High-order harmonic generation with Rydberg atoms by using an intense few-cycle pulse

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We demonstrate that high-order harmonic generation (HHG) with both high cutoff frequency and high conversion efficiency can be realized by using a Rydberg atom in a few-cycle laser pulse. This is because a Rydberg state has a large electron orbital radius and small binding energy; therefore an electron in the Rydberg state can be ionized easily and accelerated directly toward the core under the interaction of a few-cycle laser pulse, leading to emission of harmonic photons. In this case, the tunneling process of the electron is not involved and, hence, the conversion efficiency and the cutoff frequency of harmonic generation can be higher than that predicted by the conventional three-step model.

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I. INTRODUCTION

High-order harmonic generation (HHG) has been the subject of intense theoretical and experimental studies since its discovery in the late 1980s [1,2]. Harmonic emission is now becoming a very useful source of coherent light in the extreme ultraviolet and soft x-ray regions of the spectrum (see, e.g., Refs. [3,4] for recent reviews). It is also the starting point for the generation of isolated or a series of attosecond pulses that has potential applications in ultrafast spectroscopy [5–8] of atoms, molecules, and materials. HHG is commonly described by a recollision model [9–11] that involves three steps: (1) the tunneling ionization of a bound-state electron, (2) the acceleration of the free electron in the laser field, and (3) the recombination of the electron with its parent ion producing high harmonics. Recently, it has been found that HHG can be used for imaging the structure of atomic wave functions [12,13] and molecular orbitals [14]. For example, we found that the electronic density distribution of a Rydberg state can be probed directly using a broadband harmonic spectrum because of the one-to-one correspondence relationship between the initial position of the electron and the harmonic order [12].

There are two important aspects in HHG that we need to be concerned about, i.e., the cutoff frequency of the harmonic spectrum and the conversion efficiency of the harmonic generation. The cutoff frequency is predicted by the cutoff law, $I_p + 3.17U_p$ [10], where I_p and U_p are the atomic binding and electron ponderomotive energies in the laser field, respectively. On the other hand, if the gas medium is ionized before the arrival of the peak of the laser pulse, the maximum value of U_p is limited by the saturation of ionization regardless of the peak intensity of the laser pulse. U_p can be increased by using atoms with large ionization potential, allowing atoms to remain neutral to a higher laser intensity before being ionized. Since the saturation intensity of ions is high, the harmonic emission from ions can, in principle, be extended to very high energies. For example, Gibson *et al.* [15] have demonstrated the generation of very high order harmonics, up to 250 eV, using Ar ions. However, the HHG conversion efficiency is usually low because of the small ionization probability of the ions. The increase of harmonic conversion efficiency by preparing the initial state as a coherent superposition of two bound states has been studied in Refs. [16–18]. The role of the excited state is to enhance the ionization probability, thus increasing the conversion efficiency.

In this paper, we investigate HHG in an ultrashort laser pulse with the initial state being prepared as a superposition of the ground state and a Rydberg state. The proposed scheme has the advantage of producing high-order harmonic photons and, at the same time, maintaining a high conversion efficiency. The primary motivation of using a Rydberg state is that, since the Rydberg electron has small binding energy and is far from the core, it can be ionized easily and accelerated directly toward the core under the interaction of a few-cycle laser pulse. As a result, the energy of the emitted photon can be beyond the $I_p + 3.17U_p$ cutoff law predicted by the three-step model. This is in sharp contrast with the conventional three-step process, where the electron has to be tunnel-ionized first and then propagate away from the core before it is driven back by the laser field. Moreover, since the Rydberg electrons participate directly in the harmonic generation, the conversion efficiency of HHG with Rydberg atoms can be much higher than that of the conventional recollision model.

II. NUMERICAL METHOD

We investigate the interaction of a one-active electron atom with a few-cycle laser pulse by solving the three-dimensional time-dependent Schrödinger equation (TDSE) (in which atomic units are used throughout, unless otherwise stated),

$$i\frac{\partial}{\partial t}\psi(\mathbf{r},t) = \left[-\frac{1}{2}\nabla^2 - \frac{Z_{\text{eff}}}{r} + \mathbf{r}\cdot\mathbf{E}(t)\right]\psi(\mathbf{r},t),$$

where the effective nuclear charge $Z_{\text{eff}} = \sqrt{2I_p} = 1.2592$ with I_p being the ionization threshold of the ground state of Ne. The laser electric field $\mathbf{E}(t)$ is along the z axis. The time propagation of the wave function is performed by integrating the TDSE in the spherical coordinate system using the finite-difference

technique [19]. Our integration grid is confined in a finite space $r < 600$. In order to avoid the reflection of the wave packets from the boundaries, a $\cos^{1/8}$ mask function, which varies from 1 to 0, is utilized in the range of 500–600 after each time interval [12,20].

III. HHG BY A RYDBERG ATOM

The initial state is prepared as a superposition of the ground state $|1s\rangle$ and a Rydberg state $|np\rangle$, i.e.,

$$\psi(r,t \to -\infty) = \frac{1}{\sqrt{2}}(|1s\rangle + |np\rangle). \quad (1)$$

Here, n is the principal quantum number of the Rydberg state and the phase difference between the two component states is set to be zero for simplicity. We note that such a superposition state may be achieved directly by a one-photon resonant excitation process. Moreover, Avetissian *et al.* [21] suggested that such a superposition state can also be achieved by a multiphoton resonant excitation of an atom by an appropriate optical pulse with a moderate strong intensity. Recently, they have also predicted that various coherent superposition states in atomic or ionic systems with high nuclear charges can be obtained by a multiphoton excitation based on an analytical solution of the Dirac equation [22]. Another effective method to obtain the superposition state is the coherent control of the atomic states by electromagnetically induced transparency [23].

We first consider the HHG spectra for a laser pulse with $E(t) = E_0 \sin^2(\pi t/\tau)\sin(\omega t + \varphi)$, where the pulse duration is $\tau = 5$ fs and the central frequency is $\omega = 0.056$ a.u. φ is the carrier envelope phase (CEP). Figure 1 presents HHG spectra when the peak laser intensity is 5×10^{14} W/cm^2 and the initial states are the superposition states $\frac{1}{\sqrt{2}}(|1s\rangle + |np\rangle)$ with $n = 3$ (dot-dashed line), 6 (solid line), and 9 (dot-dot-dashed line). For comparison, we also present the result when the initial state is the ground state only (dashed line). Our results indicate that the cutoff of the HHG spectrum for the ground state is 44ω, whereas for the superposition state with $6p$ the cutoff extends to 96ω, corresponding to $I_p + 4.03U_p$. Further increase of n does not change the cutoff frequency (for example, $9p$ in Fig. 1), but only lowers the conversion efficiency of HHG. Figure 2(a) presents the cutoff frequency of the harmonic spectrum as a function of the principal quantum number n. It shows that the value of the cutoff frequency increases with n at first and then reaches its maximum at $n = 6$. This indicates that the optimal number for HHG is $n = 6$ in our present atom-laser condition. We also find that the HHG spectra with Rydberg atoms are sensitive to the laser condition. Figure 2(b) presents the cutoff frequency for the superposition state with $6p$ as a function of the pulse duration (solid cycles). For comparison, we also present the result for the ground state (open triangles). It shows that the extension of the cutoff due to the presence of a Rydberg state is evident only when the pulse duration is less than two optical cycles. For longer pulses, the cutoff converts to the conventional $I_p + 3.17U_p$ cutoff law, which is attributed to the contribution from the ground state. We then study the CEP effects on the HHG spectra. Figure 2(c) presents the CEP dependence of the cutoff frequency when the initial state is a superposition state with $6p$, the laser peak intensity is 5×10^{14} W/cm^2, and the pulse duration is 5 fs. The cutoff frequency varies from $I_p + 4.03U_p$ (96ω) to $I_p + 2.1U_p$ (57ω) as the CEP changes from 0 to 0.4π. Finally, Fig. 2(d) shows the cutoff frequency as a function of the laser intensity with the CEP being zero. It indicates that the cutoff of the HHG spectrum decreases from $I_p + 4.4U_p$ to $I_p + 4.0U_p$ as the intensity increases from 10^{14} to 10^{15} W/cm^2 with the pulse

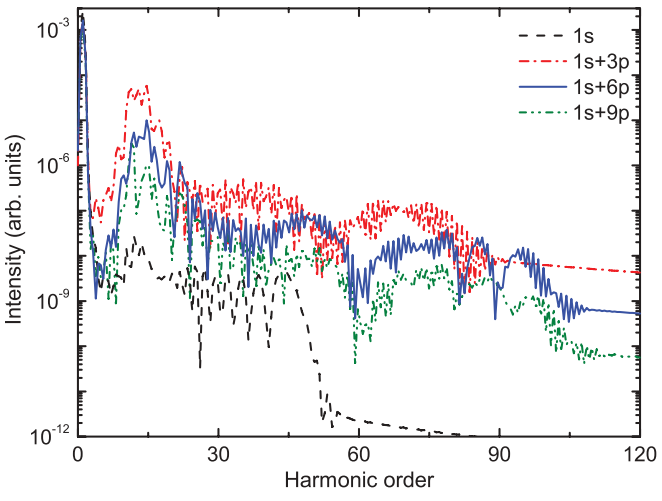

FIG. 1. (Color online) Harmonic spectra from np Rydberg atoms with $n = 3$ (dot-dashed line), 6 (solid line), and 9 (dot-dot-dashed line) by a 5-fs 800-nm laser pulse. The ionization threshold of the ground state of the model atom is the same as that of the Ne atom. The peak intensity of the pulse is 5×10^{14} W/cm^2. For comparison, the harmonic spectrum from the ground-state atom (dashed line) is also included.

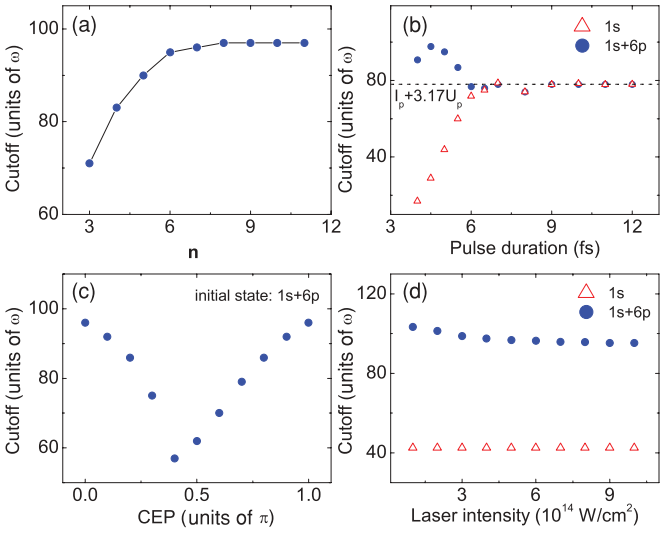

FIG. 2. (Color online) (a) Cutoff frequency of the harmonic spectrum vs the principal quantum number n of the state np with the same laser condition as in Fig. 1. (b) Cutoff frequency as a function of the pulse duration with a wavelength of 800 nm and an intensity of 5×10^{14} W/cm^2. (c) Cutoff frequency of the HHG spectrum vs the carrier envelope phase (CEP). (d) Cutoff frequency vs the laser intensity with a pulse duration of 5 fs. The wavelength is 800 nm and the CEP is zero.

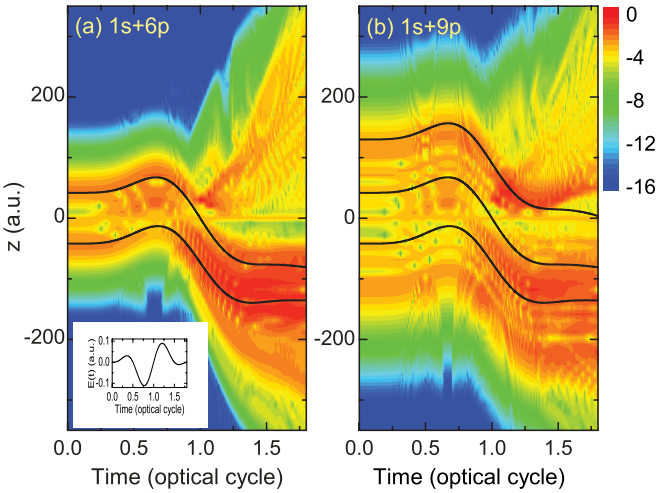

FIG. 3. (Color online) Time evolution of the electron density distribution on the z axis for the superposition state with $6p$ (a) and $9p$ (b). The solid lines are typical classical trajectories. The inset in (a) shows the laser electric field with a duration of 5 fs. The results are plotted on a logarithmic scale.

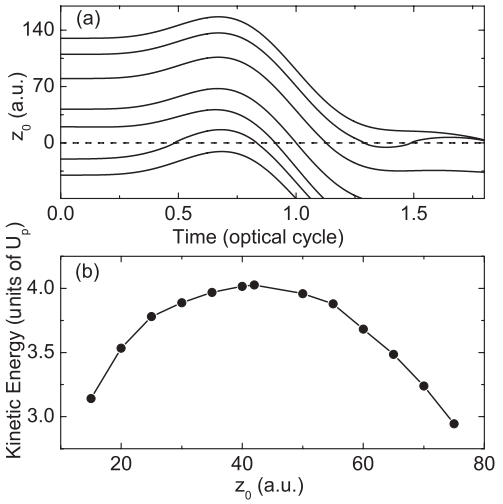

FIG. 4. (a) Classical trajectories of electrons under the combined interaction of the Coulomb and laser fields (solid line). (b) The kinetic energy of the electron when it collides with the core vs the initial position z_0. The laser parameters are the same as those in Fig. 3.

duration being 5 laser cycles. In contrast, the cutoff frequency for the ground-state case remains at $I_p + 1.5U_p$ as the intensity changes.

To clarify the mechanism of HHG with Rydberg atoms, we study the time evolution of the electron probability density in a few-cycle laser pulse with a duration of 5 fs and peak intensity of 5×10^{14} W/cm^2. Figure 3 presents the contour plots of the time-dependent electron density distribution on the z axis for $6p$ [Fig. 3(a)] and $9p$ [Fig. 3(b)]. It shows a quite different picture from that of the three-step model of HHG. Specifically, the Rydberg electron, which spreads over a wide region, is weakly bounded by the ion core. When a few-cycle laser pulse is applied, the electron will move by the combined interaction of the Coulomb and laser fields. Then the electron initially located in a certain region will be accelerated toward the core by the laser field. When the electron collides with the core, the harmonic photon is emitted by the coherent oscillation between the returning electron wave packet and the ground-state wave function. The energy of the harmonic photon can be larger than $I_p + 3.17U_p$ predicted by the three-step model. On the other hand, most electrons will undergo a collision and finally leave the core. Therefore, unlike the three-step process of HHG in a long-duration laser pulse, in our case, the collision between the electron and the core occurs only once during the HHG process.

We now investigate how the principal quantum number n affects the HHG cutoff. The average distance between the Rydberg electron and the core increases with n. For $n < 6$ the returning electron can acquire higher kinetic energy from the field as the initial distance increases, leading to a higher cutoff frequency. Now, let us compare the contour plots of the electron probability densities for $6p$ and $9p$. For the former case [Fig. 3(a)], initially the electron spreads in a region of $|z_0| \lesssim 100$ whereas the electron located in the region $z_0 > 0$ will be driven to the core and will contribute to the harmonic generation. By contrast, for the latter case, $9p$ [Fig. 3(b)], although the electron spreads over a wider region of $|z_0| \lesssim 150$, only the electron located initially in the region $0 < z_0 \lesssim 100$ can be driven to the vicinity of the core. Therefore, the cutoff frequencies are about the same for $6p$ and $9p$. However, since the probability of the electron being driven to the core is less for $9p$, the conversion efficiency of HHG decreases (see Fig. 1).

HHG with Rydberg atoms can be understood by a simple classical model. We calculate the trajectory of an electron under the combined interaction of the Coulomb and laser fields by using the Newtonian equation. The initial position of the electron is assumed to be z_0, while the initial velocity is assumed to be zero because the Rydberg electron has very small kinetic energy. Also, the elastic collision between the electron and the core is ignored in our calculations. Figure 4(a) presents the classical trajectories that show that only electrons initially located at $0 < z_0 \lesssim 100$ can be driven back to the core and thus contribute to the harmonic generation. Especially, the electron with initial position $z_0 = 42$, where $z_0 = 42$ is the most probable position of the Rydberg state 6p [Fig. 3(a)], can collide with the core at the moment of 1.0 optical cycle and acquire the highest kinetic energy from the field. This is consistent with the quantum calculations (see Fig. 3). The cutoff of HHG with Rydberg atoms can also be predicted by the classical model. Figure 4(b) presents the kinetic energy, E_k, of the electron when it collides with the core versus the initial position z_0. It shows that E_k increases with z_0 until $E_k = 4.03U_p$ for $z_0 = 42$, corresponding to the energy $I_p + 4.03U_p$ of the harmonic photon. This is because for trajectories with initial position smaller than 42 the acceleration time of the electrons by the field is longer for larger z_0. On the other hand, electrons with initial distance $z_0 > 100$ cannot return to the core and, hence, have no contribution to HHG, as shown in Fig. 3(b) for the case of $n = 9$. We note that since the electron trajectories directly respond to the time evolution of the electric field, the cutoff is sensitive to the CEP, as shown in Fig. 2(c). In Fig. 4 we set the CEP of the laser pulse to zero.

Another important issue in HHG is the conversion efficiency of the harmonic generation. It is well known that in the

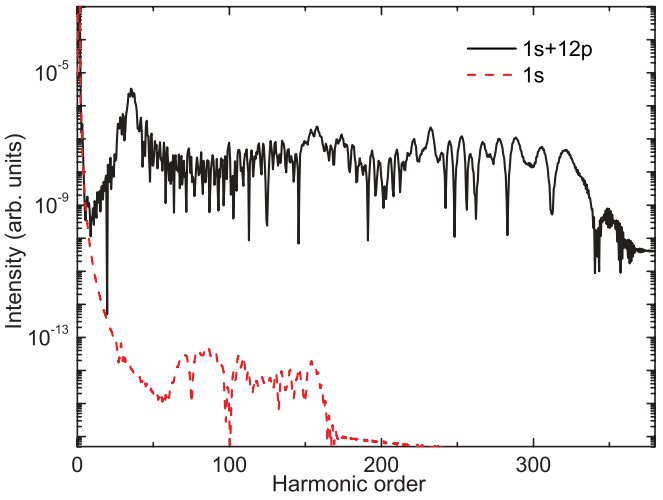

FIG. 5. (Color online) Harmonic spectrum of the He^+ ion with the superposition state of the ground state and the Rydberg state $12p$ (solid line) when the laser peak intensity is 3×10^{15} W/cm^2 and the pulse duration is 5 fs with the CEP being zero. The harmonic spectrum for the ground state is also presented (dashed line).

conventional HHG process only ionized electrons are involved in the harmonic generation; thus, the conversion efficiency is determined by the ionization rate of the atom. By contrast, in HHG with Rydberg atoms, all electrons located initially in a relatively wide region will participate in the harmonic emission [see Fig. 3(a)]. Therefore the conversion efficiency will be higher if initially there is a large coherence between the ground state and the Rydberg state, and the laser intensity does not cause population depletion of the ground-state. This feature is more evident if atoms with large I_p are used. Figure 5 presents the harmonic spectrum of the He^+ ion with the superposition state of the ground state and the Rydberg state $12p$ when the laser peak intensity is 3×10^{15} W/cm^2 and the pulse duration is 5 fs. For comparison, we also present the HHG spectrum when the initial state is the ground state and the laser has the same peak intensity and pulse duration. Compared to the conventional three-step model, HHG with Rydberg atoms has both higher cutoff frequency and higher conversion efficiency.

One important application of HHG is the production of pulses with attosecond duration [8,24–26]. Attosecond pulses are generated during electron-ion collisions. Thus, the duration of the attosecond pulse is largely determined by the kinetic energy of the electron. Since HHG with Rydberg atoms has both higher cutoff frequency and higher conversion efficiency, we expect that attosecond pulses of shorter duration can be generated with higher efficiency. Also, a single attosecond pulse can be produced because the collision between the electron and the core occurs only once during the HHG process. Finally, when the Rydberg electron collides with the core, besides the harmonic generation, it may free another bound electron in the core [27]. This process is similar to nonsequential double ionization, which has been studied in the framework of electron recollision. Our method can also be applied to molecules, which are interesting because of the presence of nuclear motion.

IV. CONCLUSION

In summary, we have proposed a method of HHG with the initial state being prepared as a superposition of the ground state and a Rydberg state. The mechanism of HHG with Rydberg atoms is different from that of the conventional three-step model of HHG. Specifically, in the three-step model, the key step is the tunneling ionization of a bound-state electron because only after ionization can the electron acquire high kinetic energy from the laser field for the subsequent recollision. By contrast, in our case because the Rydberg electron has small binding energy and is already far from the core, it can respond directly to the few-cycle laser pulse. As a consequence, the energy of the emitted photon can be beyond the $I_p + 3.17U_p$ cutoff law of the three-step model. Moreover, since HHG with Rydberg atoms does not rely on tunneling ionization, the conversion efficiency can be high if a large coherence between the ground and the Rydberg states exists initially. We note that although some methods have been proposed to produce high harmonics with energies beyond the cutoff law [28,29], they are always accompanied by low conversion efficiency. Also, HHG with coherent superposition of the ground and low excited states has been proposed to increase the conversion efficiency of the harmonic generation [16–18]. However, since this scheme is still based on the three-step model, the cutoff frequency cannot be extended. Compared to these methods, the advantage of using Rydberg atoms in HHG is that we can obtain both higher cutoff frequency and higher conversion efficiency. Moreover, single attosecond pulses can be produced.

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