High Harmonic Generation from Ultrafast Pump Lasers

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Using an ultrafast pump pulse affects the spectral and temporal characteristics of high order harmonics in an unexpected and fortuitous way. Calculations of spectra for rare gases using 10–100 fs, 800 nm pulses show that as the pump pulse length decreases the highest harmonics become correspondingly shorter while their conversion efficiencies increase dramatically. Especially significant is that we find these highest harmonics have phase characteristics that allow for the possibility of compression to subfemtosecond time scales. [S0031-9007(96)02239-9]

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Recent advances in terawatt Ti:sapphire laser technology have made it possible to produce 800 nm high intensity pulses less then ten optical cycles ($\sim$20 fs) in duration [1,2]. A major topic of current research focuses on the effects of using increasingly shorter, high intensity pulses on excitation and ionization processes. Recent experiments [3] on harmonic generation using 25 fs pulses have exhibited two such effects: an increase in the saturation intensity (the highest intensity experienced by a reasonable fraction of atoms) leading to higher energy photoemission as compared to longer pulses and a substantial broadening of the spectral widths of the harmonics. These results and the prospect of experiments using even shorter pulses raise many fundamental questions. Can the spectral width of these harmonics be used to produce subfemtosecond vacuum ultraviolet (VUV) pulses? Is there an optimal pump pulse duration for producing ultrafast high harmonics? How is the conversion efficiency effected by the pulse length? What is the upper limit on harmonic photon energy from neutral atoms?

In this Letter we explore the limits of high harmonic emission from this new generation of ultrashort pulse lasers via a theoretical study of the interaction of rare gas atoms with intense laser pulses which have half-widths of 5 to 40 optical cycles. We find that the single atom harmonics have broad spectral widths similar to the experimental results. The highest harmonics, those at the end of the plateau and in the cutoff region, exhibit two notable properties: their conversion efficiencies increase rapidly with decreasing pulse duration and they have simple quadratic phase structures (corresponding to a linear frequency chirp in time), which, if removed, will yield compressed pulses lasting less than 2 fs. We also find that several adjacent harmonics at the end of the plateau have almost the same linear chirp and roughly the same phase relative to the driving field, meaning that they could, in principle, be combined to yield a pulse lasting perhaps 400 attoseconds. Though the shortest pump pulses studied yielded the highest energy photoemission, pulses lasting less than ten cycles were found to impart significant phase distortions to even the harmonics at the end of the plateau. This implies there will be an optimum pulse length for producing compressible harmonic pulses.

Harmonic generation in a gas is a coherent process involving both the emission from each individual atom and the propagation of the emitted fields as well: good phase matching is essential for high conversion efficiencies [4]. The macroscopic harmonic fields within the excited medium are profoundly influenced by the phase characteristics of the induced atomic dipoles, which, in turn, depend on the focal properties of the driving laser field. In this Letter we ignore phase matching effects to concentrate on the single atom emission for two reasons: (1) most of the extremal properties of the full (single atom + phase matching) problem are constrained by the single atom behavior (in particular, the minimum pulse length and maximum photon energy of the harmonics are so constrained), and (2) the advent of multiterawatt lasers means that harmonic generation experiments with unfocused lasers are now possible, leading to simplified phase matching, at least for intensities below saturation.

We calculate the harmonic generation spectrum using the single active electron approximation (SAE) [5]. To do this, we numerically integrate the time-dependent Schrödinger equation for an atom in a linearly polarized laser pulse with a Gaussian envelope assuming that only the outermost valence electron responds to the field. The emitted radiation spectrum is proportional to the Fourier transform of the acceleration of the active electron

$$\mathcal{A}(\omega) = \int dt\, e^{i\omega t} a(t) \equiv A(\omega) e^{i\phi(\omega)}, \qquad (1)$$

where $A(\omega)$ is the (real) spectral envelope function. The electron's acceleration $a(t)$ is given by

$$a(t) = \frac{d^2\langle z\rangle}{dt^2} = -\langle\psi|[H,[H,z]]|\psi\rangle, \qquad (2)$$

where H is the full (atomic + laser interaction) Hamiltonian. For the SAE pseudopotentials the commutator must be evaluated numerically. This form has been found to

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be computationally more stable than the usual dipole expression $\mathcal{D}(\omega)$, obtained by Fourier transforming $\langle z(t)\rangle$, when significant ionization is present [6]. An approximate $\mathcal{D}(\omega)$, calculated as outlined in Ref. [6] by taking into account only transitions back to the ground state, agrees with $\mathcal{A}(\omega)/\omega^2$ in both amplitude and phase for all of the harmonics in the plateau and cutoff. This result emphasizes the importance of excited electrons that return to the ion core in the harmonic generation process.

Figure 1 shows the high end of a representative short pulse high harmonic spectrum [both the square of the dipole spectrum and the phase $\phi(\omega)$] for an argon atom interacting with a 27 fs, 810 nm pulse with a peak intensity of 3×10^{14} W/cm^2. At this intensity approximately half the atoms will be ionized by the peak of the pulse. We find that only the last few harmonics are distinct and that they are very broad compared to even what we would expect from this short pulse, i.e., $k^2 \Delta\lambda_k \gg \Delta\lambda_1$, where k is the harmonic order and λ_1 is the fundamental wavelength. Surprising also is that the phases of these highest harmonics behave in a simple and uniform fashion. The spectrum below the vicinity of the cutoff is highly structured with the expected odd harmonics being hard to distinguish and their frequency dependent phases varying rapidly. We find this spectrum to be typical of the short pulse regime.

At this intensity and wavelength, the ionization and photoemission dynamics are dominated by tunneling: every half optical cycle, a wave packet of electron probability escapes into the continuum via a tunneling process. As the wave packet drifts away a significant fraction of the density recollides with the ion core due to the large amplitude, oscillatory motion imposed by the driving field. The observed broadening of the plateau harmonics is most easily understood in terms of the rapid change in the laser intensity during the pulse and its effect on this tunneling wave packet. An electron that enters the continuum while the laser intensity is increasing experiences an additional acceleration before returning, earlier than it would have in a CW field, to rescatter off of the parent ion. This produces a blueshift. If the laser amplitude increases linearly with time, the shift is approximately constant [7]. By the same token, electrons ionized after the peak of the pulse are decelerated, returning later, and therefore the spectrum is redshifted. Harmonics at the end of the plateau (~49th harmonic in this case) are created only during the cycle or two near the peak of the pulse and have a simple linear frequency chirp from blue to red, as exemplified by the quadratic phase dependence of $\phi(\omega)$. Harmonics in the plateau are created over many optical cycles. As expected from these arguments, these harmonics experience much larger phase distortions and a broader range of frequency shifts compared to the harmonics at the end of the plateau. Since the amplitude for emitting a harmonic photon is the coherent sum over all of the different pathways leading to a given photon energy [Eq. (1)], harmonics that are made over a substantial portion of the pulse have a greater opportunity to exhibit rapid phase variations due to numerous interfering pathways than those harmonics that are made only near the peak of the pulse. Because of the extensive interference, the integrated emission strength within the limits of a given harmonic peak rises only slowly with increasing peak intensity after the harmonic in question has joined the plateau.

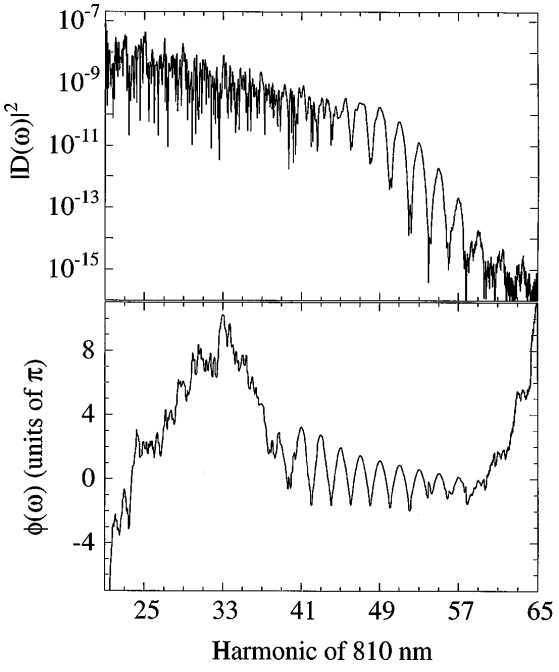

FIG. 1. Argon at 810 nm, 3×10^{14} W/cm^2 peak intensity and 27 fs FWHM pulse width. Upper panel: $|\mathcal{D}(\omega)|^2$; lower panel: $\phi(\omega)$.

We can examine the electric field envelope of a typical harmonic near the end of the plateau and its dependence on the pump pulse duration. The emitted electric field of the kth harmonic is obtained by back transforming the acceleration spectrum after applying a filter function around the frequency ω_k:

$$\mathcal{E}_k(t) = e^{-i\omega_k t} \int d\omega e^{-i(\omega-\omega_k)t} [\mathcal{A}(\omega) F(\omega - \omega_k)], \tag{3}$$

where $F(\omega - \omega_k)$ is a square filter that roughly duplicates the action of a grating in picking out a specific harmonic. For the distinct harmonics, the shape of the filter function is not crucial.

As an example, we focus on the 49th harmonic in argon at 810 nm (~75 eV) and a peak intensity of 3×10^{14} W/cm^2. Figure 2 illustrates the electric field envelope for three different pulse durations: $\tau_p = 108$, 27, and 13.5 fs corresponding to 40, 10, and 5 optical cycles. As the pump duration is decreased the harmonic pulse length decreases rapidly (from 21 to 7 to ~2 fs). The total area, however, actually increases even though

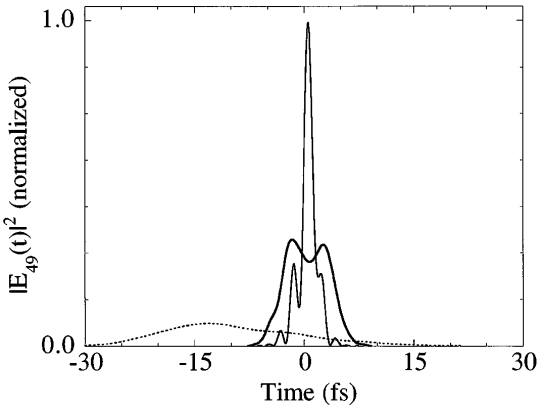

FIG. 2. Electric field envelope of the 49th harmonic at 810 nm and 3×10^{14} W/cm^2 peak intensity. The pulse duration is 108 fs (dashed), 27 fs (heavy line), or 13.5 fs (thin line).

the pump pulse energy is proportional to τ_p. Thus the relative conversion efficiencies for 108, 27, and 13.5 fs pulses are 1:6.5:11 at this single atom level.

We note that the saturation intensity for a 108 fs pulse is less than 3×10^{14} W/cm^2, which means that the 49th harmonic is made on the rising portion of the pulse only, as can be seen from its electric field envelope, and the energy is blueshifted in accordance with our earlier discussion. A 27 fs pulse is just below saturation at this intensity and a 13.5 fs pulse is unsaturated. Along with this rise in saturation intensity comes a concomitant rise in the highest energy harmonic in accord with the cutoff rule $E_{\max} = E_0 + 3U_p$, where E_0 is the ionization energy and $U_p = I/4\omega^2$ is the intensity dependent ponderomotive energy [6]. This raises the question of how high ultimately the harmonic cutoff can go for emission from a neutral atom. Calculations using a 13.5 fs pulse for helium, which has the highest ionization potential and therefore should have the highest saturation intensity, indicate that more than half of the atoms can survive to an intensity of about 2×10^{15} W/cm^2, where the cutoff harmonic is more than 380 eV. We believe that this is a good estimate for the upper limit in photoemission energy from a neutral atom.

Next we turn to the question of whether the broad spectral width of the harmonics can be used to produce compressed harmonic pulses. In Fig. 3(a) we show the envelope and phase of the 49th harmonic from Fig. 1. The phase (dotted line) is almost purely quadratic in ω. As we have discussed elsewhere [8], this quadratic component can be entirely removed with a grating pair arranged to provide positive group velocity dispersion, while at the same time separating the harmonic from surrounding wavelengths. The phase minus its quadratic component is shown as a solid line above the envelope. Fig. 3(b) shows the time history of the harmonic electric field envelope before and after compression. The resulting compressed

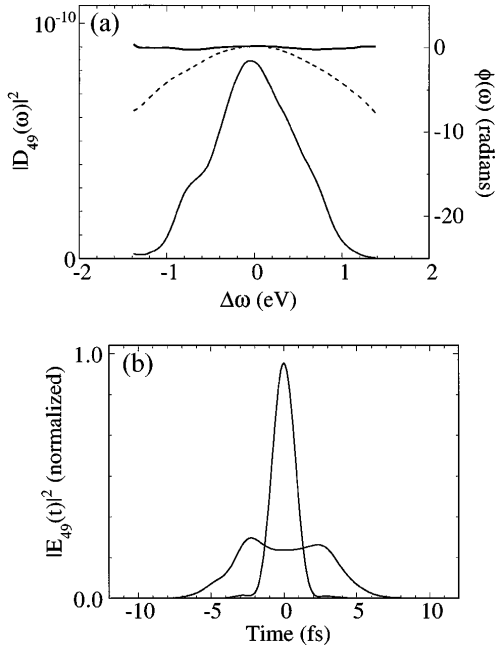

FIG. 3. (a) Envelope and phase of the 49th harmonic from Fig. 1. Initial phase (dotted line) and the phase with its quaratic component removed (upper solid line). (b) Electric field envelope of the 49th harmonic field before and after compression.

pulse width, 1.8 fs, is within a few percent of the transform limit for the spectral envelope function. The compression, by a factor of 4 in time, leads to a corresponding increase in peak intensity.

We have repeated the calculations illustrated in Figs. 1–3 for a range of peak intensities in argon and neon at 810 nm using 27 fs pulses. Qualitatively the results are the same. The harmonics at the end of the plateau exhibit a simple linear chirp which can be removed to provide <2 fs pulses from a single harmonic. The highest efficiency is obtained by compressing harmonics that are at the end of the plateau for peak intensities just below the saturation intensity. Using neon instead of argon allows us to reach higher photon energies as is illustrated in Fig. 4(a). Here we show the spectrum in the region near the end of the plateau at 4×10^{14} W/cm^2. Each of the harmonics shown has a quadratic phase structure and is compressible to a pulse width of 1.6–2.0 fs. In addition, as is clear from the figure, all three of the harmonics have almost the same group delay ($d\phi/d\omega$). Again, we have found this to be a general feature in all of our short pulse harmonics calculations. Removing the average group delay of the three harmonics and back transforming yields the intensity profile shown in Fig. 4(b). The width of the pulse is $\approx$400 as, roughly 2 orders of magnitude shorter than the pump pulse.

Interestingly, we find that the general trend that the harmonics at the end of the plateau have well-behaved

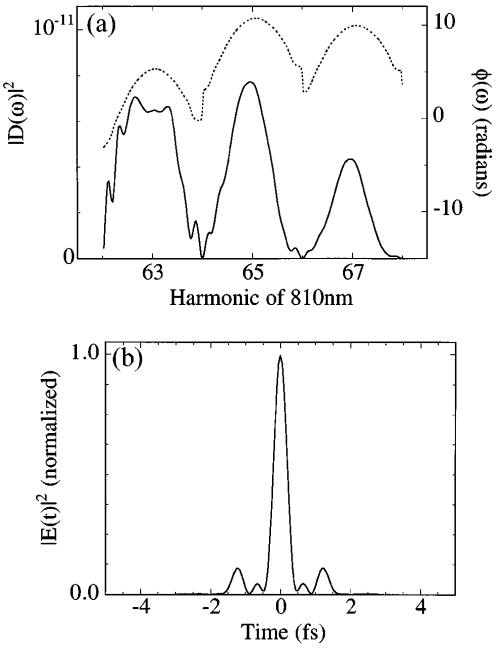

FIG. 4. (a) Part of the high harmonic spectrum for neon at 810 nm, 4×10^{14} W/cm^2 peak intensity, and 27 fs pulse width. Solid line: $|\mathcal{D}(\omega)|^2$; dotted line: $\phi(\omega)$. (b) Electric field envelope of the three harmonics in shown (a) after removing the average group delay.

phases breaks down when using a 13.5 fs pump pulse. The intensity changes so rapidly near the peak of such a pulse that the spectrum of even the cutoff harmonics shows considerable structure. These harmonics, though individually short, cannot be simply combined to give subfemtosecond pulses.

The measurement of single femtosecond VUV pulses presents its own challenges [8]. We note that with continuously adjustable dispersive optics it is possible to continuously change the pulse duration of the harmonic in question. By focusing this harmonic with a multilayer mirror, it should also be possible to reach intensities sufficient to induce a two photon transition to a high lying radiative level of a noble gas. By measuring the rate of change of an appropriate two photon induced fluorescence with respect to the separation of the gratings in the compressor it may be possible to estimate the chirp present on the pulse and the minimum pulse duration of the harmonic.

We have demonstrated that the high order harmonics emitted when a nonlinear medium interacts with an ultrafast, intense laser pulse are, in principle, compressible to subfemtosecond time scales. Our proposal to use several harmonics in combination to produce attosecond pulses is very much in the spirit of an earlier proposal by Harris *et al.* [9], recently revived by Antoine *et al.* [10]. They suggested combining several plateau harmonics to produce a train of subfemtosecond pulses, one pulse each half cycle of the driving field. Corkum [11] has proposed using a pump pulse which has a rapidly varying ellipticity to take advantage of the strong polarization dependence of the harmonic emission process and produce subfemtoseconds bursts of VUV photons. This also leads to a train of pulses [12]. By using an ultrashort pump pulse and compressing the harmonics at the end of the plateau, we believe it will be possible to generate *single* pulses which are on the order of 1 fs and shorter. These single VUV pulses are likely to be more useful for most experimental applications.

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