Spin Anisotropy for Excitation in Collisions between Two One-Electron Atoms

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The singlet and triplet contributions of excitation cross sections are studied theoretically for collisions between various two one-electron atoms. The spin anisotropy is shown to have a general behavior in the important impact energy range. At low energies triplet cross sections dominate completely over the singlet ones while the opposite is true when the active electron and projectile velocities are comparable. Beyond the matching velocity regime singlet and triplet contributions become identical. We propose a general dynamical interpretation based on the analysis of the time dependency of the electron probability density and probability current density.

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While the bound state two-electron problem to a high accuracy was solved by Hylleraas [1] more than 70 years ago, scattering problems remain challenging in atomic physics today. Even for the simplest systems it is awkward to achieve convergence in time-dependent and time-independent nonperturbative approaches. Calculations using close-coupling methods or direct numerical integration of the time-dependent Schrödinger equation (TDSE) can be practically carried out only on high speed parallel supercomputers. For example, the implementation of cunning boundary conditions within the close-coupling approach has only recently given excellent agreement with experimental results for differential cross sections in $(e, 2e)$ reactions [2]. Achievements have also been made in double ionization of He atoms by heavy (anti-)particles [3], photons [4], and also by strong pulsed laser field [5] (for H^- ionization, see [6]). A common feature of these studies, apart from inelasticity in the electronic degrees of freedom, is the central role played by the *electron correlation*.

From a theoretical viewpoint, heavy particle collisions present an elucidating simplification due to the short wavelength of the heavy particle relative motion which allows for a semiclassical description [7], and thereby introduces an external clock into the scattering problem [8]. Then the important features of the two-electron systems, the electron correlation and spin symmetry, may be studied dynamically by the time dependency of the electronic wave function, as previously used for one-electron systems, e.g., [9].

Within the Schrödinger formalism the total electronic spin is conserved. This divides the theoretical treatment into two separate problems which needs to be solved independently, i.e., singlet and triplet cases for general two-electron systems. So far only scattering experiments with unpolarized atom beams have been performed and, for a given electronic transition, only spin averaged cross sections are available. The confrontation between theory and experiment can then allow only partial understanding of the mechanisms responsible for the transition. Note that spin resolved experimental data are available for electron-atom scattering, cf., for example, [10].

In the present Letter we report results on excitation in collisions involving simple one-electron atoms, H(1s)-H(1s), He^+(1s)-He^+(1s), and H(1s) $-$ He^+(1s). We limit ourselves to excitation of the target, but identical conclusions can be drawn for excitation of both partners. It is interesting to note that a remarkable agreement with experiments has been obtained by one-electron models for certain such systems [11,12]. In these models the TDSE is solved for one "active" electron while the other electron is assumed "passive" and represented by an effective potential [13]. In some cases, this approach seems to have given even better agreement than full two-electron calculations [11]. This would be a true "mind-boggling" result if both calculations were complete, in the sense of spanning the full Hilbert space of each model system. The complexity of the two-electron problem is, however, so large that it is a long computational way to go to reach completeness. We believe, however, that our present calculations span the important part of the Hilbert space and that the excitation dynamics is therefore accurately described. Atomic units ($\hbar = m_e = e = 1$) are used unless stated explicitly.

The present work is based on the solution of the Schrödinger equation $[H - i\frac{\partial}{\partial t}]\Psi(\vec{r}_1, \vec{r}_2, \vec{R}(t)) = 0$ where H is the electronic Hamiltonian. Time is introduced through the internuclear coordinate, $\vec{R}(t) = \vec{v}t + \vec{b}$, which describes a straight line trajectory. The spatial part of the wave function is expanded as

$$\Psi(\vec{r}_1, \vec{r}_2, \vec{R}(t)) = \sum_{i,j} c_{ij}^{TT}(t)\Psi_{ij}^{TT}(\vec{r}_1, \vec{r}_2) + \sum_{i,j} c_{ij}^{PP}(t)\Psi_{ij}^{PP}(\vec{r}_1, \vec{r}_2, \vec{R}(t)) + \sum_{i,j} c_{ij}^{TP}(t)\Psi_{ij}^{TP}(\vec{r}_1, \vec{r}_2, \vec{R}(t)), \quad (1)$$

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where $T(P)$ refers to target (projectile) centered spin symmetrized single atom states, including electronic translational factors [14]. Note that this wave function describes not only single excitation, but also double excitation and electron transfer. By varying the basis size we have checked that the results are converged.

In Fig. 1 we compare our calculated spin averaged cross sections for $2s$ excitation in H($1s$)-H($1s$), $He^+(1s)$-$He^+(1s)$, and $He^+(1s)$-$H(1s)$ collisions with the most recent experiments and calculations. For H-H the one-electron calculations of Riley and Ritchie [12] are in very good agreement with the experimental data and our two-electron results. Their underestimate at high energies is easily understood from the fact that double-atom excitation dominates above 30 keV [15], and that process cannot be directly calculated in a one-electron model. For $2s$ excitation in He^+-He^+ collisions we have found no other calculations and experimental results. In this system, compared to H-H, we note a rather similar behavior of the cross sections, shifted to higher energies due to the much stronger endoergodicity of the process. The small peak around 50 keV/amu is due to interference with electron capture [16]. For He^+-H collisions we compare with experiments, a two-electron calculation of Ermolaev *et al.* [17] and a one-electron calculation by Kuang *et al.* [11]. It is here seen that the present two-electron calculations are in better agreement with the one-electron results [11] (and experiments) than with the calculations in [17].

If closing at this point one could be led to conclude that the one-electron model not only produces accurate cross sections, but also describes the *dynamics* correctly. A disproof of the latter is seen by introducing, for a given transition $i \rightarrow f$, a spin anisotropy parameter

$$A_S = \frac{\sigma_{i\rightarrow f}^{S=1} - \sigma_{i\rightarrow f}^{S=0}}{\sigma_{i\rightarrow f}^{S=1} + \sigma_{i\rightarrow f}^{S=0}}. \qquad (2)$$

In Fig. 2 this parameter is plotted with respect to the reduced velocity v/v_e [18] for the three $2s$ excitation processes considered above. We observe first that this parameter behaves similarly for all three systems at low velocities where triplet excitation dominates. For all systems singlet excitation becomes increasingly more important with increasing velocity until it dominates for $v \sim 1$ for H-H and He^+-He^+. The He^+-H system behaves slightly differently in this intermediate energy region for two reasons: (i) this system has a dominant capture to the $He(1s^2)$ ground state around $v/v_e \sim 1$ ($E \sim$ 25 keV/amu) which interferes strongly with the excitation channels and (ii) much larger impact parameters are important. At asymptotic high velocities the triplet/singlet asymmetry vanishes for all excitation channels, as seen in Fig. 2. Note that for the electron transfer channel the corresponding asymptotic value is $A_S = -1$ since the two-electron ground state is singlet.

The dominance of singlet excitation at intermediate energies and triplet excitation at low energies is well known from electron-atom collisions [10]. In heavy particle collisions this general behavior has not been noted earlier. In fact, we are aware of only very few works (e.g., [19,20]) which have reported singlet and triplet cross sections separately. One of these works is in disagreement with

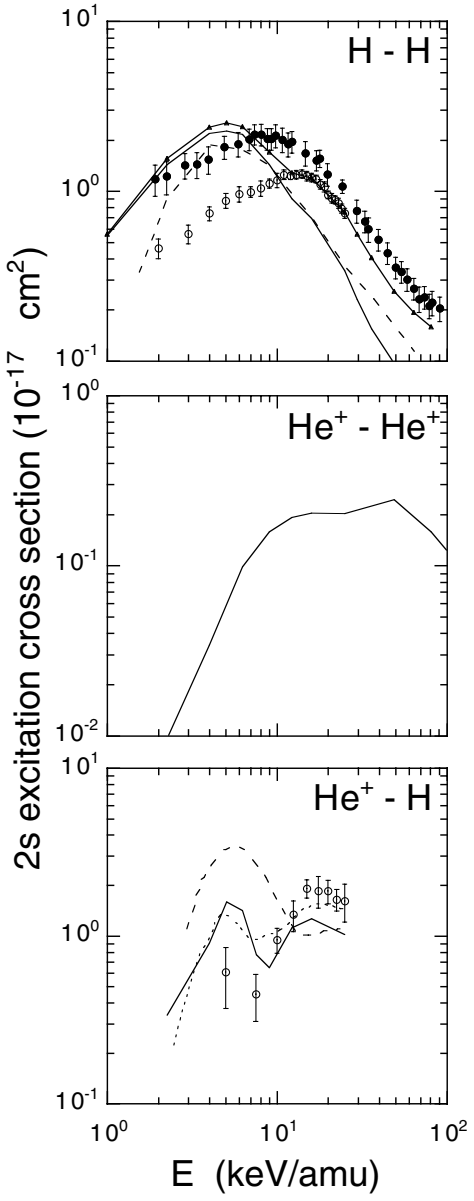

FIG. 1. Cross sections for $2s$ excitation for three different collision systems. Upper panel, H($2s$) excitation in H($1s$)-H($1s$) collisions: solid line, spin averaged two-electron calculations of Hansen and Dubois (1998) [15] (the solid line marked with triangles shows double-atom excitation); dashed line, one-electron calculations of Riley and Ritchie (1999) [12]; experimental data of Hill *et al.* (1979) [23] (○) and of Morgan *et al.* (1980) [24] (●). Middle panel, $He^+(2s)$ excitation in $He^+(1s)$-$He^+(1s)$ collisions: solid line, present spin averaged two-electron calculations. Lower panel, H($2s$) excitation in $He^+(1s)$-H($1s$) collisions: solid line, present spin averaged two-electron calculations; dashed line, spin averaged two-electron calculations of Ermolaev *et al.* (1994) [17]; dotted line, one-electron calculations of Kuang *et al.* (1995) [11]; ○, experimental data of Geddes *et al.* (1994) [25].

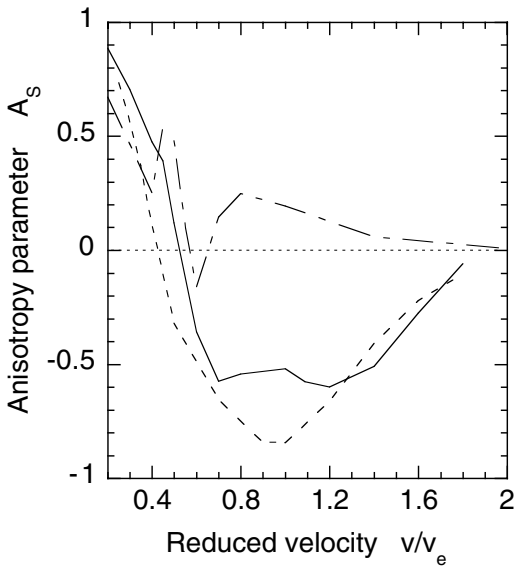

FIG. 2. Spin anisotropy parameter A_s for H($2s$) excitation vs reduced velocity v/Z_T for the three systems considered in Fig. 1. Solid line: H($1s$)-H($1s$); dashed line: He^+($1s$)-He^+($1s$); chain line: He^+($1s$)-H($1s$).

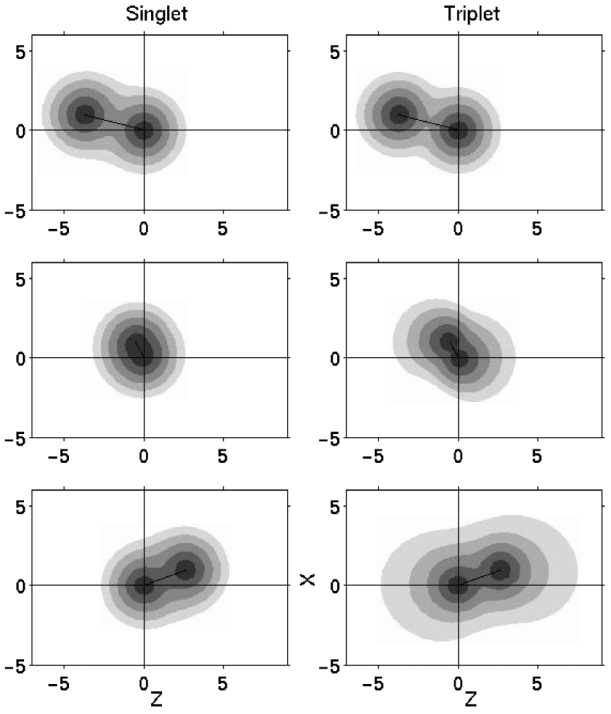

FIG. 3. Snapshots of the H($1s$)-H($1s$) one-electron density for singlet collisions (left) and triplet collisions (right) for impact velocity $v = 0.4$. The densities are computed in the collision plane, i.e., with internuclear vectors $\vec{R}(t) = [b, 0, vt] = [1, 0, -4.0]$ (top), $\vec{R}(t) = [1, 0, -0.5]$ (middle), $\vec{R}(t) = [1, 0, 3.0]$.

our results [19], while the most recent one by Borondo *et al.* [20] agrees well with the general results presented in Fig. 2.

The detailed dynamics will now be displayed by studying the one-electron probability density (charge cloud) and the electronic current density. The charge cloud related to the electron at coordinate $\vec{r}_1$ is given by

$$\rho(\vec{r}_1, \vec{R}) = \int d^3r_2\, \Psi^*(\vec{r}_1, \vec{r}_2, \vec{R}(t))\Psi(\vec{r}_1, \vec{r}_2, \vec{R}(t)). \quad (3)$$

For the H-H system, three characteristic snapshots of the charge cloud [21] in the (xz) collision plane are shown in Fig. 3 at $v = 0.4$ and for a typical impact parameter $b = 1.0$; see, e.g., [15]. Initially the triplet and singlet charge clouds look rather identical since they consist of two well separated H($1s$) states. We note that the triplet density is not exactly zero at the midpoint between the two nuclei (as it would be in a static example) due to the electron translational factors. In the singlet case a very located charge cloud is created when the two atoms are close by. On the other hand, the triplet charge cloud is seen to reach a larger spatial extension, which can be attributed to Pauli repulsion. This causes excitation to be much more effective at low energies.

The dynamics may alternatively be exposed by constructing the one-electron probability current density

$$\vec{j}(\vec{r}_1, \vec{R}) = Re\Big[\int d^3r_2\, \Psi^*(\vec{r}_1, \vec{r}_2, \vec{R}(t)) \times (-i\vec{\nabla}_1)\Psi(\vec{r}_1, \vec{r}_2, \vec{R}(t)) \Big], \quad (4)$$

which is obtained by time differentiation of the probability density. In Fig. 4 snapshots of the x-z component of this current for singlet and triplet collisions are compared at the same instants of time as in Fig. 3. We note for both collisions that the horizontal flux related to the projectile motion is dominating [22]. Zero flux is present initially on the target since the flux related to a (stationary) real state is zero.

In the singlet case, as the nuclei approach each other, the attraction causes a flux towards the projectile. For low velocities the adiabaticity allows for a return flux after the turning point as seen in the lowest left panel. At higher energies the situation is opposite: the charge localization caused by this flux allows for an effective "kick" of the electron to an excited state. This kick becomes effective enough to allow for singlet dominance of the cross section. These two mechanisms identified for H-H collisions are present for He^+-He^+ and He^+-H. However, for the He^+-H system at intermediate energies, the current snapshots show also clearly the very strong capture process to the He($1s^2$) state, as previously mentioned. This coupling perturbs sufficiently the mechanisms described above to change the general behavior of the spin anisotropy parameter; cf. Fig. 2.

In conclusion, we have demonstrated that excitation in collisions between two one-electron atoms have a strong sensitivity to the electronic total spin and that one-electron models therefore describe only the dynamics in an average sense. Our calculations indicate that the spin anisotropy follows a general behavior with respect to scaled velocity.

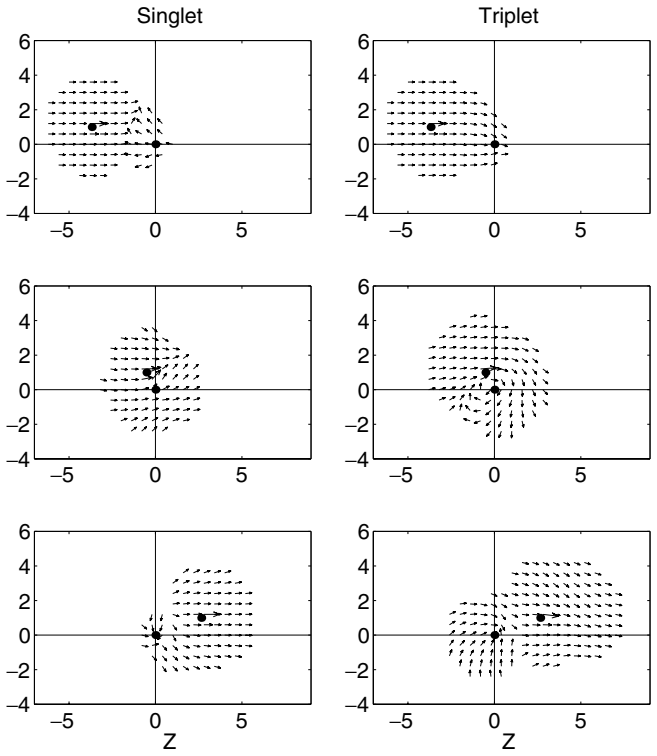

FIG. 4. Snapshots of the H($1s$)-H($1s$) one-electron current for singlet collisions (left) and triplet collisions (right). Only the x-z component of the current is shown for the same internuclear distances as in Fig. 3.

Studies of the time-dependent charge cloud and current density have identified two different dynamical mechanisms responsible for the excitation.

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