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Temporal phase control of soft-x-ray harmonic emission

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We report a strong dependence of the soft-x-ray spectra generated by high harmonic emission on the chirp of the excitation pulse, when an ultrashort laser drives the process. For identical pulse durations, distinct harmonic peaks can be observed for positively chirped excitation pulses, while for negatively chirped pulses, the harmonic peaks become irregular. This behavior is explained by simulations that combine the chirp of the laser with the intrinsic phase shift of the harmonics. This work resolves an outstanding discrepancy between theory and experiment by demonstrating that high-order harmonic generation driven by short-duration pulses can result in distinct harmonic peaks. This work conclusively demonstrates the role of the intrinsic phase in determining harmonic emission spectra, and control this phase during the emission process.
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In recent years, there has been much interest in the process of high-order harmonic generation (HHG) [1–9] because of potential applications of coherent vacuum ultraviolet (VUV) and soft-x-ray sources in science and technology [10,11]. To date, the dependence of the harmonic output spectra on laser intensity, wavelength, polarization, and pulse width has been studied both experimentally and theoretically. For example, by comparing the experimentally measured cutoff wavelength (highest energy photon produced) as a function of atomic species, the basic conceptual validity of the ''rescattering'' picture [1,12] of HHG has been verified. However, more-detailed comparisons of these models with experiment has proven to be difficult, because of the lack of detailed experimental data and greatly varying experimental parameters. One prominent outstanding question in the literature is that of the role of single-atom effects vs collective (i.e., phase-matching) effects. In this work, we consider the case of HHG at low pressures, where one expects collective effects to be minimized. We observe that, for positively chirped pump pulses, the individual harmonic peaks are well defined and discrete, while for negatively chirped pump pulses, the harmonic output spectra merge into a continuum. By comparison with both analytic semiclassical theory and quantum models, we show that these spectral features are a result of the (single-atom) intrinsic phase of the generated harmonic emission, which can be reduced or enhanced by adjusting the phase (chirp) of the excitation laser pulse.

This work explains an outstanding discrepancy between theory and experiment for HHG of very short (~25-fs) pulses. Experimentally, well-defined harmonic spectral peaks are observed, but theoretical models have indicated that the harmonic emission should be an irregular continuum. This discrepancy had been attributed to collective or spatial-averaging effects. This work demonstrates that even a modest value of positive chirp (which broadens the pulse by less than 50%) can result in spectrally resolved emission peaks. An improved understanding of these effects is important for several reasons. First, a complete understanding of the single-atom HHG process, and particularly the role of parameters such as laser chirp (which is always varied and somewhat uncertain in experiments, but until this work not considered in theoretical models) will help us to understand the role of collective effects. Second, a more fundamental understanding may allow us to optimize the HHG process for specific applications through cycle-by-cycle coherent control of the radiating electron. Third, concepts such as attosecond-duration pulse generation depend on the validity of theoretical models of the HHG process; thus validation of these models will help to guide further experimental efforts [6,13,14].

The well-established ''rescattering'' model of HHG views x-ray emission as the result of scattering or recombination of an electron, ionized in a strong laser field, with its parent ion. In this picture [1,12], the shortest x-ray wavelength emitted is directly related to the strength of the electromagnetic field at the time of ionization. The use of very short pulses makes it possible to extend high-order harmonic generation to shorter wavelengths, since a rapidly rising excitation pulse allows the atoms to survive to a higher intensity before ionizing [5–7,9]. Previous work has demonstrated that a fast-changing pulse intensity can spectrally shift the HHG radiation. For example, for a given order, the phase of the x-ray emission (with respect to the laser field) changes with laser intensity [15]. In the midplateau region, many electron trajectories contribute to emission at the same harmonic order. Each contribution has a different frequency shift, depending on when the emission occurs. These phase effects are the origin of the interference effects predicted and observed in the midplateau region [7] where the harmonic spectra are not as well resolved and discrete as near cutoff. In contrast, near cutoff the harmonics are generated by only a few electron trajectories, corresponding to electrons ionized near the peak of the laser pulse. Classically, these trajectories correspond to the case of the electron that acquires close to the maximum quiver energy from the laser field and then rescatters from its parent ion. The limited number of contributing trajectories reduce interference effects, and the harmonics near cutoff exhibit well defined individual peaks. As stated above,

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a direct consequence of this is that the harmonics near cutoff are negatively chirped [8]. This induced negative chirp can be explained by the intensity-dependent phase [16,17] of the harmonic generation process near the peak of a short laser pulse, which causes the emission from subsequent cycles on the pulse leading edge to be earlier in phase from cycle to cycle, and which also results in a higher return energy of the rescattering electron. This effect leads to a phase shift. However, near the peak of a pulse, the decrease in the shift leads to an induced negative frequency *chirp*. Finally, we note that the effects of the intrinsic phase on the spatial properties of harmonic emission have also been investigated [18,19].

For our experimental work discussed here, we used an ultrashort-pulse Ti:sapphire amplifier system described previously [20]. The near-transform-limited pulse width used in this experiment was 40 fs. The output beam diameter of the laser was 1 cm, and a 1-m focal length curved mirror was used to focus the beam onto a gas jet, producing a ~100-μm-diam focal spot. The gas nozzle diameter was 1 mm, while the gas pressure was approximately 8 torr. Typically, 10 mJ of laser energy was used to generate the harmonics, which corresponds to an intensity of 1.8×10^{15} W/cm^2 at the focus. The x rays were dispersed using a flat-field soft-x-ray spectrometer, and then detected using an image intensifier with a pair of microchannel plate detectors (MCPs). The spectrally dispersed image of the high harmonic emission was recorded using a cooled charge-coupled-device camera connected to a computer. A 0.4-μm-thick carbon filter was placed in front of the MCP to block the very bright scattered light from low-order harmonics.

The chirp of the excitation laser pulse could easily be varied by adjusting the separation of the gratings in the pulse stretcher. The resultant harmonic x-ray spectra were observed to change dramatically as a function of excitation laser chirp, as shown in Fig. 1. The harmonic peaks shift to longer wavelengths for positive chirp, when the leading edge of the pulse is redder than the trailing edge. This effect is qualitatively similar to our previous results with argon [5], and can be explained by the wavelength change on the leading edge of the laser pulse as the pulse is chirped. What was not explained at that time was the asymmetry in spectral broadening that occurred as the laser pulse was positively or negatively chirped. In the case of He, this asymmetry is even more dramatic, as shown in Fig. 1. The harmonic peaks are observed to broaden dramatically as the pump pulse is negatively chirped, merging into a continuum in the case of He. However, for positive laser chirp, the peaks become narrower and well defined.

Experimentally, we characterized the laser chirp using the technique of second-harmonic frequency-resolved optical gating [21]. The experimentally measured pulses for positive and negative chirps were quite symmetric, ruling out the possibility that the observed spectral asymmetries arose from some unexpected phase variation in the pump pulse shape for positive and negative chirps. We can therefore conclude that the observed spectral changes are indeed influenced primarily by the sign of the input laser chirp. Finally, numerical simulations show that, at the pressures and intensities used in these experiments, the effect of ionization-induced blue shift on the harmonic spectra is very small and therefore can be neglected.

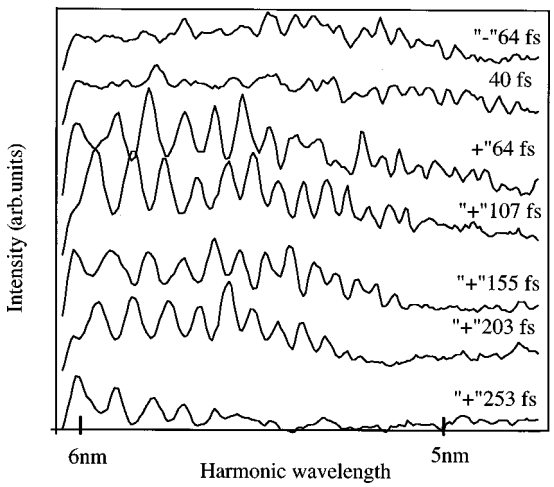

FIG. 1. Experimentally measured harmonic spectra for positively (+ sign) and negatively (− sign) chirped laser excitation pulses.

In order to understand the experimentally observed spectra, we simulated harmonic generation using the Lewenstein model [22,23]. This model is valid for harmonic orders higher than the ionization potential, and also requires that the photon energy of the laser be much smaller than the ionization potential, which is true for our experiments ($\lambda=800$ nm). We use the saddle-point integration over momenta to simplify the calculation, and use hydrogenlike dipole matrix elements to simulate the real helium atom. The depletion of the ground state was calculated using Ammosov-Delone-Krainov ionization rates [24]. The results of our simulations for He are shown in Fig. 2, which clearly show that for positively chirped excitation pulses, the HHG peaks are well defined, while for negatively chirped excitation pulses, the peaks close to cutoff are spectrally smeared, in very good agreement with our experimental data [7,25]. Similar results for Ar have been obtained recently by Salières *et al.* [16,17]. The numeric simulations alone, however, do not explain why the harmonic spectra are influenced by the chirp of the laser.

Our results can be explained intuitively by assuming that both the laser pulse and the harmonic pulse are Gaussian. The laser field at given time t is then expressed as

$$E_1(t)=E_1\exp(-\Gamma_1 t^2)\exp(i\omega_1 t), \tag{1}$$

where E_1 is the peak field, and ω_1 is the carrier frequency. The chirp of the pulse b_1 is included in the complex Gaussian parameter $\Gamma_1=a_1-ib_1$, where $a_1=2\ln(2)/\Delta t_1^2$ and Δt_1 is the full width at half maximum (FWHM) of the pulse. In our experiments, $b_1=0$ corresponds to a transform-limited pulse, which has the minimum pulse duration (40 fs). When the grating separation in the stretcher is increased, it introduces a net positive chirp ($b_1>0$) on the pulse, and increases the pulse duration. Decreasing the grating separation also broadens the pulse, but introduces a net negative chirp ($b_1<0$).

The electric field of the qth harmonic is given by

$$E_q(t)=E_q\exp(-a_q t^2)\exp[iq\omega_1 t+\Phi_{\text{total}}(t)], \tag{2}$$

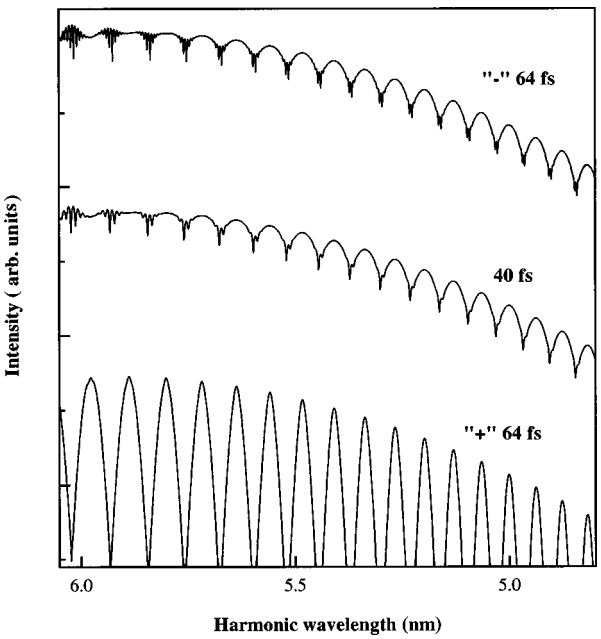

FIG. 2. Numerical simulation of high harmonic emission spectra near cutoff for different values of chirp of the excitation pulses: positively chirped pulses (+ sign) and negatively chirped pulses (− sign).

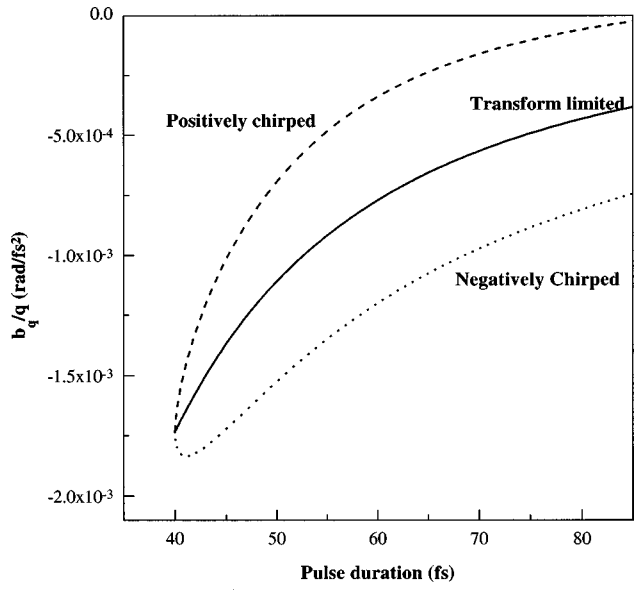

FIG. 3. Normalized chirp parameter (b_q/q) of the emitted harmonic pulse as a function of laser pulse duration. The sign of the chirp of the laser pulse is indicated from each curve.

where E_q is the peak field, and $a_q = 2\ln(2)/\Delta t_q^2$, while Δt_q is the FWHM of intensity, $\Phi_{\text{total}}(t)$ is the total phase, which can be written as the sum of two parts [26]:

$$\Phi_{\text{total}}(t) = \Phi_{\text{dipole}}(t) + \Phi_{\text{laser}}(t), \tag{3}$$

where $\Phi_{\text{laser}}(t) = q b_1 t^2$ is that phase induced by the chirp of the laser pulse, and $\Phi_{\text{dipole}}(t)$ is the phase of the induced dipole moment, which is laser intensity dependent.

It has been shown that under the quasiclassical approximation, the phase of the induced dipole is determined by the value of the action acquired along the most relevant saddle-point trajectory, i.e., $\Phi_{\text{dipole}}(t) = S$ [15]. We are only interested in harmonic orders close to cutoff, which are generated, from the classical point of view, by an electron that is released at a phase of 0.3 rad, and that returns at a phase of 4.37 rad [for a constant cos ($\omega_1 t$) field] [1]. The classical action of such an electron path is

$$S = \frac{2\pi}{h}\int_{0.3}^{4.27} \frac{p^2(t'')}{2m} dt'', \tag{4}$$

where h is Planck's constant, m is the mass of the electron, and p is the momentum of the electron. This integration gives $S = 2.92 U_p/\hbar\omega_1$, which is close to the results of previous quantum models ($S = 3.2 U_p/\hbar\omega$) [15]. Note that the contribution of I_p to S is a dc phase shift, which is not relevant to our discussion here, and is therefore omitted. Since U_p is proportional to the laser intensity, it is clear that the phase of dipole is intensity dependent. Therefore, for short pulse excitation, the dipole phase is also time dependent.

At the peak of the laser pulse where the harmonics near cutoff are generated, the pulse shape can be approximated by a parabola, so that $\Phi_{\text{dipole}}(t) = (3.2 U_{\text{po}}/\hbar\omega_1^*)(1 - 2a_1 t^2)$ and U_{po} is the ponderomotive potential at the peak of the pulse. It is interesting to note that since the cutoff harmonic order q is given by $(I_p + 3.2U_{\text{po}})/\hbar\omega_1$, for very high orders we can approximate $3.2U_{\text{po}}/\hbar\omega$ by q. After omitting the dc phase shift, we then obtain

$$\Phi_{\text{total}}(t) = q(-2a_1 + b_1)t^2. \tag{5}$$

By combining Eq. (2) with Eq. (5), we can thus describe the harmonic field by

$$E_q(t) = E_q \exp(-\Gamma_q t^2)\exp(iq\omega_1 t), \tag{6}$$

where $\Gamma_q = a_q - i b_q$ is the complex Gaussian parameter, and $b_q = q(-2a_1 + b_1)$ is the chirp parameter of the qth harmonic.

The effect of the laser chirp on the chirp of the harmonic can be seen in Fig. 3, which plots the predictions of Eq. (5). When the pump pulse is transform limited ($b_1 = 0$), the harmonics in the cutoff region are negatively chirped ($b_q < 0$), since $a_1 > 0$. When the laser pulse is negatively chirped ($b_1 < 0$), then the negative chirp of the harmonics is enhanced. Finally, if the laser pulse is positively chirped ($b_1 > 0$), it will compensate for the chirp induced on the harmonic field to some extent, depending on the relative values of b_1 and a_1. The observation of an optimum laser chirp to spectrally shape harmonic emission is similar to recent observations of quantum control in molecular systems using tailored laser pulses [27].

The FWHM spectral width of the qth-order harmonic $\Delta\omega_q$ is given by [28].

$$\Delta\omega_q = \sqrt{8(\ln 2) a_q \left[1 + \left(\frac{b_q}{a_q}\right)^2\right]}. \tag{7}$$

The induced chirp b_q on the harmonic pulse will broaden the bandwidth by a factor of $\sqrt{1 + (b_q/a_q)^2}$ compared to the transform-limited harmonic case for an identical pulse width.

Our simulations show that the HHG pulse duration is the same for positively or negatively chirped pump pulses differing only in the sign of the chirp. However, the harmonic chirp $|b_q|$ induced by a negatively chirped laser pulse is much larger than for a positively chirped pulse, as shown in Fig. 3 and predicted by Eq. (7). These simple simulations explain intuitively why spectral asymmetries are observed experimentally and predicted numerically in the harmonic output for positively and negatively chirped excitation pulses.

The chirp parameter b_1 initially changes very rapidly as the pulse duration increases from the transform limit. Further increase in pulse duration results in smaller changes in b_1. As a result, the chirp of the qth-harmonic b_q also follows the same trend, as shown in Fig. 3. Thus, most of the effect of laser chirp on the harmonic spectral structure should be observed for small values of detuning from the transform-limited case. This is precisely what is observed in both our experimental and numerical simulations, as shown in Figs. 1 and 2. A small amount of positive chirp can cancel the intrinsic phase of the harmonics, resulting in distinct harmonic peaks. This explains what had been thought to be an inconsistency between theory and experiment, where experimental data showed better-defined harmonic peaks than theory predicted [8]. In contrast, negatively chirped excitation pulses will induce a larger chirp and bandwidth on the harmonics than in the case of transform-limited excitation, and may allow subsequent recompression of the harmonics using a grating pair. [8].

In conclusion, we have demonstrated experimentally and theoretically that high harmonic generation is very sensitive to the chirp of the excitation pulse. Our results conclusively demonstrate the importance of the intrinsic phase in determining the harmonic emission spectra. Moreover, the effects of the intrinsic phase were observed and controlled by the instantaneous phase of the excitation laser pulses. The excellent agreement between our experimental results and theoretical calculations based on simple models of HHG validates our understanding of this process, and therefore also lends credence to recent theoretical predictions, including the possibility of generating attosecond-duration light pulses [6,8,13].

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