# Lithium ionization by an intense laser field using classical ensemble simulation

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The classical ensemble method is applied to study ionization processes of a one-dimensional model lithium atom interacting with an intense laser pulse. The motion of electrons is described by the classical Hamiltonian canonical system of equations. The ratio of double-to-single ionization with the increasing laser intensity is calculated and explained in terms of the energy distribution of electrons. The triple ionization of lithium is also investigated and the primary triple ionization path is found. Our results show a clear transition from nonsequential to sequential double ionization as the intensity increases, which is in agreement with the quantum calculation.

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## I. INTRODUCTION

Extensive research into the nature of multiphoton ionization processes has improved theoretical and experimental capabilities over the last decade [1–4]. Many experimental improvements have been derived from the higher brightness of the third-generation synchrotron light source [5]. The study of behaviors of multielectron atoms in strong laser fields is a subject of increasing interest [6]. Due to advances in capabilities of experimental measurements and in new technologies, dynamics of simple systems has been studied thoroughly both theoretically and experimentally, such as nonsequential double ionization [7]. However, the study of more complex systems is needed in many circumstances. Experimental measurements of triple photoionization have recently been carried out for Li at photon energies well beyond the threshold for three-electron escape [8]. There have been several theoretical studies of these multiple ionization processes in Li. For example, van der Hart and Greene [9] investigated the double and triple photoionization of Li using an independent-electron model in the high-energy limit. Ruiz *et al.* [10] studied the interaction of a 1D model lithium with a strong laser field quantum mechanically. Yan [11] also calculated the double photoionization of Li and $Be^+$ at the high-energy limits using Hylleraas coordinates.

In principle, the treatment of laser-matter interaction involving an atom or molecule needs full quantum theory. However, it has been demonstrated that a classical treatment is valid for the case of superintense and ultrashort laser pulses, where the Planck constant $\hbar$ is negligible compared with the system's action [12]. The classical method can be used to describe a high-order ionization process and can include correlations among electrons. Recently, classical simulations of $H_2^+$ and $H_2$ have also been performed [13,14] which can reproduce qualitative features of the corresponding quantum mechanical calculations. Particularly, Eberly and co-workers [15–18] have done a series of studies on nonsequential double ionization (NSDI) and nonsequential triple ionization (NSTI) using classical simulation.

The purpose of this paper is to investigate ionization dynamics for a one-dimensional (1D) model lithium interacting with an intense laser pulse using the method of classical ensemble simulation. We will calculate the ratio of double-to-single ionization and study its dependence of laser intensity at different wavelengths. We will demonstrate how double ionization shifts from nonsequential to sequential as the intensity increases. We will also look into the possible paths that lead to a triple ionization.

## II. CLASSICAL ENSEMBLE METHOD

From a classical point of view, lithium can be considered as a dynamic system which consists of four classical particles: one nucleus and three electrons. When the external field is absent, this four-body system is stable. When a laser field is applied, the motion of each particle depends not only on the laser field but also on the Coulomb field due to the other three particles.

Given the present computing resources that we can access, we only treat the 1D model lithium in this paper. For the 1D model lithium, since the electric field is linearly polarized, the motion of the electrons and nucleus is along the direction of electric field. We take the nucleus as the origin of our coordinate system. Thus the classical Hamiltonian of lithium in an intense laser field can be given by (atomic units are used throughout unless otherwise stated)

$$H(x_1,p_1;x_2,p_2;x_3,p_3;t) = \mathcal{T}(p) + \mathcal{V}(q,t), \tag{1}$$

where the kinetic energy $\mathcal{T}$ and the potential energy $\mathcal{V}$ are given, respectively, by

$$\mathcal{T}(p) = \frac{p_1^2}{2} + \frac{p_2^2}{2} + \frac{p_3^2}{2}, \tag{2}$$

$$\mathcal{V}(q,t) = -\sum_{i=1}^{3} \frac{3}{\sqrt{\alpha^2 + x_i^2}} + \sum_{\substack{i,j=1 \\ i>j}}^{3} \frac{1}{\sqrt{\beta^2 + x_{ij}^2}} + E_{\mathrm{ex}}(t)\sum_{i=1}^{3} x_i. \tag{3}$$

In the above equations, $q=(x_1,x_2,x_3)$ stands for the positions of the three electrons, $p=(p_1,p_2,p_3)$ is for their correspond-

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ing conjugate momenta, $x_{ij}=|x_i-x_j|$, $E_{ex}(t)$ is the laser field, and $\alpha$ and $\beta$ are the soften parameters, which are generally used to remove the singularity in 1D Coulomb problems. In this work, we choose $\alpha=\beta=0.7$ a.u. The canonical system of equations for Li is

$$\frac{dp}{dt}=-\frac{\partial \mathcal{V}(q,t)}{\partial q}, \tag{4}$$

$$\frac{dq}{dt}=\frac{\partial \mathcal{T}(p)}{\partial p}. \tag{5}$$

We assume that the atom is in its ground state $^2S_{1/2}$ and thus the initial energy is the energy of the ground state of Li which is approximately equal to −7.33 a.u. or −199.44 eV [10]. We choose a set of initial stable states $\{x_i(0),p_i(0)\}_{i=1}^3$ by the method of classical mechanical ensemble [12–14] and solve the above canonical equations numerically in order to obtain the time evolutions of the electron positions and the corresponding momenta $\{x_i(t),p_i(t)\}_{i=1}^3$. The Hamiltonian system (1) is a separable system in the sense that $q$ and $p$ are contained separately [14,19] in $\mathcal{V}(q,t)$ and $\mathcal{T}(p)$. Also, the Hamiltonian is an explicit function of the time variable $t$. We may use an explicit symplectic scheme [14] to solve these equations in order to obtain the classical trajectories for the electrons in the intense laser field. For the field-free case, the energy can be preserved numerically in time by using the symplectic method, even for a long-time evolution. Thus, we can prepare the initial conditions for the atom-laser interacting system by first solving the field-free Hamiltonian using the symplectic method and then pick up a large number of points randomly along the field-free trajectory. These points consist of a microcanonical ensemble and will be served as the initial conditions for the full Hamiltonian (1).

The ionization yields are computed by partitioning the Hilbert space as follows [10]:

$$\mathrm{Li}+\mathrm{laser}\rightarrow \mathrm{Li},\quad |x_i|<15,\quad |x_j|<15,\quad |x_k|<15, \tag{6}$$

$$\mathrm{Li}+\mathrm{laser}\rightarrow \mathrm{Li}^+ + e,\quad |x_i|<15,\quad |x_j|<15,\quad |x_k|>15, \tag{7}$$

$$\mathrm{Li}+\mathrm{laser}\rightarrow \mathrm{Li}^{2+} + 2e,\quad |x_i|<15,\quad |x_j|>15,\quad |x_k|>15, \tag{8}$$

$$\mathrm{Li}+\mathrm{laser}\rightarrow \mathrm{Li}^{3+} + 3e,\quad |x_i|>15,\quad |x_j|>15,\quad |x_k|>15, \tag{9}$$

where $i,j,k=1,2,3$. These four processes are defined as the survival, the single ionization, the double ionization, and the triple ionization, respectively.

There are several definitions for the one-particle energy of the electron in a many-electron atomic system in the literature [16–18,20]. Each definition depends on the approach used in the calculation. In this work, we define the one-particle energies for each electron during the laser pulse as follows:

$$\epsilon_1=\frac{p_1^2}{2}-\frac{3}{\sqrt{\alpha^2+x_1^2}}+\frac{1}{\sqrt{\beta^2+x_{12}^2}}+\frac{1}{\sqrt{\beta^2+x_{13}^2}}, \tag{10}$$

$$\epsilon_2=\frac{p_2^2}{2}-\frac{3}{\sqrt{\alpha^2+x_2^2}}+\frac{1}{\sqrt{\beta^2+x_{21}^2}}+\frac{1}{\sqrt{\beta^2+x_{23}^2}}, \tag{11}$$

$$\epsilon_3=\frac{p_3^2}{2}-\frac{3}{\sqrt{\alpha^2+x_3^2}}+\frac{1}{\sqrt{\beta^2+x_{31}^2}}+\frac{1}{\sqrt{\beta^2+x_{32}^2}}. \tag{12}$$

This definition, which is similar to the one used in Refs. [18,20] for the case of helium, includes the kinetic energy of the electron and all the electron interaction energies with the remaining particles. The $i$th electron is considered to be ionized when the condition of $\epsilon_i>0$ is satisfied.

Suppose $T$ is the pulse duration of the laser field. Let the time step be $\tau=T/Z$ with $Z$ being a sufficiently large positive integer and denote $t_k=k\tau$, $k=0,1,2,\ldots,Z$. We further let $\Omega$ be the number of initial conditions. We can then obtain $\Omega$ classical trajectories for the lithium-laser system by using the symplectic method. For each time $t_k$, we compute the numbers of classical trajectories for the processes of the survival $S_{sur}$, the single ionization $S_{SI}$, the double ionization $S_{DI}$, and the triple ionization $S_{TI}$ according to the criteria Eqs. (6)–(9), respectively. Then we can calculate the corresponding probabilities for these processes at each time $t_k$ according to

$$P_{sur}=\frac{S_{sur}}{\Omega},\quad P_{SI}=\frac{S_{SI}}{\Omega},\quad P_{DI}=\frac{S_{DI}}{\Omega},\quad P_{TI}=\frac{S_{TI}}{\Omega}. \tag{13}$$

## III. RESULTS AND DISCUSSION

In the present work, we first choose an ultrashort laser pulse $E(t)=E_0f(t)\sin(\omega_0 t)$, with the frequency $\omega_0=1.169$ a.u. (39 nm in wavelength) and the pulse shape

$$f(t)=\begin{cases}\sin^2\left(\dfrac{\pi t}{T}\right), & 0<t<T\\ 0, & \text{otherwise},\end{cases} \tag{14}$$

where $T=20T_0$ is the pulse duration with $T_0=2\pi/\omega_0$ being the period of the pulse, and $E_0$ is the peak intensity. In this work, we utilize a microcanonical ensemble which consists of $5\times10^5$ three-electron "trajectories."

Figure 1 shows the single, double, and triple ionization probabilities calculated by using the classical ensemble method. We can see that the single ionization probability increases with the intensity. When the intensity is further increased to the saturation point, the single ionization starts to decrease; this is because the sequential production of double ionization begins to deplete the single ionization ion yield. The double ionization also increases with the intensity, but the probability is much smaller than the single ionization one at first, then it increases with the intensity rapidly and finally exceeds the single ionization probability. The triple ionization starts to turn on at a much higher intensity and follows a similar tendency as the double ionization. Furthermore, the triple ionization probability is even much smaller

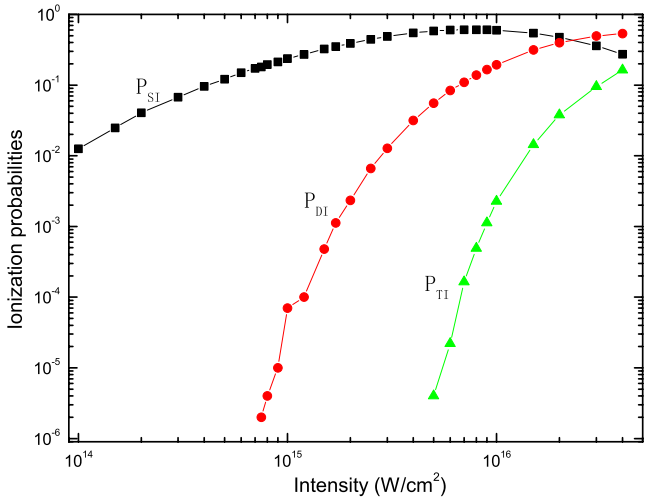


FIG. 1. (Color online) Single ($P_{SI}$), double ($P_{DI}$), and triple ($P_{TI}$) ionization probabilities at different intensities. The laser wavelength is 39 nm.

than the double ionization probability. Finally, we cannot observe the NSTI for the wavelength we have used. If the NSTI is important, then we should see the "knee," as pointed out by Ho and Eberly [15]. We think the reason for this is that the wavelength used is so short that the sequential triple ionization (STI) is predominant and the NSTI is seldom to occur.

In general, the theory of Ho and Eberly [15], established for a model atom, may still suit for a real atom. In Fig. 2 we have used a 20 fs laser pulse of 780 nm in wavelength as used by Ho and Eberly and found the NSTI phenomenon for lithium, where an obvious "knee" is presented on the double- and triple-ionization probability curves. Our results show a similar feature as predicted by Ho and Eberly [15]. Moreover, comparing to Fig. 1, we can see that the NSDI and NSTI are more likely to occur for a low-frequency laser pulse.

The intensity dependence of the ratio of double-to-single ionization $Li^{2+}/Li^{+}$ can be used to indicate if a double ionization is nonsequential or not. As has been well established for helium, if this ratio is only weakly intensity dependent, the precursor to $Li^{2+}$ is not $Li^{+}$ and thus the ionization involves a nonsequential process [21]. Figure 3 shows the double-to-single ionization ratio of Li as a function of the intensity at the laser wavelength of 39 and 780 nm, respectively, computed at about the end of the laser pulse. We can see that the nonsequential ionization is not obvious at the laser wavelength of 39 nm, whereas a flattening of the ratio curves (the "knee" structure) is obvious at the laser wavelength of 780 nm, which may indicate NSDI. The change in the slope of this ratio as the intensity increases in Fig. 3(b) is the signature of the transition from nonsequential to sequential double ionization [10]. Hence, Fig. 3(b) shows that the double ionization of lithium also shifts from nonsequential to sequential as the intensity increases, which is similar to the results of Ruiz *et al.* using the *ab initio* quantum method [10].

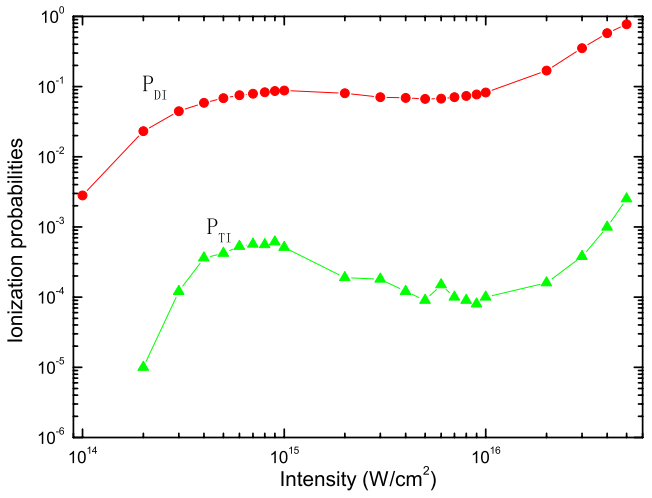


FIG. 2. (Color online) Double ($P_{DI}$) and triple ($P_{TI}$) ionization probabilities at different intensities. The laser wavelength is 780 nm.

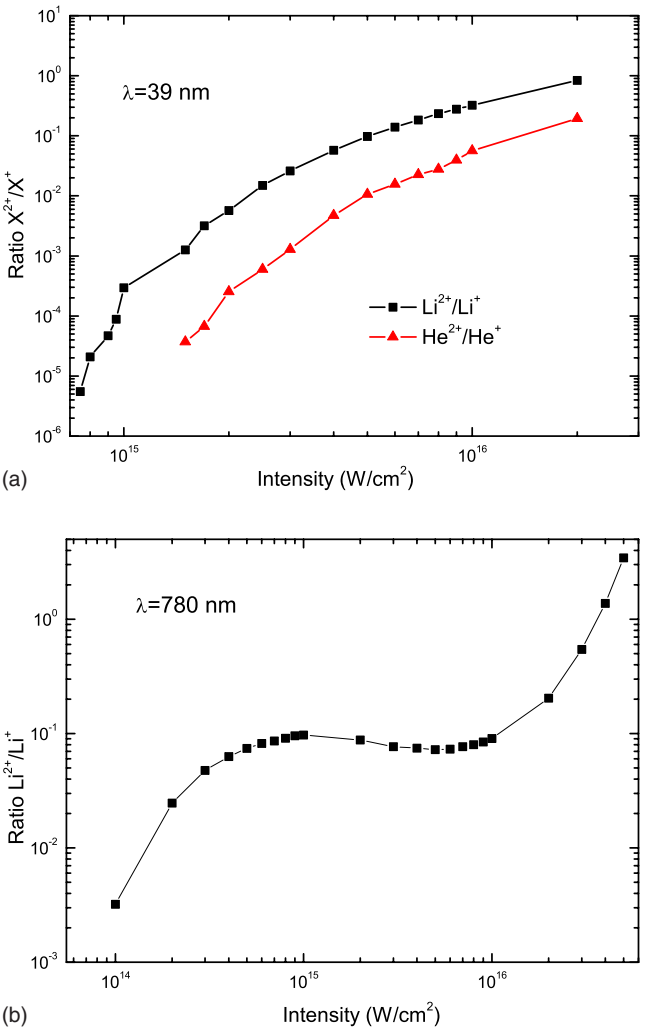


FIG. 3. (Color online) Ratio of $Li^{2+}$ to $Li^{+}$ ionization at different intensities. (a) The laser wavelength is 39 nm. (b) The laser wavelength is 780 nm. For comparison, we also show the intensity dependence of $He^{2+}$ to $He^{+}$ ionization ratio in (a).

It may be probable that there is a physically different process in a three-electron system not existing in a two-

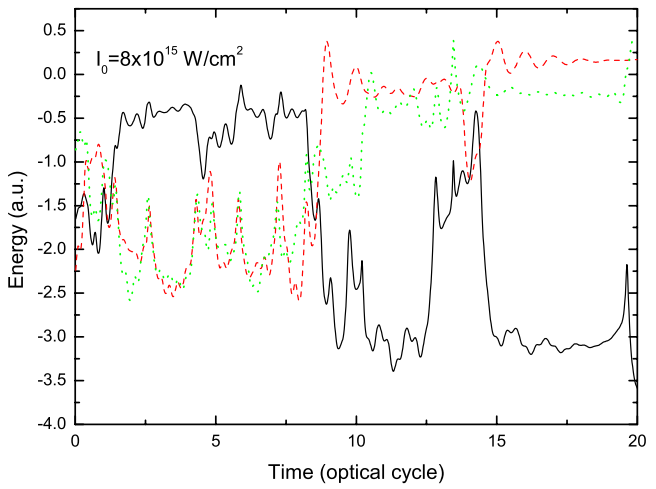


FIG. 4. (Color online) An example for a classical double ionization of Li via the decay of an autoionizing state in terms of the one-particle energies. The solid, dashed, and dotted lines track the energies of three electrons, respectively.

electron system. In addition to the triple ionization process, there may exist indirect ionization channels in double ionization [9]. For comparison, we also show the ratio of $He^{2+}$ to $He^+$ ionization yield at different intensities in Fig. 3(a) and find that the ratio of $Li^{2+}$ to $Li^+$ is much larger than that of $He^{2+}$ to $He^+$ under the same conditions. We think it may be due to the fact that the initial ionization of Li can produce a highly excited autoionizing state of $Li^+$. The autoionizing state of the ion could easily decay in an electric field by emitting one electron, causing a double ionization. This could be a potentially important channel to the double ionization continuum, which is absent from helium. Autoionization is a correlation effect which appears in highly excited states in a multielectron system. Its spectrum is neither completely discrete nor completely continuous. In order to illuminate this mechanism, an example is given in Fig. 4 for a classical double ionization of Li via the decay of an autoionizing state in terms of the one-particle energy. We can see that an electron (the dotted line) is in an excited state for a while with high energy and eventually is ejected. We find a series of autoionizing states of $Li^+$ calculated by Bhatia [22]. From a classical point of view, if the one-particle energy of an electron is equal to the energy of one of these autoionizing states at time $t$ so that this electron is emitted to the continuum, then we can say that the electron is ionized via the autoionization pathway. We use an ensemble of 100 000 "trajectories" and take $^1S^e$ state (70.5502 eV) as an example. The number of trajectories of the double ionization via this autoionization pathway at the laser intensity $I_0=8\times10^{15}$ W/cm$^2$ is 431. We also calculate the numbers of double ionization trajectories via the $^1P^o$ (71.3449 eV) and the $^3D^e$ (84.2145 eV) autoionization states and find that they are 579 and 717, respectively, and so on. Our calculations can be served as a rough approximation to the real situation. We may appeal to the full quantum method to obtain more accurate results. However, our results may explain the existence of the autoionization channels qualitatively.

Generally speaking, there are three different mechanisms which contribute to the double-photoionization (DPI) process [23–25]: the shake-off mechanism, the two-step mechanism, and the ground-state correlation. Thus, compared to He, the additional 2$s$ electron in Li not only affects the overall strength of the interaction but also affects the different mechanisms which are responsible for ionization.

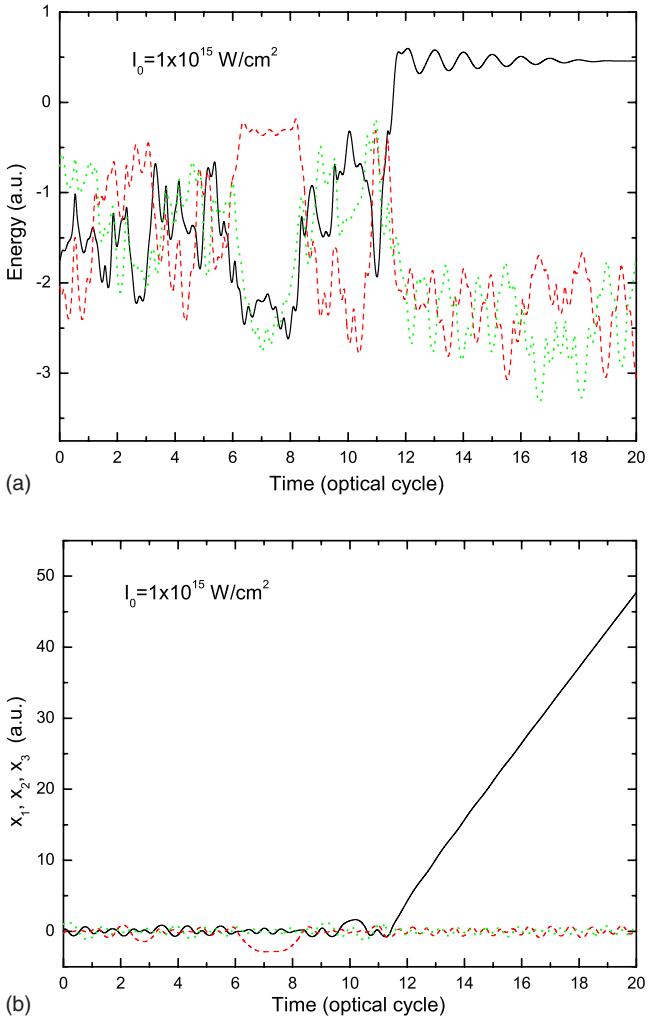


FIG. 5. (Color online) An example for classical single ionization in terms of the one-particle energies and the electron positions. (a) The solid, dashed, and dotted lines track the energies of three electrons, respectively. (b) The solid, dashed, and dotted lines track positions of three electrons, respectively.

In the following, we will calculate the energy and the corresponding position distributions of the three electrons in a high-frequency laser field (39 nm in wavelength) and explain which ionization process they may stand for. Figure 5 shows an example for a single ionization at $1\times10^{15}$ W/cm$^2$. We can see from the energy distribution of Fig. 5(a) that the ionized electron (the solid line) gains energy from the electric field and the remaining two electrons still stay in a bound state with lower energy. One observes from the electron positions of Fig. 5(b) that the electron which leaves first moves in the same direction as the other electrons. Then, this electron (the solid line) is ionized, whereas the remaining two electrons move in the opposite directions periodically, keeping the $Li^+$ ion stable.

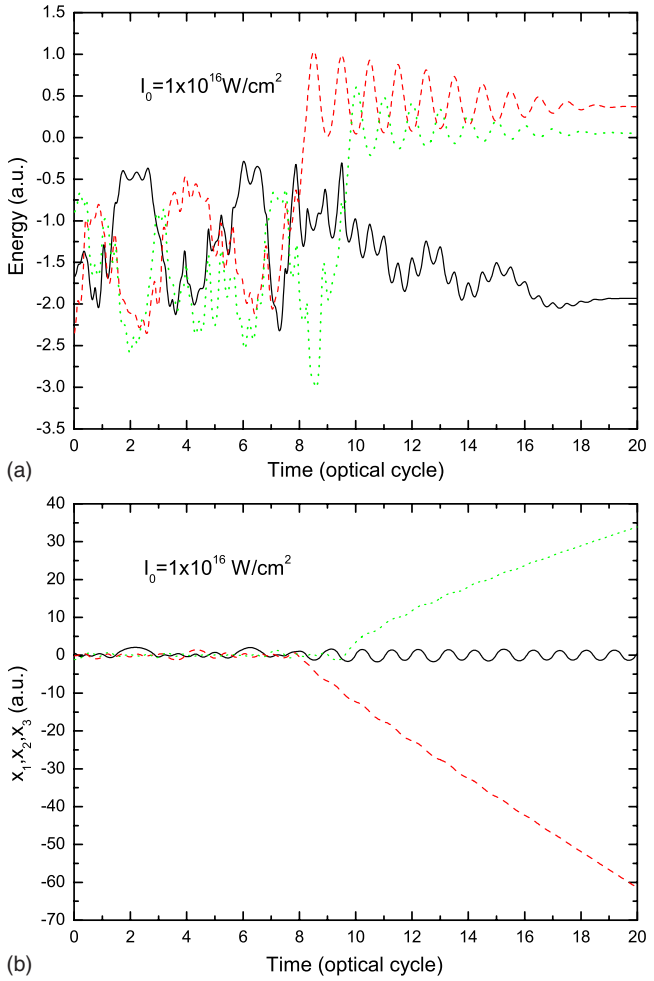


FIG. 6. (Color online) An example for classical sequential double ionization in terms of the one-particle energies and the electron positions. (a) The solid, dashed, and dotted lines track the energies of three electrons, respectively. (b) The solid, dashed, and dotted lines track positions of three electrons, respectively.

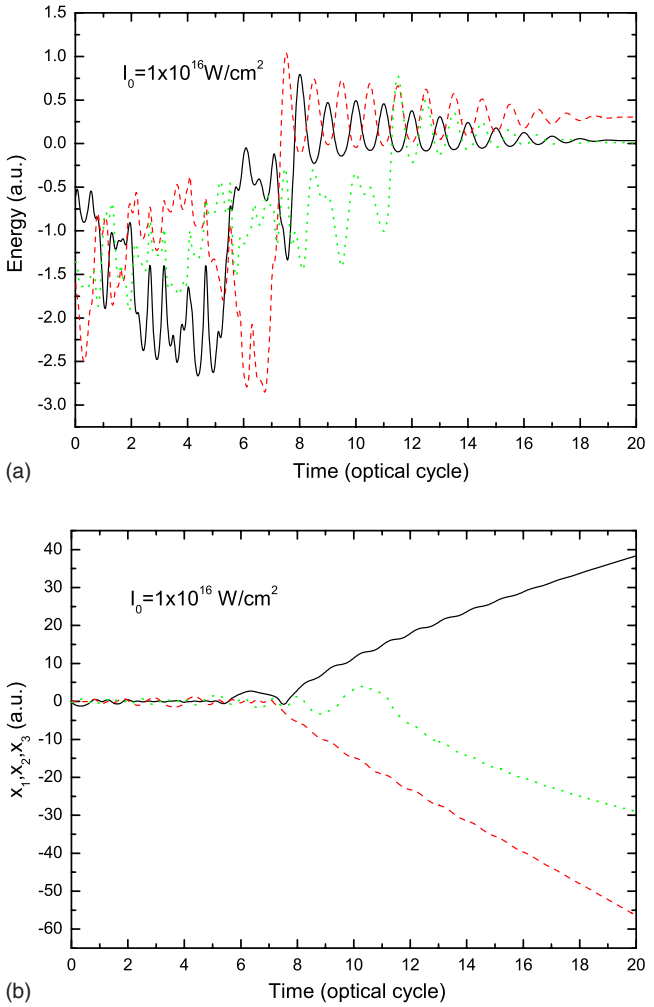


FIG. 7. (Color online) An example for classical sequential triple ionization in terms of the one-particle energies and the electron positions. (a) The solid, dashed, and dotted lines track the energies of three electrons, respectively. (b) The solid, dashed, and dotted lines track positions of three electrons, respectively.

In order to describe it qualitatively, we have done a statistic. Here we utilize a microcanonical ensemble which consists of $10^5$ three-electron "trajectories." The total number of single ionization "trajectories" at $1\times10^{15}$ W/cm$^2$ is 23 782; whereas the number of single ionization trajectories similar to Fig. 5(a) is 22 670, which is about 95.32% of the total number of single ionization "trajectories." Thus we believe that Fig. 5(a) is more realistic. We also have calculated this percentage at other laser intensities and the results also show the same tendency.

Figure 6 presents an example for double ionization at $1\times10^{16}$ W/cm$^2$. At this field strength, the sequential double ionization process is more probable than the NSDI one. We can see from the energy distribution of Fig. 6(a) that one electron (the dashed line) is ionized first followed by the second electron (the dotted line), which means that a sequential double ionization occurs. They also exhibit jitter oscillations, characteristic of free electrons. One can further see from the electron positions of Fig. 6(b) that, after one electron (the dashed line) is ionized, the second electron (the dotted line) is ionized two cycles later, which is in correspondence with the energy distribution of Fig. 6(a). Then the remaining electron moves regularly, forming a stable $Li^{2+}$ ion.

We also calculated the double ionization probability at $I_0=1\times10^{16}$ W/cm$^2$ with an ensemble of 100 000 "trajectories," and then singled out the sequential double ionization trajectories. We found that the total number of double ionization trajectories is 19 371 and the number of sequential double ionization is 19 053, which is about 98.36% of the total double ionization. Thus, the sequential double ionization is the predominant process under this condition.

The triple ionization process involves two steps [26]: after single photon absorption by one electron, redistribution of the energy due to the electron correlations takes place, resulting in a three-electron escape to the continuum. In our calculations, there are at least two possible paths responsible for the triple ionization. First, three electrons are ionized in a sequential way. A typical example is displayed in Fig. 7, where the one-particle energies and the corresponding electron positions are plotted. We can clearly see from Figs. 7(a)

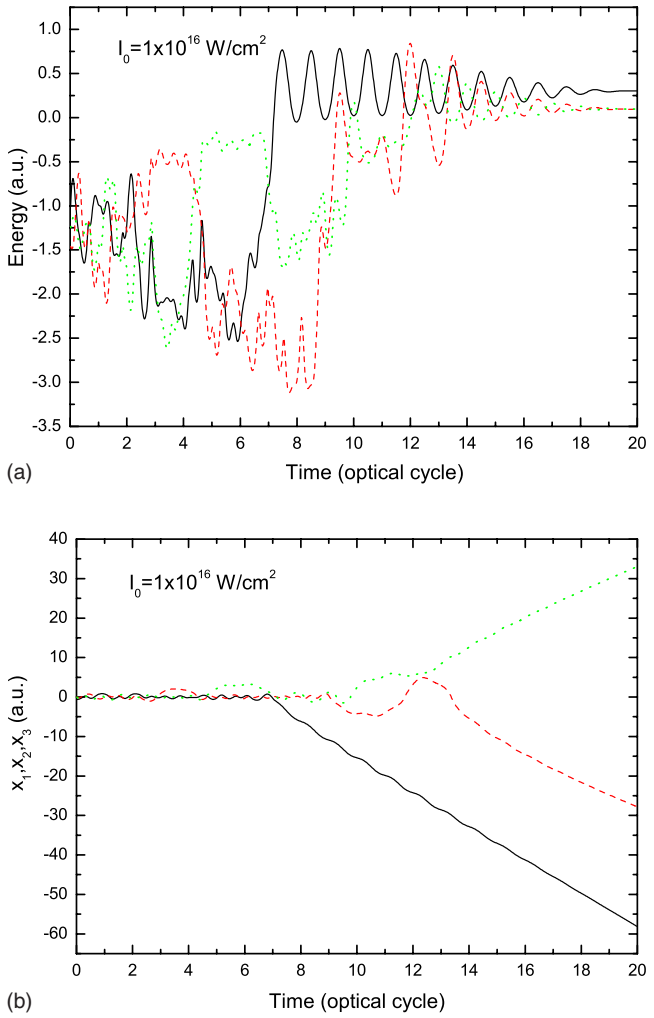


FIG. 8. (Color online) An example for classical nonsequential triple ionization in terms of the one-particle energies and the electron positions. (a) The solid, dashed, and dotted lines track the energies of three electrons, respectively. (b) The solid, dashed, and dotted lines track positions of three electrons, respectively.

and 7(b) that the three electrons are ionized one after another sequentially. Second, one electron tunnels out through the field-induced potential first, followed by the second electron ionization. This second electron then returns back to the atomic core with higher energy and collides with the remaining electron, causing a triple ionization. Such an example is displayed in Figs. 8(a) and 8(b). In order to describe it qualitatively, we can single out all of the triple ionization trajectories at the intensity range used. Setting $I_0=1\times10^{16}$ W/cm$^2$ as an example, there are 38 triple ionization trajectories (here the definition of the triple ionization is that the energy of each electron is greater than zero at the end of the pulse) in an ensemble of 100 000 "trajectories." Most of them are sequential triple ionization trajectories; only two trajectories are nonsequential ones which are similar to the case of Figs. 8(a) and 8(b). The triple ionization at other laser intensities is also calculated. Our results show that the NSTI always exists although the number is much smaller than that of STI. In general, if the wavelength is too short, then the quiver energy becomes much smaller than the ionization energy, so that recollisional ionization cannot occur [27]. The maximum kinetic energy is $K_e=3.17U_p$, where the time-averaged quiver energy is given by $U_p=E_0^2/(4\omega_0^2)$. For the case treated here, at $I_0=1\times10^{16}$ W/cm$^2$, one has $K_e\approx0.1657$ a.u., much smaller than the energy for ionizing the third electron. Besides, the probability of triple ionization by collision at high-frequency laser pulse is rare; the probability should be larger in a 1D approximation. Thus we think that the STI is the predominant process and Fig. 8(b) is a rare coincidence in our calculations. It might be an artifact of the restriction to the one spatial dimension. It should be noted that new issues still need to be addressed to reveal the multielectron ejection mechanism in lithium.

## IV. CONCLUSIONS

The classical dynamics of lithium in an intense laser field has been studied by the classical ensemble method. The 1D model lithium has been used, with the bare Coulomb potential replaced by a soften potential to avoid the singularities. We have demonstrated that the double ionization of Li shifts from nonsequential to sequential as the intensity increases at the laser wavelength of 780 nm. We have also demonstrated that the triple ionization channel is turned on at high intensity and the triple ionization increases with the intensity. The primary triple ionization path has been found in terms of the one-electron energy. Under the condition of high laser frequency, the STI is predominant and the NSDI is not obvious. The classical motion of lithium in an intense laser field has revealed some interesting dynamical features. A study of multielectron atoms using the classical method is in progress.

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