Correlated two-electron dynamics in strong-field double ionization

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Vector momenta of electrons and ions have been measured for strong-field (25 fs, 0.25 to 1.0 PW/cm^2) double ionization of Ar using a ''reaction microscope.'' Correlated two-electron momentum distributions along the field direction ($P_{1\parallel}, P_{2\parallel}$) are explored at different transverse momenta $P_{1\perp}$ of one of the electrons. Whereas a distinct ($P_{1\parallel}, P_{2\parallel}$) correlation, strongly varying with $P_{1\perp}$ is observed for nonsequential double ionization at small light intensity, only a weak, but still significant correlation is found at large intensity where sequential ionization dominates.

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A profound knowledge of many-electron dynamics in intense, femtosecond laser fields interacting with single atoms or molecules is basic and indispensable for our understanding of laser-matter interactions in general and, thus, for a large variety of present and future application. Among them are high-order harmonic generation, acceleration of electrons and ions, strong-field induced fusion or the realization of intense table-top x-ray sources. Theoretically, this requires a consistent quantum description of the time-dependent correlated motion of several electrons in the combined fields of the laser light and of the ionic core on a subfemtosecond time scale, i.e., for times that are short compared to one optical cycle of a typical laser pulse. Experimentally, the ultimate goal would be to trace the electronic dynamics on such short time scales, which has not been feasible up to now, leaving this domain completely unexplored.

The development of multielectron-ion imaging techniques [1] and their successful combination with femtosecond laser systems with kilohertz pulse-repetition rates enabled first single-differential measurements on strong-field multiple ionization of He [2,3], Ne [4], Ar [5], and Xe [6]. Surprisingly, the determination of ion recoil momenta alone already settled a 15-yr controversial debate on the mechanisms responsible for nonsequential (NS) double ionization and stimulated a series of theoretical papers: Only the ''rescattering model'' proposed by Corkum [7] eight years ago was found to be kinematically consistent with experimental-ion momentum distributions [4,8]. Recent theoretical *S*-matrix results [9–12], various numerical time-dependent two-electron quantum calculations [13,14] as well as predictions based on classical dynamics [15] all obtain the experimentally observed, characteristic double-peak structure in the recoil-ion momentum along the polarization direction $P_{R\parallel}$. Thus, they all provide strong evidence that rescattering dominantly contributes to NS multiple ionization in the intensity-regimes investigated.

Though being generally accepted that rescattering is the leading mechanism at intermediate intensities, the various calculations strongly disagree among each other in predicting the details of the underlying dynamics, i.e., of the electron-impact-ionization $(e,2e)$ kinematics in the presence of the field. Moreover, none of them is in quantitative agreement with first experimental results for argon double ionization [16]. Here, the correlated electron-momentum distributions along the field direction of the linear polarized light were found to be strongly peaked for emission of the two electrons in the same direction with a significant contribution of back-to-back scattered electrons.

Whereas all three-dimensional (3D) *S*-matrix calculations [10–12] do predict the unidirectional emission of the electrons at moderate light intensities, none of them can account for back-to-back ejection of the two electrons [16]. Moreover, even the details for unidirectionally emitted electrons differ strongly in the various 3D models. One of them, based on a Coulomb form factor for the $(e,2e)$ scattering process, predicts strongly asymmetric energy (momentum) sharing between the two electrons [10,12]. The other one, assuming a hard-core contact potential for the electron-electron interaction, on the other hand yields symmetric longitudinal momentum sharing [11,13].

Thus, in essence, though rescattering as being the trigger for multiple NS double ionization seems to be out of question, the details of the electron-scattering dynamics, i.e., of the $(e,2e)$ kinematics in the presence of the electric field are far from being well understood and remain the subject of intense debate.

In a previous paper [17] we have shown for 0.25 PW/cm^2, 25-fs light pulses interacting with an argon target, that back-to-back emitted electrons may consistently be interpreted as being due to excitation during rescattering [18] at a phase where the electric field is small with subsequent and independent field (tunnel) ionization of the second electron. Within the assumptions of the model [classical $(e,2e)$ dynamics without energy exchange with the light wave during recollision] both processes may be separated experimentally,

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leaving us now with the question on the correlated dynamics of unidirectionally emitted electrons.

In this paper, we present differential experimental information on correlated electron-momentum spectra in the longitudinal direction for unidirectionally emitted electrons as a function of one-electron's transverse momentum, shedding light on the details of the correlated electronic motion in the field and providing benchmark data for theory. Strong effects are observed in that an asymmetric sharing of the longitudinal momenta is favored if the transverse momentum space is restricted. Surprisingly, a weak though significant correlation is also found at an increased light intensity where sequential double ionization is expected to dominate. Recently, similar results were reported by Weckenbrock *et al.* for 150-fs laser pulses [20].

The experiments were performed at the Max-Born-Institut in Berlin using a Kerr-lens mode-locked Ti sapphire laser at 795-nm central wavelength. The laser pulses were amplified at 1 kHz repetition rate up to pulse energies of 500 μJ and focused by an on-axis spherical mirror ($f=100$ mm) to a spot of ≈ 10 μm diameter (full width at half maximum) within an ultrahigh vacuum chamber (base pressure less than 7×10^{-11} mbar). The pulse peak intensity was adjusted in the range between 0.25 and 1.0 PW/cm^2. Intensity fluctuations were controlled throughout the experiment and kept below 5%.

The laser beam was focused on a low density (10^8 atom/cm^3) supersonic gas jet formed by expanding Ar at a stagnation pressure of 5 bar through a cooled, 10-μm-diam nozzle. The beam was collimated over a total path of 2 m to a rectangular shape of 0.2×4 mm^2 at the focal spot, oriented with the broad side perpendicular to the laser beam. This yielded an overlap volume between laser beam and argon jet of 200 μm in length and 10 μm in diameter.

Vector momenta of ions and electrons emerging from the source volume were recorded using a "reaction microscope" that has been described in detail before [19]. Low-energy ions and electrons are accelerated in opposite directions by a 1-V/cm electric field applied perpendicular with respect to the propagation directions of the laser as well as the atomic beam and parallel to the laser-polarization direction. The transverse motion of the electrons is confined by an additional homogeneous solenoidal magnetic field. In this way all electrons with transverse energies smaller than 50 eV and longitudinal energies below 15 eV are guided onto a 75-mm-diam two-dimensional position-sensitive microchannel plate detector placed 20 cm away from the reaction volume. From the measured absolute positions and flight times the ion and electron trajectories are reconstructed and their initial momenta are calculated. For the electrons a momentum resolution in transverse and longitudinal directions of 0.05 a.u. is achieved. The ion-momentum resolution in the direction of the jet expansion is limited by the internal jet temperature to 0.3 a.u. Transverse to the jet expansion a resolution of 0.1 a.u. is achieved due to collimation of the jet.

In Fig. 1 the correlated two-electron longitudinal momentum spectrum is shown for argon double ionization at a power density of 0.25 PW/cm^2, where nonsequential ionization dominates. In this plot, which has been published before [17], the longitudinal electron momenta are presented without any further restriction on the other momentum components. As discussed, a significant correlation is observed: Preferentially the two electrons are emitted into the same direction having both either positive or, equivalently, negative momenta of similar magnitude. Here, at relatively low light intensity, a kinematical analysis within the conditions described in Ref. [17] demonstrated, that events due to $(e,2e)$ ionization during rescattering are exclusively restricted to unidirectional emission of the two electrons into the area surrounded by the solid line. Excitation during rescattering followed by tunnel ionization of the second electron, instead, allow for two-electron momenta inside the area indicated by the broken lines.

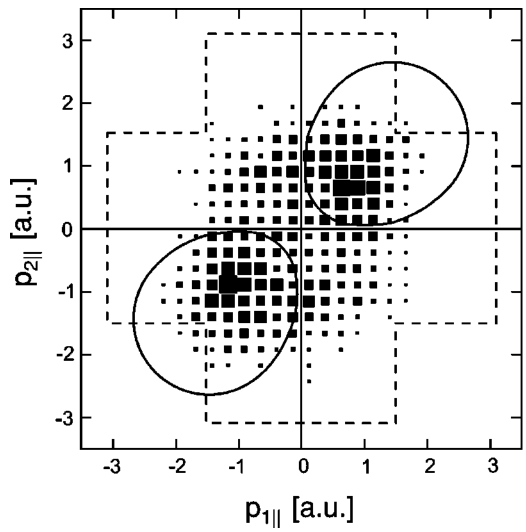

FIG. 1. Longitudinal momentum of electron one ($P_{1\parallel}$) versus that of electron two ($P_{2\parallel}$) for Ar double ionization by 25-fs laser pulses at a peak intensity of 0.25 PW/cm^2. Box sizes correspond to the intensity on a linear scale between zero and maximum intensity. Full line: borderline for $(e,2e)$ kinematics. Dashed line: borderline for excitation-tunneling events (see also Ref. [17]).

In order to explore the electronic motion during rescattering in more detail we have plotted the correlated electron momenta for unidirectional emission, where the $(e,2e)$ events occur, under the additional condition that the transverse momentum of one electron ($P_{1\perp}$) is either larger or smaller than 0.5 a.u. (Fig. 2). This splits the total number of events into two contributions of about identical size.

Distinct differences are found. If the transverse momentum of electron "one" is large $P_{1\perp}\geqslant0.5$ a.u., the correlation in the longitudinal electronic motion becomes even more pronounced [Fig. 2(a)]. Unequal momentum sharing is widely suppressed and the electrons most likely emerge with quite similar final momenta along the diagonal in the two-dimensional plot. Therefore, both electrons must have been created with similar or very small momentum components $P_{1\parallel},P_{2\parallel}$ at the same phase in the light wave and, thus, have received similar drift momenta after their creation.

This tendency might be understood within the rescattering model assuming energy conservation $P_{1\parallel}^2+P_{2\parallel}^2+P_{1\perp}^2+P_{1\perp}^2=2(E_0-I_p)$ during recollision (E_0: energy of the returning

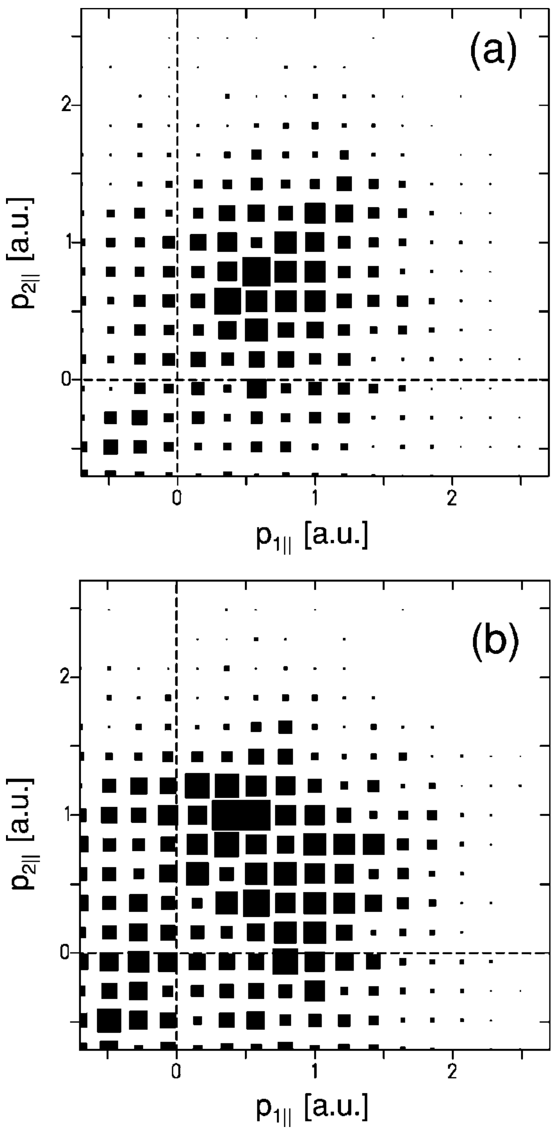

FIG. 2. Enlarged view of the uppermost right-hand part of Fig. 1 with additional conditions on the transverse momentum of electron "one:" (a) $P_{1\perp} \geqslant 0.5$ a.u. and (b) $P_{1\perp} \leqslant 0.5$ a.u.

electron; $I_p = 27.62$ eV: ionization potential of Ar). In a field-free $(e,2e)$ collision at moderate E_0, "binary collisions" between the electrons would dominate for significant transverse momentum transfer to one of them $(P_{1\perp} \geqslant 0.5$ a.u.), leaving the second one with a similar $P_{2\perp} \geqslant 0.5$ a.u. Consequently, this reduces the kinematically allowed regime for the longitudinal momenta from $P_{1\parallel}^2 + P_{2\parallel}^2 = 2(E_0 - I_p) = 1.47$ a.u. for $P_{1\perp} = P_{2\perp} = 0$ at maximum $E_0 = 3.17U_p$ to $P_{1\parallel}^2 + P_{2\parallel}^2 \leqslant 0.97$ a.u. for $P_{1\perp}$, $P_{2\perp} \geqslant 0.5$ a.u. (U_P is the ponderomotive potential). Thus, a significantly reduced, still quite large momentum spread between the two electrons in the longitudinal direction remains possible and asymmetric sharing is expected from field-free $(e,2e)$ dynamics. This is definitely not observed experimentally [Fig. 2(a)]. Assuming further that $P_{1\parallel}^2 \approx P_{2\parallel}^2$, which might be justified at small excess energies, finally leads to similar longitudinal electron momenta of less than 0.7 a.u., Depending on

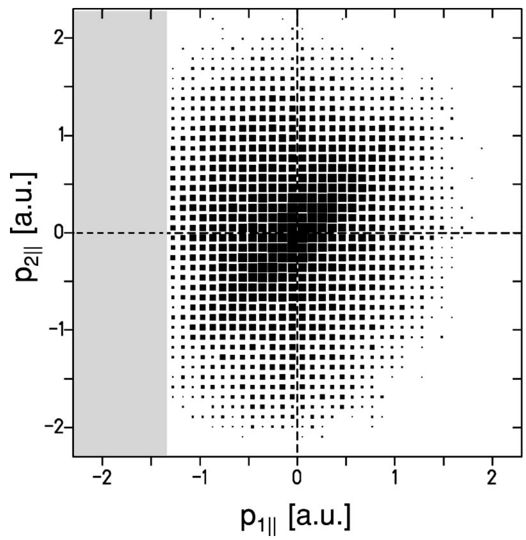

FIG. 3. Same as Fig. 1 for an intensity of 1.0 PW/cm^2. Shaded area indicates a region of no acceptance for electron "one."

the recollision phase in the oscillating field, both electrons would then acquire nearly identical drift momenta of less than 1.5 a.u. each, in qualitative agreement with the experimental data.

Obviously, the Coulomb repulsion between the two electrons in the longitudinal direction is strongly suppressed and quite similar momenta do occur if comparably large relative momenta of the electrons in the second, i.e., in the transverse dimension, are admitted. Thus, one-dimensional calculations will never be able to give longitudinal momentum distributions in agreement with the experimental data. In essence, they predict zero intensity for equal momentum sharing, i.e., along the diagonals in Figs. 1 and 2 at a light intensity comparable with the one used here [13,14]. Clearly, the deficiency of such models lies in their reduced dimensionality resulting in a strong overestimation of the electron-electron repulsion. As a consequence, equal longitudinal electron momenta are strictly forbidden if they are created in a rescattering event at identical phase.

If small transverse momenta of electron "one" $P_{1\perp} \leqslant 0.5$ a.u. are selected instead, i.e., if the two-electron phase space in the second dimension is reduced, the $(P_{1\parallel}, P_{2\parallel})$ pattern changes significantly [Fig. 2(b)]. Unequal momentum sharing is strongly favored now, with typical momenta around 1 a.u. for one and 0.5 a.u. for the other electron. This behavior is well known from field-free $(e,2e)$ dynamics, where unequal energy sharing dominantly contributes to the cross section at small scattering angles of the projectile electron. In the rescattering scenario, the impinging electron is little deflected and only looses a small part of its longitudinal momentum in the $(e,2e)$ collision, producing a low-energy secondary electron. Thus, directly after recollision, the start momenta of both electrons are very different, resulting in a most likely asymmetric momentum sharing in the final state.

Basically, such a behavior has been theoretically predicted for Argon at identical light intensity within an S-matrix approach using a Coulomb form factor to describe the $(e,2e)$ collision [10,12]. It is not consistent, however, with the results of similar calculations using a hard-core potential [11,12]. Moreover, since the momentum space is reduced in the transverse direction, Coulomb repulsion between the two electrons after recollision, which is not included in any of the 3D calculations [10,12], is expected to reduce events with similar longitudinal momenta. Reduction of the transverse momentum of one of the electrons essentially limits its motion in this direction, making the electron propagation ''one-dimensional'' with increased importance of the electron-electron repulsion in the longitudinal direction. Thus, it is no surprise that the two-electron momentum distribution is now in better qualitative agreement with results of one-dimensional calculations.

Finally, two-electron spectra have been investigated in Fig. 3 at a high peak intensity of 1 PW/cm^2 where sequential electric-field (tunnel) ionization is expected to be the dominant mechanism. We cannot completely exclude possible volume effects, but any contribution from regions with lower intensities should result in the appearance of a structure similar to that of Fig. 1, namely, two well-separated maxima in the momentum pattern. For sequential and therefore noncorrelated electron emission one would expect a spherically symmetric two-electron momentum distribution. Surprisingly, more events appear along the diagonal line indicating that some correlation is left. Both electrons are preferentially emitted into the same direction for small longitudinal-momentum components, i.e., for electrons that have been created close to the maxima of the oscillating electric field. Such a pattern might be expected if two electrons either leave by tunneling (sequential or collective) or by ''shake-off'' during the same cycle of the laser pulse.

In summary, we have presented differential correlated two-electron longitudinal-momentum spectra for different transverse momenta of one of the electrons. It has been demonstrated that our experimental data can help to distinguish between predictions of different calculations, thus, serving as benchmark data for theory.

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[1] R. Dörner *et al.*, Phys. Rep. **330**, 96 (2000).
[2] T. Weber *et al.*, Phys. Rev. Lett. **84**, 443 (2000).
[3] R. Lafon *et al.*, Phys. Rev. Lett. **86**, 2762 (2001).
[4] R. Moshammer *et al.*, Phys. Rev. Lett. **84**, 447 (2000).
[5] T. Weber *et al.*, J. Phys. B **33**, L127 (2000).
[6] B. Witzel, N. A. Papadogiannis, and D. Charalambidis, Phys. Rev. Lett. **85**, 2268 (2000).
[7] P. B. Corkum, Phys. Rev. Lett. **71**, 1994 (1993).
[8] B. Feuerstein, R. Moshammer, and J. Ullrich, J. Phys. B **33**, L823 (2000).
[9] A. Becker and F. H. M. Faisal, Phys. Rev. Lett. **84**, 3546 (2000).
[10] S. V. Popruzhenko and S. P. Goreslavskii, J. Phys. B **34**, L239 (2001); Opt. Express **8**, 395 (2001).
[11] R. Kopold *et al.*, Phys. Rev. Lett. **85**, 3781 (2000).
[12] R. Kopold, S. V. Popruzhenko, S. P. Goreslavskii, and W. Becker, Phys. Rev. A **64**, 053402 (2001).
[13] M. Lein, E. K. U. Gross, and V. Engel, J. Phys. B **33**, 433 (2000).
[14] M. Lein, E. K. U. Gross, and V. Engel, Phys. Rev. Lett. **85**, 4707 (2000).
[15] B. Eckhardt and K. Sacha, Phys. Scr. **T90**, 185 (2001).
[16] T. Weber *et al.*, Nature (London) **405**, 658 (2000).
[17] B. Feuerstein *et al.*, Phys. Rev. Lett. (to be published).
[18] H. van der Hart, J. Phys. B **33**, L655 (2000).
[19] R. Moshammer *et al.*, Nucl. Instrum. Methods Phys. Res. B **108**, 425 (1996).
[20] M. Weckenbrock *et al.*, J. Phys. B **34**, L449 (2001).