Extended phase matching of high harmonics driven by mid-infrared light

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We demonstrate that phase-matched frequency upconversion of ultrafast laser light can be extended to shorter wavelengths by using longer driving laser wavelengths. Experimentally, we show that the phase-matching cutoff for harmonic generation in argon increases from 45 to 100 eV when the driving laser wavelength is increased from 0.8 to 1.3 μm. Phase matching is also obtained at higher pressures using a longer-wavelength driving laser, mitigating the unfavorable scaling of the single-atom response. Theoretical calculations suggest that phase-matched high harmonic frequency upconversion driven by mid-infrared pulses could be extended to extremely high photon energies. © 2008 Optical Society of America

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High-order harmonic generation (HHG) is a unique source of femtosecond-to-attosecond duration soft x-ray beams that has made new studies of atoms, molecules, and materials possible, as well as enabling high-resolution coherent imaging using a tabletop source. To date, however, most applications of HHG radiation employ relatively low-energy extreme-UV wavelengths, because the efficiency of the HHG process decreases rapidly at high photon energies. This decrease is not fundamental to the nonperturbative HHG process, whose effective nonlinearity scales relatively slowly with photon energy, but rather results from the large phase mismatch between the harmonic and the driving laser fields. Phase-matching high harmonic upconversion at very short wavelengths using a 0.8 μm driving laser is difficult because of the required high incident laser intensity that strongly ionizes the medium, creating a large free-electron dispersion that prevents phase matching [1]. True phase matching of the HHG process is thus limited to a parameter range where the level of ionization of the medium is low, so that neutral atom dispersion can balance the anomalous free-electron plasma dispersion [2,3]. For a 0.8 μm driving laser, the "critical" ionization levels above which true phase matching is not possible are $\eta_{cr} \approx 3.8\%$ for argon and $\approx 0.5\%$ for helium, corresponding to photon energies of ~ 45 and ~ 130 eV, respectively. Quasi-phase matching (QPM) techniques have been employed for generating short-wavelength harmonics at ionization levels where true phase matching is not possible [4–7]. However, by their nature, all QPM techniques only partially correct the phase mismatch and therefore are fundamentally less efficient than true phase matching.

Another important limit in HHG is the maximum photon energy (cutoff energy) that can be generated by the laser, regardless of phase matching. This cutoff energy is given by $h\nu_{\max} = I_p + 3.2U_p$, where I_p is the ionization potential of the gas, $U_p \propto I_L \lambda_L^2$ is the quiver energy of the recolliding electron, and I_L and λ_L are the intensity and wavelength of the driving laser. The favorable λ_L^2 scaling of the cutoff has motivated studies of HHG with mid-IR driving pulses [8]. Significant extension of the cutoff energy has been demonstrated in several experiments [8,9]. However, it was recently found theoretically that the single-atom yield scales as $\lambda_L^{-5.5\pm0.5}$, which greatly reduces the efficiency of HHG driven by longer wavelengths [10]. Thus increasing the HHG yield by phase matching the conversion process is critical to obtain a usable flux. Indeed, it was recently suggested theoretically that favorable "self-phase matching" conditions might be realized with mid-IR pulses [11].

In this Letter, we show experimentally and theoretically that it is possible to extend true phase matching of the high harmonic generation process to significantly higher photon energies using mid-IR wavelengths. Using a 1.3 μm driving laser and pressure-tuned phase matching in a hollow waveguide, we show that the region of phase matching using Ar gas extends from ~ 45 eV (0.8 μm driver) to ~ 100 eV (1.3 μm driver). Moreover, we show that phase matching is obtained at higher pressures using longer driving wavelengths, thereby helping to mitigate the unfavorable scaling of the single-atom efficiency. Most importantly, as a result of the ability to implement phase matching of the HHG process at high gas pressure, the high harmonic output from Ar in the phase-matched region of 70–100 eV driven by 1.3 μm light is of comparable brightness to that of phase-matched He using a 0.8 μm driver. This is, to our knowledge, the first demonstration that mid-IR lasers can generate sufficient HHG flux for application experiments.

Pressure-tuned phase-matched high harmonic generation involves creating a near plane-wave propagation geometry by proper coupling of high-intensity light into a hollow waveguide [3]. In this geometry, the phase mismatch is a sum of contributions from the pressure-dependent neutral atom and free-electron dispersion, and the pressure-independent waveguide dispersion:

$$\Delta k = q\left\{\left(\frac{u_{11}{}^2\lambda_L}{4\pi a^2}\right) - P\left((1-\eta)\frac{2\pi}{\lambda_L}\Delta\delta - \eta N_{atm} r_e \lambda_L\right)\right\}.$$

Here q is the harmonic order, u_{11} is the mode factor, a is the inner radius of the 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 $\Delta\delta$ is the difference between the indices of refraction of the gas at the fundamental and harmonic wavelengths. Phase matching ($\Delta k = 0$) can be achieved by simply tuning the gas pressure inside the waveguide. However, this is only possible as long as η is lower than the critical ionization level: $\eta_{cr}(\lambda_L) = [\lambda_L{}^2 N_{atm} r_e / (2\pi\Delta\delta(\lambda_L)) + 1]^{-1}$ [3]. In the context of phase matching, the critical ionization results in a "phase-matching cutoff energy," which is the largest photon energy that can be generated before the ionization of medium reaches η_{cr}. This phase matching cutoff changes only slightly for shorter laser pulses and thus represents a fundamental limit for conventional phase matched upconversion.

We first evaluate the dependence of the phase-matching cutoff energy on the wavelength of the driving laser by calculating the ionization level of the gas using the Amossov–Delone–Krainov tunneling model [12]. We assume a hyperbolic secant pulse of eight cycles at FWHM (35 fs at 1.3 μm), with the peak intensity selected such that the ionization level on axis at the peak of the pulse corresponds to the wavelength dependent critical ionization [e.g., $\lambda_L = 1.3\ \mu$m, $I_L(\text{Ar}) = 1.58\times10^{14}\ \text{W/cm}^2$, $I_L(\text{Ne}) = 3.72\times10^{14}\ \text{W/cm}^2$, and $I_L(\text{He}) = 5.2\times10^{14}\ \text{W/cm}^2$]. Figure 1 shows the result of this calculation, which indicates a significant increase in the maximum phase-matched photon energy with increasing driving laser wavelength. In fact, for $\lambda_L = 2.8\ \mu$m (or $10^3\times$ lower single atom yield compared with $\lambda_L = 0.8\ \mu$m), phase-matched conversion can be obtained up to photon energies approaching 1 keV. Although the critical ionization level decreases as a function of λ_L, the quiver energy U_p of the returning electron increases with wavelength. Thus, a given energy harmonic can be generated at lower laser intensities where the medium is less ionized. Therefore the phase-matching region moves to significantly higher photon energy. Finally, we note that when modal averaging is taken into account, the peak intensity can increase further (<20%), which leads to an approximately 10% higher phase-matching limit [3].

Experimental data verifying the extension of the pressure-tuned phase-matching cutoff using long driving-laser wavelengths is presented in Fig. 2. High-energy (up to 2.1 mJ), 35 fs pulses at 1.3 μm were generated through three-stage optical parametric amplification (OPA) of a chirped white light continuum. Conversion efficiencies of 28% were achieved for the OPA signal at 1.3 μm (47% total efficiency for both signal and idler) when driven by a 7.5 mJ, 10 Hz, 800 nm pump pulse of duration 25 fs. These pulses were then focused into a hollow waveguide (250 μm inner diameter) filled with Ar. The gas was injected into the waveguide through two laser-drilled holes, allowing a static pressure to be maintained over an interaction length of $L_{med} = 10$ mm, while two 5 mm end sections enabled differential pumping to a vacuum of 10^{-3}–10^{-7} Torr.

Figure 2 shows that using a 1.3 μm driving laser, true phase matching in Ar extends up to 100 eV. In comparison, using a 0.8 μm driving laser phase matching in Ar is limited to 45 eV [1–3]. Based on the driving laser intensity of $2\times10^{14}\ \text{W/cm}^2$ and the pulse duration of 35 fs, calculations show that the single-atom cutoff reaches 105 eV at the instant when the ionization level reaches $\eta_{cr} = 1.5\%$. Consistent with this calculation, the experimental phase-matching limit is estimated to be 100 eV at the point where the usable signal drops 50% from the plateau

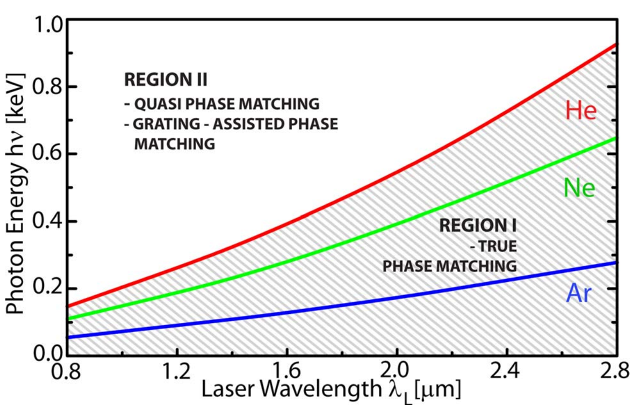

Fig. 1. (Color online) Theoretically predicted "phase-matching cutoff energies" as a function of the laser wavelength λ_L showing that true phase matching could be extended to ~1 keV at ionization levels below η_{cr} (region I). Quasi-phase matching or grating-assisted phase-matching techniques must be implemented at higher energies (region II).

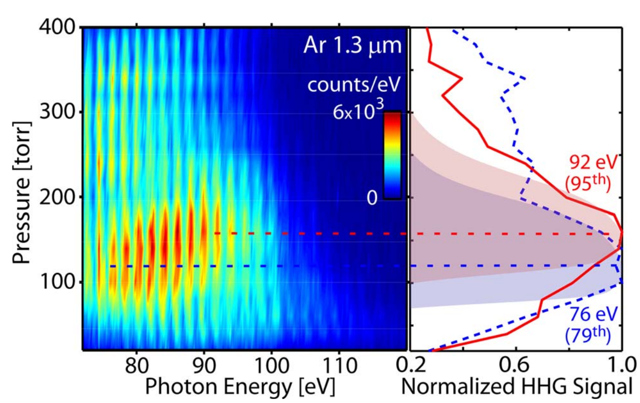

Fig. 2. (Color online) (Left) Experimental HHG spectrum through 200 nm Zr filter as a function of pressure, demonstrating phase-matched emission from Ar up to 100 eV using a 1.3 μm driving pulse. (Right) Lineouts showing the pressure dependence of the harmonics around 76 eV (dashed line) and 92 eV (solid line) together with theoretical curves.

level. The HHG spectrum as a function of pressure shows that higher harmonics are phase matched at higher pressures, as expected for pressure-tuned phase matching. Since higher harmonic orders are generated at higher η, higher neutral atom pressures are required to compensate for the increasing free-electron induced phase mismatch. A theoretical fit that includes absorption in the medium shows that the most efficient upconversion for harmonics around 76 and 92 eV is confined to ionization level intervals of 1.0–1.3% and 1.2–1.3%, respectively.

Although our findings are applicable to HHG in gas jets and waveguides, the hollow-waveguide geometry is particularly useful in the case of mid-IR driving wavelengths. First, it enables an optimal pressure-length product for phase matching. The higher pressures of 70–200 Torr required for phase matching at 1.3 μm (compared to 10–50 Torr at 0.8 μm [3]) would be difficult to obtain in a free-space jet. Second, guiding of the laser beam maintains high intensity over an extended interaction distance, even for a relatively long-wavelength driver (the experimentally measured waveguide transmission was >70%). This is in contrast to a free-focusing geometry, where for the same waist the confocal parameter shrinks as $1/\lambda_L$, corresponding to a drop of $1/\lambda_L^2$ in harmonic intensity. Thus, although the $\lambda_L^{-5.5}$ scaling of the single-atom efficiency corresponds to an 11–18-times weaker emission at $\lambda_L = 1.3$ μm compared with 0.8 μm, the ability to phase match the process allows the photon flux to still be high. For a direct comparison of harmonic flux in the extended phase-matching region, we used a 0.8 μm beam to generate phase-matched harmonics in He, using approximately 2.6× higher peak laser intensity than for a 1.3 μm beam, so that the same 130 eV cutoff was achieved. The similar absorption lengths at optimal pressures for He ($L_{abs} = 1.31$ mm at 500 Torr) and Ar ($L_{abs} = 1.34$ mm at 160 Torr) around 100 eV ensure qualitatively the same coherent buildup of HHG over the interaction length. This distance was selected to be close to the saturation length of $\sim 10L_{abs}$ at coherence length $L_c = \pi/\Delta k \gg L_{abs}$ [13]. Figure 3(a) shows that the HHG flux from Ar driven by a 1.3 μm beam is comparable to that of the brightest source in the region, phase-matched He at 0.8 μm. At low pressure and almost full ionization, HHG emission from Ar driven by 1.3 μm light extends up to 200 eV [Fig. 3(b)] with reduced conversion efficiency (>5 times). In this case the well-defined plateau and the sharp cutoff observed under optimal phase-matching conditions [Ar in Fig. 3(a)] disappear owing to increased free-electron density.

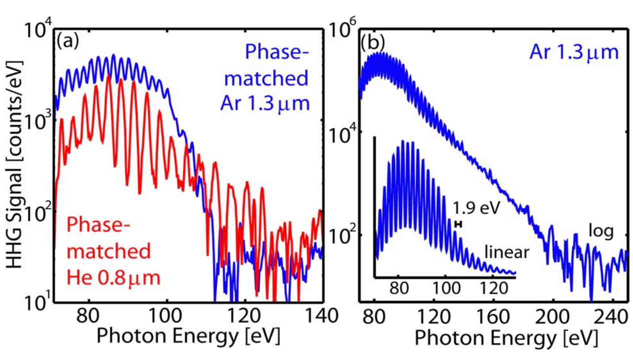

Fig. 3. (Color online) (a) Comparison between HHG from phase-matched Ar (160 Torr, 2.0×10^{14} W/cm^2) driven by 1.3 μm light and phase-matched He (500 Torr, 5.2×10^{14} W/cm^2) driven by 0.8 μm light. (b) Harmonic spectrum from almost fully ionized Ar at lower pressure (60 Torr) driven by a 1.3 μm pulse at an intensity of 4.5×10^{14} W/cm^2, showing emission up to 200 eV.

In summary, we demonstrate experimentally and theoretically for the first time (to our knowledge) that increasing the wavelength of the driving pulse in high harmonic generation significantly increases the cutoff energy for true phase matching. We also show that pressure-tuned phase matching of the HHG process in a hollow waveguide using longer wavelength drivers can mitigate the low single-atom response. This result demonstrates a path for producing bright coherent x-rays for biological and materials imaging up to photon energies approaching 1 keV.

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