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Molecular-dynamics simulations of atomic ionization by strong laser fields

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We use molecular-dynamics simulations to study the competition between sequential and collective ionization in low-Z atoms by strong laser fields. The model shows sequential ionization at low fields, changing to collective ionization at large fields. The field strength at which this occurs is lower for higher frequencies. We also study the ionization as a function of pulse length and find that collective ionization is favored for shorter pulses.

I. INTRODUCTION

The development of high-powered, fast-pulsed lasers allows one to study the response of a many-electron atom to a strong electromagnetic field. The nature of this response is currently a topic of much debate. In small fields the atom ionizes to the state with the lowest available energy, in which one electron has absorbed the minimum number of photons necessary for ionization and the remaining ion is left in its ground state. In strong fields, it is observed that the atom can ionize to higher energy states. In particular, the ionized electrons are observed to have absorbed more than the minimum number of photons needed for ionization and, in strong enough fields, multiple ionization occurs.[1] In this paper, we concentrate on the mechanisms by which multiple ionization occurs, particularly whether the ionization occurs sequentially or collectively. In sequential ionization, one electron ionizes, leaving the resulting ion with little or no excitation. This is followed by the next electron ionizing, leaving the double-charged ion in its ground state, and so on. This response, which is observed to dominate in small fields, concentrates all of the energy being absorbed from the field in one electron at a time. In contrast, one can imagine collective ionization occurring, in which several electrons simultaneously absorb energy and ionize.

Multiple ionization of noble-gas atoms at intensities up to 10^{14} W/cm^2 at wavelengths of approximately 500 nm and larger have been studied by several groups, who find that their data can be well fit by models that assume sequential ionization.[2] Studies of this multi-ionization at stronger fields and smaller wavelengths ($\sim$200 nm) have found evidence that sequential ionization might be breaking down in the production of the higher charge states.[3] Geltman *et al.* have suggested that the charge states produced can be accounted for by a model that assumes the electrons ionize independently, rather than sequentially.[4]

Theoretical studies have also suggested the possibility that sequential ionization will break down in strong fields. Kulander has integrated the time-dependent Hartree-Fock (TDHF) equations for helium and seen evidence that a doubly excited state will play a major role in the ionization.[5] Using many-body perturbation theory, L'Huillier and Wendin have suggested that the ionization dynamics will have a frequency dependence, with multiply excited states playing a more important role at higher frequencies.[6] Such excitations could result in the ionization occurring other than sequentially.

We use molecular-dynamics simulations to study these issues. The advantage of these simulations is that one can solve exactly the classical equations of motion for a many-body system numerically, something that cannot be done for the corresponding quantum equations. This allows us to study exactly, within these models, the effect of a strong applied field of an electron-electron repulsion. In particular, the atom is free to respond to an external field without any imposed constraints. In addition, classical simulations allow easy contact with all the observable quantities. A defect is that resonant processes are ignored. The specific potentials we use are due to Kirschbaum and Wilets.[7] These potentials simulate the effects of the Heisenberg uncertainty and the Pauli exclusion principles.

This paper is organized as follows. In Sec. II, we discuss the molecular-dynamics model employed and review its static properties. In Sec. III, we use it to study the response of helium and beryllium to various applied fields. Section IV contains our conclusions.

II. MODEL

Our model of an N-electron atom follows Ref. 7. The N classical electrons interact with each other via two-body potentials and with the nucleus via a one-body potential. In addition to the standard Coulomb terms, one adds potentials $V_H(r_i,p_i)$ and $V_P(r_{ij},p_{ij})$ that simulate the Heisenberg and Pauli principles by not letting the quantities r_ip_i and $r_{ij}p_{ij}$ become too small. Here $\mathbf{r}_i$ and $\mathbf{p}_i$ are the coordinates and momenta of the ith electron and $r_{ij}=|\mathbf{r}_i-\mathbf{r}_j|$ and $p_{ij}=|\mathbf{p}_i-\mathbf{p}_j|$. Specifically, the Hamiltonian for the atom is

$$H_{\text{atom}}=\sum_i\left[\frac{p_i^2}{2m}-\frac{Ze^2}{r_i}+V_H(r_i,p_i)\right]+\sum_{i<j}\left[\frac{e^2}{r_{ij}}+V_P(r_{ij},p_{ij})\delta_{s_i,s_j}\right], \quad (1)$$

where

$$V_H(r_i,p_i)=\frac{(\xi_H\hbar)^2}{4\alpha r_i^2 m}\exp\left\{\alpha\left[1-\left(\frac{r_i p_i}{\xi_H\hbar}\right)^4\right]\right\} \tag{2}$$

and

$$V_P(r_{ij},p_{ij})=\frac{(\xi_P\hbar)^2}{4\alpha r_{ij}^2 m}\exp\left\{\alpha\left[1-\left(\frac{r_{ij} p_{ij}}{2\xi_P\hbar}\right)^4\right]\right\} . \tag{3}$$

Here, M and $-e$ are the electron's mass and charge, $+Ze$ is the nuclear charge, and $s_i=\pm\frac{1}{2}$ is the conserved spin projection. The values of the potential parameters are $\alpha=5$, $xu_H=0.9535$, and $\xi_P=2.767$. The parameter α controls the rigidity with which the Pauli and Heisenberg phase-space constraints are implemented classically. For a given α, ξ_H is chosen to fit the binding energy and Bohr radius of hydrogen and ξ_P is chosen to give the correct Fermi energy of a uniform electron gas.[7]

The equations of motion for the atom in the presence of a uniform external force field $\mathbf{F}_{\text{ext}}(t)$ are

$$\begin{aligned} \frac{d\mathbf{r}_i}{dt}&=\frac{\partial H}{\partial \mathbf{p}_i} , \\ \frac{d\mathbf{p}_i}{dt}&=-\frac{\partial H}{\partial \mathbf{r}_i}+\mathbf{F}_{\text{exp}}(t) . \end{aligned} \tag{4}$$

The total energy of the atom evolves in time as

$$\frac{dH_{\text{atom}}}{dt}=\sum_i \frac{\partial H}{\partial \mathbf{p}_i}\cdot\mathbf{F}_{\text{exp}}(t)=\sum_i \mathbf{v}_i\cdot\mathbf{F}_{\text{ext}}(t) , \tag{5}$$

where $\mathbf{v}_i=d\mathbf{r}_i/dt$ is the velocity.

The ground state of the atom is determined by minimizing the energy of the system, for example, by solving the classical equations of motion with a dissipative term. The Heisenberg term serves to bound the total energy from below, resulting in a finite binding energy. The resulting atoms have ground-state energies and rms radii that agree reasonably well with experimental data.[7] A particularly relevant quantity for the study of ionization is the separation energy, shown in Table I for systems with $Z=2-5$. This model is clearly able to reproduce the structure of the 1s and 2s shells although the 2s shell is overbound by a factor of 2.

There are an infinite number of degenerate classical configurations corresponding to the ground state of the atom and any calculation of the response of the atom must average over all of these initial states. From a given configuration, one can generate the other configurations by using the symmetry properties of the Hamiltonian. The Hamiltonian, Eq. (1), is invariant under separate rotations of all the position vectors and all the momentum vectors. In addition, one can independently reflect all the position or all the momentum vectors. We generated successive initial states by performing all of these operations randomly on one "parent" configuration.

We integrated Eq. (4) numerically using a Runge-Kutta method. We verified the accuracy of the integration by checking that our results stayed compatible with Eq. (5) to an accuracy of at least 10^{-4} and that angular momentum along the direction of the applied-field was conserved to the same accuracy.

TABLE I. Energy required to remove a single electron from an ion of charge N for atoms with atomic number Z in the molecular-dynamics model (eV). Experimental values are given in parentheses.

N	$Z=2$	$Z=3$	$Z=4$	$Z=5$
0	28.9 (24.5)	12.2 (5.4)	22.6 (9.3)	10.2 (8.3)
1	54.5 (54.4)	83.4 (75.6)	38.3 (18.2)	54.4 (25.2)
2		123 (122)	165 (160)	77.6 (37.9)
3			218 (218)	265 (259)
4				341 (340)

III. MANY-ELECTRON RESPONSE

We now present results on the response of these model atoms to various applied electric fields, concentrating on the mechanisms by which ionization occurs. We first analyze the response to monochromatic waves in order to understand the effect of varying the frequency and the amplitude on the ionization dynamics. We then study the response to the more realistic situation of pulsed waves.

We study the ionization dynamics of focusing on the behavior of the individual electrons. For sequential processes, we expect only one electron to be active at a time, while for collective processes we expect to see several electrons active simultaneously. To study the nature of the response, we look at several quantities.

The first quantity is the time at which the electrons are ionized. An ionized electron is one which will escape to spatial infinity with a nonzero velocity. The time at which the electron is ionized corresponds to the moment at which the electron obtains enough energy to escape and then uses this energy to escape the atom. Unfortunately, such a time is not uniquely defined. Rigorously, we can only determine whether an electron is ionized by turning off the field and observing whether or not the electron remains unbound. Thus, by turning off the field after a given amount of time, we can determine how many electrons were ionized, but this tells us nothing about how they were ionized. Nonetheless, we can establish a criteria for when an electron has obtained a level of activity in the presence of the field that is consistent with ionization, and this allows us to determine a time of ionization. We operationally define an electron as being ionized when its single-particle energy (to be defined shortly) has become positive and it has moved a distance R_0 (typically $R_0=4$ Å) away from the nucleus. Except where noted, we will use the term "ionized" to refer to an electron that has met this criteria. One might worry that this definition breaks down in strong fields if the electron is driven in a large amplitude oscillation about the nucleus, as might occur when the quiver distance of a free electron becomes comparable to R_0. In such cases, the transverse momentum of the electron causes the electron to leave the vicinity of the atom, so a stable oscillation about the nucleus is, in general, not set up.

The single-particle energy of the ith electron at an instant in time is calculated as

$$E_i=\frac{p_i^2}{2m}-\frac{Ze^2}{r_i}+V_H(r_i,p_i)+\frac{1}{2}\sum_{j(\neq i)}\left[\frac{e^2}{r_{ij}}+V_P(r_{ij},p_{ij})\delta_{s_i,s_j}\right], \tag{6}$$

where the potential energy between pairs of electrons is arbitrarily divided equally between them. The definition of single-particle energy is somewhat arbitrary. This definition has the useful property that $E=\sum_i E_i$. An alternate definition that is more conventional is

$$E_i'=\frac{p_i^2}{2m}-\frac{Ze^2}{r_i}+V_H(r_i,p_i)+\sum_{j(\neq i)}\left[\frac{e^2}{r_{ij}}+V_P(r_{ij},p_{ij})\delta_{s_i s_j}\right]. \tag{7}$$

E_i' is the energy required to ionize an electron in the limit that the other electrons are static. Hence, in the limit of sequential ionization this is the appropriate single-particle energy to use. On the other hand, if all the electrons respond equally, then E_i is the appropriate single-particle energy.

In general, the true response will be somewhat between these two extremes. Since $E_i<E_i'$, ionization according to E_i will also correspond to ionization according to E_i'. If we use the latter definition, however, there could be multielectron excited states that are bound but would count as ionized by this definition of single-particle energy. A problem with the E_i definition is that if a particle ionizes sequentially it could still have a negative value of E_i. However, since $E_i\rightarrow E_i'$ as the electron escapes to infinity, the E_i definition will also record the electron as being ionized, although with a longer ionization time. In strong fields, the ionization usually occurs deep enough into the continuum that this is not a significant limitation. Hence, we use the E_i definition of single-particle energy in most of our analysis.

The second quantity we examine is the energy of the remaining atom when a given electron is ionized. For sequential ionization, the atom will be left in the ground state, while for collective ionization we should see some excitation.

The single-particle energy also contains information on the role of the electron-electron interaction in the ionization process. Since $E=\sum_i E_i$, Eq. (5) implies that the single-particle energy evolves in time as

$$\frac{dE_i}{dt}=\mathbf{F}_{\mathrm{ext}}\cdot\mathbf{v}_i+\left.\frac{dE_i}{dt}\right|_{e\text{-}e}, \tag{8}$$

where the first term is the rate at which the field supplies energy to the particle and the second term is the rate at which the particles distribute energy among themselves. As such, it satisfies

$$\sum_i\left.\frac{dE_i}{dt}\right|_{e\text{-}e}=0\ . \tag{9}$$

We can calculate the fraction of the energy used to ionize a particle that comes directly from the field (ε), as opposed to from the other electrons as

$$\varepsilon=\frac{E_f^*}{E_f-E_i}, \tag{10}$$

where E_f is the energy of the electron at the time of ionization t_f, E_i is the initial energy of the electron, and E_f' is the net energy supplied directly by the field

$$E_f^*=\int_0^{t_f}\mathbf{F}_{\mathrm{ext}}\cdot\mathbf{v}_i dt\ . \tag{11}$$

This provides a direct measure of the role of the electron-electron force in the ionization process. If the ionization is accomplished solely by the field acting on the electron, then $\varepsilon=1$, while if most of the energy is transferred from the other electrons, ε is small. Our definition of single-particle energy is not a conserved quantity for motion in the mean field of the other electrons. Thus, ε measures energy gained from the mean field of the other electrons, as well as correlations beyond the mean field. For instance, in the limit that the nonionizing electrons are static

$$\varepsilon=\frac{E_f-E_i'}{E_f-E_i}<1\ . \tag{12}$$

As the ionization becomes more collective $\varepsilon\rightarrow 1$. If collisions are important in the ionization process then they will tend to diminish ε for the first electrons to ionize and enhance it for the later ones.

We begin by analyzing the response of our model atoms to sinusoidal electric fields with an initial linear rise in amplitude,

$$F_{\mathrm{ext}}(t)=\hat{\mathbf{z}}F_0(t)\sin(\omega t)\ , \tag{13}$$

where $F_0(t)$ varies linearly from 0 to F_0 over a time t_{rise} and subsequently remains fixed.

We first study the response of helium to fields of varying strengths and frequencies. In particular, we choose fields of strength $F_0=10-80$ V/Å (corresponding to intensities of $1.3\times10^{13}-8.4\times10^{16}$ W/cm^2) and frequencies $\omega=5-60$ fs^{-1} (corresponding to photon energies of 3.3–39.5 eV), corresponding to 1–10 photon ionization of the first electron. These field strengths result in ionization rates in the femtosecond range. Processes over longer time scales require too much computer time to be studied. The simulations are typically run for 20 fs or until both electrons are ionized. The rise time is chosen to be $t_{\mathrm{rise}}=0.5$ fs. For each set of field parameters, runs are done on 10 different randomly chosen initial atomic configurations, as described in Sec. II. We find that, even with these few initial configurations, we can get good enough statistics to study the general properties of the ionization.

Results are tabulated in Tables II and III. Table II shows the average number of electrons ionized for vari-

TABLE II. Helium ionization times. N is the average number of electrons ionized in 20 fs and t_i is the average time at which the ith electron is ionized.

ω_0 (fs^{-1})	F_0 (V/Å)	N	t_1 (fs)	t_2 (fs)	t_2/t_1
5	10	0.2	15		
	20	1.2	4.1		
	30	1.9	1.1	6.0	5.5
	40	2.0	0.79	2.2	2.7
	50	2.0	0.47	1.0	2.1
	60	2.0	0.48	1.0	2.1
	70	2.0	0.37	0.61	1.6
	80	2.0	0.37	0.61	1.6
10	10	1.1	11		
	20	1.7	2.4	10	4.1
	30	1.9	0.74	3.6	4.9
	40	2.0	0.66	1.4	2.1
	50	2.0	0.53	0.93	1.8
	60	2.0	0.50	0.63	1.3
20	10	1.5	5.0	14.0	2.8
	20	1.8	1.5	3.4	2.3
	30	2.0	0.93	4.3	4.6
	40	2.0	0.62	1.3	2.1
	50	2.0	0.59	0.96	1.6
	60	2.0	0.49	0.68	1.4
40	10	1.2	7.2		
	20	1.7	2.4	3.4	1.4
	30	1.8	0.94	1.4	1.5
	40	2.0	0.87	1.3	1.5
60	10	1.4	5.5		
	20	1.8	2.2	5.8	2.6
	30	2.0	0.83	2.0	2.4
	40	2.0	0.77	1.3	1.6

TABLE III. Helium ionization energies: Average energy of the second electron (E) and value of ε of the first electron at the moment of ionization of the first electron.

ω_0 (fs^{-1})	F_0 (V/Å)	E (eV)	ε
5	10	-50	0.55
	20	-52	0.82
	30	-49	0.92
	40	-48	0.89
	50	-46	0.91
	60	-40	0.91
	70	-37	0.92
	80	-21	0.94
10	10	-48	0.72
	20	-50	0.82
	30	-51	0.82
	40	-47	0.92
	50	-37	0.93
	60	-28	0.94
20	10	-49	0.56
	20	-37	0.76
	30	-49	0.82
	40	-36	0.84
	50	-37	0.85
	60	-14	0.93
40	10	-44	0.66
	20	-23	0.80
	30	-24	0.81
	40	-11	0.86
60	10	-36	0.72
	20	-31	0.81
	30	-17	0.73
	40	-12	0.78

ous field configurations, the average times at which the first and second electrons were ionized in the double ionization events, and the ratio of these two times. At small fields, we do not always observe double ionization because of the limited duration of the simulation. Table III shows the average energy of the second electron and the value of ε of the first electron at the moment the first electron has ionized. Figures 1 and 2 plot representative data showing trends with increasing field strength and increasing frequency.

First consider the ionization rate data (Table II). Notice that while the average number of particles ionized and the rate at which the first particle is ionized increase with field strength [Figs. 1(a) and 1(b)] as expected, these quantities are roughly independent of the frequency [Figs. 2(a) and 2(b)]. This agrees with the TDHF calculations of Kulander.[5]

The rate that the second particle ionizes also increases with field strength and becomes closer to the rate that the first particle ionizes [Fig. 1(b)]. By looking at the ratio of these ionization times, we can say something about the ionization dynamics. If the two electrons ionize independently with equal probabilities, as in the Geltman model,[4] then one expects this ratio to be 3. If the ionization takes place sequentially, then each ion is more tightly bound giving it a slower ionization rate that results in this ratio being greater than 3. On the other hand, if the electrons are correlated in such a way that the ionize together then this ratio will be reduced below 3. The statistics on our calculation of this ratio are not particularly good, but one can see the general trend of this ratio to decrease with increasing field strength (Table II). This provides some evidence that the ionization process is changing from sequential to collective.

One has to be careful with these results since the ionization times, and particularly their ratio, are sensitive to both the definition of ionization and the details of the applied field. As one increases R_0, the electrons have to travel farther to be declared ionized and thus the ratio of their ionization times will tend towards one. Also, as the ionization rate tends toward the rise of the pulse, the second electron sees a stronger field than the first and thus ionizes quicker resulting in a ratio of ionization times nearer one. By varying these parameters, we have observed that the trend for the ratio of ionization times to decrease with field strength is not affected by them, although the absolute magnitudes are. As one takes T_0 to zero the value of the ratio tends nearer three at high fields.

We now consider the ionization energy data shown in

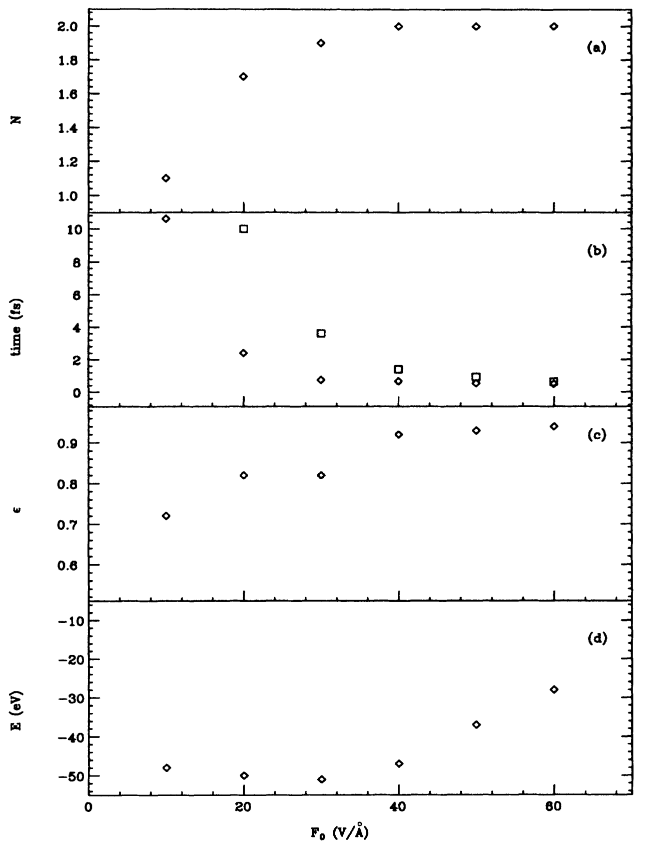

FIG. 1. Ionization properties of helium as a function of F_0 for $\omega = 10\ \mathrm{fs}^{-1}$: (a) average number of electrons (N) ionized in 20 fs, (b) average time of ionization of the first electron (diamonds) and the second electron (squares), (c) ε of the first electron to ionize, and (d) energy of the second electron (E) at the time the first electron ionizes.

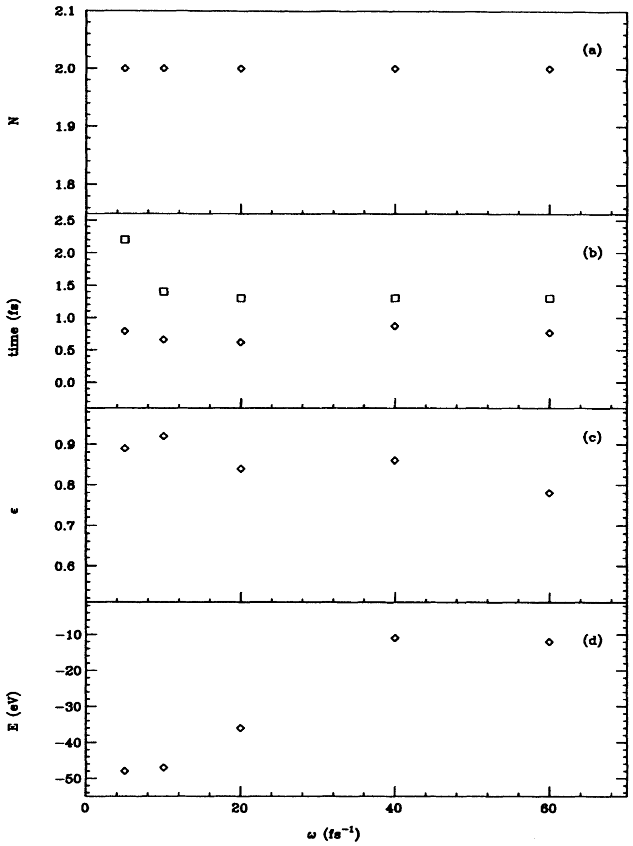

FIG. 2. Ionization properties of helium as a function of ω for $F_0 = 40$ V/Å: (a) average number of electrons (N) ionized in 20 fs, (b) average time of ionization of the first electron (diamonds) and the second electron (squares), (c) ε of the first electron to ionize, and (d) energy of the second electron (E) at the time the first electron ionizes.

Table III. At low frequencies and field strengths the remaining electron is left near the ground state (within several eV of -54 eV) indicating that sequential ionization has occurred. For sequential ionization in which the second electron is static, Eq. (8) gives a value of $\varepsilon = 0.7$; the actual values found in this low-frequency, low-field regime are comparable to this. At the lowest fields the values are smaller, indicating that some energy is absorbed from the applied field by the nonionizing electron and transferred to the ionizing one. Figure 3(a), which plots the single-particle energies versus time for a representative run, is an example of this process. We see that the time scale over which the actual ionization takes place (the transition from being tightly bound to unbound) is very fast (0.1 fs) compared to the ionization rate. During the ionization process, the ionizing electron gets all the mutual Coulomb energy from the remaining electron, leaving the resulting ion near its ground state.

As the field strength is increased at a given frequency, the ionization rate increases rapidly, becoming of order the period of the field, the nonionizing electron is left in an excited state [Fig. 1(c)] and the value of ε of the ionizing electron increases towards one [Fig. 1(d)]. This latter is consistent with both electrons more equally sharing their mutual Coulomb energy. In strong fields, the ionization rates of the two electrons become close enough that the second electron tends to become excited before the first electron has had a chance to leave the vicinity of the atom. This allows the second electron to recoil while the first is ionizing, resulting in increasing values of ε and E

As the frequency is increased at a given field strength we also find that the atom is left in increasingly excited states [Fig. 2(b)]. The ionization rate [Fig. 2(b)] and ε [Fig. 2(c)] are observed to be insensitive to the frequency, indicating that the energy transfer between electrons is primarily determined by the ionization rate. On the other hand, the excitation of the second electron increases, indicating that there is significant interaction between the two electrons prior to ionization at higher frequencies. Figure 3(b) shows a representative example of the high-frequency ionization process. The ionization starts as in

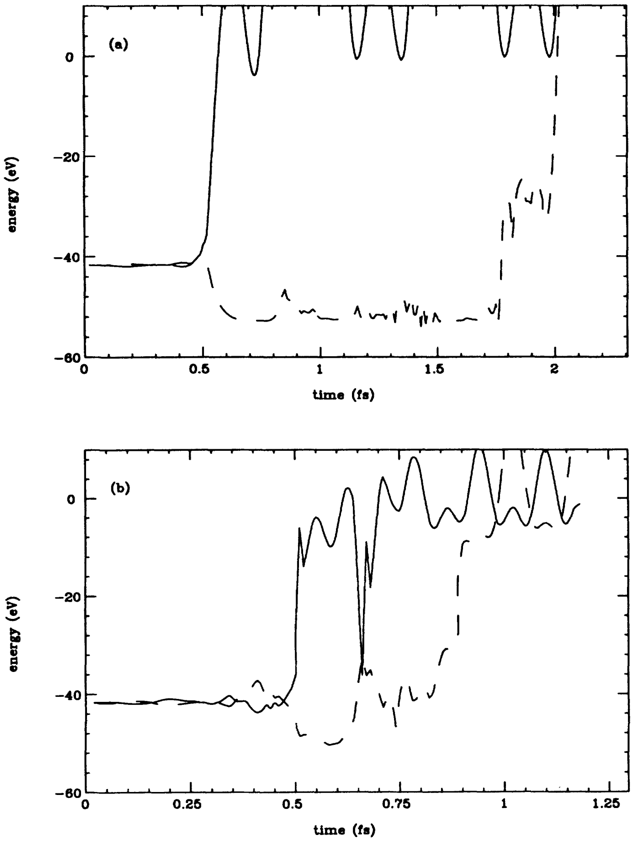

FIG. 3. Single-particle energies for representative responses of He to monochromatic waves with (a) $\omega=10$ fs^{-1}, $F_0=40$ V/Å and (b) $\omega=40$ fs^{-1}, $F_0=40$ V/Å. (a) is an example of sequential ionization and (b) is an example of collective ionization.

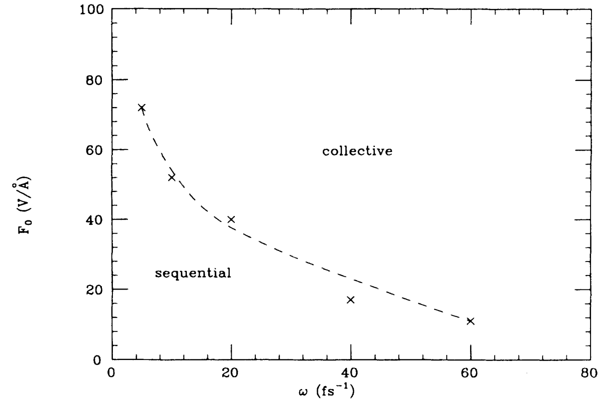

FIG. 4. Parameter ranges of F_0 and ω corresponding to sequential and collective ionization of helium by a monochromatic field as defined in the text.

Fig. 3(a), but then the first electron stops short of ionizing and returns near to its ground state before barely ionizing into the continuum. The key criteria that makes nonsequential ionization easier at high frequencies is that the ionizing electrons tend not to ionize very far into the continuum due to the smaller "quiver" energy associated with the motion of the electron in the external field (the quiver energy is proportional to F_0^2/ω^2).[8] This means that the ionizing electron spends a longer amount of time near the atom which enhances its likelihood of interacting with the bound electron during ionization.

The above discussion has shown that the ionization dynamics of an atom changes from sequential to collective with increasing field strength. In strong fields, the collective ionization appears as a result of the ionization rate becoming comparable to the limit it takes an ionizing electron to leave the atom. At higher frequencies one also sees significant collective interaction between the electrons prior to ionization. This results in the transition to collective ionization occurring at lower field strengths for higher frequencies. This is summarized in Fig. 4 where we arbitrarily define the transition as occurring when the second electron has obtained a single-particle energy of −35 MeV that corresponds roughly to half the energy needed to populate the first excited state of a real He$^+$ atom.

We can further study the role of electron correlations in helium by altering the way one of the electrons responds to the field. Consider the case in which only one electron feels an interaction with the external field. This changes the equation of motion for the momenta in Eq. (4) to

$$\begin{aligned} \frac{d\mathbf{p}_1}{dt} &= -\frac{\partial H}{\partial \mathbf{r}_1} + \mathbf{F}_{\text{ext}}(t) \,, \\ \frac{d\mathbf{p}_2}{dt} &= -\frac{\partial H}{\partial \mathbf{r}_2} \,. \end{aligned} \tag{14}$$

Table IV shows results of simulations under these conditions for $F_0=20$ and 40 V/Å at frequencies that correspond to sequential ($\omega=10$ fs^{-1}) and collective ($\omega=20$ and 40 fs^{-1}) ionization in the fully interacting case. The results for $\omega=10$ fs^{-1} are identical to those when both electrons interact with the field, indicating that the interaction of the nonionizing electron with the applied field plays no role in sequential ionization. In contrast, in the collective ionization regime, the interaction of the

TABLE IV. Helium ionization: one interacting electron. Same notation as Table II is used. R is the ratio of the corresponding values of t_1 from this table and Table II.

ω_0 (fs^{-1})	F_0 (V/Å)	E (eV)	t_1 (fs)	R
10	20	−50	2.4	1.0
	40	−51	0.64	1.0
20	20	−44	2.7	1.8
	40	−39	0.79	1.3
60	20	−48	3.4	1.4
	40	−41	1.1	1.4

field with the second electron plays a significant role. Even though the second electron does not interact with the external field, it is still left in an excited state after ionization, although with less energy than in the fully interacting case. Thus collective response can be generated by only forcing one electron. However, the ionization of the first electron takes place more slowly than it does when both electrons are forced, indicating that in the collective regime the external force on the second electron plays a significant role in ionizing the first, increasing the ionization rate by 50%.

We now consider the response of the beryllium atom to the same monochromatic fields, Eq. (13). In this model, as well as the actual atom, neutral Be consists of two shells of electrons, allowing us to study processes involving more than one shell. In our model, the outer electrons have an energy [as defined by Eq. (6)] of -58.4 eV and are located 0.5 Å from the nucleus, while the inner two electrons have an energy of -163.4 eV and are located 0.1 Å from the nucleus. These values are to be compared with the helium atom in which each of the two electrons has an energy of -41.7 eV and a radius of 0.3 Å, values roughly comparable with the outer shell of beryllium. For a given applied field configuration, we ran simulations for 10 different initial atomic configurations to a maximum time of 5 fs.

We analyze the ionization of the outer shell of beryllium and compare it with the ionization of helium. Any significant differences are likely due to the presence of the inner shell of beryllium, which affects the outer shell in two ways. First, the static core changes the potential in which the outer electrons move and Pauli blocks these electrons away from the center of the nucleus. Second, the core can dynamically interact with the valence shell and the applied field thereby transferring energy to and from the valence shell.

We analyze the ionization as we did for helium. Table V shows the number of electrons ionized and the average ionization times. Due to the tight binding of the core, no more than two electrons are ionized in the time period studied (5 fs). Table VI shows the energy of the second valence shell electron when the first electron is ionized (E_1), the total energy of the core electrons when the second electron ionizes (E_2), and the fractions of energy applied by the field in ionizing each of the first two electrons (ε_1 and ε_2, respectively). If the singly ionized atom is left in the ground state, then $E_1 = -67.5$ eV and if the double ionized atom is left in the ground state, then $E_2 = -382.9$ eV. Representative results are plotted in Figs. 5 and 6 that show the trends with increasing field strength and frequency.

The variation of the ionizing dynamics of the outer shell with field (Fig. 5) shows many of the same features as helium. The number ionized, ionization rates ε_1 and

TABLE V. Beryllium ionization times. Same notation as in Table II is used.

ω_0 (fs^{-1})	F_0 (V/Å)	N	t_1 (fs)	t_2 (fs)	t_2/t_1
5	10	1.2	2.6	3.9	1.5
	20	2.0	1.0	1.6	1.6
	30	2.0	0.72	0.99	1.4
	40	2.0	0.41	0.55	1.3
	50	2.0	0.39	0.47	1.2
10	10	1.7	2.7	3.5	1.3
	20	2.0	0.82	1.1	1.3
	30	2.0	0.61	0.87	1.4
	40	2.0	0.52	0.60	1.2
	50	2.0	0.49	0.52	1.1
20	10	1.7	1.9	3.6	1.9
	20	2.0	0.90	1.4	1.6
	30	2.0	0.75	1.0	1.3
	40	2.0	0.55	0.99	1.8
	50	2.0	0.50	0.70	1.4
40	10	0.8			
	20	1.5	1.5	2.7	1.8
	30	2.0	0.96	2.0	2.1
	40	2.0	0.67	1.5	2.2
	50	2.0	0.58	1.3	2.2
60	10	0.3			
	20	1.3	1.8	3.2	1.8
	30	1.8	0.86	2..5	2.9
	40	2.0	0.69	1.4	2.0
	50	2.0	0.76	1.8	2.4

TABLE VI. Beryllium ionization energies. E_1 is the average energy of the second outer-shell electron when the first is ionized, E_2 is the average energy of the core when the second electron is ionized, and ε_i is the average value of epsilon for the ith electron at the moment of ionization.

ω_0 (fs^{-1})	F_0 (V/Å)	E_1 (eV)	E_2 (eV)	ε_1	ε_2
5	10	-57.9		0.54	
	20	-44.8	-373	0.56	1.20
	30	-39.0	-374	0.73	1.04
	40	-30.6	-375	0.77	0.96
	50	-12.3	-375	0.77	0.97
10	10	-33.4	-369	0.69	0.85
	20	-40.2	-367	0.68	0.98
	30	-22.7	-373	0.77	0.89
	40	18.4	-373	0.82	0.96
	50	65.3	-374	0.85	1.00
20	10	-28.1	-364	0.59	0.93
	20	-20.0	-366	0.64	0.84
	30	-7.0	-362	0.81	0.90
	40	-12.9	-368	0.75	0.85
	50	52.2	-367	0.80	0.85
40	10	-19.6	-375	0.60	0.67
	20	-15.4	356	0.64	0.85
	30	-21.7	-344	0.79	0.84
	40	-8.7	-350	0.61	0.82
	50	-20.2	-341	0.62	0.95
5060	30	-22.5		0.54	
	30	-28.5	-344	0.51	0.70
	40	-13.8	-355	0.64	0.66
	50	-16.2	-317	0.57	0.72

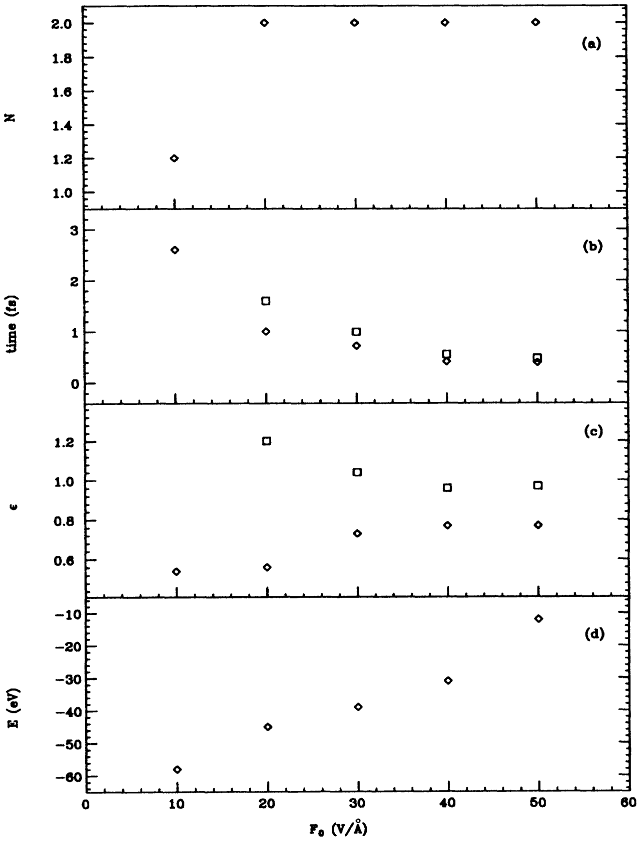

FIG. 5. Ionization properties of beryllium as a function of F_0 for $\omega=5$ fs^{-1}: (a) average number of electrons (N) ionized in 5 fs, (b) average time of ionization of the first electron (diamonds) and the second electron (squares), (c) ε of the first electron to ionize (diamonds) and the second electron to ionize (squares), and (d) total energy of the core (E) at the time the second electron ionizes.

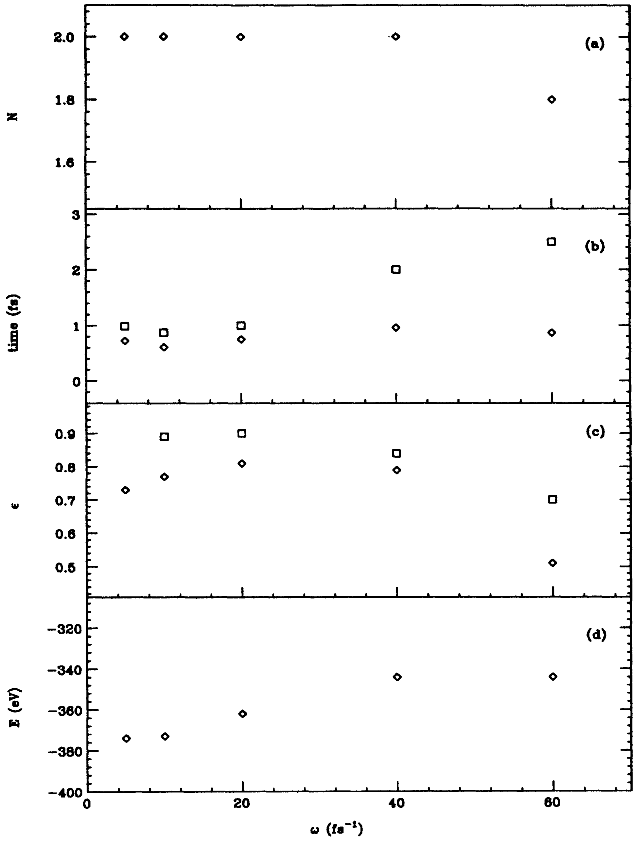

FIG. 6. Ionization properties of beryllium as a function of ω for $F_0=30$ V/Å: (a) average number of electrons (N) ionized in 5 fs, (b) average time of ionization of the first electron (diamonds) and the second electron (squares), (c) ε of the first electron to ionize (diamonds) and the second electron to ionize (squares), and (d) total energy of the core (E) at the time the second electron ionizes.

E_1, all increase, and the ratio between the times of the two electrons tends to decrease with the field. Note that the fields and frequencies required to give a similar excitation are less for Be than He. One difference is in the behavior of ε_2. For ionization in which the remaining electrons are treated as static, one expects $\varepsilon_1=0.4$ and $\varepsilon_2=0.7$. Although ε_1 tends towards this value at low fields and frequencies as expected for sequential ionization, ε_2 tends towards much larger values at low frequencies and fields. This could be caused by a coupling between the core and the second electron which drains energy from the second electron.

The fact that the core is playing a large role is seen by examining the variation of the ionization with frequency (Fig. 6). The number of particles ionized, the time at which the second electron is ionized, and the energy of the core all increase at higher frequencies. This excitation of the core acts to inhibit the ionization of the second electron, although it appears to have little effect on the ionization of the first electron. Also note that ε diminishes at high frequencies for both electrons, indicating that as the core excites the external field needs to do less work on the outer electrons to ionize them.

In Table VII, we show ionization data when only one outer-shell electron is forced. This exhibits the same behavior as in the helium case. At low frequencies, the ionization times are about the same as in the fully interacting case and the atom is left in the ground state when the forced electron ionizes. At higher frequencies, the ionization of the forced electron is delayed and leaves the atom in an excited state. Thus the effect of collective ionization is to enhance the ionization of the first electron relative to the single-electron rate. We also tried forcing one inner-shell electron and found that one could transfer energy to the outer-shell electrons this way. At $\omega=60$ fs^{-1} and $F_0=50$ V/Å, we find that one out of ten runs results in an outer-shell electron ionizing.

Hence, Be and He both exhibit collective ionization for large frequencies and fields. These excitations occur in a regime in which the ionization rate is of order the fre-

TABLE VII. Beryllium ionization: One interacting electron. Same notation as Table V is used.

ω_0 (fs^{-1})	F_0 (V/Å)	t_1 (fs)	E_1 (eV)	R
5	10	3.0	−61	1.2
	20	1.0	−59	1.0
	30	0.61	−63	0.8
	40	0.45	−61	1.1
	50	0.41	−64	1.1
40	20	2.2[a]	−32	1.2[a]
	30	0.95	−54	1.0
	40	1.1	−44	1.6
	50	0.873	−46	1.4

[a] Only seven out of ten electrons ionized in 5 fs so these are lower bounds to the actual values.

quency of the wave and so the ionization will be very sensitive to the shape of the wave. Experimentally, one exposes the atoms to pulsed waves. Since the ionization rates in the strong fields considered in this paper are in the femtosecond range, one must use pulses with rise times of this order in order to expose the atom to these strong fields. We examine the effect of pulse shape by exposing the model atoms to Gaussian pulses

$$\mathbf{F}_0(t)=\hat{\mathbf{z}}F_0\exp\left[-\frac{(t-3\sigma)^2}{\sigma^2}\right]\sin(\omega t)\ , \qquad (15)$$

and observe show the ionization varies with pulse width.

In Fig. 7 we present results for the response of helium obtained by integrating the equations of motion from $t=0$ to $t=6\sigma$ for $F_0=60$ V/Å and $\omega=10\ \mathrm{fs}^{-1}$. When exposed to monochromatic fields of this frequency we have previously observed collective ionization at the peak field strength and sequentially ionization at slightly lower field strengths.

In Fig. 7(c) the values of E correspond to collective ionization for femtosecond pulse widths and sequential ionization for pulse widths that are smaller or larger. This can be understood because the ionization takes place at the peak field of the pulse [Fig. 7(b)] and also the finite pulse width introduces high-frequency components into the applied field, thus encouraging collective ionization. If the pulse length becomes too small, then there is not enough time for multiple ionization to occur and what ionization does occur is the energetically easiest case in which the second electron is left in the ground state. Notice that the transition from sequential to collective ionization is dramatic and is reflected in a small enhancement in the average amount of ionization [Fig. 7(a)]. Here ionization refers to the number of electrons with positive energy after the pulse has diminished, so there is no ambiguity in determining ionization as there is when the applied field is on.

Examining the ionization times in units of 3σ [Fig. 7(b)], we see that the ionization occurs earlier along the pulse, and hence in weaker fields, as the pulse width is increased. Thus, we would expect that when the pulse width is increased significantly beyond the femtosecond range the ionization will continue to be sequential. This corresponds to the current experimental situation in which picosecond pulses are used and only sequential ionization is observed.[9,10]

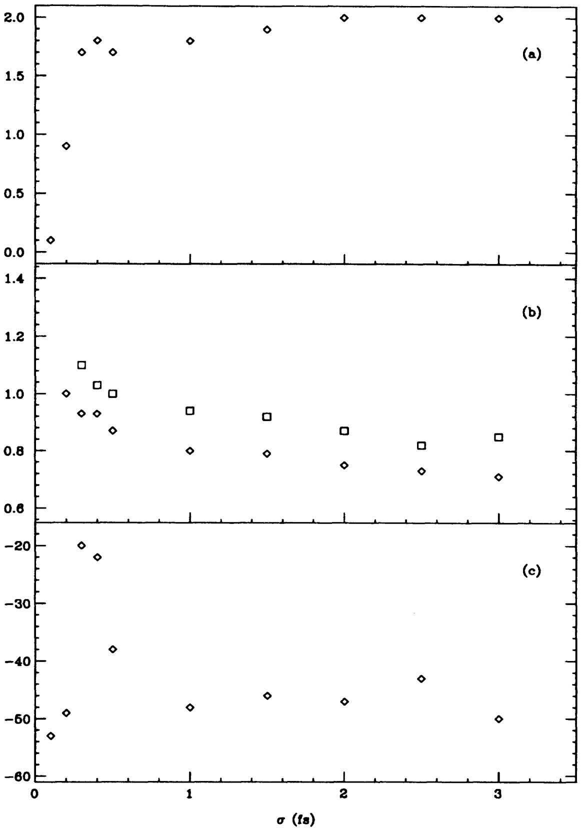

FIG. 7. Ionization properties of helium as a function of pulse length σ as defined by Eq. (15) with $\omega=10\ \mathrm{fs}^{-1}$ and $F_0=60$ V/Å: (a) average number of electrons (N) ionized by the pulse, (b) average time of ionization in units of 3σ (so that a value of 1.0 corresponds to the peak of the pulse) for the first electron (diamonds) and the second electron (squares), and (c) energy of the second electron (E) at the time the first electron ionizes.

IV. CONCLUSIONS

We have used molecular-dynamics simulations to study the double ionization of helium and beryllium. These models exhibit sequential ionization in low fields, changing to collective ionization as the strength of the field is increased, with less field required at higher frequencies. Sequential ionization is characterized by one electron ionizing, leaving the rest of the atom in its ground state. The interaction of the other electrons with the applied field plays little role in this process. Collective ionization is characterized by the atom being left in an excited state when an electron ionizes. It is observed to occur in the regime in which the ionization time is comparable to the periods of the applied field. In this case, the interaction of the field with all the electrons plays a crucial role in the response of the atom. We have observed that collective effects within a shell tend to enhance the ionization rate. For Be, we also observed coupling between the inner and outer shells. At high frequencies the inner shell becomes excited, decreasing the ionization of the outer shell.

Finally, to generate experimentally collective ionization, one will have to use fast pulses in the femtosecond range (or very strong pulses of longer duration). We have seen that such short pulses can enhance the likelihood of collective ionization when the pulse length is of order the ionization rate. This is due to the generation of high-frequency components in the short pulse and the increased likelihood for the atom to see the peak of the pulse.

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