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Coherent control of high-order harmonics generated with intense femtosecond laser pulses

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Abstract. Experimental results and theoretical analysis on the coherent control of high-order harmonics with chirped femtosecond laser pulses are presented. The coherent control of high-order harmonic generation resulted in sharp harmonic spectra by compensating for induced harmonic chirp with the control of applied laser chirp and it was found to be crucial also in producing sharp and bright harmonics.

PACS. 42.65.Ky Frequency conversion; harmonic generation, including higher-order harmonic generation – 32.80.-t Photon interactions with atoms – 42.65.Re Ultrafast processes; optical pulse generation and pulse compression

Advances in high-power femtosecond laser technology have made it possible to investigate high harmonic generation processes in a high-intensity regime. Owing to the expectation that it may be utilized to develop ultrashort extreme ultraviolet sources, the subject of high harmonic generation has drawn considerable attention from researchers working in the area of intense field-atom interaction. The coherence of high-order harmonics from the pump laser pulse is very good [1,2], and the duration of a high-order harmonic pulse can be much shorter than the duration of a driving laser pulse [3,4]. Thus, for applications such as X-ray interferometry [2,5] or femtosecond time-resolved X-ray spectroscopy [6], the harmonic X-ray sources have definite advantages over other sources such as synchrotrons [7] and X-ray lasers [8]. For diverse applications of the high harmonics as a coherent femtosecond X-ray source, however, the harmonic spectrum and conversion efficiency still needs to be improved. Recent investigations have shown that the harmonics are negatively or positively chirped and this chirp induces the broadening of the harmonic spectrum, decreasing the brightness of harmonic beams [9]. Due to the intensity-dependent harmonic phase, the negatively chirped harmonics are produced in a rapidly increasing laser field [10]. On the other hand, when the propagation of the laser pulse in the nonlinear medium is considered, harmonics can have positive chirp due to the self-phase modulation (SPM) of the propagating laser pulse [9,11]. The laser pulse can attain positive chirp in an ionizing medium, and this induced laser chirp is directly transferred to the temporal structure of harmonics and positively chirped harmonics are emitted. Therefore, the active manipulation of temporal structure of the laser pulse is needed to coherently control the high-order harmonic generation process for the production of spectrally sharp and strong harmonics.

In this work, we present the temporal structure of high-order harmonics using a time-frequency analysis technique and experimentally demonstrate the coherent control of high-order harmonics using chirped femtosecond laser pulses.

The interaction between an intense laser field and an atom can be described using a time-dependent Schrödinger equation (TDSE). As the intense laser field can provide an electric field comparable to or stronger than the Coulomb field in an atom, a perturbation approach cannot be used and TDSE needs to be solved numerically. The high-order harmonic spectra calculated for neon atom exposed to a laser pulse of duration of 27 fs and intensity 1×10^{16} W/cm^2 are shown in Figure 1. The numerically obtained spectrum in Figure 1a shows a very irregular noise-like harmonic spectrum, making it nearly impossible to distinguish individual harmonic as shown in Figure 1a. When the harmonic components other than the short path component are removed using the coherent sum method [12], well-defined harmonic peaks appeared as shown in Figure 1b. Thus, the irregular structure comes from the contribution from several different harmonic components.

At the higher-order harmonics from $59\omega_0$ to $99\omega_0$, the harmonic spectrum still shows a complicated structure even after the coherent sum (see Fig. 2b). For clear understanding of this result, we apply the Wigner distribution (WD) [13] function in the analysis of high-order harmonics [14] to reveal the harmonic structure in time-frequency domain. In Figure 2b the WD corresponding to Figure 2a is shown. The temporal variation of harmonic

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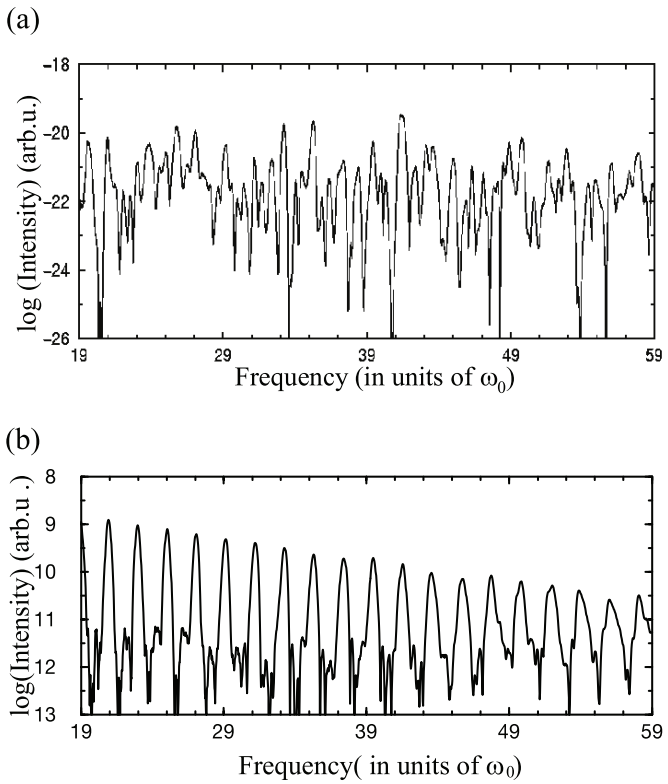

Fig. 1. High-order harmonic spectrum from a neon atom irradiated with a 27-fs, 817 nm laser pulse of intensity $I = 1 \times 10^{16}$ W/cm^2. (a) Single-atom harmonic spectrum and (b) harmonic spectrum using the coherent sum method.

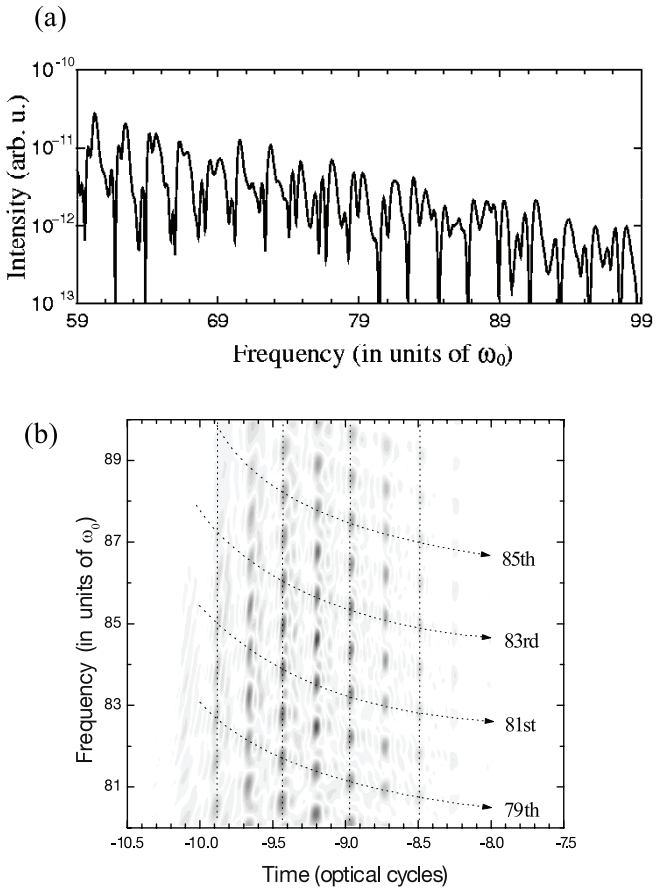

Fig. 2. (a) High-order harmonic spectrum between $75\omega_0$ and $99\omega_0$, and (b) Wigner distribution of high-order harmonics for the same parameters as in Figure 1. Positive values are colored black, and negative values are colored white (in arbitrary units).

frequency indicated by the dotted lines shows the frequency change of harmonics in time. In spite of a large frequency spread greater than $2\omega_0$, the interval between adjacent harmonics, the spectral structure of harmonics is resolved in this time-frequency analysis. The WD clearly shows that harmonics are blueshifted and negatively chirped. For example, in the case of the 85th harmonic, as time evolves it sweeps from $90\omega_0$ to $87\omega_0$, with a peak near $(87.5-88.2)\omega_0$. As a consequence, the large spectral spreading due to the chirp causes neighboring harmonics of different orders to overlap and the distinction of individual harmonic is not possible in the Fourier spectrum of harmonics shown in Figure 2a.

In addition to the single atom effect of harmonic chirp, the propagation effect of the pump laser pulses in ionizing medium has to be taken into account for the full understanding of temporal structure of the harmonic beam. At laser intensity above the saturation intensity of optical-field ionization, the ionization effect in the leading edge of the laser pulses is not negligible. The self-phase modulation (SPM) of the laser pulse propagating in an ionization medium alters its spectral structure and temporal shape. The change of the spectral structure of the laser pulse can be revealed in the time-frequency domain using the Wigner distribution function. The medium (neon) density and length for the results in Figure 3 are 10^{18} cm^{-3} and 0.7 mm, respectively. It is seen in Figure 3 that, due to the phase modulation induced by the plasma formation, the frequency of laser pulse increases with time (becomes positively chirped) in the leading edge of the pulses and this effect is more dramatic when we increase the laser intensity from 1×10^{15} W/cm^2 (Fig. 3a) to 1×10^{16} W/cm^2 (Fig. 3b). When the laser intensity is sufficiently high enough to significantly ionize the atoms, high-order harmonics mostly occur much earlier in time than the peak of the laser pulse [15,16]. The positively chirped structure in the leading edge of the laser pulse will be directly transferred to the frequency of harmonics. Consequently, when this SPM-induced positive chirp becomes larger than the dynamically induced negative chirp (harmonic chirp from single atom effect), the sign of harmonic chirp changes from negative to positive [11,17].

We experimentally examined the effect of laser chirp on the spectral structure of high-order harmonics emitted from neon atoms. In our experiments, Ti:sapphire laser pulses with a duration of 27 fs and a center wavelength of 820 nm were applied to an elongated gas jet from a 9×0.5 mm^2 slit nozzle. The peak gas pressure was 40 torr. The laser chirp condition was controlled by changing the grating separation in the pulse compressor. The reduction

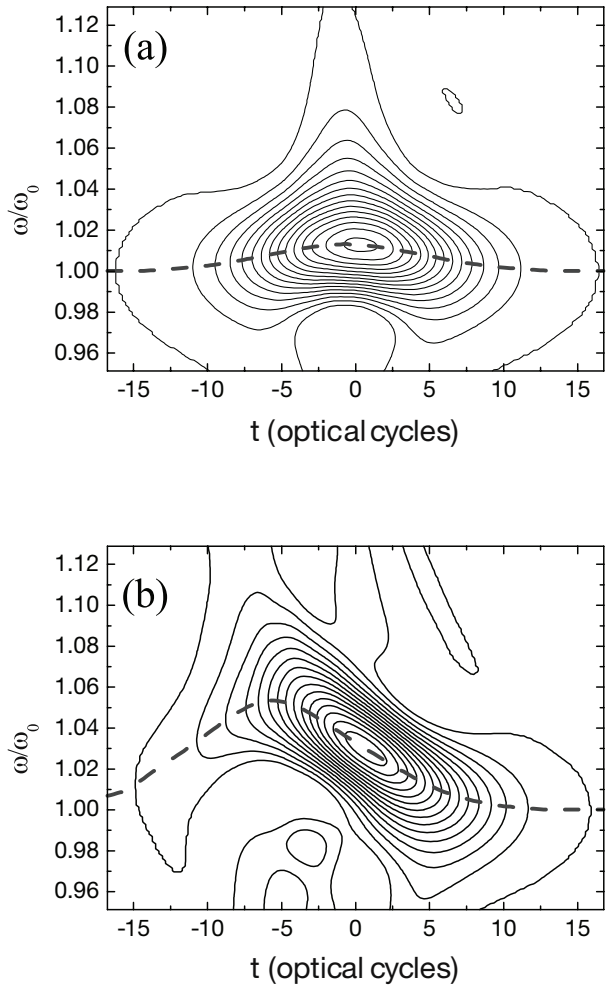

Fig. 3. Wigner distributions of 30-fs, 800 nm laser pulses after propagating through a Ne gas medium of length $l = 0.7$ mm and density $n_0 = 10^{18}$ cm^{-3}. The laser intensity is given by (a) $I = 1 \times 10^{15}$ W/cm^2 and (b) $I = 1 \times 10^{16}$ W/cm^2.

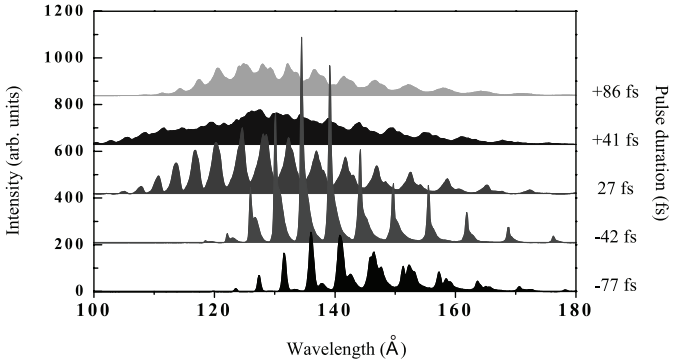

Fig. 4. Experimentally observed harmonic spectra with the 9-mm long gas jet at $z = -15$ mm driven by 27-fs laser pulses with different laser chirps. The peak laser intensity of the chirp-free pulse and neon density were $I = 1 \times 10^{15}$ W/cm^2 and 40 torr, respectively.

of the grating separation from the chirp-free condition generates positively chirped pulses, and the increase of the separation provides negatively chirped pulses. The pulse duration and phase information of chirped laser pulses were characterized with the second-harmonic generation frequency-resolved optical gating technique. The generated harmonics from the interaction of laser pulses with a gas jet were detected using a flat-field extreme-ultraviolet (XUV) spectrometer equipped with an X-ray charge-coupled device (CCD). Zr X-ray filters with a total thickness of 0.4 μm were used in front of CCD to block scattered light in the XUV spectrometer.

Figure 4 shows the harmonic spectra from the 9-mm long gas jet irradiated with a chirp-free 27 fs laser pulse and with negatively and positively chirped laser pulses. The gas jet was placed -15 mm before the laser focus to increase the harmonic intensity by reducing the plasma defocusing effects with converging laser pulses [18]. The peak laser intensity of the chirp-free pulse at the center of the gas jet was 1×10^{15} W/cm^2. With positively chirped pulses the harmonic spectra became weaker and broader than that of the chirp-free case. On the other hand, with negatively chirped pulses the harmonics became sharp and strong, but the cutoff harmonics turned out to be disappearing and the shortest harmonic wavelength increased. When harmonics are produced with negatively chirped pulses of 42 fs, the harmonic of 13.4 nm wavelength was so strong that it saturated the X-ray CCD in a single laser shot of 4.5 mJ. This shows that, in the case of 9-mm long propagation with sufficiently high laser intensity for OFI, the harmonic chirp structure is strongly affected by the SPM-induced laser chirp in an ionizing medium and, therefore, the total harmonic chirp became positive.

In conclusion, we have investigated the time-frequency characteristics of high harmonics and the plasma induced laser chirp using the Wigner distribution analysis, and demonstrated that high harmonic generation processes can be coherently controlled using chirped femtosecond laser pulses to produce sharp and strong harmonic spectra. This shows that a rigorous analysis of harmonic spectral structure is crucial for proper understanding of the interaction processes between intense femtosecond laser field and atoms. In other words, the proper coherent control of high harmonics must be based on the integral treatment of the interaction between atoms and a driving laser pulse.

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