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

Bruno Eckhardt,[1] Jakub S. Prauzner-Bechcicki,[2] Krzysztof Sacha,[3] and Jakub Zakrzewski[3]

[1]*Fachbereich Physik, Philipps-Universität Marburg, D-35032 Marburg, Germany*
[2]*Faculty of Conservation and Restoration of Works of Art, Academy of Fine Arts, Smoleńsk 9, Kraków, Poland*
[3]*Marian Smoluchowski Institute of Physics and Mark Kac Complex Systems Research Centre, Jagiellonian University, ul. Reymonta 4, 30-059 Kraków, Poland*

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} 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 a substantial number of the electrons escape 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 [6,7] reasoning for double ionization after collision or photoexcitation. 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 a 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_2 and N_2 molecules where the initial states are of triplet and singlet character, respectively [15]. Recent experiments [16,17] observed differences between O_2 and N_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 ψ to continuum final states of well-defined electron momenta $\vec{k}_1$ and $\vec{k}_2$, i.e., $\langle \vec{k}_1 \vec{k}_2 | U | \psi \rangle$ where U 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 behavior $\langle \vec{r}_1 \vec{r}_2 | \vec{k}_1 \vec{k}_2 \rangle \rightarrow (2\pi)^{-3} \exp(i\vec{k}_1 \cdot \vec{r}_1 + i\vec{k}_2 \cdot \vec{r}_2 + i\chi)$, where the phase $\chi = 2\ln(k_1 r_1 + \vec{k}_1 \cdot \vec{r}_1)/k_1 + 2\ln(k_2 r_2 + \vec{k}_2 \cdot \vec{r}_2)/k_2 - \ln(k r_{12} + \vec{k} \cdot \vec{r}_{12})/(2k)$ with $\vec{k} = (\vec{k}_1 - \vec{k}_2)/2$ 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 labeled α,

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

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 is the evolution operator $U = P \exp(-i\int_0^T dt H/\hbar)$, where P is the time-ordering operator, corresponding to the Hamiltonian (in atomic units)

$$H = \sum_{i=1}^{2} \left(\frac{\vec{p}_i^2}{2} - \frac{2}{r_i} \right) + \frac{1}{|\vec{r}_1 - \vec{r}_2|} + F(t)(z_1 + z_2), \quad (2)$$

where $F(t)$ contains the time dependence of a linearly polarized laser pulse of T duration. It is symmetric under ex-

change of electrons. Therefore if the initial wave function belongs to the ^3S state of He, the sum in Eq. (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 gasses 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 $R=\rho_1=\rho_2$, $p_R=2p_{\rho_1}=2p_{\rho_2}$, $Z=z_1=z_2$, $p_Z=2p_{z_1}=2p_{z_2}$, $|\phi_1-\phi_2|=\pi$, and $p_{\phi_1}=p_{\phi_2}=0$. The Hamiltonian (2) in the C_{2v} subspace reduces to

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

The potential energy in Eq. (3) [and obviously also the potential energy in Eq. (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., $F(t)=$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. Note that the selection rule also applies to the static field problem and will modify the quantum cross section for simultameous electron escape if the initial state corresponds to ^3S.

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

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

$$(4)$$

Comparing Eq. (4) with Eq. (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 π. 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 the 1+1-dimensional model below, but we emphasize that the cleanest 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

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

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 and 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
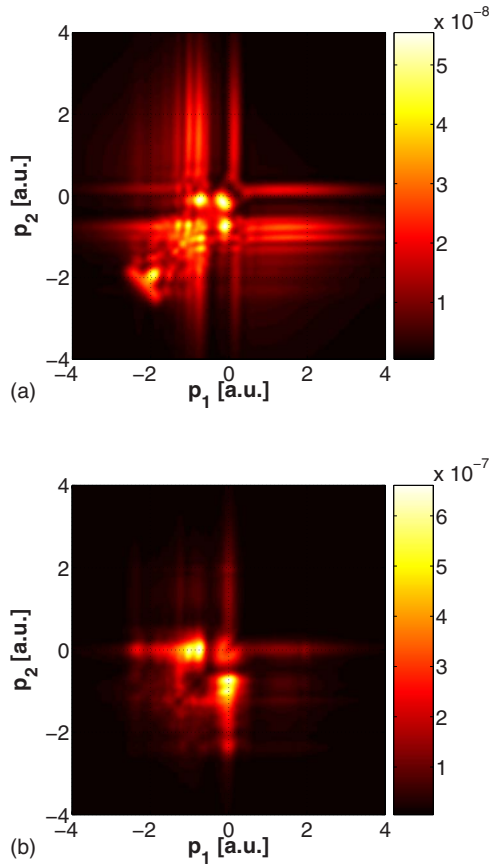
FIG. 1. (Color online) Electron momentum distributions for double ionization in a strong linearly polarized laser field starting from the ground state of the unperturbed atom (a) and from the first excited state (b). The distributions are obtained from Fourier transforms of the parts of the final wave functions 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 rule.

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 choose as an initial state the first excited state, which is antisymmetric with respect to electron exchange (in analogy to the ^3S state), the wave function 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 $1/x \rightarrow 1/\sqrt{x^2 + e}$ with $e = 0.6$, starting with the ground and first excited states of the unperturbed Hamiltonian. The ground state energy, for the chosen parameter e 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
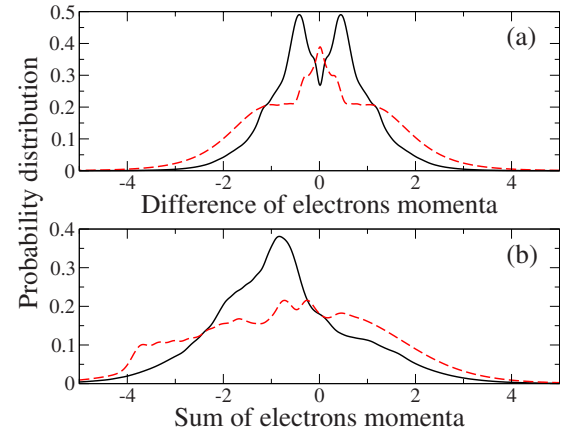

FIG. 2. (Color online) The probability distributions for differences, $p_{R_1} - p_{R_2}$, [panel (a)], and for sums, $p_{R_1} + p_{R_2}$, [panel (b)], of electron momenta for double ionization in a strong linearly polarized laser field. Data for the initial ground state (red dashed curves) are compared with those obtained for the initial first excited state (solid black lines). The minimum in the difference distribution in panel (a) reflects the suppresion of symmetric escapes. All parameters are the same as in Fig. 1.

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 exessively 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 wave functions in the regions $|R_1|$, $|R_2| > 50$ and they are Gaussian (of width 0.07) averaged in order to mimic the experimental resolution. For double ionization starting from the symmetric ground state there is a symmetric distribution of parallel final momenta with a maximum along equal momenta, $p_{R_1} = p_{R_2}$. That the distribution reaches only into the negative momentum sector is due to the short pulse which contains only one cycle [24]. When the initial state is the antisymmetric state, the corresponding distribution shown in Fig. 1(b) has a clear minimum along the axis $p_{R_1} = p_{R_2}$, reflecting the absence of the perfectly correlated escape, as predicted by the selection rule. The small residual contribution at the center is due to averaging with the Gaussian that models experimental resolution.

The signatures of the effect can be enhanced, as shown in Fig. 2(a), 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 minium when the initial state is the antisymmetric first excited state. Note again that the minimum in Fig. 2(a) does not reach zero because of the averaging; but Fig. 2(a) clearly demonstrates that the reso-

lution attainable in current experiments is sufficient to observe effects of the selection rule we analyze here.

In Fig. 2(b) 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 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. 2(b) 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. (We would like to point out that the selection rule, and hence the suppression, is not restricted to triplet states but also applies to singlet states, provided the quantum number M is odd.) 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 is O_2 molecules with triplet initial state oriented along the polarization axis. Recent experiments with aligned N_2 molecules [17] probed a singlet initial state only.

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