Generation of Bright Isolated Attosecond Soft X-Ray Pulses Driven by Multi-Cycle Mid-Infrared Lasers

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Abstract: By driving the high harmonic generation process with multi-cycle mid-infrared laser pulses, we demonstrate bright isolated, attosecond soft X-ray pulses for the first time.

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High harmonic generation (HHG) is the most extreme coherent nonlinear optical process in nature, making it possible to generate coherent beams that span from the EUV to > keV photon energies. The ability to generate ultrashort femtosecond-to-attosecond pulses in the EUV region of the spectrum has already resulted in new understanding of charge and spin dynamics in molecules and materials on the fastest timescales relevant to function. To date, most HHG work has been done using multi-cycle 0.8 μm driving lasers, where HHG generally consists of a train of attosecond pulses in the EUV. Most schemes for creating isolated attosecond pulses rely on driving the process using either few-cycle 0.8 μm pulses, or complex polarization modulation, or a combination of multicolor fields, or phase matching gating using ≈ 15 fs lasers. All these techniques are limited in pulse energy, and moreover, the resulting isolated attosecond pulse is extremely sensitive to the carrier envelope phase (CEP) of the driving laser pulse. Furthermore, all these approaches to date have been limited to the EUV region of the spectrum because using near-IR Ti:Sapphire lasers limits the bright phased-matched upconversion to <≈ 100 eV photon energies.

In this paper, we, for the first time, generate isolated soft X-ray attosecond pulses at photon energies up to 180 eV, which emerge as linearly chirped 300 *as* pulses with a transform limit pulse duration of 35 *as*. Moreover, when mid-infrared lasers are used to drive the high harmonic generation process, the conditions for optimal bright soft X-ray generation naturally coincide with the generation of isolated attosecond pulses that are also shorter in duration if compressed. Most surprisingly, advanced theory shows that long-duration, 10-cycle, driving laser pulses are required to generate isolated soft X-ray bursts efficiently, to mitigate group velocity walk-off between the laser and the X-ray fields that otherwise limit the conversion efficiency [1].

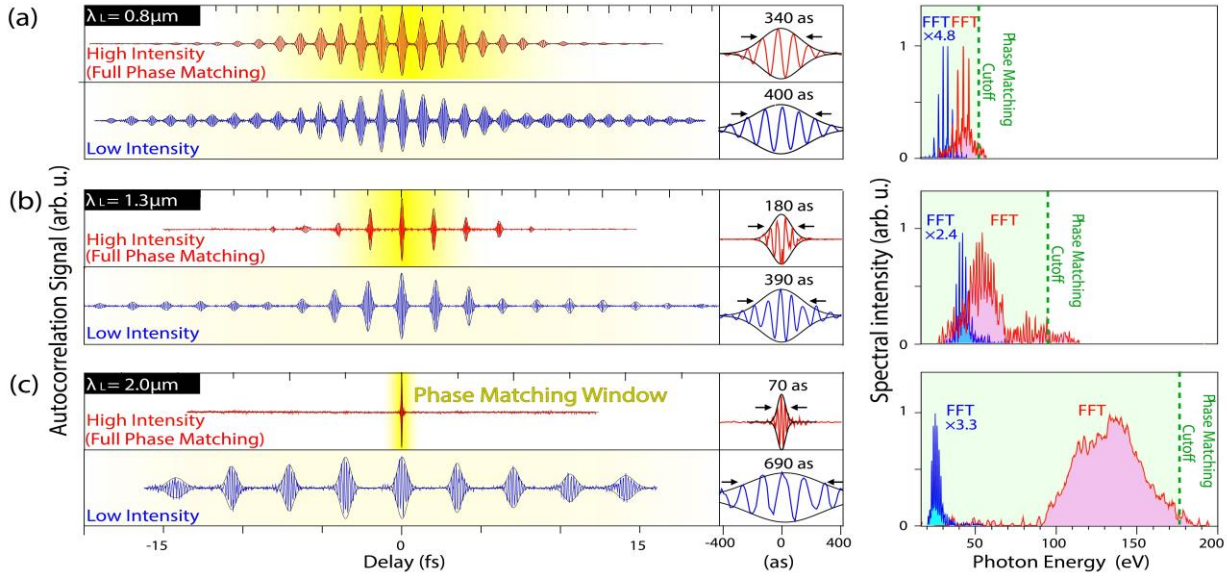

Fig 1: Comparison of experimental HHG autocorrelation data (normalized) from Ar driven by ~10 cycle laser pulses at (a) 0.8 μm, (b) 1.3 μm, and (c) 2 μm for high (fully phase matched) and low laser intensity conditions [red and blue]. Left: field autocorrelation of the HHG field. The phase matching temporal window is highlighted in yellow. Note that the bandwidth-limited pulse duration is half this coherence time. Right: HHG spectra obtained using the fast Fourier transform of the field autocorrelation traces (filled-area plots). The predicted phase-matching cutoffs are also shown (green) [2]. For longer laser wavelengths, the phase matching window is much narrower, leading to isolated attosecond pulses.

In our experiment, laser pulses at wavelengths of 0.8 μm, 1.3 μm, and 2.0 μm are generated using a 1 kHz Ti:Sapphire laser pumping a three-stage optical parametric amplifier (OPA) [3]. The pulse durations of all three wavelengths are adjusted to be ~ 10 cycles in duration. Each beam is focused into a 2 mm long, Ar-filled cell, with the backing pressure and focus position both varied to obtain optimal phase matching. The temporal structure of the generated HHG beam is measured through a soft X-ray field autocorrelator. A fast Fourier transform of the field autocorrelation trace, captured by an X-ray CCD, is performed to obtain the HHG spectra. The attosecond field autocorrelation is sufficient to fully characterize how the temporal gating of phase matching scales with driving laser wavelength by a direct measurement of the number of attosecond bursts contained in the HHG emission, i.e., a total of 2*n*-1 fringes will be measured if there are *n* bursts in the pulse train. Figure 1 shows that for all driving laser wavelengths, as the laser intensity is increased, the number of individual bursts in the attosecond pulse train decrease. The high laser intensity corresponds to optimal phase-matching at the highest photon energies possible in Ar, which generates the brightest flux. Remarkably for 2 μm lasers, when the optimal phase-matching intensity is used, and the HHG flux optimized, the emission corresponds to an isolated attosecond pulse with a central photon energy of 140 eV (spanning from 90 to 180 eV), and with a FWHM bandwidth of ≈ 60 eV, which is capable of supporting a ≈ 35 *as* transform-limited pulse. This pulse duration is corroborated by the measured 70 *as* field autocorrelation trace, which as expected, corresponds to twice the transform-limited pulse duration.

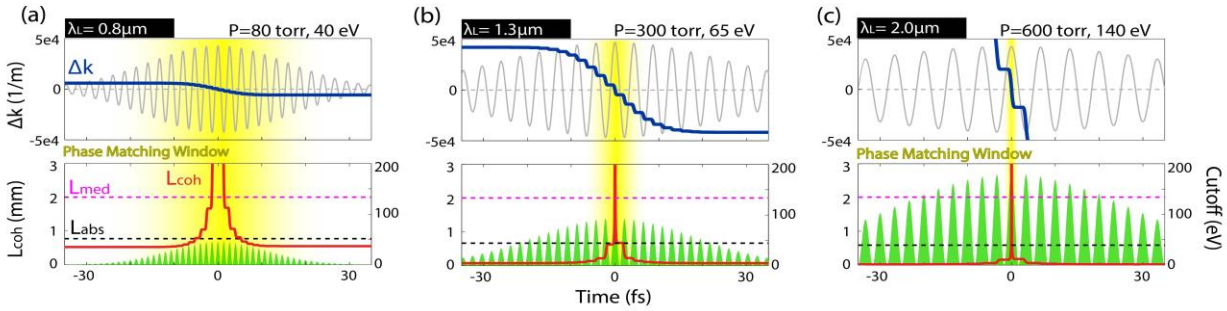

Fig 2: Calculated phase mismatch Δk [blue line], $L_{coh} = \pi/\Delta k$ [red line], both of which illustrate the shrinking of the phase matching temporal window [yellow highlight] at longer laser wavelengths. Also shown is the instantaneous HHG cutoff photon energy [green] for HHG in Ar driven by 10-cycle laser pulses. (a) 0.8 μm, 2.4x10^{14} W/cm^2; (b) 1.3 μm, 1.9x10^{14} W/cm^2; and (c) 2.0 μm, 1.5x10^{14} W/cm^2.

To understand why the phase-matching temporal window shrinks as the driving laser wavelength is increased, it is useful to look at the time derivative of the phase-mismatch [2]

$$\frac{\partial \Delta k(\tau)}{\partial \tau} \approx -P \cdot q \left[\delta n \cdot \frac{2\pi}{\lambda_L} + N_{atm} \cdot r_e \cdot \lambda_L\right] \cdot \frac{d\eta(\tau)}{d\tau} \propto P \cdot q \cdot \lambda_L \cdot \frac{d\eta(\tau)}{d\tau}, \qquad (1)$$

where Δk is the phase-mismatch, τ represents time measured in cycles of the driving laser, P is the gas pressure, q is the harmonic order, δn is the difference between the indices of refraction at the fundamental and harmonic wavelengths, λ_L is the central wavelength of the driving laser, N_{atm} is the number density of atoms at 1 atm, r_e is the classical electron radius, and $\eta(\tau)$ is the instantaneous ionization fraction. In the high intensity case, where optimal phase-matching conditions are obtained, the optimal phase-matching pressure, P_{PM}, and the central harmonic order, q_{PM}, scale by the driving laser wavelength as $P_{PM} \propto \lambda_L^2$, and $q_{PM} \propto 1/2 \cdot \lambda_L^{2.7}$, as have been observed experimentally. Thus, under optimal phase-matching conditions Eqn 1 can be approximated by: $\mathrm{dk}(\tau)/\mathrm{d}\tau \propto \lambda^{5.7} \cdot \mathrm{d}\eta(\tau)/\mathrm{d}\tau$. Figure 2 plots the phase mismatch and coherence length as a function of time during the laser pulse for different driving laser wavelengths, which were calculated using ADK ionization rates and optimal phase matching conditions. The temporal window during which phase matching occurs shrinks rapidly as the driving laser wavelength is increased. For 2 μm lasers, the phase-matching window is only one half-cycle long, thus leading to an isolated attosecond pulse, in excellent agreement with the data of Figure 1. Furthermore, group velocity walk-off becomes important at mid-IR laser wavelengths since the envelope of the pulse starts to reshape the electric field in the phase matching window. This effect can be mitigated by using multi-cycle driving lasers where the change in the electric field strength from cycle to cycle is negligible.

In summary, under optimal phase-matching conditions, long wavelength driving lasers naturally produce bright isolated attosecond soft x-ray pulses of shorter transform-limited duration, and do not require any complex pulse compression or multi-color driving lasers. This work demonstrates the first isolated attosecond pulses in the soft X-ray regime and readily provides an accessible and reliable route for broad application of attosecond science [4].

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