Double ionization of He in an intense laser field via a rescattering process

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We investigate the ratio of double to single ionization of He in an intense laser field based on the rescattering model. Folding the rescattering energy spectra with the electron impact inelastic cross sections, we obtain the probability of double ionization due to the nonsequential ionization process. Our results are in reasonable agreement with the experiment [Walker *et al.*, Phys. Rev. Lett. **73**, 1227 (1994)]. Furthermore, we investigate the physical insights of the nonsequential double ionization by analyzing the rescattering energy spectra at different intensities and the contributions from individual returns. This study confirms the reliability of the rescattering energy spectra obtained from *ab initio* calculations. The rescattering information can be used to analyze many other dynamical processes in intense laser-matter interactions, such as molecular imaging.

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I. INTRODUCTION

The precision measurement [1] of double to single ionization of He in an intense laser field has stimulated active theoretical studies [2–7]. The enhanced double ionization cannot be explained by the two sequential single-ionization processes. The laser intensity dependence also rules out the shakeoff process [8], which is due to the relaxation caused by the loss of the first electron. Corkum [9] has proposed a three-step rescattering model, in which the first electron is ejected to the outer regime by tunneling ionization, and then the ejected electron revisits the ion (parent core) when the laser field reverses its direction. The returning electron may knock out the second electron as in the $(e,2e)$ process [10] when it collides with the ion and induces nonsequential double ionization (NSDI). Although the physical picture of the rescattering model is very intuitive, a direct quantum simulation in a full dimension has not been carried out yet. There are several quantum simulations using one-dimensional [11–13] and two-dimensional [3,14] models or a semiclassical model [15]. Such studies provided some physical insight into the NSDI process. Yudin and Ivanov [16,17] have studied NSDI using a semiclassical method based on the rescattering model and Fu *et al.* [18] have investigated the process using a classical Monte Carlo method. From those and many other theoretical works, it is now widely accepted that NSDI is originated from the rescattering process.

The difficulties in direct quantum simulations [19] are due to (1) the full quantum simulation for a two-electron system in a laser field remaining out of the reach of present supercomputers and (2) the fact that the rescattering electron wave packet is only a small portion of the total wave function and it is embedded in a huge background of the ground state. Recently, rescattering information was obtained directly from a quantum simulation [20]. Morishita *et al.* [21] extracted the rescattering information from the above-threshold ionization. In this way they can provide the rescattering electron distribution without the detailed information, such as the contributions from each return. Since the rescattering process plays a central role for understanding many dynamical processes in intense laser-matter interactions, it is preferable to test whether the obtained rescattering information can be used to explain NSDI. We first calculate the rescattering energy spectra at the time when the ejected electron revisits the ion, and then the double ionization (and excitation) probability of the returning electron colliding with the ion using the electron impact inelastic cross sections documented by Yudin and Ivanov [17]. The single ionization (SI) probability is calculated by solving the time-dependent Schrödinger equation with a single-active-electron (SAE) approximation. Furthermore, by analyzing the rescattering energy spectra of different laser intensities at different returns, we explain why the first return has large contributions at the high-intensity and the low-intensity sides. By comparing our results to the experiment, we see that the rescattering information provided by our *ab initio* calculation can be used to study the NSDI process, and naturally it can be also used to analyze other dynamics in intense laser-matter interactions, such as in molecular imaging experiments [22].

II. THEORETICAL METHOD

In the long-pulse experiment, the electron tunnels to the outer region when the laser field reaches its peak value every half optical cycle and we only need to trace the electron wave function of a wave packet created in a half cycle to study the NSDI process. Within the SAE approximation, the corresponding time-dependent wave function is expressed as [20]

$$\Psi(t) = -i \int_{-T/4}^{T/4} e^{-i\int_\tau^t H(t')\,dt'} V_{\rm ext}(\tau) e^{-iH_0\tau} \Phi_0 \, d\tau, \qquad (1)$$

where Φ_0 is the helium ground-state wave function, and $H_0 = -\frac{1}{2}\nabla^2 + V_{\rm eff}(r)$ the atomic Hamiltonian with a model potential $V_{\rm eff}(r)$ [23] (and where atomic units $\hbar = m = e = 1$ are used unless stated otherwise). The electron-laser interaction is given by $V_{\rm ext}(\tau) = -zE_0\cos(\omega\tau)$, where z is the electron z coordinate, E_0 is the laser field strength, $\omega = 2\pi/T$ is the laser frequency, and $H(t) = H_0 + V_{\rm ext}(t)$

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is the total Hamiltonian. We trace the motion of the electron wave packet after it was ionized. Both direct ionization [24] and rescattering information [20] are contained in the time-dependent wave function $\Psi(t)$ of Eq. (1). We extract the rescattering information by projecting the wave function in an inner region onto a continuum state as

$$C_l(E,t) = \int f(r,R_c)Y_{l0}^*(\hat{\mathbf{r}})\psi_l^*(E,r)\Psi(t)\,d\mathbf{r}, \tag{2}$$

where $\psi_l(E,r)$ is the laser-field free atomic continuum wave function normalized in energy for a given partial wave ℓ and energy E [20]. The function $f(r,R_c)$ is introduced to define the size of the inner region R_c with a boundary width Δ as

$$f(r,R_c) = \begin{cases} 1.0 & \text{for} \quad r < R_c, \\ e^{-(r-R_c)^2/\Delta^2} & \text{for} \quad r \geqslant R_c. \end{cases} \tag{3}$$

In the present calculation, R_c and Δ are chosen as 7 and 2 a.u., respectively. Varying R_c from 5 to 10 a.u. and Δ from 1 to 3 a.u. we find that the results are not sensitive to the choice of R_c and Δ. The time propagation of Eq. (1) is carried out using a generalized pseudospectral grid method in the energy representation [25]. The detailed numerical procedure can be found in Ref. [20]. With the coefficients $\{C_l(E,t)\}$, the rescattering energy spectra at time t for partial wave ℓ is given by

$$\frac{dP_\ell(E,t)}{dE} = |C_l(E,t)|^2. \tag{4}$$

Since we trace the motion of the electron wave packet as a function of time t explicitly, the mean electron current density for the nth return can be expressed as

$$I_n(E) = \frac{2}{T}\sum_\ell \int_{nT/2}^{(n+1)T/2} \frac{1}{S_\ell(E)}\frac{dP_\ell(E,t)}{dE}dt, \tag{5}$$

with $S_\ell(E) = \pi b^2$, where $b = \sqrt{(\ell+0.5)^2/(2E)}$ corresponds to the impact parameter [26] in the semiclassical interpretation. The NSDI due to the electron colliding with the ion in the nth return is written as

$$B_n^{2+} = \int \sigma(E)I_n(E)\,dE, \tag{6}$$

where $\sigma(E)$ is the total inelastic cross section of the ion colliding with the returning electron. If $\sigma(E)$ is larger than $S_\ell(E)$, $S_\ell(E)$ is replaced by $\sigma(E)$ since the double ionization probability cannot be larger than the probability of the returning electron. In contrast to the conventional electron beam experiment, the two electrons are correlated in spin functions since the ground state of He is a singlet state. We use the empirical inelastic cross sections documented by Yudin and Ivanov [17]. Once the electron collides with the ion and induces double ionization, it will no longer contribute to the later returns. In Eq. (6), this depletion effect is not taken into account. To take it into account the effect, we define a depletion factor for the $(n+1)$th return as

$$A_{n+1} = \left(1 - \frac{B_n^{2+}}{D_n}\right)A_n \tag{7}$$

with $A_1 = 1$ and

$$D_n = \frac{2}{T}\sum_\ell \int\int_{nT/2}^{(n+1)T/2} \frac{dP_\ell(E,t)}{dE}dt\,dE, \tag{8}$$

which stands for the portion of a single electron passing through the ion during the nth return. Now the NSDI probability due to the nth return can be written as

$$P_n^{2+} = B_n^{2+}A_n, \tag{9}$$

and the total NSDI probability is

$$P^{2+} = \sum_n P_n^{2+}. \tag{10}$$

For single ionization, we calculate the total single ionization probability of He in a pulsed laser by solving the time-dependent Schrödinger equation numerically [25] and then obtain the single ionization probability P^+ within a half optical cycle by dividing the probability by the number of half cycles in the pulse.

III. RESULTS AND DISCUSSION

From the calculated NSDI and SI probabilities, we investigate NSDI of He in an intense laser field. To compare with the experiment [1], we choose a laser wavelength of 780 nm and a laser intensity in the range from $2I_0$ to $20I_0$ with $I_0 = 10^{14}$ W/cm^2. Our results and the experimental data are shown in Fig. 1. The contribution of the depletion effect is about a few percent. Overall, the simulated results are in reasonable agreement with the measurement. To check the convergency, we vary the number of grid points and partial waves as well as the time steps. The parameters used in the present calculations are 1024 grid points in the radial part, 80 partial waves, and 800 time steps per optical cycle. The box size is varied from 150 to 250 a.u., which is several times larger than the quiver distance of a free electron in the laser field.

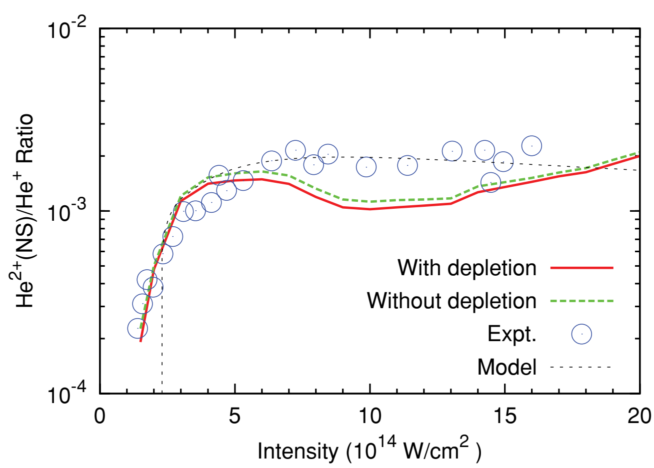

FIG. 1. (Color online) The ratio of double to single ionizations of He in the intense laser fields calculated with the depletion (solid line) and without the depletion (dashed line) effects in comparison with the experimental values [1] (open circles). The prediction of the simple model from Eq. (12) is also presented with an adjustable constant (dotted line).

The classical rescattering model [9] predicts that the highest rescattering energy is $3.2U_p$, which is 0.67 a.u. at I_0 for a 780-nm laser. Here $U_p = I/(4\omega^2)$ is the ponderomotive energy and I is the laser intensity. Since the lowest excitation energy (the excitation threshold energy) of He^+ is 1.5 a.u., the NSDI probability should be zero when the laser intensity is lower than $2.2I_0$. Thus, when the laser intensity is lower than this critical intensity, the NSDI channel is closed. As the intensity increases, the portion of the returning high-energy electron increases and this increases the NSDI probability. The general trend of the ratio of NSDI to SI can also be estimated by a simple model. From the classical rescattering model [9], the returning electron carries the highest energy when the electron tunnels out at $\omega t = \theta = 17^\circ$. Thus the ratio of the returning electron with $3.2U_p$ to the total ionized electron in a half cycle is

$$R_s(I) \propto \sqrt{E_0} e^{-2\kappa^3(1/\cos\theta - 1)/(3E_0)}, \quad (11)$$

where E_0 is the laser field strength and $\kappa = \sqrt{2I_p}$ with I_p the ionization potential of He. This equation is derived from Eqs. (9) and (10) in Ref. [27]. This ratio increases as the laser intensity increases. Suppose only the $3.2U_p$ electron contributes to NSDI, then the ratio of NSDI to SI is approximated as

$$R(I) = R_s(I)\sigma(3.2U_P). \quad (12)$$

Apart from an overall constant, this simple model predicts a correct trend that the ratio increases rapidly in the low-intensity range, reaches a plateau, and then decreases gradually as the intensity increases further as shown in Fig. 1. Although the simple model shows better agreement with the measurements than the present quantum simulation around 10^{15} W/cm^2, this agreement could be fortuitous given the simplified assumption that the return energy is fixed at 3.2 U_p. Our elaborate simulation shows that the energy spreads more [20]. In the plateau region, our results are lower than the measured ones. As the laser intensity increases above $12I_0$, the simulated results increase again. This might be due to the fact that tunneling ionization dominates in the plateau region while, at the high intensity ($>12I_0$), over-barrier ionization [28] is dominant.

To investigate the details of the rescattering process, we propagate the wave packet for 40 cycles after the direct ionization and find that the later returns after the 10th return contribute less than 3.0% in total at maximum and the later returns after the 20th return contribute less than 0.3%. So we focus on the contributions of the first several returns. Figure 2 shows the contributions of the first five returns normalized to the total NSDI probability P^{2+}. At the lower intensity, the first return is the only contributor. As the laser intensity increases, the contribution from the 3rd return increases quickly and then the 5th return increases. The even returns (the 2nd and the 4th returns) also contribute around 10% in the plateau region above $5I_0$. As the laser intensity increases further, the contribution of the first return increases again.

Although the highest rescattering energy is $3.2U_p$ from the classical prediction, the rescattering energy has a broad distribution in the quantum simulation [20,26] and it is higher than $3.2U_p$. Figure 3 shows the rescattering energy spectra at three intensities, which represent the low-, middle-, and high-intensity cases. For the later returns, the rescattering energies are always lower than the one of the first return. For the lower intensity case, only a small portion of the returning electron can reach the threshold energy so that the NSDI probability is very small and there is no contribution from the later returns. As the laser intensity increases, the rescattering energy increases almost linearly for the first return and the portion of the returning electron which contributes to the NSDI process also increases. Meanwhile, the rescattering energies of the later odd returns also increase and reach the threshold energy. Thus the later odd returns also contribute to NSDI. In the upper panel of Fig. 3, the even returns almost have no contribution to NSDI at $3I_0$. This explains why the ratio of NSDI to SI probabilities increases from the lower intensity up to $5I_0$ and the relative contribution of the first return decreases. As the laser intensity increases further, as shown in the middle panel of Fig. 3, the rescattering energies of both even and odd returns are higher than the threshold energy and all the first several returns contribute to NSDI. This makes the contribution of the first return even lower. As the laser intensity increases further, as shown in the lower panel of Fig. 3, although the rescattering energies of the later returns are larger than the threshold energy, the relative strengths (indicated by the color coding) of the later returns are smaller than the corresponding ones for $7I_0$. This explains why the contribution of the first return increases when the laser intensity increases again.

There are two interactions which force the electron return to the parent core. One is the electron-core Coulomb interaction and the other is the electron-laser interaction. The Coulomb interaction becomes less important for high-energy electrons when the laser intensity is high. Thus the portion of later returning electrons decreases as the laser intensity increases. The ratio of the first returning probability to the SI probability increases as shown in Eq. (11). Thus, as the laser intensity increases further, the ratio of NSDI to SI probabilities also increases. Meanwhile the SAE approximation becomes less reliable [29] and the sequential double ionization also becomes important in the high-intensity region. All these factors make the problem more complicated. In our present simulation, we do not take into account interference effects.

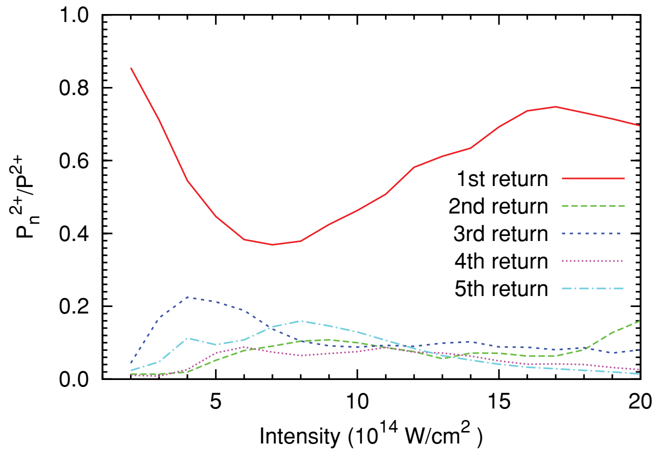

FIG. 2. (Color online) The contributions of the first five returns normalized to the total NSDI probability as a function of the laser intensity.

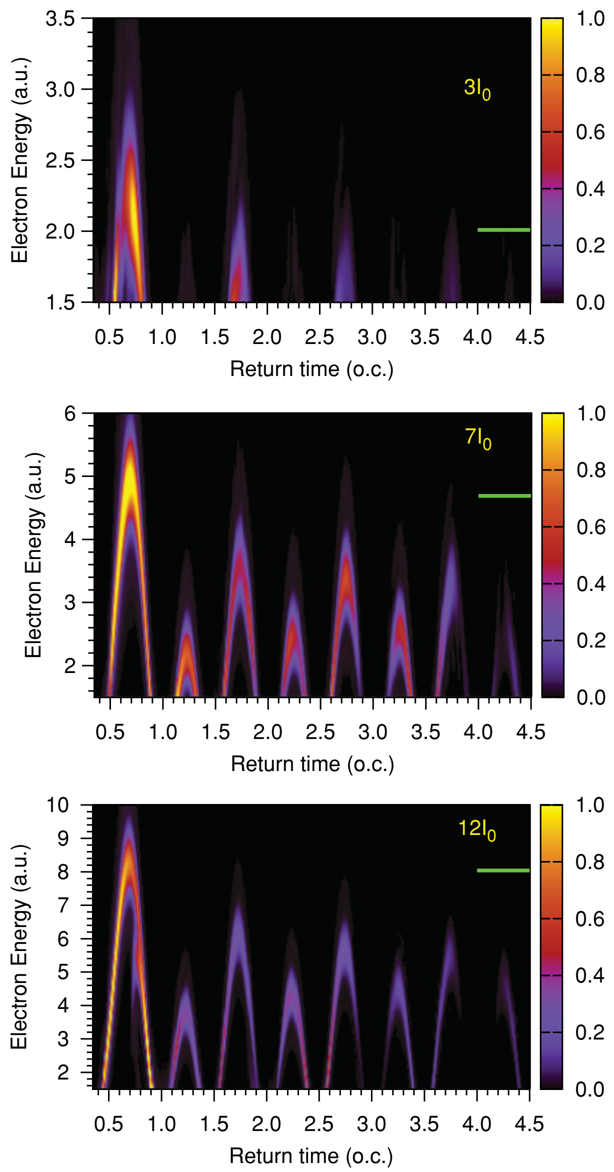

FIG. 3. (Color online) The rescattering energy spectra at the time when the electron revisits the ion at $3I_0$ (upper panel), $7I_0$ (middle panel), and $12I_0$ (lower panel), respectively. The horizontal bars indicate the $3.2U_p$ position. The color bar indicates the relative spectra strength.

There are four types of interference involved in the NSDI: the interference between the different returns, intercycle interference, intracycle interference [30], and the interference between the direct NSDI and the indirect NSDI (via impact excitation). The interference affects the ionized electron momentum distribution [31,32]. Such interference is less important for the total NSDI probability unless the electron energies are very close to the ionization threshold.

Our simulation is for a long pulse, which is close to the experimental condition [1] and we see that each return contributes differently to the NSDI in Fig. 2. Although our ratios of NSDI to SI are close to the semiclassical results [7,17], the contributions from each individual return differ significantly. The contributions of the later returns from the semiclassical simulation are much larger than the present results. Recently, a single-cycle pulse laser has been developed [33] and hence the contribution from each individual return can be studied experimentally by tuning the pulse duration. There are several theoretical studies on pulse duration dependence [7,34] or carrier-envelope phase dependence [35] of NSDI. All these parameters are related to the details of the rescattering energy spectra in the pulsed laser field. The discrepancies between the present quantum simulation and the semiclassical simulations in the contributions of the individual return will be clarified in future experiments.

In conclusion, we have investigated the NSDI process of He in an intense laser field using the rescattering model. The rescattering energy distribution is obtained by a quantum simulation. Our results are in reasonable agreement with the experiment. The present work not only confirms the validity of the rescattering model, but it also provides quantitative information of the rescattering electron from the quantum simulation. Such information is very useful for molecular imaging experiments and it cannot be obtained in the experiment. Numerical simulation is the only way to provide this important information directly and the present work confirmed the reliability of the rescattering energy spectra from the *ab initio* calculations.

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