Time-dependent effects in the nonsequential ionization of helium at various wavelengths

P.P. CORSO,1 D.G. LAPPAS, 2 AND P.L. KNIGHT 2

1Istituto Nazionale di Fisica della Materia and Dipartimento di Scienze, Fisiche ed Astronomiche dell’Università, via Archirafi 36, 90123, Palermo, Italy
2Optics Section, Blackett Laboratory, Imperial College, London SW7 2BZ, United Kingdom

(Received 25 May 1999; Accepted 25 June 1999)

Abstract

Calculations of the double ionization of a model 1D helium atom interacting with an intense short laser pulse are presented. A substantial signature of nonsequential double ionization is found for all the wavelengths that are considered, from 248 nm to 1064 nm, provided that sufficiently short pulses are employed. We show how one can modify the size of the “knee” structure of the double ionization yield curve by varying the laser pulse duration. The underlying mechanism of the correlated double electron ejection is investigated and discussed.

1. INTRODUCTION

In the last few years, experiments have revealed an unexpectedly high signal for the nonsequential (NS) ionization in multielectron atomic systems subjected to intense laser pulses. The first observation of NS ionization in noble gases was made by L’Huillier et al. (1983). In subsequent experiments by Fittinghoff et al. (1992), with 120 fs pulses from a 614 nm dye laser, and later by Walker et al. (1994), with 160 fs, 780 nm pulses from a Ti:Sapphire laser, very accurate measurements of the ion yields of helium were obtained. In both cases, a rather particular behavior has been observed: while single ionization rates can be very well predicted by a single-active electron (SAE) model of He (Kulander, 1987), the double-ionization rates cannot. The presence of a first saturation regime in the double ionization yield (known as “knee”) is strong evidence of the role played by electron–electron interaction and cannot be predicted by a sequential model such as SAE.

Recently, NS ionization has been experimentally detected also in diatomic molecules by Talebpour et al. (1997), at 800 nm, with a Ti:Sapphire laser. Further experimental efforts have revealed multiple NS ionization of rare gas atoms at various wavelengths [see, e.g., Larochelle et al. (1998) and references therein].

A variety of theoretical approaches have been developed in the last four years in order to explain the very high degree of NS double ionization. S-matrix theory (Faisal et al., 1999; Becker & Faisal, 1999), “exact” fully correlated 1D models (Pindzola et al., 1991; Grobe & Eberly, 1992; Lappas et al., 1996; Lappas & van Leeuwen 1998) and numerical models with partial correlation (Watson et al., 1997; Burnett et al., 1998; Sanpera et al., 1998) have successfully reproduced the knee structure. Other attempts that involve the full 3D dynamics of the two correlated electrons have been made with large scale supercomputers (Dundas et al., 1999), but they have been limited so far to rather high frequencies and a restricted range of intensities. In this work, we use both the exact and the partially correlated models introduced by Lappas and van Leeuwen (1998) and by Watson et al. (1997), in order to investigate the two-electron dynamics for a wide range of laser pulse parameters.

2. EXACTLY SOLVED TWO-ELECTRON MODEL

In order to study the two-electron phenomena and understand the basic physics, an exact treatment of the electron correlation is needed. For this purpose, numerical calculations have been widely used in the past (Pindzola et al., 1991; Grobe & Eberly, 1992; Lappas et al., 1996, 1998). All these works reduce the dimensional complexity of the system and solve the two-electron Schrödinger equation exactly in one space dimension.

Address correspondence and reprint requests to: D.G. Lappas, Optics Section, Blackett Lab., Imperial College, London SW7 2BZ, UK. E-mail: d.lappas@ic.ac.uk
The basic scheme of the two-electron system interacting with an external classical electric field is described by the following Hamiltonian in the minimal coupling form, in atomic units (a.u.):

\[
H(t) = \frac{i}{2}[\left( p_1 - A(t) \right)^2 + \left( p_2 - A(t) \right)^2] + V(x_1) + V(x_2) - V(x_1 - x_2)
\]

where

\[
x_1, x_2 = \text{spatial coordinates of electron 1 and 2 respectively}
\]

\[
p_1, p_2 = \text{their momenta}
\]

\[
A(t) = \text{vector potential of the laser field}
\]

The initial ground state is propagated in time with the above Hamiltonian, according to a standard numerical split-operator technique (Feit et al., 1982; Grobe & Eberly, 1992). \(V(x)\) is the Coulombic potential that describes the interaction of each electron with the core and the electron–electron repulsion. In a 1D model, we employ the following soft-core potential, in order to avoid overestimating these interactions around the origin:

\[
V(x) = \frac{-Z}{\sqrt{1 + x^2}}
\]

which is also known as the “Rochester” potential (Grobe & Eberly, 1992). The two-electron ground state energy is found to be -2.238 a.u. In what follows we will assume the electric field to be linearly polarized and given by:

\[
E(t) = E_0(t)\sin(\omega t) = -\frac{\partial}{\partial t} A(t)
\]

where the envelope function \(E_0(t)\) takes into account the turn on and turn off effects of the laser pulse. The Schrödinger equation for the full two-electron wave function \(\psi(x_1, x_2; t)\) at time \(t\) is then given by:

\[
i \frac{\partial}{\partial t} \psi(x_1, x_2; t) = H(t) \psi(x_1, x_2; t).
\]

We will return to this model in Section 5. In the meantime we will introduce an approximate model that will not only make our calculations easier and faster, but will allow for more flexibility in probing separately the electron–laser and the electron–electron interactions as well.

### 3. APPROXIMATE MEAN-FIELD MODEL

Although the exact model outlined above describes the experimental features in a very good qualitative manner, it is rather computationally demanding, even in one space dimension, and cannot be used easily for very long pulses, very high electron energies or for large regions of space. A first approximation to the previous model has been introduced by Watson et al. (1997). In the intensity regime of the relevant experiments, one of the helium electrons ionizes rapidly, while there is only a small relative probability that the second electron also ionizes. It is then assumed that we can distinguish between the two electrons as if they were identifiable as an external and an internal one. The external electron, because of fast tunneling ionization, can be considered as bound only by a soft-core effective potential, the result of the nuclear attraction and screening. The internal electron is subjected to the nuclear Coulombic potential and to the Coulombic mean field interaction with the external electron. It has been shown (Lappas & van Leeuwen, 1998; Petersilka & Gross, 1999) that if both electrons are treated equally (as in a time-dependent Hartree–Fock or density functional theory approach), the knee structure cannot be reproduced, a fact that indicates the limitations of some standard mean-field approximations.

Here, the total wave function is written as a product of the wave functions of the two electrons, which are now distinguishable. Neglecting the exchange term we simply have (Watson et al., 1997):

\[
\psi(x_E, x_I; t) = \psi_E(x_E; t) \cdot \psi_I(x_I; t)
\]

where \(x_E\) and \(x_I\) refer to the external and internal electron coordinates, respectively.

We then consider two separate one-particle Schrödinger equations. For the external electron we have:

\[
i \frac{\partial}{\partial t} \psi_E(x_E; t) = \left( \frac{(p_E - A(t))^2}{2} + V_E(x_E) \right) \psi_E(x_E; t)
\]

where

\[
V_E(x_E) = -\frac{(Z - 1)}{\sqrt{a_E^2 + x_E^2}}
\]

is the effective potential with \(Z = 2\). We take the soft-core parameter to be \(a_E = 0.697\), so that the ground state of \(V_E\) is matched with the one-electron ionization potential of the real He atom.

The equation for the internal electron contains an additional external field term due to the Coulombic interaction with the external electron:

\[
i \frac{\partial}{\partial t} \psi_I(x_I; t) = \left( \frac{(p_I - A(t))^2}{2} + V_I(x_I; t) \right) \psi_I(x_I; t)
\]

where

\[
V_I(x_I; t) = -\frac{Z}{\sqrt{a_I^2 + x_I^2}} + \int dx_E \frac{\phi^*_E(x_E; t) \cdot \psi_E(x_E; t)}{\sqrt{a_{E,I}^2 + (x_E - x_I)^2}}
\]
In this case, \( a_I = 0.7 \) and \( a_{E,I} = \sqrt{2} \) yield the best match for the case of the real He atom.

The great advantage of this approach is that the two equations of motion are essentially decoupled, because Eq. (6) does not depend on \( x_I \). Therefore, the 2D problem is reduced to two 1D Schrödinger equations which can be easily solved numerically even on a personal computer.

### 4. DEFINITION OF IONIZATION

In our exact model, where the joint two-electron probability distribution can be calculated numerically, the double ionization probability is defined [somewhat arbitrarily, but in a rather useful and realistic way; see also Dundas et al. (1999)] as the total probability that both electrons are at least \( R = 5 \) a.u. away from the origin, at the end of the laser pulse, that is:

\[
P_2 = \int_{|x_I|>R} dx_I \int_{|x_E|>R} dx_E |\psi(x_I, x_E; t_{end})|^2. \tag{10}
\]

Accordingly, the single-ionization probability is defined as the probability that one and only one of the two electrons is within the distance \( R \) from the origin:

\[
P_1 = 2 \int_{|x_I|<R} dx_I \int_{|x_E|>R} dx_E |\psi(x_I, x_E; t_{end})|^2 \tag{11}
\]

where the factor of 2 is due to the symmetry under exchange of the two electrons. In such a model the two electrons are completely indistinguishable and full correlation is considered.

In most of the following, where we use the approximate model to study the response of a He atom subjected to laser pulses of different wavelengths and durations, the above definition should be modified. A first simple definition is related to the distinction we have made between the internal and external electrons: the single ionization probability is derived from the complement of the projection of the propagated external electron wavefunction on to its initial ground state and is indicated by \( P_{E,\text{out}}^{\text{ion}(t)} \). The double ionization probability is deduced from the projection of the propagated internal electron wave function on to its initial ground state and is indicated by \( P_{I,\text{out}}^{\text{ion}(t)} \) (Watson et al., 1997). Therefore:

\[
\begin{align*}
P_{E,\text{out}}^{\text{ion}(t)} &= 1 - \left| \int \psi_E^I(x_E; t) \psi_E(x_E; t_0) \, dx_E \right|^2, \\
P_{I,\text{out}}^{\text{ion}(t)} &= 1 - \left| \int \psi_I^E(x_I; t) \psi_I(x_I; t_0) \, dx_I \right|^2. \tag{12}
\end{align*}
\]

Obviously, the sum of these two ionization probabilities and the probability of the neutral atom is not strictly equal to 1.

Another more accurate definition of ionization probability has been considered. This requires the use of a region in space of radius \( R \) around the atom, inside which the electrons are not considered ionized. The single ionization probability \( P_I(t) \) is given by the probability that the internal electron is inside, times the probability that the external electron is outside, plus the probability the external electron is inside, times the probability the internal electron is outside. The double ionization probability \( P_2(t) \) is given by the probability that the internal electron is outside, times the probability that the external electron is outside too, that is:

\[
\begin{align*}
P_I(t) &= (1 - P_{E,\text{out}}^{\text{ion}(t)})(1 - P_{E,\text{out}}^{\text{ion}(t)}) + P_{I,\text{out}}^{\text{ion}(t)}(1 - P_{E,\text{out}}^{\text{ion}(t)}), \\
P_2(t) &= P_{I,\text{out}}^{\text{ion}(t)}(1 - P_{E,\text{out}}^{\text{ion}(t)}). \tag{13}
\end{align*}
\]

Equation (12) does not mix the behavior of the two electrons; the single ionization probability is only related to the external electron while the double ionization probability is related only to the internal electron. Equation (13) mixes these two probabilities, as in standard time-dependent Hartree–Fock and density functional theory approaches (see, e.g. Lappas & van Leeuwen, 1998 and references therein) and introduces the indistinguishability of the two electrons. In an equivalent way, one can start from the symmetrization of wave function in Eq. (5) and then use the more general definitions of Eqs. (10) and (11) for the ionization probabilities in a fully correlated system. One can then arrive at formulae in Eq. (13) by simply ignoring the small exchange integral of the two orbitals.

### 5. NONSEQUENTIAL IONIZATION YIELDS

#### 5.1. Results for different wavelengths

We have analyzed the case of a 1D He atom subjected to an external laser pulse using mainly the approximate scheme described in Section 3. The laser is described by a classical electric field whose pulse shape is given by a \( \sin^2 \) function in a reasonably realistic way:

\[
E(t) = E_0 \sin^2 \left( \frac{\pi t}{n_c T_L} \right) \sin(\omega_c t). \tag{14}
\]

\( E_0 \) is the field strength, \( n_c \) is the total number of optical cycles (o.c.) in the pulse, and \( T_L \) is the laser optical period.

We have considered five different wavelengths: 1064 nm, 780 nm, 532 nm, 390 nm, and 248 nm, that correspond to the Nd:Yag and Ti-Sapphire lasers, their second harmonics, and the KrF laser, most commonly used in strong-field physics experiments. For each of these, we have investigated three different cases corresponding to three different pulse durations: 4 o.c., 8 o.c., and 32 o.c. For each case of pulse duration and wavelength, we have varied the field strengths in the range 0.05 a.u.–0.6 a.u. (laser intensity \( 10^{14} – 10^{16} \text{W/cm}^2 \)). We have also looked at the He\(^+\) ionization yield, in order to distinguish between the sequential and the NS contributions to the total double ionization yield of He.
Fig. 1. Single (solid lines) and double (dashed lines) ionization yields of He as functions of intensity, together with the ionization yields of the He$^+$ ion (dotted lines). The wavelength in each case is shown in the inserted box. The pulse duration is 32 optical cycles. The approximate model of Section 3 has been used. All the curves are superimposed in the lower right corner figure for ease of comparison.
In the case that is close to the experimental conditions, for a pulse duration of 32 o.c., the single ionization yields are in good agreement with those that are obtained from the SAE model for He, at least before the complete saturation region. Our results for all wavelengths, obtained from the approximate model introduced in Section 3, are shown in Figure 1. We see that for every wavelength, except for 390 nm and 248 nm, the single ionization yield is characterized by an almost linear behavior for low intensities (on a bilogarithmic scale). We can also observe, by looking at Figure 1, that the relative amplitude of the single ionization signal changes as one varies the wavelength of the laser for a fixed pulse duration (measured in optical cycles, not in real time). For a fixed laser intensity, the single ionization signal increases as the wavelength decreases.

We have also found a rather high NS ionization signal at 248 nm, in agreement with Lappas and van Leeuwen (1998), but in disagreement with Becker and Faisal (1999). We believe that the very long laser pulses (longer than 600 o.c.) that were used in the latter case (Becker & Faisal, 1999) caused saturation of the single ionization at very low intensities, thus making the detection of the NS signal essentially too difficult to discern. In Figure 2 we show our calculations of the ion yields using the exact 1D model that we introduced in Section 2. The pulse here is only 8 o.c. long and has a trapezoidal shape with two cycles for turn on, four cycles of constant field strength, and two cycles for turn off. We see that the NS process can add more than one order of magnitude to the double ionization signal.

When the wavelength is 390 nm or 248 nm, the raising part of the single ion yield curve is not as smooth as in the other cases, but has a rich structure characterized by the presence of small jumps (see Fig. 1). These jumps must presumably be connected with resonance atomic effects which

![Figure 2](image_url)

**Fig. 2.** Single (solid line) and double (dashed line) ionization yield of He as a function of intensity, together with the ionization yield of the He$^+$ ion (dotted line), at wavelength 248 nm, with the “exact” fully correlated two-electron model. The pulse duration is 8 optical cycles.
appear at wavelengths such that multiphoton (or resonant) ionization is more important than tunneling ionization.

A common feature in all of the above cases of Figure 1 is the clear appearance of three distinct regions of intensities for the double ionization yields. The first region extends up to intensities of $10^{15}$ W/cm$^2$ and is characterized by a monotonic increase with intensity. Beyond $10^{15}$ W/cm$^2$, the single ionization yields saturate, and, therefore, the NS double ionization yields also saturate ("knee" structure). A third region is evident for even higher intensities when the double ionization becomes completely sequential.

For the double ionization yields there are large discrepancies between the two different approaches that we consider: the double ionization yields from Eq. (13), and the sequential model, that is, the one that is obtained using the single-electron system of He$^+$ for the double ionization probability. We immediately note that there is a good agreement between the different approaches for laser intensities large enough for the double ionization yields to be greater than 0.01. For smaller values of laser intensity, the agreement is very poor.

Also, substantial differences between our two ionization probability definitions appear in the intermediate and low-intensity region. In particular, if we use the first ionization probability criteria, given by Eq. (12), for every wavelength, we obtain a knee which is wider than that obtained using Eq. (13). For lower intensities, Eq. (12) overestimates the

![Fig. 3](image_url)

Fig. 3. Single (solid lines) and double (dashed lines) ionization yields of He as functions of intensity, together with the ionization yields of the He$^+$ ion (dotted lines), calculated from the approximate model, for various laser pulse durations, at a wavelength of 780 nm. In each case the pulse duration in optical cycles (o.c.) is shown in the inserted box. All the curves are superimposed in the lower right corner figure.
double ionization, because the probability of the outer electron being inside the atom is not taken into account. This explains the already noted discrepancy (Sheehy et al., 1998) between the experimental data and a previous model calculation (Watson et al., 1997) regarding the double ionization yields for low intensities.

5.2. Results for different pulse durations

After having analyzed the behavior of our He atom model when subjected to different laser wavelengths, we now investigate the behavior of this system under laser pulses of various durations.

We have studied three different cases corresponding to pulse durations of 32, 8, and 4 optical cycles. The main results are shown in Figure 3, for a 780 nm laser. Again, it is possible to observe some general features. At first, as we decrease the total pulse duration, passing from 32 to 8 and 4 o.c., we observe that all curves move towards higher laser intensity values. Such behavior can be simply explained by the fact that, as the pulse duration decreases, the same effect is approximately obtainable with a higher intensity.

An interesting result is connected with the shape of the knee. As we decrease the pulse duration, the knee structure tends to become smoother and eventually disappears—as we shall see in the next section as well. This phenomenon is due to the fact that the position of the single ionization saturation intensity moves towards higher intensities and closer to the double ionization curve as the pulse duration is decreased, and thus restricts the knee structure to a narrower intensity region.

Given the sensitivity of the knee structure on the pulse duration, one cannot make easy conclusions regarding the existence (or not) and the shape of the knee at various wavelengths. New experiments at shorter wavelengths and with shorter pulses will be needed to verify our conclusions.

6. DISCUSSION

Two main mechanisms for the double electron ejection have been proposed in the last decade. One of them, the “recoalescence” mechanism, is based on the quasi-classical strong-field approximation that has been introduced in order to describe high-harmonic generation in atomic gases irradiated by long-wavelength infrared laser pulses. According to this proposed mechanism (Corkum, 1993), the external electron of He is ionized rapidly due to tunneling, and subsequently propagates freely in the laser field. When some phase conditions are met, this electron may return to the atomic core within one optical cycle. The subsequent rescattering event is considered to be responsible for the emission of high-energy photons and for the enhanced ionization of the internal electron through energy transfer. The second proposed mechanism assumes that the fast ejection of the external electron leaves the internal one in a superposition of He\(^+\) bound states (shake-off); this excited state contribution then leads to a fast double ionization due to the strong laser field (Fittinghoff et al., 1992; Sheehy et al., 1998).

In order to investigate the influence of these two different mechanisms on the double ionization, we have made model calculations that can explicitly identify the two processes. First, in order to exclude any possibility for rescattering we

![Fig. 4. Single (solid lines) and double (dashed lines) ionization yields of He as functions of intensity, together with the ionization yields of the He\(^+\) ion (dotted lines), for a laser pulse that is half an optical cycle long. The approximate model of Section 3 has been employed. The wavelengths are shown in the boxes.](image)
use a laser pulse with a duration of only one-half of the optical cycle. In this case there is no time for the ejected external electron to interact with the internal one after its propagation in the laser field. Therefore, the only process that could then create a NS signal should be the shake-off effect. In Figure 4 (for 780 nm and 390 nm) we show the single and double ionization yields of He, together with the ion yield of He$^-$, after irradiation with a half-cycle pulse. We see that, while in the 390 nm case the NS contribution is barely observable, in the 780 nm case this contribution is still negligible and insufficient to create any knee structure. We conclude that the shake-off effect is absent here.

Furthermore, in order to estimate the effect of the electron rescattering, in accordance with the recollision mechanism, we can artificially switch off the laser coupling with the internal electron in Eq. (8), thus leaving only the electron–electron interaction term as an external field that can cause double ionization. As we can see from Figure 5, this term alone can reproduce the very high NS signal of double electron ejection with excellent agreement, up to the end of the knee region, where the NS process saturates due to the complete ionization of the external electron. For higher intensities there is no external electron packet remaining to have an effect on the dynamics of the internal one, and the electron–electron interaction becomes insignificant. This result confirms a previous study by Sanpera et al. (1998), where the internal electron was considered to interact only with the dipole radiation field (harmonics) that is generated by the motion of the external electron in the strong laser field.

Finally, we emphasize that the suggested interaction of the core electron with the outer electron has essentially nothing to do with the standard process of single-impact ionization. It is an electron–electron scattering process that occurs periodically, over many laser cycles, in a time interval of the
order of the pulse duration. The energy that is transferred to the internal electron is provided not simply by the kinetic energy of the returning external electron in a single collision event, but by the laser field itself through the multiphoton absorption of unlimited energy by the outer active electron and subsequent electron–electron interaction [for a detailed review and discussion see Lambropoulos et al. (1998)].

ACKNOWLEDGMENTS

We thank John Watson for helpful discussions and for providing us with some of his numerical codes. We acknowledge the Imperial College Parallel Computing Centre for the allocation of its computer facilities and for technical support. This work was supported by the UK Engineering and Physical Sciences Research Council.

REFERENCES

Becker, A. & Faisal, F.H.M. (1999). Phys. Rev. A 59, R1742.
Burnett, K., Watson, J.B., Sanpera, A. & Knight, P.L. (1998). Phil. Trans. R. Soc. Lond. A 356, 317.
Corkum, P. (1993). Phys. Rev. Lett. 71, 1994.
Dundas, D., Taylor, K.T., Parker, J.S. & Smyth, E.S. (1999). J. Phys. B: At. Mol. Opt. Phys. 32, L231.
Faisal, F.H.M., Becker, A. & Muth-Böhm, J. (1999). Laser Phys. 9, 115.
Feit, M.D., Fleck, J.A. & Steiger, A. (1982). J. Comput. Phys. 47, 412.
Fittinghoff, D.N., Bolton, P.R., Chang, C. & Kulander, K.C. (1992). Phys. Rev. Lett. 69, 2642.
Grobe, R. & Eberly, J.H. (1992). Phys. Rev. Lett. 68, 2905.
Kulander, K.C. (1987). Phys. Rev. A 36, 2726.
Lambropoulos, P., Maragakis, P. & Zhang, J. (1998). Phys. Rep. 305, 203.
Lappas, D.G., Sanpera, A., Watson, J.B., Burnett, K., Knight, P.L., Grobe, R. & Eberly, J.H. (1996). J. Phys. B: At. Mol. Opt. Phys. 29, L619.
Lappas, D.G. & van Leeuwen, R. (1998). J. Phys. B: At. Mol. Opt. Phys. 31, L249.
Larocheille, S., Talebpour, A. & Chin, S.L. (1998). J. Phys. B: At. Mol. Opt. Phys. 31, 1201.
L'Huillier, A., Lompré, L.A., Mainfray, G. & Manus, C. (1983). J. Physique 44, 1247.
Petersilka, M. & Gross, E.K.U. (1999). Laser Phys. 9, 105.
Pindzola, M.S., Griffin, D.C. & Bottcher, C. (1991). Phys. Rev. Lett. 66, 2305.
Sanpera, A., Watson, J.B., Shaw, S.E.J., Knight, P.L., Burnett, K. & Lewenstein, M. (1998). J. Phys. B: At. Mol. Opt. Phys. 31, L841.
Sheehy, B., Lafon, R., Widmer, M., DiMauro, L.F., Agostini, P.A. & Kulander, K.C. (1998). Phys. Rev. A 58, 3942.
Talebpour, A., Larocheille, S. & Chin, S.L. (1997). J. Phys. B: At. Mol. Opt. Phys. 30, L245.
Walker, B., Sheehy, B., Di Mauro, L.F., Agostini, P., Shafer, K.J. & Kulander, K.C. (1994). Phys. Rev. Lett. 71, 1227.
Watson, J.B., Sanpera, A., Lappas, D.G., Knight, P.L. & Burnett, K. (1997). Phys. Rev. Lett. 78, 1884.

Time-dependent effects in the nonsequential ionization of Helium