Quasi-Phase-Matching of High Harmonic EUV Generation at Very High Ionization Levels

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Summary. We experimentally demonstrate enhanced conversion efficiency for high harmonic generation in fully ionized gas using quasi-phase-matching. We report the first observation of high harmonic generation in Argon up to 180 eV.

High-order harmonic generation (HHG) is a useful source of coherent light in the extreme ultraviolet to soft x-ray region of the spectrum. In HHG, an intense laser pulse is focused into a material, typically a gas. The extreme nonlinear interaction with the atoms results in generation of high-order harmonics of the fundamental laser that emerge as a coherent, low-divergence beam. The conversion efficiency, however, is limited by the difficulty of phase-matching the process, particularly at high ionization levels where the higher energy harmonics are generated.

One method to improve the conversion efficiency of HHG is to focus the fundamental light into a gas-filled hollow-core glass waveguide [1]. Phase-matching of the HHG conversion process can then be achieved by adjusting the gas pressure to balance the various contributions to the phase velocity of the light from the plasma, waveguide and neutral gas. Since these contributions are of opposite sign, phase-matching can be achieved and the harmonic signal increases quadratically over the interaction length, neglecting absorption. However, this method of phase-matching only works for low levels of ionization (\(~5\%\)). At higher ionization levels, the plasma contribution becomes much greater than the neutral gas contribution - making phase-matching impossible. When the phase-velocities are not equal, the harmonic emission builds up periodically over a coherence length, and then interferes with out-of-phase light generated later along the length of the interaction. Since the highest harmonic orders are created at the peak of the pulse where the ionization level is greatest, this effectively limits the highest EUV photon energy that can be generated efficiently.

In recent work [2, 3], we demonstrated that by modulating the diameter of the waveguide, we could quasi-phase-match (QPM) the EUV conversion process up to ionization levels of \(~10\%\). The effect of the fiber modulations is to periodically change the driving laser intensity along the interaction length. Because the EUV phase is dependent on the driving laser intensity, the fiber modulations can both adjust the phase of the EUV light, and also suppress...
the EUV generation in certain regions of the fiber. This periodic dependence of the strength and phase of the nonlinearity enables QPM. In our past work, a large enhancement of the highest order harmonics was observed in Helium and Neon, corresponding to compensating for ionization levels below 10%. In this work, we show it is possible to quasi-phase-match HHG at dramatically high ionization levels of 100% in Argon and 35% in Neon. This work overcomes a critical limitation for extending HHG to higher energies where ionization destroys the build-up of signal. As a result of QPM, we enhance the harmonic emission between 160 - 180eV in Neon and Argon, which are the highest harmonics to be phase matched to date.

In this experiment, we focus light from a 1kHz Ti:sapphire laser system producing 23fs duration pulses with a typical pulse energy of 1.1mJ into either a straight or modulated 150μm diameter waveguide filled with Argon or Neon. The modulated fiber we use is shown in Figure 1. The modulations of the fiber inner diameter are approximately sinusoidal with a depth of ~ 10% and a period of 0.25mm and the total fiber length is 2.5cm. The harmonic emission from the fiber passes through 0.4μm of Zirconium to block the fundamental light, and is spectrally dispersed by a Hetrick grazing-incidence EUV spectrometer and focused onto an Andor CCD camera. The HHG spectra are calibrated by recording the positions on the CCD of the Silicon and Boron absorption edges, at 99.9eV and 188.35eV respectively, to obtain the linear conversion from CCD position to wavelength.

Figure 2 shows the harmonic emission from both a straight and a modulated waveguide filled with Neon. Both spectra are taken with the same driving pulse characteristics, gas pressure, fiber coupling efficiency, and CCD exposure time. The harmonics generated in the straight waveguide only extend to ~ 135eV. For the higher harmonic orders, ionization creates a large a phase mis-match which greatly reduces the signal. In contrast, the harmonics from the modulated waveguide extend to ~ 170eV. Note that for the higher harmonic orders, the modulated waveguide increases the signal by orders of magnitude over the straight fiber.

Figure 3 shows HHG emission in Argon from straight and modulated waveguides at low pressures. The modulated waveguide increases the HHG

![Fig. 1. Optical microscope image of a 0.25mm period modulated waveguide.](image)