Generation of Nonadiabatic Blueshift of High Harmonics in an Intense Femtosecond Laser Field

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By applying intense 30-fs Ti:sapphire laser pulses to Ar gas well above the saturation intensity for optical-field ionization, a strong blueshift of high harmonics 2 times as large as the laser frequency was generated. A semiclassical calculation showed that the observed large blueshift resulted from a rapidly increasing electric field, existing much earlier in time than the peak of the laser pulse, i.e., a nonadiabatic effect.

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With the technological advancement of intense femtosecond lasers, experiments on high-harmonic generation (HHG) have extended its emission wavelengths and improved its efficiency [1,2]. The understanding of the HHG processes has also progressed along with theoretical developments, especially semiclassical theory [3] and the strong-field approximation (SFA) model [4]. The former provided the basic picture of HHG, i.e., tunneling ionization under strong laser field, acceleration of an ionized electron along the field, and generation of high harmonics during its recombination after gaining kinetic energy. The SFA model has been successful in explaining various experimental observations by providing direct physical interpretations of HHG processes [5,6]. One important feature derived from SFA is that the phase of dipole moment induced on an atom by a driving laser, or equivalently harmonic phase, depends on the laser intensity. This property of intensity-dependent harmonic phase led to the prediction that the blueshift of harmonics would occur in a rapidly increasing electric field due to a nonadiabatic response of atoms to the field [7]. The blueshift observed in previous experiments [8,9], however, was explained by considering the change of the refractive index during ionization, which modulates the spectrum of a propagating laser pulse—the self-phase modulation (SPM) in an ionizing gas. In this Letter we report the first experimental generation of a nonadiabatic blueshift of high harmonics twice as large as the laser frequency, and we show that a semiclassical calculation based on the SFA model can provide a good explanation of the observed blueshift.

When atoms are exposed to a laser field with large electric-field variation over one optical cycle, the high harmonics emitted would reflect the nonadiabatic response of the atoms. A large electric-field variation, achievable from an extremely short (~10 fs) laser pulse, can also be found in a slightly longer (~30 fs) laser pulse if the temporal structure is carefully looked into. The electric-field variation of a laser with a Gaussian temporal profile increases linearly from zero at the peak of the laser pulse as time goes negative. Figures 1(a) and 1(b) show the electric-field profile and relative electric-field variation of a 30-fs Gaussian laser pulse. If high harmonics are generated by a femtosecond laser with an intensity less than the saturation intensity for optical-field ionization (OFI), most of the harmonics are emitted near the peak of the laser pulse. In this case high harmonics would exhibit modest spectral broadening due to the negative chirp of harmonic pulse [5,10]. The quadratic time dependence of the harmonic phase near the peak of the laser pulse generates blueshift in the rising side of the laser field and redshift in the falling side, resulting in the modest spectral broadening due to the small electric-field variation near the peak of the laser pulse. However, if the intensity of a femtosecond laser significantly exceeds the saturation intensity, HHG would occur much earlier in time than the peak of the laser pulse, reflecting the characteristics of atoms exposed to a laser field with large electric-field variation.

High-harmonic emission from a single atom can be obtained by calculating the dipole acceleration of a returning electron which is ionized and accelerated by an oscillating electric field. Figures 1(c) and 1(d) show the temporal distributions of dipole acceleration for harmonics between the 21st and 39th orders from an Ar atom driven by a 30-fs Ti:sapphire laser pulse with peak intensities of 3×10^{14} and 9×10^{15} W/cm^2, respectively. The dipole acceleration was calculated using the SFA model

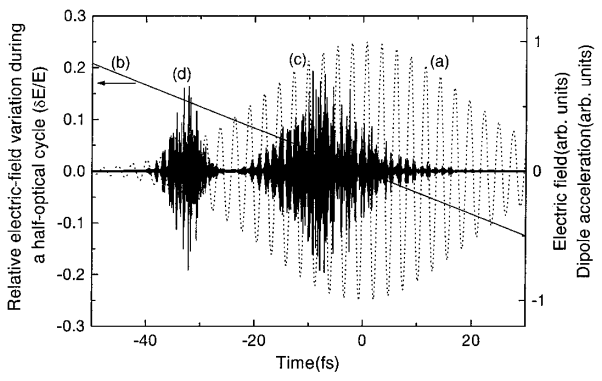

FIG. 1. Electric-field profile (a) and relative electric-field variation (b) of a 30-fs Gaussian laser pulse, and calculated dipole acceleration of a recolliding electron at the laser intensities of 3×10^{14} (c) and 9×10^{15} W/cm^2 (d).

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considering neutral depletion by the Ammosov-Delone-Krainov formula [4,11]. In the case of Fig. 1(c), with laser intensity comparable to the saturation intensity of Ar, most harmonics are generated near the peak of the laser pulse in which the electric-field variation is rather small. In the case of laser intensity far above the saturation intensity [Fig. 1(d)], harmonics are generated long before the peak of the laser pulse, and Ar atoms during strong HHG experience an electric-field variation of as much as 15% during a half-optical cycle. The decrease of recolliding electron phase due to rapidly increasing electric field will result in blueshift of high harmonics—a nonadiabatic effect in HHG, which should be observable.

The HHG experiment on nonadiabatic blueshift was performed with a femtosecond chirped-pulse amplification Ti:sapphire laser operating at 10 Hz [12]. The Ti:sapphire laser consisted of a femtosecond oscillator, a reflective-type grating pulse stretcher, two multipass amplifiers, and a grating pulse compressor in vacuum. It utilized a long-wavelength injection method to generate a broad amplified spectrum. The laser beam was focused into a gas jet for HHG. A gas jet with a small nozzle of 200-μm diameter was used to confine the interaction region well within the confocal parameter of 10 mm. The interaction region was set to 250 μm above the nozzle tip; the full width at half maximum of the gas-density profile was about 700 μm. The gas-density profile was measured by applying a focusing method [13], which detected the refraction of incident light due to nonuniform gas-density distribution. The generated harmonics 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 XUV spectrometer employed a toroidal mirror in front of the entrance slit of the spectrometer to compensate for the astigmatism occurring at the grazing-incidence concave grating; this also served to enhance x-ray collection efficiency [14]. The resolving power of the spectrometer was better than 1000, limited mainly by the pixel size of the CCD, thus allowing a high-resolution detection of harmonic spectra.

The generation of strong high harmonics requires the application of an intense laser field to an interacting medium while neutral atoms are abundant. Figure 2 shows the HHG spectra obtained from Ar gas driven by a 30-fs Ti:sapphire laser with spectral bandwidth of 41 nm centered at 813 nm. The laser was focused on the center of the gas jet with laser intensity ranging from 3×10^{14} to 9×10^{15} W/cm^2. Laser intensity was adjusted by controlling laser energy before the pulse compressor using a combination of a half-wave plate and dielectric polarizers. Great care was taken to prevent any spectral modulation during energy control. The temporal characteristics obtained using a second-harmonic generation frequency-resolved optical gating showed a pulse shape close to Gaussian with a duration of 30 fs. Observation of harmonics was performed using Al filters with a total thickness of 0.4 μm in front of the CCD to block scattered laser light in the XUV spectrometer. The harmonic emissions from Ar with a peak density of 8×10^{17} cm^{-3} were observed in orders 23 through 39, as shown in Fig. 2. As laser intensity increased, a strong blueshift of harmonics was clearly visible. The 39th harmonic generated in the laser intensity of 9×10^{15} W/cm^2 shifted twice as large as the laser frequency, that is, the 39th order harmonic was observed at the position of the unshifted 41st order harmonic. The harmonic blueshift therefore could cover the whole frequency interval between adjacent orders and offers means of generating a tunable coherent femtosecond XUV source.

Figures 3(a) and 3(b) show the summarized results of the density and intensity dependence of the harmonic blueshift, respectively. For analysis of the blueshift of high harmonics, density dependence was first examined to determine the contribution from SPM. A laser pulse propagating through an ionizing gas is known to shift its spectrum towards a shorter-wavelength region due to SPM in an ionizing gas. The frequency shift due to the refractive-index change in an ionizing gas is proportional to the gas density [8,9], since the OFI rate is independent of gas density. The amount of frequency shift due to SPM was easily identified by looking at the density dependence of the blueshift. The density dependence of the blueshift of harmonic emissions obtained with 30-fs pulses at an intensity of 9×10^{15} W/cm^2 is shown in Fig. 3(a). The peak density of Ar varied from 1×10^{17} to 8×10^{17} cm^{-3}. Even though the harmonic blueshift increased with gas density, the amount of blueshift extrapolated to zero gas density was still significant. Consequently, the harmonic blueshift observed in this experiment should have a mechanism other than SPM.

A characteristic behavior of harmonic blueshift was observed as the laser intensity was scanned, as shown in Fig. 3(b). When the laser intensity was increased from 3×10^{14} to 2×10^{15} W/cm^2, the blueshift rapidly increased to a significant value. It then slightly increased further as the intensity was raised to 9×10^{15} W/cm^2. As the amount of calculated blueshift based on a single-atom response was roughly proportional to the relative electric-field variation of a driving laser pulse [15], the qualitative understanding of the characteristic behavior could be obtained if the electric-field variation at the time of strong harmonic emission is estimated. The actual laser intensity at the time of maximum dipole acceleration was almost constant, varying only in the range of $(2-3) \times 10^{14}$ W/cm^2 as shown in Figs. 1(c) and 1(d), even though the peak laser intensity increased from 3×10^{14} to 9×10^{15} W/cm^2. It is then possible to estimate the electric-field variation by finding the time of strong harmonic emission. Consequently, in the case of Gaussian laser pulses the intensity dependence of harmonic blueshift has the following functional form, whose shape closely matches the observed characteristic behavior:

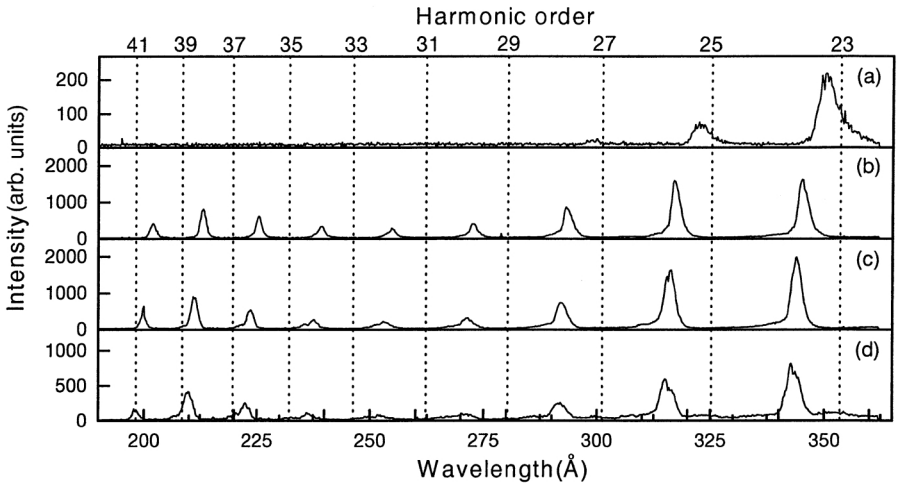

FIG. 2. High-harmonic spectra obtained from Ar driven by a 30-fs Ti:sapphire laser centered at 813 nm. Peak gas density at the interaction region was 8×10^{17} cm^{-3}; laser intensities were 3×10^{14} (a), 9×10^{14} (b), 3×10^{15} (c), and 9×10^{15} W/cm^2 (d).

$$\frac{\Delta\omega_q}{\omega_q} \propto \sqrt{\ln(I/I_p)}, \tag{1}$$

where $\Delta\omega_q$ is the frequency shift of the qth harmonic with frequency ω_q, I is the scanned laser intensity, and I_p is the laser intensity in the case of maximum harmonic emission at the peak of laser pulse.

The blueshift of harmonics from atoms in a rapidly increasing laser field can be calculated using a semiclassical theory based on the SFA model [4]. In this model, an electron first tunnels through the atomic potential modified by the laser field and appears in the continuum with zero velocity. Harmonics are then emitted when the electron driven by the oscillating laser electric field returns to the atom. The phase, θ_q, of the qth harmonic emitted by the electron may be written as [7]

$$\theta_q = q\omega_0 t_q - S(t_q), \tag{2}$$

where ω_0 is the laser frequency and t_q corresponds to the returning time of the electron with the kinetic energy of $q\hbar\omega_0 - E_I$, where E_I is the ionization potential of Ar. The classical action, $S(t_q)$, which is the accumulated phase of the wave function of an electron ionized at time t' with zero initial velocity, is given by

$$S(t_q) = \frac{1}{\hbar}\int_{t'}^{t_q} dt\left[\frac{1}{2}m_e\left(\frac{dx}{dt}\right)^2 + E_I\right], \tag{3}$$

where the first term in the integrand is the kinetic energy gained by the electron. The frequency shift of the qth harmonic is then calculated by considering the rate of harmonic-phase change:

$$\Delta\omega_q = -\frac{\Delta\theta_q}{\Delta t}. \tag{4}$$

In the plateau region, two different electron trajectories with same recolliding kinetic energy may contribute to a given harmonic, corresponding to pre- and post-3.17 trajectories of which ionized electron returns before and after the trajectory with the maximum kinetic energy of 3.17 times the ponderomotive potential of the laser field, respectively [7]. The poor phase matching of the post-3.17 trajectory during the propagation through the HHG medium, however, makes the post-3.17 harmonics less visible [7]. As a result, the calculation of the frequency shift was performed for the pre-3.17 trajectory.

The calculated fractional frequency shift for the 27th harmonic due to a Gaussian electric field of 30-fs duration is shown in Fig. 4(a), along with the experimental result obtained with Ar density of 2×10^{17} cm^{-3}. The harmonic blueshift calculated using the semiclassical theory could reproduce the characteristic behavior, i.e., the initial rapid increase and then slow increase. Quantitative comparison, however, requires consideration of the

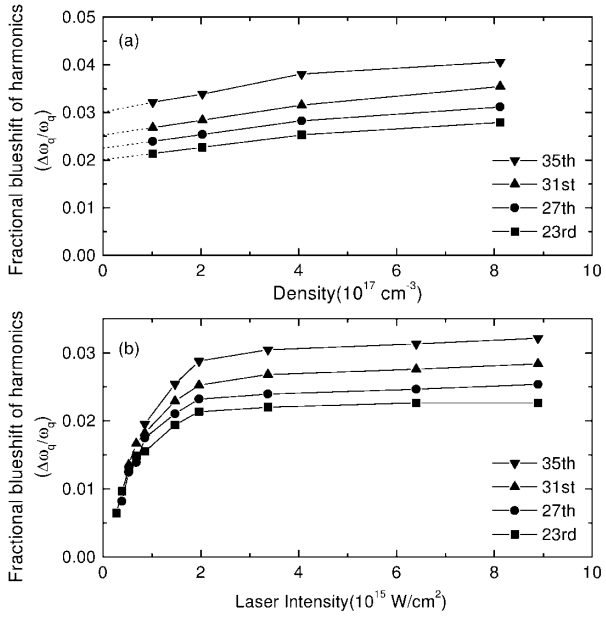

FIG. 3. Fractional blueshift of harmonics from Ar driven by a 30-fs laser pulse with respect to gas density (a) and to laser intensity (b). The laser intensity of (a) was 9×10^{15} W/cm^2, and the gas density of (b) was 2×10^{17} cm^{-3}.

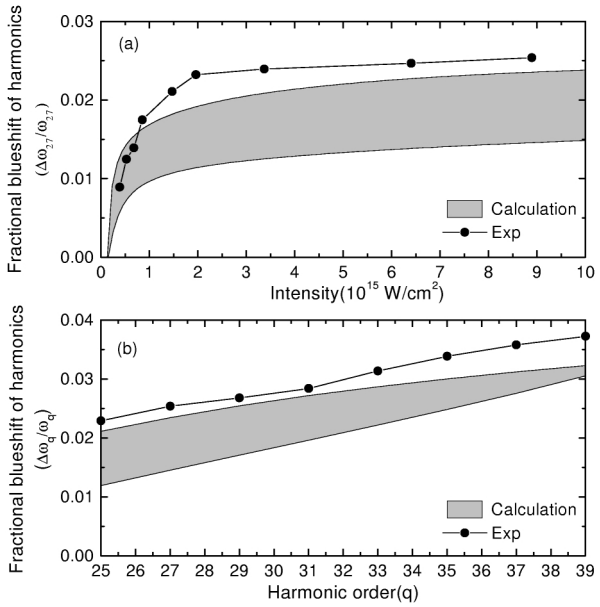

FIG. 4. (a) Fractional blueshift of the 27th harmonic with respect to laser intensity. Solid line with circles indicates an experimental result obtained at Ar density of 2×10^{17} cm^{-3}. The lower bound of the shaded area was calculated by assuming the harmonic generation at the maximum dipole acceleration; the upper bound was determined by assuming it at the cutoff intensity of the 27th harmonic. (b) Order dependence of harmonic blueshift. Experimental result of fractional blueshift obtained at the laser intensity of 9×10^{15} W/cm^2 and the Ar density of 2×10^{17} cm^{-3} is compared with the semiclassical calculation performed as in (a).

HHG over an extended period of time. If only a single-atom response is considered, the harmonic intensity would be strongest at the time of maximum dipole acceleration. At this moment about 30% of neutrals are ionized by the driving laser pulse. This may not yield the maximum observed harmonic intensity, since phase matching is less favorable over the harmonics emitted earlier in time; it would provide a lower bound of the fractional frequency shift. The upper bound of the fractional frequency shift can be estimated by considering the lowest possible laser intensity to generate a given harmonic, i.e., the cutoff intensity for a given harmonic order [7]. The shaded area in Fig. 4(a) shows the lower and upper bounds of the calculated fractional frequency shift. As the experimental result obtained with an Ar density of 2×10^{17} cm^{-3} contains a blueshift of about 10% due to the SPM effect, the observed blueshift matches the calculated range. For intensity above 1×10^{15} W/cm^2 in which the nonadiabatic effect is significant, the blueshift of harmonics has values close to the upper bound—harmonics emitted as early as possible, indicating the importance of the phase-matching effect.

Another feature of nonadiabatic blueshift as distinguished from the SPM effect can be found in the order dependence of harmonic blueshift. The fractional blueshift due to SPM has no order dependence; however, the nonadiabatic effect shows a linear increase of the fractional shift with the harmonic order. Figure 4(b) shows the order dependence of the experimental results obtained at an intensity of 9×10^{15} W/cm^2 and Ar density of 2×10^{17} cm^{-3}, along with the calculated result obtained in Fig. 4(a). The fractional blueshift increases linearly from 0.023 at the 25th order to 0.037 at the 39th order with values near the upper bound of the calculated fractional blueshift, further confirming the nonadiabatic blueshift.

In conclusion, the intensity, density, and order dependence of the harmonic blueshift generated in an intense, 30-fs laser field confirmed the nonadiabatic blueshift of harmonics; this is an important consequence of the intensity-dependent harmonic phase derived from HHG theory. The observed blueshift was sufficient to cover the interval between odd harmonics, which opens a way to utilize HHG as a tunable coherent femtosecond XUV source. The current result demonstrated that properties of high harmonics can be controlled by applying the intensity-dependent harmonic phase, which would play a crucial role in generating phase-locked harmonics to produce attosecond (10^{-18} sec) pulses.

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