Grating-Assisted Phase Matching in Extreme Nonlinear Optics

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We propose a new technique for phase matching high harmonic generation that can be used for generating bright, tabletop, tunable, and coherent x-ray sources at keV photon energies. A weak quasi-cw counterpropagating field induces a sinusoidal modulation in the phase of the emitted harmonics that can be used for correcting the large plasma-induced phase mismatch. We develop an analytical model that describes this grating-assisted x-ray phase matching and predicts that very modest intensities ($< 10^{10}$ W/cm^2) of quasi-cw counterpropagating fields are required for implementation.

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Phase matching techniques such as quasi-phase matching [1,2] (QPM) and grating-assisted phase matching [2,3] (GAPM) play an important role in converting light from one frequency to another, expanding the wavelength range of coherent light sources. Recently, the generation of coherent laser-like soft x-ray beams using the process of high-order harmonic generation (HHG) has received considerable attention [4]. However, efficient up-conversion into the soft x-ray region of the spectrum is challenging, because ionizing radiation is strongly absorbed by all matter. Hence, traditional phase matching techniques that rely on anisotropic crystalline solids or periodically poled materials cannot be used and novel techniques must be devised.

In high harmonic generation, an electron is first ionized by the driving laser. Once free, the electron oscillates in the continuum in response to the laser field. A small fraction of the ionized electrons recombine with the parent ion and liberate their excess energy as a short-wavelength photon. This process represents an extreme limit of nonlinear optics, where dozens, hundreds, or even thousands of visible photons, each with energy 1–2 eV, are combined together, resulting in coherent beams at photon energies up to a few keV [5]. A major limitation to date, however, is the relatively low conversion efficiency from laser light to harmonics, particularly to photon energies >130 eV. This low efficiency is *not* due to the effective nonlinearity of the process, which is nonperturbative and thus scales relatively slowly with increasing photon energy [6]. Rather, the problem to date has been the inability to efficiently phase-match this high-order conversion process [4]. In the ideal case of phase matched nonlinear conversion, the high-order polarization and the generated harmonic propagate with the same phase velocity, so that the harmonic signal builds up coherently over the entire length of the medium. If the phase velocities of the two waves differ, the signal buildup is limited to the coherence length L_c, which corresponds to that distance over which a phase slip of π accumulates between the two waves. Medium lengths longer than L_c result only in oscillation of the generated signal due to repeated destructive and constructive interference. In HHG, optimum phase matched conversion can only be obtained for photon energies below $\approx$ 130 eV. In this case, use of a hollow waveguide [7,8] or a shallow-focus geometry [9] makes it possible to balance geometrical dispersion with material and plasma dispersion. This phase matching technique relies on the presence of neutral atoms in the medium, and is therefore limited to the case of weak ionization (i.e. <0.5%–5%, depending on the gas). However, higher photon energies are generated at higher laser intensities where the medium is more highly ionized and where this type of phase matching is impossible.

Few approaches have been discussed for partial phase matching at high photon energies. Modulated waveguides partially readjust the phase slip between the driving laser and harmonic waves by periodically modulating the laser intensity [10–12]. More recently, all-optical QPM was implemented using a train of counterpropagating pulses [13]. In that work, the counterpropagating light suppresses the HHG process in regions where it intersects with the driving pulse [14–16]. A train of counterpropagating pulses can be used for implementing QPM by suppressing emission from out-of-phase regions [13]. However, these QPM implementations are limited to the case where the coherence length is larger than ~10 μm. At keV energies, however, the coherence length is typically in the micron range [17,18]. An approach that can increase the coherence length at high energies is nonadiabatic self-phase matching (NSPM) [17,18]. In NSPM, L_c is extended somewhat due to the subcycle evolution of an intense, 5 fs, pulse that results from rapid ionization of the medium. However, the increase in L_c is maintained for a short propagation distance (few microns) because of the inevitable phase slip resulting from absorption and defocusing of the driving laser. NSPM is thus not a general technique for phase matching over an extended distance. Finally, high-order difference frequency mixing has been proposed as a way to compensate for the effect of plasma on phase matching conditions [19–21].

In this Letter, we propose a new technique for phase matched frequency conversion into the x-ray region of the spectrum. A weak quasi-cw counterpropagating field induces a sinusoidal modulation on the phase of the generated harmonics, which is formally equivalent to a modulation in the refractive index for the driving laser. Exploiting this correspondence, we show that phase matching of HHG with a quasi-cw counterpropagating field is equivalent to conventional low-order harmonic generation under grating-assisted phase matching conditions [2,3]. A simple analytical model predicts the optimal conditions, such as the intensities and wavelengths of the forward and backward propagating waves, for implementing GAPM in HHG. This is the first technique that appears to be feasible for phase matching very high-order harmonics over extended distances in plasma waveguides, to generate bright, tunable, and narrow bandwidth x-ray beams at keV photon energies.

A distinctive property of HHG is that the emitted harmonics are phase shifted relative to the driving laser. This extra phase, which is primarily acquired by the electron along its femtosecond "boomerang" path under the influence of the laser field, is very large, reaching tens or hundreds of radians. It is also proportional to the intensity of the driving laser [22]. Thus, by inducing a shallow modulation in the laser intensity along the propagation direction, for example by interfering the driving laser pulse with a weak counterpropagating beam, leads to modulated phase change that can be used to correct the phase mismatch of the HHG process.

We develop a simple analytical model of the induced phase matching process. Consider a driving pulse, $E_F(t, z) = E_0 A_1(z, t)\cos(\omega_1 t - 2\pi n_1 z/\lambda_1)$, at wavelength λ_1 that propagates along z and interferes with a weak counterpropagating beam, $E_B(z, t) = E_0 r\cos(\omega_2 t + 2\pi n_2 z/\lambda_2)$, at wavelength λ_2 where E_0 is the peak field, $A_1(z, t)$ is a normalized envelope, $r \ll 1$ is a field ratio parameter, $\omega_{1,2} = 2\pi c/\lambda_{1,2}$ are the angular frequencies, and $n_{1,2} \approx 1$ are the refractive indices [Fig. 1(a)].

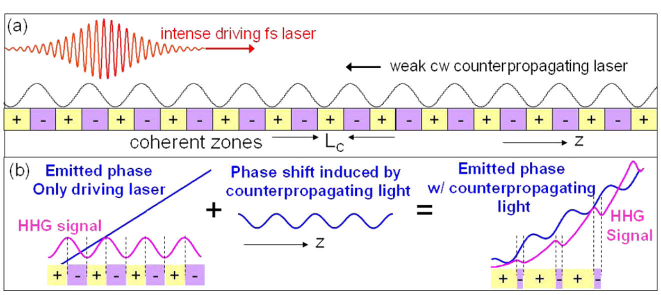

FIG. 1 (color online). (a) Schematic of grating-assisted phase matching (GAPM) in high harmonic generation (HHG) by quasi-cw counterpropagating light. (b) The combination of the medium phase mismatch and the optically-induced sinusoidal oscillation in the phase of the emitted harmonics results in a partial correction of the phase mismatch associated with the frequency conversion process.

Transforming into a frame moving in the forward direction at phase velocity of the driving field c/n_1, where c is the velocity of light at vacuum gives $E_F(\tau, z) = E_0 A_1(\tau, z) \times \cos(\omega_1 \tau)$ and $E_B(\tau, z) = E_0 r\cos(\omega_2 \tau + 2\pi z/\Lambda)$, where $\Lambda = \lambda_2/[n_1 + n_2] \approx \lambda_2/2$ and $\tau = t - n_1 z/c$. The joint intensity $I(\tau, z) \propto [E_F(\tau, z) + E_B(\tau, z)]^2$ has a component that is proportional to r and is modulated along the propagation direction with periodicity Λ (we neglect the very week term that is proportional to r^2). For example, the intensity modulation at $\tau = 0$ is proportional to $\Delta I(z) \propto 2{E_0}^2 r\cos(2\pi z/\Lambda)$. Thus, the phase change of the emitted harmonics is given by

$$\Delta\Phi_{\mathrm{HHG}}(z) = \pi z/L_c + A\cos(2\pi z/\Lambda) \tag{1}$$

where the first term is the linear growth in the phase change due to the phase mismatch of the conversion process and the second term describes the induced phase modulation with amplitude $A \propto r$ and periodicity Λ by the counterpropagating light.

To study the effect of the induced modulations on the phase matching conditions, we calculate the coherent buildup of the high-order harmonic field by

$$E_{\mathrm{HHG}} = \int_0^L E^0_{\mathrm{HHG}} \exp(i\Delta\Phi_{\mathrm{HHG}})dz \tag{2}$$

where E^0_{HHG} is the harmonic field generated in interval dz (which is insensitive to the shallow modulation in the intensity) and L is the propagation distance. It is interesting to note that Eqs. (1) and (2) were studied in the context of grating-assisted phase matching in conventional (i.e., low-order) nonlinear optics [2,3]. In that case, the sinusoidal term in Eq. (2) results from periodic variations in the linear susceptibility. It was shown [2,3] that for $\Lambda = 2mL_c$ and $L = a\Lambda$, where m and a are positive integers, inserting Eq. (1) into Eq. (2) leads to

$$E_{\mathrm{HHG}}(z = L) = E^0_{\mathrm{HHG}} L J_m(A) \tag{3}$$

where J_m is the mth-order Bessel function of the first kind. Note that the product $E^0_{\mathrm{HHG}}L$ is the harmonic field that would be obtained under perfect phase matching conditions. It is now clear that to implement GAPM in HHG, one first needs to measure or calculate L_c [16] and select λ_2 such that $\Lambda = 2mL_c$. Then, $A(A_1, A_2)$ is calculated (as shown below) and the optimal intensities for which J_m is maximized are determined. Under such optimal conditions, the conversion efficiencies (the generated power normalized by the power generated for perfect phase matching) in the first four orders ($m = 1$–4) of GAPM are 0.338, 0.237, 0.185, and 0.16, respectively. For comparison, the conversion efficiency for mth-orders conventional QPM is given by $(2/m\pi)^2$ which for the first four orders are 0.405, 0.101, 0.045, and 0.025, respectively [2]. In HHG, on the other hand, the optimal conversion efficiencies in QPM techniques [10–13] are not known but are expected to be somewhat larger than the efficiency for 2nd order QPM ($\cong 0.1$) which corresponds to complete elimination of the signal generation in out-of-phase zones. Therefore, in HHG, GAPM is expected to be more efficient than QPM techniques [10–13] in correcting the phase mismatch. Interestingly, the condition $\Lambda = 2mL_c$ corresponds to the phase matching condition, $k_{\mathrm{HHG}} = qk_1 \pm mk_2$, of a wave mixing process $\omega_{\mathrm{HHG}} = q\omega_1 \pm m\omega_2$, where k_1, k_2, and k_{HHG} are the wave vectors while ω_1, ω_2, and ω_{HHG} are the temporal frequencies of the forward, backward, and harmonic beams. This correspondence indicates that mth-order GAPM corresponds to the case in which the backward propagating beam contributes m photons to the conversion process. Notably, the most efficient conversion is obtained for $m = 1$, i.e., one backward photon. Thus, our simple model predicts what intensities of the backward and forward waves are necessary to obtain optimal high-order wave mixing.

It is instructive to emphasize that GAPM is very different from all-optical QPM because it uses a continuous-wave counterpropagating field to influence the x-ray emitted phase and not a pulse train. Moreover, in GAPM, the emitted x-ray phase is sinusoidally modulated to achieve phase matching. This is in contrast to all-optical QPM, where the x-ray emission in out-of-phase zones is suppressed by the counterpropagating light. All-optical QPM is thus akin to an amplitude modulation phase matching scheme. In contrast, GAPM represents a general x-ray phase modulation technique for efficient conversion of light into the hard x-ray region of the spectrum, that manipulates the nonlinearity and brings it closer to a true phase matching condition than all-optical QPM does.

Next, we study numerically GAPM in HHG in a specific example—HHG in a preformed plasma waveguide [23]. We consider a 20 fs driving laser beam at $\lambda_1 = 0.8\ \mu$m, and peak intensity $I_L = 5.5 \times 10^{15}\ \mathrm{W/cm^2}$ ($E_0 = 2 \times 10^9$ V/cm), propagating in a medium that consists of doubly ionized Ne ions (ionization potential is 63.45 eV) at pressure $P = 70$ torr. In addition, a weak beam at $\lambda_2 = 1.6\ \mu$m (which can be generated by using an optical parametric amplifier) propagates in the backward direction [Fig. 1(a)]. The propagations of the forward driving pulse, $E_F(z, t)$, and harmonic fields are calculated using the model in Ref. [17,18]. The backward propagating beam is very weak and therefore propagates linearly in the medium. The total optical field $E(z, \tau) = E_F(z, \tau) + E_o r \cos(\omega_2 \tau + 2\pi z/\Lambda)$ is used for calculating the ionization rate using the ADK model [24], the generated harmonic field, and the phase of the q-order generated harmonic, using the generalized Lewenstein model [17,18].

According to the scheme above, the first step is to calculate the amplitude $A(r)$. To simplify this calculation, we set the pressure to zero, thus, isolating the effect of the counterpropagating beam on the harmonic phase. Figure 2(a) shows typical change in the phase of the harmonics. As expected, the counterpropagating beam induces a sinusoidal oscillation in $\Delta\Phi$, with a periodicity

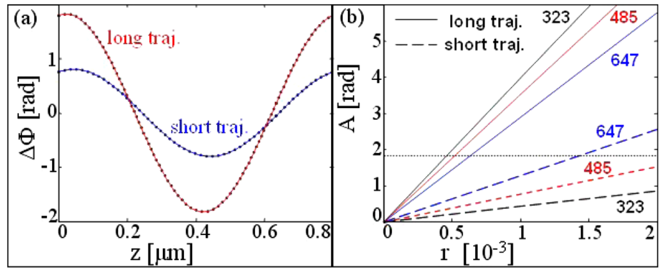

FIG. 2 (color online). Influence of weak counterpropagating light on the harmonic phase. (a) Modulation of the harmonic phase of the "short" and the "long" paths of the 647-order polarization (solid line) and fit to a cosine function (dotted line) induced by the counterpropagating beam. This is calculated for a driving laser intensity of $5.5 \times 10^{15}\ \mathrm{W/cm^2}$ and a counterpropagating laser intensity of $2.35 \times 10^9\ \mathrm{W/cm^2}$. (b) Amplitude of the phase oscillations as a function of the ratio between the peak fields of the backward and forward propagating beams. The dotted horizontal line correspond to $A = 1.84$ which is the optimal value for first order GAPM.

$\Lambda = \lambda_2/2 = 0.8\ \mu$m. Figure 2(b) shows that the amplitude of the induced oscillation (A) is indeed proportional to the field ratio r. Also, A is larger for the long path, in which the electron spends more time in the continuum and therefore interacts longer with the counterpropagating beam. (The short (long) quantum paths correspond to trajectories in which the time between ionization and recollision is $<T/2$ ($>T/2$), where $T = 2.6$ fs is the optical cycle of the driving laser.) [25] Next, we estimate the coherence length by examining the oscillations in the harmonic signal in the absence of the backward propagating beam. We find that the coherence length of harmonic order $q = 647$ is approximately $L_C \sim 0.4\ \mu$m, thus matching the condition for first order GAPM, $\Lambda = 2L_c$. From Fig. 2(b), we find that the expected optimal ratio, r, for $q = 647$ for the short and long trajectories are $r_{\mathrm{long}} = 6.34 \times 10^{-4}$ and $r_{\mathrm{short}} = 14.4 \times 10^{-4}$, respectively. We calculated the growth of the harmonic flux for r in the interval 0–5×10^{-3} and found that largest enhancement is obtained with $r = 8 \times 10^{-4}$ ($I_B \sim 3.5 \times 10^8\ \mathrm{W/cm^2}$) which is between the optimal ratios for the long and short trajectories. Figures 3(a) and 3(b) show the spectrum and flux at harmonic orders 647 ± 36 (corresponding to photon energies 990 ± 50 eV) with this optimal intensity, showing enhancement of $\approx 4 \times 10^4$ after propagation distance of $L = 200\ \mu$m. The enhancement is far from saturation, and therefore we expect that the signal would continue to increase over more extended propagation lengths ($L = 200\ \mu$m was selected to demonstrate phase matching using a reasonable computation time). Note that under the conditions assumed in our calculation, the absorption depth of the generated radiation (below the Ne K edge at $\sim$900 eV) is $>$10 cm—thus, the limiting factor is likely to be ionization loss of the driving laser. We note that phase matching over such extended 10 cm distances is experimentally feasible. Although GAPM does not require the use of waveguide propagation,

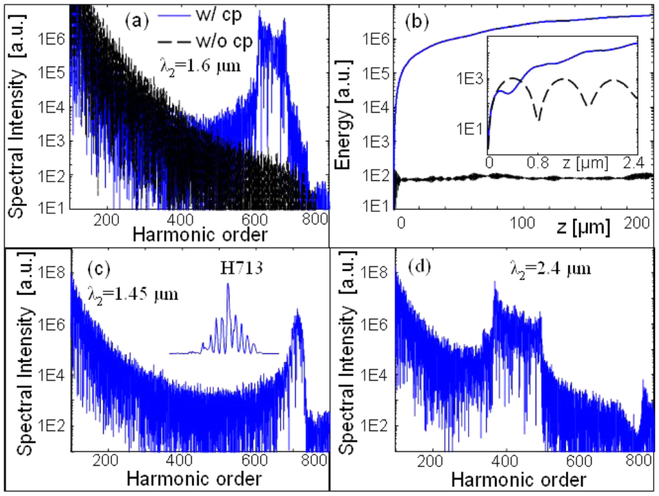

FIG. 3 (color online). Grating-assisted phase matching by counterpropagating light. (a) Harmonic spectral intensity with (solid line) and without (dashed line) a $\lambda_2 = 1.6\ \mu$m counterpropagating field. (b) Harmonic signal versus propagation distance in the spectral window $q = 647 \pm 36$, with (solid line) and without (dashed line) a counterpropagating field. The inset shows the signal in harmonic order $q = 647$ in the first 2.4 μm, showing that $L_c = 0.4\ \mu$m. Harmonic spectrum with (c) $\lambda_2 = 1.45\ \mu$m and (d) $\lambda_2 = 2.4\ \mu$m. The inset in (c) shows the enhanced region in linear scale.

plasma waveguides of 5–10 cm length have already been implemented [23]. A counterpropagating 10 cm pulse corresponds to a pulse duration of 0.6 ns, or $\sim 3 \times 10^4 \times$ the duration of the driving pulse. Given the required intensity ratio of $< 10^{-6}$, such a pulse would require a small fraction of the energy of the driving pulse. Furthermore, the counterpropagating pulse propagates in a relatively benign environment in a waveguide at low intensity and will be unaffected by the driving laser pulse until the interaction point. Finally, Figs. 3(c) and 3(d) show that the spectral window of the enhancement can be controlled by tuning the wavelength of the counterpropagating beam. In Fig. 3(c), the phase matching window partly extends beyond the cutoff energy, leading to a harmonic field with a narrow bandwidth.

Future work can make use of shaped (e.g., chirped) counterpropagating pulses to optimize the output for particular experimental requirements (e.g., spectral purity, temporal pulse duration, or chirp). Note that the temporal profile of the counterpropagating pulse maps directly onto a collision point within the medium, and thus the counterpropagating pulse amplitude and instantaneous frequency can be adjusted to compensate for local variations throughout the propagation, for example, for ionization loss of the driving laser. This provides a unique capability to phase match over extended lengths even in the extremely complex and dynamically changing environment of HHG. One can expect that adaptive (i.e., learning control) feedback of both pulse shapes will allow for further optimization of this process. Finally, multiple weak counterpropagating waves can be used to induce complex structures in the phase of the generated harmonic beams that may be used for spatiotemporal manipulation of the x-ray beams.

In conclusion, we demonstrate that weak quasi-cw counterpropagating field leads to a sinusoidal modulation of the phase of the generated high-order harmonics, which is formally equivalent to a modulation in the refractive index of the driving laser. Exploiting this correspondence, we demonstrate that phase matching in HHG with a weak counterpropagating beam is equivalent to harmonic generation in conventional nonlinear optics under grating-assisted phase matching conditions. This formalism enables us to predict the optimal intensity of the counterpropagating beam, with the finding that modest intensity ($< 10^{10}$ W/cm^2) is required for very large enhancements. This technique shows great promise for generating bright, coherent, ultrafast, and spectrally tunable hard x rays in the keV region of the spectrum, enabling applications in bio- and magnetic imaging, nanoimaging, and molecular imaging on the fastest attosecond time scales.

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[1] J. A. Armstrong *et al.*, Phys. Rev. **127**, 1918 (1962).
[2] M. M. Fejer *et al.*, IEEE J. Quantum Electron. **28**, 2631 (1992).
[3] S. Somekh and A. Yariv, Appl. Phys. Lett. **21**, 140 (1972).
[4] H. C. Kapteyn, M. M. Murnane, and I. P. Christov, Phys. Today **58**, No. 3, 39 (2005).
[5] J. Seres *et al.*, Nature (London) **433**, 596 (2005).
[6] P. B. Corkum, Phys. Rev. Lett. **71**, 1994 (1993).
[7] A. Rundquist *et al.*, Science **280**, 1412 (1998).
[8] C. G. Durfee *et al.*, Phys. Rev. Lett. **83**, 2187 (1999).
[9] E. Constant *et al.*, Phys. Rev. Lett. **82**, 1668 (1999).
[10] I. P. Christov, H. C. Kapteyn, and M. M. Murnane, Opt. Express **7**, 362 (2000).
[11] A. Paul *et al.*, Nature (London) **421**, 51 (2003).
[12] E. A. Gibson *et al.*, Science **302**, 95 (2003).
[13] X. Zhang *et al.*, Nature Phys. **3**, 270 (2007).
[14] S. L. Voronov *et al.*, Phys. Rev. Lett. **87**, 133902 (2001).
[15] J. Peatross, S. Voronov, and I. Prokopovich, Opt. Express **1**, 114 (1997).
[16] A. L. Lytle *et al.*, Phys. Rev. Lett. **98**, 123904 (2007).
[17] G. Tempea *et al.*, Phys. Rev. Lett. **84**, 4329 (2000).
[18] M. Geissler, G. Tempea, and T. Brabec, Phys. Rev. A **62**, 033817 (2000).
[19] P. L. Shkolnikov, A. E. Kaplan, and A. Lago, Opt. Lett. **18**, 1700 (1993).
[20] S. Meyer *et al.*, Phys. Rev. Lett. **76**, 3336 (1996).
[21] O. Cohen *et al.*, Phys. Rev. Lett. **98**, 043903 (2007).
[22] M. B. Gaarde *et al.*, Phys. Rev. A **59**, 1367 (1999).
[23] D. M. Gaudiosi *et al.*, Phys. Rev. Lett. **96**, 203001 (2006); B. A. Reagan *et al.*, Phys. Rev. A **76**, 013816 (2007).
[24] M. V. Ammosov, N. B. Delone, and V. P. Krainov, Sov. Phys. JETP **64**, 1191 (1986).
[25] C. Kan *et al.*, Phys. Rev. A **52**, R4336 (1995).