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Generation of circularly polarized multiple high-order harmonic emission from two-color crossed laser beams

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We present a scheme for the production of circularly polarized multiple high-order harmonic generation (HHG). The proposed experimental setup involves the use of two-color laser fields, consisting of a circularly polarized fundamental laser field and a linearly polarized second-harmonic laser field, in crossed-beam configuration. The feasibility of such a scheme is demonstrated by an *ab initio* quantal study of the HHG power spectrum of He atoms by means of the time-dependent density-functional theory with optimized effective potential and self-interaction correction recently developed. The theoretical study also provides insights regarding the different mechanisms responsible for the production of HHG in different energy regimes as well as the mechanism for the generation of continuous background radiation. [S1050-2947(98)50410-6]

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Multiple high-order harmonic generation (HHG) is one of the most rapidly developing topics in the field of laser-atom interaction. The generation of harmonics of orders well in excess of 100, from noble-gas targets, has been demonstrated by a number of experiments using high intensity pump lasers [1,2]. The shortest wavelength generated by the HHG mechanism demonstrated today is approximately 2.7 nm [3], using ultrafast pulses (26-fs pulse duration) from a Ti:sapphire laser centered at 800 nm. Most of the investigations of HHG so far have focused on the production of HHG using one laser field. Recently, there also has been growing interest, both experimentally [4] and theoretically [5], in the exploration of the feasibility of coherent control of HHG by means of two-color or mixed laser fields. All of the previous studies, however, were concerned only with the production of *linearly polarized* (LP) HHG. In this Rapid Communication, we propose a scheme for the generation of *purely circularly polarized* (CP) HHG, which can be performed within the current experimental capabilities. The scheme involves the use of two-color laser fields, consisting of a CP fundamental laser field and a LP second-harmonic field, in crossed-beam configuration. We demonstrate the feasibility of such a scheme by performing an *ab initio* calculation of HHG in He by means of the *time-dependent density-functional theory* recently developed [6].

The proposed crossed-beam setup is shown in Fig. 1: a CP laser beam (with fundamental frequency $\omega_c = \omega$) is oriented towards the z direction, and a LP second-harmonic laser beam (with frequency $\omega_L = 2\omega$) is oriented towards the x direction with the electric polarization along the z direction. With such an experimental configuration, one can obtain purely CP high harmonic generation emitted along the incident CP laser beam (z) direction and purely LP high harmonic generation emitted along the incident LP laser beam (x) direction.

The mechanism for the production of purely CP HHG is illustrated in Fig. 2. Due to the dipole selection rule, the HHG can only be emitted from a (real or virtual) excited state with magnetic quantum number $m=0, \pm 1$, if the initial state is at the $m=0$ level. To reach the $m=0$ excited level, the ground-state electron can only absorb LP photons. To reach the $m=1$ excited level, the ground-state electron can only absorb an even number of LP photons and one CP photon. [Without the loss of generality, we shall assume that the incident CP laser field has the *right* circular polarization (RCP) in this paper.] To reach the $m=-1$ excited level, the electron can only absorb an even number of LP photons and emit one CP photon. Once the electron is in the excited state, there are three possible routes for the emission of HHG in the crossed-beam configuration: (a) The excited electron in the $m=0$ state can emit $(2n+1)\omega_L = (4n+2)\omega$ LP HHG in the incident LP laser beam (x) direction; (b) the excited electron in the $m=1$ state can emit $(2n)\omega_L + \omega_c = (4n+1)\omega$ RCP HHG along the incident CP laser beam (z) direction; and (c) the excited electron in the $m=-1$ state can emit $(2n)\omega_L - \omega_c = (4n-1)\omega$ left circularly polarized (LCP) HHG in the z direction. Note that the above discussion refers to the leading channel only. Thus, the net absorption of n photons may include higher-order processes, such as the absorption of $(n+n')$ photons and the emission of n' photons, etc. We also note that while the schematic diagram

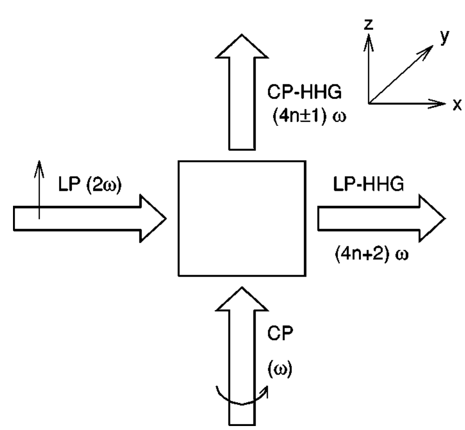

FIG. 1. The proposed crossed laser beam scheme for the production of circularly polarized multiple high-order harmonic generation.

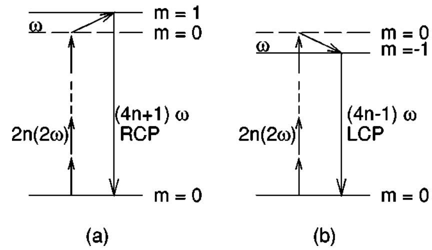

FIG. 2. Schematic diagram to show the leading channels for the production of CP HHG.

in Fig. 2 refers mainly to the *multiphoton* mechanism for the production of CP HHG, the same selection rules apply to the case of HHG due to the *tunneling* mechanism.

To demonstrate the feasibility of the proposed scheme for the production of purely CP HHG, we have performed an *ab initio* quantal study of the HHG of He atoms in the crossed-beam configuration by means of the newly developed *time-dependent density-functional theory* (TDDFT) with *optimized effective potential* (OEP) and *self-interaction correction* (SIC) [6]. The three-dimensional (3D) TDDFT/OEP-SIC equations can be solved numerically, accurately and efficiently, using the *time-dependent generalized pseudospectral* method in *energy representation* [7]. As illustrated in recent works [6], the TDDFT/OEP-SIC formalism provides a powerful nonperturbative approach for the treatment of multiphoton processes of many-electron systems in strong fields, taking into account the effects of electron correlations and inner-shell excitations. We refer to Refs. [6, 7] for the technical details of the procedures.

The laser parameters used in the present work are as follows. The wavelength for the fundamental CP field used is 1064 nm, and the wavelength for the second-harmonic LP field is 532 nm. The laser intensity for the LP field is fixed at $I_2 = 2\times10^{14}$ W/cm^2, and several CP laser intensities are considered: $I_1 = 0$, 2×10^{12}, 2×10^{13}, and 2×10^{14} W/cm^2. The laser field pulse shape used is $\sin^2(\pi t/T)$ for both LP and CP laser beams with $T = 90$ optical cycles of the fundamental field.

We first show the results of the LP HHG power spectra in Figs. 3(a)–3(d), corresponding to the incident CP laser intensities $I_1 = 0$, 2×10^{12}, 2×10^{13}, and 2×10^{14} W/cm^2, respectively. These harmonics are emitted along the incident LP laser beam (x-) direction and can only be generated by the induced dipole moment in the z direction, $d_z(t)$. Figure 3(a) corresponds to the case of one-color HHG by the incident LP field only (with wavelength 532 nm) and displays the characteristic HHG pattern: initial decrease of the harmonic intensity for the first few harmonics, followed by a plateau of many harmonics of similar intensity and then a sharp cutoff. For a comparison with the crossed-field results, we duplicate the one-field HHG spectra by filled circles in Figs. 3(b)–3(d). When the fundamental CP field intensity is relatively weak [Fig. 3(b)], we see that the LP HHG power spectra do not exhibit significant changes, apart from some broadening and enhancement beyond the cutoff regime. As the CP laser intensity is further increased, we see significant suppression of the background radiation intensity in the emitted light and in the HHG peak intensities in the ‘‘plateau’’ regime [see, particularly, Fig. 3(d)]. However, the peak intensities of the lower harmonics seem to be only slightly affected by the

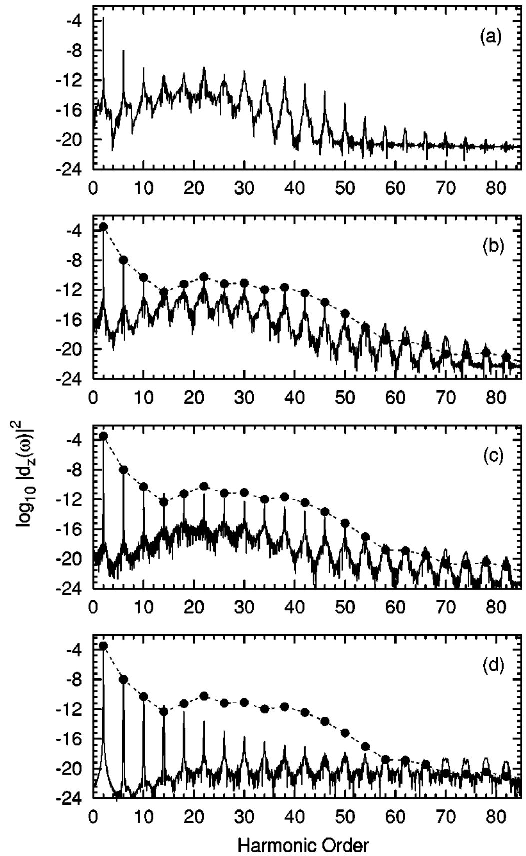

FIG. 3. Linearly polarized HHG power spectra of He atoms in a one-color LP laser field (a) and in crossed-beam configuration (b)–(d). The incident CP fundamental laser wavelength is 1064 nm and the peak intensity of the incident LP second-harmonic field (532 nm) is fixed at $I_2 = 2\times10^{14}$ W/cm^2. (a)–(d) correspond to incident CP field intensity $I_1 = 0$, 2×10^{12}, 2×10^{13}, and 2×10^{14} W/cm^2, respectively. For comparison, the one-color results (denoted by filled circles) are also included in (b)–(d).

strength of the incident CP field. Also, the enhancement of the highest harmonics beyond the cutoff regime seems to persist.

Figures 4(a)–4(c) show the results of CP HHG power spectra emitted along the incident CP laser beam (z-) direction, corresponding to $I_1 = 2\times10^{12}$, 2×10^{13}, and 2×10^{14} W/cm^2, respectively. For comparison, we also include the LP HHG power spectra for the one-field case (denoted by filled circles). Note that the CP HHG can only be produced by the x-y components of the induced dipole moments $d_{xy}(t)$. Figures 4(a)–4(c) show that purely CP HHG can indeed be produced by means of the crossed-beam configuration. Assuming that the incident CP field is of right circular polarization, the emitted RCP HHG can only occur at the $(4n+1)\omega$ positions, whereas the LCP HHG can only occur at the $(4n-1)\omega$ positions. When the intensity of the incident CP field I_1 is increased, several salient features of the power spectral changes are observed. First, we see that both the *lower* CP harmonics and the *highest* harmonics above the cutoff regime are enhanced with increasing incident CP field intensity I_1. Second, the CP HHG intensity in the plateau regime first increases and then decreases with increasing incident CP field intensity, and there is an optimal I_1 intensity range somewhere between 2×10^{12} and

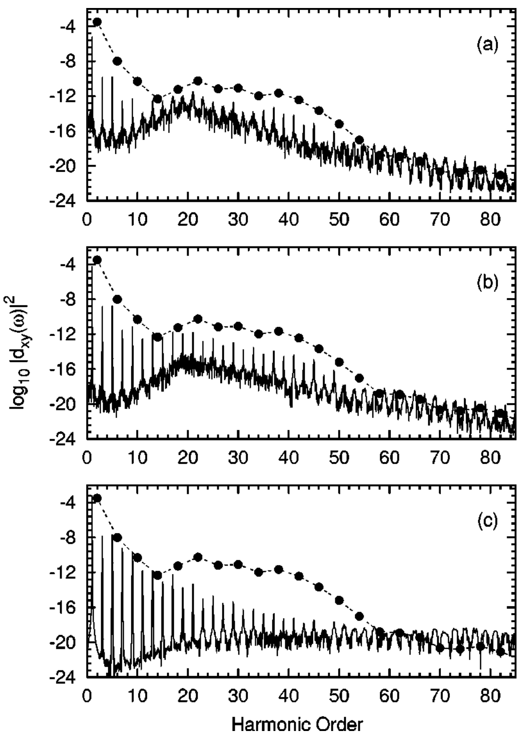

FIG. 4. Circularly polarized HHG power spectra of He atoms in crossed laser beam configuration. (a) $I_1=2\times10^{12}$ W/cm^2, (b) $I_1=2\times10^{13}$ W/cm^2, and (c) $I_1=2\times10^{14}$ W/cm^2. The incident LP field intensity is fixed at 2×10^{14} W/cm^2. Laser wavelengths are the same as those in Fig. 3. For comparison, the one-color LP HHG data (from Fig. 3) (denoted by filled circles) are also included.

2×10^{13} W/cm^2. Finally, the background radiation intensity is significantly suppressed, both in the lower harmonic and in the plateau regime, as the CP field intensity I_1 is increased.

To explore the mechanisms responsible for the observed phenomena of the suppression of both the HHG plateau and the continuous background intensities at stronger incident CP fields, we have performed the following additional calculations and analysis. First we note that the Keldysh parameter [8], corresponding to LP 2ω field at intensity 2×10^{14} W/cm^2, is $\gamma=1.54>1.0$. This may imply that the multiphoton mechanism is the dominant process at work here. However, the Keldysh parameter is, at best, a qualitative indicator regarding the HHG mechanism, particularly when γ is not far from one. More quantitative information can be obtained by a time-frequency analysis of the HHG spectrum in different energy regimes. It is generally expected that in the tunneling process, the electrons are emitted in bursts near the maxima in the oscillating electric field, while the multiphoton excitation is constant throughout the optical cycle [9]. Further, the multiphoton ionized electrons appear in the continuum near the nucleus while tunneling electrons originate at the outer turning point of the instantaneous potential barrier. The temporal profile of a given qth harmonics $d_q(t)$ can be obtained by the inverse Fourier transformation of the frequency-dependent dipole spectrum $d(\omega)$ in the frequency interval $[(q-1)\omega,(q+1)\omega]$ [10–12]. For the harmonics produced by the tunneling process, it has been shown that for a given qth harmonic below the cutoff, there exists two returning electron wave-function trajectories of suitable energy $q\omega-I_p$ with different phases (where ω is the incident laser frequency and I_p is the ionization potential of the atom), leading to coherent beating in the time-dependent dipole spectrum $d_q(t)$ [11]. In contrast, $d_q(t)$ is expected to show nonoscillatory smooth temporal profile in the multiphoton dominant processes [12]. We have thus performed the calculation of $d_q(t)$, corresponding to different CP field intensity I_1. We found that for the case of weak CP field, $I_1=2\times10^{12}$ W/cm^2, the time-dependent dipole spectra $d_q(t)$ for plateau harmonics show oscillatory patterns superimposed on a major smooth profile, indicating that both multiphoton and tunneling mechanisms contribute to the HHG processes. As the CP field intensity increases, the oscillatory pattern decreases, and at the highest CP field considered here, $I_1=2\times10^{14}$ W/cm^2, $d_q(t)$ shows a nonoscillatory smooth temporal profile, suggesting that the plateau harmonics are produced mainly by the multiphoton mechanism.

Based on this time-frequency analysis, we see that as the CP-field intensity is sufficiently strong, multiphoton excitation becomes dominant and the probability of absorbing more than one CP photon increases substantially. Since an atom that absorbs more than one CP photon cannot emit harmonics back to the ground state (due to the dipole selection rule), the HHG height in the plateau regime is suppressed as seen in Figs. 3(d) and 4(c).

A similar explanation can account for the observed suppression of the continuous background intensity. We note that although there is some discussion [13] on the possible source for the production of the background radiation associated with HHG processes, the actual mechanism seems not yet settled. From our analysis, we infer that the background radiation is likely to be due to the bremsstrahlung mechanism, namely, the rescattering of the electron with the nucleus in the laser fields [14,15]. This is consistent with the time-frequency analysis indicated above, namely, the probability for rescattering decreases with increasing CP field intensity, leading to the quenching of the bremsstrahlung process. As an additional support of this view, we have performed an analysis of the pulse-length dependence of the HHG spectrum and background radiation. It is known that there is no background radiation in the HHG spectrum for atoms in purely periodic fields or infinitely long pulses, due to the destructive interference of the bremsstrahlung radiation produced in each optical cycle [14]. As the pulse length becomes sufficiently short and the laser pulse becomes nonperiodic, continuous background radiation can occur. We have performed a separate calculation of the He HHG processes in two-color crossed laser beams with substantially shorter laser pulses ($T=30$ optical cycles). We found that the background suppression behavior at strong CP fields is essentially the same as that observed for the longer pulse case ($T=90$ optical cycles). This supports the view that the background radiation is mainly due to the rescattering of the higher energy photoelectrons with the parent nucleus.

In summary, we have proposed and demonstrated a feasible experimental scheme for the production of purely CP HHG in a crossed laser beam configuration. Although both LP and CP HHG are produced simultaneously, they propagate in perpendicular directions and are completely separable. The suppression of the background intensity, leading to cleaner HHG, is another potential advantage. For a given incident LP field intensity, one can also tune the incident CP field intensity to find the optimal range for the production of CP HHG. Finally, by performing a time frequency of the HHG spectra, with respect to the variation of the incident CP laser intensity, insights can be obtained regarding the mechanisms responsible for the production and suppression of HHG, as well as the origin of the continuous background radiation.

The calculations in this work were performed on the SGI/CRAY Origin2000 supercomputer. We acknowledge the Kansas Center for Advanced Scientific Computing for the grant of computer time.

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