

Frequency modulation of high-order harmonic fields with synthesis of two-color laser fields

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Abstract: We report periodical frequency modulation of high-order harmonic fields observed by changing the delay between the driving two-color laser fields consisting of the fundamental and its second harmonic (SH) field. The amplitude of modulation has been up to ~ 0.4 eV, which is larger than the bandwidth of the fundamental field. Experimental results show that the intensity and chirp of the fundamental field can control this phenomenon. Numerical analysis by solving the time-dependent Schrödinger equation approves of these results and shows that anharmonic frequency components of the SH field have a crucial role in this phenomenon.

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1. Introduction

There has been growing interest to investigate high-order harmonic generation (HHG) by using two laser fields with different wavelengths, since it makes it possible to analyze or control HHG processes in situ by changing the shape of an instantaneous electric field with alternation of the delay (relative phase) between the two laser fields. HHG by a Ti:sapphire laser field superposed with its second harmonic (SH) field has been particularly interesting due to inversion symmetry breaking, resulting in generation of even-order harmonic fields in the plateau region.

Such dense harmonic spectra are on the way approaching generation of an isolated attosecond pulse (IAP). For example, Oishi et al. have demonstrated generation of a continuous spectrum in the extreme ultraviolet (XUV) spectral region when the pulse duration of the fundamental field approaches 9 fs [1]. Feng et al. have succeeded in observing an IAP generated from a 28-fs multi-cycle laser pulse by adopting the polarization gating technique on a two-color laser field [2]. We also note that it is advantageous to generate the IAP by a two-color laser field synthesized from the fundamental field and its anharmonic field, rather than its exact SH field. Takahashi et al. have clearly observed generation of a continuous spectrum by extending the cut-off region. This result has been obtained from a 30-fs pulse of a Ti:sapphire laser superposed with an infrared (IR) laser pulse with a wavelength of 1300 nm [3].

The theoretical work of Fleischer et al. predicts the position of the even-order harmonic fields when the frequency of the superposed field is detuned from that of the fundamental field [4]. This has been verified experimentally by Vozzi et al. [5] and Bandulet et al. [6] by using different IR frequencies in a two-color laser field. Although generation of new frequency components is observed, neither of these works has predicted or observed frequency modulation of high-order harmonic (HH) fields, which is the main concern of this paper. Generation of new spectral components using a two-color laser field synthesized from the fundamental field and its anharmonic field can result in generation of a quasicontinuum in the HH spectra, which can moderate the conditions of IAP generation. Therefore, frequency modulation of the HH fields simply by changing the delay between the two fields could add another degree of freedom to relaxing the conditions of IAP generation.

Not only being useful in IAP generation, two-color laser fields have been also applied in in-situ measurement and control of the HHG processes by controlling the electron trajectory which happens at a sub-fs time scale. Mauritsson et al. have verified the effects of the delay on generation of attosecond pulse trains using a two-color laser field [7], and Dudovich et al. have made an attempt to measure and control the birth of attosecond XUV pulses by changing the delay [8]. Dahlström et al. have similarly used a two-color laser field for atomic measurement of attosecond pulse trains and have also compared the results to those obtained by macroscopic measurements [9]. Furthermore, quantum path selection in HHG by a two-color laser field has been investigated by Ishii et al. [10]. An orthogonally polarized two-color laser field has been even applied to increase the efficiency of the HHG process [11]. All of these experiments are implemented with a pulse duration longer than 20 fs and we could not find any notable modulation of the frequency of the HH fields in the above results.

In this paper, we report on the first observation of periodical frequency modulation of HH fields depending on the relative phase difference between the two-color laser fields, owing to the fact that the amplitude of the frequency modulation is sufficiently large to be resolved by an XUV spectrograph and the relative phase difference can be finely controlled by a two-color interferometer. In the next section, we will briefly introduce the experimental setup, and then in Sec. 3, show the experimental results for investigating essential laser parameters related to the frequency modulation of HH fields. These sections are followed by numerical analysis of the HHG process with a two-color laser field by solving the time-dependent Schrödinger equation (TDSE) followed by two discussions to explain how the frequency modulation can be controlled by alternation of the delay and how to estimate the amplitude of frequency modulation (Sec. 4). Finally, we will summarize the results.

2. The experimental setup

We used a chirped pulse amplification (CPA) system of Ti:sapphire laser, which is capable of generating 9.9-fs pulses with a peak power of 1.1 TW without using a nonlinear spectral broadening method such as a hollow-core fiber filled with gaseous medium. One of the key optical elements for spectral broadening (pulse shortening) of the laser system is a gain narrowing compensator (GNC) inserted in the cavity of the regenerative amplifier. The details of the laser system was reported in Ref. [12]. We have used a fundamental pulse with a pulse energy of 15 mJ and a pulse duration of ~ 15 fs yielded by a brief modification in the GNC in the regenerative amplifier to realize a higher pulse energy and a more stable spectrum behind the multi-pass amplifier for this sensitive experiment.

The spectrum of the fundamental field is shown by the solid curve in Fig. 1(a) and has a foot-to-foot bandwidth of ~ 0.33 eV (in photon energy units) at a central wavelength of ~ 808 nm. The dashed curve in Fig. 1(a) shows the spectral phase after control by a liquid-crystal spatial light modulator (LC-SLM). The SH field has been generated by a self-standing Type I BBO crystal with a thickness of $100 \mu\text{m}$ to result in a Fourier-limit pulse duration of 20 fs (FWHM) at a central wavelength of 417 nm with pulse energy of $780 \mu\text{J}$, the spectrum of which is shown in Fig. 1(b) by the solid curve. Although the central wavelength was tunable in a range of ± 7 nm, we could not get a strong SH field at a central wavelength of 400 nm. We will see that the anharmonic frequency components in the SH field have a crucial role in the observed

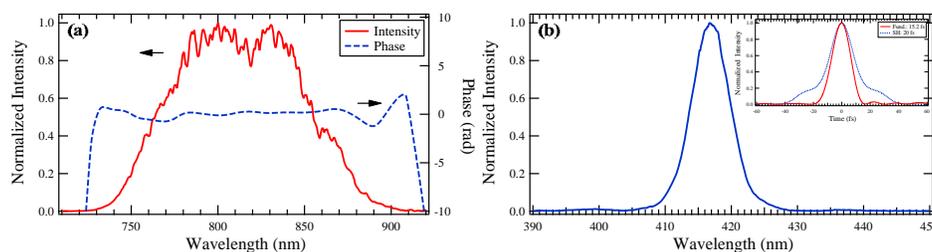


Fig. 1. Spectra and pulse shapes of the driving two-color laser fields. (a) Spectrum of the fundamental field (solid curve) together with the spectral phase after control by the LC-SLM (dashed curve). (b) The solid curve shows the spectrum of the SH field. The dotted blue curve in the inset shows the Fourier-limit temporal profile (FWHM 20 fs) of the SH field compared to the reconstructed temporal profile of the fundamental field with FWHM of 15.2 fs (solid red curve).

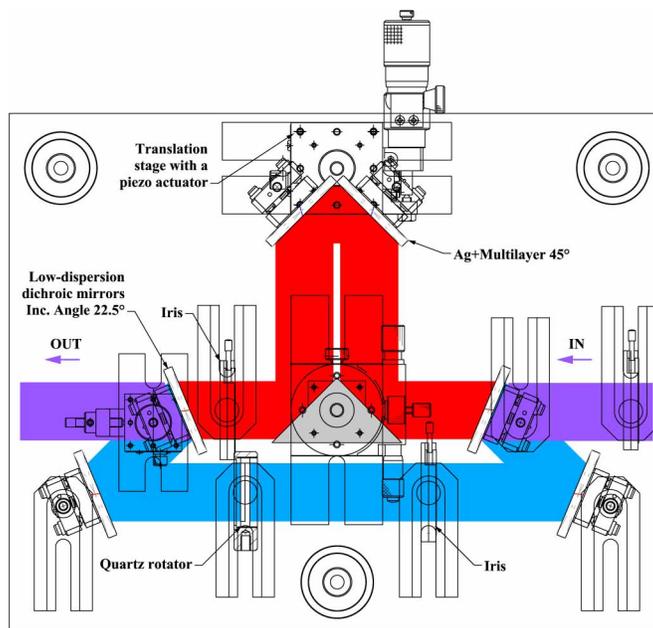


Fig. 2. Schematic of the two-color interferometer implemented to synthesize the broadband two-color laser field having an adjustable beam spot size at the focus.

novel phenomenon (Sec. 4). The dotted blue curve in the inset of Fig. 1(b) shows the Fourier-limit temporal profile of the SH field compared to the reconstructed temporal profile of the fundamental field with FWHM of 15.2 fs, shown by the solid red curve. The generated SH field and the residual fundamental field were sent into a two-color interferometer, the schematic of which is shown in Fig. 2. The SH field was reflected with a broadband dichroic mirror (BDM) at the entrance of the interferometer with an incident angle of 22.5° , while the fundamental field transmitted the BDM. This exceptional incident angle was needed to keep high reflectivity and low dispersion in the broad wavelength region of 355 nm up to 445 nm. The polarization of the SH field was rotated by 90° with a quartz rotator. The fundamental field passed through a delay line controlled by a translation stage with a piezo actuator, and combined spatio-temporally with the SH field on another BDM at the exit of the interferometer. Owing to the different paths of the two laser fields, we were able to independently control the focusing conditions of the two laser fields by adjusting the diameters of two iris diaphragms placed in the arms of the interferometer.

The pulse duration of the SH field is slightly stretched by the dispersion of the rotator (Quartz, $t=1.85$ mm) and also the input window (UVFS, $t=2$ mm) of a focusing chamber and has an FWHM of ~ 40 fs. Using a broadband 45° dichroic mirror, the two-color synthesized laser field is reflected into the focusing chamber consisting of an Al-coated off-axis parabolic mirror with a focal length of 500 mm to be focused into a gas cell with an interaction length of 12 mm. The HH spectra are measured with an XUV imaging spectrograph consisting of an entrance slit, a flat field grating, and an X-ray CCD camera. An aluminum foil with a thickness of 150 nm supported by a mesh has been placed between the slit and the grating in the XUV spectrograph to prevent saturation of the X-ray CCD camera by strong irradiation of the fundamental field. The X-ray CCD camera and the driver of the Piezo actuator are controlled by a personal computer to make the measurements of the XUV spectra full automatic. The schematic of the whole HH

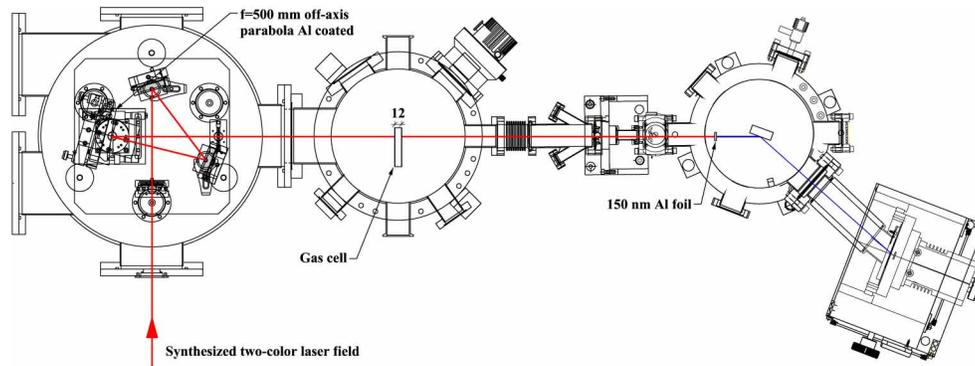


Fig. 3. Schematic of the HH generation and measurement setup.

generation and measurement setup is shown in Fig. 3.

3. HHG by the two-color laser field

For the first trial of HHG by the two-color synthesized laser field, the pulse energy of the fundamental laser field was 8.5 mJ and that of the SH field was 125 μ J both attenuated by using the iris diaphragms placed in the arms of the two-color interferometer. The two laser fields have almost the same beam spot at the focus, which is ~ 100 μ m to get a high HH yield. The target gas was argon (Ar) with a backing pressure of ~ 20 torr. The HH spectrum consisting of only odd-order harmonic fields generated by the fundamental field alone is shown by the dashed curve in Fig. 4, and that generated by the two-color laser field at a fixed delay at which the peaks of the two fields overlap, is shown by the solid curve. The two-color laser field has resulted in generation of even-order harmonic fields as well and has decreased the total integrated HH yield to $\sim 65\%$ of the yield obtained by the fundamental field alone. We suppose the lower HH yield originates from high degrees of ionization of the target Ar gas with high field amplitude of the two-color laser field, even though the increase of the pulse energy is only $\sim 1.5\%$. Spectral blue shifts of the odd-order harmonic fields (for example, ~ 0.26 eV at the 27th order) also support this speculation.

By changing the delay between the fundamental field and the SH field in 20 nm steps, we recorded the HH spectra for 100 laser shots per delay scan. The resultant spectrogram is shown in Fig. 5(a). After normalizing the delay with the optical period of the fundamental field (T_f), we can see the modulation of the intensity of each even-order harmonic field with a period of $T_f/4$, consistent with the results of Dahlström et al. [9], rather than those of Dudovich et al. [8], who reported a modulation period of $T_f/2$. Numerical analysis also agrees with our experimental results and those of Dahlström et al., yielding a modulation period of $T_f/4$, as will be shown in Fig. 8.

According to the theory of in-situ measurement of HHG process [8], the phase of the intensity modulation of each even-order harmonic field could give us information on the emission time of the adjacent odd-order harmonic fields, if the SH field did not significantly perturb the motion of the electrons. In fact, the intensity of the SH field in our experiment is too high to execute the in-situ measurement of attosecond pulses, because even the intensity of the odd-order harmonic fields is modulated by alternation of the delay. Instead, we have found a novel feature in each frequency component of even-order harmonic fields. In Fig. 5(a), saw-toothed waveforms appear in the 18th up to the 28th even-order harmonic spectra. These waveforms

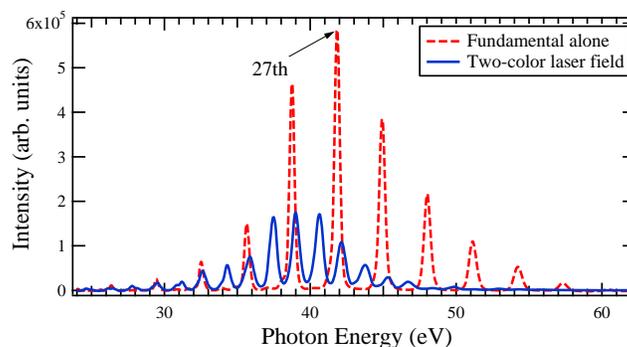


Fig. 4. HH spectrum generated by the fundamental field alone (dashed curve). The solid curve shows the spectrum generated by the two-color laser field.

are the consequences of the periodical peak frequency modulations of the even-order harmonic fields. This delay-dependent periodical frequency modulation has an amplitude of ~ 0.4 eV for the 22nd harmonic field as shown in Fig. 5(b). This amplitude is larger than the foot-to-foot bandwidth of the fundamental field and accounts for $\sim 26\%$ of the HH separation generated by the two-color laser field. This novel phenomenon has been also observed in krypton gas (Kr) and has almost the same characteristics compared to the case of Ar gas [13].

To verify the reason why the periodical frequency modulations appeared in this experiment, we changed the intensity of the fundamental field and kept the other parameters fixed and repeated the correlation experiment. In the later experiment, we faced a failure in the properties of two laser mirrors used in the delay line of the two-color interferometer to reflect the fundamental field and after replacing them, we could get a much better beam spot and therefore less amount of fundamental pulse energy was necessary for HHG. With fundamental pulse energy of 5.7 mJ and SH pulse energy of 85 μJ at the central wavelength of ~ 417 nm, we could observe a large amplitude of frequency modulation for the even-order harmonic fields as shown in Fig. 6(a). Figure 6(b) shows the spectrogram obtained by a lower fundamental pulse energy of 4.2 mJ and SH pulse energy of 60 μJ to keep the relative intensity of the two laser fields almost constant. The other parameters such as the backing pressure of the Ar gas are the same.

While frequency modulation could be observed in the lower-order harmonic fields generated by a higher fundamental pulse intensity, the frequency of the harmonic fields generated by a lower fundamental pulse intensity is not modulated to that extent. For example, Fig. 6(c) shows that the amplitude of frequency modulation of the 20th harmonic field (~ 0.3 eV) is larger when the harmonics are generated by a pulse energy of 5.7 mJ (diamonds with solid curve) compared to that generated by the lower pulse energy of 4.2 mJ with an amplitude of ~ 0.17 eV (diamonds with dashed curve). Other even-order harmonic fields are also in a similar relation. Therefore, the intensity of the fundamental field is a key parameter which can control this phenomenon. We also investigated the effects of the intensity of the SH field, but we noticed that its increase does not affect this phenomenon. Moreover, we also investigated the effects of phase matching on the amplitude of frequency modulation by changing the position of the gas cell. We found out that the amplitude gets larger when the gas cell is placed slightly before the focus rather than behind the focus, where total HH yield is higher.

We also investigated the effects of the chirp of the fundamental field on this phenomenon. After investigation, we found out that the position of the grating compressor being shifted to compensate for the dispersion of the BDMs used in the two-color interferometer and the

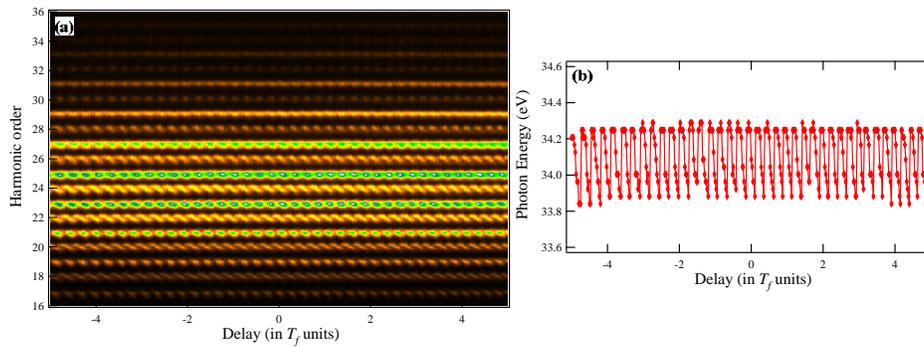


Fig. 5. XUV spectra collection by scanning the delay between the two-color laser fields and the first observation of frequency modulation of HH fields. (a) Spectrogram of HH fields. The HH spectra are scaled with the optical frequency of the fundamental field. The delay is normalized with the optical period of the fundamental laser field (T_f). (b) Peak frequency of the 22nd harmonic field against the delay, which is extracted from panel-(a) by calculating the frequency of the HH field when its intensity reaches a maximum. All of the spectrograms in the following figures are depicted in the same manner.

input window of the vacuum chamber was slightly imposing a minus chirp on the fundamental field. The amount of the minus chirp considering the group delay dispersion (GDD) when the frequency modulation appeared strongly as in Fig. 6(a) was $\sim -75 \text{ fs}^2$. We imposed a higher amount of minus chirp of the order of $\sim -125 \text{ fs}^2$ on the fundamental field by changing the position of the grating compressor and repeated the correlation experiment with keeping the fundamental pulse energy of 4.2 mJ and SH pulse energy of 70 μJ at the central wavelength of 417 nm.

The result is shown in Fig. 7(a), showing typical HH fields having frequency modulation in the lower-order harmonics. As we moved the compressor back to the position which is considered to be very close to get a Fourier-limit pulse, the amplitude of frequency modulation shrank as shown in Fig. 7(b) with the same experimental conditions but SH pulse energy of 80 μJ at a central wavelength of 417.8 nm, which is due to moving the grating compressor. To get a clearer view of the effects of the chirp of the fundamental field, the amplitudes of frequency modulation of the 24th harmonic field, which is selected for having the largest amplitude of frequency modulation, are compared in Fig. 7(c), which shows a larger amplitude of $\sim 0.26 \text{ eV}$ when the fundamental field has minus chirp (diamonds with solid curve) compared to the case of unchirped fundamental field with an amplitude of $\sim 0.17 \text{ eV}$ (diamonds with dashed curve). Therefore, the chirp of the fundamental field is also another key parameter which can control this phenomenon.

4. Numerical analysis and discussions

As a further step towards understanding this phenomenon, we numerically solved the time-dependent Schrödinger equation (TDSE) for Ar atom within the single-active-electron (SAE) approximation [14]. One of the parameters we could not consider during the experiment was the wavelength of the SH field in a broad range. Hence, we investigate the effects of the SH wavelength as well as those of the intensity and chirp of the fundamental field on the frequency

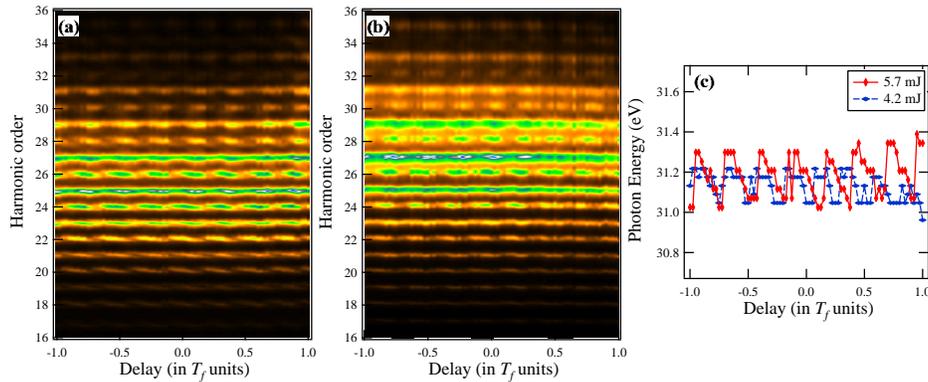


Fig. 6. Spectrograms to investigate how the intensity of the fundamental field affects the frequency modulation of the HH fields. (a) Spectrogram obtained by a high-energy fundamental field (fundamental 5.7 mJ and SH 85 μJ). (b) Spectrogram obtained by a low-energy fundamental field (fundamental 4.2 mJ and SH 60 μJ). (c) Peak frequency of the 20th harmonic field extracted from panel-(a) shown by the diamonds with solid curve and that of panel-(b) (diamonds with dashed curve).

modulation of HH fields.

We consider a two-color laser field consisting of a top-hat 800-nm fundamental field with intensity (pulse duration) of 1.4×10^{14} W/cm² (17 fs) and SH intensity (pulse duration) of 2.4×10^{12} W/cm² (40 fs). The intensity of the fundamental field is kept lower than the estimated intensity in the experiments ($\sim 2.4 \times 10^{14}$ W/cm²) and its pulse duration is slightly stretched by 2 fs to make the calculated spectrograms less complicated. The wavelength of the Fourier-limit SH field is switched between 400 nm and 417 nm to consider the effects of SH wavelength. Figure 8(a) shows that adding SH wavelength of 400 nm to the Fourier-limit fundamental field is not capable of bringing up the frequency modulation of the HH fields, which corresponds to the previous studies [8,9]. By contrast, the same conditions at SH wavelength of 417 nm shown in Fig. 8(b) slightly modulate the frequency of the HH fields and give them a double-peak structure which depends on the delay between the two laser fields. Since lower-order harmonic fields have a much higher intensity compared to those near the cut-off region, we applied a broad Gaussian filter around the 29th harmonic field to moderate the intensity and get a wider view of the HH fields, in all cases.

Figure 8(c) shows that adding a slight amount of minus chirp (Gaussian-profile-equivalent $\text{GDD} = -30$ fs²) to the fundamental field, while keeping the SH field (417 nm) unchanged, results in a larger amplitude of frequency modulation. For example, the frequency modulation of the 30th harmonic field resulted by the Fourier-limit fundamental field leading to an amplitude of ~ 0.2 eV (diamonds with dashed curve) and that by the chirped fundamental field leading to an amplitude of ~ 0.4 eV (diamonds with solid curve) are compared in Fig. 8(f). The 30th harmonic field has been selected for its relatively large amplitude of frequency modulation.

Keeping the other parameters fixed and slightly increasing the intensity of the fundamental field to 1.5×10^{14} W/cm² and that of SH to 2.5×10^{12} W/cm² to keep the relative intensity of the two laser fields constant, makes the amplitude of frequency modulation much larger [Fig. 8(d)]. Comparing the frequency modulation of the 18th harmonic field in Fig. 8(c) with a lower fundamental field intensity shown by the diamonds with dashed curve in Fig. 8(g)

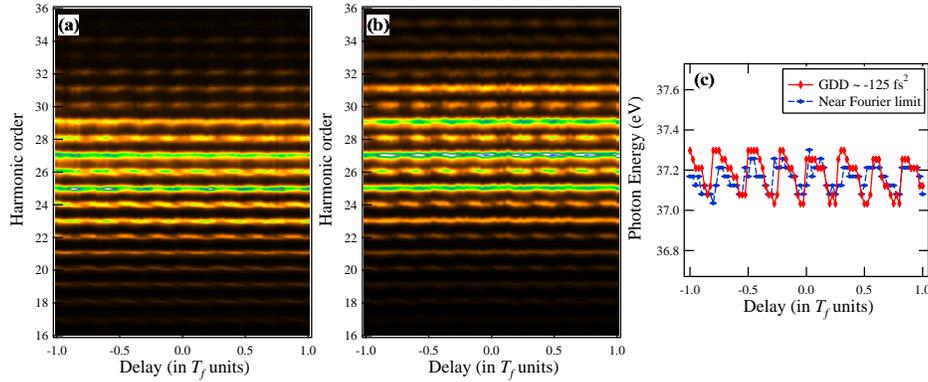


Fig. 7. Spectrograms to investigate how the chirp of the fundamental field affects the frequency modulation of the HH fields. (a) Spectrogram obtained by a two-color laser field in which the fundamental field has minus chirp ($GDD \sim -125 \text{ fs}^2$). (b) Spectrogram obtained by a two-color laser field in which the fundamental field is nearly Fourier limited. (c) Peak frequency of the 24th harmonic field extracted from panel-(a) shown by the diamonds with solid curve and that of panel-(b) (diamonds with dashed curve).

with an amplitude of $\sim 0.15 \text{ eV}$ to that of Fig. 8(d) with an amplitude of $\sim 0.3 \text{ eV}$ [diamonds with solid curve in Fig. 8(g)], shows that a higher fundamental field intensity can increase the frequency modulation amplitude. While the 18th harmonic field has been selected for its visually noticeable difference in Figs. 8(c) and 8(d), other even-order harmonic fields are also in a similar relation.

All of these observations are consistent with the experimental results in the preceding section and show that the intensity and chirp of the fundamental field can control this phenomenon and lead to a larger amplitude of frequency modulation. To make sure that SH field with wavelength of 400 nm does not lead to the same results as those obtained by 417 nm , we have calculated the spectrogram with the same parameters used in Fig. 8(d), but with SH wavelength of 400 nm [Fig. 8(e)]. The result shows that a large amplitude of frequency modulation comparable to those obtained experimentally or numerically by SH wavelength of 417 nm does not exist for even-order harmonic fields and therefore emphasizes the crucial role of the anharmonic frequency components of the SH field in this phenomenon assisted by the high intensity and minus chirp of the fundamental field.

Although the above numerical analysis reveals the important laser parameters for observation of the frequency modulation of HH fields, it does not give us a physical insight into how the frequency modulation can be controlled by alternation of the delay between the two fields. To explain this point, let us begin with Eq. (1) of Dudovich et al. [8] i.e.,

$$S_2(t, \phi) = S_1(t) - \sigma(t, \phi). \quad (1)$$

S_2 in Eq. (1) is the action in the two-color laser field and S_1 is the unperturbed action, σ is the additional phase induced by the second field, ϕ is the relative phase between the two fields, controlled by the delay. If the second field is the exact SH field ($\omega_2 = 2\omega$), then we obtain $\sigma(t + \pi/\omega, \phi) = -\sigma(t, \phi)$, which leads to generation of even-order harmonic fields.

If the central angular frequency of the second field is detuned from that of the exact SH field

by $-\delta\omega_{SH}$ ($\omega_2 = 2\omega - \delta\omega_{SH}$), as in the experiment, it satisfies

$$\cos[\omega_2(t + \pi/\omega) + \phi] = \cos[\omega_2 t + \{\phi - (\pi/\omega)\delta\omega_{SH}\}]. \quad (2)$$

Then using Eq. (2) we obtain

$$\sigma(t + \pi/\omega, \phi) = -\sigma(t, \phi - (\pi/\omega)\delta\omega_{SH}) \approx -\left(\sigma(t, \phi) - \frac{\partial\sigma}{\partial\phi}\left(\frac{\pi}{\omega}\delta\omega_{SH}\right)\right). \quad (3)$$

Provided that the fundamental pulse is sufficiently short so that $\delta\omega_{SH} \cdot \tau < 2\pi$, with τ being its pulse duration, using Eq. (3) and similar dependence of $\sigma(t, \phi)$ on the frequency of HH fields, we would expect that the even-order harmonic fields appear not exactly at $\omega_{HH} = 2n\omega$ (n : integer), but at slightly modulated frequency of $\omega_{HH} = 2n\omega - \delta\omega_{HH}$ with a modulation ($\delta\omega_{HH}$) that satisfies

$$\frac{\partial\sigma}{\partial\phi}\left(\frac{\pi}{\omega}\delta\omega_{SH}\right) - \frac{\pi}{\omega}\delta\omega_{HH} = 0. \quad (4)$$

From Eq. (4) we obtain

$$\delta\omega_{HH} = \frac{\partial\sigma}{\partial\phi}\delta\omega_{SH}. \quad (5)$$

Since $\frac{\partial\sigma}{\partial\phi}$ depends on ϕ , the modulation of the frequency of the HH fields can be controlled by alternation of the delay as shown in Eq. (5).

Further remaining issue left from the above discussion is the reason why the amplitude of the frequency modulation can be larger than the bandwidth of the fundamental field. The spectrum of the top-hat fundamental field used in the above analysis in the frequency domain has a sinc² function with broadly spanning frequency components. The main peak in this spectrum has a broad foot-to-foot bandwidth of ~ 0.45 eV (in photon energy units), and the adjacent pedestal peaks are located at ± 0.33 eV aside from the main peak. Thus, the amplitude of frequency modulation in the numerical analysis should not be directly compared to that in the experimental data obtained from a fundamental field with a finite bandwidth, shown in Fig. 1(a).

Instead, we consider multi-photon picture of HHG process contributed by many photons of the fundamental field and a single photon of the SH field to elucidate the large frequency modulation amplitude of an even-order harmonic field. The frequency of the $2n$ th harmonic field should correspond to two in-situ processes of (i) the sum of $2(n-1)$ fundamental photons ($v_{2(n-1)}$) and the SH photon (v_{SH}) and (ii) the sum of $2(n+1)$ fundamental photons ($v_{2(n+1)}$) with the difference of the SH photon. Note that in both processes an odd number of total photons are involved in even-order harmonic generation [9]. In the experiment, v_{SH} is slightly detuned from the exact SH frequency of 2ν by $-\delta v_{SH}$, resulting in $v_{SH} = 2\nu - \delta v_{SH}$. We have evaluated $h\delta v_{SH}$ to be ~ 0.125 eV. Here, ν is the frequency of the fundamental field and h is the Plank constant.

Furthermore, we have found that the peak frequency of each odd-order harmonic field is blue shifted from the exact odd-multiple frequency of the fundamental field, even without adding the SH field. For example, the energy shifts of the odd-order harmonic fields [under experimental conditions of Fig. 6(a)] are shown in Fig. 9(a). Indicating this blue shift by δv_{2n-1} , the frequency of the $(2n-1)$ th harmonic field becomes $v_{2n-1} = (2n-1)\nu + \delta v_{2n-1}$. We can approximate that δv_{2n-1} linearly depends on the harmonic order. Thus, we express $\delta v_{2n-1} = (2n-1)\delta v - q$, where q is a constant. Fitting the experimental data shown by the solid circles in Fig. 9(a) results in $h\delta v$ of 0.055 eV (slope of the linear fit shown by the dashed line). This blue shift in the multi-photon picture of HHG process can be represented as a frequency difference in the fundamental frequency to result in $v_{2(n-1)} = 2(n-1)(\nu + \delta v) - q$. Note that the spectral blue shift of the HH fields is commonly observed when the intensity of

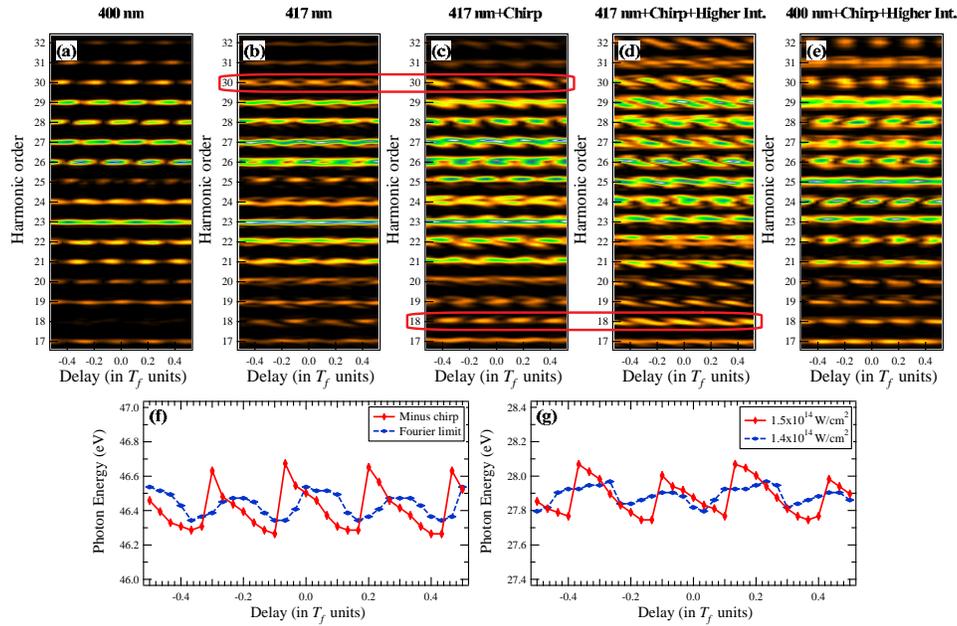


Fig. 8. Numerical analysis results to verify the effects of the wavelength of the SH field and the intensity and chirp of the fundamental field on the frequency modulation of HH fields. (a) Calculated spectrogram by SH wavelength of 400 nm added to Fourier-limit fundamental field. (b) Calculated spectrogram by SH wavelength of 417 nm added to the same fundamental field. (c) Same conditions as panel-(b), but with a chirped fundamental field ($GDD=-30 \text{ fs}^2$). The introduction of the GDD results in 17.7 fs pulse duration, for which the peak intensity of the fundamental field is kept unchanged. (d) Same conditions as panel-(c), but with a higher fundamental pulse intensity ($1.5 \times 10^{14} \text{ W/cm}^2$). (e) Same conditions as panel-(d), but with SH wavelength of 400 nm. (f) Effects of minus chirp on the frequency modulation of the 30th harmonic fields of panel-(c) (diamonds with solid curve) and panel-(b) shown by the diamonds with dashed curve. (g) Effects of intensity on the frequency modulation of the 18th harmonic fields of panel-(d) (diamonds with solid curve) and panel-(c) shown by the diamonds with dashed curve.

the fundamental field is reasonably high. This is due to the rapid change of macroscopic dipole phase and the suppression of the HH yield at the trailing edge of the fundamental field with crucial ionization of the target atoms [15], which is not included in our numerical model of single atom response.

The frequency of the $2n$ th harmonic field (ν_{2n}) generated with the in-situ process (i) is

$$\begin{aligned}
 \nu_{2n} &= \nu_{2(n-1)} + \nu_{SH} \\
 &= [2(n-1)(\nu + \delta\nu) - q] + \{2\nu - \delta\nu_{SH}\} \\
 &= 2n\nu + 2(n-1)\delta\nu - q - \delta\nu_{SH},
 \end{aligned} \tag{6}$$

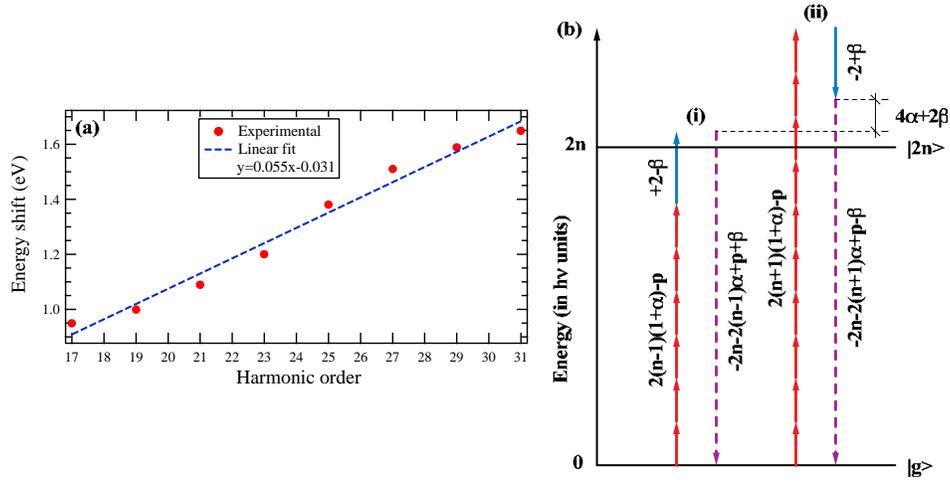


Fig. 9. Blue shift of the odd-order harmonic fields and energy level diagram showing generation of even-order harmonic fields by two in-situ processes. (a) Blue shift of the odd-order harmonic fields generated by a high-intensity fundamental field alone, expressed using photon energy shifts of the peak frequencies. Experimental data shown by the solid circles are those obtained under conditions of Fig. 6(a), showing an $h\delta\nu$ of 0.055 eV (slope of the linear fit shown by the dashed line). (b) Photon energy of the even-order harmonic fields generated by the in-situ processes (i) and (ii), expressed in units of the photon energy of the fundamental field ($h\nu$). For clarity in the figure, $\delta\nu_{SH}/\nu$ is denoted by β , $\delta\nu/\nu$ by α , and q/ν by p . This results in even-order harmonic fields with energy of $2n + 2(n-1)\alpha - p - \beta$ and $2n + 2(n+1)\alpha - p + \beta$ respectively, having energy difference of $4\alpha + 2\beta$, which is equal to $h(4\delta\nu + 2\delta\nu_{SH})$.

while that with the in-situ process (ii) is

$$\begin{aligned}
 v'_{2n} &= v_{2(n+1)} - v_{SH} \\
 &= [2(n+1)(\nu + \delta\nu) - q] - \{2\nu - \delta\nu_{SH}\} \\
 &= 2n\nu + 2(n+1)\delta\nu - q + \delta\nu_{SH}.
 \end{aligned} \tag{7}$$

Thus, the difference between the two frequencies obtained in Eqs. (6) and (7) becomes

$$v'_{2n} - v_{2n} = 4\delta\nu + 2\delta\nu_{SH}. \tag{8}$$

This situation is schematically shown in Fig. 9(b). For more clarity, energy has been shown in units of the photon energy of the fundamental field. The energy difference calculated in Eq. (8), $h(4\delta\nu + 2\delta\nu_{SH})$ is estimated to be 0.47 eV, which is sufficiently large to explain that the experimental maximum frequency modulation amplitude [~ 0.4 eV in Fig. 5(b) or the 28th harmonic field in Fig. 6(a)] exceeds the bandwidth of the fundamental field. Frequency modulation can appear by changing the contributions of the two processes due to alternation of the delay between the two laser fields. Considering the resolution of the XUV spectrograph, a frequency modulation with an amplitude larger than ~ 0.15 eV can be measurable. To get such an amplitude without considering the assist of the blueshift due to ionization, SH field at a central wavelength detuned from 400 nm by more than 10 nm could be enough to observe this phenomenon.

5. Summary

By performing a high-resolution correlation experiment using a two-color synthesized laser field consisting of the fundamental field of a terawatt 15-fs laser system and its SH field, we have been able to observe delay-dependent periodical frequency modulation of HH fields, for the first time. The amplitude of the frequency modulation depends on the intensity and chirp of the fundamental field as demonstrated by the experimental results. The numerical analysis revealed the crucial role of the anharmonic frequency components of the SH field in this phenomenon and the obtained results with SH wavelength of 417 nm highly approve of the experimental results and show that a fundamental field with a slight amount of minus chirp and a higher intensity can result in a larger amplitude of frequency modulation. Also, the frequency modulation amplitude has been estimated by considering the frequency difference of even-order harmonic fields generated by two in-situ processes when the SH field has anharmonic frequency components and the HH fields are blue shifted due to the high intensity of the fundamental field. The estimated amplitude agrees with the experimentally observed one. This novel phenomenon could be applied in in-situ precise control of the intensity and pulse duration of attosecond pulse trains.

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