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LETTER TO THE EDITOR

Electronic correlation studied by neutron scattering

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Abstract

The use of inelastic low-energy neutron scattering from a many-electron target is envisaged as a tool for the investigation of electronic correlation. Specifically, numerical calculations are carried out to estimate the probability that an incoming neutron beam knocks out the nucleus of a helium atom, leaving behind two interacting continuum electrons. The signatures of the electronic correlation and of the electron–electron scattering dynamics in the calculated cross sections are discussed and the proposed method is contrasted with other existing many-particle spectroscopic tools.

For the investigation of electronic correlation and few-particle scattering dynamics in atoms and molecules a number of methods have been put forward (see the recent monograph [1] for an overview). The basic principle has been to prepare a system in a well defined state and to monitor its response to an external perturbation. The perturbation is usually induced by an impinging charged particle beam, e.g. electrons, positrons, protons, etc, or due to the absorption of photons. The advantage of these probing beams is that they are widely available and their properties can be controlled with sufficient accuracy. In addition, the coupling of these beams to atomic matter is sizeable so that the scattering cross sections are measurable with present day technology. On the other hand, one encounters several undesirable effects when it comes to extracting information exclusive to the target: a charged beam interacts with the constituents of atoms via the infinite-range Coulomb force, which implies that multiple scattering between the beam's particles and the excited atoms is generally strong and persists to very large distances. Therefore, it is, in general, not straightforward to disentangle from the measured scattering cross sections information that can be assigned purely to the target dynamic. This undesirable feature can be avoided, to a certain extent, by choosing appropriate scattering geometries, as specified below, or by utilizing single VUV photons to disturb the target. However, photons in the VUV regime provide only energy but no appreciable momentum to the target. In addition, only certain partial waves are excited due to the nature of the electric-dipole photon–electron interaction.

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This study explores the possibility of utilizing neutrons as a tool to investigate the electronic correlation in atoms. The idea is to switch off the nucleus by an instantaneous interaction that is transparent for the electrons [2]. After the nucleus is removed the initially bound electronic cloud fragments into individual interacting electrons that are then resolved asymptotically in energies and escape angles. There are two main sources of neutron-induced perturbations: the strong interaction with the nuclei and the magnetic interaction with existing (nuclear and electronic) magnetic moments of the target. In addition, there are a number of higher-order coupling terms that are described in [3] and will not be of relevance for the present study. The strong interaction potential V of neutrons with the nucleus is sizeable: however, its extension is much smaller than the wavelength of the neutron (we are looking at neutrons with energies less than 100 keV). Therefore, the potential V is well approximated by a contact, isotropic interaction. Within the Born approximation, its form is given by the Fermi pseudo-potential [4] as

$$V(\boldsymbol{r}_n) = g\delta(\boldsymbol{r}_n) \tag{1}$$

where $\boldsymbol{r}_n$ is the relative distance between the neutron and the nucleus. The coupling constant g is given by $g = b(2\pi\hbar^2/\mu_{n\alpha})$ where $\mu_{n\alpha}$ is the reduced mass of the neutron–nucleus system and b is the scattering length which depends on the specific nucleus and on the net nuclear spin. For the sake of simplicity we consider $\mathrm{He}(^1\mathrm{S}^\mathrm{e})$ as a target in which case the contributions of the two electrons to the magnetic scattering cancel each other and the nucleus (the α particle) has no net magnetic moment, i.e. the potential (1) is the only source of perturbation we consider for the neutron scattering from $\mathrm{He}(^1\mathrm{S}^\mathrm{e})$.

The aim of the present letter is to provide, within a realistic model, a numerical estimate of the cross section for the knock-out of the α particle and the subsequent emission of the two electrons with momenta $\boldsymbol{k}_1$ and $\boldsymbol{k}_2$.

When the neutron collides with the helium nucleus, the two electrons of He will always stick to the moving α particle if the momentum transfer is small. As we will see explicitly below, this is due to the small mass of the electron (as compared to that of the α particle). Therefore, events with very large momentum transfer are necessary to get the electrons off the nucleus. To catch these fine details in a theoretical description it is imperative to account for effects due to the finite nuclear mass. For a formulation in the centre-of-mass system we therefore utilize the set of Jacobi coordinates which are displayed in figure 1.

The fully differential cross section in the centre-of-mass system is given by [5] (atomic units (au) are used hereafter, i.e. $\hbar = 1 = a_0$ and the unit mass and unit charge are those of the electron)

$$\sigma(\boldsymbol{k}_1, \boldsymbol{k}_1, \boldsymbol{K}_n) = (2\pi)^4 \frac{\mu_{n\,T}}{K_{n0}} |\mathcal{T}|^2 \; \delta(E_i - E_f)\, \mathrm{d}^3\boldsymbol{k}_1\, \mathrm{d}^3\boldsymbol{k}_1\, \mathrm{d}^3\boldsymbol{K}_n. \tag{2}$$

Here $\boldsymbol{K}_{n0}$ and $\boldsymbol{K}_n$ are the initial and final channel momenta, respectively, that are conjugate to the coordinate $\boldsymbol{R}_n$ (cf figure 1) and $\mu_{n\,T} = (m_n(m_\alpha + 2))/(m_n + m_\alpha + 2)$ (where m_n and m_α are the masses of the neutron and the α particle, respectively). The momenta $\boldsymbol{k}_1$ and $\boldsymbol{k}_2$ are conjugate to $\boldsymbol{r}_1$ and $\boldsymbol{r}_2$, respectively. The initial and the final channel total energies are denoted by E_i and E_f. The transition matrix element $\mathcal{T}$ is given by

$$\mathcal{T} = g\left\langle \varphi_{\boldsymbol{K}_{n0}}(\boldsymbol{R}_n)\phi'(\boldsymbol{r}_1, \boldsymbol{r}_2, \boldsymbol{r}_{12})\, |\delta(\boldsymbol{r}_n)|\, \varphi_{\boldsymbol{K}_n}(\boldsymbol{R}_n)\phi(\boldsymbol{r}_1, \boldsymbol{r}_2, \boldsymbol{r}_{12})\right\rangle \tag{3}$$

$$= \frac{g}{(2\pi)^3}\left\langle \phi'(\boldsymbol{r}_1, \boldsymbol{r}_2, \boldsymbol{r}_{12})|\exp(-\mathrm{i}\boldsymbol{q}\cdot[\boldsymbol{r}_1/(m_\alpha + 1) + \boldsymbol{r}_2/(m_\alpha + 2)])|\phi(\boldsymbol{r}_1, \boldsymbol{r}_2, \boldsymbol{r}_{12})\right\rangle. \tag{4}$$

Here $\varphi_{\boldsymbol{K}}(\boldsymbol{R}_n)$ is a plane wave describing the neutron–target relative motion with a momentum vector $\boldsymbol{K}$ and $\boldsymbol{q} = \boldsymbol{K}_{n0} - \boldsymbol{K}_n$ is the momentum transfer vector. The initial bound state and the final continuum state of the helium atom are described by the wavefunctions $\phi(\boldsymbol{r}_1, \boldsymbol{r}_2, \boldsymbol{r}_{12})$ and $\phi'(\boldsymbol{r}_1, \boldsymbol{r}_2, \boldsymbol{r}_{12})$, respectively.

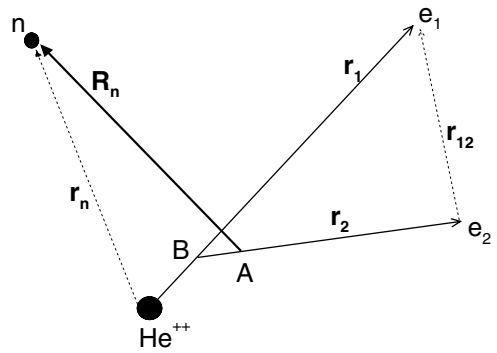

Figure 1. The set of Jacobi coordinates for the neutron–He system, as used in this letter. $\boldsymbol{r}_1$ is the relative coordinate between the electron labelled e_1 and the α particle. The relative coordinate between the centre-of-mass of this system (marked by $\boldsymbol{B}$) and the second electron (labelled e_2) is denoted by r_2. The relative position of the neutron with respect to the centre-of-mass of the He atom (A) is $\boldsymbol{R}_n$, whereas the relative distance of the neutron with respect to the α particle is denoted by $\boldsymbol{r}_n$.

Before evaluating numerically the integrals (4), the formal structure of the transition matrix element (4) deserves several remarks:

- It is clear from equation (4) that, if q is small, $\mathcal{T}$ becomes a direct overlap of different eigenstates of the same Hamiltonians and therefore vanishes. Therefore, the value of q must be of the order of m_α, i.e. the incident energy of the neutron has to be large enough and the neutron must recoil back, transferring its whole kinetic energy to the nucleus.
- The perturbation operator in equation (4) is a product of single-particle operators. Therefore, if the initial and final state are expressible in terms of independent single-particle orbitals, i.e. if the electronic correlation is neglected, the transition matrix element vanishes.
- For the double ionization by charged-particle impact, e.g. by electrons or charged ions, one obtains transition matrix elements that have a similar structure as shown by equation (4): however, this is the case only under the assumption that the projectile moves freely in the initial and final channels. Conventionally, this condition is justified for swift (relativistic) collisions with a small momentum transfer [5, 6], i.e. close to the optical limit. From a formal point of view the neglect of the distortion of the charged projectile motion during the collision is based on perturbative arguments whose range of validity is *a priori* not clear, in particular for Coulomb systems. In contrast, for neutrons equation (4) is valid in as much as the Fermi pseudo-potential (1), i.e. as long as the wavelength of the neutrons is appreciable (on the length scale of the strong force). This makes neutrons vital for investigating, in a non-perturbative way, collisions in atomic systems with very large momentum transfer. The price to be paid for this advantage is that the neutron scattering cross sections are very low, as detailed below.

The cross section (2) scales as the square of the scattering length which is of the order of a few fermis. Thus, one can expect the cross section for neutron scattering to be much smaller than for charged-particle or photon impact. For a more precise estimate of the cross section one can model the initial state wavefunction of helium $\phi(\boldsymbol{r}_1, \boldsymbol{r}_2, \boldsymbol{r}_{12})$ by a symmetrized three-parameter Hylleraas type wavefunction. The final three-body continuum state $\phi'(\boldsymbol{r}_1, \boldsymbol{r}_2, \boldsymbol{r}_{12})$ is described approximately by a three-body Coulomb (3C) wavefunction, which consists of a product of three Coulomb waves each describing the two-body scattering between the electrons and the α

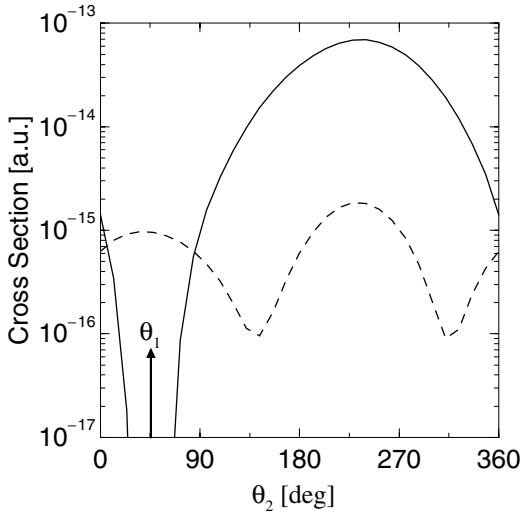

Figure 2. The cross section as a function of the emission angle θ_2 of one of the electrons with respect to the momentum transfer direction. The other electron is detected under a fixed angle $\theta_1 = 45^\circ$, as marked by the arrow. The two electrons have the same energy of 50 eV. The initial kinetic energy of the neutron–He relative motion is 10 keV. Upon collision with the nucleus, the neutron is reflected at an angle of 175° with respect to its initial direction. The full curve shows the full numerical calculations as described in the text whereas the broken curve is the result when the electron–electron final-state interaction is switched off.

particle and the mutual electron–electron scattering. Such a model yielded reasonable results for the double ionization by photon [7] and electron impact [8]. As we were dealing with fast ejected electrons (in the target frame) we did not attempt to employ more elaborate models that correct for some of the shortcomings of the 3C theory, for such corrections can be deemed marginal at higher escape energies of the electrons.

As stated above, for a sizeable cross section, one has to ensure high momentum transfer to the nucleus. Therefore, for the sake of illustration we consider the situation where the incoming neutron has a 10 keV incident energy (with respect to the target's centre-of-mass). Upon encounter with the nucleus the neutron is back-reflected at 175° with respect to the incidence direction. This case with a recoil velocity of about 0.3 au does not really correspond to a swift removal of the residual ion. However, as shown below, fingerprints of the initial-state electronic correlation can still be traced down. The advantage of choosing this 'adiabatic situation' is that the cross section is still sizeable (compared to a sudden removal of the nucleus) and the energies involved are well below the nuclear fragmentation threshold. Figure 2 shows the angular distribution of one of the electrons (electron 2) when the other electron is detected under an angle θ_1 with respect to $\hat{\boldsymbol{q}}$. Both electrons have the same energy of 50 eV. The most favourable situation for the two-electron emission is the back-to-back configuration. This is not only due to the electron–electron repulsion which leads to a vanishing electronic density of state around the fixed electron: if we switch off the final-state electron–electron interaction (broken curve in figure 2) (while maintaining the use of a correlated initial state), the cross section becomes smaller since electronic correlation is essential for the two-electron transition. However, the two electrons still prefer to recede in opposite directions, i.e. with zero total linear momentum of the electron pair. Roughly speaking, one can say after the α particle has been swept out by the neutron the electron pair is left behind without a confining

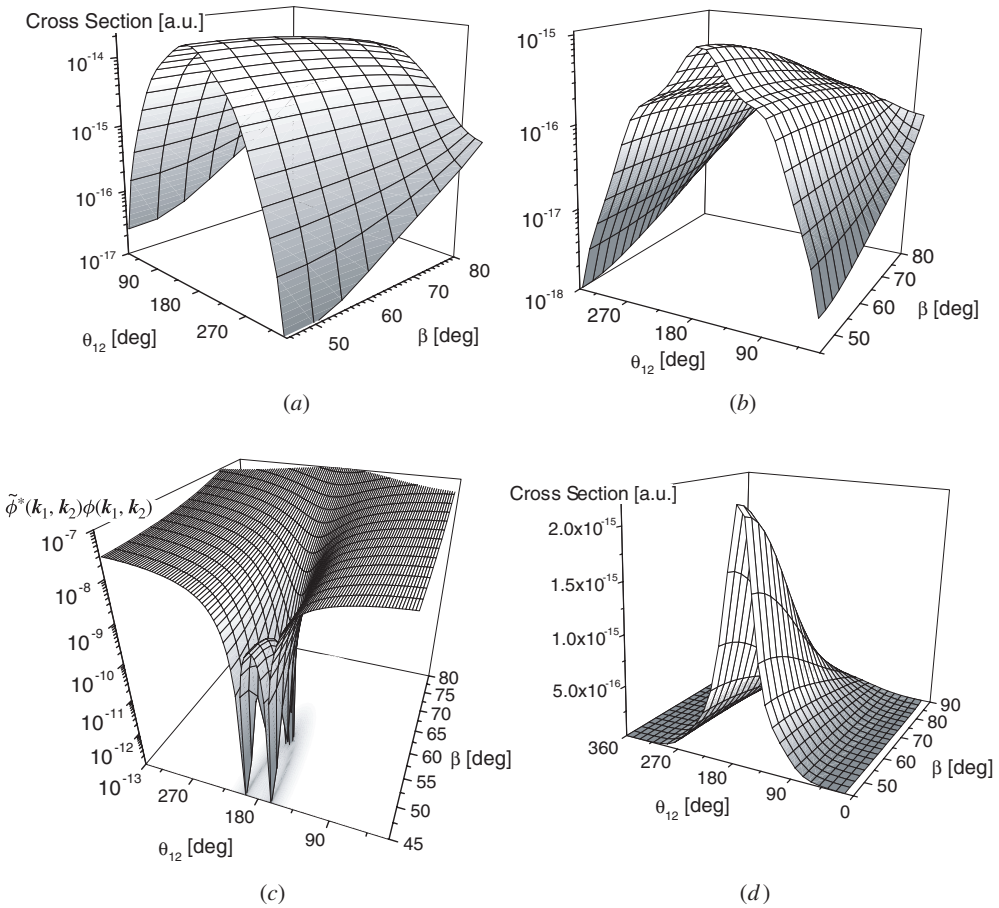

Figure 3. (*a*) The cross section as a function of the mutual electron–electron emission angle θ_{12} and of the angle $\beta = \tan^{-1}(k_1/k_2)$. The total energy of the electron pair is constant and equal to 170 eV. Both electrons are emitted with equal angles to the left and to the right of the incidence direction. The energy and the scattering angle of the neutron are as in figure 2. (*b*) The same as in figure 3(*a*): however, the total energy of the electron pair is increased to 400 eV. (*c*) For a total energy of 400 eV of the electron pair, the initial two-electron probability density is shown in the momentum space, i.e. $\tilde{\phi}^*(\boldsymbol{k}_1, \boldsymbol{k}_2)\phi(\boldsymbol{k}_1, \boldsymbol{k}_2)$ is shown as a function of the variables $\beta = \tan^{-1}(k_1/k_2)$ and $\theta_{12} = \cos^{-1}(\hat{\boldsymbol{k}}_1 \cdot \hat{\boldsymbol{k}}_2)$ that have been defined in figure 3(*a*). Here $\tilde{\phi}(\boldsymbol{k}_1, \boldsymbol{k}_2)$ is the double Fourier transform of the (position space) two-electron initial bound state used in figures 3(*a*) and (*b*). (*d*) The same as in figure 3(*b*): however, an initial state without angular electron–electron correlation has been employed.

potential and therefore fragments in the opposite direction. We recall that this fragmentation mode is forbidden for electric dipole transitions due to parity conservation, i.e. for small q/m_α (cf equation (4)) the angular distribution of figure 2 has a node instead of a peak for electron emission in opposite directions.

To trace the footprints of initial-state correlation in the cross section the following situation is considered in figure 3. The emission angles θ_1' and θ_2' of the two electrons are varied simultaneously with respect to the incident direction such that $\theta_1' = -\theta_2'$. For a fixed total energy $E_{\rm tot}$ of the electron pair we quantify the ($E_{\rm tot}$) energy sharing between the two escaping electrons by the angle $\beta = \tan^{-1}(k_1/k_2)$. In figure 3(*a*) $E_{\rm tot} = 170$ eV and the cross

section is shown as a function of the electron–electron mutual angle $\theta_{12} = \theta_1' - \theta_2'$ and β. As has been observed in figure 2, the two electrons propagate into the asymptotic region, most probably in opposite directions and with equal energies ($\theta_{12} = 180^\circ$, $\beta = 45^\circ$). As demonstrated by figure 3(b), this situation changes when the total energy of the pair is increased to $E_{\rm tot} = 400$ eV. Here we notice the appearance of additional shoulder structures whose origin can be pinned down by considering, as done in figure 3(c), the momentum-space two-electron probability density of the initial state which is given by $\tilde{\phi}^*(\boldsymbol{k}_1, \boldsymbol{k}_2)\tilde{\phi}(\boldsymbol{k}_1, \boldsymbol{k}_2)$. Here $\tilde{\phi}$ is the double Fourier transform of ϕ. The structures in the cross section depicted in figure 3(b) are readily comprehensible from figure 3(c) and from the fact that the final-state electron–electron interaction implies a vanishing two-electron density of state at $0^\circ = \theta_{12} = 360^\circ$. For completeness we show in figure 3(d) the cross section for the case of figure 3(b) evaluated with a Slater-type initial-state wavefunction. In this case $\tilde{\phi}^*(\boldsymbol{k}_1, \boldsymbol{k}_2)\tilde{\phi}(\boldsymbol{k}_1, \boldsymbol{k}_2)$ does not show any angular dependence and consequently the cross section possesses only one peak when the electrons escape with zero total linear momentum $\boldsymbol{k}_1 = -\boldsymbol{k}_2$.

In conclusion, we considered theoretically the possibility of knocking out the nucleus from an atomic target by means of the strong nuclear force exerted by an incoming neutron beam. To utilize this reaction for the study of the electronic correlation we investigated the cross section for the emission of two electrons, and proposed and implemented a numerical method for the evaluation of the cross sections. The purpose of this letter has been to provide, at least theoretically, a rough picture of the physics one may expect in such a process. Clearly, the absolute values of the fully differential cross sections in the situations we discussed in this letter are far too small to be detected at present. However, over the past few years impressive progress has been made in multi-particle coincidence techniques [9, 10] and in neutron beam treatment which gives hope for the future realization of the proposed experiments.

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