Coherent XUV Generation from Gases Ionized by Several Cycle Optical Pulses

C. Kan, N. H. Burnett, C. E. Capjack, and R. Rankin

Department of Electrical and Computer Engineering, University of Alberta, Edmonton, Alberta, Canada T6G 2G7

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Recent advances in solid state laser technology enable efficient generation of optical frequency laser pulses a few cycles in duration with focused intensities in excess of 10^{15} W/cm^2. When such pulses are propagated in neutral gases such as helium, ionization rates comparable to the optical frequency are induced. The very high ionization rates and the corresponding optical field strengths to which neutral atoms can be exposed with few cycle pulses present an intriguing possibility for coherent XUV generation. In this Letter, we will examine phase matching issues associated with this phenomenon. [S0031-9007(97)04254-3]

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Recent spectacular developments in ultrashort pulse solid state laser technology have led to the capability to generate pulses of a few optical cycles duration at repetition rates of about 1 kHz and pulse energy of about 1 mJ [1]. Such short duration lasers have important implications on the XUV conversion process previously studied in the context of high harmonic generation by many cycle optical pulses in ionizing gases [2].

Harmonic generation for many cycle optical pulses can be described by a quasiclassical picture [3–5] in which XUV generation results from the recombination of tunnel ionized electrons which have been accelerated by the optical field. The semiclassical picture predicts a cutoff in the single-atom XUV spectrum at an energy corresponding to $h\nu \approx I_p + 3.17U_p$, where U_p denotes the ponderomotive potential in the optical field. For pulses longer than several hundred cycles, the maximum value of U_p is limited by the saturation of ionization regardless of the peak intensity of the fundamental. Recently several single-atom theoretical studies have addressed the XUV conversion process for 5 to 25 fs driving optical pulses [6–9]. Analysis of the resulting dipole spectrum indicates that (i) ionization and XUV conversion can persist to much higher x-ray energies than is the case with longer duration pulses and (ii) the high frequency components in the dipole spectrum represent an x-ray continuum and can constitute an attosecond x-ray pulse in the temporal domain [7]. Practical consideration of the conversion process using ultrashort pulses also requires an understanding of propagation effects, which have not been previously investigated. In this Letter, we address phase matching issues associated with this process.

In the semiclassical picture, the frequency and phase of the induced nonlinear polarization relative to the phase of the driving fundamental is simply described by the quasiclassical action integral over the classical trajectory of the electron while it resides in the continuum [10]. Phase mismatch between XUV emission in the various regions of the nonlinear medium ultimately limits coherent growth. Contributing mechanisms include a geometric phase advance of the fundamental due to its focusing, intensity (of the fundamental) dependent changes in the harmonic phase through the quasiclassical action due to focusing [11], refraction or absorption of the fundamental, and phase velocity mismatch between the fundamental and harmonic due to the process of free electrons [12]. Another contribution to the phase mismatch originates from self-phase and amplitude modulation (SPAM) of the fundamental through modification of the classical trajectory and hence the quasiclassical action for electrons of a given final energy. Because the magnitude of the SPAM of the fundamental is dependent upon the gradient of the ionization front [13], this contribution becomes more significant for shorter pulses. Phase mismatch due to focusing can be ameliorated by making the gas target thin compared to the Rayleigh range [12]. This becomes an increasingly practical proposition with ultrashort driver pulses since large confocal parameters can still be achieved with low energy fundamental pulses making it meaningful to consider frequency conversion in something closely approximating a one dimensional geometry. There are no obvious solutions to the phase matching limitations imposed by ionization dependent phase velocity dispersion. We will address this aspect of the problem in the one dimensional limit.

One might anticipate that free electron induced phase mismatch coupled to self-phase and amplitude modulation of the fundamental would impose insurmountable obstacles to efficient phase matched frequency conversion under conditions where substantial ionization is occurring in a fraction of an optical cycle. However, this is not necessarily the case. We show that phase matching for several cycle driver pulses can be significantly enhanced over that anticipated from simple free electron induced phase velocity dispersion as applicable to longer duration drivers.

The square of the Fourier transform of the dipole expectation for a single He atom as predicted by the Lewenstein-Ivanov model [4] is shown in Fig. 1 for driver pulses with Gaussian temporal profiles and durations of 5 and 200 fs. The two spectra have been normalized by the respective fundamental pulse widths (energies). The fundamental wavelength has been chosen as 750 nm and the peak laser intensity was 2×10^{15} W/cm^2. A

hydrogenlike ground state was assumed [4]. Bound state depletion was included based on the Ammosov-Delone-Krainov (ADK) ionization rate [14]. For the 5 fs pulse, a fractional ionization of 40% was reached at the peak of the pulse, while for the 200 fs pulse, ionization was substantially complete when the laser intensity reached 1.9×10^{15} W/cm^2 on the leading edge of the pulse. It is apparent that the typical odd harmonic structure usually associated with frequency conversion is largely absent with a 5 fs driver and that the cutoff energy in the harmonic spectrum is extended towards shorter wavelengths. This increase is predicted to become even more dramatic as the peak driver intensity is increased up to about 4×10^{15} W/cm^2 at which point saturation of ionization is predicted for the 5 fs pulse. At this intensity, the 5 fs dipole acceleration spectrum is predicted to extend beyond 500 eV.

The time dependent square of the predicted dipole expectation for the 5 fs driver is plotted in Fig. 2(a) after applying a spectral filter to isolate only those frequency components between $h210$ and $h230$ and then applying an inverse transform. Also shown are the amplitude wave form of the driving pulse and the ionization probability of He. In terms of the semiclassical model, the high energy end of the XUV spectrum is entirely due to a pair of free electron trajectories, which originate in the continuum near the indicated time t' and reencounter the parent ion near the time t''. The phase difference between these trajectories increases as the frequency is reduced below cutoff until they interfere destructively for the first time near $h210$. At lower energies, a larger number of classical trajectories can contribute and the interference between them gives rise to the complicated structure seen in the frequency domain in Fig. 1(a).

In addition to a significant atomic response as seen in Fig. 1(a), practical XUV conversion also requires favorable phase matching lengths. We will address phase matching issues in coherent XUV generation by numerically solving the one dimensional wave equation for the driver field in the presence of field dependent ionization as given by the ADK (dc) formula [14]. The solution is coupled to an approximate equation for the high frequency field in which we neglect the influence of free electrons on propagation. This is justifiable because we are interested only in XUV frequencies, where $n_e/n_c \ll 1$ (n_c is the critical electron density). We solve the full Maxwell wave

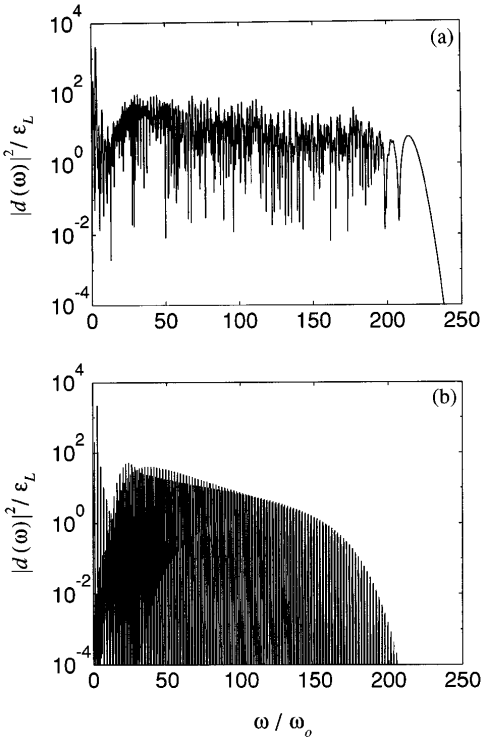

FIG. 1. Calculated power spectra for the dipole expectation for He illuminated by pulses of peak intensity 2×10^{15} W/cm^2 from the quasiclassical model for (a) a 5 fs FWHM Gaussian, 750 nm fundamental and (b) a 200 fs FWHM, 750 nm fundamental. The spectra have been normalized to the respective pulse lengths.

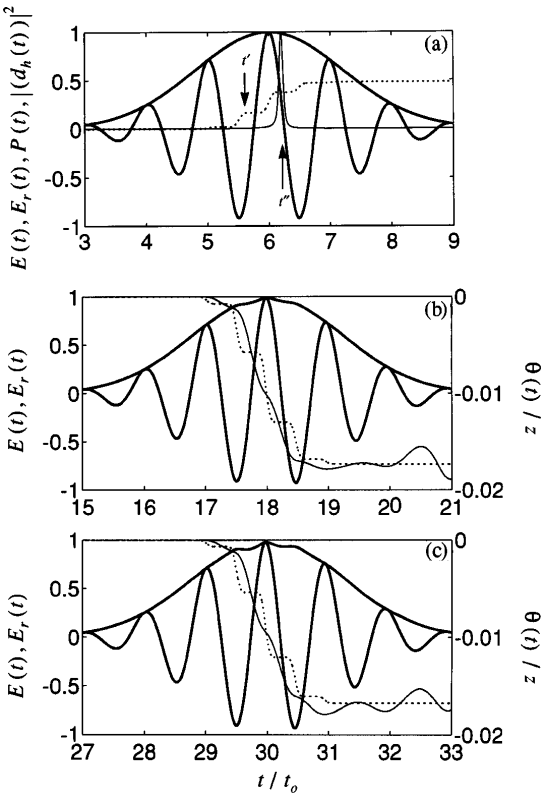

FIG. 2. (a) The incident 5 fs fundamental pulse envelope and real field (thick solid lines) at the entrance to a medium consisting of 500 Torr of He, the time dependent ionization probability (dotted line), and the time dependence of the square of the dipole expectation filtered to isolate the spectral region from $h210$ to $h230$ (thin solid line). (b) The real envelope (thick solid line) and phase modification (thin solid line) calculated for the fundamental at a distance of 9 μm into the medium compared to the expected WKB phase modification from the self-consistent time dependent ionization (dotted line). (c) Same as (b) but for a propagation distance of 18 μm. The propagation time is normalized to the fundamental laser period t_0.

equation for the fundamental field in envelope form; i.e.,

$$\frac{\partial^2 E_1}{\partial z^2} - \frac{1}{c^2}\frac{\partial^2 E_1}{\partial t^2} + \frac{2i\omega_0}{c}\left(\frac{\partial E_1}{\partial z} + \frac{1}{c}\frac{\partial E_1}{\partial t}\right) - \frac{\omega_{pe}^2(\xi_1, t)}{c^2} E_1 = 0, \tag{1}$$

where the real field is defined as $\xi_1 = \mathrm{Re}\{E_1 \exp[i(k_0 z - \omega_0 t)]\}$. Besides affording some computational advantages, solving the wave equation in envelope form provides direct access to phase and amplitude deviations of the fundamental from those corresponding to vacuum propagation. The solution for the fundamental field is then inserted into the wave equation for the XUV field neglecting dispersion; i.e.,

$$\frac{\partial^2 E_h}{\partial z^2} - \frac{1}{c^2}\frac{\partial^2 E_h}{\partial t^2} = \frac{4\pi}{c^2} n_0 d_a^h(\xi_1, t), \tag{2}$$

where d_a^h is the dipole acceleration calculated from the Lewenstein-Ivanov model. This is solved using a Fourier transform method.

Figure 2(a) shows the phase and amplitude of the numerically propagated fundamental, which was defined at the entrance ($z = 0$) to the medium (500 Torr He) as $E_0(t)\exp(i\omega_0 t)$, where $E_0(t)$ was taken as a 5 fs FWHM Gaussian. In Figs. 2(b) and 2(c) we show the fundamental and its phase and amplitude modifications due to ionization at depths of 9 and 18 μm into the medium. At 9 μm the calculated phase error in the XUV field around $h230$ is about π as will be discussed below. Thus the propagation of the fundamental over distances much in excess of 9 μm is not relevant to the problem of coherent XUV generation. Also shown is the expected WKB phase modification due to the free electron induced phase velocity (refractive index) modification ($\theta_{\mathrm{WKB}} = \pi n_e z/\lambda_0 n_c$). It is apparent that although the wave in the medium can still be well described by the incident fundamental with a small phase and amplitude distortion which grows linearly with propagation distance, distortion is no longer completely determined by the instantaneous electron density. One can use the observed linearity of the phase and amplitude distortion over lengths of relevance to the XUV phase matching problem to derive a simple first order ordinary differential equation for these quantities expressed as a complex phase (velocity) modification. This equation can be derived if one assumes that the complex envelope of the propagating fundamental can be written in the retarded frame ($z' = z, t_R = t - z/c$) as $E(z', t_R) = A(t_R)\exp[iS(z', t_R)]$, where $S = (z'/\lambda_0)F(t_R)$, and $A(t_R)$ describes the (real) field envelope at the entrance to the medium. Then making the appropriate substitution into the wave equation in envelope form [Eq. (1)], neglecting terms of order F^2, and assuming that $n_e/n_c \ll 1$ and $S(z', t_R) \ll 1$ one obtains

$$\frac{dF}{d(t_R/t_0)} - 2\pi i F - 2\pi^2 i \frac{n_e(t_R)}{n_c} = 0. \tag{3}$$

It can be verified that this equation fits quite well the observed ionization induced amplitude and phase modulation observed over the propagation lengths shown in Fig. 2.

The effects of free electron production on phase matching of the induced nonlinear polarization can be described in terms of (i) a phase velocity error associated with the deviation from c of the velocity of the release phase (t' in Fig. 2) for electrons which return with the correct energy to produce a given frequency and (ii) the effect of self-phase and amplitude modulation of the fundamental on the quasiclassical action integral of these electrons while they reside in the continuum (between t' and t''). For a long pulse driver, only the first term is significant and coherence is limited only by the deviation of the WKB phase velocity from c. That is to say, the WKB phase velocity of an electromagnetic wave in a plasma is given by $v_p = c(1 + \omega_{pe}^2/\omega_0^2)^{1/2} \approx c(1 + n_e/2n_c)$. This gives the usual coherence length limitation from phase velocity dispersion, $L_{\mathrm{coh}} \approx n_c/n_e\lambda_0/h$. For example, the 200 fs driver pulse which produces the single-atom response seen in Fig. 1(b) would produce a level of ionization of 100% by the time its intensity was sufficient to contribute a nonlinear polarization at a harmonic frequency $h = 201$. Thus the coherence length for the right end of the spectrum shown in Fig. 1(b) should be about 0.4 μm.

The calculated growth of the harmonic field for $h201$ with rigorous one dimensional numerical propagation of the 200 fs fundamental is shown in Fig. 3(a) along with the growth of the continuum emission in the region $h210$–$h230$ for the 5 fs driver pulse. The calculated phase mismatch in the harmonic field for the 5 fs driver is shown in Fig. 3(b). It is apparent that the effective coherence length is increased by about an order of magnitude with the shorter pulse.

The increased coherence length for the 5 fs pulse seems to stem from two effects. The first is that the electron density that exists just after the fundamental achieves sufficient intensity to produce the short wavelength emission is less (by about a factor of 3) in the short pulse case. The second effect is that the breakdown of the WKB approximation leads to a phase velocity error of the release phase t' which is lower (also by about a factor of 3) than that given by the instantaneous electron density and the WKB plasma dispersion relation. In Fig. 3(b) we have plotted the phase mismatch calculated solely from the phase velocity error at $t'(z)$ as observed from the fundamental propagation results. We also plot a phase error calculated solely from self-phase and amplitude modulation of the fundamental. This phase error is derived from the quasiclassical action of the appropriate electron trajectory (i.e., that which produces emission at the desired frequency) calculated for a field of the form $E_1 = GE_0(t, \omega_0)$ for $t < t'$ ($z = 0$) and $E_1 = GE_0(t, \omega_0 + \delta\omega)$ for $t > t'$ ($z = 0$), where E_0 is the driving field at $z = 0$, and G and $\delta\omega$ are used, respectively, to model the amplitude distortion and frequency shift as observed from the

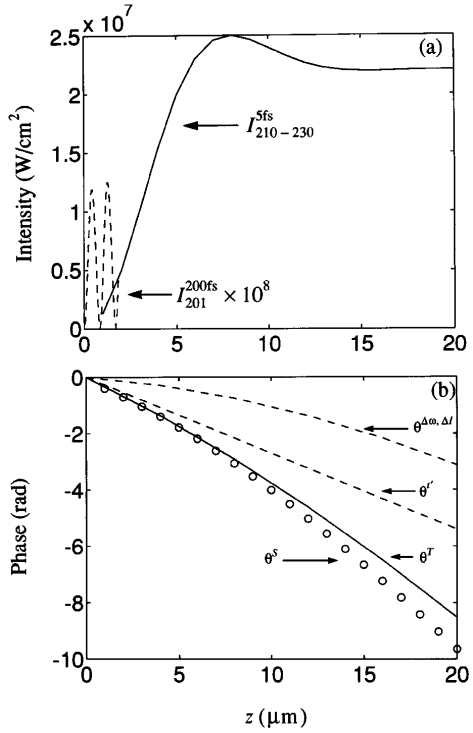

FIG. 3. (a) The calculated growth in the peak intensity of XUV emission in the region of $h200$ for 5 and 200 fs fundamental pulses propagating in 500 Torr of He. (b) The numerically calculated phase error in harmonic emission in the region of $h210-h230$ for the 5 fs fundamental in 500 Torr of He (open circles, θ^S) compared to the phase error contributions from pure self-phase modulation, $\theta^{\Delta\omega\Delta I}$, phase velocity error of the release trajectory, $\theta^{t'}$, and the sum of these contributions, θ^T.

fundamental propagation results. It is apparent that phase mismatch for the 5 fs case can still be thought of largely in terms of phase velocity dispersion although at a rate not entirely determined by the local electron density when the relevant electrons first appear in the continuum. As the fundamental pulse is shortened even further, one can anticipate that the self-phase and amplitude modulation contribution to phase mismatch will grow and eventually dominate. Our calculations confirm that this is the case for a 2.5 fs driver.

Several aspects of the strong field model used here for the atomic dipole expectation may be severely tested in the short pulse-high intensity limit, in particular the ionization rates and the rate of transverse spread of the free electron wave packet may be impacted by the onset of barrier suppression [15]. One can anticipate, however, that the XUV phase will still be determined through the quasiclassical action integral. Thus, the observations regarding the effect of ultrashort pulses on phase matching that are made here should be quite general. Somewhat similar considerations may also impact the practicality of using electron recollision to pump a short wavelength laser in the traveling wave excitation regime.

In conclusion, we have shown that the use of a few cycle driver in conjunction with frequency conversion in ionizing gases promises an economically feasible (finite pulse energy) route towards a 1D conversion geometry and improved coherence lengths for high energy XUV emission. In view of the recent spectacular progress in amplifying 5 fs Ti:AlO_3 pulses to near millijoule energies, one can anticipate that experimental studies of XUV conversion in the few cycle regime will soon be available. Such experiments will pose an important test for current models of high field atomic physics and the origin of coherent XUV generation, and may well lead to a practical source of coherent XUV radiation extending into the water window.

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Note added.—Since the submission of this Letter, several groundbreaking experiments have confirmed the extension of coherent XUV generation to wavelengths below the carbon K edge at 280 eV with ultrashort driver pulses. The group at the Technical University of Vienna [16] uses a 5 fs driver pulse and has recently observed coherent emission beyond 500 eV. The group at Michigan (Chang *et al.* [17]) uses a 25 fs driver pulse.

[1] M. Nisoli *et al.*, Opt. Lett. **22**, 522 (1997).
[2] See, for example, A. L'Huillier *et al.*, in *Atoms in Intense Laser Fields*, edited by M. Gavrilla (Academic Press, New York, 1992), pp. 139–206, and references therein.
[3] P. B. Corkum, Phys. Rev. Lett. **71**, 1995 (1993).
[4] M. Lewenstein *et al.*, Phys. Rev. A **49**, 2117 (1994).
[5] K. C. Kulander, K. J. Schafer, and J. L. Krause, in *Proceedings of the Workshop on Super Intense Laser Atom Physics (SILAP III)*, edited by B. Piraux (Plenum Press, New York, 1993).
[6] K. J. Schafer and K. C. Kulander, Phys. Rev. Lett. **78**, 638 (1997).
[7] I. P. Christov, M. M. Murnane, and H. C. Kapteyn, Phys. Rev. Lett. **78**, 1251 (1997).
[8] J. Zhou *et al.*, Phys. Rev. Lett. **76**, 752 (1996).
[9] I. P. Christov *et al.*, Phys. Rev. Lett. **77**, 1743 (1996).
[10] C. Kan *et al.*, Phys. Rev. A **52**, R4336 (1995).
[11] P. Salières, A. L'Huillier, and M. Lewenstein, Phys. Rev. Lett. **74**, 3776 (1995).
[12] T. Ditmire *et al.*, J. Opt. Soc. Am. B **13**, 408 (1996).
[13] E. Esary, G. Joyce, and P. Sprangle, Phys. Rev. A **44**, 3908 (1991).
[14] M. V. Ammosov, N. B. Delone, and V. P. Krainov, Zh. Eksp. Teor. Fiz. **91**, 2008 (1986) [Sov. Phys. JETP **64**, 1191 (1986)].
[15] S. Augst *et al.*, Phys. Rev. Lett. **63**, 2212 (1989).
[16] Ch. Spielmann *et al.*, postdeadline paper to the Conference on Quantum Electronics and Laser Science, QELS '97 (unpublished).
[17] Z. Chang *et al.*, preceding Letter, Phys. Rev. Lett. **79**, 2967 (1997).