Suppression of correlated electron escape in double ionization in strong laser fields

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The effect of the Pauli exclusion principle on double ionization of He atoms by strong, linearly polarized laser pulses is analyzed. We show that correlated electron escape, with electron momenta symmetric with respect to the field polarization axis, is suppressed if atoms are initially prepared in the metastable state $^3S$. The effect is a consequence of selection rules for the transition to the appropriate outgoing two-electron states. We illustrate the suppression in numerical calculations of electron and ion momentum distributions within a reduced dimensionality model.

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The interaction of atoms with strong laser pulses of intensities in the range of $10^{14} \text{W/cm}^2$ may result in the production of doubly charged ions. Early experiments indicated that the rates for this process are much higher than expected on the basis of a single active electron model (see [1] and references therein). The commonly accepted explanation is based on the rescattering process, where a temporarily ionized electron is driven back to the atom by the electric field [1, 2]. The acceleration by the field provides additional energy which can then be transferred to the second electron during the collision with the residual ion. Measurements of the doubly charged ion and electron momentum distributions, in addition to the total ionization yield, [1, 3, 4] revealed that substantial number of the electrons escapes with equal momenta along the field polarization axis. The origin of this correlation was analyzed as a consequence of the Coulomb repulsion between the outgoing electrons, their Coulomb attraction to the nucleus and the pulling force from the electric field [5]. Except for the presence of the field during the decay of the complex formed in the rescattering this analysis has many parallels to Wanniers reasoning for double ionization after collision or photo excitation. Since in the Wannier case it can be shown that the observed cross sections are constrained by the symmetries of the problem [8–11], we here search for the corresponding effects in strong field double ionization.

Consequences of the exchange symmetry on double ionization yield have previously been studied [12, 13] within the aligned electron model [14]. These authors found an reduction in ionization rate in the antisymmetric state, but overestimation of the Coulomb electron repulsion in the aligned electron model did not allow them to treat the final momentum distributions. These studies were motivated by experimental observation of the difference in single and double ionization of O\textsubscript{2} and N\textsubscript{2} molecules where the initial states are of triplet and singlet character, respectively [15]. Recent experiments [16, 17] observed differences between O\textsubscript{2} and N\textsubscript{2} molecules when the electron momentum correlations for components parallel to the light polarization axis were measured.

The constraints on the double ionization cross section arise from the symmetries of the wave functions for the outgoing electrons and can be derived from the study of the selection rules for transitions to two electron continuum states by Maulbetsch and Briggs [11]. They analyze transition matrix elements from an initial state $\psi$ to continuum final states of well defined electron momenta $\vec{k}_1$ and $\vec{k}_2$, i.e., $\langle \vec{k}_1\vec{k}_2|T|\psi \rangle$ where $T$ is the appropriate transition operator. As $r_1$, $r_2$ and $r_{12} = |\vec{r}_1 - \vec{r}_2|$ tend to infinity, the continuum states for a two electron case of He approach the asymptotic behaviour $\langle \vec{r}_1\vec{r}_2|\vec{k}_1\vec{k}_2 \rangle \rightarrow (2\pi)^{-3} \exp(i\vec{k}_1 \cdot \vec{r}_1 + i\vec{k}_2 \cdot \vec{r}_2 + i\chi)$, where the phase $\chi = 2 \ln(k_1r_1 + \vec{k}_1 \cdot \vec{r}_1)/k_1 + 2 \ln(k_2r_2 + \vec{k}_2 \cdot \vec{r}_2)/k_2 - \ln(k_{12} + \vec{k} \cdot \vec{r}_{12})/(2k)$ with $k = (\vec{k}_1 - \vec{k}_2)/2$ accounts for the long range of Coulomb potentials. These states and the full transition matrix elements can now be projected onto a complete set of two electron states characterized by total angular momentum $L$, its projection $M$ on a chosen quantization axis $z$, total spin $S$ and any other necessary quantum numbers, collectively labelled $\alpha$.

$$\langle \vec{k}_1\vec{k}_2|T|\psi \rangle = \sum_{\alpha LMS} \langle \vec{k}_1\vec{k}_2|\alpha LMS \rangle \langle \alpha LMS|T|\psi \rangle. \quad (1)$$

Selection rules are now obtained for sets of $\vec{k}_1$, $\vec{k}_2$, $L$, $M$ and $S$ for which the overlaps $\langle \vec{k}_1\vec{k}_2|\alpha LMS \rangle$ vanish identically. Various situations are discussed in [11]. The case most appropriate for correlated double escape with equal energy and equal momenta parallel to the field axis is their case (I), where $k_1 = k_2$, both momenta lie in a plane, i.e., $|\phi_1 - \phi_2| = \pi$, and their projections on the quantization axis coincide, $\theta_1 = \theta_2$: then states with $(S + M)$ odd do not contribute to the transition matrix elements. Therefore, if the initial wave function belongs to this subspace and if the evolution operator preserves this subspace, these final states will be suppressed.

For the case of strong field double ionization the transition operator $T$ is the evolution operator corresponding
to the Hamiltonian (in atomic units)

$$H = \sum_{i=1}^{2} \left( \frac{p_{i}^2}{2} - \frac{Z}{r_i} \right) + \frac{1}{|r_1 - r_2|} + F(t)(z_1 + z_2), \tag{2}$$

where $F(t)$ contains the time dependence of a linearly polarized laser pulse. It is symmetric under exchange of electrons. Therefore, if the initial wave function belongs to the $^3S$ state of He, the sum in (1) remains restricted to $M = 0$ and $S = 1$, since both the $z$ component of the total angular momentum and the spin operators commute with the Hamiltonian. As a consequence, the matrix element of the transition operator between the initial metastable state and continuum states with $\vec{k}_1$ and $\vec{k}_2$ symmetric with respect to the field polarization axis vanishes by the above selection rule. Thus, the differential cross section for the double ionization process is exactly zero whenever the momenta of outgoing electrons are symmetric with respect to the field axis. Note that in experiments starting with the He ground state, i.e. $^1S$, and in the double ionization of noble gases the symmetric escape is not forbidden — correlated electron escapes are clearly visible in electron momentum distributions in the double ionization of Ne and Ar atoms [1, 3, 4].

On the level of the classical dynamics of the two-electron system, the selection rule eliminates a subset of phase space that includes the $C_{2v}$ subspace of symmetric electron motion, which has been shown to capture much of the observed momentum distributions [5]. In cylindrical coordinates, the $C_{2v}$ subspace is defined by $R = r_1 = r_2$, $p_R = 2p_{\rho_1} = 2p_{\rho_2}$, $Z = z_1 = z_2$, $p_Z = 2p_{\phi_1} = 2p_{\phi_2}$, $|\phi_1 - \phi_2| = \pi$ and $p_{\phi_1} = p_{\phi_2} = 0$. The Hamiltonian (2) in the $C_{2v}$ subspace reduces to

$$H = \frac{p_R^2 + p_Z^2}{4} - \frac{4}{\sqrt{R^2 + Z^2}} + \frac{1}{2R} + 2ZF(t). \tag{3}$$

The potential energy in (3) (and obviously also the potential energy in (2)) possesses a saddle located at $R = R_s \sin \theta_s$, $Z = R_s \cos \theta_s$, where $R_s = 3^{1/4}/\sqrt{|F(t)|}$ and $\theta_s = \pi/6$ or $5\pi/6$ depending on the sign of $F(t)$. Electrons moving in the subspace may simultaneously escape from the atom by going over the saddle. Trajectories living in the full space that approach the saddle sufficiently symmetrically may lead to simultaneous escape of electrons with highly correlated final momenta [5]. For the static electric field (i.e. $F(t) = \text{const}$) the stability analysis of the saddle allows one to estimate the energy dependence of the classical cross section for correlated double escape for energy close to the saddle energy, i.e. to obtain a counterpart of the Wannier threshold law in the presence of the electric field [18]. Note that for the energy equal to the threshold value the only trajectory leading to the double escape corresponds to two electrons moving symmetrically in the same direction along the field axis. This is very different from the original Wannier problem where the electrons escape symmetrically but in opposite directions. The analysis of the quantum selection rule for double ionization of the metastable He atoms by linearly polarized laser pulses implies that, in terms of classical mechanics, this entire $C_{2v}$ symmetric subspace is forbidden for electrons.

With perfectly symmetric escape excluded, we have to look for escape with almost symmetric electrons. Relaxing the condition on the angular degrees of freedom, i.e. by allowing for arbitrary $\phi = \phi_1 - \phi_2$ but $p_\phi = 2p_{\phi_1} = -2p_{\phi_2}$ (i.e. $M = 0$), we are led to consider the invariant subspace of $C_v$ symmetry (the $C_v$ subspace contains obviously the $C_{2v}$ subspace) [5]. Then the Hamiltonian of the system reads

$$H = \frac{p_R^2 + p_Z^2}{4} + \frac{p_{\phi}^2}{R^2} - \frac{4}{\sqrt{R^2 + Z^2}} + \frac{1}{2R|\sin (\frac{\phi}{2})|} + 2ZF(t). \tag{4}$$

Comparing (4) with (3) we see that in the present case electrons can perform a symmetric bending motion around the field axis. In the $C_v$ subspace electrons may pass over the saddle considered previously and ionize but their momenta need not lie in the plane $|\phi_1 - \phi_2| = \pi$. In the quantum description there is no selection rule for electrons with $k_1 = k_2$ and with momenta that have the same projections on the polarization axis, but with $|\phi_1 - \phi_2| \neq \pi$.

The consequences of the selection rule should show up most clearly in angle resolved double ionization cross sections, as they result in a zero for the symmetric momenta and an azimuthal angle difference of $\pi$. However, it will also show up as a reduction in intensity in the angle integrated cross sections, like the ones where only the parallel momenta components of ionizing electrons are measured [1, 3]. We will demonstrate this with calculations in 1+1-dimensional model below, but we emphasize that the clearest verification is possible with fully momentum resolved cross sections as obtained in recent experiments [4].

Full three-dimensional (3D) calculations of the double ionization process are barely feasible [19, 20], so we take recourse to low-dimensional models. Aligned electron models [14] are not suitable for our purpose since there the symmetric simultaneous escape of electrons is forbidden by the overestimated electron repulsion. The 1+1-dimensional (1+1D) model proposed in [21] does allow for symmetric escape by confining the electrons to move along one-dimensional (1D) tracks that form angles $\pi/6$ and $5\pi/6$ with respect to the field polarization axis. The Hamiltonian of the system, in atomic units, reads

$$H_{1,D} = \sum_{i=1}^{2} \left( \frac{p_{R_i}^2}{2} + \frac{Z}{|R_i|} \right) + \frac{1}{\sqrt{(R_1 - R_2)^2 + R_1 R_2}} + \frac{F(t)\sqrt{3}}{2} (R_1 + R_2), \tag{5}$$

where $R_1$ and $R_2$ denote positions of electrons on the chosen tracks and $p_{R_1}$ and $p_{R_2}$ are conjugate momenta. In this model the rescattering process, single and sequential
double ionization are present but importantly also the symmetric simultaneous escape of electrons can be properly simulated [22]. Indeed, in the 3D case, there is a single saddle around which two electrons can escape simultaneously and that saddle possesses two unstable directions. The 1+1D model also has a saddle located at the same place as in the 3D case, \( R_1 = R_2 = -\text{sgn}(F(t))R_s \), also with two unstable directions. Hence, the key topology elements of the phase space of the 3D case are properly reproduced in the model. The symmetric subspace of the classical phase space in the model is defined by \( R_1 = R_2 \) and \( p_{R_1} = p_{R_2} \). If, in quantum simulations, we start with the ground state of the unperturbed system symmetric electron escape is not forbidden. However, if we chose as an initial state the first excited state, which is antisymmetric with respect to electron exchange (in analogy to the \(^1\text{S}\) state) the wavefunction vanishes at \( R_1 = R_2 \). The symmetric subspace is then forbidden.

We have performed numerical integration of the Schrödinger equation corresponding to the Hamiltonian (5) with Coulomb singularities smoothed by the replacement \( 1/x \rightarrow 1/\sqrt{x^2 + \epsilon} \) with \( \epsilon = 0.6 \), starting with the ground and first excited states of the unperturbed Hamiltonian. The ground state energy, for the chosen parameter \( \epsilon \) is \(-2.83\) while the first excited state is at \(-2.21\). We have used a single cycle pulse of the form \( F(t) = F_0 f(t) \sin(\omega t) \), where the pulse envelope \( f(t) = \sin^2(\omega t/2) \), with \( \omega = 0.06 \) and \( F_0 = 0.15 \). Single ionization in the case of the initial first excited state is very efficient (with probability equal to 0.98) and much more probable than in the case of the ground state (with probability \( 10^{-4} \)). That is due to the higher energy of the first excited state, which allows electrons to easily pass over the Stark barrier and to escape near the peak intensity of the field. Probabilities for double ionization are comparable in both cases, about \( 10^{-6} \), thus indicating that the difference in initial energy does not have an excessively strong effect on double ionization.

Figure 1 shows electron momentum distributions in the double ionization of He for the initial ground and first excited states. The distributions have been obtained adopting the method proposed in [23] as described in [22]. The momentum distributions are obtained by Fourier transforms of parts of the final wavefunctions in the regions \( |R_1|, |R_2| > 50 \) and are averaged with a Gaussian of width 0.07 in order to model experimental resolution. The time-dependance of the field is \( F(t) = F_0 f(t) \sin(\omega t) \) where \( F_0 = 0.15 \), \( \omega = 0.06 \) and one-cycle pulse duration with an envelope \( f(t) = \sin^2(\omega t/2) \). The concentration of the outgoing momenta in one quadrant is a result of the single cycle laser pulse. The low intensity along the diagonal in panel (b) is the signature of the suppression of symmetric double escape by the selection.

The signatures of the effect can be enhanced, as shown in Fig. 2a, by considering the distributions of the momenta differences, \( p_{R_1} - p_{R_2} \). One finds a strong maximum at \( p_{R_1} - p_{R_2} = 0 \) for double ionization starting from the symmetric ground state and a profound minimum when the initial state is the antisymmetric first excited state. Note again that the minimum in Fig. 2a does not reach zero because of the averaging. But Fig. 2a clearly demonstrates that the resolution attainable in current experiments is sufficient to observe effects of the selection rule we analyze here.

In Fig. 2b the distributions of momenta sum (equivalent to ion momentum distributions), i.e. \( p_{R_1} + p_{R_2} \), in double ionization of He atoms are plotted. We see that the momenta sum distribution shows no simple signature.
FIG. 2: The probability distributions for differences, $p_R_1 - p_R_2$, [panel (a)], and for sums, $p_R_1 + p_R_2$, [panel (b)], of electron momenta for double ionization in a strongly linearly polarized laser field. Data for initial ground state (red dashed curves) are compared with those obtained for the initial first excited state (solid black lines). The minimum in the difference distribution in panel (a) reflects the suppression of symmetric escapes. All parameters are the same as in Fig. 1.

of the absence of highly correlated electron escape in the case of the initial first excited state except, possibly a small narrowing of the distribution. By comparison, the existence of correlated pairs of electrons for the initial ground state case broadens the corresponding sum distribution (dashed line). We also would like to point out that the asymmetry of the ion momentum distribution seen in Fig. 2b reflects the carrier-envelope phase as expected [24].

Finally, we would like to stress that the numerical results presented are obtained in the reduced dimensionality model where the 1D electron tracks are chosen in two-dimensional space containing the polarization axis. Direct application of the reduced 1+1D model to experimental data is thus possible only if events with electron momenta lying in a plane that contains the field polarization axis are collected only.

We have shown that the selection rules of Maulbetsch and Briggs [11] imply a difference in the final momenta distributions in double ionization by strong linearly polarized laser pulses depending on the symmetries of the initial state. For double ionization in He starting from the metastable state 3S correlated electron escape with final electron momenta symmetric with respect to the polarization axis is suppressed. Numerical simulations in a reduced dimensionality model show a clear dip in the momentum distributions, and demonstrate that the effect appears despite differences in the initial energy. The effect should be within reach of todays experiments with their resolution and detailed kinematic characterization of the final state. Extensions to double ionization in molecules are immediate. A prime candidate are O2 molecules with triplet initial state oriented along the polarization axis. Recent experiments with aligned N2 molecules [17] probed a singlet initial state only.

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