Nonsequential Double Ionization of Helium

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Electron correlation effects in strong laser fields are investigated by using a simplified two electron model to calculate the double ionization rate in helium. In our model we make a correction to the single active electron approximation by including the effect of the outer electron on the inner one through a time-dependent potential. Using this approach we are able to investigate the nonsequential double ionization observed in recent experiments. [S0031-9007(97)02687-2]

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Electron correlation is at the heart of the description of many important physical phenomena. The elucidation of its role in atomic systems has led to the understanding of many important phenomena such as autoionization, dielectronic recombination, and Wannier threshold effects. In the case of atoms irradiated by moderate strength fields the role of electron correlation has been discussed for laser dressing of autoionizing states as well as for laser induced continuum structure. In superintense fields, however, the role of electron correlation is more difficult to assess theoretically.

To date the most common numerical approach for studying the time-dependent response of multielectronic atoms to superintense laser fields has been the single active electron (SAE) approximation. In this approximation the correlation between the electrons is only included at the first stage of the calculation, i.e., in the initial wave function. It is then assumed that all the electrons but one are frozen in their orbitals, and therefore the atomic response to the external field is entirely due to this single, outermost, electron. Any multiple ionization is therefore assumed to occur by a stepwise process [1]. For most cases of atomic interaction with superintense laser fields the experimental results are accurately reproduced by SAE calculations, indicating that the dynamics are apparently dominated by independent electron evolution.

One case where one can see a clear effect of the dynamics of one electron on the other is in the experiments reported by Fittinghoff *et al.* [2] and more recently by several other groups [3,4]. In these experiments the single and double ionization yields of He in a linearly polarized field are measured to very high accuracy. The single ionization yield is accurately predicted by calculating the response of neutral helium using the SAE approximation [4]. The results are also found to be in reasonable agreement with the Ammosov-Delone-Krainov tunneling rate for ionization [5]. The case of double ionization is, however, more intriguing. For very high intensities ($I \gg 10^{15}$ W/cm^2), where there is strong double ionization, there is good agreement with the numerical SAE calculations for He^+, clearly demonstrating the essentially sequential character of double ionization at such intensities. At lower intensities, however, the double ionization yield is much higher than that predicted by the sequential SAE models. Initially the yield increases rapidly as a function of the laser intensity, with an appearance intensity significantly lower than that predicted by the SAE. This extra component then rolls over before merging with the SAE prediction at higher intensity. This behavior is commonly referred to as a "shoulder" or "knee."

In order to explain the experimental observations, two distinct mechanisms have been proposed. On one hand, Fittinghoff *et al.* [2] proposed a nonsequential ionization process due to the "shake-off" mechanism. In this case it is the rapid change in the potential experienced by the inner electron, due to the escape of the outer electron that is responsible for the increased ionization rate. Corkum [6] proposed an alternative mechanism based on electron-electron inelastic rescattering. This is directly related to the recollision model that has been so successful in explaining the structure of the harmonic spectrum. In this picture it is assumed that the outer electron is promoted into the continuum, by some combination of tunneling and multiphoton absorption. This electron is then driven by the oscillating laser field, and has some probability of returning to the core, where it can excite or ionize the inner electron. Consistent with this proposed rescattering mechanism, experiments have shown that in elliptically polarized light the nonsequential double ionization rate is greatly reduced [7].

The object of this Letter is to investigate the features of this nonsequential double ionization. For relatively modest intensities it can be assumed that the ionization occurs sequentially and the atomic response can be obtained through the single active electron approach. This approach, developed first by Kulander *et al.*, has recently been used to demonstrate the contribution of ions to the harmonic spectrum [8]. If, on the other hand, the laser intensity is sufficiently high, the electron-electron interaction can be neglected in comparison with the effect of the field, and thus the electrons ionize independently.

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In this regime the dynamics are adequately described by Hartree-Fock type models as discussed by Kulander [9], Pinzola *et al.* [10], and Geltman [11].

For the range of intensities used in the experiments by Fittinghoff and Walker, neither of the above models give an adequate description of the behavior of the electrons. Since the ionization is still, to a good approximation, sequential, the Hartree type models, where the electrons ionize independently, are not appropriate. On the other hand, the shoulder is clear evidence of nonsequential behavior, indicating that we need to go beyond the single active electron approximation. Despite the recent progress in the development of computational models of the fully correlated two-electron wave function for helium in both one [12,13] and three [14] spatial dimensions, they remain too computationally demanding for making a large number of calculations using realistic laser pulse lengths. In this paper we propose a novel approach that employs a quasi-independent electron configuration to study the onset of electron-electron interactions. This approach, which can be thought of as a correction to the SAE approximation, allows us to reproduce the shoulder structure observed experimentally.

In the SAE approximation it is assumed that there is an "outer" electron that moves in a constant effective potential due to the nucleus and the inner electron; the external field is then the only time-dependent influence on the escaping electron. When this electron ionizes we are left with a He^+ ion that does not experience any effect due to the first electron. An obvious first correction to this model is to include the effect of the outer electron on the inner one. In doing this we neglect the nonlocal exchange term between the electrons, but include the Coulomb correlation term. We assume that the overlap between outer and inner electrons is sufficiently small for the exchange term to be negligible in comparison to the Coulomb repulsion term. The total wave function can, therefore, be written as a product form

$$\Psi(\vec{r}_1, \vec{r}_2, t) = \psi_1(\vec{r}_1, t)\psi_2(\vec{r}_2, t) + \psi_2(\vec{r}_1, t)\psi_1(\vec{r}_2, t). \tag{1}$$

Since we are neglecting the exchange interaction, we need only consider the first of these terms.

The time evolution of the outer and inner electrons is calculated by solving two equations of the form

$$i\frac{\partial \psi_n}{\partial t}(\vec{r}_n, t) = \left[-\frac{\nabla_n^2}{2} + V_n(\vec{r}_n, t) + V_{\text{int}}(\vec{r}_n, t)\right] \times \psi_n(\vec{r}_n, t), \tag{2}$$

where $n = 1, 2$ refers to the outer and inner electrons, respectively, and $V_{\text{int}}(\vec{r}_n, t)$ is the potential due to the external field. For the outer electron we use the SAE approximation where $V_1(\vec{r}_1, t) = V_1(r_1)$ is the time-independent Hartree-Fock effective potential. For the inner electron, $V_2(\vec{r}_2, t)$ is the time-dependent potential due the nucleus plus the outer electron. The wave function of the inner electron $\psi_2(\vec{r}_2, t)$ depends on the time-dependent wave function of the outer electron $\psi_1(\vec{r}_1, t)$ through this potential.

We have used this model in both one and three dimensions to study the double ionization of helium for the same wavelength ($\lambda = 780$ nm) used in the experiments of Walker *et al.* [4] and intensities ranging from $I = 10^{14}$–10^{16} W/cm^2. In our one-dimensional calculation we use the soft-core Rochester potential [15] $V(x) = -1/\sqrt{a^2 + x^2}$ for the outer electron, where the value of a has been chosen so that the lowest eigenstate of the potential matches the ionization energy of helium. In the case of the inner electron we solve a similar TDSE, but with a time-dependent potential given by

$$V_2(x_2, t) = \frac{-2}{\sqrt{a_2^2 + x_2^2}} + \int dx_1 \frac{\psi_1^*(x_1, t)\psi_1(x_1, t)}{\sqrt{a_{12} + (x_1 - x_2)^2}}. \tag{3}$$

The first term represents the atomic core, and has a value of a_2 chosen so that without the second term, the lowest eigenstate matches the binding energy of He^+. The second term represents the effect of the outer electron on the inner electron, where we have assumed a soft-core interaction in order to avoid the singularity at $x_1 = x_2$. The choice of the soft-core parameter a_{12} is somewhat arbitrary; we have chosen $a_{12} = 2$, the parameter that gives a binding energy equal to that of hydrogen for a single electron system.

Figure 1(a) shows the single and double ionization yields calculated in 1D for a 48 cycle (i.e., 120 fs) trapezoidal pulse as a function of laser intensity at 120 different intensities in the range 8×10^{13}–10^{16} W/cm^2. The single ionization yield is calculated using the SAE, while the double ionization yield is calculated using both the SAE and our model (solid curve). The similarity between this simple calculation, and the experimental data is striking. In both cases, for sufficiently high intensities, the double ionization is in agreement with the SAE calculation for He^+, while for lower intensities there is a clear shoulder structure. The ionization yields shown in Fig. 1(a) are obtained by calculating the probability of finding the He^+ ion in its initial (ground) state. To confirm these results we have also calculated the probability of finding the electron within ± 12.5 and ± 25 a.u. of the origin, at the end of the pulse. In both cases the ionization as a function of laser intensity shows exactly the same behavior. This is significant since it confirms that our model predicts double ionization rather than population of the excited states.

The simple one-dimensional model described above is able to reproduce, qualitatively at least, the shoulder observed in the experimental data. To ensure that the numerical results are not an artifact of the reduced dimensionality we have extended our approach to a more realistic three-dimensional model of the He atom. To

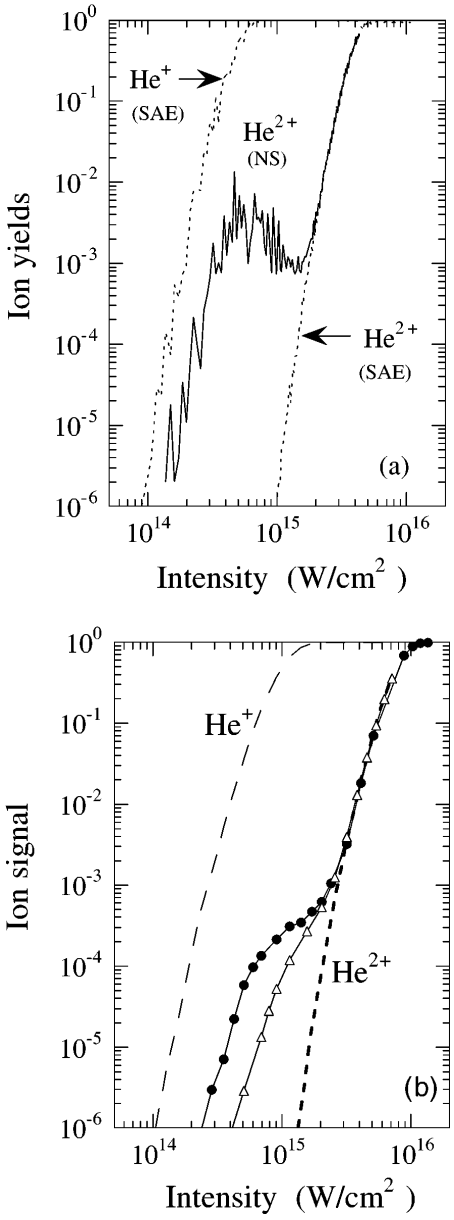

FIG. 1. Numerically calculated single and double ionization yields of He in our 1D (a) and 3D (b) models. The dashed lines correspond to the SAE calculation for He and He^+; the solid line corresponds to the double ionization calculated with our model (nonsequential). In (b) the full circles correspond to our model and the triangles to the case where an absorbing boundary has been used to inhibit the recollision of the outer electron.

reduce the computing time in the 3D case we take 32 cycle (80 fs) sine-squared pulses, and compute the ionization yields for 20 different intensities. As in the 1D case, for the outer electron we use a static Hartree-Fock-Slater effective potential, while for the inner electron we include a time-dependent effective potential due to the outer one,

$$V_2(\vec{r}_2, t) = \frac{-2}{r_2} + \int \frac{\psi_1(\vec{r}_1, t)\psi_1^{\star}(\vec{r}_1, t)}{|\vec{r}_2 - \vec{r}_1|}\, d\vec{r}_1 . \quad (4)$$

The time-dependent effect of the outer electron on the inner one can be thought of in terms of a multipole expansion;

$$\frac{1}{|\vec{r}_2 - \vec{r}_1|} = \sum_{\ell=0}^{\ell=\infty} \frac{r_<^{\ell}}{r_>^{\ell+1}} P_{\ell}(\cos(\theta_{12})). \quad (5)$$

The monopole term ($\ell = 0$) simply changes the central potential that the inner electron experiences by adding a constant term if $r_1 > r_2$, while the dipole term can be thought of as being a modification of the external field. (We consider as a first approximation that higher multipole orders can be neglected.) The monopole term alone does not give the shoulder structure of the experimental results; however, when the multipole expansion is truncated after the dipole term $\ell = 1$, the numerical calculation once again reproduces the shoulder observed in the experimental results, as can be seen in Fig. 1(b).

In order to make a quantitative comparison between our results and the experimental data, we have calculated a spatial average of the single atom response assuming a Gaussian beam profile (TEM_{00}) with a constant waist. Our results, along with the experimental data, are shown in Fig. 2. From the figure it is clear that our model, which contains no free parameters, is in remarkable quantitative agreement with the experimental results, confirming that the increased double ionization occurs due to the influence of the outer electron on the inner.

For peak intensities above $I > 2 \times 10^{15}$ W/cm^2 the outer electron ionizes so rapidly that the interaction between the electrons is only important during the rising edge of the pulse. Furthermore, the laser intensity is sufficient to generate significant ionization in He^+, making

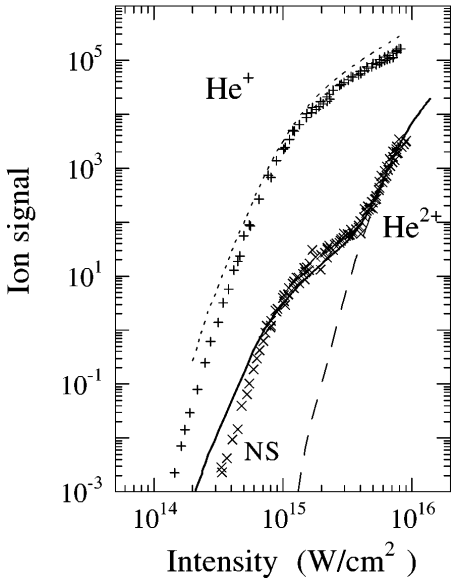

FIG. 2. Comparison of the He^+ and He^{2+} yields predicted by our 3D model with the experimental He^+ (+) and He^{2+} (×) yields measured by Walker *et al.* [4]. The theoretical curves are obtained by spatially averaging the single atom data over a Gaussian laser profile.

the electron-electron interaction negligible in comparison. As a result, at high intensities the double ionization is dominated by a sequential process. This is confirmed by the agreement between the SAE for He^+, our model, and the experimental results in this intensity regime.

To elucidate the mechanism responsible for the nonsequential double ionization, we have recalculated the double ionization while restricting the possible recolliding trajectories of the outer electron by using an absorbing boundary for the outer electron. The position of this boundary is critical: if it is too far from the core then there will still be a significant probability of high energy electron recollision, while if it is too close to the core there will be spurious results due to absorption of the outer electron from bound states. We have performed this calculation in one and three dimensions using absorbing boundaries at distances 10 and 20 a.u. from the core. For the range of intensities used in the simulation the free excursion parameter α_0 ranges between $27-150$ a.u., so our absorbing boundary is sufficient to greatly reduce the effects of recollision. The results show that the shoulder structure of the double ionization yield is still clearly present when recollision is inhibited; however, the signal is reduced by almost an order of magnitude in both the one and three dimensional calculations. This can be seen in Fig. 1(b) where we have included the results (triangles) for the case where the probability of recollision is reduced.

This result clearly indicates that the nonsequential ionization occurs as a result of the return of the outer electron; when it is inhibited the nonsequential ionization is reduced by an order of magnitude. One should, however, be careful in thinking of the nonsequential double ionization in terms of an electron impact process. Such a mechanism would only be effective if the kinetic energy of the returning electron was greater than the binding energy of the inner electron. The change in the potential experienced by the inner electron as the outer electron returns can also be effective in producing double ionization.

In summary, we have presented a novel model, containing no fitted parameters, that is able to describe the dynamics of helium interacting with a laser field for a wide range of intensities. By considering the effect of the escaping electron on the inner electron, this model quantitatively reproduces the experimentally measured double ionization yield, without the complexity of a fully correlated two-electron approach. Furthermore our model allows us to make a preliminary investigation of the mechanism responsible for the shoulder in the double ionization yield. When the return of the outer electron is inhibited by an absorbing boundary the nonsequential double ionization rate is reduced by an order of magnitude, indicating that the nonsequential ionization occurs as a result of the outer electron returning to the nucleus.

In future this quasi-independent electron approach will be used to study other aspects of the dynamics such as harmonic generation and above threshold ionization. The results will be presented elsewhere.

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