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Multielectron response to intense laser fields

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In this paper we will describe recent theoretical approaches to calculating the response of multielectronic atoms in intense laser fields. This work is based on the soultion of the time-dependent Schrödinger equation for a single atom driven by a linearly polarized laser field. Results obtained in recent experimental measurements of multiple ionization rates suggest that the interaction between the electrons in helium enhances the double ionization rate. Solving the full two-electron Schrödinger equation is a very computer-intensive problem, and therefore we have introduced a new model that can reproduce the experimental results. This model is described in detail in this paper, and used to investigate the origin of the double ionization.

Keywords: electron correlation; multielectron atoms; double ionization; non-sequential double ionization; recollision mechanism; Crapola model

1. Electron correlation effects in physics

Producing an accurate description of electron correlation in diverse physical systems still constitutes one of the major challenges of theoretical physics. The success of independent particle methods, e.g. the self-consistent Hartree–Fock method in describing the qualitative features of atomic, molecular and condensed matter systems is remarkable but there are situations where it fails spectacularly. In atoms, configuration interaction and its manifestation in resonance and autoionization phenomena displays clearly the failure of the independent particle view. In condensed systems many situations e.g. itinerant magnetism, high-temperature superconductors, etc., display marked deviations from the behaviour expected from self-consistent descriptions (even in their extended renormalized quasi-particle versions). In atoms, powerful techniques have been developed to treat electron correlation in all its gory details for bound states. For the case of atoms in ultra-intense laser fields the theory is now being developed. For moderate intensity fields where resonance effects are still important the electron correlation has been shown to give rise to spectacular effects such as laser-induced continuum structure. The treatment of these phenomena first relied on the essential states configuration interaction method (Protopapas *et al.* 1997). More recent studies have used the *R*-matrix Floquet technique to make quantitative predictions of electron correlation effects in complex atoms (Kylstra *et al.* 1995). For ultra-intense fields, where the laser field becomes comparable to the internal Coulomb field alternative techniques, based on the direct numerical integration of the Schrödinger equation are required.

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2. The single active electron approximation

To fully describe the dynamics of a multielectron atom interacting with an intense laser field it is necessary to solve the full many-body Schrödinger equation. Although solutions of the TDSE for a single electron are now commonplace, the numerical solution of the TDSE for more than one electron is a very tough problem. As a result there has been a great deal of discussion as to how much of the atomic response can be described using less demanding models of the dynamics. To date, one of the most common approaches is to use the so-called single-active-electron (SAE) approximation.

In this approach, first pioneered by Kulander *et al.* (1992*b*), it is assumed that the response of a target atom is entirely dominated by the dynamics of a single outermost electron. It is therefore assumed that all of the electrons except for one are 'frozen' in their original orbits, with the remaining electron moving in the mean field due to the frozen core. In order to calculate the dynamics of a given atom in the SAE approximation the first step is to calculate the Hartree–Fock ground state of the atom (in the field free limit), and hence an initial state and effective potential for the outer electron. The atom response is then obtained by solving the one-electron TDSE. The SAE approximation has been extensively used by many authors, and has been found to reproduce many of the experimental results.

More recently the SAE approximation has been used to study the response in the regime where one would expect significant contributions to the harmonic spectrum from ions. In such a calculation it is assumed that the response from each of the ions can be calculated independently using seperate SAE calculations. The contribution to the emitted spectrum from each ion can be weighted by the probability of reaching that ion state.

This approach has been successfully used by Sanpera *et al.* (1994) to study the contribution to the harmonic spectrum by He^+. Simulations of the experimental results obtained by Sarukura *et al.* (1991) indicate that the highest-order harmonics are in fact due to the response of the helium ion at an intensity of $10^{16}\,\mathrm{W\,cm^{-2}}$. Preston *et al.* (1996) have also used a similar approach to analyse the harmonics obtained from a neon target. In the case of neon it is more convenient to use a one-dimensional model of the atomic dynamics. In this case the results show clear evidence of harmonic response from the neutral and also the first and second excited states.

Figure 1 shows the experimental results they obtained using a KrF laser (248 nm) at intensity $10^{17}\,\mathrm{W\,cm^{-2}}$ using a 350 fs pulse. The curves on the plot show the response of neutral neon and also Ne^+ and Ne^{2+}, calculated in the SAE approximation using a one-dimensional model of the atom. In order to make a direct comparison between experiment and theory the single-atom response has been spatially averaged over the focus of the laser (assuming a Gaussian beam profile). From the figure it is clear that by taking into account the response of the various charge states of neon, it is possible to reproduce the experimentally observed spectrum. Furthermore, this calculation clearly demonstrates that the neon ions make a significant contribution to the harmonic response.

3. Beyond SAE: non-sequential double ionization

Despite the success of the SAE approximation in describing the dynamics of atoms in super-intense laser fields, there has still been a great deal of interest in fully cor-

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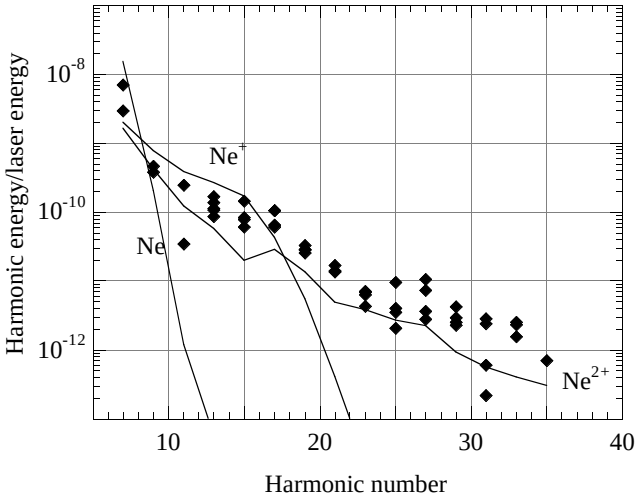

Figure 1. Comparison between the experimentally observed harmonics from neon and a one-dimensional SAE simulation of the response of neon and its ions. (Taken from Preston *et al.* (1996).)

related models of the electron dynamics. One reason for this is in order to find out just how good the SAE approximation is. The fact that the SAE accurately reproduces the experimentally observed harmonic spectrum radiated by the atom does not necessarily imply that it gives a full description of the electron dynamics. Interest in going beyond SAE has increased in recent years due to a series of experiments demonstrating significant non-sequential ionization in the intense field regime. In the first of these experiments, performed by Fittinghoff *et al.* (1992), the single- and double-ionization rates of helium were accurately measured as a function of the laser intensity. It was found that the single-ionization rate could be reproduced by tunnelling rates calculated using the Ammosov, Delone and Krainov (ADK) theory (Ammosov *et al.* 1986). For high intensities, close to saturation of the double ionization, the ADK rates also gave a reasonable description of the He^{++} yield, but at lower intensities there was a dramatic disagreement between the predicted and observed rates. For intensities below 10^{15} W cm^{-2} the He^{2+} yields observed in the experiment were found to be up to two orders of magnitude higher than predicted by the single-electron theories. As the intensity increases there is, initially, a saturation of the double-ionization yield, which converges with the theoretical prediction before once again increasing rapidly with increasing intensity. This increased double ionization was initially referred to as a knee, but has more recently become known as the 'shoulder'. Since this phenomenon was first reported similar results have been obtained in helium by Walker *et al.* (1994) and in both neon and helium by Dietrich *et al.* (1994). The results obtained by Walker *et al.* (1994) are plotted in figure 2. In this figure the closed symbols are the experimental single ionization yields while the open symbols denote double-ionization yields. The figure also shows the theoretical ionization yields obtained by solving the TDSE in the SAE approximation. As was the case with the Fittinghoff experiment, the theoretical model gives an excellent description of the single-ionization rate, but fails to reproduce the shoulder observed in the double ionization yield. The shoulder must therefore be due to some non-sequential double ionizaton process.

In order to explain the origin of the non-sequential double ionization two mecha-

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Figure 2

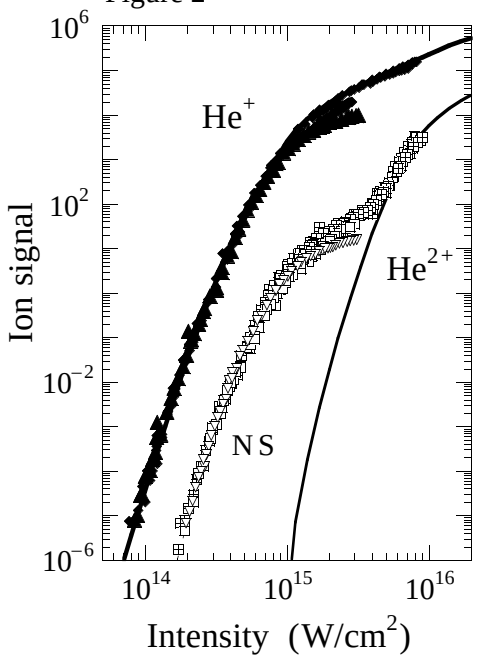

Figure 3

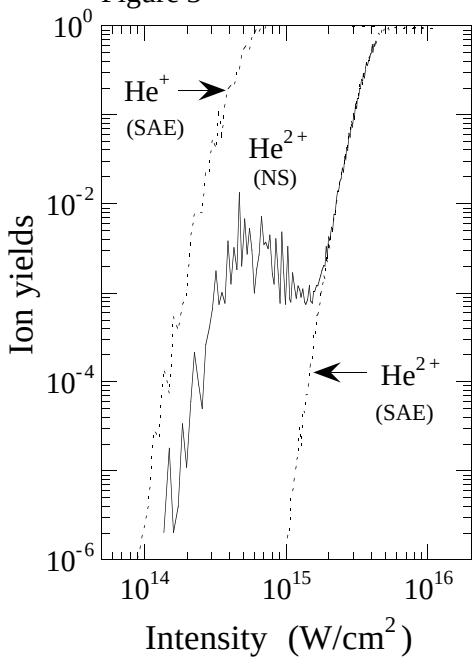

Figure 2. Experimentally measured single- and double-ionization yields in helium for linearly polarized, 100 fs, 780 nm light. The solid curve is the calculated sequential He^{2+} yield. (Taken from Walker *et al.* (1994).)

Figure 3. Numerically calculated single- and double-ionization yields of He calculated using our one-dimensional model. The dashed lines correspond to SAE calculations for He and He^+, the solid line corresponds to the one-dimensional Crapola model.

nisms have been proposed. The first is known as the 'shake-off' mechanism while the second is known as the 'recollision' mechanism. The shake-off mechanism was first discussed in detail by Walker *et al.* (1994). The basic idea is that when the field is applied one of the electrons is rapidly ionized, leaving a He^+ ion. In the sequential model the double ionization rate is small due to the large binding energy of the helium ion. In the shake-off mechanism it is assumed that because the ionization of the outer electron is rapid, the remaining electron does not have time to relax into the ground state of the ion. In other words there is a significant probability of leaving the ion in an excited or even continuum state, from where it can be ionized. By making an estimate of the probability of leaving the ion in an excited state Walker *et al.* (1994) have been able to reproduce the shoulder structure. The alternative mechanism is the recollision mechanism proposed by Corkum (1994). In common with the recollision model that has been so succesful in explaining the harmonic spectrum, the recollision mechanism is based on the idea that when the outer electron is ionized it has a significant probability of returning to the core. When it returns, it can recombine emitting harmonics or it can reduce the binding of the other electron leading to enhanced double ionization. As with the shake-off mechanism it is possible to reproduce the experimental results using the recollision mechanism, but once again a free parameter is required—in this case the impact parameter. A further advantage of the recollision model is that it gives a very simple explanation of the double ionization in elliptically polarized fields. It has been demonstrated (Walker *et al.* 1993; Dietrich

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et al. 1994) that increasing the ellipticity of the laser field reduces the non-sequential double-ionization rate. The explanation of this phenomenon is simply that increasing the ellipticity of the laser is equivalent to introducing a component of the field transverse to the main axis. This component reduces the probability of the electron returning to the core, and therefore reduces the probability of recollision-induced double ionization (and also the probability of harmonic generation).

4. Alternative approaches to calculating the multielectron response

(*a*) *Solving the full TDSE in one dimension and three dimensions*

The experimental observation of non-sequential double ionization of helium, characterized by the appearance of a shoulder in the double-ionization yield, has led to renewed interest in calculating the multielectron response of atoms in intense laser fields. In order to fully calculate the dynamics of a two-electron atom in an intense field one would like to solve the full two-electron time-dependent Schrödinger equation. The use of this approach has been limited by the computational resources required. There has, however, been much recent progress in this direction made by Parker *et al.* (1996). Using the massively parallel Cray T3D supercomputer at the University of Edinburgh they have been able to solve the two-electron Schrödinger equation in three spatial dimensions. Their approach is based on the decomposition of the wavefunction into a set of angular momentum basis states. By assuming that the laser is linearly polarized, they are able to reduce the number of spatial dimensions; however, they are still left with a large number of coupled partial differential equations. The computational size of this problem means that they are limited to intensities below 10^{16} W cm^{-2}, with very-short laser pulses.

Perhaps the most obvious method of reducing the complexity of the calculation is to only consider a single spatial dimension. One-dimensional models of the single-electron response have been used extensively by many groups. It has been well established that the qualitative results obtained using one-dimensional models are very similar to those obtained using more realistic three-dimensional models. The use of one-dimensional two-electron models has been pioneered by Grobe *et al.* (1991) to study the dynamics of helium and H^-. More recently this approach has been used by Lappas *et al.* (1996) to study the double ionization of helium.

Solving the two-electron Schrödinger equation, even in one spatial dimension, is an extremely computer intensive problem, which places restrictions on the laser pulses that can be modelled. Furthermore to make a detailed study of the effect of electron correlation, (to study the origin of the shoulder for example) one would like to make a large number of calculations, in order to scan the parameter space. This is clearly not practical using the fully correlated models, and therefore there has been considerable interest in less demanding models of the electron dynamics.

(*b*) *Time-dependent Hartree–Fock (TDHF)*

One method that has been used to study the dynamics of multielectron atoms is the time-dependent Hartree–Fock method. This approach was first used by Kulander to study the multiphoton ionization of both helium (Kulander 1987) and later xenon (Kulander 1988), and has also been studied by Pindzola *et al.* (1991). The difficulty in using the Hartree–Fock method, as discussed by Kulander (1987) and by Pindzola *et al.* (1991), is that this method imposes the constraint that the atom wavefunction

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is the product of single-electron wavefunctions which, for a helium atom initially in its ground state, are identical. In other words the wavefunction is of the form

$$\Psi(\boldsymbol{r}_1, \boldsymbol{r}_2, t) = \psi(\boldsymbol{r}_1, t)\psi(\boldsymbol{r}_2, t). \tag{4.1}$$

The reason why the TDHF approach fails becomes clear if we consider the effect of exciting (or ionizing) the helium atom. If there is significant coupling to the excited states then we can write the wavefunction in the form

$$\Psi(\boldsymbol{r}_1, \boldsymbol{r}_2, t) = [c_{1s}(t)\phi_{1s}(1) + c_{2s}(t)\phi_{2s}(1) + \cdots][c_{1s}(t)\phi_{1s}(2) + c_{2s}(t)\phi_{2s}(2) + \cdots]. \tag{4.2}$$

In this representation there is always a significant probability of double excitation (when the probability of excitation P_{ex} is small, the probability of double ionization scales as P_{ex}^2). This double ionization is an artifact of the Hartree–Fock representation. Furthermore, since in the TDHF model the electrons only interact through their mean fields, the double ionization leads to a reduction of the screening of the nuclear charge. Therefore as the electrons escape, the binding energy of the bound states increases leading to unphysical population trapping.

One possible method of avoiding these problems is to use a configuration interaction (CI) picture. In this case we write the two-electron wavefunction in the form

$$\Psi(\boldsymbol{r}_1, \boldsymbol{r}_2, t) = c_{1s^2}\phi_{1s}(1)\phi_{1s}(2) + c_{1s2p}[\phi_{1s}(1)\phi_{2p}(2) + \phi_{2p}(1)\phi_{1s}(2)] + c_{2p^2}\phi_{2p}(1)\phi_{2p}(2) + \cdots. \tag{4.3}$$

In general, unless the field is extremely strong, the doubly excited state populations are small compared with the singly ionized states (i.e. $c_{2p^2} \ll |c_{1s2p}|^2$).

In the next section we develop a model that shares certain features of both the CI and TDHF representations, and show that it gives a remarkably good description of the dynamics of helium in an intense laser field. Our model, which is closely related to the unrestricted Hartree–Fock model is based on the fact that the helium ground state can be approximated by a wavefunction of the form

$$\Psi(\boldsymbol{r}_1, \boldsymbol{r}_2, t) = (1/\sqrt{2})[\phi_{in}(1)\phi_{out}(2) + \phi_{out}(1)\phi_{in}(2)], \tag{4.4}$$

where ϕ_{in} and ϕ_{out} are single-electron wavefunctions with ϕ_{in} more strongly localized close to the nucleus. Starting from this wavefunction it is clear that single ionization dominates at relatively low intensities since ϕ_{in} is more strongly bound than ϕ_{out}.

5. The Crapola model of helium

While each of the approaches described above can be used to describe the dynamics of helium under certain circumstances, none of them are particularly suitable for investigation of the origin of the shoulder. Clearly since the shoulder is due to a non-sequential process we cannot use the SAE approximation. On the other hand, we are not in the regime where the motion of the two electrons can be considered completely independent and therefore the Hartree–Fock method is not appropriate. It is, of course, possible to reproduce the dynamics by solving the fully correlated two-electron Schrödinger equation. However, as we have already discussed, the computational intensity of this method limits it to very short pulses for a limited set of parameters. Therefore, in order to study the non-sequential ionization, we have developed a new model of the dynamics of helium which we will refer to as the Crapola model.

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Perhaps the best way to think of the Crapola model is as a first correction to the single-active-electron approximation. Previous investigations of the SAE have shown that for most situations it gives a very accurate description of the dynamics of a helium atom in an intense laser field. Indeed, even in the experiments by Walker *et al.* (1994) SAE calculations give an excellent description of the single ionization. In our model we will therefore follow the SAE in assuming that we can identify an inner and an outer electron. Perhaps the best justification for this assumption is simply that it is well known that in this intensity regime one of the helium electrons ionizes rapidly, while there is only a small probability of both electrons ionizing. This initial choice of wavefunction can also be obtained by making a variational calculation of the ground state of the atom. Furthermore we will assume that the dynamics of the outer electron are accurately described by the SAE. The justification for this last assumption is simply that it has been established experimentally that one of the electrons ionizes much more rapidly than the other. Since the motion of the outer electron occurs on a much shorter timescale than that of the inner it is reasonable to assume that the potential experienced by the outer electron due to the inner does not change significantly. Clearly this is not the case for the inner electron, and therefore when we calculate the dynamics of the inner electron we include a time-dependent potential due to the outer electron. This approach is clearly related to the Hartree–Fock approach, except that we are assuming that the initial state is of the form

$$\Psi(\boldsymbol{r}_1, \boldsymbol{r}_2, t) = (1/\sqrt{2})[\phi_{\rm in}(1)\phi_{\rm out}(2) + \phi_{\rm out}(1)\phi_{\rm in}(2)]. \tag{5.1}$$

We then follow Gross *et al.* (1996) in assuming that for helium it is possible to neglect the exchange interaction. In this limit we can consider just a simple product wavefunction rather that the superposition given above. We also assume that the time-dependent potential on the outer electron due to the inner can be replaced by a time-independent potential. The justification for this second approximation is simply that in the intensity regime where the shoulder is observed the probability of ionizing the inner electron is small, and therefore the potential due to the inner electron doesn't change significantly during the pulse. The above approximations lead to a simple model that can be applied to realistic pulse lengths for a large number of different parameters using a conventional workstation. Furthermore, as we will discuss below, our model gives remarkable agreement with the experimental results of Walker *et al.* (1994) and can be used to investigate the physical mechanism responsible for the non-sequential double ionization.

We therefore have two coupled equations of the form

$$\mathrm{i}\frac{\partial \psi_n}{\partial t}(\boldsymbol{r}_n, t) = [-\tfrac{1}{2}\nabla_n^2 + V_n(\boldsymbol{r}_n, t) + V_{\rm int}(\boldsymbol{r}_n, t)]\psi_n(\boldsymbol{r}_n, t), \tag{5.2}$$

where $n = 1, 2$ for the outer and inner electrons, respectively, $V_{\rm int}(\boldsymbol{r}, t)$ is the potential due to the external field, and $V_n(\boldsymbol{r}, t)$ is the potential due to the nucleus plus the interaction with the other electron. As we have already discussed, we calculate the dynamics of the outer electron using the SAE approximation and therefore $V_1(\boldsymbol{r}, t)$ is a time-independent potential given by the Hartree–Fock effective potential for an electron in helium. For the case of the inner electron we use a time-dependent potential made up of the static potential due to the atomic core plus the time-dependent potential due to the effect of the outer electron acting on the inner.

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6. The one-dimensional Crapola model

In one-dimensional calculations the use of Coulomb potentials is not appropriate due to the singularity at $x = 0$. Several model potentials have been suggested for the one-dimensional calculations, as with many of the authors studying multiphoton physics we use a soft-core (Rochester) potential, first introduced by Eberly *et al.* (1992), of the form

$$V(x) = \frac{-Z}{\sqrt{a^2 + x^2}}, \tag{6.1}$$

where a is a parameter that is chosen so the lowest eigenstate of the potential matches the binding energy of the atom under consideration. For the 'outer' electron the potential V_1 is simply a soft-core potential scaled to have the same binding energy as the neutral helium atom. For the inner electron the potential is given by

$$V_2(x_2, t) = \frac{-2}{\sqrt{a_2^2 + x_2^2}} + \int \mathrm{d}x_1 \frac{\psi_1^*(x_1, t)\psi_1(x_1, t)}{\sqrt{a_{12} + (x_1 - x_2)^2}}. \tag{6.2}$$

In this equation the first term represents the attractive potential due to the nucleus, the value of a_2 is chosen to match the binding energy of He^+. The second term represents the potential experienced by the inner electron due to the motion of the outer electron. In this case the choice of the parameter a_{12} is somewhat arbitrary. We have chosen $a_{12} = 2$, the same value as is used in single-electron calculations to model hydrogen. We have no clear justification for the use of this value, however, in this one-dimensional calculation we will only consider qualitative results. We are confident that changing the value of this parameter, or any of the parameters in our model, will not change the qualitative features of our results.

The simplicity of our model has allowed us to simulate the 100 fs pulses used in the experiment by Walker *et al.* (1994) for 200 different intensities between 10^{14} W cm^{-2} and 10^{16} W cm^{-2}. Figure 3 shows the single- and double-ionization probabilities plotted as a function of the laser intensity. In the case of the inner electron the yield calculated using the SAE approximation is also plotted. The most striking feature of this plot is that there is a clear shoulder in the double ionization yield calculated using the Crapola model. For intensities above 10^{15} W cm^{-2} our model agrees with the SAE calculation. At lower intensities, however, our model predicts a significantly higher yield then the SAE. We should point out that the results plotted in figure 3 are from a single-atom calculation, and therefore cannot be directly compared with the experimental results of Walker *et al.* (1994), however, the qualitative features are remarkably similar in each case. In order to obtain a better qualitative agreement with the experiment, we have extended our model to three spatial dimensions. As we will show, using a simple technique to average over the laser intensity profile, the three-dimensional model gives quite remarkable qualitative agreement with the experimental data.

7. The three-dimensional Crapola model

As in the one-dimensional model, in three-dimensions we assume that the dynamics of the outer electron are well described by the SAE approximation. In three dimensions we assume the potential experienced by the outer electron due to the inner electron and nucleus can be modelled by a Hartree–Fock–Slater time-independent

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potential. The potential for the inner electron is then given by the Coulomb potential due to the nucleus plus a time-dependent potential due to the mean field of the outer electron

$$V_2(\boldsymbol{r}_2, t) = \frac{-2}{r_2} + \int \frac{\psi_1(\boldsymbol{r}_1, t)\psi_1^\star(\boldsymbol{r}_1, t)}{|\boldsymbol{r}_2 - \boldsymbol{r}_1|}\,\mathrm{d}\boldsymbol{r}_1. \tag{7.1}$$

In order to calculate the effective potential it is convenient to make a multipole expansion of the electron–electron interaction

$$\frac{1}{|\boldsymbol{r}_2 - \boldsymbol{r}_1|} = \sum_{\ell=0}^{\ell=\infty} \frac{r_<^\ell}{r_>^{\ell+1}} P_\ell(\cos(\theta_{12})), \tag{7.2}$$

where P_ℓ are the Legendre polynomials. The monopole ($\ell = 0$) term in this expansion only contributes to the spherically symmetric central potential experienced by the atom. It can therefore be thought of as producing a screening of the nuclear charge. We have calculated the double-ionization yield using the monopole term only, and have found that the ionization yield does not change greatly in comparison with the SAE calculation. The second term in the expansion is the dipole ($\ell = 1$) term. It is possible to think of this term as a modification of the laser field driving the atom. Our calculations have shown that it is this term that is responsible for the increased double-ionization yield, and therefore the shoulder.

As with the one-dimensional calculation, the results shown in figure 4 are the probability of single and double ionization for a single atom, and therefore are not directly comparable with the experimental results. However, the qualitative similarities between figures 2 and 4 are striking. In both cases there is a clear shoulder, and the intensity at which the shoulder merges with the SAE calculation is similar in both plots. In the next section we will use a simple spatial average over the beam profile in order to make a quantitative comparison between our theory and the experimental results.

8. Comparison of the Crapola model with experiment

As we have stated previously, it is not possible to make a direct quantitative comparison between figures 2 and 4. The experimental response shows the number of ions produced by a laser pulse focused into an ensemble of atoms, whereas the theoretical results presented in the previous section are the probability of ionizing a single atom at the focus of the laser profile. A full calculation of the ionization yield would require a calculation of the propagation of the laser pulse through the target, something that is not practical for this case. There is, however, a less-demanding approach that can be used to get an estimate of the full ion yield. We simply assume that the laser pulse propagates as a perfect Gaussian, so that atoms at different points in the target experience identical pulses, except that they are scaled to have a different peak intensity. The response is then calculated by taking a weighted average over the intensities experienced at different points in the medium. This approach has been succesfully used in previous work by Preston *et al.* (1996) to model harmonic spectra.

The single- and double-ionization yields calculated by this method are plotted in figure 5, along with the experimental data provided by Walker *et al.* (1994). In making this calculation we have two undetermined parameters. The first of these is the laser profile, which we have assumed to be a perfect Gaussian beam using

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Figure 4

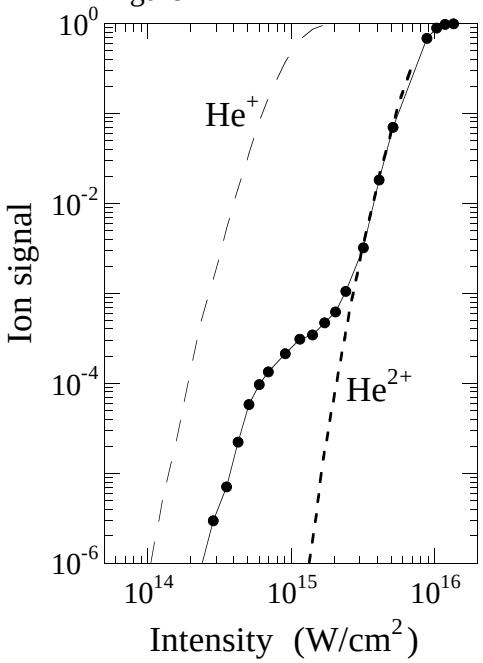

Figure 5

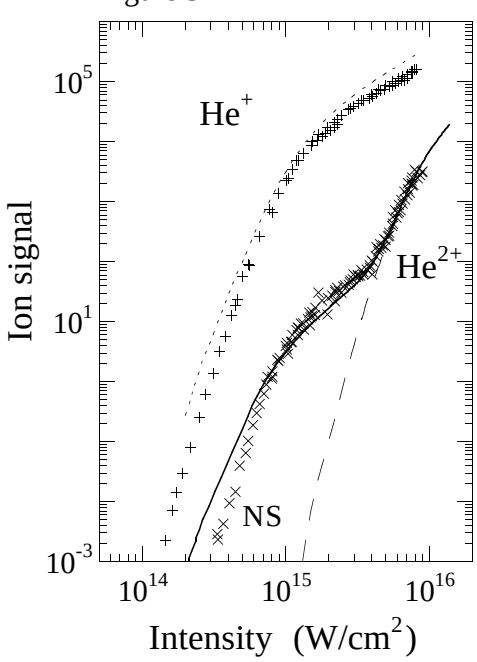

Figure 4. Numerically calculated single- and double-ionization yields of He calculated using our three-dimensional model. The dashed lines correspond to SAE calculations for He and He^{+}, the solid line corresponds to the three-dimensional Crapola model.

Figure 5. Comparison of the He^{+} and He^{2+} yields predicted by our three-dimensional model with the experimental He^{+} (+) and He^{2+} (×) yields measured by Walker *et al.* (1994). The theoretical curves are obtained by spatially averaging the single atom data over a Gaussian laser profile.

the same beam waist as assumed by the experimental group. The second parameter, the only fitted parameter in our model, is the density of atoms in the target. Since we have chosen this parameter in order to obtain the best possible fit to the single ionization rate, we can justifiably claim that our calculation of the double-ionization yield contains no free parameters.

9. Mechanism of the double ionization

In addition to allowing us to accurately reproduce the experimental results, our model also allows us to investigate the mechanism responsible for the non-sequential double ionization. Before considering the mechanism in detail, we make the point that the fact that our model works gives us information about the origin of the shoulder. By comparing our model with the SAE approximation it is clear that the shoulder occurs as a result of the potential experienced by the inner electron as a result of the outer electron. We can, however, go a step further in examining the mechanism. If we consider the two possible explanations discussed previously then there is a clear difference between the two. In particular, the recollision mechanism depends on the fact that the outer electron oscillates in the continuum before returning to the nucleus. Therefore one would expect the double ionization yield to decrease significantly if we inhibit the return of continuum electrons. We can do this easily

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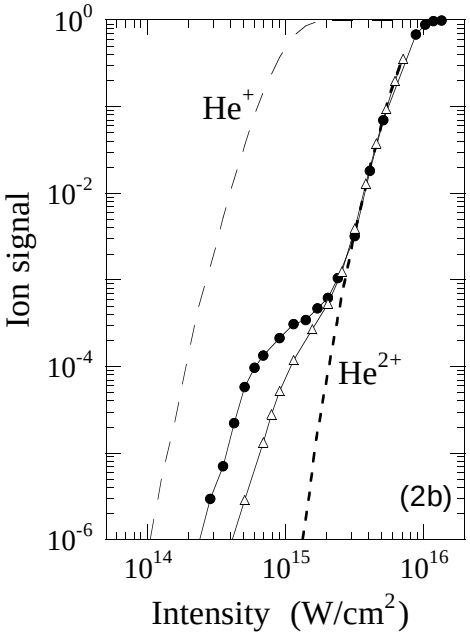

Figure 6. Single- and double-ionization yields in helium. The double ionization yield has been calculated using our three-dimensional model with an absorbing boundary to inhibit the recollision of the outer electron (open triangles). The yield obtained with no absorber is also shown (filled circles).

by introducing an absorbing boundary for the outer electron. The position of this boundary is critical—too close to the nucleus and there is significant absorption from the bound states of the atom, while if the boundary is too far from the core the electron can oscillate and return without ever reaching the boundary. The single-atom response is plotted in figure 6.

In the figure the filled circles show the double-ionization yield calculated by our model without the absorber, while the open triangles show the yield calculated with an absorbing boundary. It is clear from the figure that when the return of the outermost electron is inhibited, the double ionization yield is decreased by an order of magnitude. This clearly indicates that the return of the outer electron plays a critical role in generating the shoulder. It is interesting to note that our absorbing boundary does not completely eliminate the shoulder. Whether this is due to recollision of part of the outer-electron wavepacket that does not reach our absorber or whether it is due to some other mechanism is not yet clear.

10. Conclusions

In this paper we have introduced a model which we have recently developed (Watson *et al.* 1997) in order to describe the dynamics of a helium atom in a super-intense laser field. Our model, which can be thought of as an extension to the single-active-electron approximation, has been used to investigate the non-sequential double ionization observed by Fittinghoff *et al.* (1992) and also by Walker *et al.* (1994). Despite the simplicity of our approach, we are able to reproduce the shoulder using both one-dimensional and three-dimensional versions of our model. Furthermore, by taking a simple spatial average over a Gaussian laser profile, we have shown that our model

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is in excellent quantitative agreement with the experimental results published by Walker *et al.* (1994).

Our model also allows us to investigate the mechanism responsible for double ionization, and in particular, to look for evidence of either the shake-off or recollision mechanisms. The simple fact that our model matches the experiment so well, clearly indicates that it is the influence of the outermost ionized electron that is responsible for the enhanced ionization of the remaining electron. We can, however, go further than this. We have also shown that inhibiting the return of the outermost electron to the atomic core significantly reduces the probability of non-sequential double ionization. while this does not fully confirm the recollision model, or rule out the shake-off mechanism, it does demonstrate that the return of the outermost electron plays an important role in the double ionization process.

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