

Quasi-phase matching of high-order harmonic generation at high photon energies using counterpropagating pulses

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We extend all-optical quasi-phase matching of high-order harmonic generation into spectral regions where conventional phase matching is not possible. The high laser intensities required to generate harmonics at energy >130 eV, coupled with the resulting high level of ionization, preclude conventional phase matching in all nonlinear media. Selective enhancement factors between 40 and 150 in the flux of harmonics at photon energies around 140 eV are demonstrated using a train of two counterpropagating pulses. © 2008 Optical Society of America

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High-order harmonic generation (HHG) is a useful source of fully coherent ultrafast light pulses in the extreme-ultraviolet (EUV) and soft-x-ray regions of the spectrum. The unique characteristics of this source, including high spatial coherence [1] and femtosecond-to-attosecond pulse duration [2], make it particularly attractive for new types of time-resolved x-ray spectroscopy [3,4], x-ray driven dynamics [5], and coherent imaging [6]. To date, however, experiments that make use of HHG light sources have predominantly been performed with photons of energy <100 eV, because the conversion efficiency falls dramatically at higher photon energies. This diminishing efficiency is not intrinsic to the nonlinear response of the medium used to generate the harmonics but rather is due to poor phase matching.

Phase matching of the high harmonic upconversion process over extended propagation distances can be achieved in a guided-wave geometry [7] or in a gas cell [8] by balancing the effective contribution to the phase mismatch of the neutral gas, the free-electron plasma, and the geometry (waveguide or free focus). However, when the ionization in the medium exceeds a critical ionization (η_{cr}) determined by the dispersion of the gas medium [7] (e.g., $\eta_{cr}=0.5\%$ for He), dispersion due to the free-electron plasma dominates, precluding phase matching. Even for very short driving laser pulses of 10–30 fs duration, the resultant phase mismatch limits the photon energies that can be generated with reasonable efficiency to less than ≈ 130 eV [9].

Quasi-phase matching (QPM) provides a potential alternative when conventional phase matching is not possible. Recent work demonstrated that by using a counterpropagating (CP) laser field [10], all-optical QPM can be implemented in a manner that is both flexible and experimentally practical [11,12]. Using a sequence of three CP pulses, strong ($>300\times$) enhancement in HHG emission from low-pressure argon (Ar) gas was demonstrated at photon energies around 65 eV. However, conventional pressure-tuned

phase matching is possible in helium (He) at these photon energies, because the higher ionization potential of He (24.6 eV) compared with Ar (15.8 eV) allows harmonics in this energy range to be generated at lower ionization levels. Nevertheless, the flux obtainable using helium was lower than that for the all-optical QPM case because of the relatively strong absorption and weak emission of He in this region.

In this work, we extend all-optical QPM techniques to the case of He in the photon energy range around 140 eV, a photon energy that cannot be phase matched using conventional methods. Using a sequence of two CP pulses, we show that the 87th and 89th harmonic orders can be selectively enhanced by more than 2 orders of magnitude at pressures around 110 torr, and by $40\times$ as compared with the flux at the optimum He pressure (without QPM) of 500 torr. This result demonstrates that all-optical QPM techniques can be extended to higher photon energies and in dense gases to selectively phase match harmonic generation in regimes where conventional phase matching is not possible using any gas.

Figure 1 shows a schematic of the experimental setup. The output of a 1 kHz, Ti:sapphire amplifier system is separated into two beams before pulse compression. Approximately 1 mJ of the laser energy is compressed to a pulse duration of 27 fs FWHM to

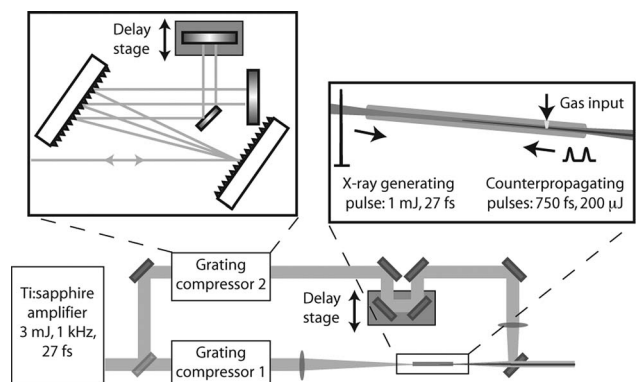


Fig. 1. Experimental setup for all-optical QPM in He.

generate high-order harmonics. This beam is then focused into a 6 cm long, 150 μm inner diameter, fused silica waveguide. He gas is introduced into the waveguide through a small laser-drilled hole 5 mm from the exit, creating a slow ramp-up in pressure throughout most of the interaction region, and a rapid pressure drop at the exit. The remaining laser pulse energy (~ 1 mJ) is partially compressed to ~ 1.5 ps FWHM in a separate grating pair compressor. This pulse propagates through a motorized delay stage and is then coupled into the opposite end of the waveguide via a mirror with a 3 mm diameter hole drilled in the center. This hole allows the forward-going high harmonic beam to pass through, while directing ~ 400 μJ of the CP pulse energy into the waveguide. The forward-going fundamental light is blocked past the center-drilled mirror using a 200 nm silver and a 200 nm zirconium filter, which transmit the photon energy band of interest. The spectrometer was calibrated using a removable 400 nm Si filter, which has an absorption edge at 99 eV. The HHG spectrum is detected using a flat-field, imaging x-ray spectrometer (Hettrick Scientific) and an x-ray sensitive CCD camera (Andor Technology).

To create two individually controllable CP pulses, a mirror in the compressor directs half the dispersed spectrum to a separate retroreflector (see Fig. 1). This retroreflector is mounted on a delay stage to control the timing of one half of the spectral energy in the beam relative to the other. Because the dispersion of the CP pulse is dominated by linear chirp, each pulse is approximately 750 fs FWHM in duration. Thus both the width and the separation of the two CP pulses are independently and continuously variables, which is important for implementing all-optical QPM.

Without the CP pulses present, harmonics are observed from He at pressures of 110 torr up to the 87th order at 134 eV (Fig. 2(a)). The presence of even one CP pulse in the center of the waveguide increases the brightness of several harmonic orders by suppressing harmonic production that would otherwise cause destructive interference [12,13]. This indicates that the falloff at the 87th harmonic is due to phase mismatch at ionization levels greater than η_{cr} . Moreover, at the laser intensity used, the cutoff photon energy is well above the highest photon energies observed. The enhancement is selective, occurring only over a limited range of photon energies, because the coherence length is inversely proportional to harmonic order and the level of ionization in the gas. Using a quantitative analysis, we recently predicted that for a sech^2 CP pulse, the largest enhancement is obtained when the CP FWHM corresponds to $0.92L_C$ [14].

The experimentally measured enhancement factor using two CP pulses is shown in Fig. 2(b). The second pulse enhances the harmonic emission still further if it is positioned at an adjacent out-of-phase coherence region, at an effective distance of twice the coherence length of the nonlinear conversion process [11,14]. The effect of the two CP pulses is thus to enhance the brightness of the 87th and 89th harmonic orders by

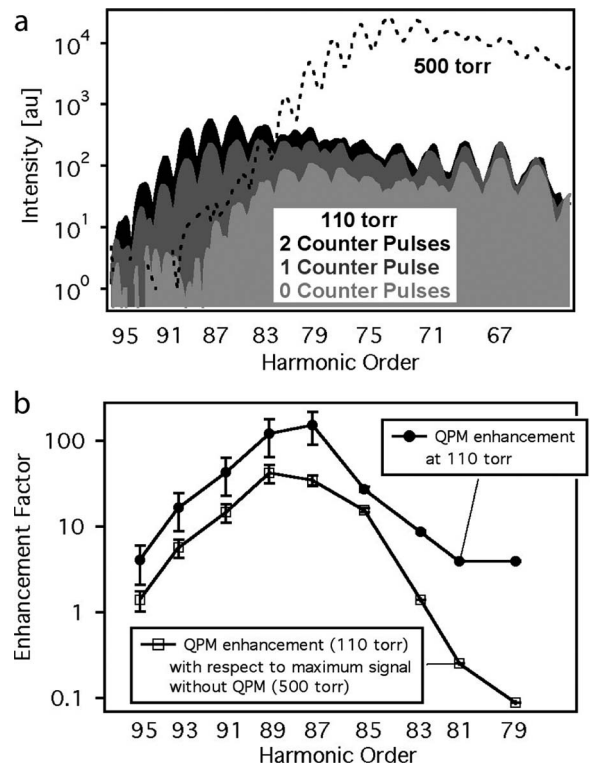


Fig. 2. (a) HHG spectra from 110 torr He with with zero (light gray), one (medium gray), and two (black) counter-propagating pulses. The dotted black curve shows HHG spectra under similar laser conditions, but at a pressure of 500 torr, which is optimal for conventional phase matching of lower-order harmonics up to $q \approx 77$ (120 eV), at ionization levels below critical. (b) Enhancement factors for harmonic orders 79–95. The circles show the ratio of QPM emission using two counterpropagating pulses to that without using counterpropagating pulses at a pressure of 110 torr. The squares show the ratio of QPM emission at 110 torr to that without counterpropagating pulses at a pressure of 500 torr.

factors of approximately $150\times$ and $120\times$, respectively. The highest observable harmonic is also extended, owing to partial suppression of out-of-phase zones for those harmonic orders. To obtain the maximum enhancement, both the CP pulse width and separation were optimized, resulting in a pulse duration of ~ 750 fs FWHM and a separation of ~ 3 ps.

The coherence length, for example, for the 89th order, can be determined in this experiment through two independent measurements. First, the CP pulse was scanned through the interaction region [12], with the observed output oscillations corresponding to a coherence length of 217 ± 25 μm for the 89th harmonic. Second, the separation between a pair of CP pulses can be varied, and an optimum of 3 ps (~ 900 μm), corresponding to $4L_C$, yields $L_C = 225 \pm 25$ μm . Also, the single-pulse FWHM of 225 ± 15 μm approximately matches the prediction that the largest QPM enhancement occurs when the pulse width corresponds to $0.92L_C \sim 203$ μm . This measured coherence length can then be converted to a phase mismatch, $\Delta k = \pi/L_C$, which can be modeled in terms of the observed experimental parameters as

$$\Delta k = q \left\{ \left(\frac{u_{11}^2 \lambda_0}{4\pi a^2} \right) - P \left((1 - \eta) \frac{2\pi}{\lambda_0} \delta n - \eta [N_{\text{atm}} r_e \lambda_0] \right) \right\}, \quad (1)$$

where q is the harmonic order, u_{11} is the lowest-order waveguide mode factor, λ_0 is the center wavelength of the driving laser, a is the inner radius of the hollow waveguide, P is the pressure, η is the ionization level, r_e is the classical electron radius, N_{atm} is the number density of atoms at 1 atm, and δn is the difference between the indices of refraction of He at the fundamental and harmonic wavelengths. This expression can then be solved for the ionization level at which the 140 eV harmonics are generated, giving $\eta \sim 1.7\%$ for the above experimental parameters. This number is also consistent with the ionization level predicted at the time when the cutoff intensity reaches 140 eV, as calculated using Ammosov–Delone–Krainov ionization rates [15]. The critical ionization for He, η_{cr} , may be determined by solving for the ionization level at which the dispersion from the neutral gas is balanced by that of the plasma [16] (neglecting the dispersion from the waveguide):

$$\eta_{\text{cr}} = \left[\left(\frac{\lambda_0 N_{\text{atm}}}{2\pi \delta n} \right) \left(1 - \frac{1}{q^2} \right) + 1 \right]^{-1}. \quad (2)$$

The critical ionization for He at this photon energy is 0.5%, and so it is clear that the measured harmonics are generated at a level of ionization $3\times$ higher than can be conventionally phase matched in any gas and in any geometry, including jets, cells, or waveguides.

In the experiments discussed above, the He pressure at the input to the waveguide was kept constant at 110 torr. To determine the absolute value of the enhancement we obtain using all-optical QPM, we also optimized the He pressure, particularly for highest emission at these orders around $q=89$, without any CP pulses present. Overlaid in Fig. 2(a) is a spectrum taken at the optimum 500 torr pressure. Figure 2(b) shows the enhancement factor obtained by all-optical QPM at 110 torr and also the enhancement of two-pulse QPM at 110 torr compared with the maximum emission without QPM at 500 torr, i.e., the most conservative measure of the enhancement factor. It is seen that the two-pulse QPM signal at 110 torr enhances the HHG output for harmonics >83 rd in all measures.

In the future, further enhancements are possible using a longer train of counterpropagating pulses, allowing the coherent addition of HHG over a longer distance. The absorption depth of He at this pressure and photon energy is nearly 2 cm, making it likely that much larger enhancements will be possible. Efficient implementation of all-optical QPM will require pulse trains with individually adjustable width

and separation to match the spatially varying coherence length present in the waveguide. Since higher photon energies have increasingly shorter coherence lengths, pulse trains with several pulses will be necessary for high enhancements. This work extends the potential of all-optical QPM for creating bright, coherent EUV and soft-x-ray sources based on extreme nonlinear optics.

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References

1. R. A. Bartels, A. Paul, H. Green, H. C. Kapteyn, M. M. Murnane, S. Backus, I. P. Christov, Y. W. Liu, D. Attwood, and C. Jacobsen, *Science* **297**, 376 (2002).
2. M. Hentschel, R. Kienberger, Ch. Spielmann, G. A. Reider, N. Milosevic, T. Brabec, P. Corkum, U. Heinzmann, M. Drescher, and F. Krausz, *Nature* **414**, 509 (2001).
3. J. Itatani, J. Levesque, D. Zeldler, H. Niikura, H. Pepin, J. C. Kieffer, P. B. Corkum, and D. Villeneuve, *Nature* **432**, 867 (2004).
4. N. L. Wagner, A. Wuest, I. P. Christov, T. Popmintchev, X. B. Zhou, M. M. Murnane, and H. C. Kapteyn, *Proc. Natl. Acad. Sci. U.S.A.* **103**, 13279 (2006).
5. E. Gagnon, P. Ranitovic, X.-M. Tong, C. L. Cocke, M. M. Murnane, H. C. Kapteyn, and A. S. Sandhu, *Science* **317**, 1374 (2007).
6. R. L. Sandberg, A. Paul, D. A. Raymondson, S. Hadrich, D. M. Gaudiosi, J. Holtsnider, R. I. Tobey, O. Cohen, M. M. Murnane, H. C. Kapteyn, C. Song, J. Miao, Y. Liu, and F. Salmassi, *Phys. Rev. Lett.* **99**, 098103 (2007).
7. A. Rundquist, C. G. Durfee, Z. H. Chang, C. Herne, S. Backus, M. M. Murnane, and H. C. Kapteyn, *Science* **280**, 1412 (1998).
8. P. Salieres, A. L'Huillier, and M. Lewenstein, *Phys. Rev. Lett.* **74**, 3776 (1995).
9. H. Kapteyn, O. Cohen, I. Christov, and M. Murnane, *Science* **371**, 775 (2007).
10. S. L. Voronov, I. Kohl, J. B. Madsen, J. Simmons, N. Terry, J. Titensor, Q. Wang, and J. Peatross, *Phys. Rev. Lett.* **87**, 133902 (2001).
11. X. Zhang, A. L. Lytle, T. Popmintchev, X. Zhou, M. M. Murnane, H. C. Kapteyn, and O. Cohen, *Nat. Phys.* **3**, 270 (2007).
12. A. L. Lytle, X. Zhang, J. Peatross, M. M. Murnane, H. C. Kapteyn, and O. Cohen, *Phys. Rev. Lett.* **98**, 123904 (2007).
13. J. Peatross, S. Voronov, and I. Prokopovich, *Opt. Express* **1**, 114 (1997).
14. O. Cohen, A. L. Lytle, X. Zhang, M. M. Murnane, and H. C. Kapteyn, *Opt. Lett.* **32**, 2975 (2007).
15. M. V. Ammosov, N. B. Delone, and V. P. Krainov, *Zh. Eksp. Teor. Fiz.* **91**, 2008 (1986).
16. A. Rundquist, "Phase-matched generation of coherent, ultrafast x-rays using high harmonics," Ph.D. thesis (Washington State University, 1998).