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Multiple ionization of helium clusters by long wavelength laser radiation

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Abstract. We applied the method known as Fermion Molecular Dynamics to the description of a small cluster of helium atoms interacting with a short pulse of intense, long wavelength laser radiation, for both linear and circular polarization. We discuss the results of these calculations, elucidating questions relating to the mechanisms of multiple ionization in small clusters.

PACS. 36.40.-c Atomic and molecular clusters – 34.80.Qb Laser-modified scattering

1 Introduction

Within the past several years experiments have been performed by a number of different groups in which isolated noble gas atoms were multiply ionized by short pulses of high intensity, long wavelength, laser radiation [1–4]. Based on the earliest of these works it was suggested that, at the lowest laser intensities for which multiple ionization of isolated atoms could be observed, some new mechanism, distinct from sequential ionization [5], became dominant; but, only if the laser was linearly polarized. A form of laser-induced electron-electron interaction was proposed [6].

In the case of double ionization, this interaction mechanism was visualized as a recollision of the first electron to be photoionized with the residual +1 ion, leading to impact ionization of the second electron. Based on a classical argument, it was noted that the laser-induced *boomeranging* of the first electron could occur with an impact energy of up to $3.17U_p$, where the electron quiver energy was $U_p = (E_0/2\omega)^2$ in atomic units (a.u.), for a peak laser electric field strength of E_0 [6]. This implied a threshold laser intensity for correlated double ionization which was well below the threshold for over-the-barrier ionization (OBI) of the second electron, provided the laser wavelength was long enough. For example, for a helium atom and for a laser wavelength of 455 nm, the preceding argument implies a threshold intensity for double ionization of $9 \times 10^{14}\,\mathrm{W/cm^2}$, whereas the OBI threshold intensity for ionization of the second electron is approximately $10 \times$ larger.

Quasiclassical calculations, based on Fermion Molecular Dynamics [7] (FMD), have provided additional support for this model of laser-induced correlated double ionization [8]. These FMD calculations displayed an increasing dominance of boomeranging trajectories in the double ionization of helium, as the peak laser intensity was reduced below $10^{15}\,\mathrm{W/cm^2}$, for $\lambda = 455\,\mathrm{nm}$, and for linear polarization. This effect was absent in FMD calculations performed for circular polarization [9].

New experiments on the multiple ionization of atoms bound in clusters have very recently been reported [10], for laser conditions similar to those described above. These experiments have shown that there is a lower threshold laser intensity for the multiple ionization of an atom bound in a cluster than for an isolated atom of the same element. Speculations have arisen as to the nature of the mechanisms at work in this multiatom environment [11–15].

Simple classical arguments akin to those in reference [6] lead one to believe that the threshold for multiple ionization within a cluster can be reduced below its value for an isolated atom; *e.g.*, for helium atoms and for $\lambda = 455\,\mathrm{nm}$, the threshold laser intensity can be reduced to $4 \times 10^{14}\,\mathrm{W/cm^2}$, for a recollision taking place at a distance of approximately 30 a.u. from the site of the initial photoionization, for linear polarization. Thus, the threshold is lowered for a fraction of those events in which a photoionized electron, still under the influence of the laser, collides with a nearby +1 ion, but not with its parent ion. The amount of threshold lowering depends on the separation of the two ions, decreasing at very large separations; see Appendix A. Similar arguments apply for circular polarization, even though there is no threshold lowering for isolated atoms in this case.

We have performed a series of FMD calculations for helium atoms formed into clusters, and interacting with a short pulse of long wavelength laser radiation, both linearly and circularly polarized. These quasiclassical calculations clearly demonstrate the plausibility of the classical ideas: As the size of the cluster is increased, the probability

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for producing +2 helium ions (normalized to the number of atoms in the cluster) increases at all values of the peak laser intensity. This is especially notable at the lowest laser intensities for which double ionization occurs.

2 Formalism

The quasiclassical method known as Fermion Molecular Dynamics (FMD) has been applied successfully by us earlier to the description of the ionization of isolated helium atoms by pulsed laser light [8]. We have outlined the FMD method in that work, and in still earlier publications referenced therein. The first application of FMD to laser-atom interaction phenomena was by Wasson and Koonin [16].

Very briefly, the FMD method is based on an extension of Hamilton's equations for a system of electrically charged particles and external fields:

$$d\mathbf{r}_j/dt = \boldsymbol{\nabla}_{\mathbf{p}_j} H$$
$$d\mathbf{p}_j/dt = -\boldsymbol{\nabla}_{\mathbf{r}_j} H. \tag{1}$$

The Hamiltonian H may be decomposed as,

$$H = H_0 + U + V_H + V_P \tag{2}$$

where

$$H_0 = \sum_i [p_i^2/2M_i + Z_i \sum_{j<i} (Z_j/r_{ij})] \tag{3}$$

$$U = -\mathbf{E}(t) \cdot \sum_i Z_i \mathbf{r}_i, \tag{4}$$

and for electrons $Z_i = -1$ and $M_i = 1$. The applied electric field is,

$$\mathbf{E}(t) = \hat{\mathbf{z}} E_0(t) \cos(\omega t + \phi) \tag{5}$$

for linear polarization, and

$$\mathbf{E}(t) = \hat{\mathbf{z}} E_0(t) \cos(\omega t + \phi) + \hat{\mathbf{y}} E_0(t) \sin(\omega t + \phi) \tag{6}$$

for circular polarization, all in atomic units (a.u.). The function $E_0(t)$ is an envelope function, and the phase ϕ is arbitrary (determined randomly as a part of the initial conditions). The FMD extensions of the Hamiltonian function are contained in the effective potentials V_H and V_P, which are written as,

$$V_{H,P} = A_{H,P} \sum_i \sum_{j<i} (1/r_{ij}^2) \exp(-B_{H,P} r_{ij}^4 p_{ij}^4) \tag{7}$$

where the relative coordinates and momenta are, respectively,

$$r_{ij} = |\mathbf{r}_i - \mathbf{r}_j|$$
$$p_{ij} = |M_j \mathbf{p}_i - M_i \mathbf{p}_j|/(M_i + M_j). \tag{8}$$

The potentials V_H and V_P mimic the effects of the Heisenberg relations and the Pauli principle, respectively. V_H operates only between particles of opposite electrical charge,

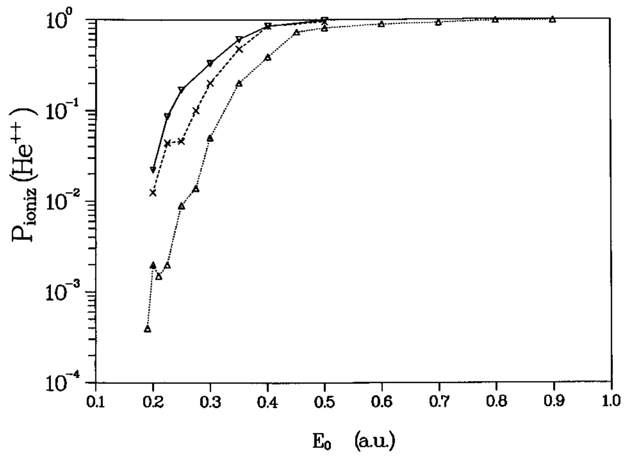

Fig. 1. Probability of helium double ionization *vs.* laser peak electric field strength, for $t_{pulse} = 80$ fs and $\lambda = 455$ nm, all for linear polarization: single atom (dotted line); 8 atom cluster (dashed line); 27 atom cluster (solid line).

while V_P operates only between electrons of the same spin; in FMD, electrons are deemed to be labled by a two-state spin variable. The constants $A_{H,P}$ and $B_{H,P}$ (both positive real numbers) are chosen in a straightforward manner [7]. For the singlet state (spins opposed) of He, $A_P = 0$, so that only V_H contributes.

As in reference [8], we chose the envelope function to have a pure sine-squared form; *i.e.*,

$$E_0(t) = E_0 \sin^2(\pi t/t_{pulse}) \tag{9}$$

for $0 \leq t \leq t_{pulse}$, for which the full-width at half-maximum is $\Delta t_{1/2} = t_{pulse}/2$.

3 Results and discussion

We used a wavelength of $\lambda = 455$ nm ($\omega = 0.1$ a.u.), and a pulse length of $t_{pulse} = 80$ fs (3300 a.u.) Clusters containing 8, 27, and 64 helium atoms were considered separately. The clusters were always of simple cubic symmetry initially, with a "lattice constant" of 6.6 a.u. (3.5 Å). The case of an isolated helium atom was also considered as a reference. Calculations were performed for both linear and circular polarization.

In Figure 1 we plot the probability of double ionization per atom for the four helium cluster sizes considered (isolated atom also), for linear polarization. As described in the Introduction and as is seen in the figure, P_{ioniz}(He^{++}) increases at all values of the peak laser intensity as the size of the cluster increases.

In Figure 2 we plot the corresponding data for circular polarization. In this case, there is no recollisional ionization observed for the isolated atom. However, within a cluster the amount of enhanced double ionization is generally as pronounced as for linear polarization, especially at the lower laser intensities. This is consistent with the

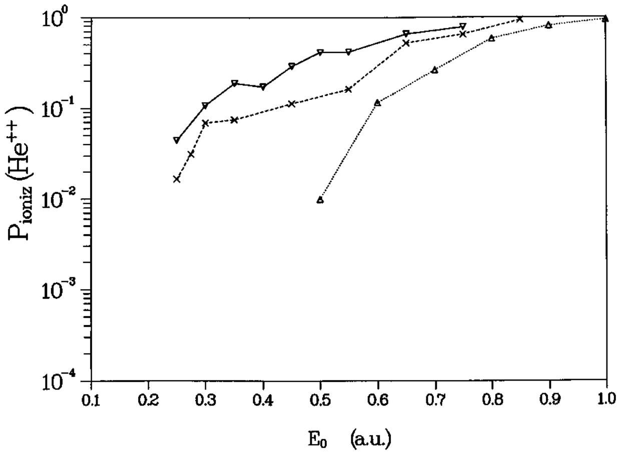

Fig. 2. Conditions as in Figure 1, but for circular polarization.

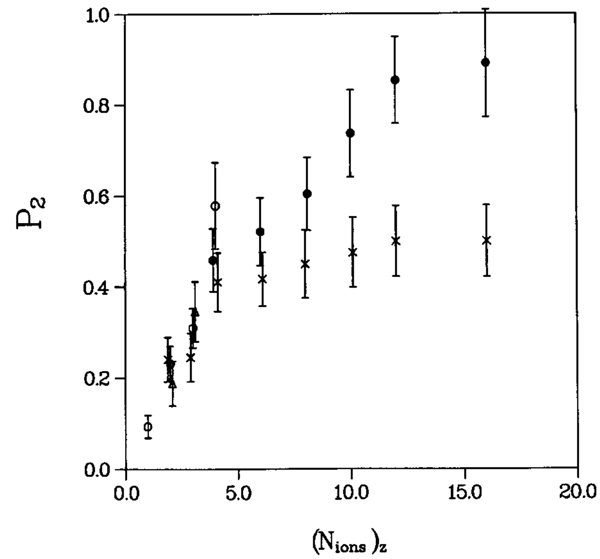

Fig. 3. Probability of helium double ionization *vs.* cluster size, for $E_0 = 0.30$ a.u., $t_{pulse} = 80$ fs, and $\lambda = 455$ nm, all for linear polarization. Atoms are arrayed along the Cartesian directions as: $1 \times 1 \times N$ (x); $2 \times 2 \times N$ (•); $1 \times N \times N$ (△); $N \times N \times N$ (○); and the laser electric field points in the z-direction.

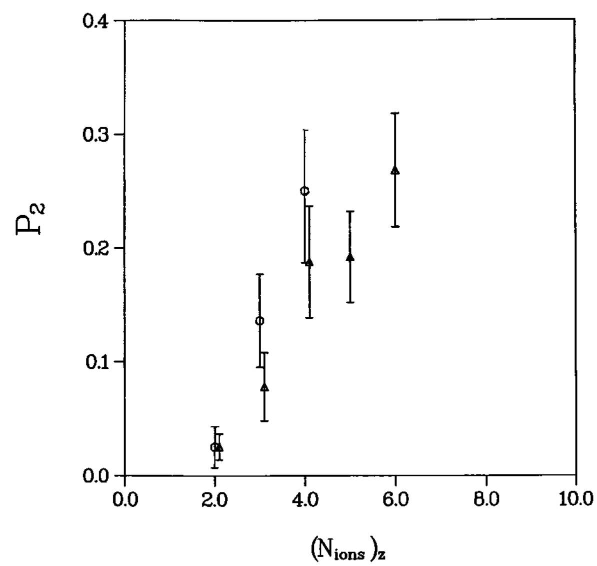

Fig. 4. Conditions as in Figure 3, but for circular polarization. Atoms are arrayed according to: $1 \times N \times N$ (△); $N \times N \times N$ (○).

mechanism outlined in the Appendix A. Since for circular polarization one has that $r(t) > 0$ for all $t > 0$, the first electron to be ionized never returns to its parent ion. However, it can still gain energy in the field of the laser and then collide inelastically with a neighbor ion, resulting in the enhanced emission of secondary electrons and an excess production of multiply charged ions. As described in the Appendix A, the maximum energy of recollision in a cluster is $8U_p$ for both linear and circular polarization. This contrasts with the maximum energy of recollision for an isolated ion, which is just $3.2U_p$, and then only for linear polarization.

For fixed peak field strength $E_0 = 0.3$ a.u., and with $\lambda = 455$ nm and $t_{pulse} = 80$ fs, we have the results displayed in Figure 3 for linear polarization, and Figure 4 for circular polarization. For linear polarization, we obtained results for purely linear arrays of atoms ($1 \times 1 \times N$), for elongated shapes ($2 \times 2 \times N$), and for cubic arrays ($N \times N \times N$); the laser electric field was always parallel to the long axis of the array (z-direction). For circular polarization, we obtained results for platelets ($1 \times N \times N$), as well as for cubic arrays; the laser electric field always lay in the plane of the platelet ($y{-}z$ plane).

It is clear from Figures 3 and 4 that the degree of ionization increases rapidly with N, and that nearly complete double ionization is achieved for very small clusters. More importantly, it is apparent that the geometry of the cluster is as significant a factor in promoting double ionization as is the number of atoms in the cluster. For these very small clusters, enhanced double ionization depends upon aligning the long axis of the asymmetric cluster with the laser electric field. This suggests that: (*i*) Laser driven collisions of electrons with ions other than their parent is a dominant factor in double ionization within small clusters. (*ii*) There are very few off-axis collisions occurring for linear polarization, and very few out-of-plane collisions occurring for circular polarization, within very small clusters. Evidently, however, as the cluster size increases more off-axis and out-of-plane collisions do occur. FMD simulations of multiple ionization of isolated atoms by lasers [17] suggest that these collisions are as likely to lead to excitation, with subsequent photionization from the excited state, as to direct collisional ionization.

It appears that neither of the ionization mechanisms described in reference [10] is required to explain our results. That is, in the context of our FMD calculations for very small clusters of He atoms, neither the so-called coherent electron motion model (CEMM) [18], nor the ion ignition model(IIM) [11], seems compelling. Instead, the collisional model of reference [6] appears to suffice. One should realize, however, that actual experiments have employed large atoms which can become very highly charged. This is significant since the continuum lowering in bulk plasma is given approximately by the expression Z_{eff}/R, where Z_{eff} is the average degree of ionization and R is the average distance between ions. For our case, this implies a continuum lowering of only ~ 0.2 a.u., much less than the binding energy of the He^+ ion in its ground state. Thus, our calculation is not a fair test of the IIM.

Also, as described in reference [13], a plasma resonance might lead to enhanced absorption of laser energy when $n_e/n_{crit} = 3$, where n_e is the free electron density inside the cluster and $n_{crit} = \sqrt{4\pi n_e}$ in a.u. This resonance occurs for spherical plasmas, and the laser energy absorbed goes into free electron collective motion. If we assume in our simulations that, at some stage of the laser pulse, one electron has been ionized per He atom, and that the average interatom spacing has not yet changed appreciably from its initial value of 6.6 a.u., then the frequency at which such critical absorption would occur is $\omega_{crit} = \sqrt{4\pi n_e/3} = 0.12$ a.u. This implies that a plasma resonance might be relevant under the conditions of our simulations.

We estimated the expected size of this effect using simple formulae derived to describe resonance absorption near an interface with bulk plasma, in one-dimension; reference [19], equations (4.9) through (4.13). The energy absorbed was estimated to be

$$U_{abs} = \frac{\pi}{4}\frac{L^3}{\lambda}cE_d^2 \tag{10}$$

where L is the linear dimension of the (square) microplasma, λ is the wavelength of incident light, and E_d is the amplitude of the light wave inside the plasma, related to the free-space amplitude by

$$E_d \approx 0.5(\frac{\lambda}{L})^{\frac{1}{6}}E_{FS}. \tag{11}$$

These formulae suggest that the total energy which could be absorbed resonantly by our $N \times N \times N$ (micro)plasmas would be approximately 0.05 a.u. for $2\times2\times2$, 0.30 a.u. for $3\times3\times3$, and 0.90 a.u. for $4\times4\times4$. Even if all this energy were to appear for a time as the kinetic energy of a single electron, it would still be too little energy to dominate the ionization state of these microplasmas.

Finally, since our simulations did not extend to times greater than the laser pulse length (80 fs), and the clusters were very small, possible hydrodynamic effects appearing during cluster expansion [13] were unobservable. Instead ionized electrons, after colliding with neighbor ions once or twice, quickly exited the cluster. The cluster rapidly accumulated net positive charge and began to expand due to mutual Coulomb repulsion of the ions.

4 Summary

We have simulated the multiple ionization of helium atoms, formed into small clusters, and interacting with a short pulse of long wavelength laser radiation, using the method known as Fermion Molecular Dynamics (FMD). Results suggest the dominant effect of laser-induced electron collisions, with ions or atoms other than the parent ion, on the double ionization probability in small clusters. This appears to be a vindication of the ideas first proposed in reference [6]. for single atoms, and an extension of their relevance into the multiatom domain. It is important to note that the maximum energy of recollision within the cluster is $8U_p$, and not just $3.2U_p$, as it is for an isolated ion. Moreover, this recollisional energy is accessible within clusters for both linearly and circularly polarized laser beams.

Future FMD studies, for larger systems, will endeavor to assess the role played by the evolving *longitudinal* electric field, set up by ions and free electrons inside the cluster, on the multiple ionization probability of a "test ion"; *i.e.*, above and beyond the dominant role played by the very rapidly varying electric fields arising in laser-induced collisions. As we have argued, the components of this many-particle generated, slowly varying, field could play a significant role in ionization for somewhat larger clusters than those which we have simulated here. The important effect of neighbor ions on ionization dynamics has already been suggested in reference [11] (the Ion Ignition Model). In bulk plasma this is usually called pressure ionization. Of comparable importance in bulk plasma can be the screening of outer electrons bound to test ions by free electrons, known as continuum lowering. Both of these contributors to the internal cluster electric field could affect ionization dynamics. We will attempt to measure them directly in future FMD simulations. The correct assessment of the role played by resonance absorption on ionization, in larger systems, will require the coupling of a Maxwell solver to our FMD system. We have argued earlier that the role played by resonance absorption is negligible for very small systems.

Appendix A

For an electron moving under the influence of an oscillating electric field, homogeneous in space and pointing initially in the z-direction, the equation of motion is

$$\ddot{z}(t) = -E_0\cos(\omega t + \phi) \tag{12}$$

where ϕ is the phase of the field at $t = 0$. The corresponding electron velocity is

$$\dot{z}(t) = -(E_0/\omega)[\sin(\omega t + \phi) - \sin\phi] \tag{13}$$

and the electron displacement is

$$z(t) = -(E_0/\omega^2)[\cos\phi - \cos(\omega t + \phi) - \omega t\sin\phi] \tag{14}$$

assuming both that $\dot{z}(0) = 0$ and $z(0) = 0$. For the ionization problem, $t = 0$ is the instant at which the electron emerges into the continuum.

In order for a recollision between an ionized electron and its parent ion to occur, it must be that $z(t) = 0$, for some $t > 0$. For a fixed value of ϕ, this condition determines ωt. In fact, ϕ and ωt must be related by

$$\tan\phi = (\cos\omega t - 1)/(\sin\omega t - \omega t). \tag{15}$$

Then, subject to this condition, upon maximizing $|\dot{z}(t)|$, one finds that

$$|\dot{z}(t)|_{max} = 1.260(E_0/\omega) \tag{16}$$

for $\phi = 0.010\pi$ and $\omega t = 1.30\pi$; *i.e.*, within the first cycle following release. The corresponding maximum electron kinetic energy is

$$|\dot{z}(t)|^2_{max}/2 = 3.17U_p \tag{17}$$

where U_p is the electron quiver energy $U_p = (E_0\omega)^2/4$. The preceding is similar to the argument first given in reference [6]. For recollision during the second cycle following release, the maximum kinetic energy is $2.40U_p$, achieved for $\phi = 0.033\pi$ and $\omega t = 3.43\pi$; for recollision during the third cycle, the maximum kinetic energy is $2.25U_p$; *etc.* The average kinetic energy of recollision is U_p (average over many cycles.) Of course, this analysis ignores the effect of the Coulomb forces.

In a cluster, a collision between an ionized electron and some ion other than the parent may occur; in this case, $z(t) \neq 0$. Now ϕ and ωt are related by

$$\begin{aligned}\cos\phi &= (-ab - c\sqrt{b^2 + c^2 - a^2})/(b^2 + c^2)\\ \sin\phi &= (-ac + b\sqrt{b^2 + c^2 - a^2})/(b^2 + c^2)\end{aligned} \tag{18}$$

where

$$\begin{aligned}a &= R\omega^2/E_0\\ b &= 1 - \cos\omega t\\ c &= \sin\omega t - \omega t\end{aligned} \tag{19}$$

and $R = |z(t)|$. Subject to this condition on ϕ, we then maximized $|\dot{z}(t)|$ at each value of R. Results are displayed in Figure 5 (solid curve). Now larger values of the electron kinetic energy may be generated than was the case for the isolated atom. In fact, energies up to $8U_p$ are now possible. Specifically, maxima occur at the energies $KE/U_p = 7.27$, 7.84, 7.94, 7.97, ..., for the displacements $\omega^2 R/E_0 = 2.29$, 5.01, 8.05, 11.14, ...

For circular polarization, the equations of motion are

$$\begin{aligned}\ddot{x}(t) &= -E_0\cos(\omega t + \phi)\\ \ddot{y}(t) &= -E_0\sin(\omega t + \phi)\end{aligned} \tag{20}$$

with the velocities

$$\begin{aligned}\dot{x}(t) &= -(E_0/\omega)[\sin(\omega t + \phi) - \sin\phi]\\ \dot{y}(t) &= -(E_0/\omega)[\cos\phi - \cos(\omega t + \phi)]\end{aligned} \tag{21}$$

and the displacements

$$\begin{aligned}x(t) &= -(E_0/\omega^2)[\cos\phi - \cos(\omega t + \phi) - \omega t\sin\phi]\\ y(t) &= -(E_0/\omega^2)[\omega t\cos\phi + \sin\phi - \sin(\omega t + \phi)].\end{aligned} \tag{22}$$

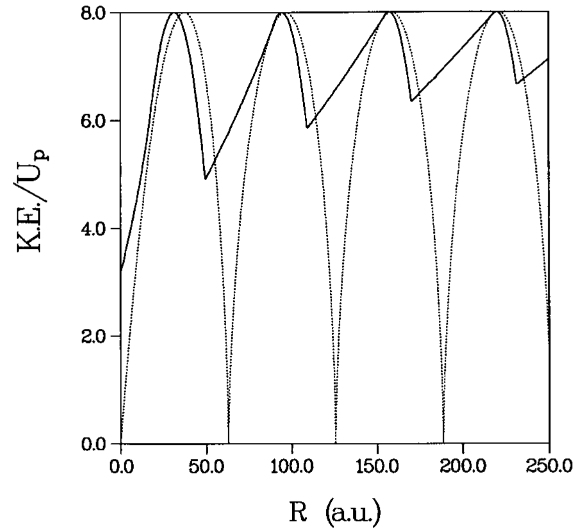

Fig. 5. Maximum kinetic energy of continuum electron, in units of U_p, *vs.* the distance R between the parent ion and the target ion, for linear polarization (solid line), and circular polarization (dotted line), and for $\omega = 0.1$ a.u.; see Appendix A.

However, the time dependent kinetic energy is independent of ϕ, as is the quantity $r(t) = [x(t)^2 + y(t)^2]^{1/2}$. The kinetic energy is just

$$[\dot{x}^2(t) + \dot{y}^2(t)]/2 = (E_0/\omega)^2[1 - \cos(\omega t)] \tag{23}$$

with maxima at $\omega t = (2n+1)\pi$, for $n = 0, 1, 2, ...$; *i.e.*, all at $8U_p$. None of these maxima are accessible at $r(t) => 0$. In fact, since

$$r^2(t) = 2(E_0^2/\omega^4)[1 + \omega^2 t^2/2 - \cos(\omega t) - \omega t\sin(\omega t)] \tag{24}$$

at kinetic energy maxima the displacements are given by

$$r(t) = 2(E_0/\omega^2)[1 + (n + 1/2)^2\pi^2]^{1/2}; \tag{25}$$

i.e., at the values $\omega^2 r(t)/E_0 = 3.72$, 9.63, 15.83, 22.08, ... Generally, the maximum kinetic energy for circular polarization takes on the values displayed in Figure 5 (dotted curve).

References

1. D. Fittinghoff, P. Bolton, B. Chang, K. Kulander, Phys. Rev. Lett. **69**, 2642 (1992).
2. K. Kondo, A. Sagisaka, T. Tamida, Y. Nabekawa, S. Watanabe, Phys. Rev. A **48**, R2531 (1993). In these experiments wavelength dependence was demonstrated: Enhanced two electron emission from isolated He atoms appeared only at the longest wavelengths.
3. B. Walker, E. Mevel, B. Yang, P. Berger, J.P. Chamberet, A. Antonetti, L.F. Dimauro, P. Agostini, Phys. Rev. A **48**, R894 (1993). In these experiments polarization dependence was demonstrated: Enhanced two electron emission from isolated He or Xe atoms was reduced by changing from linear to circular polarization.

4. S. Augst, A. Talebpour, S.L. Chin, Y. Beaudoin, M. Chaker, Phys. Rev. A **52**, R917 (1995). In these experiments enhanced two and three electron emission was observed for Ar, at low laser intensities with linear polarization.
5. P. Lambropoulos, Phys. Rev. Lett. **55**, 2141 (1985). If the laser pulse is long enough and its peak intensity high enough, then the first electron ionizes well before the peak of the pulse has been reached. The second electron, being more tightly bound, ionizes later as the pulse moves closer to peak intensity. By this time, the first electron will generally have moved off to large distances from the ion, so that electron-electron interaction plays no essential role in the second ionization. The two ionizations are said to be independent of each other and sequential in time.
6. P. Corkum, Phys. Rev. Lett. **71**, 1994 (1993).
7. C.L. Kirschbaum, L. Wilets, Phys. Rev. A **21**, 834 (1980).
8. P.B. Lerner, K.J. LaGattuta, J.S. Cohen, Phys. Rev. A **49**, R12 (1994).
9. P.B. Lerner, K.J. LaGattuta, J.S. Cohen, J. Opt. Soc. Am. B **13**, 96 (1996).
10. E.M. Snyder, S.A. Buzza, A.W. Castleman, Jr., Phys. Rev. Lett. **77**, 3347 (1996).
11. C. Rose-Petruck, K.J. Schafer, C.P.J. Barty, Proc. SPIE Int. Soc. Opt. Eng. **2523**, 272 (1995).
12. T. Ditmire, T. Donnelly, R. Falcone, M. Perry, Phys. Rev. Lett. **75**, 3122 (1995).
13. T. Ditmire, T. Donnelly, A. Rubenchik, R. Falcone, M. Perry, Phys. Rev. A **53**, 3379 (1996).
14. T. Ditmire, J. Tisch, E. Springate, M. Mason, N. Hay, J. Marangos, M. Hutchinson, Phys. Rev. Lett. **78**, 2732 (1997).
15. T. Ditmire, R. Smith, J. Tisch, M. Hutchinson, Phys. Rev. Lett. **78**, 3121 (1997).
16. D.W. Wasson, S.E. Koonin, Phys. Rev. A **39**, 5676 (1989).
17. K. LaGattuta, J. Cohen (unpublished).
18. B.D. Thompson, A. McPherson, K. Boyer, C.K. Rhodes, J. Phys. B **27**, 4391 (1994).
19. W.L. Kruer, *The physics of laser plasma interaction* (Addison-Wesley, 1988).