Intense-Field Double Ionization of Helium: Identifying the Mechanism

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We present quantum mechanical calculations of the electron and ion momentum distributions following double ionization of a one-dimensional helium atom by ultrashort laser pulses (780 nm) at various intensities. The two-electron momentum distributions exhibit a clear transition from nonsequential to sequential double ionization. We provide strong evidence that rescattering is responsible for nonsequential ionization by calculating the momentum spectrum of the He^{2+} recoil ions—which we find in excellent agreement with recent experiments—and by analyzing the electronic center-of-mass motion via Wigner transforms.

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The fundamental processes underlying the interaction of strong laser fields with matter still bare many open questions. One of the most striking surprises was found in experiments [1–4] studying single and multiple ionization of atoms by intense short laser pulses. While single ionization yields are well understood by considering only one of the electrons as active, double and multiple ionization processes turned out to be much more complex: If double ionization was a stepwise process of the form $A \rightarrow A^+ + e^- \rightarrow A^{2+} + 2e^-$, without correlation between the two steps, then the double-ionization yield would simply follow from rate equations involving the single-ionization rates of A and A^+. Experimental double-ionization yields of helium were found to be *several orders of magnitude* larger than the values calculated in this way, leading to a distinct "knee" in the double-ionization yield plotted as a function of intensity. This gigantic structure rules out a sequential ionization mechanism and hence represents one of the most dramatic manifestations of electron-electron correlations in nature. A variety of theoretical approaches, such as diagrammatic S-matrix techniques [5], one-dimensional (1D) time-dependent calculations [6,7], and three-dimensional time-dependent simulations [8] have succeeded in producing nonsequential ionization. However, the physical picture of the ionization mechanism has remained rather controversial. Essentially, three models of double ionization have been proposed: (i) a shakeoff process [1], where one electron is very quickly ionized, so that the second electron gets excited and is easily ionized a short time later, (ii) a rescattering process [9], in which the first electron is accelerated back towards the core when the electric field reverses its sign, with the effect of e-$2e$ scattering at the time when the electron hits the core, and (iii) collective tunneling [10], where both electrons tunnel out simultaneously.

Until recently, only total ionization yields could be obtained from experiment, giving little insight into the ionization mechanism. A series of new experiments (see [11] for He, [12] for Ne, and [13] for Ar) has provided information about the electron-electron correlation through measurements of the recoil-ion momenta: Since the laser photons carry negligible momentum, the recoil momentum of a doubly charged ion balances the sum of the two electron momenta. Measurements of the full two-electron momentum distribution have been published very recently [14]. One crucial observation in these experiments is that, within a certain intensity range (roughly $5 \times 10^{14} - 1 \times 10^{15}$ W/cm^2), the doubly charged recoil ions most probably receive *nonzero momenta* (in contrast to singly charged ions, which exhibit a distribution with a maximum at zero momentum). Simple classical arguments show that nonzero recoil momenta are not compatible with shakeoff or collective tunneling, but are possible in a classical rescattering picture [11,12]: If an ion is created with zero initial velocity at a time when the electric field is at its maximum, then its final momentum will be zero. In a shakeoff or a collective-tunneling process, two electrons are ejected within a short time interval, most likely at maximum field strength. The result would be a distribution of ion momenta around zero. In the rescattering model, however, up to one optical cycle may pass between the first and second ionization step, which can give rise to nonzero ion momentum, depending on the phase when the initial ionization takes place. Rescattering is also supported by previous time-dependent numerical simulations [15,16].

In this paper, we present numerical results for the two-electron momentum distributions and for the recoil-ion momentum distributions at various intensities. We employ a linear model of the helium atom, where the motion of both electrons is restricted to the direction of the laser polarization. Experience has shown that 1D models qualitatively reproduce strong-field phenomena such as the double-ionization knee structure [6] or above-threshold ionization [17]. The simulations presented in this paper reproduce the experimental recoil-ion momentum spectra very well, thereby confirming the rescattering model. A crucial advantage of our calculations is that they provide the full correlated two-electron wave function. Once this

quantity is available, it allows one to further pin down the ionization mechanism at hand. For this purpose we analyze in detail the Wigner transform of the wave function with respect to the electronic center-of-mass coordinate.

Another benefit of this paper is the possibility to study *low* intensities ($\sim 10^{14}$ W/cm^2). Those are particularly interesting as they yield the largest ratio of double to single ionization [3]. Measurements of differential spectra are hard to perform in this regime because of the small ion yields.

We use the velocity-gauge model Hamiltonian,

$$H = \frac{p_1^2}{2} + \frac{p_2^2}{2} + (p_1 + p_2)A(t) - \frac{2}{\sqrt{z_1^2 + 1}} - \frac{2}{\sqrt{z_2^2 + 1}} + \frac{1}{\sqrt{(z_1 - z_2)^2 + 1}}, \quad (1)$$

with $A(t) = -\int_0^t E(t')\,dt'$. Here z_1 and z_2 are the electron coordinates, which can be positive or negative, p_1 and p_2 are the canonical electron momenta, and $E(t) = E_0 f(t)\sin(\omega t)$ is the electric field of a laser pulse with amplitude E_0, envelope function $f(t)$, and frequency ω. The Coulomb interactions between the particles are thus modeled by softened potentials, which is a way of allowing in 1D for the possibility that the particles pass by each other without probing a Coulomb singularity. We split the two-dimensional space into three parts: (A) $\{|z_1|, |z_2| < a\}$, (B) $\{|z_1| < a, |z_2| \geq a\}$, or $\{|z_1| \geq a, |z_2| < a\}$, and (C) $\{|z_1|, |z_2| \geq a\}$ with $a = 200$ a.u. In region A, the wave function is propagated exactly by means of the split-operator method [18] with 2100 time steps per optical cycle, starting from the symmetric ground state. In region B, one of the electrons is considered as ionized and its interaction with the other particles is neglected, so that the propagation of this electron can be accomplished by multiplications in momentum space. Owing to the symmetry of the wave function with respect to the interchange of electrons, it is sufficient to treat only that part of B where electron 1 is ionized. In region C, we consider both electrons as ionized and propagate the entire wave function in momentum space. The transfer from A to B is performed by smoothly cutting off the outer part of wave function A and adding its momentum representation coherently to wave function B. In an analogous way, the transfer from B to C is done. Variation of the grid size a gives very little change in the spectra, showing that the neglect of interactions in regions B and C is well justified. The splitting method has the advantage that we retain information about the momentum distribution, even if the wave function becomes too extended to fit on a grid in configuration space. This is essential in order to deduce final momentum distributions from the simulation. Note that the previously published momentum-space snapshots [16] were taken of the inner part only. A similar splitting technique was used in Ref. [7]. The double-ionization spectrum as a function of the two electron momenta p_1, p_2 is simply obtained as the modulus squared of the final momentum-space wave function in region C. Via $p(\mathrm{He}^{2+}) = -(p_1 + p_2)$, the momentum spectrum of the He^{2+} ions follows immediately.

Our calculations use 780 nm laser pulses with a total length of 8 optical cycles ($\equiv$20.8 fs), switched on and off linearly over 2 cycles. Note that such a linearly ramped pulse behaves similar to a long pulse in the sense that no nonadiabatic change of a free particle's drift velocity is induced during the switch-on/switch-off processes, even though they are very short. The absence of nonadiabatic effects was implicitly assumed when conclusions were drawn from experimental recoil-ion momenta in Refs. [11,12].

We have varied the laser intensity in the range from 1×10^{14} to 2×10^{15} W/cm^2. Figure 1 shows the resulting double-ionization two-electron momentum distributions at a time of 16 fs after the end of the pulse, when the evolution is practically finished. Most of the spectra show the tendency that p_1 differs from p_2, which is a result of the repulsive electron-electron interaction.

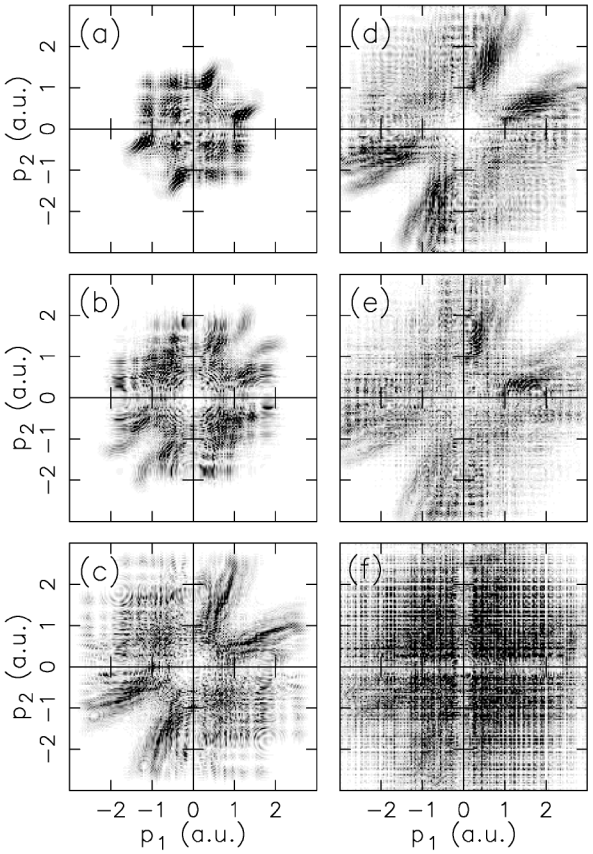

FIG. 1. Two-electron momentum distributions for double ionization at the intensities (a) 1×10^{14} W/cm^2, (b) 3×10^{14} W/cm^2, (c) 6.6×10^{14} W/cm^2, (d) 1×10^{15} W/cm^2, (e) 1.3×10^{15} W/cm^2, and (f) 2×10^{15} W/cm^2. Grey scales from 0 to (a) 1.5×10^{-6}, (b) 4×10^{-3}, (c)–(d) 1.5×10^{-2}, (e)–(f) 2.5×10^{-2}.

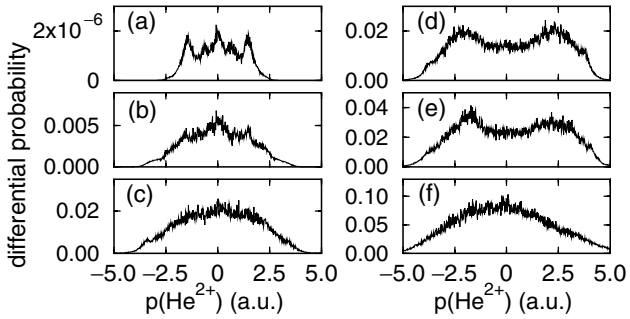

FIG. 2. Calculated momentum distributions of He^{2+} recoil ions for intensities as in Fig. 1.

At 1×10^{14} W/cm^2, four maxima are found in addition to some broadly distributed density. They correspond to a process with a slow (momentum below 0.5 a.u.) and a fast (momentum beyond 1 a.u.) electron. This is evidence for an energy transfer among the particles since a single electron from direct ionization can classically acquire at most $E_0/\omega = 0.9$ a.u. momentum in a field of 10^{14} W/cm^2, (cf. Ref. [19]). The probabilities for ejection in the same or in opposite directions are roughly equal. As we increase the intensity, the moduli of the electron momenta move towards larger values. Between $I = 6.6 \times 10^{14}$ W/cm^2 and $I = 1.3 \times 10^{15}$ W/cm^2, we find strong maxima in the regions where the electrons leave in the same direction, which is a sign that the Coulomb repulsion between the electrons loses some of its importance in the presence of the strong field. This is in agreement with experiment [14]. At an intensity of 2×10^{15} W/cm^2, the pulse is sufficiently strong to make correlation effects irrelevant: The density along the diagonal $p_1 = p_2$ is no longer suppressed, and the preference for ejection in the same direction has disappeared. Furthermore, vertical and horizontal lines indicate that the distribution resembles a product of two single-particle momentum distributions. This shows that double ionization is sequential at $I = 2 \times 10^{15}$ W/cm^2.

By integration of the two-electron spectra we arrive at the momentum distributions of He^{2+} ions, (see Fig. 2). The He^{2+} momenta extend to about ± 2 a.u. for the lowest intensity and up to ± 5 a.u. for the highest intensity. There is an intensity regime where the momentum distribution exhibits two maxima, located symmetrically with respect to the origin, while there is a minimum at zero. This general structure is predicted by the classical rescattering model [12]. The nonzero positions where these maxima are located move towards smaller momenta as the intensity is lowered. Additionally, a peak at zero appears, most clearly visible at the lowest intensity while, at 6.6×10^{14} W/cm^2, a single broad maximum is formed. In the high-intensity regime, where sequential ionization is dominant, a single maximum is observed as well. In this case, the reason is that the electrons are most likely and independently of each other emitted at times of maximum electric field, giving rise to zero final ion momentum [12]. In Fig. 3, experimental spectra from Ref. [11] are shown for comparison. The experimentally determined intensities have an uncertainty of up to 50%, and it seems that we must compare Fig. 2(d) ($I = 1 \times 10^{15}$ W/cm^2) to Fig. 3(b) ($I = 6.6 \times 10^{14}$ W/cm^2). Here we find excellent agreement, not only in the appearance of two maxima but also in the total width of the base and in the rather shallow minimum at zero momentum. If one lowers the intensity, theory and experiment are consistent as well: The double-peak structure disappears and the width of the spectrum decreases.

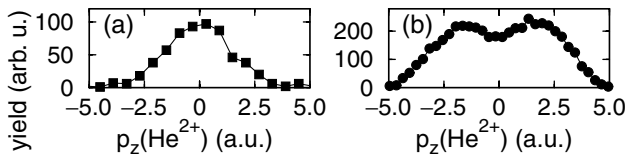

FIG. 3. Experimental distributions of He^{2+} momentum components along the laser polarization direction for the intensities (a) 2.9×10^{14} W/cm^2 and (b) 6.6×10^{14} W/cm^2. Cf. Fig. 3 of Ref. [11].

Of the proposed double-ionization mechanisms, the rescattering model is the only one that can explain the peaks at nonzero He^{2+} momenta. Yet, e-$2e$ scattering is not allowed in the classical sense at an intensity of 10^{14} W/cm^2 because the maximum recollision energy ($3.17U_p = 18$ eV, cf. Ref. [9]) is lower than the binding energy of He^+ (54 eV). Instead, we may think of a *field-assisted* rescattering process to which such constraints do not apply because the field can provide part of the energy which is needed to remove the second electron. By using semiclassical model calculations, it has been argued that rescattering cannot quantitatively account for the observed double ionization [20]. However, a recent reinvestigation of the recollision model took into account several corrections, among them the lowering of the ionization potential by the field, to show that the calculated double-ionization rates are indeed compatible with experiment [21]. Reference [22] shows that quantum interference effects on the recollision might further enhance double ionization as compared to semiclassical rescattering.

Further insight can be gained by a detailed inspection of the time evolution. However, snapshots of the wave function itself give only restricted information because the wave packets are very extended (up to hundreds of atomic units). We build a bridge to the classical way of thinking by taking the Wigner transform of the wave function, $\Psi(z_1, z_2)$, with respect to the center-of-mass coordinate, $Z = (z_1 + z_2)/2$, of the electrons,

$$w(Z, P, z) = \int dy\ \Psi^*(Z - y/2 - z/2, Z - y/2 + z/2)\Psi(Z + y/2 - z/2, Z + y/2 + z/2)e^{-iPy}\, dy\,, \quad (2)$$

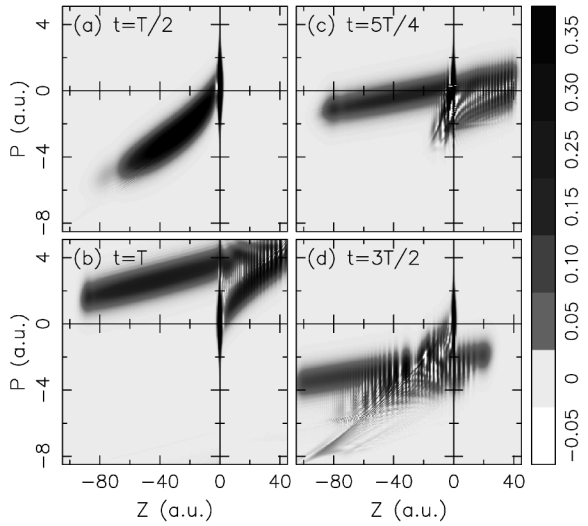

FIG. 4. Snapshots of the center-of-mass phase-space distribution $w(Z,P)$ [see Eq. (3)] in a laser field of constant intensity 10^{15} W/cm^2.

and subsequently integrating over the relative coordinate, $z = z_2 - z_1$:

$$w(Z,P) = \int dz\, w(Z,P,z)\,. \tag{3}$$

This function is interpreted as a probability distribution in the (Z,P) phase space with P being the electronic center-of-mass momentum, even though $w(Z,P)$ can have slightly negative values due to nonclassical dynamics. We have used the length-gauge formulation for a laser field of 10^{15} W/cm^2 intensity, suddenly turned on at time $t = 0$, to calculate the snapshots of $w(Z,P)$ shown in Fig. 4. After half a period, at $t = T/2$, a broad maximum has departed from the ground state at about $(Z,P) = (0,0)$ and moves towards negative values of Z. This wave packet unambiguously corresponds to single ionization, since there is negligible double ionization during the first half-cycle [16,23], i.e., Z can be identified with half of an electron's coordinate. At $t = T$, the wave packet has reversed the direction of its velocity and moves back towards positive values of Z. At its right edge, it has already crossed the line $Z = 0$, where the electron scatters from the He^+ core, which gives rise to the structure observed for $Z > 0$. Additionally, a new single-ionization wave packet has been formed at lower momenta P. Out of the scattered wave packets evolve distinct structures which pass the line $Z = 0$ at about $P = -2$ a.u. at $t = 5T/4$, moving into the negative Z direction. Simple integration of the classical equation of motion shows that these structures continue to move corresponding to two free electrons accelerated by the laser field. After 1.5 cycles, double ionization is clearly visible in the lower left region of Fig. 4(d) with a broad maximum at about $P = -7.5$ a.u. Note that this corresponds to an electronic center-of-mass drift momentum of -1.7 a.u., and thus a He^{2+} momentum of 1.7 a.u., which agrees well with the maximum about 2 a.u. in the He^{2+} spectrum [Fig. 2(d)]. That the double-ionization structures evolve from rescattered wave packets is evident in the phase-space pictures, whereas it could hardly be inferred from the time evolution in configuration [23] or momentum space [16].

To summarize, we have used a linear model of He to study how the distributions of electron and ion momenta, following double ionization, depend on the laser intensity. The calculated distributions agree well with experiment. The two-electron momentum distributions show that, for intermediate intensities (6.6×10^{14} to 1.3×10^{15} W/cm^2), both electrons prefer to depart from the nucleus in the same direction. These results and our analysis of the phase-space motion of the electronic center of mass via the Wigner transform of the two-electron wave function strongly support the rescattering mechanism for nonsequential double ionization in the intermediate intensity regime.

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