Phase matching techniques for coherent soft-x-ray generation


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Abstract

Coherent beams at soft x-ray wavelengths can be generated using extreme nonlinear optics by focusing an intense laser into a gas. In this paper we discuss phase matching and quasi-phase matching techniques that use gas-filled modulated waveguides to enhance the frequency conversion process. This leads to the generation of soft-x-ray beams that are both spatially and temporally coherent.

Introduction

Nonlinear-optical (NLO) techniques for converting light from one frequency to another play a pivotal role in expanding the range of wavelengths over which coherent light sources operate. Techniques such as optical frequency doubling, frequency mixing and parametric amplification allow laser light at a fixed wavelength to be converted to wavelengths that span the entire visible, near infrared, and near-ultraviolet regions of the spectrum. Laser sources making use of NLO techniques have found broad application in many areas of science and technology.

One area that has received increasing attention in recent years is the generation of coherent light in the extreme-ultraviolet (EUV) and soft x-ray (SXR) regions of the spectrum, using “extreme” nonlinear optics. In general, the EUV/SXR spectral region is challenging for nonlinear optics because all materials strongly absorb this radiation, with absorption depths at solid density in the micron to sub-micron range. As a result, traditional frequency-conversion techniques that rely on anisotropic crystalline solids as the nonlinear medium cannot be used. Nevertheless, coherent, low-divergence, laser-like beams of EUV light can be generated when an intense femtosecond laser is focused into a gas at intensities in excess of $\sim 10^{14}$ W/cm$^2$. This process represents an extreme limit of nonlinear optics: hundreds of visible photons, each with energy 1-2 eV, are combined together using a frequency upconversion process called high harmonic generation, resulting in coherent light at photon energy up to $\approx$1 keV. However, as will be explained below, this high harmonic generation (HHG) process is radically different from traditional nonlinear optics in several important respects.

Several strong factors that motivate the development of small-scale EUV light sources. The EUV region of the spectrum corresponds to photon energies above the ionization potential of any neutral atom or molecule. Therefore, intrinsic absorption resonances in atoms, molecules and solids can be used as a structural probe for materials characterization, or for monitoring ultrafast dynamic changes in electronic, chemical or catalytic systems. Moreover, the short wavelength of EUV light allows it to be focused more tightly. This property enables a host of applications in nano-imaging, in lithography for integrated circuit fabrication, and for biological and materials
microscopy. These applications currently can be pursued only at large-scale synchrotron facilities. Furthermore, the very broad bandwidths generated using high-harmonic generation make it possible to access very short, attosecond time-scales, making it possible to elucidate new physical phenomena and test our fundamental assumptions about the nature of light-matter interactions.

In this paper, we will discuss work in developing techniques for phase matching the high harmonic frequency upconversion process, to optimize the generated flux and coherence of the upconverted light. In particular, phase matching and quasi phase matching techniques using gas-filled hollow waveguides will be discussed.

**Coherent Soft-X-Ray Generation using “Extreme” Nonlinear Optics**

In many nonlinear-optical processes, an intense coherent beam of light (the “fundamental”) passes through a material, driving the bound electrons in the material strongly enough that their motion becomes anharmonic. The resulting induced nonlinear polarization means that some of the fundamental light is re-emitted by the driven electrons at a different frequency. Since the nonlinear polarization is created by the fundamental light, this conversion process can retain the coherent characteristics of the original beam. High harmonic generation (HHG) [1-3] takes this concept to its extreme by increasing the driving laser intensity to the point where the valence electron begins to ionize due to optical field-induced ionization. Once ionized, the electron will move freely in response to the field, oscillating sinusodally. These oscillating free electrons do not emit harmonics in the nonrelativistic limit. However, some of them can recollide with their parent ions after being driven for a fraction of a single optical cycle, and can recombine. [4-6] If this happens, the electron must release its kinetic energy at the time of the recollision, as well as the binding energy of the electron. It does this by emitting an energetic photon. The extremely anharmonic motion of the ionizing and recombining electron is the origin of the high-order harmonic emission.

In the classical picture of HHG, the maximum (or cutoff) photon energy emitted is calculated by considering the maximum possible total energy of the recolliding electron, and is given by [6]:

$$E_{\text{max}} = I_p + 3.2U_p$$

(1)

where $I_p$ is the ionization potential of the atom, and $U_p$ is the ponderomotive or wiggle energy of the electron in the laser field, given by $U_p = e^2E^2/4m\omega^2 \propto I_L\lambda^2$ where $e$, $E$, $m$, $\omega$, $I_L$, and $\lambda$ are the electron charge, field amplitude, electron mass, laser frequency, intensity, and wavelength respectively. In Eqn. 1, $U_p$ is the average kinetic energy of an electron oscillating in response to light with intensity $I_L$, while $3.2 U_p$ is the maximum kinetic energy of the electron at the time of recollision. Electrons ionized at different times within the optical cycle recollide with different energies. Neutral atoms, molecules or ions exposed to lasers with focused intensity of $10^{13}$-$10^{16}$ W/cm$^2$ can emit high-order harmonics in the range of tens to several hundreds of eV under the correct conditions.

The beauty of the high harmonic generation process is that much of the dynamics can be understood using classical physics. Many of the characteristics of the emission, such as the overall range of photon energies emitted, or the existence of discrete harmonics, can be derived from the simple and intuitive classical recollision model for the electron. However, other
characteristics of high harmonic generation depend fundamentally on the fact that the electron is a quantum-mechanical object.\textsuperscript{(6-11)} The importance of the quantum nature of high harmonic generation can be intuitively seen by considering what happens as the electron begins to field-ionize from an atom. As part of the electron wave function is accelerated away from the ion by the electric field of the laser, the wavelength of the electron follows the deBroglie relationship $\lambda = \frac{\hbar}{p}$. Between the time that the electron is released, and when it recollides with the ion a fraction of an optical cycle later, the electron wave function advances in phase by several cycles. The phase and energy of the emitted high-harmonic light is then directly related to the phase and energy of the returning electron. This quantum phase can also have a contribution due to the phase and shape of the laser pulse itself, since these determine the exact trajectory of the electron.\textsuperscript{(6-11)} The interesting and important point to note is that the quantum phase of the electron, and hence the harmonics, can be manipulated by shaping the laser field. Also, because of the finite time between ionization and recombination, and because different harmonics are generated from electrons that have traveled slightly different paths, the harmonic phase is therefore not locked in phase with the driving laser. This quantum phase (sometimes referred to as the intrinsic phase) has a profound influence on the spatial coherence (or divergence) of the high harmonic beam,\textsuperscript{(9)} as well as on the emitted spectrum and pulse duration.\textsuperscript{(8)} Because the quantum phase is dependent on the driving laser intensity, it also provides a means for manipulating the emission by controlling the laser field and the interaction geometry.\textsuperscript{(10, 11)}

The existence of the quantum phase corresponds to a fundamental difference between high-harmonic generation and other nonlinear-optical processes such as second harmonic generation, where the light emission originates from electrons that never escape the atom. HHG is a highly nonlinear process that is purely “electronic” (i.e. it results only from electronic motion, and not from any motion of the atoms themselves), but with a finite response time, corresponding to the length of time the electron propagates in the laser field—a few hundred attoseconds ($1 \text{ as} = 10^{-18} \text{ sec}$). Moreover, high harmonic generation must take place in plasma that is both strongly absorptive and strongly dispersive, further complicating the phase matching of the conversion process. Nevertheless, concepts from traditional nonlinear optics have proven very useful in enhancing this extreme frequency conversion process, allowing for significant increases in the generated flux and coherence properties of HHG. The result is a practical compact, coherent, ultrashort-pulse EUV light source.

**Phase matching techniques for frequency conversion to the EUV**

For any nonlinear optical process to be efficient, the emission from a large number of atoms that are driven nonlinearly must radiate coherently. As the driving laser beam passes through the medium, a coherent harmonic signal builds-up that co-propagates with it. However, to build-up coherently over an extended propagation distance, the fundamental and harmonic beams must travel with the same phase velocity; i.e. the process must be phase-matched. In a phase matched frequency conversion process, the nonlinear response from the medium continues to add constructively to the signal beam, leading to a bright output signal at a new wavelength.

Consider the electric field wave vector of the fundamental laser $E_\omega \propto e^{i(k_\omega x - \omega t)}$ and the generated harmonic light of order $q$, $E_q \propto e^{i(k_q x - q\omega t)}$. The phase mismatch $\Delta k$ between the two waves as they propagate depends on the fraction of neutral atoms and plasma present in the medium. It also
includes geometric terms that depend on the focusing conditions and intrinsic phase arising from the radial intensity profile of the laser. The phase mismatch can be expressed as \[\Delta k = qk_\omega - k_{q\omega} = P[\eta N_{\text{atm}} r_e (q\lambda - \lambda/q) - \frac{2\pi(1-\eta)q}{\lambda} \Delta \delta] + \text{Geometric terms}\]

Here, \(\lambda\), \(\eta\), \(P\), \(N_{\text{atm}}\), and \(r_e\) are the fundamental wavelength, ionization fraction, gas pressure in atmospheres, number density at 1 atm, classical electron radius, and \(\Delta \delta = \delta(\lambda) - \delta(\lambda/q)\) is the difference in the index of refraction at the fundamental and harmonic frequencies for the gas at 1 atm. In Eqn. 1, \(\omega/k = v_p\), where \(\omega\), \(k\) and \(v_p\) are the frequency, wave vector, and phase velocities of the light respectively. If a frequency conversion process cannot be phase matched, the phase slip between the fundamental laser field and the generated harmonic will eventually reach \(\pi\), resulting in destructive interference and back-conversion into the fundamental field. The propagation distance over which this occurs is defined as the coherence length, \(l_c\). In the case of harmonic generation of order \(q\), the coherence length is related to the phase mismatch \(\Delta k\), by \(l_c = \pi/\Delta k\). In a non-phase matched regime, the generated nonlinear signal thus arises only from the last few coherence lengths of the medium, and is much less efficient than when phase matched.

First let us consider the (overly simplified) geometry of a plane-wave with uniform intensity. In this case, the contributions to the phase mismatch due to geometric terms can be neglected. Phase matching \((\Delta k=0)\) occurs at a specific ionization level (independent of pressure) – called the critical ionization \(\eta_c\) - for each gas species.[13, 14] This critical ionization can be derived from Eqn. 2 and, assuming \(q\) large so that terms proportional to \(1/q\) are small, is given approximately by:

\[\eta_c = \left(1 + \frac{N_{\text{atm}} r_e \lambda^2}{2\pi \delta \delta}\right)^{-1}\]

Values of the critical ionization are \(\approx 0.5\%\), \(1\%\) and \(5\%\) in He, Ne and Ar respectively. For a laser pulse incident on a gas, high-order harmonics are generated during the entire time when the gas is undergoing field-induced ionization. However, at some point in time, the critical ionization level is reached. At this time the coherence length becomes infinite, and the conversion efficiency is limited only by the absorption depth of the EUV light.[15] Past this critical ionization level, the HHG conversion efficiency again drops.

During the time atoms are being ionized, high-order harmonics are being generated with energies up to the cutoff photon energy (Eqn 1), which is directly proportional to the laser intensity. Using a short pulse duration will delay ionization, and result in a higher cutoff photon energy when the critical ionization level is reached. This result can be seen from Table 1 and Figure 1. Table 1 lists the calculated values of the cutoff photon energy at the critical ionization value for several laser pulse durations and gas species. For this calculation, the ionization is calculated using ADK (Ammosov-Delone-Krainov) tunneling ionization rates[16] in the case of Table 1, or using a more-rigorous nonadiabatic quantum simulation[17] for the data of Figure 1. Figure 1 plots the ionization levels present in Helium gas at the peak of a pulse, for that laser pulse intensity that generates the given cutoff photon energy (Fig. 1 x-axis). From Fig. 1 and Table 1 we see that, even in the case of He, it is not possible to phase-match HHG at photon energies in excess of \(\approx 100 - 150\text{eV}\), because the ionization exceed the critical ionization at the required laser intensities to generate these photons. Moreover, since He is a small atom, the magnitude of the recollision...
cross-section, and therefore the effective high-order nonlinear susceptibility, is low. Therefore, the emitted harmonics are not as bright as from larger atoms such as Xe or Ar. These calculations show in a simple way that to make further progress in extending high-order harmonic generation to photon energies >100 eV with reasonable flux, it is necessary to overcome the detrimental effects of ionization on phase matching.

Using waveguides for Extreme Nonlinear Optics

From the discussion above, it can be seen that the phase-mismatch between the laser fundamental and the generated high-harmonic light can vanish if an atom is driven with a pulse short enough that harmonics are generated while the gas is still only slightly ionized. The exact conditions for this phase matching depend on the focusing geometry. However, the interaction distance over which the HHG light is generated will still be limited, due to ionization-induced defocusing, the short confocal parameter of a beam focused tightly enough to reach the required intensities, and the resulting geometric (or Guoy) phase shift through the focus that results in a finite phase mismatch for the harmonic generation process. With enough energy in a laser pulse (>10 mJ), and using a very large focal spot (obtained by using a ~5 meter focal length lens to focus a TEM$^{00}$ laser beam), this type of quasi plane-wave phase matching can be implemented in some gasses.[18] However, at the lower per-pulse energies (~1 mJ) that can be readily obtained using kilohertz or higher repetition rate lasers, the focusing requirement makes the confocal parameter of the beam too short to obtain optimal phase matching. Furthermore, gas loads for the required windowless geometry become very challenging. A method for controlling the interaction geometry to extend the confocal parameter thus becomes very desirable. Fortunately, this is possible. The interaction geometry for HHG can be engineered, making it possible to adapt a number of techniques from visible-wavelength nonlinear optics for high harmonic generation.

In nonlinear optics, the technique of using a waveguide to increase the interaction length of an NLO process is well established. A waveguide makes it possible to create a well-defined plane-wave-like interaction over an extended distance. This concept can be applied to the HHG process by guiding intense light inside a gas-filled hollow waveguide (see Fig. 2).[12, 14, 19, 20] The light is guided by glancing-incidence reflection from the walls of the waveguide,[21] allowing the laser and high harmonic beams to co-propagate over an extended interaction length with a well-defined and controlled intensity and phase profile. In the hollow waveguide, the fundamental and harmonic light propagate in waveguide modes that correspond to a modified propagation vector. The waveguide affects the fundamental light propagation the most, since the waveguide counteracts diffraction of the light, which is strongest for the longer wavelengths. The high-order harmonics are nearly unaffected by the waveguide. The effect of the waveguide is to speed-up the phase velocity of the guided mode compared with free-space propagation. Thus, the contribution to the propagation wave vector has a different sign for the neutral gas compared with the plasma and the waveguide, making it possible to tune the phase mismatch between of the driving laser and the harmonic light. In the presence of guiding, Eqn. 2, becomes[12] —

$$
\Delta k = P \bigg[ \frac{2\pi (1-\eta)q}{\lambda} \Delta \delta - \eta N_{\text{atm}} r_e (q\lambda / q) - \frac{q u_{11}^2 \lambda}{4\pi a^2} \bigg] = q \bigg[ P \left( \frac{2\pi}{\lambda} \Delta \delta - \eta \left( \frac{2\pi}{\lambda} \Delta \delta + N_{\text{atm}} r_e \lambda \right) \right) - \frac{u_{11}^2 \lambda}{4\pi a^2} \bigg] \quad (4)
$$
where $a$ and $u_{11}$ are the waveguide radius and the first zero of the Bessel function $J_0$, respectively. From Eqn. 4 we can see that for low levels of ionization ($\eta$ small), phase matching ($\Delta k = 0$) of high harmonic generation can be accomplished by adjusting the gas pressure so that the waveguide and plasma dispersion balance the neutral atom dispersion.[14] Since phase matching is achieved primarily by adjusting the phase velocity of the fundamental laser, and since the high-harmonic light travels at a phase velocity $\sim c$ due to its minimal interaction with the waveguide, the bandwidth of the pressure-tuned phase matching is very broad, and can encompass many harmonic orders. When phase matched, the harmonic signal initially increases quadratically with interaction length, ultimately limited by the background gas absorption. Other advantages of the waveguide geometry include the generation of EUV light that has all the characteristics of a laser beam, i.e. possessing full spatial coherence.[22, 23] The waveguide helps to establish a well-behaved laser mode with uniform radial phase, even in the presence of some level of ionization.[24] This laser mode is in-turn upconverted into an EUV beam with a radial profile and phase that are also well behaved. This behavior is in contrast to a gas jet geometry, where the measured coherence values are limited to $\approx 50\%$ due to spatial nonuniformities and lack of phase matching. Second, since the interaction length possible in the waveguide can exceed the confocal parameter of a free-focus beam by up to an order of magnitude or more, HHG in waveguides around 50eV can be implemented using very low laser pulse energies of as little as 50µJ. Finally, the gas load on the vacuum system is also very low. The basic implementation of hollow waveguide HHG uses a 3-segment capillary tube with gas feed such that the center section is kept at constant pressure (see Fig. 2). The gas load on the system corresponds to the (small) conductance of the end section of the waveguide—a ~150µm tube a few mm long. The gas load can generally be handled using a small turbo pump, and if UHV pressures are required a thin-film filter can be used to isolate the high vacuum from the ultra-high vacuum portions of the system.

Even in the case of upconversion in a hollow waveguide, however, phase matching is still possible only in the case where the gas is not fully ionized. The left-hand side of Fig. 3 shows a parametric plot that illustrates traditional phase matching in the hollow waveguide, based on Eqn. 4 and as a function of ionization fraction of the gas. Essentially, traditional phase-matching is possible only when the harmonics are generated at ionization levels less than $\eta_c$, and the phase-matching conditions depend on both the pressure and the ionization fraction. This prediction is somewhat pessimistic regarding the tolerable ionization level, since it assumes that the gas in the waveguide is uniformly ionized. In actuality, only the central axis along the waveguide is fully ionized, while laser beam mode occupies the entire waveguide. The outer portions of the beam propagate in neutral gas, and as a result, propagate at phase velocities less than or close to $c$, the speed of light. Through modal averaging, this helps to counteract the increasing phase velocity in the (more ionized) center of the waveguide. This effect reduces the phase mismatch due to the plasma, and increases the allowed ionization levels for which phase matching can still be achieved to $\approx 2\eta_c$. Nevertheless, ultimately the tolerable level of ionization of the gas to obtain phase-matched conversion is quite limited.

Even using waveguide techniques, frequency conversion into the EUV is still more challenging and more limited than for conventional low-order nonlinear processes. Since the upconversion usually occurs in the presence of strong background absorption (since the EUV photon energies are above the ionization potential of the medium), absorption limits the conversion efficiency
that can be obtained. Optimum conversion efficiency for HHG in the range of $10^4 - 10^6$ to photon energies in the 20-50 eV range, and decreases rapidly at higher photon energy. This is far less than the $>50\%$ efficiencies that can be obtained for processes such as frequency doubling. Nevertheless, the waveguide geometry makes it straightforward to generate a useful flux of fully coherent light, with approximately $10^{12}$ photons per second at 40-60 eV, using kHz repetition rate driving lasers. This brightness is comparable to the coherent flux that can be obtained from a synchrotron bending magnet, and is useful for a variety of experimental applications such as photoelectron spectroscopy of gas-phase and condensed phase media, for lifetime and spectroscopy measurements, and for time-resolved absorption and reflectivity measurements.

**Quasi-phase-matched conversion to soft x-ray photon energies using modulated waveguides**

Further progress in developing HHG as a light source in the soft x-ray region of the spectrum can be accomplished by combining concepts borrowed from conventional nonlinear optics with the unique quantum dynamics of high-harmonic generation. In the visible region of the spectrum, it is well established that in cases where true phase-matched frequency conversion is not possible, *quasi phase-matching* (QPM) can still allow for efficient frequency conversion.[25] In QPM, the phase slip between the driving laser and the generated signal is accommodated-for, rather than eliminated. This adjustment of phase slip is achieved by modulating the frequency conversion process with a period that corresponds to the phase slip between the fundamental and harmonic waves. The most-successful application of this concept for visible-wavelength photonics has been the use of periodically poled materials for frequency doubling.[26] In these materials, the crystal orientation of the nonlinear material is reversed periodically. As the fundamental and the harmonic travel through the material, the driving laser and the harmonic signal accumulate a half-wave of phase slip. If the orientation of the nonlinear crystal then flips, the sign of the phase of the generated harmonic light also flips, and further phase slip between the fundamental and the harmonic will cause the signal to again be generated *in-phase* with the previously-generated light. This situation guarantees that the nonlinear signal builds up constructively during propagation through the medium.

In the case of high harmonic generation, there is no equivalent to orientation of the nonlinearity since an isotropic atomic gas is used as the nonlinear medium. However, because HHG is a *highly* nonlinear process, small changes in the laser intensity can strongly modulate the cutoff of the harmonic generation process, as well as the phase of the generated EUV light. For example, from Eqn. 1, a 1% change in intensity of the driving laser can result in a change in the cutoff photon energy of 1%, or 1 eV at 100 eV. Such a small change is thus enough to strongly modulate the intensity of the harmonic generated at the cutoff photon energy. Modulating the driving laser intensity can be accomplished simply by modulating the diameter of the guide itself.[27] In this way, high harmonic emission at the cutoff photon energy emerges only from certain regions where the diameter of the guide is reduced, while emission from regions where the guide diameter is larger is suppressed. The result is that the generated harmonic signal can continue to increase, with periodic “pauses” in the signal generation where the guide diameter is wider and the laser intensity is lower. This scheme automatically suppresses emissions from regions that would otherwise interfere destructively with the total harmonic signal. Mechanisms other than the simple intensity modulation, such as the intrinsic quantum phase shift of the
harmonic emission, or modulation of the plasma-induced index, also can play a role in quasi-phase-matching. Any periodic variations in the generation process can enable quasi-phase-matching to a greater or lesser extent.

Mathematically, the phase mismatch given in Eqn. 4 above can be modified to include the contribution arising from periodic modulation of period \( \Lambda \), to give -

\[
\Delta k = q \left( \frac{2 \pi}{\lambda} \Delta \delta - \eta \left( \frac{2 \pi}{\lambda} \Delta \delta + N_0 r_c \lambda \right) + \left( \frac{u_1^2 \lambda}{4 \pi a^2} - \frac{K_m}{q} \right) \right) = q \left( \frac{2 \pi}{\lambda} \Delta \delta \left( 1 - \frac{\eta}{\eta_c} \right) - \frac{2 \pi}{q \Lambda_c} (1 - m \beta) \right)
\]  

(5)

where \( K_m = 2 \pi m / \Lambda \) is the contribution to the phase mismatch due to the modulations in the waveguide, and \( m \) (integer) is the order of the quasi phase matching. First-order QPM (\( m=1 \)) corresponds to modulating the waveguide (and harmonic emission) every coherence length. In Eqn. 5 above, the terms are grouped to separate the gas and plasma contributions (first terms) from the waveguide and periodic contributions (last terms). Also, a critical periodicity, \( \Lambda_c \), can be defined where the quasi-phase-matching wave vector is equal to the wave vector for the straight waveguide. This critical periodicity is given by -

\[
\Lambda_c = \frac{1}{q} \frac{8 \pi^2 a^2}{\lambda^2} 
\]  

(6)

The ratio \( \beta \) is defined as \( \beta = \Lambda_c / \Lambda \), which is equal to 1 at the critical periodicity. Phase matching for \( \beta = 1 \) (when possible) is then independent of pressure and occurs at the critical ionization level \( \eta_c \). For \( \Lambda < \Lambda_c \) or \( \beta > 1 \), the last term in Eqn. 5 provide a net positive contribution to the \( k \)-vector of the laser light in guide, counteracting the large negative terms due to ionization. Finally, for \( \Lambda > \Lambda_c \) or \( \beta < 1 \), the last two terms in Eqn. 5 provide a net negative contribution to the \( k \)-vector of the laser light in guide, but a reduced one compared with the straight waveguide. Figure 3 illustrates the different possible regimes of phase matching, plotting the predicted, normalized, phase matching pressure-tuning curves for traditional and quasi phase matching.

For high harmonic orders generated at very high levels of ionization \( \eta \approx 1 \), the phase mismatch is dominated by plasma dispersion. In this limit, the approximate phase mismatch is given by -

\[
\Delta k \approx \frac{2 \pi m}{\Lambda} - \frac{q n_e e^2 \lambda}{4 \pi m_0 e_c c^2} = \frac{2 \pi m}{\Lambda} - q n_e r_c \lambda.
\]  

(7)

where \( \lambda \) is the laser wavelength, \( q \) is the harmonic order, and \( n_e \) is the electron density. For fully ionized argon at a pressure of 1 torr, and for \( q = 91 \), \( n_e = 3.5 \times 10^{16} \text{ cm}^{-3} \), the contribution to the phase mismatch due to the plasma and without QPM is \( \approx 132 \text{ cm}^{-1} \). Therefore, the coherence length, \( l_c \), given by \( l_c = \pi / \Delta k \), is 0.24 mm. This corresponds to a required modulation periodicity of \( \approx 0.5 \text{ mm} \) for quasi-phase matching at this ionization level and pressure. Alternatively in this limit, the modulation period \( \Lambda \) required for phase matching is given by -

\[
\Lambda \approx \frac{\eta_c \lambda}{q P \Delta \delta}
\]  

(8)
Thus, very substantial levels of ionization can be compensated for using QPM, with experimentally realizable modulation periods in the fraction of a millimeter range. The exact level of signal enhancement cannot be predicted analytically, since the level of phase/amplitude modulation of the HHG signal due to the periodic modulations is not necessarily optimum and requires a realistic calculation to predict. Simulations predict possible enhancement of > 1000 due to this QPM process in a highly optimized situation. Current experiments fall far short of demonstrating this level of enhancement, but still show large and repeatable signal enhancements.

Experimental Results—quasi-phase matching

To date, quasi phase matched high-order harmonic generation using modulated fibers has proven to be useful for photon energies in the range of 50 - >300 eV.[28, 29] In some cases, the use of this technique has resulted in increases in flux of 1-2 orders of magnitude compared with non-phase matched geometries.

In initial work on this topic, we demonstrated that use of a modulated waveguide could significantly extend the range of observable photon energies generated in Helium gas from 80 eV to ≈ 170 eV, by compensating for ionization-induced phase mismatch.[28] For this work, we used 3 cm long waveguides with modulation period in the range of 1mm to 0.5mm, filled with He at 111 torr. The laser pulse duration was 25 fs while the peak intensity was ∼ 5 x 10^{14} W/cm^2. This peak intensity corresponds to a cutoff photon energy of about 170 eV, and for a 25 fs-duration driving pulse. ADK models indicate that the Helium is ≈ 1-2% ionized at the peak of the pulse. This ionization level is above the critical ionization level in He, which is ≈ 0.5%. Therefore, conversion to 170 eV is not phase matched in a straight waveguide, but quasi phase matching can be used to enhance the effective interaction length at photon energies >100 eV. Experimental data for the spectrum at several values of waveguide periodicity are shown in Reference 28.[28] The spectrum extends to only ~80 eV in the case of a non modulated waveguide. However, it extends to progressively shorter wavelengths as shorter-period QPM waveguides are used. This behavior is consistent with a simple calculation: considering harmonic order 117 at 180 eV, and using a 0.5mm modulation period. For m=1 phase matching the contribution of the modulated waveguide to the phase mismatch is K ≈ 12,500m^{-1}. This value adds to the phase mismatch due to the waveguide (-7627 m^{-1}), the plasma (-8681 m^{-1}), and the neutral gas (4160 m^{-1}) to achieve phase matching i.e. Δk = 0.

In the case described above, the quasi-phase-matching process is demonstrably effective in a situation where the helium gas is still not strongly ionized. However, in attempting to improve the phase matching process for HHG in Argon and Neon, it is important to show that QPM can be effective even when the gas becomes nearly fully ionized. These gases have lower ionization potentials than He, and therefore will be more ionized for any given harmonic order generated. Furthermore, for generating photon energies of >200 eV even using Helium, the gas will be at least 10% ionized before photons are emitted. This is also illustrated by examining the data of Fig. 1. In order to generate photons from He in the so-called “water window” region of the
spectrum (i.e. photon energies ~300 eV), the ionization level of 10 – 50% is present, depending on pulse durations of 5 – 30 fs.

In this regime, quasi-phase matching can still be effective if modulation periods greater than the critical periodicity (Eqn 6), and relatively low gas pressures, can be used.[29] There is a limit to this, however, in that for modulation periods comparable to or shorter than the diameter of the waveguide itself, calculations indicate that the mode of the laser beam starts averaging over the modulations rather than following them. Therefore, for laser pulse widths of 20 fs, “m=1” quasi phase matching enhancement can be obtained at photon energies up to ~300 eV using a modulation period of <0.5mm, and sufficiently low pressure to limit the plasma contribution to the phase mismatch.

Figure 4 shows the harmonic emission from He at a pressure of 10 torr, from 10 cm long straight and modulated waveguides, with a periodicity of 0.25mm, and for a laser pulse duration of 20 fs.[29, 30] By using a modulated waveguide, the harmonic flux is enhanced by a factor of 5 at the carbon edge around 284 eV compared with a straight waveguide. However, the exact enhancement in x-ray emission achieved due to quasi phase matching in a modulated fiber is difficult to specify. This is because in a straight fiber at the laser intensities required to generate 150eV photons and greater, very often a form of self-quasi phase matching can occur where the laser periodically focuses. Thus, the x-ray emission from the straight fiber is likely to be enhanced.[31] For slightly lower laser intensities, shorter waveguides (≈ 3 cm) and longer pulse durations (≈ 25 fs), the emission at the carbon edge is significantly less, as can be seen in Fig. 5. Here, phase matching is possible at the lower 150 eV photon energies, but it is not possible to achieve m=1 phase matching at 300 eV due to the increased pressure and ionization levels. Nevertheless, higher order (m=3) quasi phase matching is possible at 300 eV and as a result, throughout the entire energy range, use of the modulated waveguide increases the x-ray emission. This is particularly true at photon energies > 225eV, where no emission is observed from a straight fiber. Based on estimates of detection efficiency, filter transmission, and measurements of grating efficiency, we determined a minimum flux of between $10^7 - 10^9$ photons/sec in a 10% bandwidth at the C edge. By using shorter pulses, tighter modulation periods, longer, low-loss waveguides, higher-order quasi phase matching, higher pressures and multistage waveguides, it should be possible to enhance the flux still further, possibly by several orders of magnitude.

The fact that signal enhancements can be observed in a regime where the quasi phase matching must be a higher order (m=3, or m=5) process is encouraging that this technique can provide some flux enhancement even at very high photon energies. In other work, we demonstrated that by using m=5 or m=7 QPM, we could observe high harmonic emission from Ne in the water window. This was the first demonstration of generation of coherent light in the water window using neon gas. Since emission from larger atoms such as Ne and Ar might be brighter than that from He, investigating harmonic emission from other atoms, ions and molecules at higher energies is very worthwhile.

Figure 6 shows the integrated flux for two different photon energy ranges as a function of pressure, for straight and modulated (0.5 mm) waveguides filled with Ne. Lower gas pressures often increase the highest observed harmonic orders. This effect is a result of reduced plasma defocusing and reduced absorption, both of which can suppress the intensity of the driving laser.
From Fig. 6, it is apparent that there is phase matching of higher photon energies in the modulated waveguides since the harmonic emission optimizes at a distinct pressure, as expected from Eqn. 5. In contrast, in the straight waveguide, phase matching is only achieved at lower energies – as expected because of the lower values of the phase mismatch at lower values of q.

The use of quasi phase matching in modulated waveguides continues to have a significant effect even at ionization levels up to 50%. Figure 7 compares harmonic emission in Ne between 90 eV and 180 eV, for the case of straight and modulated waveguides (periodicity 0.25 mm). The higher levels of ionization present in these experiments were compensated using tight (0.25 mm) modulation periods and low pressures of 11 torr. Using the modulated waveguides, the efficient energy range for EUV generation is extended from 105 eV to 150 eV, while the cutoff is extended from 130 eV to 165 eV. At even higher photon energies near the Carbon edge at 284 eV, the level of ionization present at the peak of a 22 fs laser pulse increases to \( \approx 80\% \), corresponding to a phase mismatch of \( \Delta k \sim 77,000 \text{ m}^{-1} \). The effective wavevector for first-order QPM for a modulation period of 0.25 mm is \( K_1 \sim 25,000 \text{ m}^{-1} \) however near the C edge, it is possible to compensate for the large phase mismatch by third order (m=3) QPM.

Figure 8 compares using Helium vs Neon as the nonlinear medium for generating light in the water window region 285 eV to 330 eV, using a time-dependent ADK ionization calculation with the laser intensity chosen to fully ionize Ne at the peak of the pulse. During the time when the atoms are ionizing, the ionization levels for Ne with an ionization potential of 21.6 eV, is approximately double that of He, with ionization potential of 24.6 eV. This larger level of ionization for Ne leads to larger values of the phase mismatch, which must be compensated using high-order QPM. Higher levels of ionization will also contribute to loss associated with ionization-induced refraction and defocusing, and with spatially-varying conditions as the pulse travels down the waveguide. In Fig. 8, the positions in the pulse and associated ionization levels are marked for generating photons of energy 285 eV and 330 eV. The rapidly varying ionization on the leading edge of the pulse leads to transient phase matching; thus, it is more preferable to phase match near the peak of the pulse where the conditions are more stationary. However, this corresponds to ionization levels of \( \approx 40\% \) and 90\% in He and Ne respectively.

In the simple picture of quasi phase matching using modulated waveguides, the modulations induce an oscillation in the laser intensity and therefore harmonics in the cutoff region are generated only in those sections of the waveguide where the laser intensity is highest. However, experimentally as can be seen in Figs. 4 – 7 and elsewhere, the harmonic emission is enhanced throughout a large photon energy range when using modulated waveguides. This broad range of enhancements can be explained by the calculations shown in Fig. 9. Figure 9 plots the instantaneous phase mismatch present during the time the laser pulse is generating harmonic orders 131, 161 and 191 in Neon. The lower-order harmonics can be phase matched using m=1 and m=3 QPM, while the higher-order harmonics are phase matched using the same modulated waveguide, but using m=3 and m=5 QPM. This explains why we observe enhancements over a broad photon energy range using QPM.

The maximum enhancement of HHG that can be achieved by quasi-phase matching depends upon how strongly the harmonic light is absorbed by the gas medium. Fortunately, at higher photon energies, the absorption of the gas decreases significantly. Figure 10 plots the absorption length, \( L_{\text{abs}} \), as a function of photon energy for Ne and He. For higher photon energies around the
C-edge (284 eV), the absorption length is quickly increasing, and reaches several meters in 100 torr helium. Thus, conversion at these very high photon energies is very strongly limited by phase-matching considerations, and not by absorption as is the case for the lower harmonic orders. For quasi-phase matching, the optimal signal is obtained at that gas pressure where the m=1 quasi-phase matching condition is met. Previous calculations have shown that the phase matched flux is optimized when the medium length is > 3L_{abs} and that it saturates at around 10L_{abs}.[15] Under our typical experimental conditions, m=1 quasi-phase matching of the harmonics at the C-edge in He is achieved at a pressure of 9 torr (for a 0.25 mm modulated waveguide) and the absorption length at this pressure is L_{abs} = 1.64 m. Therefore, by simple estimates, the maximum signal enhancement possible would be \sim (1/m\pi)^2 (10L_{abs}/L_{coh})^2 (1/e)^10 \sim 80,000. With shorter modulation periods, the same signal enhancement could be obtained at a higher pressure using a shorter waveguide. However, enhancements this large are not feasible. Phenomena that will limit the effective length over which harmonics will be generated are: 1) power loss associated with the fundamental mode propagation; 2) losses associated with ionization of the gas; 3) dispersion of the waveguide, which can stretch the pulse in time and reduce the peak intensity; and 4) group velocity mismatch of the fundamental and high harmonic. Of these, ionization induced loss is the most significant—a simple calculation of energy deposited into ionization of the gas shows that this can consume >0.1 mJ/cm of propagation. In comparison, the attenuation constant for the EH_{11} mode for 800 nm light and a waveguide radius of 75 \mu m is \alpha = 0.323 m^{-1}. Dispersion of the waveguide shows a propagation length of \sim 14 m for doubling the duration of the driving laser pulse. The group velocity walkoff is significant, but also relatively smaller.

Nevertheless, there are potential methods for ameliorating these issues and extending propagation lengths. The waveguide can be tapered to slowly focus the remaining light and make-up for losses.[27] Or a “square” pulse can be used, with a fast rise time but a longer time duration—ionization will eat the leading edge of the pulse but not immediately decrease the ultimate peak intensity. None of these concepts can be described through analytical modeling; however, numerical simulations can show the feasibility of many of these concepts.

It is currently not clear what the ultimate limit is for generating very high-energy photons using harmonic generation and quasi phase matching. Some limitations are fundamental, while others are technological. For example, the magnitude of the effective high-order susceptibilities of neutral atoms and ions for very high-order conversion has not been studied extensively. However, there is reason to believe that we are currently not near any limit. High harmonic generation has a very favorable linear scaling of cutoff photon energy with the laser intensity. In contrast, x-ray laser schemes typically scale in power requirements with the (hv)^{3-6} power. To generate very high-energy photons, it is necessary to generate the harmonics in higher stages of ionization of a gas, since neutral atoms do not survive to very high intensities. Recent experiments clearly show that the neutral atom “limit” is not as severe for high harmonic generation as was thought in the past.[32] Use of a waveguide can help to keep the laser light confined at high intensities, limiting defocusing that would otherwise occur due to generation of a plasma. This allows very high-order harmonics from ions to be observed. Other physical mechanisms, such as the relativistic motion of a solid surface[33] or of free electrons,[34] also can generate higher harmonics of the driving laser and have potential for extending nonlinear optics to even higher (KeV) energies.
Mechanisms for quasi phase matching

As mentioned previously, simply including a quasi-phase-matching wave vector in the analytical expression for phase mismatch is helpful in parametrizing QPM, but is also an oversimplification and leaves out the physics of how (and how much) the waveguide modulates the high-order harmonic emission. Exact modeling of quasi phase matching must be done using three-dimensional propagation of an optical pulse in a hollow waveguide. Numerical solution of the three-dimensional scalar wave equations written for the laser field \( E(r,t) \) and for the harmonic field \( E_h(r,t) \) in a local frame of reference, moving with the speed of light along the fiber axis (z), must be implemented. As an aid to understanding, it is useful to separately consider the physical processes that lead to modulation and enhancement of HHG in modulated waveguide structures. In terms of amplitude modulation, we know from propagation models that the effect of the diameter modulation is to change the on-axis intensity in a periodic way. In the case of the tightest modulation periods, the modulations of the waveguide radius occur over too short a distance for the laser mode to adiabatically follow the waveguide diameter. Instead, the change in diameter changes the boundary conditions, exciting a transverse wave that propagates to the center of the waveguide. When the transverse wave reaches the center of the waveguide, the interference causes oscillations in the intensity. The amount by which the intensity is modulated can be approximately estimated by assuming the adiabatic case where the peak intensity is proportional to the inverse of the waveguide radius squared.

The periodic modulation of the on-axis laser intensity has several different effects on the buildup of harmonic signal over the length of the waveguide. The most dominant effect occurs for the cutoff harmonics that are generated at the very peak of the laser pulse. These harmonics will only be generated in the higher intensity regions (i.e. near the axis of the waveguide), effectively turning on and off the generation. However, for the lower energy harmonics, the effect of modulating the laser intensity is more complicated. In a certain time-window during the leading edge of the pulse, the intensity modulations will also turn on and off the generation of a particular harmonic. It is interesting to note that for increasing harmonic orders, i.e. higher threshold intensities, the duration of this time-window increases.

The expected enhancement due to modulating the harmonic amplitude can be written as:

\[
E_{qo} \propto id_{\text{eff}}E_0^mLe^{i(\Delta k - K_m)L/2}G_m \frac{\sin(\Delta k - K_m)L/2}{(\Delta k - K_m)L/2} \tag{9}
\]

where Eqn. 9, when integrated over the length of the medium, is dominated by the term in the sum for which \( \Delta k \approx K_m \).[26] The quasi phase matching bandwidth is the same as the normal phase matching bandwidth shifted by the effective wavevector \( K_m \), so that using a longer medium with more modulated sections will narrow the bandwidth. In the case of periodically poled materials where the phase relation between the fundamental and harmonic is adjusted by 180 degrees every coherence length, the nonlinear coefficient can be expressed as a square-wave function, oscillating between +1 and -1 every coherence length, with the coefficients given by \( G_m = (2/m\pi)\sin(m\pi/2) \). In our case, the enhancement in harmonic field is half that for the case described above since we only enhance the signal every other coherence length. For the case of perfect quasi-phase matching (i.e. \( K_m = \Delta k \)), the enhancement is: \( I_q \propto (d_{\text{eff}}E_0)^2 (1/m\pi)^2 L^2 \). Thus, the enhancement in harmonic intensity is reduced by a factor of \((1/m\pi)^2\) compared to the case of
no phase mismatch. [26] The greatest enhancement will be from m=1 quasi-phase matching and will decrease for higher values of m.

Other physical mechanisms for quasi phase matching may also operate in the modulated waveguide. Typically quasi-phase matching is implemented by a modulation of the nonlinear coefficient. However, it is also possible to achieve an enhancement from quasi-phase matching by modulating the linear optical properties. Modulation of the laser intensity in the waveguide results in a modulation in the value of the phase mismatch, $\Delta k$, along the waveguide from the changing plasma dispersion, since the modulation in laser intensity directly modulates the ionization fraction. For a sinusoidal modulation in the intensity, the plasma contribution to the phase mismatch can be expressed approximately as –

$$\Delta k_{\text{plasma}}(z) = P N_{\text{atm}} e^{\delta \eta} \left[ \eta + \delta \eta \cos \left( \frac{2 \pi}{\Lambda} \right) \right]$$  \hspace{1cm} (10)

where $\delta \eta$ is the change in ionization and $\Lambda$ is the period of the waveguide modulations. To estimate the amount by which the ionization can change due to a modest laser intensity modulation, Fig. 11 shows the ionization as a function of time for two different laser intensities calculated from ADK ionization rates. At the peak of the pulse, the ionization fraction is 0.35 for an intensity of $10^{15}$ W/cm$^2$ but reaches a value of 0.41 when the intensity is increased by 5%. The value of $\Delta k_{\text{plasma}}$ therefore changes by 16%. This (and other similar mechanisms) provides another physical mechanism for enhancing the harmonic signal, because significant enhancement of the signal can be obtained if the phase mismatch itself varies sinusoidally.

Another mechanism for quasi-phase matching arises from the finite time response of the harmonic generation process, causing an intensity-dependent phase of the emitted harmonics. Due to this intrinsic phase, modulation of the intensity of the driving laser can periodically adjust the phase relationship between the harmonics and the fundamental laser. This intrinsic phase of the harmonics due to the phase that the electron acquires in its trajectory as a function of the release phase, $\phi_0$, and the return phase $\phi_f$ can be expressed as[6-8] –

$$\phi_{\text{intrinsic}} = \frac{I_p}{\hbar \omega} (\phi_f - \phi_0) + \frac{2U_p}{\hbar \omega} \int_{\phi_0}^{\phi_f} (\sin\phi - \sin\phi_0)^2 d\phi$$  \hspace{1cm} (11)

For the case of very high harmonic orders where the condition $U_p >> I_p$ is met, the first term in Eqn. 11 can be neglected, and the electron kinetic energy is approximately equal to the emitted photon energy. Using this assumption, the intrinsic phase as a function of harmonic energy can be calculated. For quasi-phase matching, it is important to know how the harmonic phase changes with laser intensity. Fig. 12 plots the trajectories for the intensity dependence of the harmonic phase for two different harmonic orders, assuming an 800 nm driving laser and the condition $I_p = 0$.[30] This simple calculation is based on a numerical calculation using the classical electron trajectories. The long trajectories have a linear intensity dependence equal to $-23 \times 10^{-14}$ Rads·cm$^2$/W. The short trajectories have a more complicated dependence on intensity, which is not exactly linear and is significantly smaller than the long trajectories. For the condition $I_p = 0$, the intensity dependence of the phase is not a function of harmonic order. For the long trajectories, an intensity change of $1.4 \times 10^{13}$ W/cm$^2$ would produce a phase shift of $\pi$, while the same phase shift for the short trajectories at the cutoff would require a change of $3.4 \times$
$10^{13}$ W/cm$^2$. This corresponds to an intensity change of 1.4 and 3.4 % respectively, using a driving laser intensity of $10^{15}$ W/cm$^2$. Therefore, the intensity-dependent quantum phase of the high harmonics does play a role in the quasi-phase matching of the emission of harmonics generated at the cutoff for intensities around $10^{15}$ W/cm$^2$ and above.

**Science using coherent short-wavelength light – Present and Future**

The high-order harmonic generation process can be used to implement compact soft-x-ray light sources with unique characteristics - femtosecond-to-attosecond duration pulses tunable throughout the UV and soft-x-ray regions of the spectrum, in a coherent beam, at multi-kHz high repetition rates, and perfectly synchronized to the driving laser pulse. Typical x-ray pulse durations range from tens of femtoseconds when driven with a 100fs pulse-duration laser,[35] to a few femtoseconds when driven with a 20fs laser pulse, to sub-femtosecond or attosecond when driven by 5fs laser pulses.[36, 37] The short pulse duration, tunability, and high repetition rate (multi kHz) of this source make it ideal for applications in spectroscopy. Because the EUV region of the spectrum corresponds to photon energies above the ionization potential of any neutral atom or molecule, intrinsic absorption resonances in atoms, molecules and solids can be used as a structural probe for materials characterization, or for monitoring ultrafast dynamic changes in electronic, chemical or catalytic systems. Moreover, because of the shorter wavelength of EUV light, it can be focused tightly. This enables a host of applications in nano-imaging, in lithography for integrated circuit fabrication, and for biological and materials microscopy. These applications currently can be pursued only at large-scale synchrotron facilities, with more limited time resolution. To date, techniques such as photoelectron spectroscopies have been used to measure excited-state electron dynamics in semiconductors and organic materials,[38] as well as to observe the site-specific position of atoms during ultrafast charge transfer reactions on catalytic surfaces[39] and molecular dissociation in gas phase reactions.[40]

New applications, that depend either on the coherent nature of the high harmonic source or its ultrafast time-duration, are also possible. Very recently, ultrafast photoacoustic spectroscopy has been extended into the EUV region of the spectrum by using a high harmonic beam as an ultra-sensitive probe of high-frequency oscillations in materials.[41] The EUV probe is more sensitive and also extends to a higher frequency range than is possible using visible lasers. Applications in high resolution coherent imaging are also possible.[42, 43] Moreover, the development of ultrashort-pulse light sources at short wavelengths makes it possible to probe fundamentally new types of electronic dynamics in atoms and molecules. One such category of experiment is time-resolved studies of high-energy processes associated with ultrafast femtosecond-to-attosecond electronic rearrangement in atoms and molecules. For example, recent demonstration experiments[44] have measured the inner-shell vacancy lifetime of the 4d electronic shell of atomic Xenon, which undergoes Auger decay. This yielded a time-resolved lifetime consistent with previous measurements of the natural linewidth of the transition. In the future, time-domain studies of ultrafast electronic inner-shell processes will be possible in molecular systems. Here, the nuclear motion and electron relaxation are coupled, and the measured decay linewidth is not uniquely related to the lifetime. Finally, the past five years have seen rapid progress in the ability to treat light as an electromagnetic waveform, by controlling all aspects of the amplitude and phase of a laser pulse. This in turn has made it possible to manipulate the internal dynamics of atoms and molecules on their fundamental, femtosecond-to-attosecond, time-scales. In atomic
systems, the ability to shape a laser pulse has made it possible to selectively enhance single harmonic orders generated during the process of high harmonic generation, by controlling the dynamics of the recolliding electrons to a precision of 12 attoseconds.[10, 11] In molecular systems, high harmonic generation from aligned molecules is leading to the development of new structural probes where the recolliding electron that generates the harmonics is used as an in-situ probe of molecular structure.[45, 46]

By extending harmonic generation to ever-shorter wavelengths, and by further increasing the efficiency, still more applications in science and technology will become possible. In the future, there are exciting prospects for developing compact x-ray microscopes and new generations of x-ray nanoprobe, as well as for control and manipulation of atoms, molecules and materials on their fundamental timescales from picosecond to attosecond.

**Acknowledgements**

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Bibliography


Figure and Table Captions

Table 1: This table lists the highest possible harmonic order generated at the given ionization level for the case when the given value occurs at the peak of the laser pulse (lower value) and the case when the given value occurs early in the pulse (upper value) for several pulse durations.

Figure 1: Ionization levels present in He at the peak of the laser pulse, for a laser pulse intensity that generates a given cutoff photon energy.

Figure 2: Schematic of a modulated waveguide for coherent x-ray generation using the process of high harmonic generation. The high harmonics are generated in the narrow regions of the waveguide where the laser intensity is highest. In between these narrow sections, the laser light and x-ray light can re-phase, so that x-ray light always contributes in phase with the existing x-ray beam. The hollow-core modulated waveguides used have modulation periods of 1mm - 0.25 mm, inner diameter of 150 µm, and modulation depths ≈ 10 µm.

Figure 3: Schematic of the pressure tuning curves (normalized) for straight and modulated waveguides, for coherent x-ray generation using high harmonic generation.

Figure 4: Harmonic emission from He at a pressure of 10 torr, from 10 cm long straight and modulated waveguides, with a periodicity of 0.25mm, and for a laser pulse duration of 20 fs. By using a modulated waveguide, the harmonic flux is enhanced by a factor of 5 at the carbon edge around 284 eV compared with a straight waveguide.

Figure 5: Harmonic emission from 15 torr of He, using 2.5 cm long straight and modulated waveguides. The modulation periods were 0.25 mm, but the pulse duration and ionization levels were slightly higher than for Fig. 4. Nevertheless, use of a modulated waveguide increases photon flux over the entire energy range, in particular for photon energies > 225eV, where no x-ray emission is observed in a straight fiber.

Figure 6: Harmonic spectra from straight and 0.5 mm modulated waveguides, for different pressures of Ne. All spectra were taken with a 10 s exposure time. Also plotted are the integrated counts for three different photon energy ranges as a function of pressure. The energy ranges are indicated on the spectra using dashed lines.

Figure 7: Harmonic emission from 11 torr of Ne, from 90 eV to 180 eV, for the case of straight and modulated waveguides (periodicity 0.25mm). Using the modulated waveguides, the efficient energy range for EUV generation is extended from 105 eV to 150 eV, while the cutoff is extended from 130 eV to 165 eV.

Figure 8: Calculation of the fractional ionization of Ne and He as a function of time during a laser pulse that fully ionizes Ne. The right axis plots the cutoff photon energy corresponding to the instantaneous pulse intensity.
**Figure 9:** Phase-mismatch for different harmonic orders during the laser pulse, for 9 torr of Neon at a laser intensity of $1.5 \times 10^{15}$ W/cm$^2$, and for a 20 fs pulse duration.

**Figure 10:** Plot of absorption length as a function of photon energy for 10 torr of Ne and for 100 torr of He. Data from the Center for X-ray Optics, LBL (www-cxro.lbl.gov).

**Figure 11:** Comparison of ionization fraction of neon as a function of time for a 22 fs duration pulse at different peak intensities.

**Figure 12:** Plot of the intensity dependence of the harmonic phase for the 25$^{th}$ and 45$^{th}$ harmonic orders. The calculation was done for a fundamental wavelength of 800 nm.
<table>
<thead>
<tr>
<th>Gas species</th>
<th>Critical ionization</th>
<th>1 ps</th>
<th>25 fs</th>
<th>20 fs</th>
<th>10 fs</th>
<th>5 fs</th>
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</thead>
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<td>76-115</td>
<td>83-122</td>
<td>94-157</td>
<td>100-272</td>
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<td>54-69</td>
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<td>29-36</td>
<td>30-38</td>
<td>3046</td>
<td>36-70</td>
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<tr>
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<td>21-26</td>
<td>21-27</td>
<td>21-33</td>
<td>22-48</td>
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<tr>
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<td>15-19</td>
<td>16-20</td>
<td>17-23</td>
<td>19-30</td>
</tr>
</tbody>
</table>

**Table 1**
Figure 1
Figure 2
Figure 3
Figure 4
Figure 5
Figure 6
Figure 8
Figure 9
Figure 10
Figure 11
Figure 12

Long Trajectories:
slope = -23 \times 10^{-14} \text{ Radians} \cdot \text{cm}^2/\text{W}