Computational methods for laser-atom interactions

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Abstract. We discuss a computational method to study the dynamics in the laser-atom interactions. There are two key ingredients to the new method. Firstly, we transform the differential time-dependent Schrödinger equation into a time-integral equation, in which the dynamics related wavefunction is separated from the background wavefunction analytically to improve the numerical accuracy. Secondly, we divide the space into an inner region and an outer region, and propagate the inner-region wavefunction in the full Hamiltonian numerically and outer-region wavefunction in the momentum space analytically. In this way, we remove the physical boundary in space. To show the effectiveness of the method, we simulate the carrier-envelop phase dependent high energy above-threshold-ionization yields, which are in good agreement with the experimental observations. Furthermore, we investigate the rescattering electron momentum spectra and provide an intuitive rescattering picture from a full quantum non-perturbative calculation.

1. Introduction

The laser-atom interactions are one of the hot topics owing to the development of the laser technology in the past one or two decades. In principle, all the dynamics of the laser-atom interactions can be studied by directly solving the time-dependent Schrödinger equation. In practise, only few cases, like the above-threshold-ionization (ATI) \(^1,2,3\) and the high-order-harmonic generation (HHG) \(^4,5\), can be studied directly solving the time-dependent Schrödinger equation based on the single-active-electron (SAE) approximation. Even within the SAE approximation, most of the important minor processes were studied by the classical model \(^6\), semi-classical model \(^7,8\) or quantum methods with various approximations, such as the Lewenstein model \(^9\) or the S-matrix method \(^10\). Such models can be used to study the dynamics qualitatively or semi-quantitatively. To provide deeper insight of the physics of the laser-atom interactions, we have to develop a full quantum, non-perturbative method.

First let us examine the difficulties of conventional time-dependent methods. As we know, in any numerical simulation, we have a limited box size and all the physical processes happen inside of the box. In most of the cases, the high energy electron moves very fast and the electron reaches the boundary (box size) well before the laser pulse is over. Thus we have to treat the boundary carefully in the simulation. Even in the moderate laser intensity, the dynamics related wave function is still a small part of the total wavefunction. The high energy ATI or the rescattering wavepacket is just a few percent of this small part. Separation of this tiny part
from a huge background is still a challenge to the numerical precision of various time-dependent Schrödinger equation methods in a wide laser intensity region.

Recently we developed a new theoretical method to circumvent the above difficulties. Instead of solving the time-dependent Schrödinger equation, we solve a time integral equation which allows us to separate the dynamics related wavefunction from the background wavefunction analytically to improve the numerical accuracy. To solve the boundary problem, we separate the whole space into an inner region, in which most of the dynamics happens there, and an outer region, in which the electron-core Coulomb interaction can be ignored. We solve the problem in the inner-region numerically and propagate the outer region wavepacket in the momentum space analytically without a physical boundary in the real space. To show the effectiveness of the method, we present two examples here. In the first example, we study the carrier-envelope (CE) phase dependent ATI processes. Our simulated CE phase dependent ATI spectra are in good agreement with the experimental measurement by Paulus et al. [11] if we shift the experimental CE phase by $0.15\pi$. This means that we have re-calibrated the CE phase more accurately by comparing our simulations with the experimental measurements. In the second example, we investigate the rescattering wavepacket, which plays a central role to understand the most processes in the laser-atom interactions. Using the present method, we succeed in extracting the rescattering wavepacket, a tiny fraction of the total wavefunction, and reproducing the motion of the rescattering wavepacket predicted by the rescattering theory [6]. We introduce our theoretical method in Sec. II and present the two examples in Sec. III.

2. Theoretical Methods

In principle, all the physical processes in the laser-atom interactions can be studied by the following time-dependent Schrödinger equation (atomic units $\hbar = e = m = 1$ are used),

$$i \frac{\partial}{\partial t} \Phi(t) = H(t) \Phi(t)$$  \hspace{1cm} (1)

with time-dependent wavefunction $\Phi(\infty) = \Phi_0$. Here $\Phi_0$ is the ground state wavefunction of laser-field-free Hamiltonian $H_0$ and $H(t) = H_0 + V(t)$ with $V(t) = -\mathbf{r} \cdot \mathbf{E}(t)$ the laser-atom interactions and $\mathbf{E}(t)$ the laser field strength. Solving the above equation, we can study many physical processes, such as the ATI spectra and the HHG when the dynamically relevant wavefunction is comparable with or not too smaller than the background wavefunction. The above numerical method does not work in the perturbation limits due to the numerical problem. Instead of solving above equation, we propagate the time-dependent wavefunction in the integral form from time $T = \infty$ to $t$ as

$$\Phi(t) = -i \int_T^t U(t, \tau) V(\tau) U_0(\tau, T) \Phi_0 d\tau + U_0(t, T) \Phi_0,$$  \hspace{1cm} (2)

with

$$U_0(t, T) = e^{-i H_0 (t-T)} \quad \text{and} \quad U(t, \tau) = e^{-i \int_\tau^t H dt'}.$$

The first term in Eq. (2) stands for the initial wavefunction $\Phi_0$ propagating to time $\tau$ under $H_0$, and interacting with the laser field through $V$ at time $\tau$ (ionization) and then propagating to time $t$ under the full Hamiltonian. The second term stands for the initial wavefunction $\Phi_0$ propagating to time $t$ under $H_0$ and it does not contribute to any dynamical processes. Thus, we will focus on the dynamics related wavefunction defined as

$$\Psi(t) = -i \int_T^t U(t, \tau) V(\tau) U_0(\tau, T) \Phi_0 d\tau.$$  \hspace{1cm} (4)
The advantages of this separation are (a) that for a relatively weak field, the background wavefunction is factored out so that the numerical accuracy can be improved; and (b) that the creation and propagation processes are separated so that we can illustrate the physics more clearly. Transforming Eq. (1) into Eq. (2), we remove the background analytically and the dynamics related wavefunction can be calculated accurately even in the perturbative region.

Eq. (2) can be used to describe various physical processes, such as the laser-atom interactions [12] and the Coulomb three-body rearrangement [13,14]. Each process has its own characteristics and we have to choose the numerical procedure accordingly. The characteristics of the intense laser-atom interactions is that the active-electron parent-core interaction is dominant in the inner region, while the active-electron laser interaction is dominant in the outer region, and there are sufficient continuum contributions in $\Psi(t)$ region, while the active-electron laser interaction is dominant in the outer region, and there are sufficient continuum contributions in $\Psi(t)$. Thus we choose the following numerical scheme to study the laser-atom interactions. First we separate the space into two regions; the inner region ($0 < r < R_c$) and the outer region ($R_c \leq r < R_{\text{max}}$). The wavefunction is also separated into two parts, the inner-region wavefunction and the outer-region wavefunction. The inner region wavefunction is propagated in the real space through the full Hamiltonian $H(t)$ ($0 < r < R_{\text{max}}$), while the outer region wavefunction is propagated in the momentum space by the Volkov states. Thus the wavefunction can be expressed as the inner region wavefunction in the space and the outer region wavefunction in the momentum space. The procedure is equivalent to that we first propagate the wavefunction from inner-region to the outer region numerically, then propagate the outer-region wavefunction in the momentum space. All dynamical information can be obtained by analyzing $\Psi(t)$.

$$\Psi(t_j) = \Psi(t_j) \cdot (1 - F_s(R_c)) + \Psi(t_j) \cdot F_s(R_c) = \Psi_I(t_j) + \Psi_{II}(t_j).$$

Here, $F_s(r,R_c) = 1/(1 + e^{-(r-R_c)/\Delta})$ is a split function which separates the whole space into the inner ($0 \rightarrow R_c$) and outer ($R_c \rightarrow R_{\text{max}}$) regions smoothly and $\Delta$ represents the width of the crossover region. $\Psi_I$ represents the wavefunction in the inner region and it is propagated under the full Hamiltonian numerically. $\Psi_{II}$ stands for the wavefunction in the outer region and it is propagated under the Volkov Hamiltonian analytically. We first calculate

$$C(p,t_j) = \int \Psi_{II}(t_j) e^{-i(p-A(t_j)) \cdot r} \frac{1}{(2\pi)^{3/2}} dr,$$

with $A(t_j)$ the vector potential of the laser field. Then we propagate $\Psi_{II}$ from $t_j$ to $t$ as

$$\Psi_{II}(t,t_j) = \int U_v(t,t_j) C(p,t_j) e^{i p \cdot r} \frac{1}{(2\pi)^{3/2}} dp,$$

with the Volkov propagator $U_v(t,t_j)$ as

$$U_v(t,t_j) = e^{-i \int_{t_j}^{t} (p-A(t'))^2/2 dt'}.$$

The time-dependent wavefunction at time $t_i$ can be expressed as

$$\Psi(t_i) = \Psi_I(t_i) + \sum_{j \leq i} \int U_v(t_i,t_j) C(p,t_j) e^{i p \cdot r} \frac{1}{(2\pi)^{3/2}} dp.$$

The first term in the above equation represents the wavefunction in the inner region and the second part stands for the wavefunction in the momentum space. All dynamical information can be obtained by analyzing $\Psi(t)$. 












3. Results and Discussion

3.1. Carrier-envelop phase dependent high-energy ATI spectra

As the first example, we investigate the ATI spectra of Xe atoms in an ultrashort laser field \( E(t) = f(t) \cos(\omega t + \delta) \). Here \( f(t) \) is the laser pulse envelop function and \( \omega \) is the laser frequency and \( \delta \) is the CE phase. We use a model potential \[18\] to describe the valence electron (5p) interacting with the parent core. In the present simulation, we choose \( R_{\text{max}} = 200 \) a.u. and \( R_c = 100 \) a.u. The continuum wavefunction as \( t = \infty \) can be expressed as

\[
\Psi_c(\infty) = \sum_j \int U_v(\infty, t_j) C(\mathbf{p}, t_j) e^{i \mathbf{p} \cdot \mathbf{r}} (2\pi)^{3/2} d^3 \mathbf{p},
\]  

(10)

and the corresponding ATI electron momentum distribution is

\[
\frac{dP(\mathbf{p})}{dE d\Omega} = \sqrt{2E} \sum_j |U_v(\infty, t_j) C(\mathbf{p}, t_j)|^2.
\]  

(11)

Following the experiment \[11\], we present our simulated ATI momentum distribution by collecting the ATI yields on the right and left sides as

\[
\frac{dP_R(E)}{dE} = \int_{S_R} \frac{dP(\mathbf{p})}{dE d\Omega} d\Omega,
\]  

and

\[
\frac{dP_L(E)}{dE} = \int_{S_L} \frac{dP(\mathbf{p})}{dE d\Omega} d\Omega,
\]  

(12)

where \( S_R \) and \( S_L \) stand for the integration over the right and left half spheres. Here the right (left) side means the negative (positive) electric field direction.

Figure 1 shows our simulated ATI spectra as a function of the CE phase. Overall we see that in the low energy region (\( E < 10 \) eV), the ATI spectra from the right and left sides are in the same order and the ATI spectra are not sensitive to the CE phase. In the high energy region (\( E > 40 \) eV), the ATI yields are very sensitive to the CE phase. As shown in the figure, the ATI yields from the right side are much larger than those from the left side for \( \delta = 0 \). As we increase the CE phase from 0 to \( 0.6\pi \), the difference decreases and finally the yields from both sides are close to each other. As the CE phase increases further, the high energy ATI yields from the left side become larger than those from the right side as shown in Fig. 1. All the simulated results are in good agreement with the experimental results \[11\]. Figure 2 shows the ratio of ionization yields from the right and left sides at several photoelectron energies as a function of the CE phase. To compare with the experiment, the ratio of the total high energy (> 20 eV) ATI yields
Figure 2. (Colour online) The ratio of the ionization yields from the right and left sides for different photoelectron energies as a function of the CE phase. The laser parameters are the same as ones used in Fig. [1]. The solid triangles are the original experimental data [11]. The experimental CE phase shifted by 0.15π are also plotted (solid circles).

are also plotted (solid line). The solid triangles are the original experimental data [11] which are in reasonable agreement with our simulated ones (solid line). If we shift the experimental CE phase by 0.15π (solid circles), we find a better agreement between our simulations and the experimental ones. Thus the original experimental calibration of the CE phase should be shifted by 0.15π. As shown in Fig. [1] the ionization yields also depend on the photoelectron energy. If we use the higher energy ATI spectra as shown in Fig. [2] to calibrate the CE phase, we may get a more accurate CE phase. The high energy ATI yields drop dramatically. Thus it requires a more reliable and accurate numerical method to study the processes and our method serves the purpose.

3.2. Rescattering electron momentum spectra

The success of the ATI studies encourages us to study the rescattering electron momentum distribution. The rescattering theory [6] plays a central role to understand many dynamical processes in the intense laser material interactions. The physical process is rather simple. The electron first tunnels through the potential formed by the laser field with the electron-core Coulomb interaction when the laser field reaches its peak value. The electron is bounced back by the laser field when the field changes its direction. Although the physical picture is simple, a direct numerical observation of the rescattering wavepacket by a full quantum calculation is still not available due to the numerical difficulty. Since the new method circumvents the difficulties, we can investigate the rescattering processes now. Taking hydrogen atoms in the intense laser fields as an example, we illustrate how to study the time evolution of the rescattering wavepacket. In the present simulation, we choose the laser wavelength 800 nm and the laser intensity 10^{14} W/cm^2 to cope with the rescattering induced experiments [19, 20]. We also assume that the laser pulse duration is infinite. Thus, we recast the wavefunction Ψ(t) into

\[ Ψ(t) = \sum_j Ψ_j(t), \]  

with

\[ Ψ_j(t) = -i \int_{t_{j-1}}^{t_j} e^{-i \int_{t'}^{t} H(t')dt'} V(τ) e^{-iH_0τ} Φ_0 dτ. \]  

(13) (14)
Figure 3. (Colour online) (a) Wave function $|\Psi_j(t)|^2$ at $t = \pi/(2\omega)$ and (b) momentum distribution $|M(E, \Omega, t)|^2$ at the first returning time. The color bars are in a relative scale.

Here, $\Psi_j(t)$ corresponds to the electron interacting with the laser field directly for the first time in the interval from $t_{j-1}$ to $t_j$ and then propagating in the full Hamiltonian to time $t$. If we choose the time interval as a half cycle period, namely, $\omega t_j = (2j + 1)\pi/2$, the time-dependent wave function $\Psi_j(t)$ is the sum of each half cycle’s contribution. All the $\Psi_j(t)$s are identical with a proper time shift so we only present $\Psi_j(t)$ with $j = 0$.

Figure 3(a) shows a typical wave function immediately after the half-cycle ionization. We see that $\Psi_j(t)$ has two components in space. One represents the excitation in the inner region and the other stands for the tunnelling ionization in the outer region. The excited part will be ionized in the next half cycle when the laser field changes its direction. To illustrate the rescattering process without the contamination of this indirect ionization, we remove the excitation component from $\Psi_j(t)$ when the “direct” ionization is over. Watching the time evolution of $\Psi_j(t)$ [21], we notice that the electron wavepacket moves back and forth and goes out with the structures similar to the momentum distribution of low energy ATI spectra [22]. However, we cannot clearly distinguish the rescattering wavepacket from $\Psi_j(t)$. The visualization of $\Psi_j(t)$ does not provide the detailed information explicitly. Our goal is to study the rescattering wavepacket quantitatively, namely, the time-dependent momentum distribution of the rescattering wavepacket.

For such a purpose, we calculate the laser-field-free continuum wave function $\psi_l(E, r)$ for a given energy $E$ and a partial wave $l$ solving the Schrödinger equation numerically as

$$H_0\psi_l(E, r) = E\psi_l(E, r),$$

with imposing $r\psi_l(E, r) \to 0$ as $r \to 0$, and

$$\lim_{r \to 0} r\psi_l(E, r) = \sqrt{\frac{2}{\pi k}} \sin(kr + \frac{l\pi}{2} + \delta_l(E)).$$

Here $k = \sqrt{2E}$ and $\delta_l(E)$ is the phase shift. We project $\Psi_j(t)$ onto $\{\psi_l(E, r)\}$ states in the inner region when the wavepacket returns to the parent-core as

$$C_l(E, t) = \int (1 - F_s(r, R_c))Y_{lm}^*(\Omega)\psi_l^*(E, r)\Psi_j(t) dr.$$

In the present calculation, $R_c$ is 5 to 10 a.u. The results are not sensitive to the choice of $R_c$. With $C_l(E, t)$ and $\delta_l(E)$, we separate the wavepackets into an incoming spherical wave $\exp(-ikr)/r$ and an outgoing spherical wave $\exp(ikr)/r$ in the asymptotic region. The rescattering wavepacket is of the form of the incoming spherical wave. Defining

$$M(E, \Omega, t) = \sum_l \sqrt{i}C_l(E, t) e^{-i\delta_l(E)}Y_{lm}(\Omega),$$

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Figure 4. (Colour online) The rescattering energy and time distributions from (a) the quantum calculation and (b) the semiclassical calculation. The solid curves are from the classical model. The color coding represents the value of $\frac{d\bar{P}(E,t)}{dE}$.

the wave function of the rescattering electron in the asymptotic region with incident energy $E$ from incident direction $\Omega$ is $M(E,\Omega,t)\exp(-ikr)/r$. The rescattering momentum distributions at time $t$ is given by

$$\frac{dP(E,\Omega,t)}{dEd\Omega} = |M(E,\Omega,t)|^2.$$  \hspace{1cm} (18)$$

$M(E,\Omega,t)$ contains all the dynamic information of the rescattering wavepacket including the phase. If we can measure the dynamics due to the recollision of this wavepacket with the core, we can interrogate the core structure with the help of the calculated $M(E,\Omega,t)$.

Are the simulated results consistent with the intuitive physical picture of the rescattering theory? Figure 3(b) shows the rescattering momentum distribution at the first returning time. It does show that the electron is bounced back by the laser field from the right side and revisits the core with the rescattering energy around $4 \sim 5U_p$, with $U_p = E_0^2/(4\omega^2)$, higher than the classical one ($3.2U_p$). The time-dependent momentum distribution [23] shows that the electron moves back and forth in the momentum space. We notice that the rescattering momentum from the right side, which corresponds to the odd number returns, is higher than that from the left side, which corresponds to the even number returns. All these are consistent with the classical predictions and we do reproduce the intuitive classical pictures of the rescattering theory.

Next, we compare the percentages of the electron wavepacket coming back to the parent-core at time $t$ with energy $E$ calculated by the quantum and semiclassical methods [24]. The associated electron spectra are defined as

$$\frac{d\bar{P}(E,t)}{dE} = \frac{100}{A_i} \int \frac{dP(E,\Omega,t)}{dEd\Omega} d\Omega,$$ \hspace{1cm} (19)$$

with $A_i$ the ionization probability in a half-cycle period calculated by the corresponding method. Figure 4 shows the energy and returning time distributions of the quantum and semiclassical calculations. First, we see that only a few percent of the ionized wavepacket revisits the parent-core. Our calculated returning times are in good agreement with the classical and semiclassical ones and the returning time is model insensitive. Nevertheless, we observe a sharp contrast between the electron energy distributions by the quantum and semiclassical methods at the returning times as shown in Fig. 3. The quantum simulation shows the striking feature that the rescattering electron reaches the higher-energy regions ($E > 4U_p$) compared with the results by the other two simulations. This finding is of great importance for proper evaluations and
understandings of some rescattering induced processes, which mostly coincides with this energy region for the intensity $1 \times 10^{14}$ W/cm$^2$. For instance the rescattering induced dissociation process [20] is responsible for the rescattering electron energy near the border of the excitation energy. The high energy rescattering electron, which is missed in the semi-classical results, also responds to the generation of attosecond pulses [25] near the cut-off region. Therefore, reliable rescattering energy distributions are needed to decide if these processes are possible or not.

This example shows that the present method can also be used to study the rescattering processes in the inner region. As a matter of fact, there are many other processes in the laser-atom interactions that can be studied by the present method.

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