EXTREME NONLINEAR OPTICS: EXPOSING MATTER TO A FEW PERIODS OF LIGHT

Intense light pulses in the single-cycle regime have opened up new avenues in nonlinear optics. These, along with expected impacts on other areas of science and technology, are discussed.

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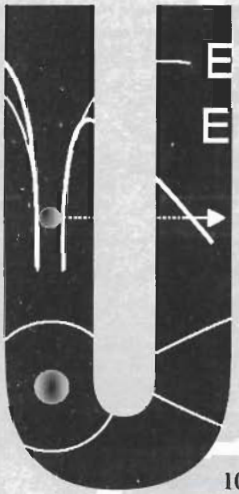

Ultrashort-pulse laser technology has progressed tremendously over the last 10 years. The invention of a new broad-band laser medium, titanium-doped sapphire,[1] a novel ultrafast mode-locking technique dubbed Kerr-lens mode-locking,[2] and a powerful device for ultrabroad-band dispersion control—chirped multilayer mirrors,[3]—for the first time opened the way to a reliable sub-10-fs laser technology.[4] Drawing on the concept of chirped-pulse amplification,[5] Ti:sapphire laser systems are now capable of generating pulses of around 20 fs in duration with millijoule energies (yielding peak powers on the order of hundreds of gigawatt, 1 GW = 109 W) at 1-kHz repetition rates,[6] and well beyond 100 mJ (resulting in multi-terawatt peak powers, 1 TW = 1012 W) in the 10-Hz regime.[7,8]

Self-phase-modulation in gas-filled capillary waveguides[9] can broaden the spectrum of millijoule-energy pulses to a supercontinuum extending from 400–1000 nm. The visible spectral components of this diffraction-limited radiation are displayed in the cover photo showing the beam exiting the capillary after being subjected to the angular dispersion of a prism. Ultrabroad-band chirped mirrors are able to compress the near-IR portion of this spectrum, resulting in the routine generation of 5-fs pulses with energies around 0.5 mJ and peak powers of 0.1 TW in a diffraction-limited beam.[10]

At somewhat lower energies (≈ 0.15 mJ), pulses as short as τ_p = 4 fs have also been demonstrated.[11] The pulse duration τ_p is defined as the length of the time interval confined by the instants at which the intensity envelope $I(t) \propto \langle E^2(t) \rangle$ drops to half the peak intensity of the pulse; the brackets denote averaging over the optical period.

Because the carrier wave oscillation cycle is $T_0 = 2\pi/\omega_0$ = 2.6 fs at the carrier wavelength of λ_0 = 780 nm, the electric and magnetic fields perform just one and a half oscillations within τ_p. These light "transients" are focusable to intensities that approach 10^{17} W/cm^2, yielding electric field strengths on the order of 10^{10} V/cm. Hence, electric field amplitudes comparable to or higher than the static Coulomb field experienced by bound electrons can be "switched on and off" within rise and fall times comparable to the oscillation period of the field. This capability now permits researchers to expose matter to extremely strong optical fields before being full ionized, and confines the effective interaction time to a single light period. The implementation of light-matter interactions under these extreme conditions is likely to significantly push the frontiers of nonlinear optics.

From linear to nonlinear optics

The electric field, $E(t)$, of a light wave polarizes an atomic specimen by slightly removing electrons from the nuclei along the direction of the field (see Fig. 1). At low intensities the induced polarization, $P(t)$, follows—with some phase retardation—the sinusoidal oscillation of a monochromatic wave $E(t) = E_0 \sin(\omega t)$. One of the most striking manifestations of this material response is the refraction of light rays upon entering a transparent dense medium at oblique incidence.

The ancient Romans were aware of the refractive power of lenses, and Al Hasan, the Egyptian natural scientist, proposed the use of this effect for correcting defective sight around 1000 A.D. Millions of people

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benefited from this invention before the underlying phenomenon was understood 900 years later.

Linear optics is based on Maxwell's wave equations and the material response $P(t) = \varepsilon_0 \chi E_0 \sin(\omega t + \varphi)$. Here ε_0 denotes the electric permeability of vacuum. The frequency-dependent material constants χ and φ are referred to as the magnitude and the phase of the electric susceptibility of the medium. This simple, but powerful theoretical framework accounts for innumerable natural phenomena of unparalleled variety and beauty, and constitutes the basis for understanding and improving the operation of numerous optical devices used in science, as well as in everyday life.

The linear relationship between P and E generally fails to give an accurate account of the medium polarization for external electric fields $E >> 1$ kV/cm. Rather, some nonlinearity in the response of material systems tends to become noticeable (see Fig. 1). Even a relatively weak nonlinearity of $P(E)$ may have striking consequences. Lasers are the only light sources capable of producing sufficiently high optical fields to induce an appreciable nonlinear response of materials.

Even though its impact on our everyday life is by far inferior to that of linear optics, nonlinear optics has established itself as an important research field over the last four decades, benefiting several areas of science and technology. Perhaps the most important practical implications of optical nonlinearities arise from the possibility of converting coherent laser radiation to new frequencies. Frequency conversion in suitable transparent optical materials (*e.g.,* dielectric solids or gases) allows researchers to develop coherent light sources with a broad tuning range, and/or at frequencies where no lasers are available. For photon energies much smaller than the energy gap between populated and the lowest unpopulated excited states, the polarization response of these materials can be approximately written as

$$P(t) = \varepsilon_0[\chi^{(1)}E(t) + \chi^{(2)}E(t)^2 + \chi^{(3)}E(t)^3 + \ldots]. \qquad (1)$$

The expansion is valid for weak nonlinearities, *i.e.,* at moderate field strengths. The contributions to the induced polarization rapidly decrease with increasing order of the nonlinear susceptibility $\chi^{(n)}(n \geq 2)$. This is the regime of perturbative nonlinear optics. The second order term in Eq. 1 gives rise to second harmonic generation (*i.e.,* radiation at 2ω).[12] The third-order contribution may result in third-harmonic generation and accounts for the intensity-induced changes of the refractive index (optical Kerr effect). The fourth-order term may lead to fourth-harmonic generation, etc.[13]

Efficient exploitation of nonlinearities of increasing order calls for an increasing laser field amplitude, E_0. The corresponding increased amplitude of the atomic dipole oscillation, in turn, greatly enhances the probability for the onset of a quantum mechanical effect, namely the liberation of an electron from its atomic bound state. This process is referred to as photoionization and sets a limit to the maximum laser intensity in a broad class of nonlinear optical interactions. In gas, photoionization depletes neutral atoms and thereby strongly reduces the polarizability of the medium. In solids, photoionization triggers an avalanche of electron impact ionization, leading to irreversible damage by optical breakdown.[14]

The availability of intense light pulses in the single-cycle regime now permits researchers to expose matter to extremely strong optical fields before being fully ionized, and to confine the effective interaction time to a single light period. This capability has several far-reaching implications.

For the first time, solids can be exposed to light intensities in excess of 10^{14} W/cm^2, without damage.[15] The corresponding optical fields are high enough to make the terms of different order comparable in the expansion of Eq. 1. Thus, the perturbative approach to the description of the material response breaks down even in the absence of resonances. Extending reversible nonlinear optics in transparent optical media (dielectrics) into the nonperturbative regime, where the traditional description drawing on Eq. 1 fails, holds promise for the exploration and exploitation of highly nonlinear optical processes never before seen in solids under reversible conditions.

In gas, the nonperturbative regime was recently accessed. Atoms can now be irradiated with much higher intensities before ionizing (up to several times 10^{15} W/cm^2), permitting researchers to generate coherent harmonic radiation up to significantly higher orders (*i.e.,* shorter wavelengths) than was previously possible, with longer laser pulses ($\tau_p \geq 100$ fs). Furthermore, theorists predict the emergence of individual soft x-ray pulses of durations much shorter than a femtosecond from gas targets illuminated with intense quasi-single-cycle laser pulses.

Optical tunnel ionization

As the laser intensity approaches or exceeds 10^{14} W/cm^2, photoionization rapidly becomes significant and can be thought of as a quasi-static tunneling process (see Fig. 2a, page 48). The laser electric field (henceforth, laser field) temporarily suppresses the static Coulomb field of the atomic core and induces a tunnel current in the direction of the instantaneous laser field that adiabatically follows the variation of the applied field. The liberated electrons are accelerated in the laser field and can collide with either their parent or surrounding ions. These elementary processes form the basis of two of the most promising concepts for compact coherent x-ray sources, namely high harmonic generation (HHG)[16, 17] and electron-excited x-ray lasers (EEXL),[18] respectively.

If the intense laser pulse consists of many optical cycles, ionization takes place over many optical cycles and tends to deplete the ground state of the atoms by the time the laser intensi-

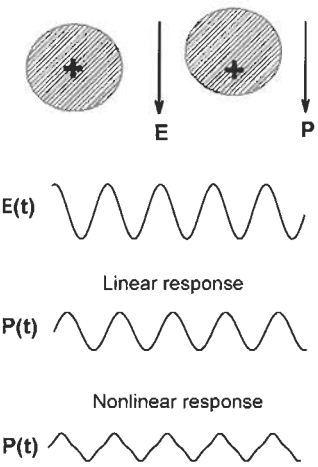

Figure 1. An external electric field tends to separate the negatively charged electrons (whose distribution is represented by the shaded area) from the positively charged nucleus (+), resulting in polarization $P(t)$ of the atom. For low and moderate fields varying sinusoidally in time, the atomic polarization follows, with some possible phase retardation, the oscillation of the electric field. With increasing field strength, however, the atomic response tends to become nonlinear, leading to a nonsinusoidal periodic evolution of $P(t)$.

ty peaks. By contrast, using few-cycle laser pulses, the process of ionization is confined to a fraction of a single oscillation cycle with both a circularly and linearly polarized field (used for EEXL and HHG, respectively), as revealed by Figure 3. Furthermore, the electrons are released into significantly higher fields than in the interaction with longer pulses comprising many cycles. EEXLs are expected to benefit from the improved energy coupling efficiency into an optically ionized plasma because the required high intensities can be achieved at much lower energy levels. HHG will be improved by both the strong temporal confinement of ionization and the higher fields the released electrons experience, as will be discussed in more detail below.

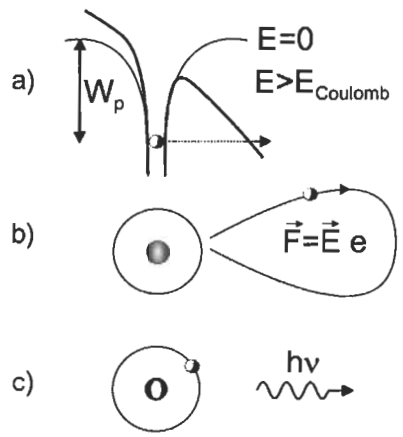

Figure 2. High harmonic generation can be understood in terms of three subsequent processes, (a) tunneling of the most weakly bound electron through the barrier formed by the atomic Coulomb field and the laser field, (b) acceleration of the quasi-free electron in the linearly-polarized laser field, and (c) recombination of the electron re-encountering its parent ion into its original bound state. The XUV emission spectrum reveals discrete (odd) harmonics of the laser radiation if this process is repeated quasi-periodically over many optical cycles.

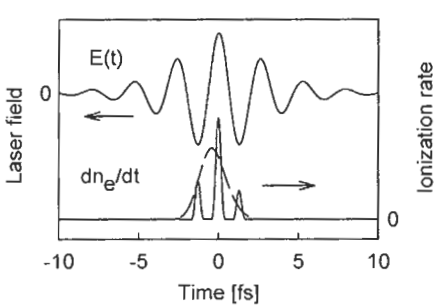

Figure 3. Upper trace: evolution of the electric field $E(t) = A(t)\cos(2\pi t/T_0)$ for a Gaussian pulse at $\lambda_0 = 780$ nm ($T_0 = 2.6$ fs) with a duration of 4 fs, representing approximately the shortest light wave packet generated to date. Lower traces: ionization rate induced in He by the 4-fs pulse with circular (dashed line) and linear (full line) polarization at a peak intensity of 2×10^{15} W/cm^2. A substantial fraction of the helium atoms are ionized by the pulse.

Coherent XUV generation by high harmonics

The most essential characteristics of HHG are accounted for by a semiclassical single-active-electron model [19, 20] In the first step, the electron tunnels through the barrier are formed by the atomic Coulomb potential and the laser field (see Fig. 2a). The quasi-free electron subsequently acquires kinetic energy from the laser field (see Fig. 2b). Depending on the phase of the field at the moment of tunneling, the electron may be brought back to its parent ion some half an optical cycle later, provided that the laser field is linearly polarized. Upon reencountering the ion, the electron may recombine, with some small probability, into its original bound state, emitting a photon with an energy $\hbar\omega$ equal to the ionization potential of the atom W_p plus the kinetic energy gained in the laser field (see Fig. 2c).

The electron passes the nucleus with maximum kinetic energy if it is "born" at a phase $\omega_0 t = 17°$ after the peak of the field, and this kinetic energy is approximately given by $3.2 \times U_p$, where U_p is the cycle-average oscillatory energy of the electron. As a consequence, the highest XUV photon energy that can be emitted upon recombination is given by

$$(\hbar\omega)_{max} \approx W_p + 3.2\ U_p. \qquad (2)$$

The spectrum of the emitted radiation generally extends from the laser frequency ω_0 to that given by the cut-off law implicit in Eq. 2 and is broken up into discrete harmonics of ω_0. Harmonics appear because the above described elementary process is repeated quasi periodically with the laser frequency (for pulses much longer than the oscillation period). In a laser pulse, U_p is time-dependent and, in units of electronvolts, is given by

$$U_p(t) \approx 9.3 \times 10^{-14} I(t)\lambda^2, \qquad (3)$$

where $I(t)$ is the intensity of the laser pulse in W/cm^2 and λ is the laser (carrier) wavelength in μm. For sufficiently high peak intensities, U_p in Eq. 2 is limited by the depletion of the neutral atoms due to ionization. Its maximum value is determined by inserting the corresponding depletion intensity I_d in Eq. 2. For reasons discussed in the preceding section, I_d increases with decreasing pulse duration, resulting in higher-order harmonics.

So far we have discussed the response of an individual atom. Practically useful XUV photon fluxes can, however, only result from the collective emission of an ensemble. Because the individual atoms in the ensemble (by the experimentator referred to as the gas target) are driven by a (gently focused) spatially coherent laser beam, the collective XUV emission of the ensemble yields a well-collimated XUV beam propagating collinearly with the laser beam. The harmonic yield critically depends on the propagation length over which contributions from individual atoms to the harmonic signal constructively interfere. The usable interaction length is limited by a difference of $\Delta\upsilon_n$ between the phase velocities of the fundamental and the nth harmonic wave. This difference gives rise to a phase mismatch $\Delta\varphi_n(z) \approx (2\pi/\lambda_n)(\Delta\upsilon_n/c)z$ between the partial harmonic waves originating from atoms at z and those at the entrance of the gas target ($z = 0$), where λ_n is the wavelength of the nth harmonic. The propagation distance that leads to $\Delta\varphi_n(z) = \pi/2$ is often referred to as the coherence length for the nth harmonic

$$L_n = (\lambda_n/4)(c/\Delta\upsilon_n), \qquad (4)$$

and provides the scale length for a coherent growth of the harmonic radiation. As Figure 4 shows, for $z > L_n$, the harmonic output tends to saturate and may even decrease as z further increases. The maximum harmonic photon yield scales with the square of L_n,

$$(P_n)_{max} \propto L_n^2. \qquad (5)$$

If the peak power of the laser pulse is high enough to reach the required intensity levels by a weak focusing of the beam, the dominant contribution to $\Delta\upsilon_n$ originates from free electrons produced by ionization. Because higher-order harmonics emerge at larger background electron densities (due to the higher fields required), the achievable coherence length L_n tends to decrease super-

linearly with the harmonic wavelength λ_n. Consequently, the efficiency of high harmonic production rapidly decreases with increasing harmonic order (or decreasing harmonic wavelength).

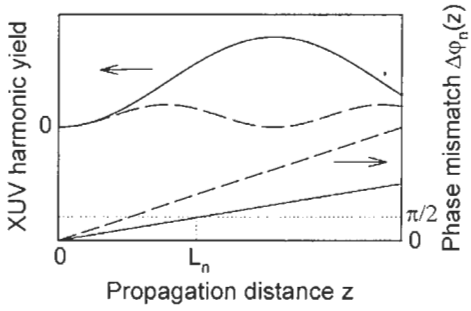

Figure 4. Variation of the XUV harmonic photon yield (at the n^{th} harmonic) and phase mismatch between harmonic radiation emitted by atoms at z and that emitted at $z = 0$ (entrance of the medium) as a function of the propagation distance across the gas target. A factor-of-two faster dephasing (or shorter coherence length L_n) implies a factor-of-four smaller maximum XUV yield of the collective emission.

The role of the pulse duration

Exploiting the above described phenomena, harmonics of 800-nm laser radiation up to the 111^{th} order (resulting in photon energies in excess of 150 eV and wavelengths below 7.5 nm) were generated by irradiating neon gas with 125-fs laser pulses at an intensity of 10^{15} W/cm^2.[16] In spite of numerous attempts, neither the harmonic photon energy nor the yield could be notably enhanced for several years after this experiment. This was because the pulse duration ($\tau_p \approx 100$ fs) has set a firm upper limit to the depletion intensity I_d and a lower limit to the minimum free-electron density accompanying the emergence of a given harmonic, thereby limiting the highest harmonic photon energy (according to Eqs. 2 and 3) and the harmonic photon yield (according to Eqs. 4 and 5). Shorter laser pulse durations allow "switching on" the high fields more abruptly and should thus give rise to an increased depletion intensity and a decreased background free electron density coupled to the appearance of a given harmonic. In fact, recent experiments[21–23] with laser pulses of $\tau_p \leq 25$ fs resulted in the generation of harmonics beyond the 300^{th} order of 800-nm light in helium gas, corresponding to photon energies of ≈ 0.5 keV and wavelengths shorter than 3 nm (see Fig. 5). These experiments constitute the first demonstration of a laboratory source of coherent X-rays at wavelengths below the K absorption edge of carbon (4.4 nm) in the so-called water window, which constitutes a spectral region of great importance for biological microscopy.

A single attosecond XUV pulse emerging

If the laser pulse duration approaches the optical period, a qualitatively new regime of strong-field laser-atom interactions is entered. Due to the pronounced nonlinear dependence of the tunneling rate on the electric field, the process of ionization is temporally confined to a single oscillation cycle as shown in Figure 3. An obvious implication is the coalescence of high harmonics to an XUV continuum (see Fig. 5). Another consequence relates to the temporal evolution of coherent XUV emission. Figure 3 reveals that electrons are liberated within a single laser period. Only a small fraction of the freed electrons are launched into trajectories that bring them back to the ion with nearly maximum kinetic energy ($\approx 3.2\ U_p$). Because they are emitted in a narrow phase interval, XUV emission at the highest photon energies (shortest wavelengths) will be confined to a small fraction of the optical period. This expectation is confirmed by the results of computer simulations[24] (see Fig. 6, page 50). The full line depicts the electric field $E(t) = A(t)\cos(\omega_o t + \psi)$ of a 5-fs pulse with Gaussian envelope for $\psi = 0$. The larger narrow spike some 500 as (attoseconds, $1 = 10^{-18}$s $= 10^{-3}$ fs) after the peak of the laser pulse ($t = 0$), represents the instantaneous intensity of the XUV emission [$\alpha\ E_{XUV}(t)^2$] near the shortest emitted wavelength at $\lambda = 3.2$ nm within an $\approx$ 10% bandwidth attosecond emerging from helium irradiated with a 5-fs pulse at a peak intensity of 2×10^{15} W/cm^2.

The confinement of the coherent emission at the high-energy end of the XUV spectrum to a single burst of attosecond duration is a general feature of HHG from gases illuminated by laser pulses consisting of a few optical cycles.[25, 26] This behavior is a consequence of the quasi-single-cycle excitation and the fact that the electric field rather than the intensity envelope governs the interaction, which is typical of the nonperturbative regime.

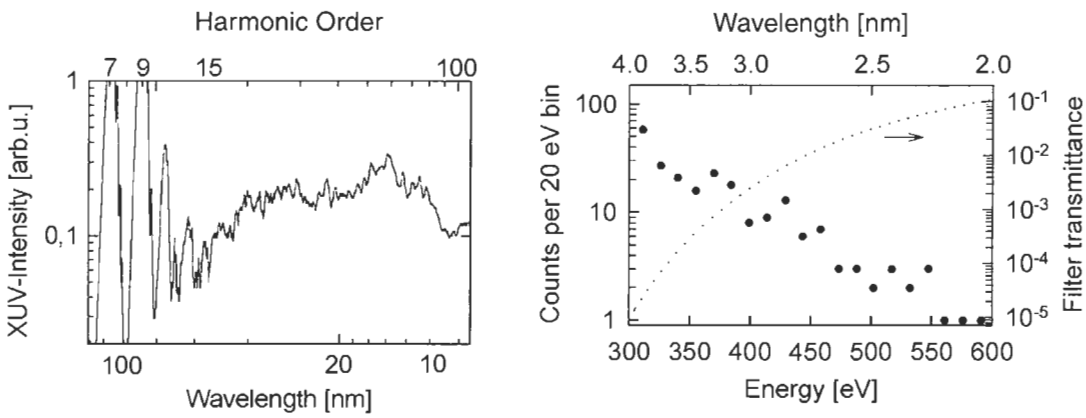

Figure 5. XUV spectra emitted by He illuminated with 5-fs pulses (λ_o 0.8 μm) at a peak intensity of 4 x 10^{15} W/cm^2. The left diagram shows the long-wavelength spectrum recorded with a grating spectrograph. Discrete harmonics merge to a continuum for $\omega/\omega_0 > 15$ as a consequence of the confinement of the relevant interaction time to a single optical period.[26] The diagram on the right shows the photon energy spectrum of the short-wavelength (cut-off) region as transmitted through a filter (dotted line).[27] This spectrum was recorded with an energy-dispersive spectral apparatus. The rapidly decreasing yield with increasing energy is a consequence of a rapidly decreasing coherence length L_n with wavelength λ_n (see Eqs. 4 and 5).

Using a laser pulse in the few-cycle regime is therefore the simplest conceivable implementation of the concept of an "atomic Pockels cell"[27] for selecting a single attosecond XUV pulse from a train, which would be produced by longer laser pulses.[28]

Does the phase of light matter

As the duration of the pulse $E(t) = A(t)\cos(\omega_0 t + \psi)$ approaches the oscillation period $T_0 = 2\pi\backslash\omega_0$, the evolution of the field tends to be significantly affected by the phase ψ of the carrier even if the amplitude envelope $A(t)$ remains unchanged. This is evident from the upper diagram of Figure 6, which shows $E(t)$ for two different values of the carrier phase ($\psi = 0$: cosine-carrier; $\psi = \pi\backslash 2$: sine-carrier) and the same $A(t)$ describing a chirp-free Gaussian pulse with a duration of 5 fs at a carrier wavelength of 800 nm. $A(t)$ changing notably within an oscillation cycle gives rise to a significant phase-sensitivity of $E(t)$. The fact that nonperturbative interactions are controlled by the electric field of radiation rather than the intensity envelope now implies that, for the first time in nonlinear optics, *the (absolute) phase of a lightwave tends to influence the polarization response of the illuminated medium.*

The XUV harmonic emission from gases beautifully exemplifies the important role of the carrier phase. In fact, Figures 6 and 7 reveal a dramatic sensitivity of the high-photon-energy x-ray emission of a 5-fs-pulse-driven coherent harmonic source to the phase. Both the harmonic yield and the temporal evolution of the highest-frequency radiation are strongly affected by ψ. At present, we do not have experimental access to the carrier phase ψ, which is subject to random variations at the output of femtosecond laser oscillators.[29] Controlling ψ will be a prerequisite for stable attosecond pulse generation.

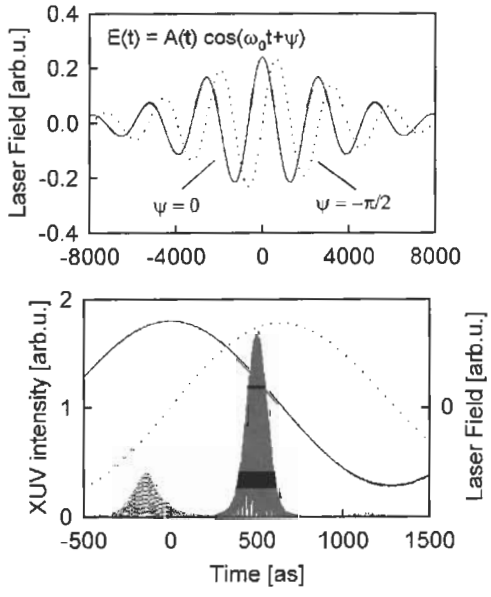

Figure 6. Upper diagram: electric field versus time in a Gaussian 5-fs pulse carried at a wavelength of 800 nm for two different values of the carrier phase ψ. Lower diagram: temporal evolution of the instantaneous XUV intensity [$\propto E^2_{XUV}(t)$] from He irradiated at a peak laser intensity of 2×10^{15} W/cm^2 within an ~10% bandwidth at $\omega \approx 250\ \omega_0$, *i.e.* $\lambda \approx 3.2$ nm for $\psi = 0$ and $\psi = \pi/2$. The cosine-carrier generates a single attosecond pulse ($\tau_p \approx 150$ as) peaking to ~ 500 as after the peak of the laser pulse. The sine carrier generates a much weaker XUV pulse of similar duration some 100 as before the peak of the laser pulse and a second, much weaker, one ~ 1,200 as behind the laser peak.

Extreme nonlinear optics now provides a possible route to the development of the first compact laboratory source of coherent soft X-ray radiation extending to the water window (2.3–4.4 nm). Radiation in this spectral range confined by the K absorption edges of oxygen and carbon offers a unique tool for the study of microscopic biological specimen (all of which contain carbon) in their aqueous surroundings with high contrast and resolution. So far, *in vivo* X-ray biological microscopy has had to rely on synchrotron radiation in large scale facilities, which strongly limited progress and proliferation of this important research. A powerful laser-driven harmonic source emitting in the water window would undoubtedly make major impacts on biological and medical research. This same source with somewhat relaxed wavelength requirements (5–20 nm) might enable x-ray lithography to become the winning technology for paving the way toward nanometer scale electronics. In this longer wavelength range, even a solid-state harmonic source may become reality, exploiting the high resistance of dielectrics to damage in the few-cycle (sub-10 fs) regime.

The predicted attosecond x-ray pulses from a sub-10 fs-laser-driven harmonic source are expected to significantly extend the scope of several existing research areas, and are likely to open up new fields of research. Clearly, these pulses would offer the capability of triggering and tracking ultrafast electronic and nuclear motion with unprecedented precision. In particular, the quantum-mechanical dynamics of bound electrons in atoms and molecules, and chemical reactions involving light elements could be investigated in the time domain for the first time. Concentration of the x-ray emission in such a short time interval, together with the excellent focusability of the harmonic x-ray beam,[22] also implies the feasibility of peak intensities that may open up the way to an extension of nonlinear optics to the x-ray regime.

Furthermore, polarization control of the sub-10-fs-IR-driver pulse allows the emission of a sub-femtosecond electron burst with well-determined energy distribution.[30] The possibility of injecting an intense subfemtosecond electron burst into a plasma in a controlled manner is likely to benefit the development of laser-driven particle accelerators[31] as well as electron-pumped keV x-ray lasers.[32]

Prospects and challenges

Our emerging capability of inducing extremely nonlinear optical processes giving rise, among others, to the emission of photons with energies 300 times that of incident photons is likely to significantly impact several fields in physical and life sciences, as well as in technology.

Before these and other intriguing applications may come true, several challenges remain to be addressed, however. First of all, the current state-of-the-art of coherent x-ray generation in the laboratory, $\approx 10^6$ photons/s X-ray yield within a 10% bandwidth at the K-edge of carbon,[22] must be improved by at least two to three orders of magnitude to make these sources useful for a wide range of applications. To this end, the coherence length for high-order harmonic generation will have to be enhanced by adapting the concept of quasi-phase-matching, which was already successfully implemented with low-order harmonics, into the XUV regime. Attosecond science, on the other hand, will not be able to emerge before experimental techniques for the temporal characterization of attosecond XUV pulses are developed and successfully implemented. Last but not least, full control of the above discussed electric-

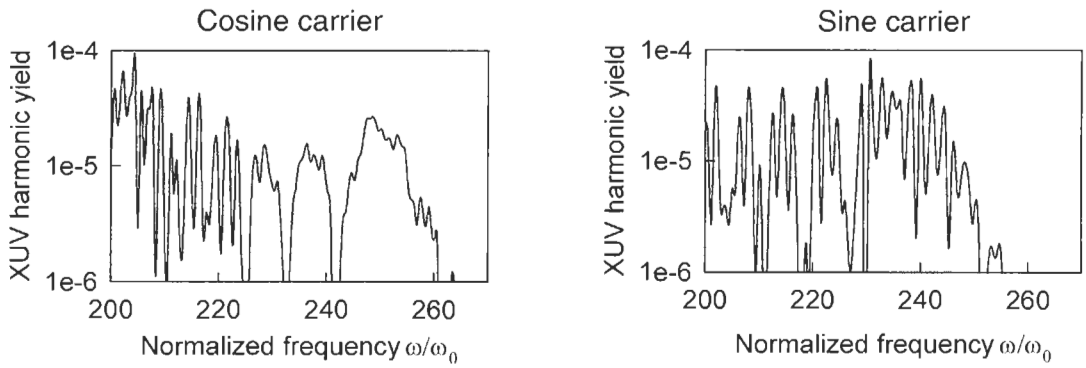

Figure 7. XUV harmonic emission spectra at the highest photon energies emitted for the parameters as listed in the caption of Figure 6 for a cosine carrier ($\psi = 0$) and a sine carrier ($\psi = \pi/2$). The strong phase-dependence of the XUV yield near cut-off translates into corresponding fluctuations of the XUV emission induced by non-phase-stabilized pulses. These "anomalous" fluctuations in the cut-off region were observed experimentally.[22]

field-governed (extreme) nonlinear optical processes calls for phase-stabilized sub-10-fs-laser-pulses.

These challenges and prospects impressively demonstrate that optics is still rapidly evolving and continues to impact other areas of science and technology.

Acknowledgments

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