The effect of Coulomb field on laser-induced ultrafast imaging methods

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The deconvolution procedure of the laser-induced ultrafast imaging schemes is usually based on strong field approximation (SFA) wherein the Coulomb interaction between the parent ion and the freed electron is ignored after ionization. In the laser-induced electron diffraction (LIED) approach, for example, the high energy part of the above-threshold ionization (ATI) spectrum used for analysis is assumed to be produced mostly by the 1st-return-recollision trajectories according to the SFA. By performing a joint theoretical and experimental investigation on the ATI spectrum, the dominant role of the 3rd-return-recollision trajectories in the high energy part of the spectrum due to the ionic Coulomb field is identified, which invalidates the key assumption adopted in the conventional LIED approach. Since the incident (return) electron beams produced by the 1st and 3rd returns possess distinct characteristics of beam energy, beam diameter and temporal evolution law due to the influence of Coulomb field, the extracted results in the LIED will be significantly altered. Such Coulomb field effect should be taken into account in all kinds of laser-induced imaging schemes based on recollision.

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As one of the most important processes in strong field physics, recollision has provided an unprecedented insight into the inner working of atoms and molecules ¹ ². In recollision, one electron is liberated from the target atom or molecule through tunneling ionization and then be accelerated in the field and pulled back by the field to collide with the parent ion. Most intriguing phenomena in strong field physics, such as high-order above-threshold ionization(HATI), high harmonics generation(HHG) and nonsequential double-ionization(NSDI), can be well understood based on the recollision physics(see, e.g., ³ ⁴ for reviews and references therein).

Since the products upon recollision carry information of the parent ion, they can be used to probe the structure of the parent ion. Various methods have been proposed to image molecules in intense laser fields based on the analysis of different products upon recollision, such as laser-induced electron diffraction (LIED) ² ¹², molecular clock¹³ ¹⁰, tomographic imaging of orbitals¹⁷ ¹⁸ and laser-induced inelastic diffraction (LIID)¹⁹. Since these methods are self-imaging approaches based on coherent electron scattering, they can provide an unprecedented spatial-temporal resolution. In general, as long as the imaging method is based on electron scattering, the resolution of extracted results will depend upon the parameters of the incident electron beam, such as the beam energy, the beam diameter and the temporal evolution of the beam. Unlike the conventional electron diffraction (CED) method in which the information of the external electron beam is well known, the incident (return) electron beam in the self-imaging method is produced by the laser-induced ionization of the target itself and is much more complicated. In LIED, for example, the temporal resolution relies on the knowledge of the temporal evolution of the beam, specifically, deconvolution of the exact recollision time. In the recollision process, however, the electron may miss the ion at the 1st return and collide with it at the subsequent returns as illustrated in Fig. ¹ (a), which results in a large uncertainty in the recollision time. To complicate matters further, the energies and the impact parameters of different returns vary. In LIED, the aforementioned complexity is largely ignored by applying the strong field approximation (SFA) wherein the Coulomb interaction between the parent ion and the freed electron is ignored. According to SFA, the maximal kinetic energy at collision for the 1st return is much higher than the subsequent returns. Thus, the high energy part of the momentum distribution selected for analysis in LIED is assumed to be produced mostly by the 1st-return-recollision trajectories. Furthermore, due to the spread of the electron wavepacket, the probabilities of the multiple-return-recollision trajectories are much smaller.
Nevertheless, more and more studies have shown that the Coulomb field plays an important role in the ionization dynamics of atoms and molecules in intense laser field. Most of the previous works on effect of Coulomb field mainly focused on the low energy electrons which seem easier to be disturbed. These low energy electrons either never experience collision or only experience a soft collision during the pulse. For hard collision trajectories which is usually picked for imaging, the Coulomb focusing effect will significantly improve the contributions of the multiple-return-recollision trajectories and in some cases they will even exceed that of the 1st return. Therefore, the basic assumption on the incident electron beam in the ultrafast imaging method may be in question, and, the impact of the Coulomb field on the imaging results needs to be carefully accessed. In this letter, based on the simulated results combined with experimental observations, we systematically analyze the incident (return) electron beam produced by laser-induced ionization of atoms in the presence of Coulomb field. We find that, for a multi-cycle laser field, the 3rd return trajectories play a dominant role in the high energy part of the ATI spectrum. Since electron beams associated with the 1st and 3rd returns show distinct characteristics, using the 3rd return for analysis in LIED will cause significant change in the imaging results.

Trajectories with multiple returns will contribute to the ATI spectrum if increasing the laser pulse duration, so the dependence of the spectrum on the pulse duration reflects the signatures of the recollision trajectories with different returns. In Fig. 1 we present the photoelectron energy spectra calculated with semiclassical model (Fig. 1(b)) and numerical solution of time-dependent Schrödinger equation (TDSE) (Fig. 1(c)), and also the measured spectra (Fig. 1(d)) for different pulse durations at wavelength of 800 nm. In the semiclassical model, the bound electron is firstly tunnel ionized in the combined laser electric field and atomic Coulomb field. The initial tunnel exit, the initial transverse velocity and the weight of the electron trajectory are calculated according to the adiabatic tunneling theory. After ionization, the evolution of the free electron is governed by the 3D Newton’s equations of motion and the Coulomb interaction between electron and parent ionic core is fully considered (for more details see Ref 32). The TDSE is solved numerically in length gauge within the single-active-electron (SAE) approximation. The wave function is expanded in spherical harmonics with each angular momentum channel expanded on a radial grid. The resulting coupled equations are solved using a Peaceman-Rachford propagator. We choose a total box size up to 2000 a.u. and 200 partial waves. The numerical convergence is checked by varying the simulation parameters. In both semiclassical and TDSE calculations, the pulse envelope takes the trapezoid shape, and the pulse duration is determined by the duration of the flat-top which is indicated by the legends.

In our experiment, the laser pulses with duration of 25 fs at 800 nm are generated by a commercial Ti:sapphire femtosecond laser system (FEMTOPOWER compact PRO CE-Phase HP/HR). To generate few-cycle laser pulses, the laser pulses from the commercial laser system are spectrally broadened by self-phase modulation in a hollow-core fiber (HCF) filled with Ne and subsequently compressed by a series of chirped mirrors. This system allows the compression of our laser pulses down to a duration as short as 5.0 fs with energy of around 400 μJ. The output pulse energy can be controlled precisely by a combination of an achromatic λ/2 wave-plate and a thin film polarizer. Note that there is no attempt to stabilize the carrier-envelope phase of the laser field during the experiment. The laser beam is introduced into the vacuum chamber of a homemade time-of-flight (TOF) photoelectron kinetic energy spectrometer with a limited detection angle (0.026 sr). The sample gas of Ar is fed through a leak valve. The base pressure of the residual gases inside the chamber is below 2 × 10⁻⁹ mbar. The photoelectrons are measured with a Microchannel Plates (MCP) detector and are recorded by a multihit time-to-digital converter, which is then transformed into a photoelectron spectrum.

All the spectra in Fig. 1 are normalized to themselves by dividing the maximum of the individual spectrum. An excellent agreement can be found between simulations (Figs. 1(b), (c)) and measurements (Fig. 1(d)).
general, they have three common features: i) all spectra show a rapid decrease within $2U_p$ followed by a plateau extending to more than $10U_p$, ii) the plateau shifts up perceptibly with increasing pulse duration, iii) the difference of the spectrum between the shortest and the intermediate pulses is much greater than that between the intermediate and the longest ones. Feature iii) indicates that some specific multiple return recollision trajectories contribute significantly to the plateau.

In the semiclassical model, the different return recollision trajectories can be distinguished according to the travel time $t_t$ defined as the interval between the ionization time and the recollision time. For trajectory with $t_t$ in the interval $[(n/2)T, ((n+1)/2)T]$ ($T$ is the optical cycle), we denote it as the $n$th return recollision trajectory $R_n$. In Fig. 2 we separate the contributions from recollision trajectories of different returns corresponding to the spectra in Fig. 1(b). In the case of the shortest duration (Fig. 2(a)), the contribution of the 2nd return exceeds that of the 1st in the low energy part of the plateau, while the contributions of the 3rd and the 4th returns can be ignored. So the plateau is mainly determined by the 1st and the 2nd returns in the case of the shortest duration. When the pulse duration increases (Fig. 2(b)), the contribution of the 3rd return increases significantly, and even exceeds that of the 1st in the high energy part of the plateau. At the same time, the 2nd return also increase a little, but the cutoff is much smaller than that of the 1st and the 3rd return. The contribution of the 4th return still can be ignored although it also increases. When the pulse duration further increases (Fig. 2(c)), the 3rd and the 4th returns increase slightly while the other two returns nearly have no change. Therefore, in the case of the longest pulse duration, the 2nd return dominates the low energy part of the plateau while the 3rd return dominates the high energy part. Apparently, the contributions of the specific multiple returns exceeding that of the 1st return is a result of the competition between Coulomb focusing effect and wavepacket spreading. In addition, the cutoff of the 3rd return in Fig. 2(c) is identical to that of the 1st and reaches almost $12U_p$, which is higher than the prediction of SFA or simple man theory ($10U_p$ for the 1st return and $8.8U_p$ for the 3rd return) and should also be attributed to the effect of the Coulomb field.

The above contradiction to SFA or simple man theory which is one of the key assumptions in LIED, can be attributed to modification of the incident (return) electron beam before recollision by the ionic Coulomb potential. The electron beam can be characterized by the temporal evolution of the beam, the beam energy (recollision energy $E_r$) and the beam diameter (impact parameter $s$) as schematically illustrated in Fig. 3(a). In Fig. 3(b) we present a simulated 2D distribution of $E_r$ and recollision time $t_r$ for recollision wavepacket of Fig. 2(c), which reveals the temporal evolution of the incident electron beam. It is worthwhile mentioning that, the ionization time of electron is restricted in the interval of $(-T/4, T/4)$, so that the four peaks in Fig. 3(b) correspond to the four returns, respectively. If compared with the results predicted by the simple man theory, the deviation is obvious. Specifically, the recollision energies of the four returns are affected by the Coulomb field in opposite ways: the energies of the 1st and 3rd returns are increased while that of the 2nd and the 4th returns are decreased. This can be attributed to the different evolution characteristics for odd and even returns, and the deviation from the simple man prediction decreases with ponderomotive energy $U_p$ and will disappear when $U_p$ is very large (see supplementary material for more details).

In Figs. 3(b) and 3(c) we plot the 2D distribution of the recollision energy $E_r$ and the impact parameter $s$ for the 1st return and the 3rd return, respectively (see sup-
for the 1st and 3rd returns. The distributions are normalized to the maximum of the 3rd return. The result predicted by simple man theory is also shown for comparison. (c) and (d) The 2D distribution of recollision energy and impact parameter for the 1st and 3rd returns. The distributions are normalized by dividing the maximum of the 3rd return.

![Graph](image)

**FIG. 4:** (Color online) (a) Simulated energy spectra with semiclassical model at wavelength of 3100nm and intensity of $1.0 \times 10^{14}$W/cm². (b) 2D distribution of recollision energy $E_r$ and recollision time $t_r$. The result predicted by simple man theory due to the very large $U_p$. Overall, the distributions in Figs. (c), (d) are similar to the results at 800 nm except some details. First, the distribution of the 1st return becomes more uniform in both energy and $s$ directions than that at 800 nm (comparing Fig. (a) with Figs. (c)), and for the 3rd return, the distribution concentrates in the small impact parameter regime for both wavelengths but distributes wider in the energy direction in the 3100 nm case. It indicates a more tight focusing at 800 nm and the difference between the contributions from the 1st and the 3rd returns to the spectrum at 3100 nm is much greater than that at 800 nm, which can be clearly seen by comparing Fig. (a) with Fig. (c). This can be attributed to the so-called defocusing effect \[ U_0 \leq 10 \text{ fs} \text{ cm}^{-1} \text{ in LIED scheme. Third, in order to obtain the field-free electron-ion differential cross section, the acceleration by the laser field after recollision, which is equivalent to the vector potential $A(t_r)$ of the field at the recollision time $t_r$, needs to be subtracted from the momentum distributions. The resulted uncertainty of $A(t_r)$ for different returns also has to be carefully checked (see supplementary material for details). Given the above, the molecular structure information extracted by LIED will be quite different if the 3rd-return-recollision trajectories instead of the 1st return ones are considered for analysis.

In conclusion, we perform a joint investigation on the dependence of ATI spectrum on the pulse duration theoretically and experimentally. The results indicate that the 3rd-return-recollision trajectories dominate the high energy part of the ATI spectrum due to the ionic Coulomb field effect, which invalidates the key assumption of SFA applied in LIED. The incident (return) electron beams associated with different returns show di-
tinct characteristics in the presence of the Coulomb field. Compared with the 1st return, the 3rd return generates an electron beam with a much smaller diameter and a much higher intensity, thus providing more high energy photoelectrons. Moreover, it is found that the Coulomb field has distinct effects on the electron beam energy for odd and even returns. More specifically, it increases the electron energy for the 1st and 3rd returns, and reversely, decreases the electron energy for the 2nd and 4th returns, when compared to the simple man’s case. The above Coulomb effects will significantly change the results extracted from the LIED approach and should be taken into account in current imaging schemes based on recollision physics.

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