Bright Coherent Ultrahigh Harmonics in the keV X-ray Regime from Mid-Infrared Femtosecond Lasers

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High-harmonic generation (HHG) traditionally combines ~100 near-infrared laser photons to generate bright, phase-matched, extreme ultraviolet beams when the emission from many atoms adds constructively. Here, we show that by guiding a mid-infrared femtosecond laser in a high-pressure gas, ultrahigh harmonics can be generated, up to orders greater than 5000, that emerge as a bright supercontinuum that spans the entire electromagnetic spectrum from the ultraviolet to more than 1.6 kilo-electron volts, allowing, in principle, the generation of pulses as short as 2.5 attoseconds. The multiatmosphere gas pressures required for bright, phase-matched emission also support laser beam self-confinement, further enhancing the x-ray yield. Finally, the x-ray beam exhibits high spatial coherence, even though at high gas density the recolliding electrons responsible for HHG encounter other atoms during the emission process.

The unique ability of x-rays to capture structure and dynamics at the nanoscale has spurred the development of large-scale x-ray free-electron lasers based on accelerator physics, as well as high-harmonic generation (HHG) techniques in the x-ray region that employ tabletop femtosecond lasers. The HHG process represents nonlinear optics at an extreme, enabling femtosecond-to-attosecond duration pulses with full spatial coherence (1-6), which make it possible to track the dynamics of electrons in atoms, molecules, and materials (7-12). X-rays can probe the oxidation or spin state in molecules and materials with element specificity, because the position of the characteristic x-ray absorption edges of individual elements is sensitive to the local environment and structure. Ultrashort x-ray pulses can capture the coupled motions of charges, spins, atoms, and phonons by monitoring changes in absorption or reflection that occur near these edges as a material or molecule changes state or shape. However, many inner-shell absorption edges in advanced correlated-electron, magnetic, and catalytic materials (Fe, Co, Ni, Cu) lie at photon energies nearing 1 kilo–electron volt (keV) (*13–15*). In contrast, most applications that use HHG light have been limited to the extreme ultraviolet (EUV) region of the spectrum (<150 eV), where efficient frequency upconversion is possible with the use of widely available Ti:sapphire lasers operating at a 0.8-µm wavelength. We therefore sought to extend bright HHG to a higher-energy soft x-ray region.

High-harmonic generation is a universal response of atoms and molecules in strong femtosecond laser fields (16, 17). In a simple analogy, HHG represents the coherent version of the Röntgen x-ray tube: Instead of boiling electrons off a hot filament, accelerating them in an electric field, and generating incoherent x-rays when the highenergy electrons strike a target, HHG begins with tunnel ionization of an atom in a strong laser field. The portion of the electron wave function that escapes the atom is accelerated by the laser electric field and, when driven back to its parent ion by the laser, can coherently convert its kinetic energy into a high-harmonic photon. The highestenergy HHG photon emitted is given by the microscopic single-atom cutoff rule: $hv_{SA cutoff} =$ $I_{\rm p}$ + 3.17 $U_{\rm p}$, where h is Planck's constant, v is the frequency, I_p is the ionization potential of the gas, and $U_p \stackrel{P}{\simeq} I_L \lambda_L^2$ is the quiver energy of the liberated electron in a laser field of intensity $I_{\rm I}$ and wavelength $\lambda_{\rm I}$.

Generating bright, fully coherent HHG beams requires macroscopic phase matching (18), wherein

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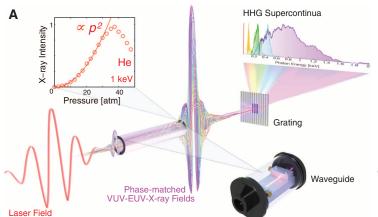
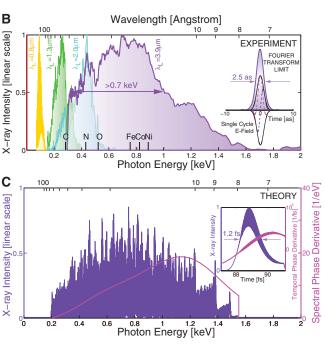


Fig. 1. (**A**) Schematic illustration of the coherent kilo–electron volt x-ray supercontinua emitted when a mid-IR laser pulse is focused into a high-pressure gas-filled waveguide. The experimental phase-matched harmonic signal grows quadratically with pressure, demonstrating excellent phase-matched coherent buildup with increasing pressure *p*. (**B**) Experimental HHG spectra emitted under full phasematching conditions as a function of driving-laser wavelength (yellow, 0.8 µm; green, 1.3 µm; blue, 2 µm; purple, 3.9 µm). (Inset) Fourier transform–limited pulse duration of 2.5 as. (**C**) Calculated spectrum and temporal structure of one of



the phase-matched HHG bursts driven by a six-cycle FWHM 3.9- μ m pulse at a laser intensity of $I_L = 3.3 \times 10^{14}$ W/cm².

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the laser and high-order nonlinear polarization propagate in phase (at the speed of light) throughout a medium to ensure that the HHG light emitted from many atoms adds coherently (1, 19, 20). Phase matching is achieved by balancing the neutral gas and free-electron plasma dispersion experienced by the laser and is only possible up to some critical ionization level that depends on the gas species and laser wavelength (fig. S1). Any geometric contributions to the laser propagation must also be considered (see supplementary text). Because ionization increases with laser intensity, the critical ionization limits the highest photon energy for which phase matching can be implemented. Recent work explored the wavelength dependence of the HHG yield (21-24), which scales as $hv_{\rm PM\ cutoff} \propto \lambda_{\rm L}^{1.7}$ under phase-matched conditions (25-27). Using 2-µm lasers (0.62-eV photons) to drive HHG, bright harmonics extend to >0.5 keV (26), demonstrating phase matching of a >800-order nonlinear process (note that only odd-order harmonics are emitted to conserve angular momentum).

In this work, bright high-harmonic x-ray supercontinua with photon energies spanning from the EUV to 1.6 keV (<7.7 Å) are generated by focusing 3.9-µm wavelength pulses from a tabletop femtosecond laser into a waveguide filled with He gas (see Fig. 1). This represents an extreme >5000-order nonlinear process while also demonstrating fully phase-matched frequency upconversion. The multiatmosphere pressures necessary for efficient x-ray generation also support laser beam self-confinement, enhancing the x-ray yield by another order of magnitude. We observe coherent, laserlike x-ray beams, despite the fact that ultrahigh-harmonic generation occurs in a regime where the laser-driven electrons encounter many neighboring atoms before they re-encounter their parent ions. Our calculations indicate that the kilo-electron volt-bandwidth coherent supercontinuum has a well-behaved chirp that, when compensated, could support a single-x-ray-cycle 2.5-attosecond pulse duration. Finally, we show that in the kilo-electron volt region, a much higher-order nonlinear process is required for phase matching than is required for harmonic emission from a single atom.

In our experiment, six-cycle full width at half maximum (FWHM) (80-fs) 10-mJ pulses, centered at a wavelength of 3.9 µm, are generated at 20 Hz as the idler output of an optical parametric chirped-pulse amplification laser system (28, 29). X-rays are generated by focusing the laser beam into a 200-µm diameter, 5-cm-long, gas-filled hollow waveguide capable of sustaining pressures of up to 80 atm in a differentially pumped geometry. The HHG spectrum is then captured with the use of a soft x-ray spectrometer and x-ray chargecoupled device camera. Figure 1B shows the phase-matched HHG emission from He, which extends to >1.6 keV (<7.7 Å). The phase-matched HHG cutoff energy agrees well with numerical predictions plotted in Fig. 2A for 3.9-µm driving lasers; that is, $hv_{PM \text{ cutoff}} \propto \lambda_L^{1.7}$ (25–27). This bright x-ray supercontinuum is ideal for x-ray spectroscopy measurements, spanning multiple inner-shell absorption edges simultaneously (Fig. 1B and fig. S2), as has already been demonstrated in the EUV region for HHG driven by multicycle 0.8-µm lasers where a quasi-continuous HHG spectrum is emitted (*15*, *30*, *31*).

The x-ray flux from He scales quadratically with pressure (number of emitters), as shown in Fig. 1A, reaching a maximum at very high gas pressures of ~35 atm, where both phase matching and laser beam self-confinement are optimized. At higher pressures, the x-ray flux decreases due to reabsorption of the generated harmonics by the high-pressure gas, as well as energy loss experienced by the laser when coupling into the waveguide. Microscopically, quantum diffusion leads to spreading of the electron wave packet, decreasing the recombination probability and, thus, the single-atom HHG yield (22–24), which scales with the laser wavelength as $\sim \lambda_L^{-6.5}$ under phase-

matched illumination. Specifically, the single-atom HHG yield is $\sim 3 \times 10^5$ smaller at 3.9 µm compared with 0.8 µm. Fortunately, the low singleatom yield can be compensated by coherently combining HHG from a large number of emitters (high gas density and medium length), which is possible in part because the gas becomes increasingly transparent at photon energies approaching the kilo-electron volt region. The HHG signal builds up over a density-length product comparable to the absorption depth of the x-ray light, leading to nearly constant brightness of the HHG emission from 0.3 to 1 keV. An approximate brightness of 10⁵ photons per shot (corresponding to 10⁶ photons/s at 20 Hz) is observed in a fractional bandwidth of 1% at 1 keV. Past work successfully made use of 0.8-µm lasers to demonstrate kilo-electron volt harmonics with a~1000-order nonlinear process but with much reduced flux (four to five orders of magnitude lower), because phase matching is not possible in

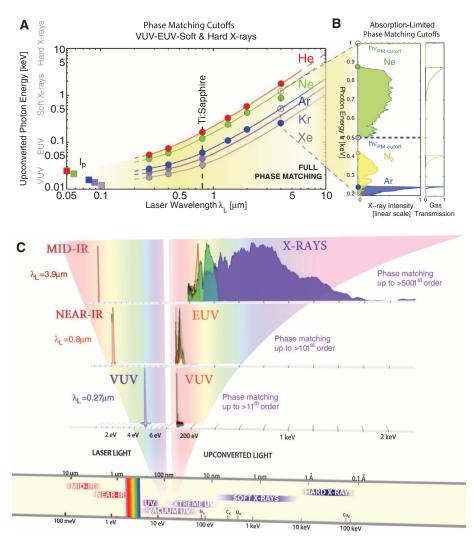


Fig. 2. (**A**) Predicted and observed HHG phase-matching cutoffs as a function of laser wavelength from the UV to the mid-IR. Solid circles show the observed cutoffs; open circles show the predicted cutoffs for Ar and Ne [which cannot be reached due to inner-shell absorption, as shown in (**B**)]. Solid squares on the left show the ionization potentials ($_{p}$) of the different atoms. (**C**) Unified picture of optimal phase-matched high-harmonic upconversion, including microscopic and macroscopic effects.

this regime (14). Thus, surprisingly, for macroscopic phase matching, the required harmonic order of >5000 is much higher than that required to generate the same photon energy from a single atom using a shorter laser wavelength.

We can now present a unified picture for phasematched high-harmonic upconversion, spanning the electromagnetic spectrum from the vacuum ultraviolet (VUV) to greater than kilo-electron volt x-ray photon energies, that includes both the microscopic and macroscopic physics. To validate theory, we tuned our driving laser to different wavelengths from the UV to the mid-infrared (mid-IR) and then implemented pressure-tuned phase matching to optimize the HHG flux at each laser wavelength with the optimal laser intensities dictated by the critical ionization of the medium (Fig. 2 and supplementary text). The required optimal pressures and interaction lengths evolve from <0.1 atm and a few millimeters in the VUV region to tens of atmospheres and multicentimeter lengths in the x-ray region. Figure 2, A and C, shows the optimized phase-matched cutoffs and spectra for different driving-laser wavelengths. To efficiently generate high harmonics, the order of the nonlinearity must increase from ~11 in the VUV to >5000 in the kilo-electron volt region. This represents an extreme for both the order of a nonlinear process and phase matching. The bright phase-matched HHG spectra evolve from a single harmonic in the VUV into a broad x-ray supercontinuum spanning thousands of harmonics in the soft x-ray region. Phase matching shuts off in the VUV at energies near the ionization potential of the nonlinear gas medium, as the HHG and driving-laser wavelengths converge (see Fig. 2A). The phase-matched HHG conversion efficiencies reach 10^{-3} to 10^{-4} in the VUV region, compared

with 10^{-5} in the EUV using 0.8-µm lasers, and 10^{-6} to 10^{-7} in the x-ray region. Moreover, in the VUV region, phase matching occurs at relatively high levels of ionization of tens of percent (fig. S1).

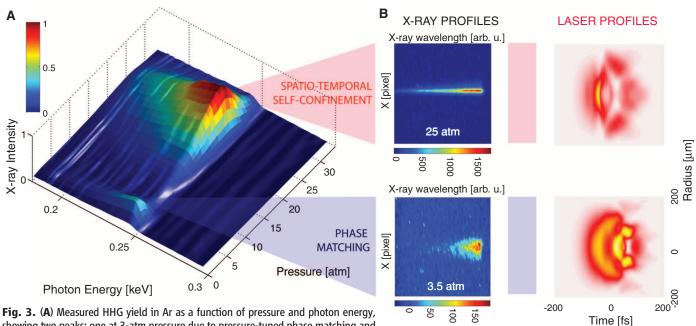
Remarkably, tunnel ionization of the atomic gas medium dominates in all phase-matching regimes. When driven by UV light, the effective potential (which is a superposition of the Coulomb and laser fields) oscillates rapidly, allowing a very short time interval for the electron to tunnel. However, the required laser intensity for HHG is extremely high (>10¹⁵ W/cm²), so tunnel ionization is more probable than multiphoton ionization. For mid-IR laser wavelengths, the slowly oscillating effective Coulomb potential can be considered quasi-static. Therefore, although the laser intensity decreases to maintain phase matching, tunnel ionization is still more probable than multiphoton ionization. Because the physics of ionization does not change, we can use an analytical description of tunneling [the Amossov-Delone-Krainov model (32)] to derive a generalized analytic HHG phase matching cutoff rule (Eq. 1), validated by comparison with experiment, as well as numerical and quantum theory (see supplementary text)

$$h\nu_{\rm PM\,cutoff} = I_{\rm p} + \frac{\alpha I_{\rm p}^3}{\ln^2 \left\{ \frac{\beta I_{\rm p}\tau_{\rm L}}{-\ln[1 - \tau_{\rm CR}(\lambda_{\rm L})]} \right\}} \lambda_{\rm L}^2 \quad (1)$$

Here, α and β are constants that depend on the laser pulse shape and the state from which the electron is tunnel ionized, τ_L is the laser pulse duration, and η_{CR} is the critical ionization. This analytical expression gives some physical insight into phase matching of the HHG upconversion process. The small deviation of $\lambda_L^{(\bar{1}.5-1.7)}$ from the λ_L^2 scaling of the ponderomotive energy incorporates the proper scaling of the laser intensity and arises from the scaling of η_{CR} , which decreases by four orders of magnitude from the UV to mid-IR driving-laser wavelengths. Short, few-cycle laser pulses make it possible to generate higher-energy photons before the critical ionization level is exceeded. However, this approach yields diminishing returns for pulses shorter than 5 to 10 cycles and leads to only modest enhancements in HHG flux and phasematching cutoff. The most substantial HHG enhancement (by orders of magnitude) arises when the right combination of laser wavelength, gas pressure-length product, and laser intensity is used.

Likewise, in contrast to conventional wisdom (see supplementary text), helium is generally the best atomic medium for harmonic generation due to the absence of inner-shell absorption (25). The absorption limit for HHG emission can be clearly seen in Fig. 2B, which shows a plot of the phasematched HHG emission from Ar, N2, and Ne when driven by 3.9-µm light. There is a sharp drop-off in signal at the inner-shell absorption edges at 0.25 (Ar), 0.41 (N₂), and 0.87 keV (Ne); therefore, the true phase-matching cutoff cannot be observed: Without absorption, the phasematching limits would be ~ 0.5 (Ar, N₂) and ~1 keV (Ne).

Generating bright kilo-electron volt harmonics from atoms driven by mid-IR femtosecond lasers takes advantage of a noteworthy convergence of favorable physics. First, the very high gas density required puts these experiments in a regime of HHG from nonisolated emitters: Spread of the ionized electron quantum wave packet over its few-femtosecond free trajectory means that the electron will encounter many neighboring atoms.



showing two peaks: one at 3-atm pressure due to pressure-tuned phase matching and

a second at 26-atm pressure due to the additional presence of laser-beam spatio-temporal self-confinement. (B) Experimental HHG beam profiles and calculated laser-beam profiles after a propagation distance of 3.8 cm in the waveguide. arb. u., arbitrary units.

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This contrasts with emission from dilute, isolated atoms for UV or EUV harmonic generation. As shown in fig. S1, for kilo-electron volt harmonics, the electron wave function in the continuum extends to ~500 Å, whereas the separation between the He atoms is ~15 Å at 10-atm pressure. However, the ionization levels are low at $\sim 0.03\%$. For VUV/EUV harmonics, the electron typically extends ~2 to 20 Å between ionization and recollision, whereas the separation between atoms is ~70 Å at ~0.1 atm pressure, and phase matching occurs at ~10% ionization levels. Thus, HHG driven by mid-IR pulses liberates 0.001 as much of the electron wave function into the continuum compared with visible driving lasers, though it is spread over a 100-times-larger distance. Fortunately, our experimental results indicate that rescattering of this large and diffuse recolliding electron wave packet from other atoms seems not to adversely influence the coherence of the emission, likely because the medium is weakly ionized. Evidence for this includes the well-formed, spatially coherent x-ray beams (Fig. 3B and Fig. 4) and the remarkable quadratic growth (Fig. 1A) that continues from 0.2 atm (when the rescattering electron wave packet can begin to encounter neighboring atoms) to more than two-orders-ofmagnitude-higher pressure.

In a second extremely favorable convergence of extreme nonlinear optics, the multiatmosphere gas pressures required for phase-matched x-ray generation also overlap with the parameter range where laser beam self-confinement is possible. Figure 3A shows a plot of the experimental x-ray emission from Ar driven by 3.9-µm lasers. The predicted phase-matching pressure is ~3 atm, and we indeed observe a peak in x-ray emission at that pressure. However, as the pressure is further increased, the x-ray yield first decreases and then increases quadratically, exhibiting a large enhancement at a pressure of 26 atm (about a factor of 10 when integrated over all soft x-ray HHG). The measured x-ray beam profile also dramatically narrows as the gas pressure increases (Fig. 3B), indicative of self-confinement of the driving laser. Essentially, the x-ray HHG beam, imaged at the exit of the fiber, shrinks to less than one-third of its former diameter, whereas the x-ray signal increases tenfold (integrated over all orders) at pressures seven times greater than those required for phase matching.

To explore theoretically how macroscopic nonlinear effects augment HHG phase matching, we numerically simulated nonlinear pulse propagation in a hollow waveguide filled with high-pressure gas by extending and expanding previous simulations to longer wavelengths (33, 34). Our simulations show that as the gas pressure increases beyond that required for phase matching, the peak laser intensity is stabilized (figs. S3 and S4). We also observe strong spatio-temporal compression and localization of the driving laser during self-confinement due to the Kerr effect and plasma generation, which also enhances the HHG yield. Figure 3B plots the calculated beam profiles at the phase-matching (3.5 atm) and higher pressures (26 atm). A stable self-confined beam forms at the higher gas pressures and persists for centimeter distances. As discussed in the supplementary text, we can experimentally and theoretically observe that self-confinement also enhances phase matching in other gases, such as He (fig. S4) and molecular N₂.

When phase matched, the spatial quality of the x-ray beam is excellent. Figure 4 shows the x-ray beam and the Young's double-slit diffraction patterns taken by illuminating 5-µm slits (separated by 10 µm) with an x-ray supercontinuum generated in He and Ne, spanning 7.7 to 43 Å and 14 to 43 Å, respectively. There is excellent agreement between the experimentally observed and theoretically predicted diffraction patterns. A plot of the expected diffraction pattern from incoherent x-ray illumination is also shown in Fig. 4, B and C, for the same experimental geometry, proving that the high fringe visibility is not due to the small pinhole size but rather to the high spatial coherence of the x-ray beam itself. This measurement is extremely challenging at short wavelengths: Very small slit widths are required so that the light from each slit diffracts sufficiently to ensure overlap and interference at the detector (3.5 m away from the slits). Thus, the throughput is very small. This spatial coherence measurement clearly demonstrates that coherent diffractive imaging will be possible with near wavelength spatial resolution, as has been achieved using HHG beams and synchrotron sources in the EUV and soft x-ray regions (35, 36).

To predict the temporal properties of the HHG radiation, we theoretically analyzed HHG driven by one- and six-cycle FWHM mid-IR laser pulses, with peak intensities of 4.1 and 3.3×10^{14} W/cm², respectively, from single atoms and also in a phase-matched regime. Our calculations, based on the strong field approximation and discrete dipole approach (*37*), confirm the femtosecond time scale of the x-ray bursts from a single atom and also after propagation (see supplementary text). Our calculated phase-matched HHG spectra agree well with those measured experimentally (Fig. 1, B and C) and show that the HHG chirp is well behaved (Fig. 1C and fig. S5) over the near—

kilo-electron volt bandwidth that, when compressed, is sufficient to support a single-cycle, 2.5-as pulse in the Fourier limit. For 3.9-µm driving lasers in the single-atom case, contributions from the short and long trajectories lead to a parabolic chirp, whereas after propagation, the phasematched short trajectory contribution leads to a positive, quasi-linear chirp. The current limit of theory allows us to simulate HHG propagation over 20-µm distances at high pressures and predicts that the uncompressed HHG temporal emission consists of a series of ~three intense bursts of 1- to 3-fs duration, due to the very long 13-fs period of the multicycle 3.9-µm driving laser field (Fig. 1C and fig. S5). However, for longer propagation distances, bright HHG emission in the form of a single isolated x-ray burst is expected. This is because phase matching is transient and favors x-ray emission from a single halfcycle of the laser pulse where the phase matching is optimal. This has been verified experimentally in the EUV, even without stabilizing the carrier wave with respect to the pulse envelope (25, 38). Interestingly, this work and past work predict that the HHG bursts are chirped, where the amount of chirp scales inversely with laser wavelength for a given spectral bandwidth (22). However, as shown in Fig. 1, the duration of each HHG burst still spans femtosecond durations (for example, 1000 times longer than their transform limit) due to the increased phase-matched HHG bandwidth, which scales almost as the square of the laser wavelength.

Experimental verification of these predictions will require the development of characterization methods that can sample ultrabroad bandwidth x-ray waveforms at different photon energies. This challenge is illustrated in Fig. 1B, where the narrow dip at 0.54 keV corresponds to oxygen K-edge absorption. It is not clear that any atomic or molecular system can interact with a kilo– electron volt bandwidth, because processes such as photoionization involve significantly slower time scales. However, the chirped x-ray supercontinua already represent a promising multiple– atomic site probe with subfemtosecond time resolution, analogous to the chirped white light (visible) continua used to probe many absorption

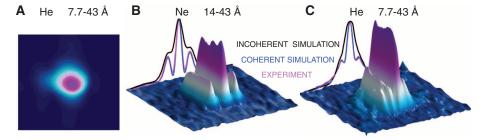


Fig. 4. (**A**) X-ray experimental beam profile. (**B** and **C**) Young's double-slit diffraction patterns taken by illuminating 5- μ m slits, separated by 10 μ m, with the beam shown in (A). There is excellent agreement between the experimentally observed (purple line) and theoretically predicted (blue line) diffraction patterns. The broad bandwidth and very low divergence of the HHG beams limit the number of fringes observed. The expected diffraction, assuming incoherent illumination, is also given for comparison (black line), illustrating the high spatial coherence of the kilo—electron volt HHG source.

features simultaneously, perfectly synchronized to the driving laser. Given our current experimental and theoretical findings, it may be possible to extend HHG to hard x-ray wavelengths and broader zeptosecond bandwidths.

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Supplementary Materials www.sciencemag.org/cgi/content/full/336/6086/1287/DC1 Materials and Methods Supplementary Text Figs. S1 to S5 References (39–46) 28 December 2011; accepted 12 April 2012

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The Heliosphere's Interstellar Interaction: No Bow Shock

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As the Sun moves through the local interstellar medium, its supersonic, ionized solar wind carves out a cavity called the heliosphere. Recent observations from the Interstellar Boundary Explorer (IBEX) spacecraft show that the relative motion of the Sun with respect to the interstellar medium is slower and in a somewhat different direction than previously thought. Here, we provide combined consensus values for this velocity vector and show that they have important implications for the global interstellar interaction. In particular, the velocity is almost certainly slower than the fast magnetosonic speed, with no bow shock forming ahead of the heliosphere, as was widely expected in the past.

The ionized solar wind flows continuously outward at speeds of ~300 to 800 km s⁻¹, incorporating interstellar neutral atoms that flow into the heliosphere and are ionized to become pickup ions (PUIs). Because the solar wind and surrounding local interstellar medium (LISM) are both magnetized plasmas and cannot penetrate each other, the solar wind inflates a bubble in the LISM called the heliosphere. Inside its boundary, the heliopause, there is a ter-

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mination shock (TS), where the solar wind and PUIs are compressed and heated. Because the heliosphere moves with respect to the LISM, the dynamic pressure plays an important role in shaping the heliosphere, with a compressed "nose" on the upwind side and a downwind "tail" (*I*). Since Parker's original work (1), there have been numerous theoretical enhancements, including the addition of an upstream bow shock (BS) (2) that was debated early on (3) but is now widely accepted [for example, see (4-7) and references therein].

NASA's Interstellar Boundary Explorer (IBEX) (8) measures neutral atoms, which move freely across magnetic fields; some of these atoms penetrate from the LISM to 1 AU (astronomical unit: Sun-to-Earth distance), where IBEX detects them. IBEX was primarily designed to measure energetic neutral atoms (ENAs) generated by charge exchange between the solar wind and PUIs (4-7) with interstellar neutrals. These observations led to the detection of an enhanced "ribbon" of ENA emissions nearly encircling the heliosphere, apparently ordered by the external LISM magnetic field and not predicted by any prior model or theory (9-11).

The IBEX-Lo instrument (12) was also designed to measure the neutral interstellar gas

Table 1. Interstellar flow parameters in ecliptic (J2000) and galactic coordinates.

Parameter	Value and 1σ uncertainty	Bounding range along Eqs. 1 to 3
Speed (V _{ISM∞})	23.2 \pm 0.3 km s^{-1}	21.3 km s ⁻¹ , 82.0°, -4.84°, 5000 K
Ecliptic longitude (λ _{ISM∞})	$\textbf{79.00^\circ} \pm \textbf{0.47^\circ}$	to
Ecliptic latitude (β _{ISM∞})	$-4.98^\circ \pm 0.21^\circ$	25.7 km s ⁻¹ , 75.5°, –5.14°, 8300 K
Interstellar He temp. ($T_{\rm He\infty}$)	$6300~\pm~390~K$	
Speed (V _{ISM∞})	23.2 \pm 0.3 km s $^{-1}$	21.3 km s ⁻¹ , 186.62°, -9.36°, 5000 K
Galactic longitude (l _{ISM∞})	$185.25^\circ\pm0.24^\circ$	to
Galactic latitude ($b_{ISM\infty}$)	-12.03° ± 0.51°	25.7 km s ⁻¹ , 183.77°, -15.22°, 8300 K
Interstellar He temp. ($T_{\rm He\infty}$)	$6300~\pm~390~K$	

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Supplementary Materials for

Bright Coherent Ultrahigh Harmonics in the keV X-ray Regime from Mid-Infrared Femtosecond Lasers

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This PDF file includes:

Methods Supplementary Text Figs. S1 to S5 Full Reference List In this supporting text, we first provide additional details about the driving laser used to generate bright keV harmonics. Next, we expand on the discussion in the main text to highlight the differences between high harmonic generation in the EUV and keV regions of the spectrum. In the final sections, we explain our calculations of 1) mid-infrared laser propagation and self-confinement; 2) the high harmonic pulse duration and phase; and 3) the analytic formula for the scaling of phase matching in the VUV and x-ray regions of the spectrum.

Femtosecond laser drivers at 3.9 µm, 0.4 µm and 0.27 µm wavelengths

The mid-IR OPCPA laser system is based on a unique femtosecond Yb:CaF₂ chirped pulse amplifier which drives a cascaded optical parametric amplifier *(28)*. The subsequent stages of the OPCPA (based on KTA crystals) are pumped by a 20 Hz ps Nd:YAG laser system and produce record 35 mJ (uncompressed) and 10 mJ (compressed) pulse energies in the signal and the idler beams at 1.46 μ m and 3.9 μ m respectively. The 0.27 μ m and 0.4 μ m laser pulses were generated from the 3rd and 2nd harmonics of a high energy, 30 mJ, 25 fs, Ti:Sapphire laser system operating at a repetition rate of 10 Hz. To optimize phase matched conversion into the VUV, pulse energies and durations of 4.5 mJ and 45 fs (3 ω), and 8 mJ and 35 fs (2 ω), were used.

Bright keV ultrahigh harmonic generation

The very high (multi atm) gas pressures required for optimum phase matching of high harmonic generation (HHG) at keV photon energies makes a differentially-pumped waveguide geometry the ideal target geometry (1). Good differential pumping is required to avoid re-absorption of the generated x-rays by the gas, defocusing of the laser prior to entering the waveguide, and destructive interference in non-optimal pressure regions. The waveguide provides for minimum (through still substantial) gas load, since the gas flow and laser propagation apertures fully overlap. The waveguide designs incorporate a 1-4 cm constant-pressure section with 0.5 cm end sections for differential pumping; the shorter fiber lengths are used for the VUV HHG where the reabsorption of the upconverted light is stronger. As discussed in the main text, the very high gas densities required for phase matching of the HHG process in the keV region necessitates a new regime of HHG from *non-isolated* emitters in a gas; extreme quantum diffusion over several fs means that the electron wavefunction will encounter many neighboring atoms, ions, or electron wavepackets in the continuum, as illustrated in Fig. S1.

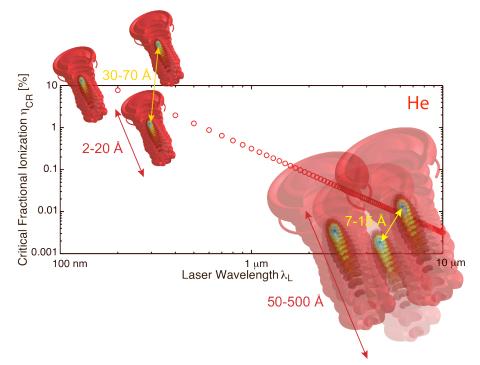


Fig. S1. The critical ionization fraction of He, above which phase matching is not possible, as a function of laser wavelength. Also shown are representations of the continuum electron wavepackets in the UV and keV regions.

Although not widely recognized previously, low-Z atoms (i.e. He) are nearly always optimum for HHG (25). Past work dismissed HHG using He as exhibiting very low efficiency;

however, this simply resulted from the fact that wavelength and the intensity of the driving laser and the gas pressure were not well optimized. However, due to its relatively low index of refraction and absorption (especially in the soft x-ray region), the optimum pressure to maximize the harmonic yield using He is typically much higher than for other gasses, and virtually impossible to achieve in a gas-jet or even a cell geometry.

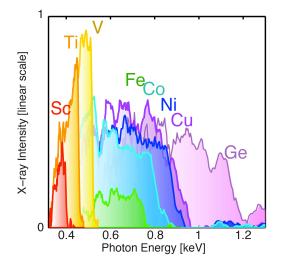


Fig. S2. The x-ray supercontinuum source enables site-specific spectroscopy at many L absorption edges simultaneously. Here thin metal films were placed between the HHG source and spectrometer.

Interestingly, even in the EUV region, when HHG is driven by 10-cycle FWHM 0.8 μ m lasers, (where the laser photon energy is ~ 1.5 eV with individual harmonics 3 eV apart), there is measurable spectral amplitude between the well-separated HHG peaks, resulting in a quasi-continuum. This enables transient reflection or absorption experiments spanning multiple M-shell absorption edges (*15, 30, 31*). For HHG driven by 3.9 μ m lasers, the laser photon energy is 0.3 eV, while the separation between individual harmonics is 0.6 eV. Thus, due to the finite width of the harmonics, the x-ray HHG spectrum is continuous, spanning multiple L-shell absorption edges (see Fig. S2). The spectral resolution of the spectrometer, ~ 200 in the keV region, cannot distinguish the individual harmonic orders.

Laser beam self-confinement in the mid-infrared

To understand how nonlinear laser beam propagation and spatio-temporal confinement contributes to phase matching, we performed 3D simulations taking into account the spatial profile of the gas in the waveguide. The gas has a uniform pressure section in the center of the waveguide, while the entrance and exit sections (~ 0.5 cm long) have a density gradient. We therefore start the field propagation from a position outside the waveguide where the density of the gas is low. For propagation in the region before the waveguide, we use the nonlinear envelope equation (NEE) (34) given by:

$$\frac{\partial E}{\partial z} = \frac{i}{2k} \left(1 + \frac{\partial}{\omega_0 \partial t} \right)^{-1} \nabla_{\perp}^2 E - i \frac{k''}{2} \frac{\partial^2 E}{\partial t^2} + \frac{ik}{n_0} \left(1 + \frac{\partial}{\omega_0 \partial t} \right) \left(n_2 \left| E \right|^2 \right) E - \frac{(\rho_n - \rho_e) W_{ADK} U}{2\left| E \right|^2} E - \frac{k}{2n_0^2 \rho_c} \left(1 + i\omega_0 \tau_c \right) \frac{\omega_0 \tau_c}{\left(1 + \omega_0^2 \tau_c^2 \right)} \rho_e \left(E - \frac{(\partial E / \partial t) \tau_c}{1 - i\omega_0 \tau_c} \right)$$

where *E* is the driving laser field, *z* is the propagation distance, *k* is the wavenumber corresponding to the central wavelength at 3.9 µm, ω_0 is the angular frequency at the central wavelength, *k*" is the group velocity dipersion parameter, n_0 is the refractive index of the gas, n_2 is the nonlinear index, ρ_n is the neutral gas density, ρ_e is the electron density, W_{ADK} is the Ammosov-Delone-Krainov (ADK) ionization rate, *U* is the first ionization energy, ρ_c is the critical density at 3.9-µm wavelength, and τ_c is the electron collision time. The terms on the right side of the equation represents diffraction, dispersion, Kerr-nonlinearity, tunneling ionization absorption, and plasma defocusing and collisional absorption, respectively. We also assume that parameters are pressure dependent (*33*) and an exponential density gradient such that $\rho(z) = \rho_0 \exp[-(z/L)^2]$, where ρ_0 is the neutral gas density at the entrance of the capillary. ADK tunneling ionization and collisional ionization are used for plasma generation. By numerically integrating the NEE, we determine the laser field profile at the input of the capillary.

For propagation inside the waveguide, we decompose the calculated input laser field into Bessel functions inside the capillary. Each Bessel mode amplitude is propagated independently, including modal dispersion and loss. After reconstructing the electric field from a superposition of the Bessel modes, we solve the nonlinear propagation in the spectral domain, including self-focusing, plasma defocusing, and absorption using the NEE. Since gases are injected through two holes that are located 5 mm from entrances of the capillary, there are also density gradients from the injection holes to two ends of the capillary. We use –

$$\rho(z) = \sqrt{\rho_0^2 + (z/L)(\rho_{\text{max}}^2 - \rho_0^2)}$$

where ρ_{max} is the maximum gas density, which is proportional to the backing pressure (39).

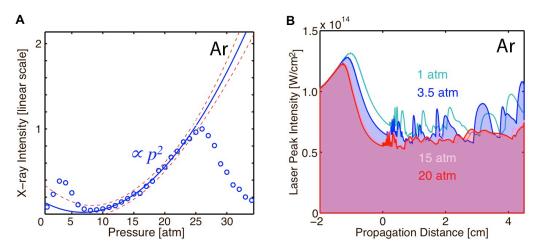


Fig. S3. A. Experimental x-ray yield as a function of pressure in Ar gas, showing two pressure peaks due to phase matching (3 atm) and laser beam self confinement (26 atm). **B**. Calculated on-axis peak laser intensity vs. propagation distance for different gas pressures. The capillary input is located at z = 0. A stable intensity is established due to spatio-temporal confinement at high > 20 atm pressures inside the waveguide.

Figures S3 B and S4 A plot the on-axis peak laser intensity as a function of propagation distance *z*, for Ar and He gas at different pressures (additional calculations for Ar are shown in Fig. 3 of the main text). For low pressures (~ 10 atm), the peak intensity inside capillary (z > 0) shows periodic oscillations due to energy exchange between the fundamental mode (i.e., lowest-order Bessel beam) and the higher-order modes. However, beam confinement due to the Kerr effect is observed at high pressures (> 15-20 atm for Ar and >30-40 atm for He) in the constant pressure region (0.5 cm < z < 2.5 cm), since the critical power for self-focusing and filamentation is inversely proportional to gas pressure (40). Here the focused beam $1/e^2$ radius at vacuum is 130 µm and the radius of the capillary is 200 µm. These findings match experiment very well.

The peak plasma density (Fig. S4 B) shows the same stabilization for higher pressures, showing that phase matching is supported by this laser beam self-confinement. The peak laser intensity and plasma density are in very good agreement with the experimental parameters for full phase matching, with minor deviations within the error of various parameters such as n_2 , ionization models, and focus position with respect to the waveguide. Figure S4 C shows examples of spatio-temporal mode profiles for two different locations and pressures for He. Compared with mode fluctuations at lower pressures due to interference between the fundamental and the higher-order modes, the spatial modes at high pressures (at values of 40 atm close to the phase-matching pressure for λ =3.9 µm) are strongly localized near the waveguide

center (r = 0). Strong spatio-temporal confinement is evident after the laser propagates 1.75 cm at high 40 atm pressure inside the waveguide.

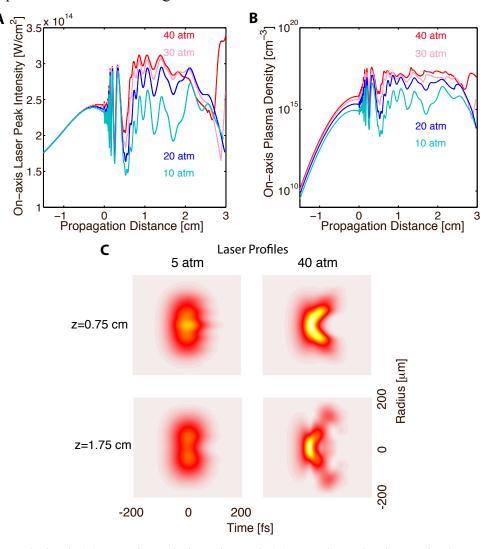


Fig. S4. Calculated (A) on-axis peak intensity and (B) on-axis peak plasma density vs. propagation distance for different He gas pressures. (C) Examples of spatio-temporal beam profiles at two different locations for high (40 atm) and low (5 atm) pressures inside the capillary. Here the capillary input is located at z = 0. Spatio-temporal confinement is evident after the laser propagates 1.75 cm at high 40 atm pressures inside the waveguide.

Further evidence for laser self-confinement is that phase-matched buildup will occur at a higher pressure for a smaller guided focal spot regardless of whether guiding is extrinsic (i.e. a waveguide) or intrinsic (i.e. self confinement). Thus, in the case of Ar, the second peak in x-ray yield as a function of pressure seen in Figs. 3 and S3 A is expected if the laser beam and x-ray beam size reduces. Figure S4 C also shows that the laser pulse compresses both in time and space as the laser propagates through the long interaction region with high density-length

product. This spatio-temporal compression supports phase matching over cm distances, and also greatly reduces any ionization-induced and normal diffraction that might occur at long wavelengths or for shorter interaction lengths (41). Thus, the waveguide geometry is also ideal for laser beam self-confinement and phase matching of the high harmonic generation process in the keV region. Finally, theoretical and experimental results using other gases such as molecular N_2 suggest that similar favorable conditions for efficient HHG upconversion in the presence of laser beam self-confinement can be achieved. Thus, remarkably, nonlinear optics and extreme nonlinear optics converge in the x ray region, where both laser self-confinement and HHG phase matching require high pressures and long propagation distances to form a self-guiding laser beam and to extend the quadratic buildup of the HHG signal to compensate for the low single-atom HHG yield.

Theoretical models of HHG phase matching in the mid-infrared

Next we present details of our simulations for how the phase-matched HHG emission at keV photon energies builds up over macroscopic distances in a high pressure medium, in order to predict the HHG pulse duration and phase. The kernel of our propagation code is the single-atom harmonic radiator solver based on the strong field approximation (SFA) (17, 42) and calculates the HHG radiation field from a single atom taking into account the particular electromagnetic field at the position of the atom. Our code is based on an extension of the strong field approximation (SFA+) (24), which increases the quantitative accuracy of the standard SFA. In order to reduce the computing time, the original SFA+ formalism has been simplified using the saddle-point approximation. This enables us to compute the single-radiator yield within minutes at laser wavelengths in the mid-IR regime, for which an exact solution of the time-dependent Schrödinger equations becomes impractical due to the extremely long computational times required. The ionization and recombination matrix elements within the SFA+ are evaluated for a Helium atom using Roothaan-Hartree-Fock wavefunctions (43). The dipole acceleration is computed from the gradient of the Coulomb potential for He given in (44).

Field propagation is then computed from the single atom response using a scheme based on the discrete dipole approximation (37) assuming a plane wave incident field, and including ionization, neutrals and group velocity effects in the fundamental field phase. Absorption was taken into account in the propagation of the harmonics. The attosecond pulses have been computed by Fourier transformation of the harmonic spectrum detected on-axis. The target is modeled as Helium gas with a density of $5x10^{19}$ atoms/cm³. The disparity of the temporal scales involved in the computations (femtosecond for the driving field and attosecond for the most energetic harmonics) together with the high density used, requires an extremely precise computation. At present we are able to demonstrate good convergence for propagation distances up to 20 microns, which correspond to the data presented here. In our calculations, the laser pulses are modeled using a sin² envelope of 1 and 6 cycle FWHM, 3.9 microns wavelength and phase-matching peak intensities of 4.1 and 3.3 x 10^{14} W/cm². The phase matched x-ray spectrum and group delay obtained for the 6 cycle case is shown in Fig. 1C of the main text, as well as the predicted x-ray pulse duration for one of its temporal bursts.

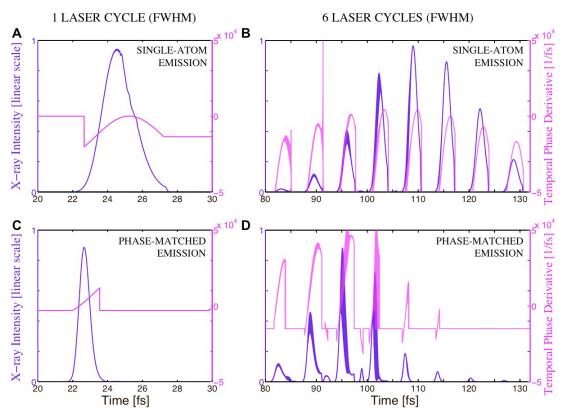


Fig. S5. Calculated HHG temporal emission and phase for both a single atom and under phase matching conditions (20 μ m propagation distance) when driven by a 1-cycle FWHM pulse with peak intensity $\simeq 4.1 \times 10^{14}$ W/cm² (**A** and **C**) and a 6-cycle FWHM (75 fs) 3.9 μ m laser pulse with peak intensity $\simeq 3.3 \times 10^{14}$ W/cm² (**B** and **D**). The target is He gas with a density of 5 $\times 10^{19}$ atoms/cm³. Note that both the duration of the x-ray burst and the number of bursts reduce in the macroscopic phase-matched HHG cases.

A comparison of the HHG temporal emission driven by 1 and 6 cycle FWHM mid-IR

laser pulses, in a phase matched regime, is shown in Figs. S5 C and D. For a 1-cycle FWHM driving laser pulse, Fig. S5 A predicts a burst of 2 fs FWHM HHG burst (associated with ionization due to the first half-cycle of the laser) with a well-behaved chirp. For a 6-cycle FWHM driving laser pulse, the phase-matched HHG emission consists of three intense pulses, each of 2-3 fs duration, also with a well-behaved chirp, and with a total FWHM envelope of 14 fs. The number of x-ray bursts is reduced compared to the single-atom calculation (Fig. S5 B) since the phase-matched emission occurs only over a fraction of the driving laser pulse, near the critical ionization level. As explained in the main text and observed in experiments using 800 nm driving pulses (38), bright HHG emission in the form of an isolated, chirped, x-ray burst is expected for longer propagation distances than those used in Fig. S5, which are at the current limit of advanced theory. The well-behaved chirp means that the x-ray pulse could be compressed in principle, to generate a single-cycle, 2.5 attosecond, Fourier-limited pulse. However, for most time-resolved x-ray experiments, compression to the Fourier limit would not be needed for attosecond time resolution experiments, since no system absorbs over a keV bandwidth. Therefore, knowledge of the chirp would suffice to capture dynamics on multiple atomic sites simultaneously, with attosecond time resolution.

Note that the simulations capture amplitude and phase noise in the x-ray burst, which shifts from the trailing edge to the leading edge of the bursts as the x-rays propagate. Suppressing the higher-order returns of the electron to the ion almost eliminates this noise. However, since emission from higher order returns phase matches under different conditions, this noise is expected to be suppressed for the longer propagation lengths used in the experiment.

In principle, short (positive slope) and long (negative slope) electron trajectories contribute to a temporal chirp. However, in a phase matched waveguide geometry, only short trajectories survive (or in the case of quasi phase matching, either the contribution from the long or short trajectories can be selectively enhanced) (45). This is another example of favorable microscopic and macroscopic physics, that in this case selects the short trajectories and reduces phase noise associated with very long trajectories for mid-IR driving wavelengths.

Analytic model of HHG phase matching from the UV to the keV

The explicit form of the analytical phase-matching cutoff rule is obtained by inserting the laser intensity at which tunnel ionization occurs (46), which is a function of the critical ionization

level, into the single-atom cutoff rule:

$$h\nu_{PM\ cutoff} = I_p + \frac{0.5I_p^{3+a}}{\ln^2 \left[\frac{0.86\tau_L I_p 3^{2n^*-1}G_{lm}C_{n^*l^*}}{-\ln(1-\eta_{CR}(\lambda_L))}\right]}\lambda_L^2$$

where a=0.5 is a correction to the analytical approximation, $n^*=Z(13.6/2I_p)^{1/2}$ and l^* are the effective principal and effective orbital quantum numbers, Z is the ionization state, $C_{n*l}=(2e/n^*)^{n*}/(2\pi n^*)^{1/2}$, where e is the base of the natural logarithms, and $G_{lm}=(2l+1)(l+|m|)!/(6^{|m|}|m|!(l-|m|)!)$. This analytical approximation is valid as long as tunneling ionization does not reach the barrier suppression regime. In barrier suppression ionization, the effective Coulomb potential is bent by the laser field below the valence electron energy.

The exact formula for η_{CR} should be used for the phase matching cutoffs, especially for VUV harmonics driven by a short wavelength laser, where the refractive indices of the gas for both the fundamental and HHG wavelengths is rapidly changing. As a result, in the VUV region, the scaling of the phase matching limits deviates from $hv_{PM cutoff} \propto \lambda_L^{(1.5-1.7)}$, as shown in Fig. 3A.:

$$\eta_{CR} = \left[\frac{\lambda_L^2 r_e N_{atm}}{2\pi\Delta\delta} \left(1 - \frac{1}{q^2}\right) + 1\right]^{-\frac{1}{2}}$$

However in the soft and hard X-ray regions, our analytic model for phase matching can be simplified, since $\eta_{CR} \ll 1$, and agrees with previous numerical calculations that predict a scaling of the phase matching as $hv_{PM cutoff} \propto \lambda_L^{(1.5-1.7)}$ (25). In the hard X-ray region of the spectrum, where η_{CR} is a tiny fraction of a percent, the analytic phase matching cutoff rule simplifies to:

$$hv_{PM\ cutoff} \approx \frac{\alpha I_p^3}{\ln^2 \left[\gamma I_p N \lambda_L^3\right]} \lambda_L^2$$

where N is the number of laser cycles and taking into account that the critical ionization drops as $\eta_{CR} \propto \lambda_L^{-2}$.

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