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Weakly relativistic electron dynamics and efficient X-ray photon emission driven by an ultraintense extreme-ultraviolet laser field

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ABSTRACT

The electron dynamics of a tightly bound system exposed to an ultraintense extreme-ultraviolet laser field is investigated by solving numerically the relativistically corrected two-dimensional time-dependent Schrödinger equation. The Coulomb interaction between the single-active electron and the parent ion is included throughout the harmonic generation process. The nondipole effects are found to be more pronounced with the increase of the ionization potential. The structure of the below-threshold harmonics changes drastically for different field intensities. The time-frequency analysis of the electron acceleration gives clearly the time-resolved harmonic spectrum and its dependence on the intensity of the pump field. With proper driving field intensities and ramp durations, hyper-Raman lines appear in the harmonic spectra for the soft-core potential. But due to multiple excitations of the atomic bound states, the hyper-Raman lines are spiked and broadened. When nearly full ionization occurs at the tail of the driving field, the HR lines disappear, while the emission of the odd harmonic lines is found to be most efficient then.

1. Introduction

The development of ultrafast laser technology brings the light-matter interaction into the strong field regime. One of the most prominent demonstrations of the nonlinear characteristics of the interaction of a strong laser field with matter is high-order harmonic generation (HHG) [1–3]. Atoms interacting with strong laser fields can emit coherent harmonic radiation at photon energies much higher than the atomic binding energy. As a promising way to produce coherent extreme-ultraviolet (XUV) and soft-X-ray radiation and attosecond pulses, high-order harmonic generation (HHG) has been widely investigated and well understood with the semi-classical three-step model [4,5] and quantum trajectory approaches [6,7]. Within the strong field approximation (SFA) [8], the electron dynamics is assumed to be dominated by the strong laser field and the influence of the Coulomb potential is neglected after tunneling. In terms of the three-step model, the electron first tunnels through the potential barrier formed by the atomic Coulomb potential and the laser field. It is then being accelerated in the laser field and gains kinetic energy. Later after some half of an optical cycle, the detached electron returns to its parent ion with some few probability, emitting a photon with the energy of the ionization potential of the atom plus the kinetic energy gained in the laser field.

The main drawback of HHG in gaseous target media is the low conversion efficiency. The spread of the electronic wave packet due to quantum diffusion decreases the recombination probability and reduces the single-atom HHG yield. The neutral gas and free-electron plasma dispersion leads to dephasing of the driving laser and the high-order harmonic fields during propagation and prevents the coherent enhancement of the harmonics generated from different atomic sites. In order to make HHG a practically useful source of coherent XUV light, efforts have concentrated on the raise of the conversion efficiency [9–11]. The enhancement of HHG can be achieved by, for example, tuning harmonic lines to atomic resonances, optimizing the focusing condition, adjusting the pressure in the gas cell, or applying a two-color laser field [12–17].

The highest energy of the harmonic photons is given by the microscopic single-atom cutoff rule $\omega_{max} = I_p + 3.17U_p$, where I_p is the ionization potential of the atom from which the harmonics are emitted, and $U_p = E_0^2/(4\omega_0^2)$ is the quiver energy of the liberated electron in a laser field of amplitude E_0 and frequency ω_0. Therefore, harmonic orders can be extended by direct or indirect increase of the field intensity. For example, it has been shown that the resonant surface plasmonic field can be used for HHG enhancement, and the confinement of the electron motion and the spatial inhomogeneity of the laser field lead to a significant increase of the cutoff [18–21]. Efficient frequency upconversion is possible up to the XUV region of the spectrum with the use of widely available Ti:sapphire lasers operating at an infrared wavelength. It was shown that ultrahigh harmonics spanning from the ultraviolet to

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a higher-energy soft X-ray region can be generated by guiding a mid-infrared femtosecond laser in a high-pressure gas, allowing in principle the generation of pulses as short as 2.5 as [22].

Improving the conversion efficiency and extending the photon energy of the radiation based on HHG are of great challenging due to the requirement on the pump field intensity and on the other hand the restriction of the relativistic effects caused by the ultraintense driving fields [23,24]. According to the above mentioned cutoff law, the harmonic spectrum can be extended up to thousands of harmonics with ultraintense field exposing. But in this case the relativistic effects become important. The relativistic drift in the electron motion suppresses the harmonic generation and imposes limitations on the achievable photon energies. The higher the kinetic energy of the released electron, the stronger the relativistic effects, which means that the high-energy part of the spectrum will be more intensively suppressed than the low-energy part.

The low-energy part of the spectrum is governed by the inner atomic dynamics. For the below- and around-threshold harmonics, the electron dynamics is strongly influenced by the atomic potential. In this case, the three-step model becomes insufficient to describe the dynamics of the electrons [25–27], and the strict solutions of time-dependent Schrödinger or Dirac equations are needed [23]. It has been shown that the major contribution to below-threshold harmonics (BTHs) comes from a specific trajectory corresponding to the electron moving straight back to the atomic core from the tunneling exit [28,29]. The quantum yield of the above threshold harmonics is severely limited by the tunneling rate and the recombination probability of the electronic wave packet. Since the BTHs are mostly generated inside the atom, involving a much larger part of the electronic wave packet, significantly higher harmonic generation probability is expected.

The photon energy of the below-threshold harmonics is limited by the ionization potential energy. For neutral atoms, it can reach only up to several electron volts. In order to get high energy X-ray photon emission from BTH generation, systems with high ionization potentials, such as highly charged ions [30,31], are needed. For highly charged ions (HCIs) with an ionization potential of hundreds to thousands of electron volts [32,33], according to the cutoff law, the harmonic spectrum may easily extend up to several ten thousand harmonics. Already the below-threshold part of the spectrum covering some hundred harmonics may well reach into the X-ray region, and the spectrum can be further extended to higher X-ray energies by using higher-Z elements.

In this paper, we investigate the weakly relativistic electron dynamics and below-threshold harmonic generation in a system of a high ionization threshold exposed to an ultraintense extreme-ultraviolet laser field by solving directly two-dimensional (2D) time-dependent Schrödinger equation (TDSE), in which the Coulomb interaction between the active electron and the remaining ion is automatically included throughout the harmonic process. The relativistic corrections are taken into account by expanding the Klein–Gordon Hamiltonian in the velocity gauge up to the second order in the ratio of the electron velocity to the speed of light. The same type of modeling has been used in our previous publication [34] for the study of the BTH generation processes pumped by the strong weakly relativistic laser field. Calculations in [34] show that generation of the BTH spectra mainly an inner atomic process are not sensitive to the simulation box size, and the agreement between the 2D and 3D results indicates that the electron dynamics in BTH generation is mainly in two dimensions determined by the polarization and the propagation directions of the field, so it is enough to solve the two-dimensional Schrödinger equation. In this paper we study the pump field intensity dependence of the electron dynamics and of the emission of the harmonic spectra. It is found that the structure of the harmonic spectra changes drastically for different field intensities. The harmonic spectra show also hyper-Raman (HR) lines at proper levels of the pump field intensity. This means that HR lines can be obtained not only in a simple two-level system, but also in systems with complex energy structures under proper pump conditions. Due to multiple excitations of the atomic bound states in the soft-core potential, the HR lines are broadened with several harmonic spikes. The strength and positions of the hyper-Raman lines are very sensitive to the parameters of the pump field, such as the field intensity and the turn-on ramp duration.

The outline of our work is as follows. Section 2 presents the theory used for the study of the weakly relativistic electron dynamics and the generation of the harmonics, including the weakly relativistic Schrödinger equation (Section 2.1), the equations for the electron acceleration and the harmonic spectra (Section 2.2), and the numerical scheme. All the formulae are given in atomic units throughout the paper unless otherwise mentioned. The results of the numerical simulations are presented and discussed in Section 3. Our findings are summarized in Section 4.

2. Theory

2.1. Weakly relativistic Schrödinger equation

According to Newton's second law of motion, the dynamics of an electron exposed to a sine-wave field is determined by equation $dv/dt = -E_0 \sin(\omega_0 t)$. The integration of the motion equation yields $v(t) = v_m \cos(\omega_0 t)$ with $v_m = E_0/\omega_0$. Therefore, when conditions of the field render the dimensionless field parameter $\xi = v_m/c = E_0/\omega_0 c \geq 1$, one must abandon the non-relativistic treatment, and adopt the fully relativistic treatment [35,36]. However, relativistic effects already start to play role when the relativistic parameter is smaller than one. In the weakly relativistic regime with ξ few times smaller than 1, which is the regime considered in this paper, the main deviations from the nonrelativistic dynamics are the drift caused by the magnetic component of the laser field, the breakdown of the dipole approximation, and the relativistic mass shift [37,38]. By subtracting the rest energy of the electron, we expand the fully relativistic Klein–Gordon Hamiltonian $H(\underline{\mathbf{r}}, t) = \sqrt{c^2(\underline{\mathbf{p}} + \underline{\mathbf{A}}/c)^2 + c^4} - c^2 + V(r)$ up to the order of $1/c^2$, and neglect the higher-order corrections. Therefore, the relativistic time-dependent Hamiltonian describing the motion of the active electron in the HCI system exposed to a strong laser field in the velocity gauge is given by

$$H(\underline{\mathbf{r}}, t) = \frac{1}{2}\left[\underline{\mathbf{p}}(\underline{\mathbf{r}}, t) + \frac{\underline{\mathbf{A}}(\underline{\mathbf{r}}, t)}{c}\right]^2 - \frac{1}{8c^2}\left[\underline{\mathbf{p}} + \frac{\underline{\mathbf{A}}(\underline{\mathbf{r}}, t)}{c}\right]^4 + V(r), \qquad (1)$$

where $\underline{\mathbf{p}}(\underline{\mathbf{r}}, t)$ is the canonical momentum operator, $\underline{\mathbf{A}}(\underline{\mathbf{r}}, t)$ is the vector potential of the driving laser field, and $V(r)$ is the atomic Coulomb potential. The second term in the right-hand-side of Eq. (1) is the relativistic mass shift. For the linearly polarized laser field involved here which is assumed to be polarized in x axis and propagates along z direction, the dynamics of the electron induced by the laser field is mainly confined in the (x, z) plane. Therefore, it is enough to perform a two-dimensional (2D) numerical integral of the TDSE in the (x, z) plane.

The response of the tightly bound system to the linearly polarized laser field is simplified by the single-active electron approximation. The 2D "soft-core" potential

$$V(x, z) = -\frac{q}{\sqrt{x^2 + z^2 + a^2}} \qquad (2)$$

is employed to model the Coulomb field experienced by the active electron. The constant parameter q is related with the charge of the ionic core, and a removes the singularity of the potential. The choice of q and a determines the depth of the potential and its smoothness