects of a few angstroms extension [2]). Secondly, protons or deuterons are normally not in paired states but each of them interacts with more than one neighbor at a time. If entanglement exists, it is therefore likely to be shared by several particles, and the pair model used here can only be seen as a model to illustrate principles for treating scattering on entangled systems (although it should be noted that coherent dissipative systems [11] are expressed in terms of ‘‘geminals,’’ i.e., two-particle wave functions). A further consequence of going beyond the two-particle entanglement in a system of identical particles would be that particles $\gamma,\delta,...$ in the labeling series $\alpha,\beta,\gamma,\delta,...$ are now also allowed as partners for the particle to be expelled in a Compton process. The strength of entanglement between, say, particles α and β and particles β and γ is also expected to vary with time, with possibility for entanglement swapping [15,16], so that suddenly γ would turn out to be the appropriate partner of α, rather than β. This would bring about a fast decrease of entanglement for particles situated at two neighboring sites.

III. THE NEUTRON SCATTERING CROSS SECTION AND ITS COMPTON LIMIT

In this section we derive the cross section for scattering by a system consisting of only two identical particles. Arising from the principle of indistinguishability of identical particles is a correlation in the quantum numbers that define the states of the system, also described as an entanglement of the system's spatial and spin degrees of freedom. The explicit form of the correlation in the quantum numbers found here is correct for two particles with arbitrary spin. There are two spatial centers, labeled 1 and 2, and the distance between them is similar to the distance between neighboring ions in the crystal. A particle has spin I, and $I=\frac{1}{2}(1)$ for a proton (deuteron). The complete wave function acquires a phase $(-1)^{2I}$ when the particles are interchanged.

The one-particle spatial orbitals discussed in the previous section are taken to be nonequivalent and purely real and they are denoted by $\varphi_1(\mathbf{R})$ and $\varphi_2(\mathbf{R})$, with

$$\int d\mathbf{R}\,\varphi_1^2(\mathbf{R}) = \int d\mathbf{R}\,\varphi_2^2(\mathbf{R}) = 1. \tag{3.1}$$

The particles are at positions $\mathbf{R}=\mathbf{R}_\alpha$ and $\mathbf{R}=\mathbf{R}_\beta$. Suitably normalized, the spatial wave function of the initial state of the two particles is

$$[2(1+\zeta S_{12}^2)]^{-1/2}[\varphi_1(\mathbf{R}_\alpha)\varphi_2(R_\beta)+\zeta\varphi_1(\mathbf{R}_\beta)\varphi_2(\mathbf{R}_\alpha)]. \tag{3.2}$$

Here, $\zeta=(-1)^J$ where the total spin of the particles, J, is an integer and $\zeta^2=1$. The overlap integral for φ_1 and φ_2 denoted in Eq. (3.2) by S_{12} is assumed to be small on account of the large distance between the two centers and, henceforth, it will be neglected. The orbitals φ_1 and φ_2, and the energy to which Eq. (3.2) corresponds, might depend on the magnitude of J or whether J is an even or an odd integer. The spinor for the two particles is

$$\chi_M^J(\alpha,\beta)=\sum_{mn}(I_\alpha m I_\beta n|JM)|I_\alpha m\rangle|I_\beta n\rangle. \tag{3.3}$$

The Clebsch-Gordan coefficient in Eq. (3.3) is defined in accord with Edmonds [16] and $\chi_M^J(\beta,\alpha)=(-1)^{2I}\zeta\chi_M^J(\alpha,\beta)$. In keeping with the nonrelativistic limit of quantum mechanics, the complete wave function of the two particles is the product of Eqs. (3.2) and (3.3), and on interchanging the particles the complete wave function does acquire a phase $(-1)^{2I}$ (In the case of a system with three or more identical particles, the spatial wave function need not necessarily be either symmetrical or antisymmetrical with respect to the interchange of any pair of particles, as the complete wave function must be.)

The wave function that describes the particles after the scattering event should also acquire a phase $(-1)^{2I}$ when the two particles are interchanged. As we shall see, in order for our model to reproduce the observed distribution of intensity as a function of neutron energy transfer, one of the two particles must be found in a state described by a plane wave, to a good approximation, with a wave vector almost equal to the wave-vector transfer, $\mathbf{k}$. The magnitude of $\mathbf{k}$ in the experiment is purposely made very large.

Let the plane wave be proportional to $\exp(i\mathbf{p}'\mathbf{R})$ and denote the second one-particle orbital by $\psi(\mathbf{R})$. The plane wave can be normalized in a box, and $\psi(\mathbf{R})$ is normalized to unity like φ_1 and φ_2. The normalization box has a volume Ω and the normalization factor attached to the plane wave will be absorbed in a momentum wave function [Eq. (3.8) below]. For the moment, $\psi(\mathbf{R})$ is not specified and a complex function is allowed.

The neutron-nuclear interaction operator is

$$V=b_\alpha\exp(i\mathbf{k}\cdot\mathbf{R}_\alpha)+b_\beta\exp(i\mathbf{k}\cdot\mathbf{R}_\beta), \tag{3.4}$$

where the scattering-length operator b is independent of the position variable. A matrix element of V taken between the initial orbital (3.2) and the proposed final-state orbital, namely,

$$\frac{1}{\sqrt{2\Omega}}[\exp(i\mathbf{p}'\cdot\mathbf{R}_\alpha)\psi(\mathbf{R}_\beta)+\zeta'\exp(i\mathbf{p}'\cdot\mathbf{R}_\beta)\psi(\mathbf{R}_\alpha)], \tag{3.5}$$

contains eight terms, four of the form

$$\int d\mathbf{R}_\alpha\exp[i\mathbf{R}_\alpha\cdot(\mathbf{k}-\mathbf{p}')]\varphi_1(\mathbf{R}_\alpha)\int d\mathbf{R}_\beta\psi^*(\mathbf{R}_\beta)\varphi_2(\mathbf{R}_\beta), \tag{3.6}$$

and four of the form

$$\int d\mathbf{R}_\alpha\exp(i\mathbf{R}_\alpha\cdot\mathbf{k})\psi^*(\mathbf{R}_\alpha)\varphi_1(\mathbf{R}_\alpha)\int d\mathbf{R}_\beta \times\exp(-i\mathbf{p}'\cdot\mathbf{R}_\beta)\varphi_2(\mathbf{R}_\beta). \tag{3.7}$$

Consider the first integral in Eq. (3.7). Because the magnitude of $\mathbf{k}$ is very large, the phase factor $\exp(i\mathbf{R}_\alpha\cdot\mathbf{k})$ contains very many oscillations, between $+1$ and -1, as $\mathbf{R}_\alpha$ varies in the volume of space in which $\varphi_1(\mathbf{R}_\alpha)$ is appreciably different from zero; a volume that is of the order of a unit cell in the crystal. In consequence, the integral in question is close to zero. The corresponding integral in Eq. (3.6) can be significantly different from zero when $\mathbf{p}'$ is chosen close to $\mathbf{k}$, so $\exp[i\mathbf{R}_\alpha\cdot(\mathbf{k}-\mathbf{p}')]$ has relatively few oscillations in a unit cell. From the conservation of momentum it follows that $\mathbf{k}-\mathbf{p}'=\mathbf{p}$ is the initial wave vector of the struck particle. With $\mathbf{p}'\simeq\mathbf{k}$ the second integral in Eq. (3.7) is close to zero and the product of integrals in the expression can be safely neglected in comparison to Eq. (3.6). For the latter we write $K_1(\mathbf{p})T_2$. Here, the momentum wave function

$$K_1(\mathbf{p})=\Omega^{-1/2}\int d\mathbf{R}\exp(i\mathbf{R}\cdot\mathbf{p})\varphi_1(\mathbf{R}) \tag{3.8}$$

satisfies

$$\sum_p|K(\mathbf{p})|^2=\frac{\Omega}{(2\pi)^3}\int d\mathbf{p}|K(\mathbf{p})|^2=1,$$

and the overlap integral

$$T_2=\int d\mathbf{R}\,\psi^*(\mathbf{R})\varphi_2(\mathbf{R}). \tag{3.9}$$

The four terms that survive the Compton limit are

$$\tfrac{1}{2}\{b_\alpha[K_1(\mathbf{p})T_2+\zeta K_2(\mathbf{p})T_1]+\zeta' b_\beta[K_2(\mathbf{p})T_1+\zeta K_1(\mathbf{p})T_2]\},$$

where $\mathbf{p}=\mathbf{k}-\mathbf{p}'$, and K_2 and T_1 are defined in accord with the foregoing definitions, Eqs. (3.8) and (3.9).

In writing down the result for the matrix element of V we will assume that the momentum wave functions constructed from φ_1 and φ_2 are almost the same and denote the common value by $K(\mathbf{p})$. One finds

$$\langle\text{final}|V|\text{initial}\rangle=K(\mathbf{p})F(J',J), \tag{3.10}$$

where

$$F(J',J)=\tfrac{1}{2}(\langle J'|b_\alpha|J\rangle+\zeta\zeta'\langle J'|b_\beta|J\rangle)(T_2+\zeta T_1) \tag{3.11}$$

and

$$\langle J'|b|J\rangle=[\chi_{M'}^{J'}(\alpha,\beta)]^+b\chi_M^J(\alpha,\beta). \tag{3.12}$$

To achieve a simple notation, in F and the matrix element of b we do not display M and M'.

The explicit form of the scattering-length operator is

$$b=A+B\mathbf{s}\cdot\mathbf{I}. \tag{3.13}$$

In this result, $\mathbf{s}$ is the operator for the spin of the neutron and A and B are linear combinations of the scattering lengths for the two possible states of the total spin, $I\pm\frac{1}{2}$. One sees that the matrix element (3.12) is purely real. The single-atom cross section is $4\pi\overline{b^2}$ where $\overline{b^2}$ is obtained by averaging b^2 over random orientations of $\mathbf{I}$, or, equivalently, random orientations of $\mathbf{s}$, and the result is

$$\overline{b^2} = A^2 + \tfrac{1}{4} B^2 I(I+1). \qquad (3.14)$$

Let us now consider the change in energy of the particles between the initial and final states. Because the duration of the scattering event is by design very small, we anticipate that the position of the struck particle is almost unchanged, and its potential energy is essentially the same in the initial and final states of the scattering event. On the other hand, the kinetic energy of the struck particle changes from $(\hbar p)^2/2M$ to $\hbar^2|\mathbf{k}-\mathbf{p}|^2/2M$. Assuming the potential energy is the same in the initial and final states, it cancels out in the conservation of energy of the struck particle, which then reads,

$$E + \frac{(\hbar p)^2}{2M} = E' + \frac{\hbar^2|\mathbf{k}-\mathbf{p}|^2}{2M},$$

where E and E' are the initial and final energies of the neutron. Moreover, measured on the scale of energy for the change in kinetic energy of the struck particle, the energy of the other particle is unchanged in the scattering event. Writing $\hbar\omega = E - E'$, the cross section for scattering is proportional to

$$\sum_p \delta\left(\hbar\omega - E_R + \frac{\hbar^2}{M}\mathbf{k}\cdot\mathbf{p}\right)|K(\mathbf{p})|^2 |F(J',J)|^2, \quad (3.15)$$

where the recoil energy $E_R = (\hbar k)^2/2M$. Regarded as a function of energy transfer the cross section (3.15) peaks at the recoil energy, in accord with the observations we aim to interpret. The width in energy of the recoil peak is related to the momentum density $|K(\mathbf{p})|^2$ in the ground state. These features of the energy dependence of Eq. (3.15) are signatures of the Compton limit of scattering, which has been extensively studied [5].

Having established the correct energy dependence of the cross section, we turn attention to the intensity that accumulates around E_R. From Eq. (3.11),

$$|F(J',J)|^2 = \tfrac{1}{4}(\langle J'|b_\alpha + \zeta\zeta' b_\beta|J\rangle)^2 |T_1 + \zeta T_2|^2. \qquad (3.16)$$

This expression depends on the total spin of the initial and final states through the matrix elements of the scattering-length operator and the phases ζ and ζ'. In the following text we evaluate expression (3.16) for the intensity, or structure factor, associated with the Compton scattering by a pair of correlated particles.

The initial and final wave functions belong to states of the two particles with widely different energies and the overlap of the wave functions is negligible. Using

$$[\chi^{J'}_{M'}(\alpha,\beta)]^+ \chi^J_M(\alpha,\beta) = \delta_{J,J'}\delta_{M,M'},$$

one finds

$$\langle \text{final}|\text{initial}\rangle = \tfrac{1}{2}\delta_{J,J'}\delta_{M,M'}(1+\zeta\zeta')[K_1(-\mathbf{p}')T_2 + \zeta K_2(-\mathbf{p}')T_1]. \qquad (3.17)$$

The required zero overlap is achieved with $J \neq J'$, for all $\mathbf{p}'$ and $\psi(\mathbf{R})$. Hence, not all values of J' are accepted in the evaluation of the structure factor. In the latter, nonzero values of the matrix element of the scattering-length operator obey the selection rule $J' = |J-1|$, J, and $J+1$, since b contains $\mathbf{I}$, which is a tensor of rank 1 [cf. Eq. (4.3) below]. The outcome of the selection rule and the orthogonality condition is to restrict J' to the values $|J-1|$ and $J+1$, whence $\zeta\zeta' = -1$.

IV. THE INTENSITY OF SCATTERED NEUTRONS

Attention in this section is on the intensity at the recoil energy appropriate to unpolarized neutrons. The intensity is calculated from Eq. (3.16), and its dependence on the total spin of the initial and final states, J and J', arises from both the matrix elements of b_α and b_β and the phases $\zeta = (-1)^J$ and $\zeta' = (-1)^{J'}$. The dependence of the structure factor $|F(J',J)|^2$ on J' is solely in the matrix element of the scattering length, for

$$\langle J'|b_\beta|J\rangle = \zeta\zeta'\langle J'|b_\alpha|J\rangle = -\langle J'|b_\alpha|J\rangle, \qquad (4.1)$$

a result that follows directly from Eqs. (3.3) and (3.12) and $J + J' =$ odd integer. Using Eq. (4.1) in Eq. (3.16),

$$|F(J',J)|^2 = \langle J'|b_\alpha|J\rangle^2 |T_1 + \zeta T_2|^2. \qquad (4.2)$$

To obtain the observed intensity we average $\langle J'|b_\alpha|J\rangle^2$ over the projections of the initial total spin and sum over the values of the projection of the final total spin. Starting from Eq. (3.13) a straightforward calculation yields

$$\tfrac{1}{2}\sum_{m_n m_n'} \frac{1}{(2J+1)} \sum_{MM'} |\langle \tfrac{1}{2} m_n'|\langle J'|b_\alpha|J\rangle|\tfrac{1}{2} m_n\rangle|^2 = \delta_{J,J'}A^2 + \tfrac{1}{4}B^2(2J'+1)I(I+1)(2I+1)\begin{Bmatrix} I & 1 & I \\ J' & I & J \end{Bmatrix}^2, \qquad (4.3)$$

where the last quantity on the right-hand side is the square of a $6j$ symbol [16]. The results (4.2) and (4.3) completely determine the intensity of the Compton scattering of unpolarized neutrons by a pair of correlated nuclei.

In considering the application of expression (4.3), thought must be given to the dependence of the energy of the particles on their total spin, arising ultimately from the principle of indistinguishability of identical particles. For $I = \tfrac{1}{2}$, to each energy level there corresponds one definite value of the total spin, 0 or 1. There is not necessarily a one-to-one correspondence between the spin values and the energy levels for particles with spin $I > \tfrac{1}{2}$, and energy levels to which there correspond symmetrical (antisymmetrical) spatial wave functions can occur for any even (odd) value of the total spin. The magnitude of the energy dependence, known as the exchange splitting, is related to the overlap of one-particle orbitals at different centers. For the moment, we will assume the energy dependence is very small and unimportant in addressing the questions at hand. Rather, all values of J and J' are now regarded as equally likely, subject to the tenets of quantum mechanics. In consequence, we will sum the intensity over the allowed values of J' and average it with respect to J.

In executing the sum on J' we recall the condition $J' \neq J$, which stems from the orthogonality of the initial and final states of the particles. From Eq. (4.3),

$$\sum_{J'\neq J} \tfrac{1}{2} \sum_{m_n m_n'} \frac{1}{(2J+1)} \sum_{MM'} |\langle \tfrac{1}{2} m_n' | \langle J' | b_\alpha | J \rangle | \tfrac{1}{2} m_n \rangle|^2 = (\sigma_{\rm inc}/4\pi)\left(1 - \frac{J(J+1)}{4I(I+1)}\right), \tag{4.4}$$

where $\sigma_{\rm inc} = \pi I(I+1)B^2$ is the single-atom incoherent cross section.

The result (4.4) is central in subsequent developments so it is fitting to record an alternative derivation of it. From the first equality in Eq. (4.1) we find $\langle J|b_\alpha|J\rangle = \langle J|b_\beta|J\rangle$ and the sum

$$\sum_{J'M'} |\langle J'M'|(b_\alpha - b_\beta)|JM\rangle|^2 = \langle JM|(b_\alpha - b_\beta)^2|JM\rangle$$

contains no contribution from the term $J = J'$. For unpolarized neutrons,

$$(b_\alpha - b_\beta)^2 = \tfrac{1}{4}B^2(\mathbf{I}_\alpha - \mathbf{I}_\beta)^2 = \tfrac{1}{4}B^2(2\mathbf{I}_\alpha^2 + 2\mathbf{I}_\beta^2 - \mathbf{K}^2),$$

where $\mathbf{K} = (\mathbf{I}_\alpha + \mathbf{I}_\beta)$. Using $\mathbf{I}_\alpha^2 = \mathbf{I}_\beta^2 = I(I+1)$ we find

$$\langle JM|(b_\alpha - b_\beta)^2|JM\rangle = B^2 I(I+1)\left(1 - \frac{J(J+1)}{4I(I+1)}\right).$$

Now, previously we have established $\zeta\zeta' = -1$ so the quantity considered here is precisely the quantity needed for the structure factor (3.16) when it is summed over all J'. Note that the algebraic factor is positive, since it is the diagonal matrix element of the square of an operator, and it reduces the matrix element below the value appropriate to simple incoherent scattering by an isolated particle.

The value of the structure factor corresponding to Eq. (4.4), namely,

$$\sum_{J'\neq J} |F(J',J)|^2 = (\sigma_{\rm inc}/4\pi)|T_1 + \zeta T_2|^2\left(1 - \frac{J(J+1)}{4I(I+1)}\right), \tag{4.5}$$

depends on whether J is an even or an odd integer, the two possibilities giving opposite signs for $\zeta = (-1)^J$. Hence, the average of Eq. (4.5) with respect to J is to be made separately for J even and J odd.

The integer $J = 0,1,2,...,2I$, and the total number of initial spin states is

$$\sum_{J=0}^{2I} (2J+1) = (2I+1)^2.$$

Also,

$$\sum_{J=0}^{2I} (2J+1)\left(1 - \frac{J(J+1)}{4I(I+1)}\right) = \tfrac{1}{2}(2I+1)^2,$$

and the sum with J restricted to odd integers is found to be one-half this value. Thus, averages of the structure factor over J-even and J-odd states generate the same numerical factor $\frac{1}{4}$. Assembling the results, the intensity per particle at the recoil energy, summed over J' and averaged with respect to J, is

$$\frac{1}{2(2I+1)^2} \sum_J (2J+1) \sum_{J'\neq J} |F(J',J)|^2 = \frac{1}{4}\left(\frac{\sigma_{\rm inc}}{4\pi}\right) \tfrac{1}{2}(|T_1 - T_2|^2 + |T_1 + T_2|^2) = \frac{1}{4}\left(\frac{\sigma_{\rm inc}}{4\pi}\right)(|T_1|^2 + |T_2|^2). \tag{4.6}$$

This result is seen as our central finding. In arriving at the final expression we assume the single-particle orbitals in T_1 and T_2 are the same for all values of J.

For the combination of overlap integrals in Eq. (4.6) we submit the inequality

$$(|T_1|^2 + |T_2|^2) \leqslant 1. \tag{4.7}$$

The inequality follows by expressing $\psi(\mathbf{R})$ as an expansion in terms of complete sets of single-particle orbitals for the two sites. Equality in (4.7) is achieved when coefficients in the expansion are zero for all orbitals except those describing the ground state, denoted in Eq. (3.2) by $\varphi_1(\mathbf{R})$ and $\varphi_2(\mathbf{R})$.

The result (4.6) is smaller than the result corresponding to total scattering by an isolated particle $\overline{b^2} = (\sigma/4\pi)$, where $\overline{b^2}$ is given in Eq. (3.14) and σ is the total single-atom cross section. In part, the shortfall in intensity in Compton scattering is due to the absence of the initial wave function Eq. (3.2) in the final state; the Compton scattering process is inelastic and incoherent, and the cross section $\sigma_{\rm inc}$ appears instead of $\sigma \geqslant \sigma_{\rm inc}$. In our model result there is also a factor $\frac{1}{4}$ in the intensity that has its origin in the same physical process, which manifests itself in the calculation by the appearance in Eq. (3.16) of the difference in scattering-length operators for the two particles and no contribution to the scattering event from states with the same total nuclear spin J.

To conclude this section, let us return to a fact already mentioned, that the energy of the particles depends on their total spin. For a system consisting of only two identical particles, the solution of Schrödinger's equation for the spatial wave function that corresponds to the lowest eigenvalue has an even value of the total spin, since the wave function for this eigenvalue is not antisymmetrical. In the case of $I = \frac{1}{2}$ the structure factor per particle for this state is

$$\tfrac{1}{2} \sum_{J'\neq J} |F(J',0)|^2 = \tfrac{1}{2}(\sigma_{\rm inc}/4\pi)|T_1 + T_2|^2.$$

Taking $T_1 = T_2$, on the grounds that the two spatial centers have the same local structure, use of Eq. (4.7) brings us to the result ($I = \frac{1}{2}$),

$$\frac{1}{2} \sum_{J' \neq J} |F(J',0)|^2 \leqslant (\sigma_{\rm inc}/4\pi). \tag{4.8}$$

Consider next $I = 1$. The state of lowest energy corresponds to $J = 0$ or 2. The structure factor per particle satisfies

$$\frac{1}{2} \sum_{J' \neq J} |F(J',0)|^2 \leqslant (\sigma_{\rm inc}/4\pi) \tag{4.9}$$

and

$$\frac{1}{2} \sum_{J' \neq J} |F(J',2)|^2 \leqslant \frac{1}{4} (\sigma_{\rm inc}/4\pi) \tag{4.10}$$

while their average is less than or equal to $\frac{3}{8}(\sigma_{\rm inc}/4\pi)$.

V. COMPARISON WITH EXPERIMENTS

The intensity per particle in Compton scattering from two identical nuclei has been shown to be less than the single-atom incoherent cross section. There are various reasons why our favored expression for the intensity per particle is the result (4.6). For one thing, the result can be interpreted as the incoherent addition of intensity for each center, and this structure in the result is consistent with the incoherent nature of Compton scattering as a probe of matter. The result (4.6) is arrived at by including all the initial states with the appropriate quantum statistical weights. We expect this to be applicable because of the energy scales in the experiment. The separation in energy of the initial states is small, as we have previously mentioned, and surely the separation is very small relative to the temperature of the sample and, also, the spread in energy sampled in the experiments.

We will evaluate Eq. (4.6) with $T_1 = T_2$, which is an assumption consistent with our earlier use of a common value for the momentum wave function of the one-particle orbitals in the initial state. In this case, the intensity per particle σ_K relative to the single-atom cross section is

$$f = \sigma_\kappa / \sigma = \frac{1}{2} (\sigma_{\rm inc}/\sigma) T_1^2, \tag{5.1}$$

and we submit the inequality $T_1^2 \leqslant \frac{1}{2}$. One finds

$$f = 0.49 T_1^2 \quad \text{for the proton,}$$

and,

$$f = 0.13 T_1^2 \quad \text{for the deuteron.} \tag{5.2}$$

The main features of the experimental results [1–3] are the following.

(a) A big shortfall in the cross section for protons in metallic hydrides; about 30% in Nb-H and about 50% in Pd-H, when the neutron scattering time (observation time) is less than 5×10^{-16} s [1,2].

(b) A cross section of normal size for protons for time larger than 10^{-15} s in the Nb and Pd hydrides [1,2].

(c) A small (about 10%) but time-independent shortfall in the cross section for deuterons in Nb-D [2].

(d) A big shortfall in the H/D cross section ratio for mixtures of D_2O/H_2O [1]. This ratio is ≈30% below the conventionally expected one for admixtures $X_D = [D]/[H+D] < 0.4$, but approaches the conventional value for $X_D = 0.9$.

The values of f in Eq. (5.2) at once admit the entanglement of spatial and spin degrees of freedom in pairs of identical particles as a candidate for the explanation of the shortfall in the observed intensity for scattering by protons or deuterons. Taking $T_1^2 = \frac{1}{2}$, a fraction 0.4 of pairs of correlated protons yields a shortfall of 30% in the intensity relative to σ, and a fraction 0.7 of pairs gives a 50% shortfall [1,2].

With a purely quantum-mechanical effect as the explanation of the anomalies one expects progressively smaller anomalies with increasing mass of the particles. It is encouraging to find our proposal fits this trend. A fraction of only 0.1 of pairs of deuterons gives a 10% shortfall in the intensity, in line with the experimental result [2]. With more massive particles even fewer pairs of correlated particles should be formed; thus the attendant shortfall in intensity will be very small and, we propose, too small to be measured.

It can be shown that, if quantum correlations are deleted in the final state, by setting $\zeta' = 0$, the conventional cross section is recovered. The gradual transition from anomalous to normal cross sections as the scattering times are increased [1,2] can be seen as due to the destruction of entanglement in the states of the unit of two particles. Such decoherence is most likely associated with the interaction of the particles with the environment. Certainly, we expect the Compton event to destroy an entangled state enjoyed by the struck particle.

VI. CONCLUSION

We report a theoretical discussion of scattering of energetic neutrons by particles in a solid, with a view to interpreting recent experiments on samples loaded with protons or deuterons. A prime objective is a complete and transparent account of the influence on scattering of entanglement of the spatial and spin degrees of freedom of the particles. This is realized by recourse to a simple model built from elementary units of two particles (nuclei), which might capture essential features expected of a many-body quantum system. The initial and final states of the two particles in a unit are represented by nonrelativistic wave functions; a wave function is the product of a spin and an orbital wave function, and each of these is a linear combination of products of nonequivalent one-particle orbitals. An interchange of the two particles in the initial or final wave functions creates a phase factor $(-1)^{2I}$ where I is the magnitude of the spin of a particle.

The initial state in the scattering event is a state of equilibrium, and use of one product wave function to describe the initial state is expected to be an acceptable approximation to the true ground-state wave function. For the final state we use a wave function in which a particle is represented by a plane wave, and the second one-particle orbital is unspecified but assumed to be spatially localized. This highly excited state of a two-particle unit has no overlap with the initial state. We demonstrate that the specified final state of the two particles produces in the cross section the observed dependence on the energy transferred to the sample, which is a typical Compton profile centered at the recoil energy of one particle.

With regard to the interpretation of experiments, our key finding is a reduction of the cross section per particle below the cross section for a single isolated particle. The reduction is caused by entanglement of the spatial and spin degrees of freedom of the two particles in a unit.

In the favored model, two factors contribute to the calculated reduction in the cross section. First, orthogonality of the initial and final states means that these states have different total spins, so in the expression for the cross section the sum over all allowed values of the final total spin excludes the value of the initial total spin. Secondly, scattering involves only the (incoherent) spin-dependent part of the nuclear scattering-length operator. Absence of the spin-independent part of the scattering length, which is equal to the coherent scattering length, means the cross section calculated for a unit of two particles is a fraction of the incoherent single-atom cross section.

The model is shown to fit key experimental results. In so doing, it is found that there are fewer correlated pairs in the system of deuterons than in the system of protons. This finding is quite consistent with an explanation based on a purely quantum-mechanical effect. Another relevant example is the isotope effect observed in the localization of μ^+ and protons in metal hydrides [9(b)].

Persuasive as our argument appears, it might be casuistic. After all, in the properties of quantum many-particle systems there is abundant evidence of great subtleties.

Note added in proof. Recent papers by Fillaux [17] and Ikeda and Fillaux [18] discuss a related scattering problem in which protons participate in hydrogen bonds. We are grateful to François Fillaux for information about this work.

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