

# All-Optical Quasi-Phase Matching in Extreme Nonlinear Optics

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In regimes where conventional phase matching is not possible, quasi-phase matching (QPM) techniques permit significant conversion efficiency in parametric nonlinear processes. QPM is especially critical for extreme nonlinear optical processes, such as high harmonic generation (HHG), where phase matching is restricted to photon energies <130 eV. In HHG, near-IR light is coherently upconverted into the extreme ultraviolet (EUV) and soft X-ray regions of the spectrum.

However, during HHG, the medium is ionized, and the associated strong plasma dispersion precludes phase matching at high energies. Previous implementations of QPM to enhance HHG used hollow waveguides with a modulated diameter to modulate the intensity of the driving laser, perturbing the HHG process to implement QPM. This work enhanced the conversion efficiency by >10×, limited by variations in phase matching conditions within the medium.

We demonstrated that counter-propagating beams of light can be used to implement quasi-phase matching of high harmonic generation over a broad energy range from the EUV to > keV.<sup>1-4</sup> The presence of the counterpropagating field perturbs the phase of the radiating electron, which in this case is in the continuum. This perturbed phase is directly mapped onto the coherent X-ray phase, providing a means for implementing QPM.

We first used a single counterpropagating pulse to map *in situ* the coherence length for the HHG frequency conversion process.<sup>1</sup> By scanning the collision point between an intense forward-propagating laser pulse that generates the harmonics, and a weak counter-propagating pulse that modulates their phase, one can observe large modulations in the output. These oscillations correspond to the coherence length of

the HHG process, which varies over the length of the gas-filled hollow waveguide.

To implement QPM, one must use a light pattern that matches the coherence length oscillations. Using three counterpropagating pulses in sequence, we enhanced the flux of a single isolated harmonic by suppressing harmonic emission from consecutive out-of-phase zones.<sup>2</sup> Selective enhancement of >300× at photon energies around 65 eV (41<sup>st</sup> harmonic) was observed in argon in a regime that cannot be phase matched using conventional approaches. In the future, by adding more pulses, and optimization of their shape,<sup>3</sup> we should obtain even greater enhancement and spectral selectivity. This technique has also been extended to higher photon energies in helium, showing >100× enhancement at the 89<sup>th</sup> harmonic order (140 eV).

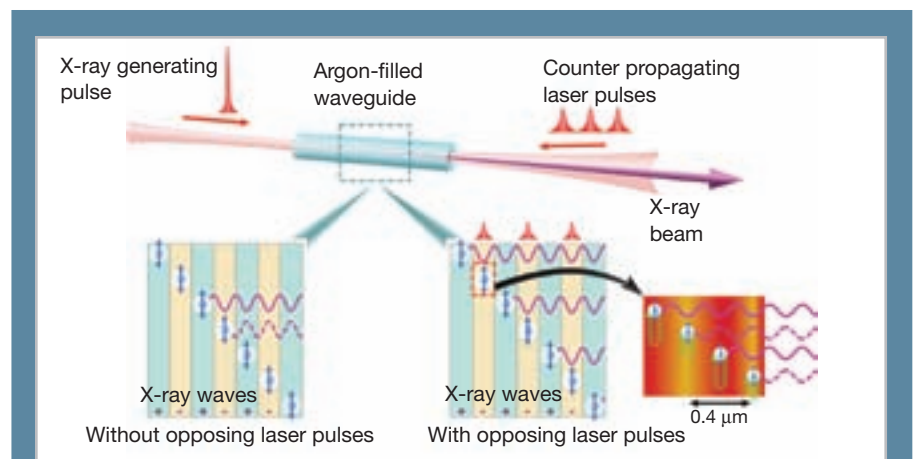
In theoretical work, we demonstrated that all-optical QPM can be extended to very high energies (keV) using a scheme that is intermediate between QPM and

true phase matching. Instead of using a sequence of pulses, one can use the oscillating field of a quasi-continuous infrared laser to adjust the phase of the emitted X-rays. This technique appears feasible even for the generation of coherent hard X-rays.<sup>4</sup> All-optical quasi-phase matching is promising for extending efficient coherent X-ray generation to high energies and for applications in atomic and molecular dynamics, nano- and bio-imaging, and materials science. ▲

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#### References

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Light patterns generated by a counterpropagating pulse-train in a hollow waveguide allow quasi-phase matching of high-order harmonic generation by manipulating the phase of the harmonic emission. Under conditions of poor phase matching (bottom left), blue and orange coherence zones destructively interfere due to the phase slip between the propagating harmonic and driving fundamental fields. When counterpropagating pulses overlap with the driving pulse in the orange regions (bottom right), the harmonic emission is suppressed by scrambling the electron and X-ray phase (inset), allowing emission from the blue regions to interfere constructively to generate a bright EUV output.