Enhanced High-Harmonic Generation Using 25 fs Laser Pulses

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We present experimental and theoretical results on high-harmonic generation in noble gases using an 805 nm, 25 fs, titanium-doped sapphire laser. The harmonic energies observed are unexpectedly high when compared with experimental and theoretical results to date for longer excitation pulses. We observe that the efficiency of harmonic production is highest for shorter pulses. Furthermore, the wavelength of the harmonics can be tuned by adjusting the sign of the chirp of the excitation pulse, demonstrating a tunable, ultrashort-pulse, <25 fs soft-x-ray source.

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The occurrence of a plateau in the conversion efficiency of high-order harmonic generation of intense laser pulses incident on gases has been well documented both experimentally [1–5] and theoretically [6–12]. A variety of wavelengths and pulse durations have been used to generate harmonics, and harmonic orders well exceeding one hundred have been observed by L'Huillier and Balcou from the two lighter noble gases using 1 ps, 1054 nm laser pulses [3]. Macklin, Kmetec, and Gordon observed harmonics up to the 109th order in neon using 125 fs, 806 nm pulses, which represents the shortest wavelength harmonic reported to date [4]. In heavier noble gases, which have smaller ionization potentials, the number of harmonics which can be generated is less, although they have higher conversion efficiencies. L'Huillier and Balcou observed up to the 55th and 27th harmonics from argon and xenon, respectively, with the 1 ps, 1054 nm laser pulses [3]. These observations are in agreement with theoretical predictions [10,11] that the photon energy of the highest harmonic emitted from a gas cannot exceed $I_p + 3.2U_p$, where I_p is the atomic ionization potential and $U_p \sim \lambda^2 I$ is the maximum ponderomotive potential that an electron may experience prior to detachment from the atom, and λ and I are the laser wavelength and intensity, respectively.

Krause, Schafer, and Kulander [10] used *ab initio* calculations of the Schrödinger equation in three dimensions to show that the breadth of the plateau in the harmonic spectrum obeys this cutoff rule, where U_p is chosen at the point where the atom ionizes, even if the peak laser intensity goes higher. They also showed that this rule can be understood with a classical picture, where the electron detaches from the atom and releases energy when it is recaptured by the atom after a laser cycle [11]. The classical picture predicts that the maximum kinetic energy acquired by an electron from the field upon return to the nucleus is $3.2U_p$. Quantum-mechanical descriptions of high harmonic generation give similar results [8,12]. All of these models agree with harmonic generation measurements to date, which have been made with laser pulses of duration greater than 100 fs. They also use the adiabatic assumption, where the laser intensity varies slowly with respect to an optical period.

For our work, we investigated high harmonics generated by a 25 fs, 10 Hz, 3 TW Ti:sapphire laser [13] in various noble gases [14]. The bandwidth of the pulses is 32 nm, centered at a wavelength of 805 nm, and the laser system can provide up to 70 mJ of energy per pulse, with shot-to-shot fluctuations of $\approx 10\%$. The laser beam is linearly polarized and focused by a curved mirror of focal length 1 m to 1.2 times the diffraction limit as measured by a charge coupled device camera, with $f/80$ focusing. A gas target ≈ 1 mm thick was placed 2.5 cm after the focus, where the laser diameter was approximately 280 ± 50 μm. The ultrashort nature of our excitation pulses (10 optical cycles FWHM) implies that at the half maximum position of the temporal pulse envelope, the laser intensity changes by more than 25% during a single cycle. This defies the adiabatic assumption, which suggests that the atomic dipole moment undergoes quasiperiodic motion from cycle to cycle, with no dependence on the history of the pulse.

The gas target [15] consists of two thin metal plates with a 500 μm hole drilled through them. The laser goes through the hole and interacts with the gas which enters the hole from between the plates. The device was backed continuously with pressures of 5–50 Torr, and the pressure inside the hole was about one-fifth of the backing pressure. Because the laser confocal parameter is much larger than the gas target thickness, the harmonics can be thought of as emerging from a plane so that geometric phase matching should not critically depend on the z position of the target. We placed that target after the focus mainly to increase the size of the laser spot and hence increase the signal. The harmonic signals are resolved with HIREF-SXR-1.75 monochromator (Hettrick Scientific) and measured with a

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microchannel plate detector. The spectrometer, which had a resolution of ≈1 Å, was scanned at a rate of ~0.02 Å per laser shot, and the harmonic signal from each laser shot was recorded. Typically, ≈12 000 points make up a given spectrum.

For the heavier noble gases, we observed harmonics with photon energies remarkably higher than previously seen. Figure 1(a) shows harmonics generated in argon, where orders up to the 61st are visible. This corresponds to a photon energy of 93 eV. The lower harmonic orders are artificially damped by the spectrometer grating efficiency. Figure 1(b) shows harmonics up to the 41st generated krypton and harmonics up to the 29th generated in xenon. These are approximately 40% higher harmonic photon energies than have been seen previously using these gases [3]. The increase in harmonic photon energy is especially interesting from the point of view that for a given intensity the ponderomotive potential U_p of our laser is 42% less than that of the 1054 nm laser used in the experiments by L'Huillier and Balcou [3]. Thus, according to the $I_p + 3.2U_p$ rule, one would expect to see harmonics of lesser photon energy with our laser, not greater. This suggests that for ultrashort-pulse excitation, atoms can survive to higher laser intensities before ionizing. In neon, we were unable to resolve harmonics past the 105th, but we saw light which may correspond to harmonic orders up to the 131st. As we reduced the laser intensity, the short wavelength edge gradually retreated to longer wavelengths, indicating that the shortest wavelength light is real. Figure 1(c) shows harmonic spectra of Ne. The lower harmonic orders seen are artificially damped by the spectrometer grating efficiency, since we had to use a different grating to observe these shorter wavelength harmonics.

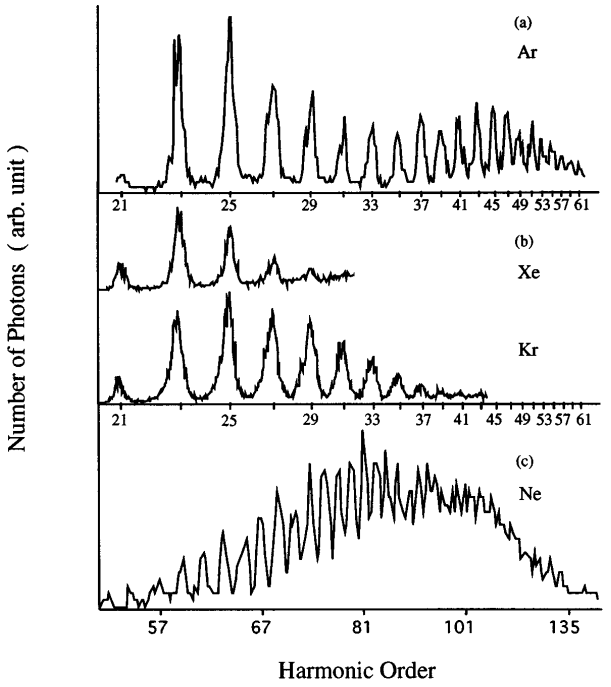

FIG. 1. Harmonic spectra generated in (a) ~5 Torr of Ar, and (b) 2 Torr of Kr and Xe, with peak laser intensity 5×10^{14} W/cm^2. (c) Harmonic spectra generated in ~10 Torr of Ne with peak laser intensity 2×10^{15} W/cm^2. The relative intensities of (a)–(c) should not be compared directly.

The spectra seen in Figs. 1(a)–1(c) were produced with a laser energy of 3.5 mJ, which for the focusing conditions described above corresponds to a peak intensity of approximately $(5 \pm 2) \times 10^{14}$ W cm^{-2}. This is significantly above (×2) the point where ionization should readily occur. The possibility therefore exists that the highest harmonics might arise from ions, in which case they would not be considered to be of unexpectedly high order. To check this, we observed harmonic generation in argon as a function of gas pressure. We observed that all of the harmonic peaks decreased in strength together as the pressure was gradually reduced from 5 to 1 Torr. If the higher-order harmonic peaks were produced by ions while the lower by neutral atoms, one would expect the harmonics to scale very differently with pressure because of a changing coherence length arising from free electrons. Thus our observations suggest that the harmonic peaks all arise from neutral atoms. Our spectra are, however, integrated over space and time from an ensemble of atoms experiencing different laser intensities.

One model for estimating the intensity at which ionization occurs is barrier suppression ionization (BSI) [16]. In this picture, the outer electron suddenly escapes from the atom when the laser field suppresses the potential barrier below the field-free binding energy of the electron. This model has worked well for predicting the intensities at which atoms ionize for 1 ps, 1054 nm, laser pulses. According to this model, the intensities at which Ne, Ar, Kr, and Xe ionize are, respectively, 8.7×10^{14}, 2.5×10^{14}, 1.5×10^{14}, and 8.6×10^{13} W/cm^2. The harmonic photon energies that we observed from Ar, Kr, and Xe well exceed the theoretical limit when the BSI assumption is used to determine U_p. In fact, under the BSI assumption, the harmonic photon energies would exceed $I_p + 5U_p$ for each gas. Thus it would seem that for ultrashort pulses the BSI model is not applicable.

We investigated numerically the dependence of ionization on pulse duration for pulses of fixed peak intensity. This was done by integrating the time-dependent Schrödinger equation in one dimension. While the solution in one dimension cannot be expected to provide an exact description of ionization, it can be interesting to compare the ionization probabilities for different applied pulse durations. The field-free potential was chosen to have the form $-1/(|x| + a)$ in atomic units [17], where $a = 3.7 \times 10^{-2}$ a.u. The calculation showed that for our experimental conditions, atoms can experience 20% higher laser intensity before the ionization probability exceeds 10% for a 25 fs excitation pulse, compared with a 100 fs pulse. The details depend on the exact shape assumed for the leading edge of the laser pulse, which has not been experimentally determined as yet. However, in

contrast with the BSI model, the implication is that, even in the tunneling regime, neutral atoms are able to experience higher ponderomotive potentials under ultrashort-pulse illumination than is possible using longer excitation pulses, since atoms can survive higher intensities before ionizing.

The $I_p + 3.2U_p$ rule for the highest possible harmonic photon energy comes from a classical consideration of the maximum energy that an electron can have when a laser of constant intensity pushes it back into its parent ion after having broken away during the previous cycle. Since this process requires the time scale of a laser cycle, the fact that our laser intensity changes during this time can influence the process. During the rising edge of the pulse, the relevant intensity for calculating the ponderomotive potential is higher than that at which the electron breaks away from the atom, since the field strength grows before the electron can return. This consideration alone cannot explain the anomalously high harmonic orders observed, but it does predict some enhancement.

To investigate how important the shortness of our laser pulse duration is to the total number of harmonic orders observed, we increased our laser pulse duration by introducing chirp. We did this by changing the position of the grating in our pulse stretcher prior to amplification. The beam alignment and pulse spectrum after amplification and compression remained unchanged. Calculations of the pulse propagation through our laser system suggest that the temporal profile of the pulse remains smooth as the chirp is varied.

We examined the effect of introducing various amounts of chirp, both positive and negative, on harmonic production in argon. Figure 2 shows harmonic spectra for different pulse durations at a pressure of 5 Torr. The harmonic spectrum generated by a transform-limited 25 fs pulse is shown in the middle. Above it are spectra from positively chirped pulses, and below it are spectra from negatively chirped pulses of similar durations. In all cases the peak intensity of the laser pulse was held fixed so that the longer pulses required more energy.

For almost all of the pulse durations shown in Fig. 2, we observe unexpectedly higher order harmonics than predicted by the BSI theory, which predicts that the 41st order should be the maximum observed. The photon energies in all cases are higher than those previously observed in neutral argon, although the highest observable harmonic order decreases gradually with increasing pulse duration. Another striking feature about the data in Fig. 2 is that the area associated with given harmonic peaks remains roughly the same for the different spectra. Since the laser energy is increased to keep the intensity fixed for the longer durations, the harmonic production process is most efficient for the shortest laser pulses. In other words, shorter laser pulses produce the most harmonic signal for a given laser energy, with the harmonic efficiency scaling inversely with pulse duration. This agrees with the findings of Kondo *et al.* [5] who reported on harmonics generated with a 120 fs Ti:sapphire laser. They stretched the laser pulse duration to 750 fs by introducing chirp and increasing the laser energy to keep the peak intensity fixed. They observed no significant change in the harmonic yield over this range. Together the two sets of experiments suggest that high-harmonic conversion efficiency is roughly inversely proportional to pulse duration for pulse widths shorter than 1 ps.

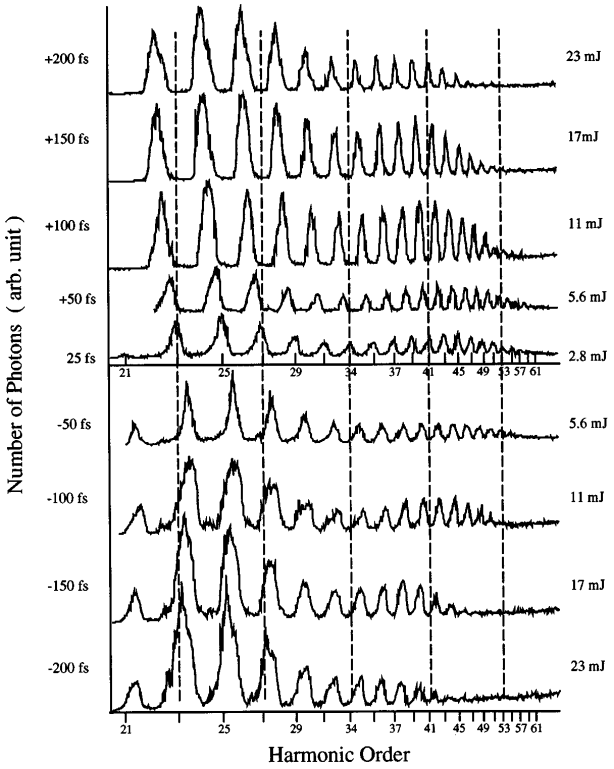

FIG. 2. Harmonic spectra generated in 5 Torr of argon for fixed peak laser intensity of 5×10^{14} W/cm^2 and for various laser pulse chirps.

In Fig. 2, a significant amount of redshifting and blueshifting of the output spectra can be seen when positive and negative chirps are introduced, respectively. This indicates that the harmonic production tends to favor the rising edge of the laser pulse and that it is unlikely that harmonic emission comes from ions. The harmonic peaks can be shifted spectrally to the degree that a continuously tunable light source is obtained for wavelengths shorter than 25 nm. The degree of blueshifting and redshifting is not the same for pulses of similar duration but with opposite chirp. For positive chirp, the redshifting of the peaks is very pronounced while the peaks remain relatively narrow. However, for the negative chirps, the harmonic peaks initially shift slightly to the blue and then broaden to approximately twice the former width. The bandwidths observed for individual peaks are sufficiently broad to support subfemtosecond soft-x-ray pulses, although it is not known as yet if the proper phase conditions are met. One possible explanation for the asymmetry observed in the shifting and broadening of the harmonic spectra as a function of laser chirp is that the negative chirp induces a

stabilizing effect against ionization. Thus, in the case of negative chirps, the atom could experience a wider range of frequencies.

Our model calculations show similar spectral shifting and broadening of the harmonics with different signs of the excitation laser chirp. The calculations show that at the point on the leading edge of the pulse profile where a negatively chirped 100 fs pulse (with the same bandwidth as a 25 fs pulse) causes 50% ionization of the atom, there is 68% ionization for a positively chirped 100 fs pulse with the same intensity. Thus, when irradiated by a negatively chirped excitation pulse, the atom survives longer and is exposed to a greater fraction of the input bandwidth than for the positively chirped case. This may explain why the harmonic peaks are broader, and shift less, for a negatively chirped excitation pulse, since harmonic emission presumably stops after ionization. Figure 3 shows calculated single-atom harmonic spectra which are less spectrally shifted for negatively chirped excitation pulses than for positively chirped pulses, in agreement with our experimental data. These results suggest that laser pulse chirp might be used to control the ionization rate of atoms, to allow the production of extremely broad bandwidth and attosecond duration x-ray pulses, with well-defined and possibly compressible chirp.

In summary, we have investigated harmonic generation using ultrashort laser pulses. In many of the noble gases, we observe unexpectedly high harmonic orders, up to 40% higher energies than have previously been predicted or observed. We also observe increased conversion efficiency of laser to soft-x-ray light by using shorter excitation pulses. Finally, we observe that the harmonic spectra are asymmetric with respect to the sign of the chirp and that the harmonics may be tuned by adjusting the chirp. Our results suggest that the BSI model is not applicable to very short excitation pulses and that the laser pulse chirp might be used to control the ionization rate of atoms. Our results have demonstrated a tunable, sub-20 fs, soft-x-ray source.

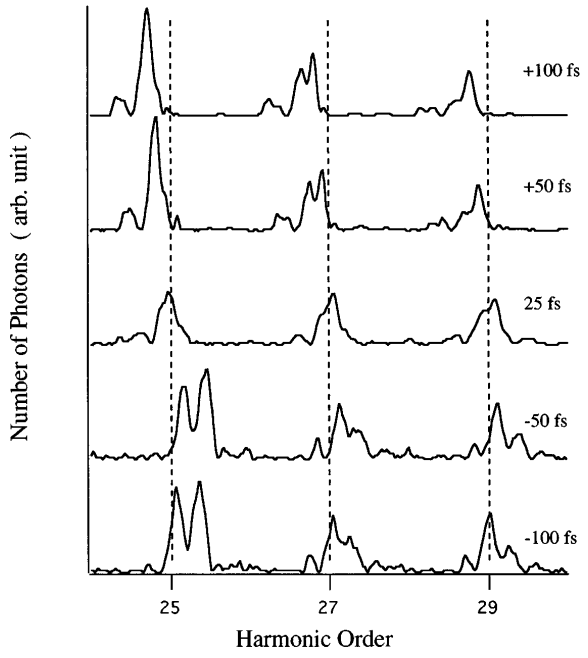

FIG. 3. Theoretical predictions for the harmonic spectra generated for various excitation laser chirp for fixed intensity and bandwidth.

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