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Published in: New Journal of Physics

Document Version: Publisher's PDF, also known as Version of record

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2011 New J. Phys. 13 013024

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Double-electron above-threshold ionization resonances as interference phenomena

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New Journal of Physics 13 (2011) 013024 (9pp)
Received 24 September 2010
Published 19 January 2011
Online at http://www.njp.org/
doi:10.1088/1367-2630/13/1/013024

Abstract. We report calculations of double-ionization energy spectra and momentum distributions of laser-driven helium due to few-cycle pulses of wavelength 195 nm. The results are obtained from full-dimensional numerical integration of the two-electron time-dependent Schrödinger equation. A momentum-space analysis of doubly ionizing wavepackets shows that the concentric-ring structure of above-threshold double ionization, together with the associated structure of peaks in the total kinetic energy spectrum, may be attributed to wavepacket interference effects, where at least two doubly ionizing wavepackets from different recollision events populate the same spatial hemisphere.

In intense low-frequency laser fields, the mechanism for double ionization of helium varies with laser intensity. At high intensities, the two electrons are ejected in two independent single-electron ionization processes [1], where an initial single ionization of the neutral atom is followed by photoionization of the He⁺ ion. At lower intensities, double ionization is described reasonably well by the recollision model [2], in which a single electron is initially ejected at or near the peak of the electric field, and is then returned to the parent ion as the field changes direction. In the simplest 1D classical recollision model, the returning electron can recollide with the parent ion with a maximum energy of $3.17U_p$, where $U_p = E^2/4\omega^2$ is the ponderomotive energy at electric field strength $E$ and frequency $\omega$. On recollision, a number of different double-ionization pathways are possible. As outlined in [3], these pathways fall into two main categories: direct and delayed. In the direct pathway, both electrons are ejected in a simultaneous fashion, immediately after the recollision. This is most easily achieved if the returning electron has sufficient energy to ionize the He⁺ electron directly. For this to occur, the

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returning electron must be born in a field of sufficiently high intensity. At 195 nm, this intensity, which we refer to as the direct double-ionization threshold intensity, is \(48 \times 10^{14}\) W cm\(^{-2}\). Below this intensity, delayed pathways are likely to be dominant. In the delayed pathway, the recolliding electron returns with insufficient energy to ionize the second electron directly, but has sufficient energy to excite the He\(^+\) ion on recollision. The ion may be subsequently ionized at a later time, either near a peak or zero of the electric field.

The predictions of the recollision model have been substantiated both in theory and experiment. A fundamental aspect of the recollision process is the time delay between the initial single-ionization step and the subsequent double ionization. Calculations of this time delay have been obtained from full-dimensional integration of the two-electron time-dependent Schrödinger equation [4], reduced-dimensionality models [5] and from classical methods [6], and have found time delays consistent with the predictions of the recollision model. Additionally, experiments performed at 800 nm [7, 8] observed a finger-like structure in the momentum distributions which was also seen in full-dimensional numerical studies of double ionization of helium at 390 nm [9], reduced-dimensionality quantum calculations [10, 11], S-matrix approaches [12] as well as in analyses of classical trajectories at a number of laser wavelengths [13]–[15].

Previous calculations of intense-field recollision-induced double ionization using the full-dimensionality approach were performed at 390 nm [9]. Such a wavelength provides a good meeting ground between theory and experiment, since it is accessible to experiments using Ti:sapphire lasers (via frequency doubling), and also provides opportunities for theoretical calculations to be carried out, whereas calculations at the fundamental 780 nm Ti:sapphire wavelength reach the limits of present supercomputer capability. Comparison between theory and experiment of single- and double-ionization energy spectra of helium at this wavelength showed excellent agreement [9]. Most recently, good agreement has been obtained with full-dimensional classical calculations of double-ionization energy spectra at a number of laser intensities [15].

In this paper, we report a theoretical double-ionization study of helium using intense 195 nm light. The results are obtained from full-dimensional numerical integration of the two-electron time-dependent Schrödinger equation [16]. Previous work at 390 nm demonstrated that the double-ionization energy spectra typically consist of a structure of peaks due to above-threshold ionization (ATI), a correlated process in which the sum of the electrons’ final-state energies is constrained to integer multiples of the photon energy above the ponderomotive-shifted ground state. In contrast to these studies, this work considers the double-ionization process as the laser pulse is progressively lengthened from as little as a single optical cycle at peak intensity. It is worth lengthening the pulse by one such half-cycle at a time, since for each half-cycle a fresh single-electron wavepacket is released near the maximum of the field, and a recollision event can subsequently occur. Thus, we can study the effect of a series of recollision events as their number increases one by one. Keeping this in mind, the full-dimensional numerical integrations are performed using laser pulses consisting of a single-cycle ramp-on, so that no double ionization occurs prior to the peak-intensity field oscillations. At the end of the laser pulse, doubly ionizing wavepackets are extracted from the two-electron wavefunction using Gaussian masks, and this portion of the wavefunction is transformed into momentum space. The angular variables of the momentum-space wavefunction are then integrated away, leaving the probability distribution of the electrons as a function of the radial momenta \(k_1\) and \(k_2\). The total kinetic energy shared by the two electrons in atomic units is given by \(E = (k_1^2 + k_2^2)/2\).
Figure 1. Double-ionization energy spectrum of helium at 195 nm (lower panels) for each laser pulse shape (top panels). The portion of each pulse highlighted in red indicates the duration for which peak intensity is maintained. The laser intensity is $48 \times 10^{14}$ W cm$^{-2}$.

Figure 1 shows the variation in the total kinetic energy spectra of doubly ionized helium with the number of half-cycles at which peak intensity of the laser field is maintained. In each case, the laser intensity is $48 \times 10^{14}$ W cm$^{-2}$, the direct double-ionization threshold intensity at 195 nm. The first column shows the total energy spectrum due to a laser pulse containing only a single–cycle at peak intensity, with a single cycle rise and fall. The spectrum is strikingly lacking the typical structure of ATI peaks seen in previous work. The lack of such structure clearly indicates a breakdown or absence of the process by which ATI resonance the peaks are created. Small fluctuations are visible in the energy spectrum given in the first column of figure 1. These fluctuations are so small that we cannot draw useful conclusions from them. Close inspection of the spectrum reveals that the fluctuations do not resemble weak ATI resonances.

The analysis presented in this paper will be based on the observation that the side lobes of the electric field (these occur during the ramp-on and ramp-off of the field) contribute negligibly to ionization in comparison with the dominant lobes marked in red in figure 1. From the time-dependent ionization yield data, we can calculate the population ejected by the side lobes. In both the examples discussed here, the contribution to the ionization yield from the side lobes is less than 2%.

In the case of a pulse containing only one full cycle at peak intensity, there is the opportunity for a single-ionization event to occur at or near each of the two full-intensity peaks of the electric field. After each of these single-ionization events, the ejected electron is returned to the He$^+$ ion, allowing a total of two recollisions to take place. The classical-trajectory analysis given in [15] establishes the relative dominance of each of
the double-ionization pathways mentioned earlier as laser intensity is varied. As has been established in these classical calculations, and in experiment [17]–[19], the dominant double-ionization pathway at intensities above the direct double-ionization threshold is the direct pathway, in which the two electrons depart unilaterally into a common spatial hemisphere. Thus, because the two successive maxima of the electric field are of opposite polarity, the first recollision event will release a doubly ionizing wavepacket into a particular hemisphere, whereas the second recollision will release a wavepacket into the opposite hemisphere.

The second column of figure 1 shows the energy spectrum due to a laser pulse consisting of three half-cycles at peak intensity, with a single-cycle rise and fall as before. A clear structure of peaks has now emerged, with each peak separated from its nearest neighbours by the photon energy. The emergence of this structure indicates that its origin lies in the effect of the third peak-intensity half-cycle of the laser field. When the pulse contains a third half-cycle at peak intensity, a third recollision event can take place. This recollision will release a doubly ionizing wavepacket into the same hemisphere which was populated after the first recollision. These two wavepackets, located in the same spatial hemisphere, now have the opportunity to interfere. No such interference was possible in the case of a single-cycle pulse, because the two portions of the wavepacket were released into opposite hemispheres. Therefore, the origin of the ATI peak structure may be attributed to wavepacket interference effects, where at least two portions of the doubly ionizing wavepacket enter the same hemisphere, but with a one laser-cycle delay between them.

The peaks seen in the second column of figure 1 are well defined over a large range of energies, although they begin to die away at the highest energies. This loss of definition indicates a weakening of the interference between high-energy components of the wavepackets born after the first and third recollisions. Such high-energy components of the first-born wavepacket rapidly spread to large radial distances, and therefore show weaker interference with the corresponding high-energy component of the wavepacket released after the third recollision. Moreover, interference effects will only be present in one spatial hemisphere. To achieve interference in both hemispheres, the laser pulse must be lengthened by a further half-cycle, so that a fourth recollision takes place. The third column of figure 1 shows the energy spectrum due to a laser pulse consisting of four half-cycles at peak intensity. The peaks are now greater in amplitude, and maintain their amplitude at noticeably higher energies than was the case for three half-cycles at peak intensity, due to combined wavepacket interference effects present in both hemispheres.

Finally, to investigate the cumulative effect of multiple interferences as the laser pulse is substantially lengthened, the fourth column of figure 1 shows the energy spectrum due to a laser pulse containing eight half-cycles at peak intensity. A total of eight recollisions now take place, allowing four doubly ionizing wavepackets to populate each hemisphere. The high degree of interference in both hemispheres greatly improves the definition of the peaks over a large range of energies, and increases their amplitude by an order of magnitude.

To test the identifications of interference phenomena observed in double ionization, we consider the analogous effects in single ionization. We review, below, results we have obtained for single ionization, noting that these confirm the findings of Arbó et al [20] for this process. A single-electron wavepacket emerges into a particular hemisphere at or near an electric field maximum, but is eventually decelerated and driven back towards the core as the laser field changes its direction. During this time, a fresh single-electron wavepacket is released (one half-cycle after the initial release) into the opposite hemisphere, making possible interference
in this second hemisphere once the returning electron wavepacket crosses the nucleus (3/4 of a cycle after its birth).

Thus, the single-ionization energy spectra should exhibit ATI resonances when the laser field consists of only two half-cycles at peak intensity. Figure 2 shows the photoelectron energy spectrum of hydrogen at 780 nm, obtained through full-dimensional integration of the one-electron time-dependent Schrödinger equation. As before, the side lobes of the electric field contribute negligibly to ionization in comparison with the major half-cycles marked in red in figure 2. This is unsurprising, given that at 780 nm, nine photons are required for ionization of hydrogen. In the weak-field limit, ionization rates would scale as $E^{18}$, where $E$ is electric field strength. At the higher field strengths considered here, the exponent is less than 18. The electric field strength of the side lobes is around 60% of that of the major half-cycles indicated in red, so that population ejected during the ramp-on will be many times smaller than that ejected during the major half-cycles. As anticipated, the energy spectrum due to a single-cycle pulse does show evidence of ATI resonances. As before, these resonances are enhanced as the number of electric field half-cycles is increased. Further consideration may be given to the case where the laser pulse contains only one half-cycle at peak intensity. In such a case, only one singly ionizing wavepacket is released, and therefore the interference effects seen for longer pulses cannot occur. The resulting single-ionization energy spectrum should therefore lack the ATI resonances seen previously. Figure 3 shows the single-ionization energy spectrum of hydrogen obtained when the laser pulse contains only a half-cycle at peak intensity $9 \times 10^{13}$ W cm$^{-2}$. The absence of resonance structure indicates that its origins lie in interference between at least two wavepackets entering a common spatial hemisphere.
Figure 3. Single-ionization energy spectrum of hydrogen at 780 nm (lower panel) due to a laser pulse containing one half-cycle at peak intensity (top panel). The portion of the pulse highlighted in red indicates the duration for which peak intensity is maintained. The laser intensity is $9 \times 10^{13} \text{ W cm}^{-2}$.

Returning to the double-ionization analysis, further insight into the nature of the double-ionization process may be gained by examining the probability distribution in momentum space, as shown in figure 4. Each panel of figure 4 shows the momentum distribution obtained at the end of the laser pulse, as the pulse is lengthened from the case of a single cycle at peak intensity (figure 4(a)) to three half-cycles at peak intensity (figure 4(b)).

The distribution shown in figure 4(a) is very different from the typical distribution seen at 390 nm [9], where a clear structure of concentric rings is visible. Figure 4(b) differs qualitatively from figure 4(a), demonstrating that after three (major) half-cycles, all the important spectral features [21] of double-ionization ATI are now well defined. As discussed in [21], population is constrained to arcs, within which the two-electron kinetic energy $E$ is a constant, $(k_1^2 + k_2^2)/2$. The energies $E$ are an integer multiple of the photon energy $\hbar \omega$ above the ponderomotive-shifted two-electron helium ground state. As first reported in [9], the region of momentum space in which the two electrons simultaneously have energy exceeding $2U_p$ is largely depleted of population. At least one of the two electrons will exhibit a cutoff in energy near $2U_p$, exactly like the single-electron ionization of hydrogen observed in figure 2, where the cutoff is characterized by the onset of rapid exponential decay in probability density. The other electron has a high-energy cutoff above $3U_p$ in energy [9]. Again, as in single ionization (figure 2), the cutoff is characterized by the onset of exponential decay in probability density. In figure 1, the onset of this final exponential decay commences at about $7U_p$, in agreement with the high-intensity results of [9], figure 3.
In the preceding discussion we looked at double ionization in an intense-field limit, where basic rescattering theory of double ionization is directly applicable. In the basic rescattering picture, double ionization can be explained very simply in terms of an energy exchange between the two electrons during collision. In the $48 \times 10^{14}$ W cm$^{-2}$ laser fields used in the above examples, the rescattering electron returns to the residual He$^+$ ion (after a delay of about three-quarters of a field period) with just-enough energy (2 au) to free the remaining electron (the ground state of He$^+$) in an inelastic collision.

We turn now to a low-intensity limit, $I = 14 \times 10^{14}$ W cm$^{-2}$ at 195 nm, where this simple interpretation is not clearly applicable. At $14 \times 10^{14}$ W cm$^{-2}$, the energy of the rescattering electron as it returns to the residual ion is $3.2U_p = 0.58$ au. The energy required to eject the ground state electron of the He$^+$ ion is 2 au, and to excite the 2p state, 1.5 au. If the return energy were great enough to excite the 2p state, then ionization of the residual He$^+$ would again have a simple explanation: the rescattering collision could populate the 2p state, which is rapidly ejected from the atom as the electric field subsequently reaches its peak about 0.25 field periods after the rescattering collision. But the threshold intensity for such excitation is $I = 36 \times 10^{14}$ W cm$^{-2}$.

As we verified in the high-intensity case, ionization of the unperturbed He$^+$ ground state (and hence double ionization) cannot be explained as a sequential process. Calculation of the ionization rate of the He$^+$ ground state—unperturbed by a rescattering collision—shows that a field intensity of $I = 14 \times 10^{14}$ W cm$^{-2}$ is too small to produce the measured yields of doubly ejected electrons. In fact, at this intensity, sequential double ionization is under 5% of total measured double-ionization yield.

Explanation of simultaneous double ionization in this low-intensity limit remains challenging. We might ask, for example, if multiple recollisions are a requirement for significant double ionization at these low intensities. If that were true we would expect a greater time delay than that observed at higher intensities (about three-quarters of a field period) between the creation of the singly ionizing rescattering electron and the appearance of significant double ionization.

In figure 5, we show the spectra obtained for pulses of peak intensity $14 \times 10^{14}$ W cm$^{-2}$. As in the intense-field case we find that, after only three (major) half-cycles, many of the important
Figure 5. Joint momentum-space probability distributions of doubly ionized helium at the end of the ramp-off of a laser pulse of intensity $14 \times 10^{14}$ W cm$^{-2}$ containing (a) two half-cycles and (b) three half-cycles at peak intensity.

features of double-electron ATI are well defined. Spectra obtained from pulses longer than three half-cycles differ insignificantly from the spectrum in figure 5(b). The lack of significant additional time delay (compared with the high-intensity results) in the production of doubly ionizing electrons and in the production of well-defined resonance peaks is not consistent with a model in which multiple recollisions are mandatory in order to generate significant double ionization. The results are, however, consistent with the model previously introduced in [9] in which the rescattering electron merely perturbs the ground state of He$^+$. This small perturbation proved to be sufficient to enable immediate ionization by the laser field. In the case examined in [9] though, the field was only slightly below the threshold intensity necessary for double ionization through an inelastic rescattering collision.

In summary, we have reported full-dimensionality calculations of two-electron energy spectra and momentum distributions of doubly ionized helium in an intense few-cycle 195 nm laser field. We have shown that ATI resonances originate from interference between doubly ionizing wavepackets located in a common spatial hemisphere. We have seen that the evolution in character of the energy spectra with laser pulse length is a useful diagnostic probe of the time-dependent ionization dynamics of two-electron atoms. In particular, both the high-intensity and low-intensity results would be difficult to explain without a process such as rescattering, in which double ionization occurs at a half-cycle or more after the single-ionization process that creates the rescattering electron. If instead the doubly ionizing wavepackets were ejected simultaneously with the singly ionizing wavepackets, then we would expect that the double-ionization ATI resonance peaks would form after just two half-cycles, in the same way as the single-ionization ATI resonance peaks of figure 2.

Acknowledgments

This work has been supported by the UK Engineering and Physical Sciences Research Council and by the Northern Ireland Department of Employment and Learning. We acknowledge the provision of computer resources through a Cray Centre of Excellence Award.
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