Phase matching, quasi-phase matching, and pulse compression in a single waveguide for enhanced high-harmonic generation

Xiaoshi Zhang, Amy Lytle, Tenio Popmintchev, Ariel Paul, Nick Wagner, Margaret Murnane, and Henry Kapteyn
JILA, Department of Physics, and National Science Foundation Engineering Research Center in Extreme Ultraviolet Science and Technology, University of Colorado at Boulder and National Institute of Standards and Technology, Boulder, Colorado 80309-0440

Ivan P. Christov
Department of Physics, Sofia University, Sofia, Bulgaria

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We demonstrate, for the first time to our knowledge, that the efficient region of high-harmonic generation can be shifted from lower to higher photon energies by combining phase matching, quasi-phase matching, and pulse compression in a single gas-filled waveguide. An intrawaveguide pulse compression process that works through a combination of ionization-induced refraction and guiding shortens the laser pulse as it propagates through an Ar-filled waveguide. This leads to enhanced harmonic emission at high photon energies near 95 eV while it reduces emission at low photon energies near 45 eV. The waveguide geometry also mitigates ionization-induced refraction, allowing Ar gas with high effective nonlinear susceptibility to be used. © 2005 Optical Society of America

High-order harmonic generation (HHG) is a useful source of coherent, ultrastart light in the extreme-UV region of the spectrum, with applications in high-resolution imaging as well as site-specific spectroscopy and microscopy. In HHG, an intense laser pulse is focused into a medium. The highly nonlinear interaction between the laser light and the atoms creates higher-order harmonics that emerge from the medium as a coherent, low-divergence beam. To generate the brightest harmonics from a medium, several conditions must be met. First, the conversion process ideally should be phase matched. This is challenging in the presence of significant levels of ionization that introduce a large plasma-induced dispersion and that prevent the laser and the harmonic light from propagating at the same phase velocity. Second, a short pulse is desirable, since this reduces the ionization at a particular laser intensity, allowing phase matching to be more easily achieved. And finally, since large atoms with high effective nonlinearity susceptibilities generate the brightest harmonics, it is desirable to use the larger noble-gas atoms. However, the efficient photon-energy range that can be reached using larger, more easily ionized noble-gas atoms is restricted by increased ionization and refraction in the medium, which refract the laser beam and prevent efficient phase matching.

Here we address all these criteria simultaneously, using a single hollow-core waveguide. We demonstrate that we can shift the efficient region of HHG from 45 to 95 eV by combining phase matching, quasi-phase matching (QPM), and pulse compression in a single gas-filled waveguide. The intrawaveguide pulse compression process, which works through a combination of ionization-induced refraction and guiding, shortens the laser pulse as it propagates through the waveguide, leading to reduced ionization for a given peak intensity and improving phase matching for higher photon energies. Moreover, the waveguide geometry reduces ionization-induced defocusing and introduces a periodic refocusing of the laser, which also improves phase matching. This shifts the maximum HHG in Ar from 45 to 95 eV for the first time to our knowledge while it reduces emission at the lower, 45 eV, photon energies. Finally, we demonstrate that QPM in modulated waveguides enhances the harmonic output still further. This combined approach to phase matching of HHG that uses compressed pulses and QPM within the same waveguide significantly increases the harmonic flux and efficiency obtained in Ar at photon energies near 95 eV, as well as simplifying the experimental geometry.

In our experiment, we focus 2.5 mJ, 27 fs pulses from a Ti:sapphire laser system centered at a wavelength of 800 nm, and at a repetition rate of 1 kHz, into a 150 μm diameter, 2.5 cm long hollow-core waveguide filled with Ar. The laser peak intensity at the input of the hollow-core waveguide is ~7 x 10^14 W/cm^2, corresponding to ~60% energy throughput through the waveguide with no gas present. This throughput reduces further to ~30% for pressures near 5 Torr. The waveguide can be either straight or periodically modulated. Figure 1 shows the laser pulse intensity both before and after propagating through the waveguide filled with 5 Torr of Ar. At these low pressures, the laser pulses are compressed from ~30 fs before the waveguide to ~15 fs after the waveguide, with a fast 10 fs rise time and without the need for dispersion compensation.
expression, $I_p$, while $Up$ $eV), while to 10 fs, respectively.

width and rise time decrease from 27 to 15 fs and from 30 to 10 fs, respectively.

The physical mechanism for the observed pulse compression is due to three-dimensional spatiotemporal reshaping of pulses by ionization-induced spectral broadening, plasma-induced refraction, and guiding in the hollow waveguide.\(^3\) The waveguide also preserves or increases the available laser intensity in the waveguide by reducing ionization-induced defocusing. This pulse compression mechanism is key to using Ar as the nonlinear medium for phase matching.

Figure 2 shows the measured harmonic emission from a straight waveguide for three gas pressures. At the laser intensities present within the guide ($\approx 5 \times 10^{14}$ W/cm$^2$), the maximum cutoff harmonic order expected in Ar is $h\nu = I_p + 3.2U_p = 100$–150 eV. In this expression, $I_p$ is the ionization potential of Ar (15.8 eV), while $U_p$ is the ponderomotive potential ($\approx I_{laser} \lambda^2$, where $I_{laser}$ is the laser intensity and $\lambda$ is the laser wavelength). However, the region of bright harmonic emission is limited not by the available laser intensity but by phase matching. Therefore, as expected, for high gas pressures near 25 Torr, the harmonic emission from Ar peaks at 45 eV under our conditions.\(^3\) At this pressure and photon energy, the positive dispersion of the neutral Ar atoms balances the negative dispersion of the plasma and waveguide. However, at low gas pressures near 5 Torr, the bright, phase-matched emission shifts to higher energies from 55 to 100 eV (Fig. 2), while the emission at 45 eV drops. This is the same gas pressure at which the plasma-induced pulse compression optimizes.\(^10\)

These observations are in contrast to what is usually observed from HHG at low pressures: The net harmonic emission intensity usually drops dramatically, while the cutoff extends to higher photon energies, and all harmonics have comparable amplitude. We also note that for a compressed pulse rise time of 10 fs the ionization level in Ar is $\approx 40\%$. The 100 eV HHG emission therefore originates from neutral, rather than ionized, Ar. Finally, in Fig. 2 the relatively higher level of continuum emission at higher energies and lower pressures is consistent with HHG from a substantially shorter pulse.\(^12\)

The mechanism for the observed enhancement and spectral shift of the harmonic emission in Ar likely results from enhanced phase matching at high energies owing to QPM and guiding of a shorter laser pulse that evolves in the waveguide. In general, the phase mismatch that is present between the fundamental and the $q$th harmonic, $\Delta k = k_q - qk_1$, which is due to the waveguide, plasma, and neutral atoms, is given by\(^3\)

$$\Delta k = \frac{qu_{11}^2}{4\pi\alpha^2} + P \eta N_{atm} r_e(q\lambda - \lambda/q)$$

$$- \frac{2\pi(1 - \eta)Pq}{\lambda} \left[ \delta(\lambda) - \delta(\lambda/q) \right],$$

(1)

where $\lambda, q, a$, $u_{11}$, $\eta, P, N_{atm}, r_e$, and $\delta$ are the fundamental wavelength, the harmonic order, the waveguide radius, the first zero of Bessel function $J_0$, the ionization fraction, the gas pressure in Torr, the number density at 1 Torr, the classical electron radius, and the index of refraction of the neutral gas at 1 Torr, respectively. On the leading edge of an intense 27 fs pulse, at pressures of 25 Torr in Ar and at photon energies of 45 eV, the phase mismatch that is due to the neutral atoms and the waveguide (each $\approx \pm 2000$ m$^{-1}$) are balanced at low levels of ionization ($< 4\%$). However, at lower pressures of 5 Torr it is not possible to phase match at 45 eV because the contribution that is due to the waveguide ($\approx 4000$ m$^{-1}$ for
$q = 65$ at 100 eV) exceeds that which is due to the neutrals ($= +420 \text{ m}^{-1}$), even with no ionization present. For phase matching of harmonics near 100 eV at the peak of the pulse, and for ionization levels of $= 50\%$, we calculate that $\Delta k = -15,000 \text{ m}^{-1}$. Therefore a large positive contribution to $\Delta k$ is needed for phase matching at 100 eV.

Such a contribution can be generated by the propagation of an intense laser beam within the guide, due to periodic refocusing of the laser.\textsuperscript{8} Simulations indicate that under the same conditions as those when pulse compression is observed, the laser beam periodically focuses with millimeter-scale periodicities. The resultant laser intensity modulations create intensity and phase modulations on the extreme-UV beam, leading to QPM of the frequency conversion process. QPM adds a positive contribution to Eq. (1) given by $K_{\text{QPM}}=2 \pi/\lambda$, where $\lambda$ is the period of the modulation. For a modulation period of 0.5 mm the contribution to the phase mismatch would be $12,600 \text{ m}^{-1}$. This term can be introduced either by physical modulation of the waveguide or by periodic focusing and defocusing in the plasma. The optimum periodicity will depend on harmonic order $q$; a periodicity that minimizes the phase mismatch at 95 eV will not phase match at 45 eV.

To further increase extreme-UV emission in the technologically significant energy region near 93 eV, we also implemented QPM in engineered modulated waveguides. Figure 3 shows harmonic emission from Ar filling of straight and modulated waveguides (diameter, 150 $\mu$m; periodicities, 0.5 and 0.25 mm) at a pressure of 5 Torr and for an input laser energy of 2.5 mJ. Three Zr filters with 0.6 $\mu$m total thickness were used to reject the laser light. The modulated waveguides enhance the harmonic emission by compensating for ionization-induced phase mismatch, contributing phase mismatches of $= 12,500 \text{ m}^{-1}$ (0.5 mm periodicities) and $= 25,000 \text{ m}^{-1}$ (0.25 mm periodicities). It is evident that QPM with 0.25 mm periodicities overcompensates for the phase mismatch and that periodicities of 0.5 mm lead to stronger enhancement of the harmonic signal near 93 eV. At the maximum enhancements observed, the HHG emission near 100 eV is within a factor of 3 of that observed from Ar when it is optimally phase matched at 45 eV. Further improvements in flux should be possible by use of a two-stage fiber or a fiber with lower intrinsic losses. Preservation of high flux at lower pressures was also verified independently by use of a vacuum diode to view the entire HHG spectrum without the use of a filter or spectrometer, where the total current increased from 2 to 6 nA as the pressure was reduced.

In conclusion, we have demonstrated extension of the efficient region of high harmonic generation in Ar to 100 eV by combining phase matching, QPM, and pulse compression in a single gas-filled waveguide. The waveguide geometry allows Ar to be used for phase matching at high energies, for the first time to our knowledge, by taking advantage of either self-induced or externally induced intensity modulations to implement QPM.

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