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LETTER TO THE EDITOR

Two-photon double ionization of He and H^-

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Abstract. Two-photon absorption processes resulting in two-electron ionization of He and H^- are calculated using a time-dependent close-coupling expansion on a two-dimensional radial lattice. The ground state atom or ion is subjected to a 10-cycle pulse of linearly polarized light with peak intensities ranging from 10^{13} to 10^{16} W cm^{-2}. Probabilities are calculated for one-photon single ionization, two-photon single ionization and two-photon double ionization. The ratio of non-sequential double ionization to single ionization following two-photon absorption is found to be somewhat larger than the similar ratio following one-photon absorption.

Due to the development of bright light synchrotron experiments, correlated photoionization processes in two-electron atomic systems have received extensive theoretical attention. The most popular methods for handling double photoionization are based on many-body perturbation theory (Carter and Kelly 1981, Hino *et al* 1994), initial-state dipole response functions (Proulx and Shakeshaft 1993, Pont and Shakeshaft 1995, Forrey *et al* 1997), asymptotically correlated final states (Maulbetsch and Briggs 1992, Teng and Shakeshaft 1993, Andersson and Burgdorfer 1993, Kornberg and Miraglia 1993), hyperspherical close-coupling theory (Tang and Shimamura 1995, Qiu *et al* 1998), convergent close-coupling theory (Kheifets and Bray 1996) and R-matrix pseudo-state theory (Meyer and Greene 1994, Meyer *et al* 1997, Marchalant and Bartschat 1997, Gorczyca and Badnell 1997). Recently, a method based on the time propagation of a coupled set of two-dimensional partial differential equations on a numerical lattice has been applied to the single photoionization of helium (Pindzola and Robicheaux 1998). The lattice results for the ratio of double to single photoionization in the energy range from 90–200 eV are in very good agreement with the most accurate previous theories and the most recent synchrotron experiments (Levin *et al* 1996, Dorner *et al* 1996, Samson *et al* 1998).

In this letter we extend the same time-dependent close-coupling method to the calculation of correlated multiphoton ionization processes in two-electron atomic systems. We begin by choosing wavelengths short enough so that two-photon absorption is above the threshold for complete fragmentation of the two-electron atom. Our choice of such wavelengths is based on a desire to stand clear of all intermediate resonances and to limit our close-coupled calculations to fairly low angular momentum. Our goal in this letter is to find the ratio of non-sequential double ionization to single ionization following two-photon absorption, which is the exact analogue of the one-photon absorption calculations and experiments.

In contrast, theoretical work to date has concentrated on much longer wavelengths where two-photon absorption is only above the first ionization threshold (Proulx *et al* 1994, van der Hart 1994, Zhang and Lambropoulos 1995, Parker *et al* 1996, Nikolopoulos and Lambropoulos 1997). Parker *et al* (1996) examined ionization yields above the threshold for the complete fragmentation of helium, but this involved a minimum of 12 photons. Although they calculated double to single ionization ratios, the likelihood of sequential processes involving ionization to He^+ or resonant excitation of doubly excited states of He is extremely high. Thus, they really did not furnish any direct information on the ratio of non-sequential double ionization to single ionization following the 12-photon absorption. The situation on the experimental side is the same. Double ionization of helium has been studied using short pulse lasers with such long wavelengths that from 16–50 photons are needed to go above the complete fragmentation threshold of 79.0 eV (Fittinghoff *et al* 1992, Kondo *et al* 1993, Walker *et al* 1994).

In the following paragraphs, we report two-photon absorption calculations for He at 45.0 eV and for H^- at 8.0 eV using a 10-cycle pulse of linearly polarized light with peak intensities ranging from 10^{13} to 10^{16} W cm^{-2}. Probabilities are calculated for one-photon single ionization, two-photon single ionization and two-photon double ionization by projecting the time-evolved lattice wavefunction onto a complete set of single-particle states. The ratio of non-sequential double ionization to single ionization following two-photon absorption is found to be somewhat larger than that found in the single-photon ionization calculations and experiments.

The time-dependent Schrödinger equation for a two-electron atom in a strong time-varying electromagnetic field is given by (in atomic units)

$$\mathrm{i}\frac{\partial\Psi(\boldsymbol{r}_1,\boldsymbol{r}_2,t)}{\partial t} = (H_{\mathrm{atom}} + H_{\mathrm{rad}})\Psi(\boldsymbol{r}_1,\boldsymbol{r}_2,t) \tag{1}$$

where the non-relativistic Hamiltonian for the atom is given by

$$H_{\mathrm{atom}} = -\frac{1}{2}\nabla_1^2 - \frac{1}{2}\nabla_2^2 - \frac{Z}{r_1} - \frac{Z}{r_2} + \frac{1}{|\boldsymbol{r}_1 - \boldsymbol{r}_2|} \tag{2}$$

and the Hamiltonian for a linearly polarized radiation field is given by

$$H_{\mathrm{rad}} = E(t)(r_1\cos\theta_1 + r_2\cos\theta_2)\cos\omega t. \tag{3}$$

In equation (2) the atomic number is Z, while in equation (3) the electric field amplitude is $E(t)$ and the radiation frequency is ω. Following a standard procedure found in time-independent scattering theory (Temkin 1962, Shertzer and Botero 1994, Wang and Callaway 1994), the total wavefunction for a given LS symmetry is first expanded in coupled spherical harmonics and then substituted into equation (1) to derive the the time-dependent partial differential equations given by

$$\begin{aligned}\mathrm{i}\frac{\partial P^{LS}_{l_1l_2}(r_1,r_2,t)}{\partial t} &= T_{l_1l_2}(r_1,r_2)\,P^{LS}_{l_1l_2}(r_1,r_2,t) + \sum_{l_1',l_2'} V^{L}_{l_1l_2,l_1'l_2'}(r_1,r_2)\,P^{LS}_{l_1'l_2'}(r_1,r_2,t)\\ &\quad + \sum_{L'}\sum_{l_1',l_2'} W^{LL'}_{l_1l_2,l_1'l_2'}(r_1,r_2,t)\,P^{L'S}_{l_1'l_2'}(r_1,r_2,t).\end{aligned} \tag{4}$$

In equation (4) the kinetic and nuclear energy operator is $T_{l_1l_2}$, the electron–electron interaction energy operator is $V^{L}_{l_1l_2,l_1'l_2'}$ and the time-varying radiation field operator is $W^{LL'}_{l_1l_2,l_1'l_2'}$ (see Pindzola and Robicheaux 1998 for detailed expressions). The same set of time-dependent close-coupling equations were used by Parker *et al* (1996) to study harmonic generation and double ionization of helium at a photon energy of 5.44 eV.

The time-dependent close-coupled equations are solved by discretization of all radial wavefunctions and operators on a two-dimensional lattice. For easy implementation on distributed-memory parallel computers, low-order finite-difference methods are employed with uniform mesh spacing. The two-dimensional radial wavefunctions are partitioned in rectangular strips over the processors, a particularly simple example of domain decomposition. The key to our numerical solution is the use of a second-order difference approximation for the time evolution of the radial wavefunctions. The method is ideal for distributed memory parallel computers since it involves only one matrix-vector multiplication at each time step.

The ground states of He and H^- are found by relaxation of the time-dependent Schrödinger equation in imaginary time. The ground state wavefunctions are then time evolved by solving the time-dependent Schrödinger equation in real time subject to a 10-cycle pulse of linearly polarized light whose electric field amplitude is given by

$$E(t) = E_0 \sin^2\left(\frac{\pi t}{T}\right) \tag{5}$$

and whose total pulse time is $T = 20\pi/\omega$. For He we employ a 200 × 200 point lattice with a mesh spacing of $\Delta r = 0.2$, so that the maximum radial extent is $R = 40$ au. For H^- we keep the same mesh spacing, but employ a 400 × 400 point lattice with $R = 80$ au. Following the radiation pulse, ionization probabilities are extracted by projecting the various LS components of the total wavefunction onto a complete set of single-particle states for He^+ and H, respectively. For example, the probability for one-photon single ionization leaving the atom or ion in the 1s state is given by

$$\mathcal{P}_{1\mathrm{s}\to kl} = 2\left(\int \mathrm{d}r_2 |\langle P_{1\mathrm{s}}(r_1)|P_{\mathrm{sp}}^{^1\mathrm{P}}(r_1, r_2, T)\rangle|^2 - \sum_{n\mathrm{p}} |\langle P_{1\mathrm{s}}(r_1) P_{n\mathrm{p}}(r_2)|P_{\mathrm{sp}}^{^1\mathrm{P}}(r_1, r_2, T)\rangle|^2\right). \tag{6}$$

The single-particle eigenstates $P_{nl}(r)$ are obtained by diagonalizing the Schrödinger equation for He^+ or H on a one-dimensional lattice with the same uniform mesh and radial extent as that used for the two-electron atom. A number of additional probabilities may be found by projection, including two-photon single and double ionization involving the ^{1}S and ^{1}D components of the total wavefunction.

To check our numerical methods we first examined the two-photon ionization of He in various approximations. The ground state wavefunction is chosen to be

$$\Phi_0(\boldsymbol{r}_1, \boldsymbol{r}_2) = \frac{1}{r_1 r_2} P_{\mathrm{ss}}^{^1\mathrm{S}} Y_{\mathrm{ss}}^{\mathrm{S}} \tag{7}$$

and we time-evolve a six-channel wavefunction given by

$$\Psi(\boldsymbol{r}_1, \boldsymbol{r}_2, t) = \frac{1}{r_1 r_2}\left(P_{\mathrm{ss}}^{^1\mathrm{S}} Y_{\mathrm{ss}}^{\mathrm{S}} + P_{\mathrm{sp}}^{^1\mathrm{P}} Y_{\mathrm{sp}}^{\mathrm{P}} + P_{\mathrm{ps}}^{^1\mathrm{P}} Y_{\mathrm{ps}}^{\mathrm{P}} + P_{\mathrm{sd}}^{^1\mathrm{D}} Y_{\mathrm{sd}}^{\mathrm{D}} + P_{\mathrm{ds}}^{^1\mathrm{D}} Y_{\mathrm{ds}}^{\mathrm{D}} + P_{\mathrm{pp}}^{^1\mathrm{D}} Y_{\mathrm{pp}}^{\mathrm{D}}\right) \tag{8}$$

where $Y_{l_1 l_2}^{L}$ is a coupled spherical harmonic in the angular coordinates. If the electron–electron interaction operator $V_{l_1 l_2, l_1' l_2'}^{L}$ is set to zero in equation (4), the ground state of He is a product of He^+ ground state wavefunctions with a total energy of −104.8 eV (i.e. a 52.4 eV ionization potential for He and another 52.4 eV to ionize He^+). Remember that the mesh spacing of the lattice is $\Delta r = 0.2$, so we do not get the exact ionization potential of 54.4 eV. For two-photon ionization of He in the 1s1s uncorrelated ground state, we choose $\omega = 60$ eV and a peak field intensity of 5.0×10^{15} W cm^{-2}. The various ionization probabilities following the radiation pulse are given in table 1. One-photon single ionization is found in the ^{1}P component at 0.238, while the ^{1}D component has 12 times as much two-photon double ionization as two-photon single ionization. This is certainly

Table 1. Multiphoton ionization probabilities for He models (intensities I in 10^{15} W cm^{-2}).

Probabilities	He(1s1s) model $\omega = 60$ eV $I = 5.0$	He(1s2s) model $\omega = 35$ eV $I = 3.0$	He(1s^2) $\omega = 45$ eV $I = 3.0$
^{1}S normalization	0.746	0.817	0.703
^{1}S total single ionization	0.000	0.001	0.008
^{1}S total double ionization	0.000	0.000	0.000
^{1}P normalization	0.242	0.152	0.267
^{1}P total single ionization	0.238	0.080	0.267
^{1}P total double ionization	0.000	0.000	0.000
^{1}D normalization	0.013	0.031	0.030
^{1}D total single ionization	0.001	0.031	0.027
^{1}D total double ionization	0.012	0.000	0.003

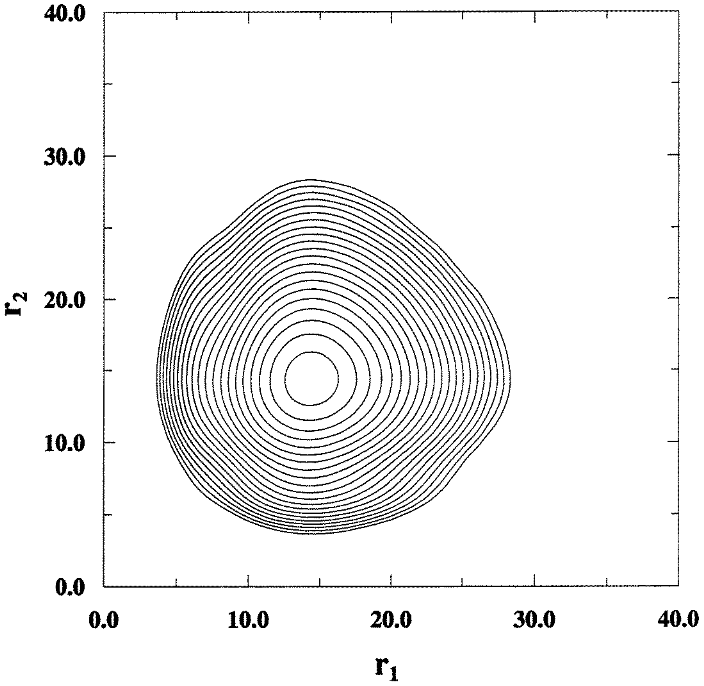

Figure 1. Multiphoton ionization for the He(1s1s) model. Contour map of the radial probability density for the (pp)^{1}P channel near the end of a 10-cycle radiation pulse.

to be expected, since the two-photon double ionization is due to the sequential process of 1s1s ^{1}S → 1skp ^{1}P single ionization of He, followed by 1skp ^{1}P → kpk'p ^{1}D single ionization of He$^+$. In figure 1 we plot $|P_{\mathrm{pp}}^{^1\mathrm{D}}|^2$ at a time near the end of the radiation pulse and find a strong peak along $r_1 = r_2$, confirming the ejection of two electrons in that channel.

Next we examined the two-photon ionization of He in the 1s2s uncorrelated excited state. Since the total energy is −65.9 eV (i.e. a 13.5 eV ionization potential for He* and another 52.4 eV to ionize He$^+$), we choose $\omega = 35$ eV and a peak field intensity of 3.0×10^{15} W cm^{-2}. The various ionization probabilities following the radiation pulse are again given in table 1. One-photon single ionization is found in the ^{1}P component at

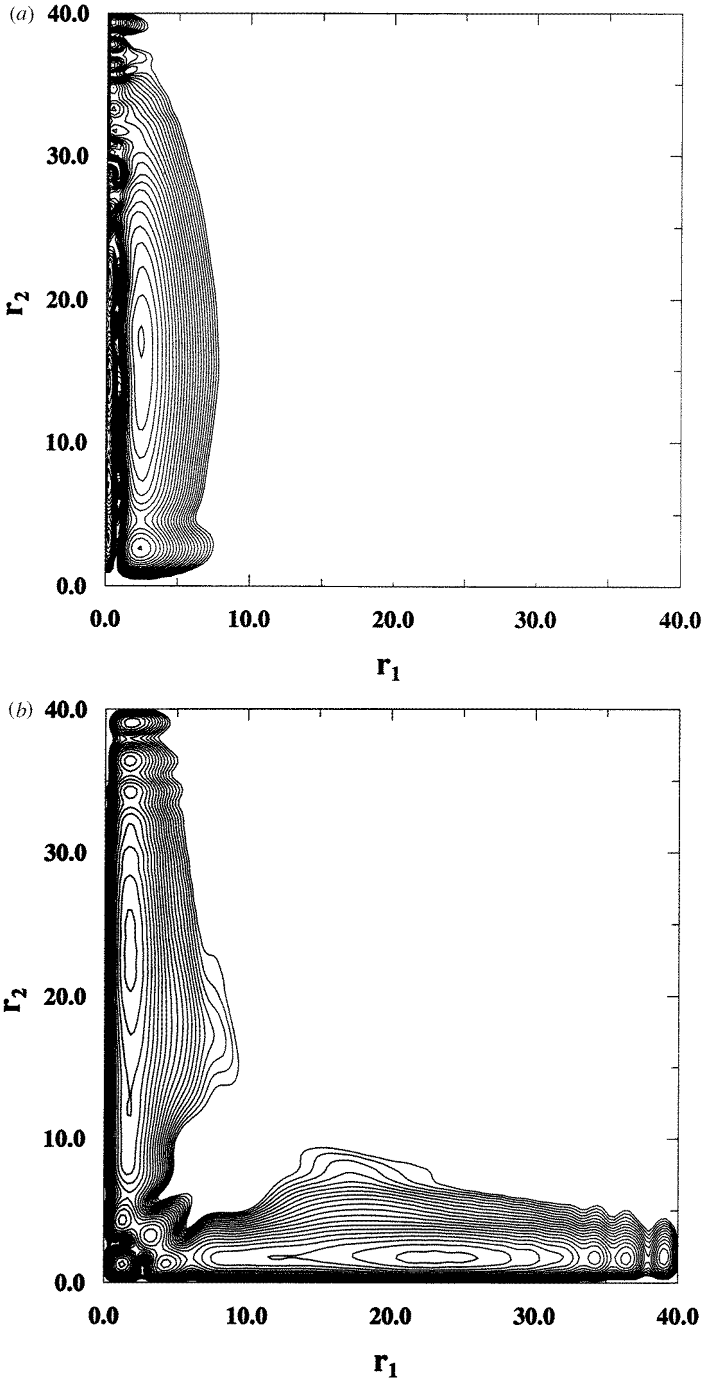

Figure 2. Multiphoton ionization for the He(1s2s) model. Contour maps of the radial probability density for the (*a*) $(sd)^1D$ and (*b*) $(pp)^1D$ channels near the end of a 10-cycle radiation pulse.

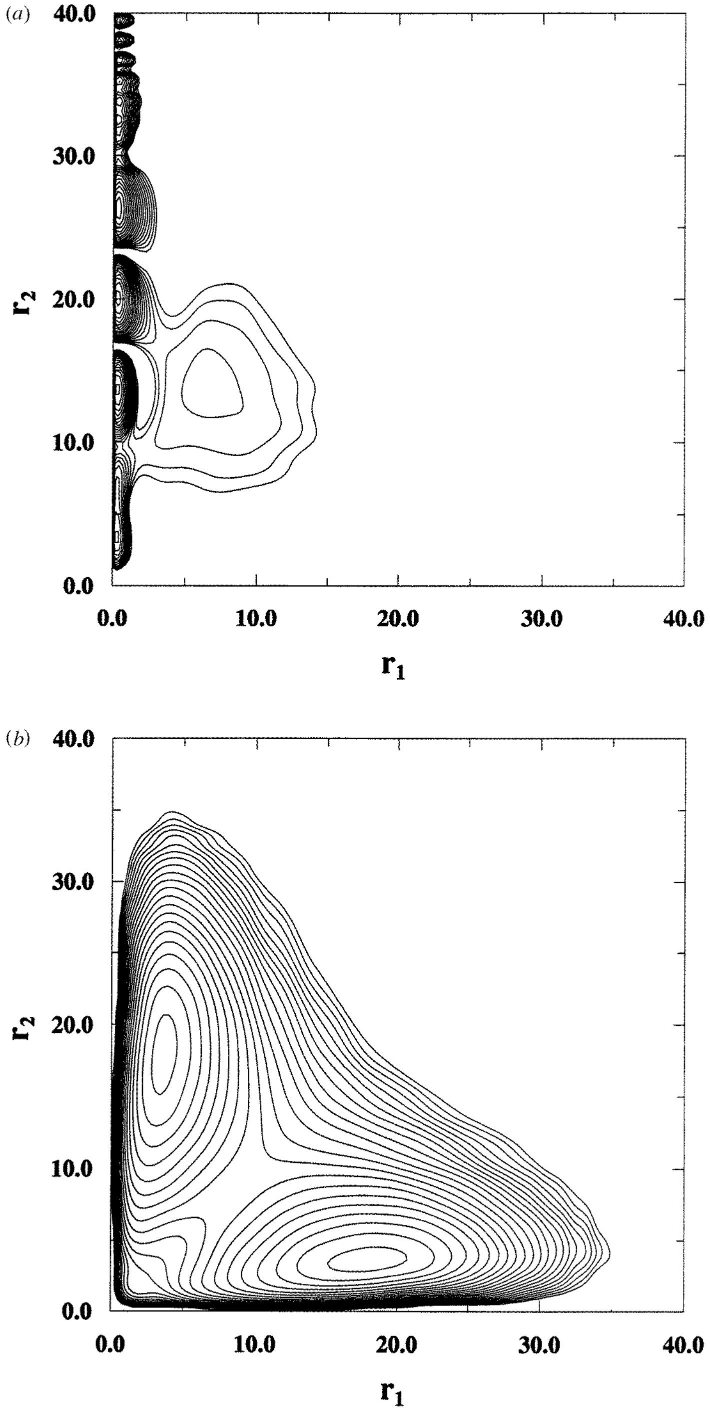

Figure 3. Multiphoton ionization for the He(1s^2). Contour maps of the radial probability density for the (*a*) (sd)^{1}D and (*b*) (pp)^{1}D channels near the end of a 10-cycle radiation pulse.

0.080, while two-photon single ionization is found in the ^{1}D component at 0.031. There is no two-photon double ionization for two reasons. First there is no sequential pathway, since ionization of He$^+$ takes a minimum of two 35.0 eV photons itself. Second there is no non-sequential path, since the electron–electron interaction has been dropped from the Hamiltonian. In figure 2 we plot $|P_{\mathrm{sd}}^{^1\mathrm{D}}|^2$ and $|P_{\mathrm{pp}}^{^1\mathrm{D}}|^2$ at a time near the end of the radiation pulse and find no two-electron ejection.

Finally, we examined the two-photon ionization of He in the 1s^2 correlated ground state. The total energy is -75.1 eV (i.e. a 22.7 eV ionization potential for He and another 52.4 eV to ionize He$^+$) and we choose $\omega = 45$ eV and a peak field intensity of 3.0×10^{15} W cm^{-2}. The various ionization probabilities following the radiation pulse for this case are given in the final column of table 1. One-photon single ionization is found in the ^{1}P component at 0.267, two-photon single ionization is found in the ^{1}D component at 0.027 and two-photon double ionization is found in the ^{1}D component at 0.003. Since there is no sequential pathway, like the He(1s2s) case, the two-photon double ionization is made possible by the addition of the electron–electron interaction into the Hamiltonian. In figure 3 we plot $|P_{\mathrm{sd}}^{^1\mathrm{D}}|^2$ and $|P_{\mathrm{pp}}^{^1\mathrm{D}}|^2$ and find evidence for two-electron ejection along $r_1 = r_2$, especially in the $P_{\mathrm{pp}}^{^1\mathrm{D}}$ channel.

Proceeding to full calculations for the two-photon ionization of both He and H$^-$, the ground state wavefunction is chosen to be

$$\Phi_0(\boldsymbol{r}_1, \boldsymbol{r}_2) = \frac{1}{r_1 r_2}\left(P_{\mathrm{ss}}^{^1\mathrm{S}} Y_{\mathrm{ss}}^{\mathrm{S}} + P_{\mathrm{pp}}^{^1\mathrm{S}} Y_{\mathrm{pp}}^{\mathrm{S}} + P_{\mathrm{dd}}^{^1\mathrm{S}} Y_{\mathrm{dd}}^{\mathrm{S}}\right). \tag{9}$$

For a lattice spacing of $\Delta r = 0.2$, the ground state energy of He is found to be -75.8 eV, while the ground state energy of H$^-$ is found to be -14.2 eV. The energy for He is within 5% of the exact value of -79.0 eV, while the energy for H$^-$ is very close to the exact value of -14.4 eV. A lattice spacing of $\Delta r = 0.1$ improves the energy for He to -78.1 eV. We time evolve a 15-channel wavefunction given by

$$\begin{aligned}\Psi(\boldsymbol{r}_1, \boldsymbol{r}_2, t) = \frac{1}{r_1 r_2}\big(&P_{\mathrm{ss}}^{^1\mathrm{S}} Y_{\mathrm{ss}}^{\mathrm{S}} + P_{\mathrm{pp}}^{^1\mathrm{S}} Y_{\mathrm{pp}}^{\mathrm{S}} + P_{\mathrm{dd}}^{^1\mathrm{S}} Y_{\mathrm{dd}}^{\mathrm{S}} + P_{\mathrm{sp}}^{^1\mathrm{P}} Y_{\mathrm{sp}}^{\mathrm{P}} + P_{\mathrm{ps}}^{^1\mathrm{P}} Y_{\mathrm{ps}}^{\mathrm{P}} + P_{\mathrm{pd}}^{^1\mathrm{P}} Y_{\mathrm{pd}}^{\mathrm{P}}\\ &+ P_{\mathrm{dp}}^{^1\mathrm{P}} Y_{\mathrm{fp}}^{\mathrm{P}} + P_{\mathrm{df}}^{^1\mathrm{P}} Y_{\mathrm{df}}^{\mathrm{P}} + P_{\mathrm{fd}}^{^1\mathrm{P}} Y_{\mathrm{fd}}^{\mathrm{P}} + P_{\mathrm{sd}}^{^1\mathrm{D}} Y_{\mathrm{sd}}^{\mathrm{D}} + P_{\mathrm{ds}}^{^1\mathrm{D}} Y_{\mathrm{ds}}^{\mathrm{D}} + P_{\mathrm{pp}}^{^1\mathrm{D}} Y_{\mathrm{pp}}^{\mathrm{D}}\\ &+ P_{\mathrm{pf}}^{^1\mathrm{D}} Y_{\mathrm{pf}}^{\mathrm{D}} + P_{\mathrm{fp}}^{^1\mathrm{D}} Y_{\mathrm{fp}}^{\mathrm{D}} + P_{\mathrm{dd}}^{^1\mathrm{D}} Y_{\mathrm{dd}}^{\mathrm{D}}\big).\end{aligned} \tag{10}$$

For two-photon ionization of He we choose $\omega = 45$ eV and a range of peak intensities from 10^{15} to 10^{16} W cm^{-2}. Ionization probabilities are given in table 2. A general rise in all the ionization probabilities is seen as the intensity increases. Two-photon single ionization is found in both the ^{1}S and ^{1}D components, and two-photon double ionization tracks at about 10% of the two-photon single ionization. For two-photon ionization of H$^-$ we choose $\omega = 8$ eV and a range of peak intensities from 10^{13} to 10^{14} W cm^{-2}. Ionization probabilities are given in table 3. The ratio of two-photon double ionization to two-photon single ionization for H$^-$ is generally somewhat less than that found for He.

At the higher intensities for both He and H$^-$, the double ionization in the ^{1}P component begins to contribute to the overall ionization process. We repeated our calculations for He at 10^{16} W cm^{-2} after expanding the wavefunction of equation (10) to include six more ^{1}F channels. The ionization probabilities for the ^{1}S, ^{1}P and ^{1}D components remained approximately the same as reported in table 2, while the ^{1}F component had almost equal amounts of three-photon single ionization and three-photon double ionization. A large contribution to the odd-parity double-ionization components is coming from the sequential process of one-photon ionization of He, followed by two-photon ionization of He$^+$. Unlike

Table 2. Multiphoton ionization probabilities for He (intensities I in 10^{15} W cm^{-2}).

Probabilities	He $\omega = 45$ eV $I = 1.0$	He $\omega = 45$ eV $I = 3.0$	He $\omega = 45$ eV $I = 5.0$	He $\omega = 45$ eV $I = 10.0$
^{1}S normalization	0.895	0.729	0.607	0.433
^{1}S total single ionization	0.014	0.024	0.042	0.104
^{1}S total double ionization	0.002	0.003	0.004	0.011
^{1}P normalization	0.101	0.244	0.332	0.410
^{1}P total single ionization	0.101	0.244	0.332	0.407
^{1}P total double ionization	0.000	0.000	0.000	0.003
^{1}D normalization	0.004	0.027	0.061	0.156
^{1}D total single ionization	0.004	0.023	0.052	0.129
^{1}D total double ionization	0.000	0.003	0.009	0.027

Table 3. Multiphoton ionization probabilities for H$^-$ (intensities I in 10^{13} W cm^{-2}).

Probabilities	H$^-$ $\omega = 8$ eV $I = 1.0$	H$^-$ $\omega = 8$ eV $I = 3.0$	H$^-$ $\omega = 8$ eV $I = 5.0$	H$^-$ $\omega = 8$ eV $I = 10.0$
^{1}S normalization	0.904	0.745	0.622	0.420
^{1}S total single ionization	0.011	0.013	0.017	0.034
^{1}S total double ionization	0.002	0.002	0.002	0.003
^{1}P normalization	0.094	0.239	0.342	0.481
^{1}P total single ionization	0.094	0.238	0.338	0.463
^{1}P total double ionization	0.000	0.001	0.003	0.018
^{1}D normalization	0.002	0.015	0.036	0.099
^{1}D total single ionization	0.002	0.014	0.034	0.091
^{1}D total double ionization	0.000	0.001	0.002	0.007

the two-photon ionization of He at $\omega = 45$ eV and of H$^-$ at $\omega = 8$ eV, the three-photon ionization is a mixture of sequential and non-sequential processes.

In summary, we have calculated two-photon double-ionization processes in He and H$^-$ by direct solution of the time-dependent Schrödinger equation on a two-dimensional radial lattice. Frequencies were chosen so that two-photon absorption is above the complete fragmentation threshold, and intensities were chosen so that one-photon single ionization is the dominant process. When the much weaker two-photon ionization processes are examined, the ratio of double ionization to single ionization is found to be surprisingly large. Due to the choice of frequencies, we can identify the two-photon double ionization as a strictly correlated non-sequential electron emission event.

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