Laser-assisted inelastic rescattering during above-threshold ionization

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(Received 28 July 1994)

As an electron is freed from an atom by an intense laser it is accelerated by the oscillating field. Before escaping entirely, it may recollide with its parent ion. During this rescattering, the remaining electrons of the ion might be excited or ionized. The influence of the laser field on these impact excitation and ionization processes is investigated. Model calculations for helium at 780 nm and intensities near 10^{15} W/cm^2 show that the effect of the field on the total probability of promoting an electron from the ionic ground state is small. However, the laser alters the final excited-state populations, rapidly ionizing any excited bound states. We discuss the importance of these results in the search for the mechanism responsible for direct double ionization in the tunneling regime.

PACS number(s): 32.80.Rm

I. INTRODUCTION

Atoms in an intense ($>10^{14}$ W/cm^2) laser field emit very-high-energy electrons and strong, high-order harmonic photons. In both cases the mechanism responsible for the emission involves an electron being promoted into the continuum, being accelerated by the oscillating electric field of the laser, and then rescattering from its original ion core [1]. For infrared or optical frequencies and high intensities, the transition to the continuum leaves the electron with an escape velocity that is small compared to its velocity of oscillation in the field. Because of this, roughly half of the time the electron recollides at least once by its parent ion before escaping entirely, either changing its orbit, which alters its drift velocity, causing high-order above-threshold ionization, and/or emitting a high-energy photon at some multiple of the driving frequency, called harmonic generation.

An additional possibility is that the returning electron may, if it has sufficient energy, excite or ionize one of the remaining electrons bound to the ion. Although electrons are generally observed to be removed sequentially from atoms in intense fields, recent experiments have shown direct, double ionization of the light inert gases [3–6]. In these experiments a second electron was observed to escape, apparently simultaneously, with the first electron as often as one out of 500 times. The underlying mechanism for this double ejection has not yet been established. It has been proposed that inelastic rescattering could be responsible [2]. However, the efficiency with which rescattering can result in the simultaneous ejection of two electrons depends on the transverse spread of the returning electrons as they revisit the vicinity of the ion core. Simple physical arguments show that this width is expected [6] to have grown too large by the time rescattering from the core occurs; few returning electrons have small enough impact parameters to cause ionization. This estimate is based on cross sections measured under field-free conditions [7]. Unless the impact ionization (e-2e) cross sections in the presence of a laser field are much larger than those measured under field-free conditions, this rescattering mechanism cannot explain the observed magnitude of the effect.

In this paper, we report model calculations which show that even though the laser electric fields encountered in these experiments can be quite large at the instant of rescattering, its effect on the total excitation cross section appears to be limited to altering the final-state probabilities. The enhancement of the total inelastic cross section is found to be on the order of 10–20 %.

Before describing our calculations we consider, in a bit more detail, the dynamics of ionization in the intensity regime appropriate to the observed double ejection. At high intensities electrons are promoted into the continuum by tunneling through or, if the field is strong enough, passing over the instantaneous barrier created by the Coulombic attraction of the core and the electric field of the laser. (See Fig. 1.) As the laser field oscillates an electron wave packet is created within the continuum each time the field reaches its maximum amplitude. This occurs twice each optical cycle. This tunneling wave packet emerges at the outer edge of the suppressed barrier. Subsequently the wave packet is driven back and forth along the direction of polarization in phase with the oscillating field. We illustrate this process in Fig. 2, where a representative rescattering trajectory of an electron ionized by tunneling is shown. Immediately after emerging from the barrier at time t_0 the field accelerates the electron away from the ion core. During the next half cycle, after the field has changed sign (at t_1), the electron is gradually stopped (t_2) and then is driven back toward the ion core. Approximately half of the emitted electrons recross the plane of the nucleus, with the possibility of exciting the remaining core electrons. Because the tunneling wave packet is well removed from the effects of the core potential during most of its evolution,

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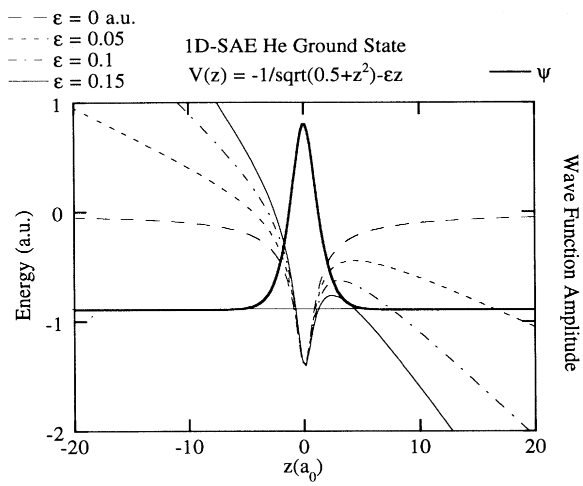

FIG. 1. Effective potential for 1D helium in the instantaneous electric field of the laser for a range of field strengths. (1 a.u. $= 5.1\times10^{9}$ V/cm). Also shown is the field-free ground-state SAE wave function. Its binding energy is 0.89 a.u.

it will spread freely in the directions transverse to the polarization axis. This spreading is illustrated in Fig. 2 by the showing that upon return, the electron can have a finite impact parameter. Knowing the electron-impact excitation and ionization cross sections of the parent ion for scattering in the presence of the laser field and the transverse distribution and kinetic energies of the returning electrons, we can determine the efficiency of the rescattering mechanism. We note that in these strong laser fields the returning electron only has to *excite* the core

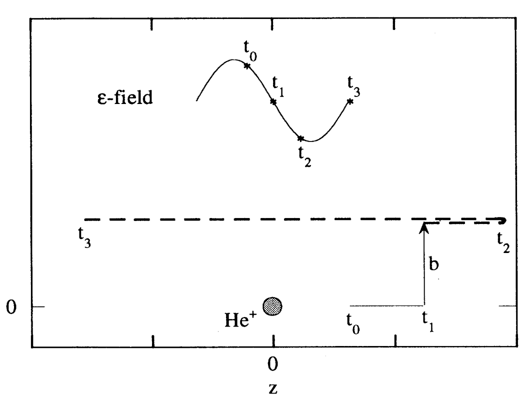

FIG. 2. Schematic of a rescattering trajectory. In the upper diagram, the amplitude of the electric field is shown for one optical cycle. At t_0 the electron tunnels free. It then propagates away from the ion until the end of the first half cycle t_1. In our calculation the electron is then given an impact parameter b and the electron-electron and laser-field–bound-electron interactions are turned on during its subsequent evolution (denoted by the heavy dashed line) in the field. During the second half cycle the field turns the electron around (t_2) and it rescatters from the helium ion core before escaping by the end of the cycle at t_3.

electron. Any excited electron will be ionized very rapidly in the field, thus contributing to the observed double-ionization signal (but technically through a sequential process). Therefore, to test this mechanism, we want to determine the total probability that an electron is removed from the ground state of the ion by the rescattering electron in the presence of the field.

II. CALCULATIONS

In our study the system parameters have been chosen to represent the experiments of Walker *et al.* [6]. The atom is helium, the laser wavelength is 780 nm, and the intensity is in the range $10^{14}-10^{15}$ W/cm^2. Since it is not at present feasible to do a complete two-electron calculation for this process, we have made some simplifying approximations which greatly reduce the computational requirements, but should not compromise our conclusions. The model is based on one first proposed and used by Pindzola, Griffin, and Bottcher [8] to study correlation effects in the multiphoton ionization of helium. In their model both electrons are constrained to move only along the direction of polarization of the laser field, reducing the wave-function spatial representation to two dimensions. In this type of model the singularities in the one-dimensional (1D) Coulombic interactions can present numerical problems, so we have used the standard procedure [9] of softening the interactions by giving the particles a finite extent,

$$\frac{q_i q_j}{|z_i - z_j|} \rightarrow \frac{q_i q_j}{\sqrt{a_{ij} + (z_i - z_j)^2}} \; . \tag{1}$$

The magnitude of a softening parameter a_{ij} depends on the particular interacting particles i and j. These parameters, which strongly effect the energy levels in the system, have been chosen to give the correct binding energies for the electrons. We will present results for a single electric-field strength $\mathcal{E}$ of 0.15 a.u., which corresponds to a laser intensity I of 7.9×10^{14} W/cm^2. This is approximately the saturation intensity for the 160-fs pulses used by Walker *et al.*, so it is expected to give the largest possible enhancement relevant to this experiment.

Here we treat only one of the 1D electrons quantum mechanically and consider the tunneling electron to be a point charge whose motion is classical. So the calculation has two separate steps. First we consider that one of the electrons e_1 has some probability to tunnel from the neutral helium atom through the suppressed barrier at each instant of time during the optical cycle. For this step we make a single-active-electron (SAE) approximation [10], where only one of the electrons is allowed to respond to the laser field, moving in the potential of the nucleus screened by the time-independent (ground-state) charge distribution of the remaining electron. The tunneling electron is assumed to be born with zero velocity at the turning point on the outer edge of the barrier at the initial-state binding energy (Fig. 1) with a rate corresponding to dc tunneling through the instantaneous barrier [11]. To determine the initial position of this electron, we use a nuclear attraction potential given by Eq. (2.1) with $q_{e_1} q_{\mathrm{He}^+} = -1$ and $a_{e_1 \mathrm{He}^+} = 0.5$. The ground-state energy for this 1D potential is 24.3 eV, which is a good approximation to the $1s^2$ He binding energy. The initial positions following tunneling are at least $5a_0-6a_0$ from the nucleus, depending on the phase of the field. In this step we follow tunneling events which occur during the first half cycle. After tunneling at some time t_0, e_1 evolves in this attractive ion core potential and the slowly varying field of the laser.

While the first electron is being accelerated away from the ion core, the second electron is assumed to relax adiabatically into the ground-state orbital of He^+. We make this adiabatic assumption regarding the relaxation of the second electron in order to test the efficacy of the rescattering mechanism. If in fact the departure of the first electron is somewhat nonadiabatic, the second electron will have some probability distributed among the excited states and the continuum of the ion. The bound excited-state components will be very rapidly ionized in this strong field because all of the field-free excited-state energies of the real helium ion lie above the suppressed barrier at the peak of the electric field. We known from single-photon ionization studies in helium that at high energies the second electron is "shaken off" directly into the continuum 3–4 % of the time in the sudden limit. As the first electron leaves, the second electron may be simultaneously freed due to the sudden loss of screening of the nuclear charge. In this limit roughly the same amount is "shaken up" to be left in excited bound states [13]. Since the multiphoton double-ionization fraction is observed to be more than an order of magnitude smaller than this, it is clear that the tunneling ionization is not in the sudden limit. However, if it turns out that the nonadiabaticity of the escape of the first electron can be shown to be large enough to account for the observed double ejection, there would be no need to invoke a rescattering mechanism. Since we are not able to estimate the degree of nonadiabaticity in tunneling, we have chosen to attempt to estimate the maximum excitation probability that rescattering, with the assistance of the field, can produce in the adiabatic limit.

At the end of the first half cycle t_1, the second electron is bound in the soft Coulomb potential of the unscreened nucleus with $q_{e_2}q_{He^{+2}}=-2$ and $a_{e_2He^{+2}}=0.5$. With this choice of parameters, the ionization potential of the 1D He^+ electron agrees exactly with that for the 3D ions, 54.4 eV.

The distribution of energies of the returning electrons is obtained by following the motion of the tunneling electrons classically [1,2]. Once an electron reaches the continuum, the density of states is high enough that classical mechanics should provide an accurate guide to the dynamics. The validity of this quasiclassical model is supported by the agreement between the predicted and measured shapes and cutoffs of the harmonic emission spectra as functions of the wavelength and intensity of the incident laser [1,2]. These spectra are produced by the returning electrons. Electrons released during most of the first quarter of an optical cycle have drift energies large enough that they never return to the nucleus. Those electrons released near or after the peak of the field are the ones which eventually return during the following half cycle. In Fig. 3(a) we show the calculated return energies and tunneling ionization rates as functions of ϕ_0, the phase of laser field at the "tunneling time." The return energies are obtained by running trajectories for different initial phases with initial conditions described above and determining the energy in excess of the binding energy when the trajectory recrosses the nucleus.

As can be seen in Fig. 1 for $\mathcal{E}=0.15$, the effective potential barrier is suppressed almost to the point that the initial state becomes unbound, resulting in a very high ionization rate. The instantaneous dc tunneling rate is very strongly peaked near the maximum of the field, indicating that only those trajectories initiated near the peak are important. Calculations for many wavelengths and intensities have shown [1] that the maximum return energy scales approximately with a multiple of the ponderomotive energy U_p $(=I/4\omega^2$ a.u.), which is the cycle-averaged energy of a free electron in the oscillating field. At this wavelength and intensity the maximum return energy for trajectories with significant tunneling probability

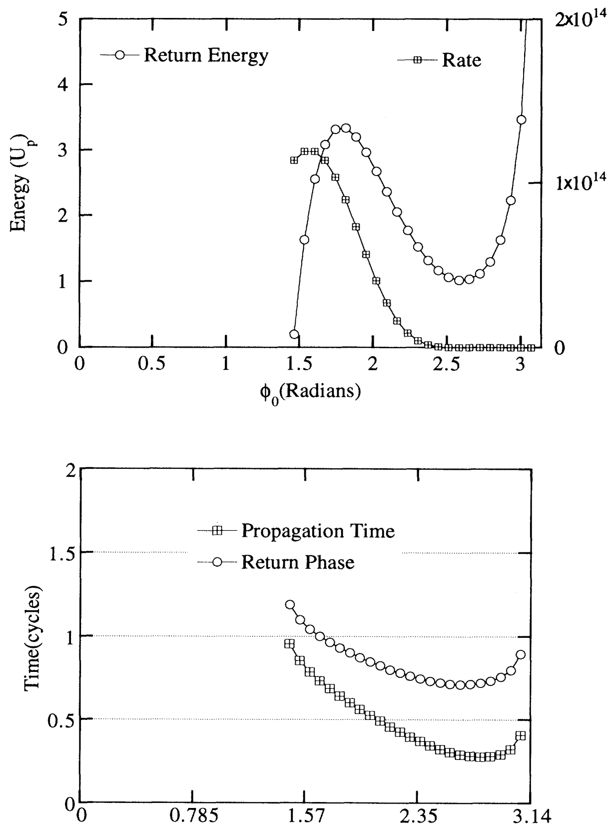

FIG. 3. (a) Return energies and tunneling rates for helium at 780 nm and 7.9×10^{14} W/cm^2 are shown as functions of their initial phase ϕ_0. Energies are scaled with the ponderomotive energy $U_p=44.8$ eV. (b) Time of propagation before rescattering from the ion core and the phase (in cycles) of the driving field at return are shown as functions of the initial tunneling phase.

is $3.3U_p$. This is slightly larger than what one finds by neglecting the ion potential and the initial displacement ($3.17U_p$) [2]. In the absence of an accompanying laser field, this return energy must exceed the lowest excitation energy for the ionic ground state to be affected. At 780 nm and 7.9×10^{14} W/cm^2, U_p is 44.8 eV, so the maximum return energy is close to 150 eV.

Because the tunneling electrons are first accelerated away from the ion, it takes approximately one-half cycle before they return to rescatter from the ion core. In Fig. 3(b) we show the phase of the field when the electron returns and the time the trajectory takes to return to the nucleus as functions of ϕ_0. These two factors are important for characterizing the rescattering process. The first defines how much the ion core states are distorted by the instantaneous electric field when the first electron returns and the second determines the transverse spread of the trajectories at the time of rescattering. The tunneling wave packet propagates outside the influence of the ion core so its transverse dimension should be comparable to that of a freely spreading Gaussian wave packet. This width in atomic units is given by $\alpha_t=\sqrt{\alpha_0^2+(2t/\alpha_0)^2}$, where α_0 is the width at $t=0$. At time t, α_t can be no smaller than $\sqrt{4t}$ corresponding to an initial width of $\alpha_0=\sqrt{2t}$. Therefore the minimum width of the returning wave packet after one-half period (54 a.u.) at this wavelength is $14.7a_0$. However, this corresponds to having an initial width of $10.4a_0$, which is unphysically broad for ionization from helium. For a more realistic α_0 of $3a_0$–$4a_0$, α_t is approximately $30a_0$.

During the second step in the calculation e_1 is turned around by the field and then rescattered by the ion core. We mimic the transverse spreading of the trajectories in our calculations by giving the trajectory of e_1 a fixed impact parameter b at the end of the first half cycle (t_1) when the electron is far from the atom, repeating the calculation for a distribution of impact parameters. The quantum-mechanical evolution of the second electron e_2 is found by integrating the time-dependent Schrödinger equation (TDSE)

$$i\frac{\partial\psi(z_2,t)}{\partial t}=\left[-\frac{1}{2}\frac{\partial^2}{\partial z_2^2}-\frac{2}{\sqrt{a^2_{e_2\mathrm{He}^{+2}}+z_2^2}}+\frac{1}{\sqrt{b^2+[z_2-z_1(t)]^2}}-z_2\mathcal{E}(t)\right]\psi(z_2,t)\ , \tag{2}$$

beginning at time t_1. The initial condition for $\psi(z_2,t=t_1)$ is the He^+ ground state. In this equation $z_1(t>t_1)$ is obtained from the solution of the classical equations of motion for e_1 in the effective potential defined using $a_{e_1\mathrm{He}^+}=b^2$. This electron accelerates as it passes the nucleus, but is not deflected because the impact parameter is held fixed. Depending on the phase of the field when e_1 is rescattered, the slowly varying field can act in concert with the passing point charge or against it. In Fig. 2 we have emphasized the fully interacting part of the calculation by the heavy dashed part of the trajectory. We neglect the exchange of energy between the two electrons, so the model is not appropriate for investigating threshold effects where the real cross sections may be small. Near threshold it is possible that the relative enhancement of the inelastic cross section by the field is large, but since we are interested only in the magnitude of the total inelastic cross section averaged over a distribution of collision (return) energies, our conclusions about the field effects on double ionization via the rescattering mechanism should be adequately addressed by these calculations.

The velocity of the returning electron is 2–3 a.u., meaning the strong interaction time between the electron, and the ion is on the order of a few atomic time units. Comparing this to the laser period 107.56 a.u., we observe that the field is effectively static during the collision. Therefore one might expect little effect from its presence save its Stark distortion of the excited states of the ion. In particular, at the peak of the field, all excited states in our model ion above the first one will be field ionized; their energies lie above the barrier. We note the first excited state in 1D He^+ lies at 29 eV, much lower than in the real ion (41 eV), making the 1D ion easier to excite.

Field-assisted cross sections are obtained as follows. We first pick an initial phase for the tunneling that will result in a trajectory for e_1 that returns to the ion core. The classical equations of motion for this electron are solved from the initiation time until the end of the first half of the period. We then choose an impact parameter and solve the TDSE for the quantum evolution of the remaining bound electron e_2. This calculation runs for one or more additional half cycles, until e_1 has moved well past the ion on its trajectory. At the end of each half cycle we can project the time-dependent wave function of e_2 onto the field-free states of the ion to determine the extent of excitation and ionization. For this wavelength and intensity the laser-induced excitation plus ionization rate is calculated to be less than 10^6 s^{-1}, so that in the absence of the rescattering electron, an extremely small depletion of the ground state is found, about 10^{-9} per cycle. With the electron-electron interaction on, substantial excitation is produced when the electron passes close enough to the ion. To determine the magnitude of the effect the laser field has on the inelastic cross section, we can omit the last term on the right-hand side of Eq. (2.2) to obtain a comparable field-free inelastic probability *for the same trajectory*. That is, e_1 still moves in response to the laser field, but e_2 is excited only through its interaction with e_1.

III. RESULTS

Performing both the field-on and field-off calculations described above for a range of impact parameters, we find the excitation probabilities for fixed rescattering energy. We define the total inelastic probability

$$P(b)=1-|\langle\psi(z_2,t_f)|\psi(z_2,t_1)\rangle|^2\ , \tag{3}$$

which is the fraction of the wave function removed from the ground state either to excited states or directly into the continuum at the end of the integration $t=t_f$. Because negligible excitation occurs of the ground state by the laser field alone, this probability becomes constant in less than a cycle after the collision. Similarly $P_0(b)$ is the inelastic probability obtained without the laser-electron interaction in Eq. (2.2). We also calculate the excitation probability for the first excited state, which we call the $2p$ state since it is of odd parity. In the field-off calculation this becomes a time-independent quantity after e_1 has moved away from the ion, but with the laser-ion interaction present, this probability decays rapidly due to ionization. We denote these respective excitation probabilities as $P_0(2p)$ and $P(2p)$, where the b dependence has been suppressed.

The collision energy is varied by changing the initial phase ϕ_0 as shown in Table I. Here we also give the phase of the laser field when the electron rescatters. Those trajectories with return phases near 6.28 rad (full cycle) rescatter as the oscillating field vanishes. Those near 4.7 rad ($\frac{3}{4}$ cycle) find the field at its maximum. We present results for three specific cases $\phi_0=1.8$, 2.0, and 2.2 rad. In these three cases the impact energy is declining as the field strength upon return is increasing. The inelastic probabilities obtained with and without the field as functions of b are plotted in Fig. 4. It is clear that the addition of the field can increase or decrease the inelastic probability, but only by a small amount.

The total inelastic cross sections as functions of rescattering energy are given by

$$\sigma(E)=2\pi\int_0^\infty bP(b)db \ , \tag{4}$$

with a similar expression for the field-free cross section σ_0. Repeating these calculations for a number of initial phases ϕ_0 corresponding to range of collision (rescattering) energies, we obtain the energy dependence of the integral cross sections shown in Fig. 5. The field-assisted results oscillate around the field-free cross sections, never differing by more than 10–20 %. In this figure we also show the field-free excitation cross section for the $2p$ state. This is the dominant channel over the whole energy range investigated. The magnitude of these cross sections is approximately 3–4 times larger than those for the 3D system, mostly reflecting, we believe, the fact that the excited states in the 1D ion have much lower transition

TABLE I. Electron rescattering parameters.

Initial phase	Rescattering energy	Rescattering phase
1.6 rad	112 (eV)	6.5 rad
1.8	150	5.8
2.0	124	5.3
2.15	95	5.0
2.2	86	4.9
2.3	67	4.8
2.8	58	4.5

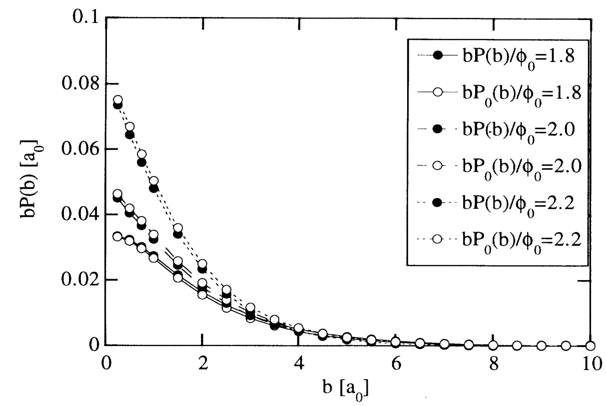

FIG. 4. Total inelastic probability as a function of the impact parameter for $\phi_0=1.8$, 2.0, and 2.2 rad. $P(b)$ is the probability obtained including the effect of the laser field and $P_0(b)$ is the field-free result.

energies and therefore are more easily excited.

The time evolution of the wave function density distribution is shown in Figs. 6(a) and 6(b) for a particular calculation. In this case the rescattering electron returns almost one-half cycle after the beginning of the calculation, time t_1 defined in Fig. 2, or about one-quarter of the way through the two-cycle evolution shown. Figure 6(a) [6(b)] shows the field-off (field-on) evolution. In the upper figures, the total 1D charge density is shown. When the electron rescatters (from right to left in the figure), some small fraction of the wave function is knocked into the forward direction by the passing electron. In the lower figure we show only the excited density after projecting out the ground state to emphasize the excitation dynamics. In the field-free case there is some prompt ionization at the time of the collision, followed by small scale oscillations of the electron density due to the large number of

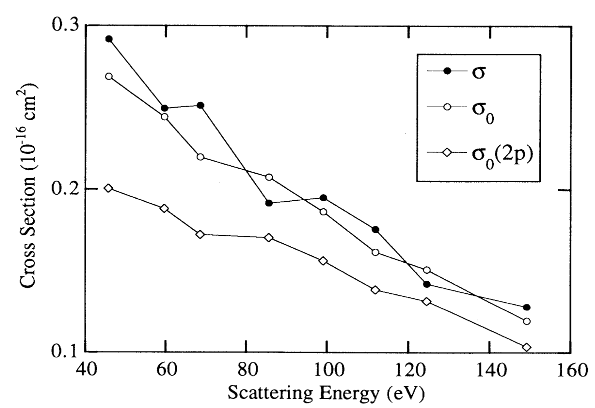

FIG. 5. Total inelastic cross sections as functions of the rescattering energy including the laser field σ and without the field σ_0. Also shown is the field-free cross section $\sigma_0(2p)$ for exciting the $2p$ state of the ion.

bound states excited. The structure of the wave function clearly shows the dominance of the $2p$-state excitation. In Fig. 6(b) polarization of the wave function due to the field is seen to occur well before the scattering electron arrives. After the prompt emission following the collision, ionization is evident flowing first to the right, then to the left, and back to the right in phase with the field. This is more clearly seen in the upper figure, which shows the total wave-function density. After a few cycles, virtually all of the excited-state density will be removed by ionization. These figures show very dramatically how the evolution of the excited probability differs when the field is present or not. However, only be performing the final projections can we conclude that the total inelastic probability is not appreciably altered by the field.

Finally, in Fig. 7 we have plotted the fractional population left in the $2p$ state after one cycle of the field for both the field-off and field-on cases as functions of the impact parameter for the initial phases considered in Fig. 4. In the field-free case, the distant collisions lead almost exclusively to $2p$ excitation. If the laser-electron interaction is present, the excited-state distribution is very different, partly due to the ionization by the field which begins immediately after excitation.

IV. CONCLUSIONS

Although the laser field used in these studies is quite strong, its effect on the total excitation plus ionization probability is quite small. This means that the rescattering mechanism for direct double ionization in helium is unlikely to be the dominant process responsible for the observed data. We found the field does affect the final-state distributions, producing more excitation and eventually complete ionization after the rescattering electron has passed. One might ask whether even higher fields might lead to larger effects, but this ionization pathway must compete with the direct ionization of the ion by the field. Just above 10^{15} W/cm^2 tunneling ionization of the helium ion becomes important and the sequential stripping begins to overwhelm the direct signal. These results give support to the idea that the double emission is a result of a small amount of nonadiabaticity during tunneling ionization.

As stated earlier, the rescattering process *does* produce the harmonic photon emission spectra [1] and the high-energy photoelectrons with structured angular distributions [14,15]. However, in the tunneling regime these emission channels involve only a small fraction of all

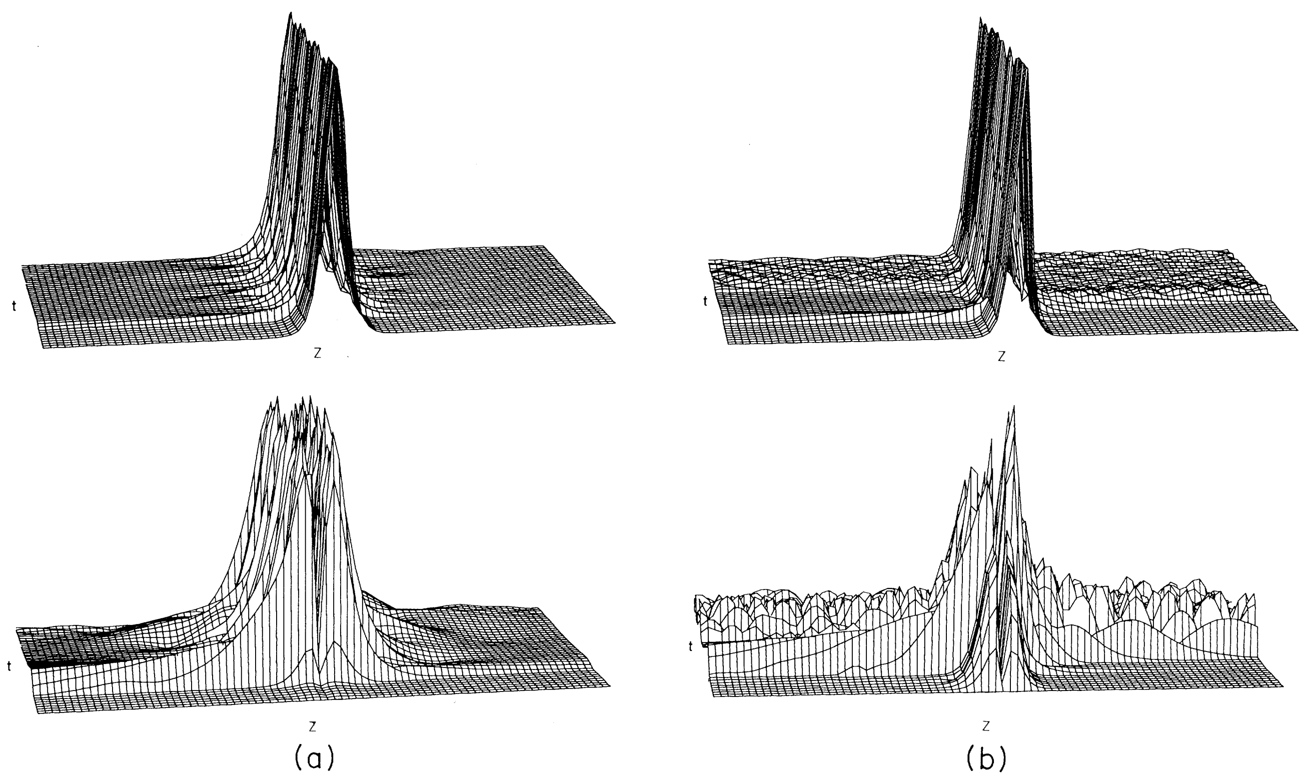

FIG. 6. Time evolution of the 1D He^+ wave function over two optical cycles for one of the cases shown in Fig. 4: $\phi_0 = 1.8$ rad and $b = 1a_0$. The horizontal axis is the spatial dimension from $-40a_0$ to $40a_0$ and the dimension into the picture is increasing time. The figures are (a) the field-free and (b) the laser-assisted cases. In each figure the upper plot is the density distribution of total wave function and the lower plot the excited component, i.e., that for the total wave function with the ground state projected out. Note the strong excitation of the $2p$ state in both cases in the lower plots.

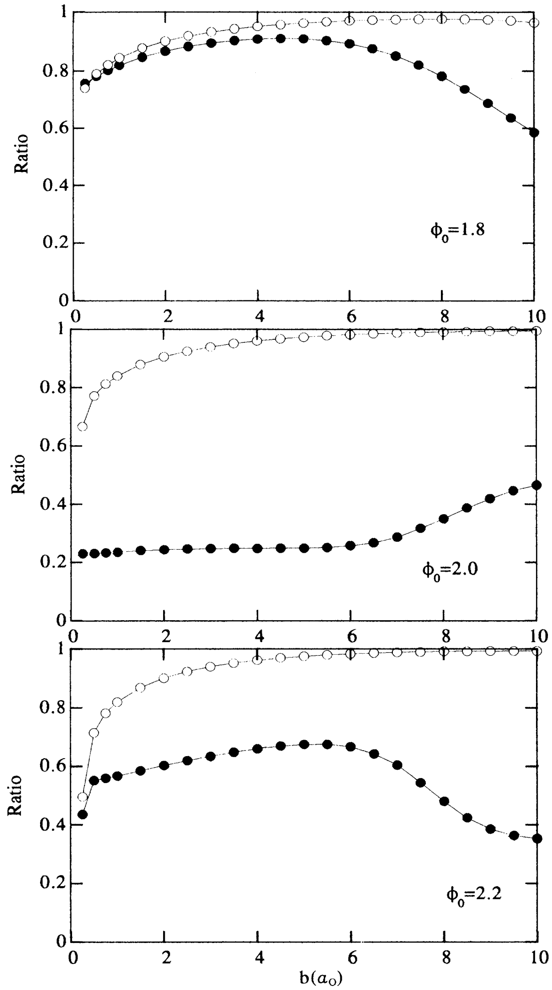

FIG. 7. Effect of the laser field on the final population in the excited $2p$ state for the three cases shown in Fig. 4. Plotted are the fractions of the total inelastic probability found in the $2p$ state as functions of the impact parameter. For the field-on (filled circles) case the projection was done after the first full cycle $(t_1+2\pi)$. Later projections in this case find less in the $2p$ state due to ionization. The field-off data are indicated by open circles.

emitted electrons, so the existence of these known processes is consistent with the conclusion that inelastic rescattering is too infrequent to cause the observed nonsequential ionization.

These results should not be too surprising since there has been a report of an extensive, thorough, and completely unsuccessful search for radiation from inner-shell excitations in atoms in very strong laser fields [12]. Since electron impact core excitation cross sections are no more than one or two orders of magnitude smaller than that for e-$2e$ in the helium ion, these measurements indicate a very small number of rescattering events occurs at small impact parameters. Irradiating xenon with 248-nm 700-fs pulses with intensities up to 3×10^{17} W/cm^2, Lee, Casperson, and Schappert put an upper limit of 2.8×10^{-7} on the probability of an atom emitting a photon from inner-shell excitation or by a bremsstrahlung mechanism. They state that "there are no measurable prompt photons of energy greater than 50 eV, regardless of the generating mechanism."

Finally, we note that in recent classical trajectory studies for a model helium atom [16], the intensity dependence of the He^{+2} yield has two components which can be interpreted to show the existence of two separate rates, one for sequential ionization and the second for direct double ionization. This is at least qualitatively consistent with the experimental data. Surprisingly a calculation for the multiphoton ionization of He^+ alone also showed a two-rate structure, which is highly improbable at these long wavelengths. Quantum calculations show no such structure. However, an analysis of the electron energy distributions indicated that, for a narrow range of intensities, correlated double emission may be occurring. This is predicted to occur only at intensities considerably above the onset observed by Walker *et al.* [6]. This discrepancy may in part be explained by the fact that trajectory calculations cannot include effects due to the nonadiabaticity during the tunneling ionization of the first electron. Tunneling is forbidden classically. Perhaps a more detailed analysis of individual trajectories might provide a clearer picture of the mechanism which leads to the classical double ejection.

ACKNOWLEDGMENTS

This research was carried out in part under the auspices of the U. S. Department of Energy at Lawrence Livermore National Laboratory under Contract No. W-7405-ENG-48. J. C. is supported in part by NSF Grant No. PHY90-12244. K.C.K. wishes to acknowledge support from the Visiting Fellow Program at JILA during the time this work was being carried out.

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