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Continuously tunable high-order harmonics from atoms in an intense femtosecond laser field

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Through the control of laser energy and chirp, continuous wavelength tuning of high-order harmonics was achieved without sacrificing spectral sharpness. This process was implemented after the amplification stage in a chirped-pulse amplification Ti:sapphire laser, without changing laser spectrum. It opens a new pathway for the realization of a continuously tunable, coherent, femtosecond x-ray source, i.e., a tabletop synchrotron.

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When gaseous atoms are exposed to an intense femtosecond laser field, the periodic modulation of the electron motion produces high-order harmonics of the laser frequency. High-order harmonics possess unique properties such as good coherence [1,2] and low beam divergence [3], as well as extremely short pulse duration [4,5], inherited from the driving laser. Although numerous harmonic wavelengths are generated, the applicability of such harmonic x-ray sources is restricted since only a discrete set of harmonic wavelengths, equal to the incident laser wavelength divided by odd integers, are available. On the other hand, synchrotron x-ray sources can provide continuously tunable x-ray radiation. However, access to synchrotron sources is limited and they usually generate x-ray pulses longer than a few tens of picoseconds [6]. The addition of tunability to harmonic x-ray sources would, therefore, offer tunable, coherent, femtosecond (or attosecond) x-ray pulses with peak brightness far greater than that of synchrotron x-ray pulses. Here, we demonstrate continuous wavelength tuning of the high-order harmonics, by simply adjusting the incident laser energy and the chirp of intense femtosecond laser pulses, without sacrificing spectral sharpness and brightness of the harmonics.

Wavelength tuning of high-order harmonics has been attempted using several different approaches. In one method, a laser pulse at the fundamental wavelength was mixed with a second pulse at a different wavelength, obtained by optical parametric generation. It has been shown that driving atoms with laser pulses mixed from two different wavelengths could generate tunable harmonics [7]. In this case, however, the new harmonics added, due to the mixing of the two pulses, were generally much weaker than those produced by the first laser pulse alone. In another method, a shift of the harmonic wavelengths was induced by the adjustment of the target position with respect to the laser beam focal spot [8]. In this case, since the applied laser intensity varies with target position, the harmonics produced at different target positions exhibited different amounts of wavelength shift. The spectral widths of the harmonics generated at different target positions, however, became quite broad, as the target position was moved away from the laser beam focus. Another possibility is the tuning of harmonic wavelengths through the tuning of the driving laser wavelength by using the optical parametric amplification technique. Though the harmonic wavelength could be tuned by this technique, the harmonic spectrum obtained was not sharp and bright [9]. Tuning of the harmonic wavelength by changing the oscillator spectrum is not practical because the adjustment of the oscillator spectrum cannot be directly transferred to the final laser spectrum due to gain narrowing and gain saturation processes in a chirped-pulse amplification (CPA) laser [10]. Moreover, the generation of the shortest possible pulse in the CPA femtosecond laser, which is needed for the production of very high-order harmonics [11] with good conversion efficiency [12], requires the use of the entire laser spectrum obtainable. This requirement severely limits the spectral tuning range of the laser. Thus, it would not be desirable to attempt to tune harmonic wavelengths by changing the laser spectrum.

We propose a new method to continuously tune the harmonic wavelengths without modifying the driving laser spectrum. Our previous experimental results [13] on the harmonics from argon, driven by 30-fs laser pulses, showed a large harmonic blueshift measuring up to twice the laser frequency—the interval between odd harmonics. When an intense femtosecond laser pulse is applied to atoms, the atoms experience a nonadiabatic increase of the laser field from one optical cycle to the next. The nonadiabatic response of the atoms to a rapidly rising laser field results in large blueshift of high-order harmonic wavelengths [13,14]. However, control of the laser intensity to induce the harmonic blueshift was accompanied by a modification of the spectral shape and brightness of harmonics, not a desirable feature for realistic applications of harmonic x rays. The spectral structure of high-order harmonics contains chirp components, i.e., positive chirp due to the self-phase-modulation (SPM) of the driving laser pulse propagating through an ionizing medium [15,16] as well as a dynamically induced negative chirp [17,18]. Since harmonic chirp results in a broadening of the harmonic spectrum, it should be appropriately compensated for to achieve sharp harmonics. In addition, the observed range of continuous tuning was limited. To achieve full continuous tuning, the harmonic blueshift should be much larger than twice the laser frequency. Consequently, we have explored the wavelength tunability of high-order harmonic x rays on the basis of the harmonic blueshift in helium atoms using chirped femtosecond laser pulses and have achieved continuous tuning of harmonics in the 10-nm soft x-ray region without sacrificing spectral sharpness.

Experiments on continuously tunable high-order harmonic generation were carried out using a femtosecond tera-

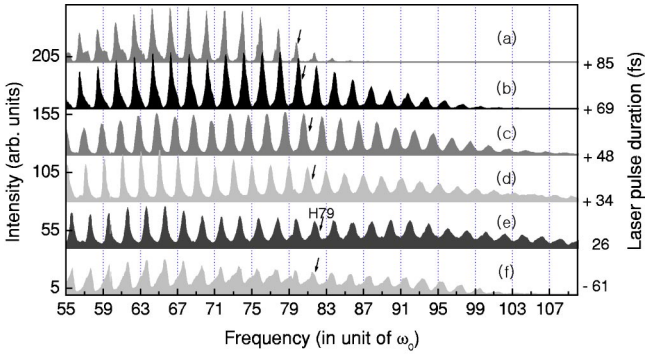

FIG. 1. High-order harmonic spectra from helium atoms obtained with positively chirped [(a)–(d) with positive signs], chirp-free (e), and negatively chirped pulses [(f) with negative sign]. The chirp-free laser pulse had a duration of 26 fs and an intensity of 1×10^{15} W/cm^2, and the helium density was 3×10^{19} cm^{-3}. The arrows indicate the 79th harmonic (H79).

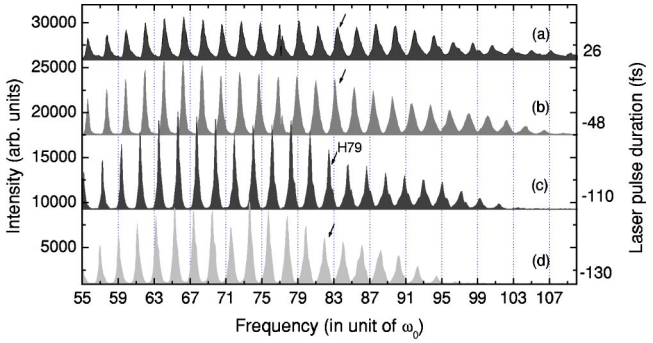

FIG. 2. High-order harmonic spectra from helium atoms obtained with chirp-free (a) and negatively chirped pulses [(b)–(d)]. The laser intensity of a chirp-free pulse was 2×10^{16} W/cm^2.

watt Ti:sapphire laser operating at 10 Hz [10]. The laser spectrum was centered at 824 nm with a spectral bandwidth of 47 nm. A helium jet, operating at 10 Hz, was positioned 2 mm after the laser beam focal spot to generate single-peaked harmonics [19]. The helium density profile in the interaction region had a peak value of 3×10^{19} cm^{-3}, with a full width at half maximum of about 0.5 mm. High-order harmonic spectra were detected by a flat-field extreme-ultraviolet (XUV) spectrometer equipped with a back-illuminated x-ray charge-coupled device (CCD) with 330×1100 pixels (Princeton Instruments). The spectral resolution set by the CCD pixel size is about 0.02 nm in the 10 nm region. Two zirconium filters of total thickness 0.4 μm were installed in front of the x-ray CCD to block stray laser light.

Helium was selected as the target medium because it is capable of providing a sufficiently large blueshift in the wavelength region near 10 nm. The harmonic intensity and range of observed harmonic orders depend on the target medium and the laser parameters. Decreasing the atomic number of the target medium decreases the intensity of the harmonics, but increases the highest obtainable harmonic order. Operating at short-wavelength harmonics helps to increase the range of harmonic blueshift, because the amount of harmonic blueshift increases with harmonic order [13]. The relatively weak harmonic intensity in helium can be overcome by the use of a high-density medium as the target, which also helps to increase harmonic blueshift because of enhanced SPM. Thus, a high-density helium gas jet can provide strong harmonic generation with sufficiently large blueshift.

The wavelength tunability of high-order harmonics was first attempted by controlling only the laser chirp. Similar investigations were carried out by Chang *et al.* [17], but the gas density employed in our experiment was two orders of magnitude higher and the pulse duration was much shorter. The laser chirp was changed by the adjustment of the grating separation in the pulse compressor. With the incident laser energy fixed, harmonic spectra from He were obtained with different amounts of laser chirp [20]. Figure 1 shows six harmonic spectra obtained with 26-fs chirp-free pulse and positively and negatively chirped pulses. The chirp-free laser intensity was 1×10^{15} W/cm^2. The arrows in Figs. 1(a)–1(f) indicate the 79th harmonic obtained with different amount of laser chirp. The frequency of 79th harmonic is blueshifted by about $0.7\omega_0$ (ω_0=laser frequency) in the case of the chirp-free pulse [Fig. 1(e)]. The 79th harmonic produced with positively chirped 85-fs pulses shows a frequency redshift of about $2\omega_0$ from that of chirp-free pulses. The intensity reduction due to the elongated pulse duration of a chirped laser pulse decreases the harmonic blueshift [13]. Moreover, harmonics produced with positively chirped laser pulses are redshifted because harmonics produced in the leading edge of the laser pulse come from the red part of the laser spectrum. Since these two effects—reduced laser intensity and positively chirped laser spectrum—both contribute towards harmonic redshift, harmonic frequencies become redshifted with positively chirped pulses. Though the frequency shift of harmonics is large enough for continuous tuning in this case, the spectral sharpness changes with the laser chirp.

Since the wavelength tuning with the control of laser chirp only was not satisfactory, the frequency shift of harmonics was investigated at much higher laser intensity. Under intense laser field, atoms are rapidly ionized in the leading edge of the laser pulse, allowing strong harmonic generation only in the leading edge [21]. The harmonics emitted are then strongly blueshifted due to both the nonadiabatic effect [13,14] and SPM [22,23] of the driving laser pulse. The nonadiabatic effect is basically a single atom effect, because it originates from the atomic response to the rapidly rising intense laser field. On the other hand, the blueshift due to SPM is a result of the propagation of a driving laser pulse through harmonic generation medium. In the ionizing medium the refractive index decreases with time, which causes SPM of the propagating laser pulse [24]. The SPM then causes a blueshift of the laser spectrum and, as a result, the high-order harmonics are also blueshifted. As the harmonic blueshift, due to both nonadiabatic and SPM effects, increases with laser intensity, a large blueshift can be obtained with intense laser pulses. As shown in Fig. 2, the harmonics, obtained with laser pulses for chirp-free intensity of 2×10^{16} W/cm^2, experience very large blueshift equal to $3\omega_0$ for 59th order and $6\omega_0$ for 99th order in the case of a chirp-free pulse.

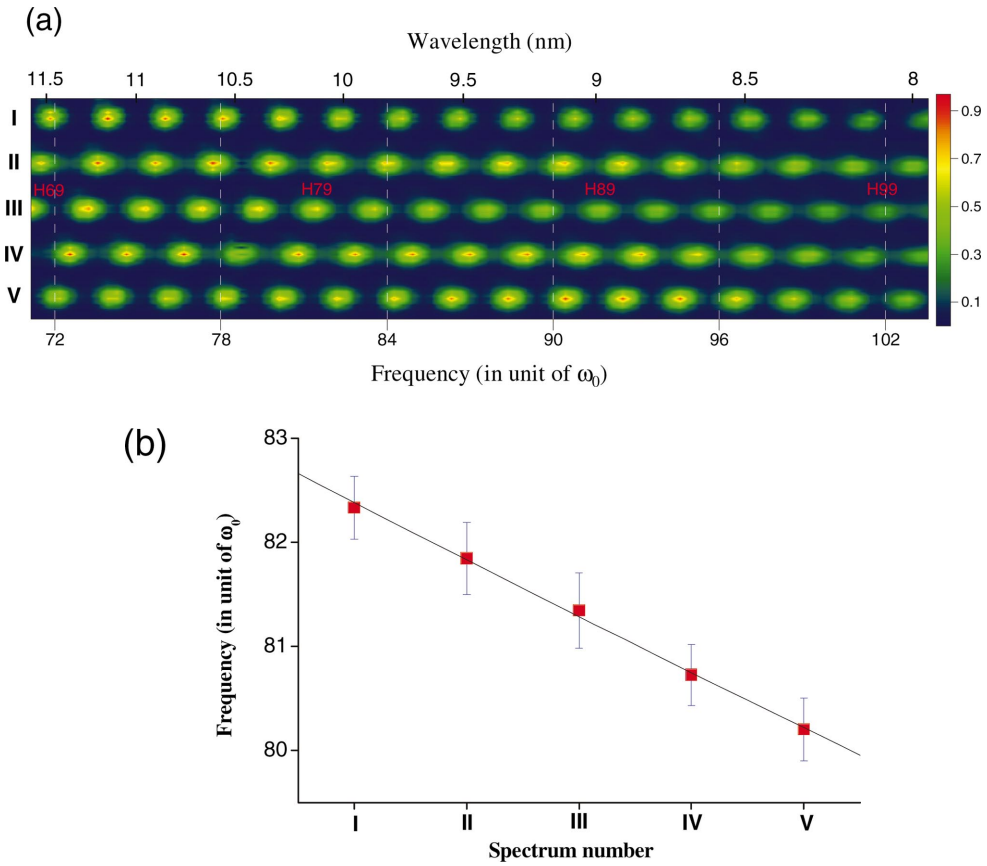

FIG. 3. (Color) Continuously wavelength-tuned high-order harmonics generated from helium atoms driven by intense femtosecond laser pulses. The wavelength tuning was achieved by adjusting the incident laser energy and laser chirp. (a) Spectral images of harmonics taken using an x-ray CCD. The laser energy and chirped pulse duration of five harmonic spectra were (I) 11 mJ and -110 fs, (II) 5.9 mJ and -87 fs, (III) 4.1 mJ and -47 fs, (IV) 1.8 mJ and 26 fs, (V) 1.0 mJ and $+44$ fs, respectively. The negative (positive) sign in the pulse duration means negatively (positively) chirped pulse. (b) Frequency and spectral width of the 79th harmonic of the five spectra shown in (a).

At high laser intensity, the harmonic chirp is dominated by the SPM-induced positive chirp and sharp harmonics are generated with negatively chirped laser pulses [15]. In this case an intense laser pulse propagating through an ionizing gaseous medium acquires a positive chirp due to SPM [16]. Figures 2(a)–2(d) show the harmonic spectra obtained with chirp-free and negatively chirped laser pulses. The chirp-free laser intensity was 2×10^{16} W/cm^2. The sharpest harmonics were produced with negatively chirped 110-fs laser pulses, as shown in Fig. 2(c), because of the compensation of the induced positive chirp by the negatively chirped laser pulses. Since the frequency redshift of harmonics due to the intensity reduction is partially canceled by the negatively chirped laser spectrum that sets the blue component of laser spectrum in the leading edge of the laser pulse, the frequencies of harmonics emitted in this case did not shift significantly from those of the chirp-free case and were still strongly blueshifted, as seen clearly from the comparison of Figs. 2(a) and 2(d). From Fig. 2(c) we observe that the harmonic of 55th order was blueshifted by more than twice the laser frequency and the harmonic of 97th order by more than four times the laser frequency.

Continuous tuning of high-order harmonics without losing spectral sharpness was achieved by controlling both incident laser energy and laser chirp. The laser intensity was varied by controlling the incident laser energy using a half-wave plate and polarizers installed in front of the pulse compressor. Spectral images of the continuously tuned high-order harmonics obtained from helium atoms are presented in Figs. 3(a) and 3(b). The gas density was the same as in Figs. 1 and 2. The wavelength of the harmonic spectrum was calibrated using argon spectral lines from Ar^{7+} and Ar^{8+} ions. Continuous wavelength tuning of sharp harmonics was achieved by changing the laser parameters from a negatively chirped 110-fs, 11-mJ pulse in spectrum (I) of Fig. 3(a) to a positively chirped 44-fs, 1.0-mJ pulse in spectrum (V). The laser chirp was controlled to generate sharp harmonics. As the incident laser energy increases, the laser chirp needed for sharp harmonics changes from positive to negative since the SPM-induced positive harmonic chirp increases with laser intensity. The harmonics of order greater than or equal to 73 in spectrum (I) overlap with the next higher-order harmonics in spectrum (V). Here we selected the harmonic frequencies such that the 79th harmonics of the five spectra in Fig. 3(a) had equal frequency intervals between neighboring spectra, demonstrating the continuous wavelength-tuning capability. The wavelength shifts of the 79th harmonic of the five harmonic spectra can be fitted by a straight line, as shown in Fig. 3(b). The spectral widths were maintained within $0.7\omega_0$

by the adjustment of the laser chirp condition. This represents a direct proof that it is possible to generate a sharp harmonic at a specific wavelength by controlling incident laser energy and laser chirp, a critical step in the realization of a continuously tunable high-order harmonic x-ray source.

The continuous tuning range of harmonics below the 71st order was slightly less than the full tuning range. The extension of the full tuning range to longer wavelength should be achievable using a higher medium density. Alternatively, adoption of a medium of higher atomic number such as neon, argon, or xenon, would also facilitate tuning at longer wavelengths because of larger blueshift of these atoms. The proposed method can be easily implemented in most harmonic experiments that use CPA lasers, because the continuous wavelength tuning is accomplished simply by adjusting the angle of the half-wave plate (for laser-energy control) and the grating separation in the pulse compressor (for chirp control).

In conclusion, we have demonstrated the continuous wavelength tuning of high-order harmonics using the property of harmonic blueshift and the chirp control of harmonics with chirped femtosecond laser pulses for spectral sharpness and brightness. The wavelength-tuning method proposed here requires only the rotation of the half-wave plate for laser-energy control and the adjustment of the grating separation in the pulse compressor for chirp control, which can be easily automated with a feedback control. Hence, the wavelength-tuning method demonstrated here has strong potential as a practical tool for applications. Tunable harmonic x-ray sources open the path to XUV or x-ray nonlinear optics, ultrafast x-ray absorption spectroscopy, and photoelectron spectroscopy in atomic, molecular, and biological materials, with superb coherence and ultrashort time resolutions not accessible with synchrotron x-ray sources. Tunable high-order harmonics should also prove useful as a seeding pulse for x-ray lasers. Instead of starting an x-ray laser from an amplified spontaneous emission (ASE), an injection-seeded x-ray laser from a high-order harmonic x-ray can have much better coherence properties and shorter pulse duration than those achievable with an x-ray laser based only on ASE. Therefore, the tunability of high-order harmonic x ray sources will significantly increase the diversity and versatility in applying high-order harmonic x-rays to many unexplored areas.

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