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Quantitative rescattering theory for nonsequential double ionization of atoms by intense laser pulses

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Laser-induced electron recollisions are fundamental to many strong field phenomena in atoms and molecules. Using the recently developed quantitative rescattering theory, we demonstrate that the nonsequential double ionization (NSDI) of atoms by lasers can be obtained quantitatively in terms of inelastic collisions of the target ions with the free returning electrons where the latter are explicitly given by a spectrum-characterized wave packet. Using argon atoms as target, we calculated the NSDI yield including contributions from direct $(e,2e)$ electron-impact ionization and electron-impact excitation accompanied by subsequent field ionization. We further investigate the dependence of total NSDI on the carrier-envelope phase of few-cycle laser pulses, and showed that the effect can be experimentally observed by measuring the yield of doubly charged ions only.

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I. INTRODUCTION

Nonsequential double ionization (NSDI) of atoms and molecules is one of the most interesting phenomena in the interaction of intense laser fields with many-electron atoms and molecules. Since its first observation on Xe atoms by L'Huillier *et al.* [1], where the Xe^{2+} yield versus laser intensity was shown to be much larger than expected from the tunneling ionization theory, many experiments have been carried out and similar enhancement has been observed for other atoms (see Ref. [2], and references therein) and molecules [3]. Recent studies on the ion momentum distributions for nonsequential double ionization include their dependence on the carrier-envelope phase (CEP) for few-cycle laser pulses [4,5] and their dependence on the laser wavelength [6].

Since "exact" *ab initio* calculations of double ionization of atoms in an intense laser field are exceedingly difficult even for the simplest two-electron atom, helium, the prediction of NSDI has to depend on theoretical models. Although additional processes may contribute to NSDI [7], experimental evidences from ellipticity dependence [8–10], as well as from other theoretical works [11–15], have favored a physical mechanism based on the rescattering picture [16]. In this picture, an electron is promoted to the continuum via tunnel ionization by the laser field. As the laser's oscillating electric field changes direction, the electron may be driven back to recollide with the target ion. The recollision may incur many different observable phenomena. Thus high-order harmonic generation (HHG) is due to the photorecombination of the returning electrons with the target ion, and high-energy above-threshold ionization (ATI) electrons are due to the elastic backscattering of the returning electrons [17–19]. For multielectron targets, NSDI is attributed to resulting from the impact ionization of the target ion by the returning electrons. In other words, the main mechanism of NSDI is the $(e,2e)$ process in the laser field. Since the returning electrons can also excite the target ion to excited states which may be ionized subsequently by the laser field, in calculating the total NSDI yield, electron impact excitation processes should also be included.

Essential to this rescattering picture is that there exists a laser-induced recolliding wave packet which can initiate collisions with the ion core. However, this recolliding wave packet (RWP) is in the presence of the laser field and is not directly measurable. Theoretically, RWP can be deduced in a straightforward manner by following the trajectories of the electrons if the motion of the electrons is treated classically. Some success has been achieved using such models to interpret rescattering in the NSDI of atoms and molecules [14,15,20,21]. In quantal calculations, the RWP becomes much more difficult to extract and depends on theoretical models [22]. Recently, it has been proposed to probe the RWP using an attosecond XUV pulse [23]. Still only partial information can be extracted this way, and the result depends on the probe pulse and the theoretical modeling.

In recent papers [24,25], we have demonstrated how to characterize the spectral distribution of the RWP by analyzing the two-dimensional momentum distribution of high-energy ATI photoelectrons generated by intense laser pulses. By analyzing the two-dimensional (2D) electron momentum spectra from solving the time-dependent Schrödinger equation, we showed that these electrons are indeed due to the elastic scattering of the returning wave packet by the ion core in the backward direction as suggested earlier [17–19], but more importantly, we established quantitatively that the high-energy electron momentum distributions can be expressed as the product of a spectrum characterized RWP with the differential elastic scattering cross sections between free electrons and the target ion. Such analysis allows us to put the rescattering picture on a firm quantitative basis. Using this quantitative rescattering theory, achievements have been made to interpret the intensity and electron energy dependence of the high-energy electron spectra for different target species in terms of their differential elastic scattering cross sections [26]. This model has also provided a robust method to simultaneously determine the laser peak intensity, pulse duration, and carrier-envelope phase (CEP) of few-cycle laser pulses [27,28]. Likewise, a similar theory has also been developed for the HHG [24,29,30]. In this paper, we use the RWP derived from the high-energy ATI electron momentum spectra to calculate the NSDI yield using electron impact

ionization and excitation cross sections weighted by the momentum distribution of the RWP. With such a model, we first calculate the NSDI yield of Ar and compared the results with earlier experimental data. We also make calculations to predict how the NSDI yield depends on the carrier-envelope phase if the Ar atoms are ionized by phase-stabilized few-cycle pulses and show that such dependence may be measured to calibrate the absolute value of the CEP experimentally.

The rest of the paper is arranged as follows. In Sec. II, we first summarize how we extract the returning wave packet from the 2D photoelectron momentum distributions and show the dependence of the RWP on the laser peak intensity. In Sec. III, we demonstrate that the variation of the NSDI yield with the laser peak intensity can be described as a recollision mechanism, where the ionization of the second electron is due to the inelastic scattering of the RWP with the target ion. The conclusions and outlook are given in Sec. IV.

II. RETURNING ELECTRON WAVE PACKET

According to the quantitative rescattering model (QRS) [24,26], the laser-generated high-energy photoelectron momentum distribution $I(p,\theta)$ can be written as

$$I(p,\theta) = W(p_r)\sigma(p_r,\theta_r), \tag{1}$$

where $\sigma(p_r,\theta_r)$ is the differential elastic scattering cross section between free electrons with momentum p_r and the target ion, and θ_r is the scattering angle with respect to the "incident" direction (the direction of the returning electron). In this equation $W(p_r)$ is interpreted as the momentum distribution of the returning electron wave packet. The validity of this interpretation is partly justified by showing that $W(p_r)$ thus obtained from Eq. (1) is independent of the angle θ_r, using $I(p,\theta)$ calculated from solving the time-dependent Schrödinger equation (TDSE) of a one-electron atom in the laser field, and $\sigma(p_r,\theta_r)$ from solving the standard quantum-mechanical electron-ion elastic scattering theory [31]. In Eq. (1), the final momentum $\mathbf{p}$ of the photoelectron (with magnitude p and angle θ with respect to the linear polarization axis) is related to the momentum $\mathbf{p}_r$ of the elastically scattered returning electron via

$$\mathbf{p} = \mathbf{p}_r - A_r\hat{\mathbf{p}}_z, \tag{2}$$

where $A_r = A(t_r)$ is the magnitude of the instantaneous vector potential at the time t_r of electron-ion collision. The second term on the right side of the above equation shows that the elastically scattered electron will gain an additional momentum (in the $\hat{z}$ direction) as it drifts out of the laser field. Atomic units are used throughout the paper unless otherwise stated.

The validity of Eq. (1) for photoelectrons with energy $\geqslant 4$ U_P (U_P is the ponderomotive energy) has been established from TDSE calculations for alkali and rare gas atoms within the single-active electron approximation [26]. Using Eq. (1), extraction of elastic differential cross sections from experimental and theoretical data have been demonstrated for short [24,27,32] and long pulses [33]. However, calculating the high-energy photoelectron momentum distribution by solving the TDSE remains computationally challenging for high laser intensities or long pulses. An alternative method is to calculate $I(p,\theta)$ using the second-order strong field approximation (SFA2) where the ionized electrons are described by Volkov states, i.e., plane waves in the laser field [34,35] and where the cross sections $\sigma(p_r,\theta_r)$ are calculated using the first Born approximation [31]. Thus from Eq. (1), the recolliding wave packet $W(p_r)$ can also be calculated. In Chen *et al.* [25], it has been demonstrated that the wave packets obtained from SFA2 and from solving TDSE are very close to each other, except by an overall normalization factor. This normalization factor is due to the fact that tunneling ionization rate calculated from SFA tends to be too small. However, the spectral shape, or more precisely, the energy or momentum distributions of the RWP are very similar. By using $W(p_r)$ from the SFA2 and accurate $\sigma(p_r,\theta_r)$ from electron-ion collision theory, the QRS model states that the high-energy electron momentum spectra generated by infrared lasers can be calculated from Eq. (1). The validity of this model has been well documented [26,27]. Using QRS one can also achieve great saving in computer time besides retaining the conceptual simplicity of the rescattering picture.

Details on SFA2 have been given in a previous paper [25]. For high-energy electrons, SFA2 is the only dominant term and the probability amplitude of detecting an electron with final momentum $\mathbf{p}$ is given by

$$f(\mathbf{p}) = -\int_{-\infty}^{\infty} dt \int_{-\infty}^{t} dt' \int d\mathbf{k} \langle \chi_{\mathbf{p}}(t)|V|\chi_{\mathbf{k}}(t)\rangle \times \langle \chi_{\mathbf{k}}(t')|H_i(t')|\Psi_0(t')\rangle a(t)a(t'). \tag{3}$$

Here

$$H_i(t) = \mathbf{r}\cdot\mathbf{E}(t) \tag{4}$$

is the laser-electron interaction, in length gauge and in dipole approximation, and the electric field $\mathbf{E}(t)$ of the laser pulse is chosen to be linearly polarized along the z axis

$$\mathbf{E}(t) = E_0(t)\cos(\omega t + \phi)\hat{z}, \tag{5}$$

where ϕ is the CEP. The envelope function is taken as

$$E_0(t) = E_0 \cos^2\left(\frac{\pi t}{T}\right) \tag{6}$$

for the time interval $(-T/2, T/2)$, and zero elsewhere, and T is the total duration of the pulse, and is related to the full width at half maximum of the intensity, or the pulse duration τ, by $\tau = T/2.75$. The function $a(t)$ describes the depletion of the ground state and is evaluated by

$$a(t) = e^{-\int_{-\infty}^{t} W(t')dt'/2}, \tag{7}$$

where the ionization rate $W(t)$ is calculated following the usual Ammosov-Delone-Krainov theory [36]. The functions $\chi_{\mathbf{p}}(t)$ are the Volkov states describing free electrons in the laser field and $\Psi_0(t)$ is the ground-state wave function. In the numerical integration of $f(\mathbf{p})$, an additional damping factor $e^{-\alpha r}$ is introduced in the model potential to avoid the singularity in the integrand. We choose $\alpha = 2$ and checked that the magnitude of $f(\mathbf{p})$ varies slightly with the value of α, but the

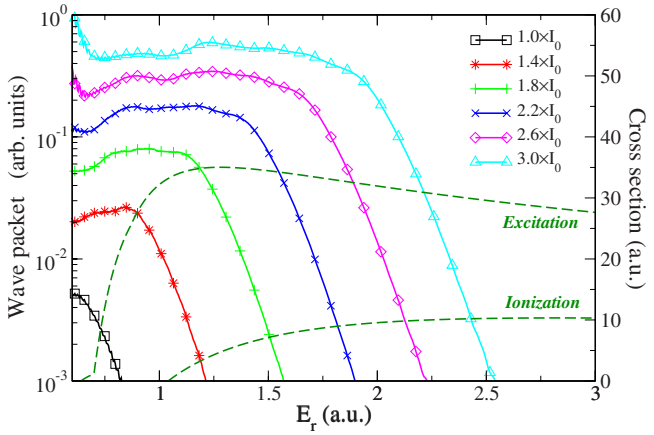

FIG. 1. (Color online) Volume-integrated electron wave packets extracted from photoelectron spectra on the "right" side, for atomic argon singly ionized by a five-cycle laser pulse with a mean wavelength of 800 nm, for CEP equals to zero, and different peak intensities (with $I_0=1.0\times10^{14}$ W/cm^2). The total electron-impact ionization and excitation cross sections from the ground state of Ar^+ are also presented.

shape remains the same. The integral is calculated within the saddle point approximation for the integration with respect to **k**, as described in Lewenstein *et al.* [37]. The electron momentum distribution is given by $I(p,\theta)=|f(\mathbf{p})|^2$. The elastic differential cross section within the plane-wave Born approximation is trivially evaluated.

Experimentally, the intensity distribution of a focused laser beam is not uniform in space. Thus to compare with experimental results, volume effects must be included in the theoretical calculations [38,39]. Assuming a Gaussian spatial distribution of the laser intensity, we define a volume-integrated returning wave packet. We show in Fig. 1 the energy distribution of volume-integrated RWP for different laser peak intensities, for a laser with mean wavelength of 800 nm, five-cycle pulse duration. The CEP is set to zero and the RWP are extracted from the "right" side (i.e., from the electron yield with a positive final momentum p_z). Increasing the laser peak intensity results in a strong increase of the yield of recolliding electrons as well as the returning energy $E_r=p_r^2/2$, since the strength of the RWP varies as the tunnel ionization rates, and its momentum depends on the vector potential A_r. We mention again that the absolute magnitude of the returning wave packet is not retrieved since SFA2 gives inaccurate electron yields [25]. In addition, we comment that such volume-integrated RWP can also be directly extracted from experimental high-energy electron momentum spectra if they are measured. This is possible because in Eq. (1), the elastic scattering cross section depends only on the returning electron energy (or momentum), not on the laser intensity.

III. NONSEQUENTIAL DOUBLE IONIZATION YIELD

In this paper we investigate if we can take the QRS model one step further by using the RWP obtained from the high-energy ATI electron momentum spectra to calculate NSDI, by replacing the differential elastic scattering cross sections with the electron impact ionization cross section. Thus the total NSDI yield is evaluated from

$$\sigma^{++}=\int dE_r[W_L(E_r)+W_R(E_r)]\sigma(E_r), \tag{8}$$

where $W_L(E_r)$ and $W_R(E_r)$ are the volume-integrated wave packets extracted from the "left" and the "right" sides, respectively, and $\sigma(E_r)$ is the electron-impact inelastic cross section from the ground state of the target ion, in the absence of the laser field. The inelastic scattering cross section includes both ionization and excitation cross sections. Indeed, an electron in the target ion can be removed either through direct $(e,2e)$ collision, or through recollision-induced excitation of the ion followed by field ionization [40]. Since the ionization potential of the first Ar^+ excited state is $I_P=0.4$ a.u., the critical classical electric field $E_t=I_P^2/4Z^*$ (where $Z^*=2$) required for the electron to overcome the potential barrier is calculated to be 0.02 a.u., which corresponds to a laser pulse with peak intensity of 1.4×10^{13} W/cm^2. For the range of intensities studied for NSDI, all the excited states of Ar^+ ions produced by electron impact excitation will be ionized.

For electron-impact ionization cross sections, we use the semi-empirical expression of Lotz [41], which has been shown to be in good agreement with experimental results for Ar^+ [42]. For the excitation cross sections, we use the empirical formula from Tong *et al.* [44]:

$$\sigma(E_r)=\alpha\frac{\pi}{\Delta E^2}e^{1.5(\Delta E-\epsilon)/E_r}f\left(\frac{E_r}{\Delta E}\right), \tag{9}$$

$$f(X)=\frac{1}{X}\left[\beta\ln X+\gamma\left(1-\frac{1}{X}\right)-\delta\frac{\ln X}{X}\right], \tag{10}$$

where ΔE is the excitation energy for a given transition. This expression has been obtained initially by fitting to the convergent-close coupling excitation cross sections for hydrogen and He^+ [43]. Keeping the other parameters as in Ref. [44], we adjust the formula by multiplying a factor α such that the cross sections reproduce the distorted-wave Born approximation (DWBA) data recommended by the International Atomic Energy Agency [45] at high energy ($E_r=20$ a.u.). The excitation cross sections for various transitions from the Ar^+ ground state are presented in Fig. 2 and the parameters are given in Table I. The excitation cross sections for the transition to $4f$ and higher excited states are small and are not considered in the following calculations. The total excitation cross sections, as well as the ionization cross section, are shown in Fig. 1. Since the energies of the RWP are close to the thresholds for both ionization and excitations, the cross sections calculated using DWBA are not valid.

A. Intensity dependence

Using Eq. (8), we calculated the NSDI yield for argon atoms as a function of the laser peak intensity for a 30 fs pulse. We comment that the RWP in Eq. (8) includes the

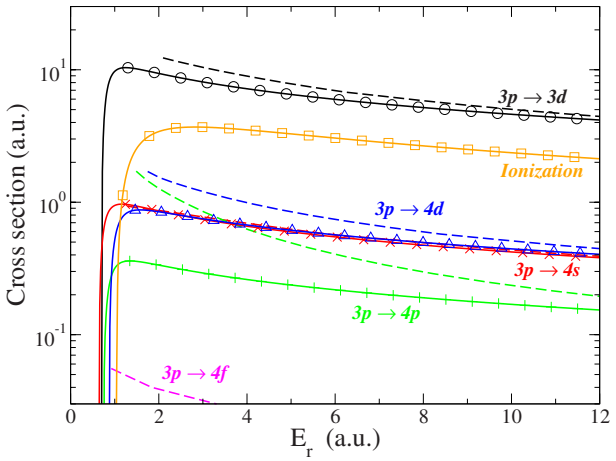

FIG. 2. (Color online) Electron-impact ionization and excitation cross sections for the ground state of Ar^+. The ionization cross section is calculated by means of the Lotz formula. The excitation cross sections (full line with various symbols) are determined from an empirical formula [44] adjusted to fit distorted-wave Born approximation calculations [45] at high-energy (dashed lines), see text.

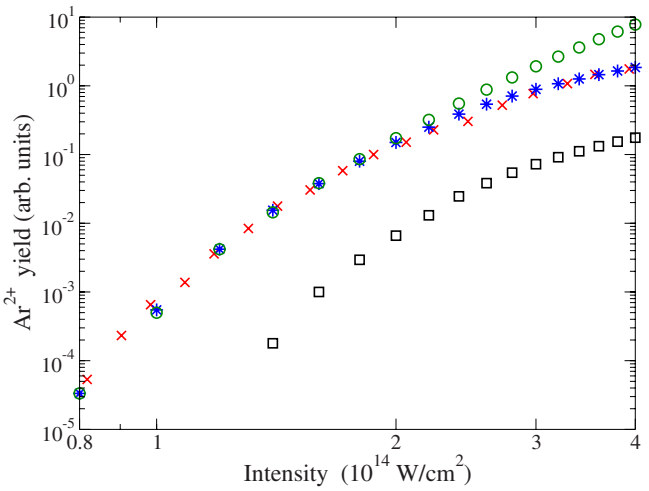

FIG. 3. (Color online) Nonsequential double ionization yield for argon atoms as a function of the peak intensity for a linearly polarized laser pulse with mean wavelength of 800 nm. The red crosses are experimental results from Ref. [46] for a 30 fs pulse duration. The blue stars are from calculations when depletion effects and collisional ionization and excitation processes are considered. The results when depletion effects are omitted are shown as green circles, and the contribution when only direct ionization is included is represented by black squares.

laser focus volume integration such that the NSDI yield calculated are double ionization yield integrated over the whole focused laser beam. This effect has not been included in most of the previous NSDI calculations. We also note that for long pulses, the momentum distribution of the volume integrated RWP is roughly independent of the pulse duration. Its magnitude would be proportional to the pulse duration except when depletion occurs and the atom is fully ionized before the laser pulse is over [25]. As discussed earlier, the depletion effect is included in our simulation. In Fig. 3, comparison with the experimental data from Guo *et al.* [46] shows that the present model works well, and indeed the intensity dependence of the NSDI can be modeled as due to electron-impact ionization of the Ar^+ ions by the returning electrons. Saturation effects at high peak laser intensities (or long pulses) are clearly observed since Eq. (8) overestimates the NSDI yield if depletion is not accounted for, see Fig. 3. Furthermore, Eq. (8) allows us to distinguish the relative contributions from direct collisional ionization, and indirect ionization through intermediate excited states. For argon atoms, the main contribution to NSDI comes from collisional excitation followed by field ionization since the Ar^{2+} yield is at least one order lower when only direct ionization is considered, in accordance with experimental findings [40]. For intensities lower than 1.2×10^{14} W/cm^2, the energy distribution of the RWP is below the Ar^+ ionization threshold, such that the only pathway to NSDI is through collisional excitation (see Fig. 1). Note that the relative branching ratio of excitation versus ionization pathways might explain the differences in the observed longitudinal ion momentum distributions of doubly charged ions for different targets [47]. We comment that such a study can be performed using the same RWP. It, however, requires accurate differential ionization and excitation cross sections and is beyond the scope of this paper.

TABLE I. Values of the parameters used in Eqs. (9) and (10) to calculate the excitation cross sections for various transitions from the ground state of Ar^+.

Transition	$3p\rightarrow3d$	$3p\rightarrow4s$	$3p\rightarrow4p$	$3p\rightarrow4d$
ΔE (a.u.)	0.70	0.64	0.74	0.87
α	8.7	0.7	0.35	1.16
β	0.7638	0.7638	0.7638	0.7638
γ	−1.1759	−1.1759	−1.1759	−1.1759
δ	−0.6706	−0.6706	−0.6706	−0.6706
ϵ	0	0.0243	0.0243	0.0243

B. CEP dependence

Liu *et al.* have shown that the longitudinal momentum distributions of Ar^{2+} from NSDI strongly varies with the CEP of few-cycle laser pulses [4]. Recently, we have studied the CEP dependence of high-energy photoelectron spectra emitted on the left and on the right theoretically and compare to results from experiment [27]. Based on the QRS, the left-right asymmetry in the electron spectra has been traced to the left-right asymmetry in the energy distributions of the RWP [28]. Here we examine whether the CEP dependence of the left-right wave packets can result in the CEP dependence of the total NSDI yields.

The "left" and "right" volume-integrated wave packets calculated for a five-cycle laser pulse with a peak intensity of 2×10^{14} W/cm^2 and different CEP are shown in Figs. 4(a) and 4(b). The energy distribution of the RWP clearly shifts with the CEP on the left RWP for ϕ from 0 to $3\pi/4$, and on the right RWP for ϕ from $3\pi/4$ to π. Note that for ϕ from π

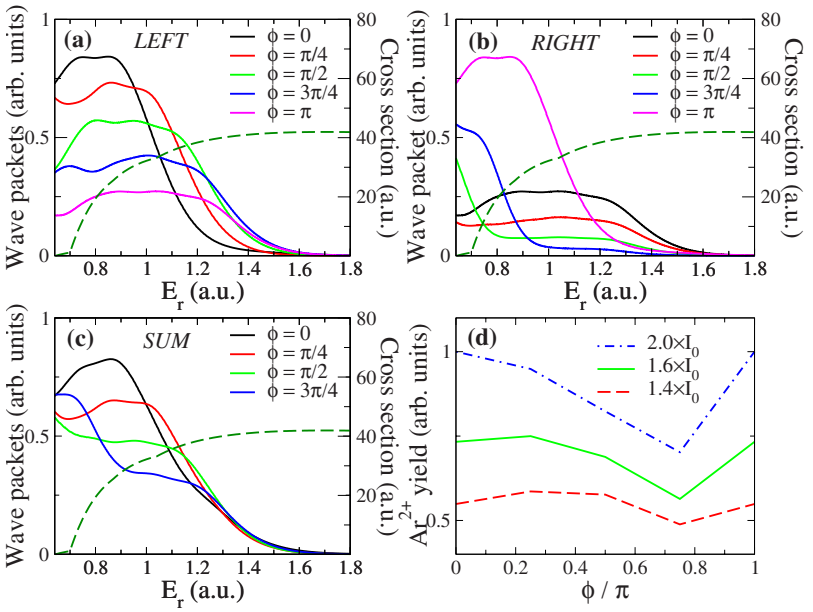

FIG. 4. (Color online) Left (a), right (b), and total (c) volume-integrated wave packets for argon atoms in a five-cycle laser pulse with a peak intensity of 2.0×10^{14} W/cm^2, a mean wavelength of 800 nm, and different carrier-envelope phases. The electron-impact inelastic cross section for the ground state of Ar^+ is also shown. (d) Nonsequential double ionization yield as a function of the absolute phase of a five-cycle laser pulse with a mean wavelength of 800 nm and different peak intensities.

to 2π, the left and right sides are simply interchanged. This smooth evolution and its effect on the high-energy ATI spectra has already been demonstrated [27]. However, for the total NSDI yield [Eq. (8)], the sum of the "left" and "right" wave packets must be used. In Fig. 4(c), we note that the sum wave packet still depends on the CEP. By multiplying the total spectral distribution of the wave packet with the total inelastic cross sections (see Fig. 4) and integrate over the returning electron energies we obtain the total NSDI. The results shown in Fig. 4(d) indicate that the NSDI has a minimum near $\phi=3\pi/4$. This minimum is due to the spectral distribution in the total wave packet [see Fig. 4(c)] and thus should be rather insensitive to the peak laser intensity, see Fig. 4(d), nor to the target used. This may provide a simple way to determine the absolute value of the CEP since this only requires the measurement of the total yield of the doubly charged ions. Note that the CEP is still undecided up to π if this method is used.

IV. CONCLUSIONS

In summary, we have shown that the recently developed quantitative rescattering theory (QRS) can be used to explain the total nonsequential double ionization (NSDI) of atoms by intense lasers. According to QRS, NSDI is due to electron impact ionization and electron impact excitation of the target ion by the returning electrons, where the electrons are described by a recolliding wave packet (RWP). The momentum distribution of the RWP can be extracted theoretically or experimentally from the two-dimensional momentum distributions of the high-energy photoelectrons. Our results quantitatively confirm the prevailing interpretation of NSDI in terms of the rescattering mechanism. The QRS model in principle can be extended to investigate electron and/or ion momentum distributions of the NSDI process, thus allowing one to distinguish contributions from direct ionization process vs excitation followed by tunnel ionization process. It can also be extended to investigate the momentum correlation between the two outgoing electrons [48,49]. Such studies would establish an even more direct connection between ionization and excitation of an atomic ion by free electrons with the NSDI which has been attributed to be due to the same processes in the laser field. On the other hand, such simulations would require differential cross sections for both electron-impact ionization and electron impact excitation processes. These cross sections are much more difficult to obtain since no empirical formula exist and they have to be calculated using advanced scattering theories.

ACKNOWLEDGMENTS

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