

Highly Efficient High-Harmonic Generation in an Orthogonally Polarized Two-Color Laser Field

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Highly efficient high-harmonic generation was achieved in helium using a two-color laser field that consisted of the fundamental and the second harmonic fields of a femtosecond Ti:sapphire laser. By applying a high intensity second harmonic, the harmonics generated in the orthogonally polarized two-color field were stronger than those obtained in the fundamental field by more than 2 orders of magnitude, and even stronger than those of the parallel polarization case. A conversion efficiency as high as 5×10^{-5} was obtained for the 38th harmonic at 21.6 nm. The physical origin of this enhancement was deduced by analyzing the electron behavior in the two-color field.

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High-order harmonic generation (HHG) is a coherent interaction process between atoms and a driving laser field and produces ultrashort coherent radiation reaching the soft x-ray region. The harmonic generation process is closely governed by the detailed structure, such as chirp, of the driving laser electric field [1,2]. The application of a two-color laser field for harmonic generation has been a fascinating topic of research since various kinds of synthesized electric fields can be applied to atoms. Theoretical investigations on harmonic generation in the two-color laser field reported the generation of all integer-order harmonics due to the breaking of inversion symmetry and an especially large enhancement of harmonic signal [3]. When the polarizations of the two combining laser fields, consisting of the fundamental and the second harmonic (SH) fields, were parallel, an enhancement of the harmonic signal by more than 2 orders of magnitude was predicted, compared to that generated only in the fundamental laser field [3,4]. The combination of two laser fields can help reduce the duration of attosecond harmonic pulses [5]. Experimentally even-order harmonics were observed in HHG when using the two-color laser field, signifying the mixing effect of the two-color field.

There have been intensive efforts towards the development of efficient harmonic conversion techniques. By maximizing the number of atoms involved in the harmonic generation using a long gas cell or a long gas jet, harmonic generation can now be achieved in the absorption-limited regime where maximum harmonic energy generated per unit cross section is determined by the reabsorption by neutral atoms. Using xenon as the target medium, strong harmonics around 53 nm (15th order) was obtained with a conversion efficiency of 4×10^{-5} [6]. Using argon, the conversion to harmonics at the 30 nm region (27th order) reached 1.5×10^{-5} [7]. On the other hand, harmonics from neon produced a strong signal at 13 nm, attaining a much lower conversion of 5×10^{-7} [8]. The strong harmonics at these regions were obtained using either a long gas cell pumped by a laser beam focused by long focal length

optics or a long gas jet driven by a profile-flattened laser beam so as to maximize the effective beam cross section at the absorption-limited regime [6–9]. However, harmonics in other wavelength regions are not as strong as in the above cases, either due to the strong absorption of harmonics by neutral atoms or due to the limit of the highest harmonic order set by applied laser intensity.

In this Letter we report the generation of exceptionally strong harmonics at the 20 nm region (38th order) from helium obtained by applying the two-color (fundamental and SH) laser field. The effects of the polarization condition and the relative phase between the fundamental and the SH fields on the harmonic generation were investigated. In addition to the observation of even harmonics, the modulation of harmonics was measured with a periodicity of π to the change of the relative phase. The two-color HHG experiments were performed using the experimental setup shown in Fig. 1. The focusing of the femtosecond laser pulse into a helium gas jet was done by a spherical

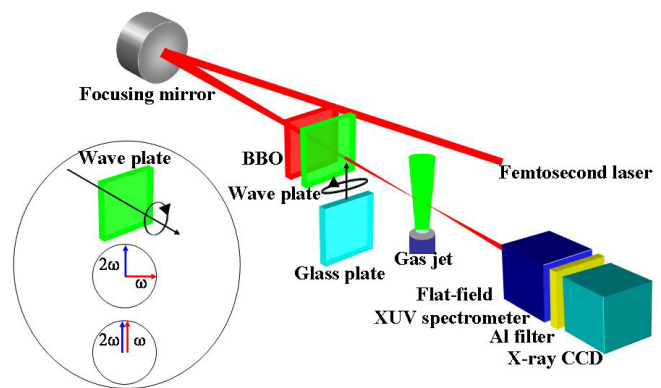


FIG. 1 (color online). Experimental setup for high-harmonic generation driven by a two-color laser field. The polarizations of fundamental (ω) and second harmonic (2ω) fields were controlled using the wave plate. For control of the relative phase between the fundamental and the SH fields, a $150 \mu\text{m}$ thick glass plate was inserted.

mirror ($f = 60$ cm) and the emitted harmonics were detected using a flat-field extreme ultraviolet (XUV) spectrometer equipped with a back-illuminated x-ray charge-coupled device (CCD; Princeton Instruments). For SH generation, a beta-barium borate (BBO) crystal ($100 \mu\text{m}$ thick, type I) was placed between the focusing mirror and gas jet, so after the BBO crystal, the laser field consisted of both the SH and the residual fundamental laser fields. The polarization of the SH field generated in this case was orthogonal to that of the fundamental laser field. By using a special quartz wave plate (Minioptic Technology) that acts as a half-wave plate for the fundamental and a full-wave plate for the SH fields, we were able to continuously rotate the polarization of the fundamental laser field, while maintaining the polarization of the SH. This simple setup allows two-color harmonic generation in parallel and orthogonal polarization conditions of the fundamental and the SH fields.

More specific experimental conditions are as follows: the output from the femtosecond chirped-pulse amplification Ti:sapphire laser operating at 10 Hz [10] had a pulse width of 27 fs and a spectral bandwidth of 42 nm centered at 820 nm. The laser intensity was 1.6×10^{15} W/cm² at the focus. For the optimum SH conversion the BBO crystal was placed 40 cm from the focusing mirror and the energy conversion efficiency was about 20%. The gas density and target position were optimized for the harmonic generation by the fundamental laser pulse. The gas pressure in the interaction region obtained with a 0.5 mm diameter circular nozzle was 950 Torr. Since the radial gradient of the electron density at the interaction region causes defocusing of the propagating intense laser pulse, the gas jet was placed 12 mm before the laser focus. When the target location is optimized this way, the converging laser beam exhibits self-guided propagation within the gas target medium [9]. The laser intensities of the fundamental and the SH pulses in the harmonic generation region were 5×10^{14} and 8×10^{14} W/cm², respectively. These estimates were made from the observed harmonic cutoff orders (77th for the fundamental and 42nd for the SH) as they represent actual laser intensities during strong harmonic generation. In 15–50 nm, the sensitivity of the XUV spectrometer varied by less than 10% when the polarization of the incident light was changed.

Harmonic spectra from helium obtained using two-color laser fields with parallel and orthogonal polarizations are shown in Fig. 2, along with those with one-color laser fields of the fundamental and the SH fields. The harmonics by the fundamental field were obtained after removing the BBO and the wave plate, and the harmonics by the SH field were obtained after filtering the fundamental field using two dichroic mirrors. When the two-color laser pulses were applied, the harmonics were enhanced by such an extent that the total thickness of two aluminum filters, used in front of the CCD to block the scattered laser light, had to be increased to $1.5 \mu\text{m}$ to prevent the saturation of CCD. In contrast, we normally used the thinnest freestanding alu-

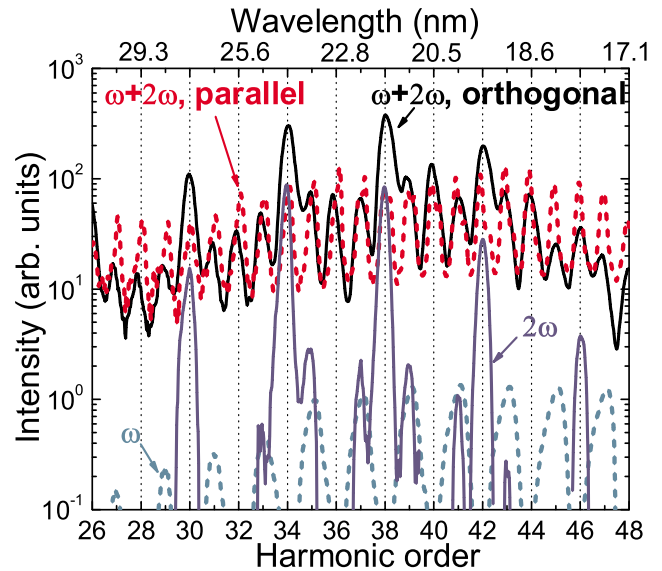


FIG. 2 (color online). Harmonic spectra from He generated in the fundamental (ω), second harmonic (2ω), and two-color ($\omega + 2\omega$) laser fields. In the two-color field, the cases of parallel and orthogonal polarizations are given.

minum filters (two 200 nm filters) in one-color fundamental experiments. In the case of a two-color field with parallel polarization, all integer-order harmonics appeared with comparable intensities. Compared to the harmonics generated by the fundamental pulse alone, the two-color harmonics were enhanced by a factor of more than 50. The harmonics generated by the SH field were much stronger than those generated by the fundamental field and were quite comparable in strength to those generated by the parallel-polarized two-color field at $2(2n + 1)$ th orders, especially at 34th and 38th orders. The side lobes that appeared near the 34th and 38th orders are the second order spectrum of harmonics generated by the residual fundamental field. The strongest harmonics, however, were obtained when the two-color laser field with orthogonal polarization was applied. In this case also all integer-order harmonics were observed, but the harmonics at $2(2n + 1)$ th orders were particularly stronger than their neighboring harmonics by factors ranging from 3 to 5 and also by similar factors compared to those of the parallel polarization case. Consequently, the enhancement factor of the two-color, orthogonal polarization case was more than 100 compared to the one-color, fundamental case. These results are clearly different from the observation by Eichmann *et al.* [11]. They showed that the harmonics generated by the parallel-polarized two-color field were more intense than those generated by the orthogonally polarized two-color laser field. Also in the latter case, the intensity of even-order harmonics was very weak, contrary to our result.

For a physical understanding of the harmonic generation in the orthogonally polarized two-color laser field, we ana-

lyzed the electron behavior in the synthesized electric field. The electric field synthesized with the fundamental and SH fields may be written as $\mathbf{E}(t) = \exp[-2\ln 2(t/\tau)^2] \times [E_1 \cos(\omega t)\hat{x} + E_2 \cos(2\omega t + \phi)\hat{y}]$, where E_1 and E_2 are the amplitudes of the fundamental and SH fields, and τ , ω , and ϕ are the pulse duration (FWHM), fundamental frequency, and relative phase of the SH field to the fundamental field, respectively. These values were chosen to match the experimental parameters, setting the same duration for the fundamental and the SH pulses. With $\phi = -\pi/2$, the Lissajous diagram of the electric field has a bow-tie shape, as shown in the inset of Fig. 3(a), and the electric field becomes nearly linearly polarized when it moves about the origin [between the points C and D in Fig. 3(a)]. As a result, the electron can recollide with the nucleus, similar to the usual one-color case. This is confirmed from the position expectation value of the electron, given in Fig. 3(a), calculated using the Lewenstein model [12].

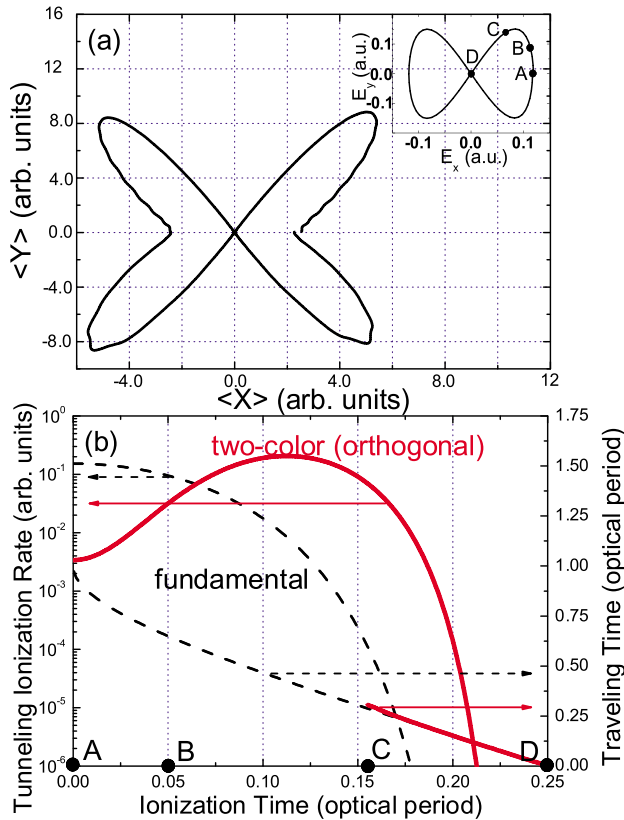


FIG. 3 (color online). (a) Position expectation value of the electron under orthogonally polarized two-color laser field. The inset shows the Lissajous diagram of the electric field ($I_\omega = 5 \times 10^{14}$ W/cm², $I_{2\omega} = 8 \times 10^{14}$ W/cm²) A, B, C, and D correspond to 0.00, 0.05, 0.16, and 0.25 optical cycles, respectively. The points B and C correspond to the ionization times for the highest-order harmonics generation in the fundamental and the two-color fields, respectively. (b) Tunneling ionization rate and traveling time of the electron in the fundamental field and orthogonally polarized two-color field.

For further verification, we calculated the classical trajectory of the electron under the two-color field using the three-step model [13]. The electron recollision can occur when the electron is ionized only during the straight section of the electric field [between the points C and D in Fig. 3(a)] and the traveling time is short [< 0.3 optical period as shown in Fig. 3(b)]. Thus the harmonics are generated mainly from short trajectories. The electrons ionized in this time period are also the main contributor to phase-matched harmonic generation even in the fundamental field, because the wave packet spreading is small and the resulting harmonic phase is insensitive to laser intensity variation. In this case, the tunneling ionization rate, calculated using the Ammosov-Delone-Krainov (ADK) formula [14], for the two-color field is much larger than that for the fundamental field, as shown in Fig. 3(b); i.e., the electron wave packet at the time of ionization is significantly denser. Consequently, orthogonally polarized two-color field can generate harmonics much more strongly than the fundamental field, agreeing with the results shown in Fig. 2. On the other hand, the overall ionization is not too high to degrade strong harmonic generation, as discussed below. According to the single-atom calculation, these behaviors are maintained from $\phi = -5\pi/8$ to $-3\pi/8$. For other relative phases, HHG becomes weak with orthogonally polarized two-color field.

Understanding the results for orthogonally polarized and parallelly polarized two-color fields requires more than just the consideration of the single-atom response. Single-atom calculations [11] show that the two-color field with parallel polarization can generate much stronger harmonics than that with the orthogonally polarized field. When an intense two-color field is applied, the effect of ionization should be taken into account in the harmonic generation. According to the calculation using the ADK formula [14], the total amount of ionization for the field with parallel polarization, under the current experimental conditions, is about 90%, much larger than that for the field with orthogonal polarization (30%). The high ionization in the parallelly polarized field makes the phase matching conditions unfavorable because, under such conditions, the field experiences more severe self-phase modulation in the ionizing medium, which leads to laser chirp and plasma defocusing. As a result, after propagation through the medium, the harmonics with the parallel-polarized field are not as strong as those with the orthogonally polarized field, as Fig. 2 shows.

The profile of the electric field synthesized with the fundamental and the SH fields is sensitive to the relative phase between the two pulses, and thus harmonic generation is also greatly affected by the shape of the synthesized electric field. As the relative phase between the fundamental and the SH fields changes in a dispersive medium, we inserted a glass plate (fused silica) with a thickness of 150 μm . With the rotation of the glass plate by slightly more than 10° , the relative phase is changed by 4π . Since a two-color field with a relative phase change of π can be

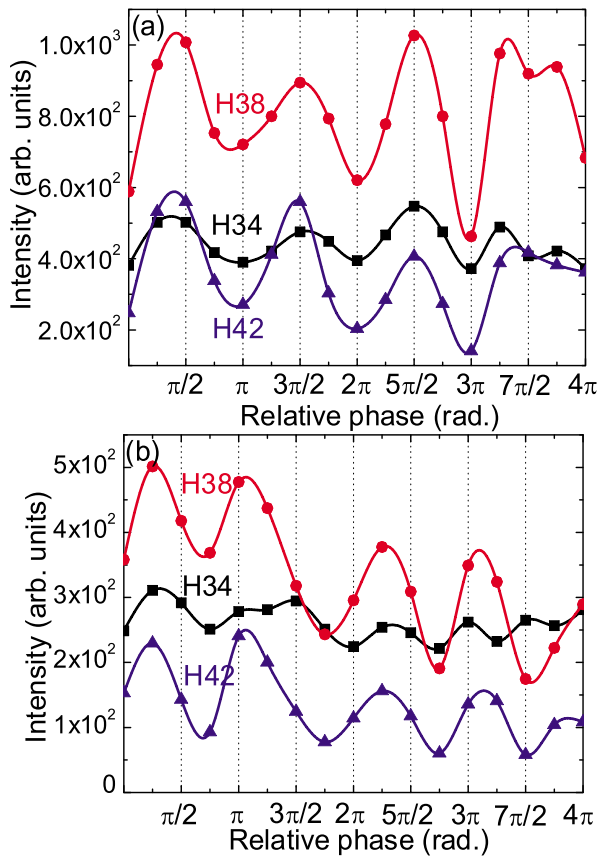


FIG. 4 (color online). Modulation of $2(2n + 1)$ th order harmonics accompanied by controlling the relative phase in a two-color laser field. This relative phase between fundamental and SH fields was changed by rotating the glass plate shown in Fig. 1. (a) Orthogonal polarization case, and (b) parallel polarization case.

mapped to the one with an opposite sign by shifting the time origin by half a cycle, the harmonics generated in the two-color field (for both parallel and orthogonal polarizations) are π periodic with respect to the relative phase change. The results shown in Fig. 4 confirm this periodicity for both polarization conditions, which, in turn, proves that the harmonics were, indeed, generated in the two-color field. The results shown in Fig. 4 also indicate that the modulation increases with harmonic order, from 0.10 for the 34th order to 0.27 for the 38th and to 0.47 for the 42nd for parallel polarization, which also agrees with the theoretical prediction [15]. A very similar trend, namely, order-dependent modulation, is also seen for orthogonal polarization. In the case of pure mixing orders, 36th and 40th, larger modulation was observed, reaching 0.47 and 0.62 for the parallel polarization case, respectively.

Another aspect we noted is the increase of the harmonic signal when the relative phase is changed. Though the signal increase in the case of the parallel polarization was larger, the harmonics generated with the orthogonal polarization was still stronger than those with the parallel polarization, at least by a factor of 2 for the 38th and 42nd

harmonics. After optimizing the phase, the 38th harmonic is stronger by more than an order of magnitude than that generated by SH pulses alone. The estimated harmonic conversion efficiency is 5×10^{-5} (150 nJ harmonic energy to the total laser energy of 2.8 mJ). For energy calibration, the total transmission of two aluminum filters of 0.7% was used (each filter with 700 nm aluminum and two 25 nm Al_2O_3 layers on both sides). This is at least 2 orders of magnitude greater than the conversion efficiency reported in the 20 nm region ($\sim 10^{-8}$) [16]. Consequently, considering the fact that lighter atoms generate harmonics at higher order but with lower efficiency [17], the achieved conversion efficiency is quite significant, even compared to those obtained at much longer wavelengths.

In conclusion, using an orthogonally polarized two-color laser field, we achieved very efficient high-harmonic generation in helium. With higher SH intensity in the two-color field, a strong enhancement of harmonics from helium was achieved in the 20 nm region. The strong modulation of harmonics obtained with the control of the relative phase between the fundamental and the SH fields, as well as the observation of all even harmonics, clearly showed the harmonic generation in the two-color field. The physical origin for the strong enhancement of the harmonics was deduced from the analysis of the electron behavior in the two-color field. The relative simplicity and effectiveness of this two-color method for strong harmonic generation will be of great use for practical applications.

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