that is responsible for such quantum-mechanical phenomena as electron diffraction through crystals. Likewise, the bound electron state possesses a quantum phase, which can change sign from place to place in the molecule. The quantum phases of the returning electron wave interfere with the different parts of the molecular bound state wave function, creating attosecond modulation of the electronic structure that is recorded in the shape of the HHG spectrum (Fig. 2).

The physics of HHG is still under active discussion, and this coherent scattering picture has many subtleties that are still unresolved. Nonetheless, the effect of molecular shape on the HHG spectrum is genuine and dramatic. For example, two simple molecules O_2 and N_2, are similar enough in their gross structure, each consisting of two atoms separated by about an angstrom, and a difference of only two electrons out of more than a dozen; but their highest occupied orbitals, the ones that will field-ionize in an intense laser field, have very different symmetries. HHG spectra reveal this difference, and recent experiments have used the spectra to perform tomographic reconstructions of the wave function responsible for ionization.

Conclusion

Only a few years ago, the direct measurement of transient phenomena lasting less than an optical cycle seemed a supreme challenge. Now scientists have brought together techniques from atomic and laser physics to make these measurements possible, if not yet routine. Attoscience is a new field, with the promise to reveal some of the fastest processes of chemistry and atomic physics, or to freeze motion that will allow us to view the structure of matter under extreme conditions. The subject is still dominated by attempts to understand and improve the sources, and to interpret the data, and there are still frontiers in source development: For example, attosecond pulses at subangstrom wavelengths have not been demonstrated. There are even more challenges in the theory of attosecond molecular dynamics: One of the most difficult issues yet to be resolved in attoscience is how we should describe the structure and motion of electrons and nuclei in physical chemistry and molecular physics to accommodate processes that are so fast that cherished concepts such as potential energy surfaces or the Born Oppenheimer approximation are no longer valid. Despite these difficulties, the rapid progress in attoscience over the past few years is not abating, and we may anticipate new physical insights in the coming decade.

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REVIEW

Attosecond Control and Measurement: Lightwave Electronics

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Electrons emit light, carry electric current, and bind atoms together to form molecules. Insight into and control of their atomic-scale motion are the key to understanding the functioning of biological systems, developing efficient sources of x-ray light, and speeding up electronics. Capturing and steering this electron motion require attosecond resolution and control, respectively (1 attosecond = 10^{-18} seconds). A recent revolution in technology has afforded these capabilities: Controlled light waves can steer electrons inside and around atoms, marking the birth of lightwave electronics. Isolated attosecond pulses, well reproduced and fully characterized, demonstrate the power of the new technology. Controlled few-cycle light waves and synchronized attosecond pulses constitute its key tools. We review the current state of lightwave electronics and highlight some future directions.

Quantum mechanics predicts the characteristic time scale for the rapidity of microscopic dynamics as $\Delta t \sim \hbar/\Delta W$, where ΔW is the spacing between the relevant energy levels of the microscopic system and $\hbar$ is Planck's constant. The milli–electron volt and multi–electron volt energy spacing of vibrational and electronic energy levels, respectively, imply that structural dynamics of molecules and solids as well as related chemical reactions and phase transitions evolve on a femtosecond time scale, whereas electronic motion on the atomic scale is to be clocked in attoseconds.

Before the invention of the laser, the resolution of time-resolved spectroscopy was limited by the nanosecond duration of pulses of incoherent light. The laser and the successive technological developments for the generation and measurement of ultrashort laser pulses improved the resolving power of pump-probe spectroscopy from several nanoseconds to several femtoseconds (*1*). The birth of femtosecond technology permitted real-time observation of the breakage and formation of chemical bonds (*2*).

We review the recent developments in the optical technology that have led to the breaking of the femtosecond barrier and provided real-time access to intra-atomic electron dynamics. Consequences include the observation of electronic motion deep inside (i.e., in inner shells of) atoms (*3*) and its control in real time (*4*). We address the underlying physical concepts and highlight the current status as well as future prospects of attosecond technology (*5*).

Femtosecond Technology: Control and Measurement with the Amplitude and Frequency of Light

Control and measurement of dynamics are intertwined. Time-resolved measurement relies on a physical quantity varying in a controlled, reproducible fashion on the relevant time scale.

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Femtosecond technology is the result of controlling the nonlinear polarization of matter with the amplitude of light. Controlling the absorption and/or refractive index in this way has yielded—along with group delay dispersion control—femtosecond pulses from laser oscillators by means of passive mode locking (*1*). Measurement of the pulses relies on the same physical effect: control of the nonlinear polarization of matter by a replica of the pulse in the presence of the pulse to be characterized (6, *7*).

The controlled, well-characterized evolution of the amplitude envelope and carrier-frequency sweep (chirp) of ultrashort laser pulses permits measurement (*2*) and control (*8*) of quantum transitions on a femtosecond time scale. Reliance on these cycle-averaged quantities implies that measurement resolution and control speed are ultimately limited by the carrier wave cycle. The carrier wave cycle period is about 3 fs in the near infrared, where low dispersion favors the generation of the shortest laser pulses.

Toward Attosecond Technology: X-ray–Pump/X-ray–Probe Spectroscopy?

Femtosecond measurement and control techniques utilizing nonlinear material response could, in principle, be extended into the attosecond time domain by using intense attosecond pulses of extreme ultraviolet (XUV) or x-ray light. Unfortunately, in these regions of the optical spectrum, the probability of two-photon absorption is prohibitively low. X-ray–pump/x-ray–probe spectroscopy and x-ray quantum control therefore rely on x-ray intensities that can be attained only with large-scale free-electron lasers (*9*). Even though these sources are expected to eventually deliver their radiation in subfemtosecond pulses (*10*) and XUV sources pumped by large-scale, high energy lasers have already pushed the frontiers of nonlinear optics to the range of several tens of electron volts (*11*, *12*), proliferation of attosecond technology and its widespread applications call for another approach—one that relies on light sources suitable for small laboratories.

An Alternative Route: Light-Induced Electronic Motion Within the Wave Cycle of Light

The electric field of visible, near-infrared or infrared (henceforth, referred to collectively as NIR) laser light, $E_L(t)$, exerts a force on electrons that varies on a subfemtosecond scale. The use of this gradient for initiating and probing the subsequent dynamics with attosecond timing precision and resolution has led to the emergence of an attosecond technology that does not rely on the existence of intense x-ray pulses.

According to theory, strong-field–induced ionization of atoms is confined to subfemtosecond intervals near the peaks of the oscillating NIR field (Fig. 1A), setting a subfemtosecond electron wave packet $\psi_{free}(t)$ free; this prediction was recently confirmed by real-time observation (*13*). The subfemtosecond ionization process, locked to the peak of NIR field oscillation with corresponding precision, may serve as a subfemtosecond "starter gun" for a wide range of dynamics (including electronic and subsequent nuclear motion and related dynamics of molecular structure).

The energetic radiation concurrent with optical-field ionization in a linearly polarized laser field (*14*) provides a means of triggering motion in a more gentle way, without applying an intense field. The broadband XUV light building up during the propagation of the driving pulse through an ensemble of atoms was predicted to emerge in a subfemtosecond burst within every half optical cycle that is sufficiently intense for ionization (*15*). In this Review, we focus on these photonic tools of attosecond technology, noting that the recolliding electron wave packet itself can also be used to explore dynamics (*16*–*18*).

The electronic (and concurrent) phenomena following the attosecond trigger can be tracked with comparable resolution with the use of the generating NIR field, the oscillations of which are synchronized with the trigger event (tunnel ionization, recollision, or XUV burst from recollision). Let us consider an electron ejected during the unfolding evolution of the system under scrutiny. In the presence of a linearly polarized NIR field, outgoing electrons collected along the direction of the laser electric field $E_L(t)$ (Fig. 1B) suffer a momentum change $\Delta p(t) = e\int_t^\infty E_L(t')dt' = eA_L(t)$. Here, e is the charge of the electron, t is the instant of its emission, and $A_L(t)$ is the vector potential of the laser field. This change varies monotonically within a half wave cycle of the laser field, mapping the temporal evolution of the emitted subfemtosecond electron wave packet to a corresponding final momentum and energy distribution of the emitted electrons. We recognize here a new embodiment of the basic concept of streak imaging, with the streaking controlled by the NIR field in this case. The electrons can gain or lose an energy of several electron volts depending on whether they have been emitted some hundred attoseconds sooner or later than the peak of the field cycle (Fig. 1B), endowing this light-field–driven streak camera with attosecond resolution (*19*, *20*).

The physical concepts and mechanisms outlined above have opened a realistic prospect of measuring and controlling electron dynamics on an attosecond time scale. However, full exploitation of the potential offered by this conceptual framework requires precise control over the applied laser field.

Steering Electrons with the Electric Field of Controlled Light Waves

In a laser pulse comprising many field cycles, the subfemtosecond electron and photon bursts born in the ionizing field emerge every half cycle, resulting in a train of subfemtosecond bursts (*21*). Its spectrum consists of equidistant lines that represent high-order odd harmonics of the driving field (*22*). The subfemtosecond pulse train and its multicycle driver constitute powerful tools for controlling and tracking electron dynamics that terminate within the NIR field oscillation cycle (*23*–*25*). The characteristics of the individual bursts in the train can be difficult to control and measure. Extension of attosecond control and metrology to a wide range of electronic phenomena (including all those extending in time over more than half an optical cycle) calls for the isolation of one to several attosecond pulses and precise control of their properties (*26*).

Isolation was predicted to be achievable by manipulating the polarization state (polarization gating) of the driving field (*27*) or shortening its duration to nearly a single cycle of the carrier wave (*28*). In fact, intense few-cycle laser pulses lasting several femtoseconds (*1*) led to the first observation of light lasting for less than 1 fs (*29*). Full control of the number and properties of bursts isolated, however, requires precise control of the driving electric-field waveform $E_L(t)$.

Intense few-cycle laser pulses with controlled waveform (*4*)—along with careful bandpass filtering of the highest-energy (cutoff) harmonics—have indeed permitted the reproducible generation of single and twin subfemtosecond pulses with cosine- and the sine-shaped electric field waveforms, respectively, as well as their reliable measurement by means of attosecond streaking (*30*, *31*) (fig. S1). The series of streaked electron spectra shown in Fig. 1C yields complete information about the driving laser field and allows determination of the temporal shape, duration, and a possible chirp of the emitted subfemtosecond XUV pulse (Fig. 1D) as well as the degree of its synchronism with its driver (*31*–*33*). These experiments reveal that few-cycle light with controlled waveform permits reproducible generation and complete characterization of subfemtosecond light. Owing to the attosecond synchronism between them, these tools allow attosecond metrology without the need for high-intensity x-rays.

Lightwave Electronics: Versatile Technology for Control and Chronoscopy on the Electronic Time Scale

Controlled light fields permit control of microscopic electric currents at the atomic scale just as synthesized microwave fields permit control of currents at the mesoscopic scale in semiconductor chips. By analogy to microwave electronics, we propose to name this new technology lightwave electronics. In marked contrast with previous quantum control through controlling transitions between quantum states with the amplitude and frequency of light (*8*), lightwave electronics gives way to controlling dynamics directly by the force that the electric field of intense light exerts on

electrons. This new approach is powerful in that (i) it provides a direct way of affecting the position and momentum of electrons and (ii) control is matched in speed to the electronic time scale.

The first notable manifestations of the power of lightwave electronics include controlling subfemtosecond XUV emission (*4*, *25*, *31*–*35*); molecular dissociation (*36*); measuring subfemtosecond XUV and electron pulses (*21*, *31*–*33*, *35*); and imaging dynamic changes in molecular structure (*16*–*18*) by means of steering electron wave packets with light fields. Sub–wave cycle (i.e., subfemtosecond) control of electron wave packet motion can be accomplished by mixing multicycle fields (*25*, *35*). However, full control over the entire system evolution from attosecond to femtosecond time scales—and, consequently, precision attosecond measurements—relies on optical waveforms that can be accurately controlled in shape and polarization state (*4*, *37*) and confined to several cycles. Exploitation of the full potential of lightwave electronics for attosecond control and metrology requires waveform-controlled broadband light.

The attosecond streaking spectrogram shown in Fig. 1C yields the complete history of photoelectron emission induced by a subfemtosecond XUV pulse as well as of the streaking optical field (*38*). Similar spectrograms can also be recorded with secondary (Auger) electrons, which uncover the history of a quantum transition of an electron deep inside an atom in real time (*3*). Attosecond streaking is not the only way of observing the temporal evolution of atomic-scale electron motion. Energetic excitation of atoms, molecules, or solids is often accompanied by the promotion of valence electrons to excited states (referred to as a shake up). The transient population of these states provides information about the instantaneous (electronic) state of the system. This population can be probed by means of attosecond tunneling induced by the time-delayed waveform-controlled few-cycle laser field (*13*).

In attosecond streaking and tunneling spectroscopy, the subfemtosecond XUV pulse serves as a pump and the controlled optical wave as the probe. Their roles can be interchanged: The optical field may—by means of tunnel ionization (*13*)—initiate an electronic process and control the unfolding dynamics (*36*), and the subfemtosecond XUV pulse may probe this process by, for example, photoelectron spectroscopy (*39*).

Attosecond technology based on a controlled optical wave and a synchronized subfemtosecond

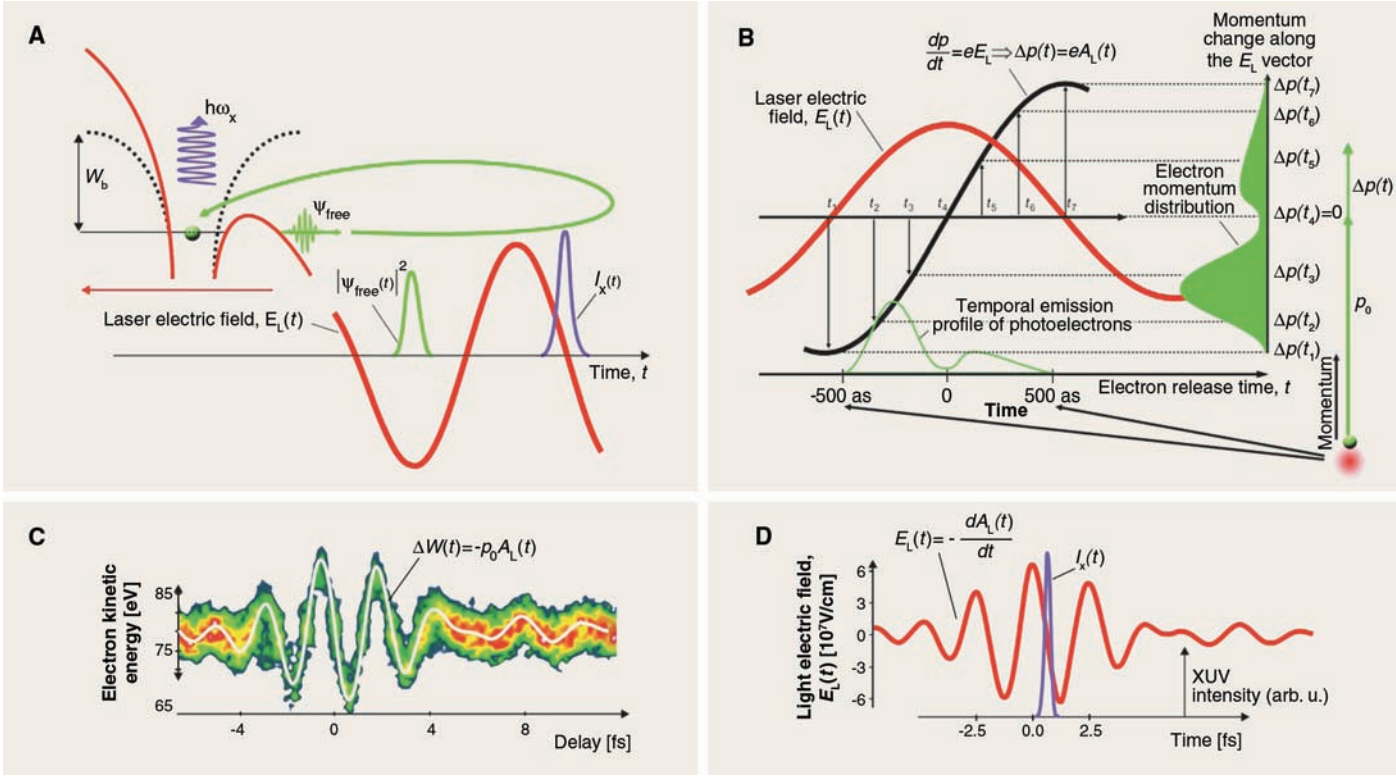


Fig. 1. The birth and measurement of a subfemtosecond (XUV) light pulse. (**A**) The field of a femtosecond NIR laser pulse, $E_L(t)$, is able to suppress the Coulomb potential in atoms sufficiently to allow a valence electron to tunnel through the narrow barrier and release a subfemtosecond wave packet, $\psi_{free}(t)$, near the peaks of its most intense oscillations. The wave packet is subsequently removed from the vicinity of the atomic core and less than a period later pushed back by the reversed field (green trajectory). Upon recollision, it interferes with the bound-state portion of the electron wave function. This interference results in high-frequency oscillations, emitting broadband XUV light. The highest-frequency portion, with intensity $I_X(t)$, is temporally confined to a small fraction of the optical period. (**B**) Concept of optical-field–driven streak imaging of electron emission from atoms. Electrons released by an XUV pulse parallel to the direction of electric field (red line) suffer a change in their initial momenta that is proportional to the vector potential of the field (black line) at the instant of release, mapping the intensity profile of the emitted electron and hence of the ionizing subfemtosecond XUV pulse into a corresponding final momentum and energy distribution of electrons. (**C**) Streaked spectra of photoelectrons released from neon atoms by a single subfemtosecond XUV pulse ($\hbar\omega_X \approx 95$ eV) recorded for a series of delays between the XUV pulse and NIR field (streaking spectrogram). The laser field causes only a moderate broadening (streaking) of the electron spectra; its main effect is the shift of the center of mass of the electron spectrum. In the limit of large initial kinetic energy of the electrons, this shift is linearly proportional to the vector potential of the field at the instant of impact of the XUV pulse (see white line). (**D**) Electric field of the NIR wave and intensity of the XUV pulse as retrieved from the streaking spectrogram shown in (C). The measurement confirms that the few-cycle waveform must be near cosine-shaped to warrant the production of a single subfemtosecond pulse and reveals a near-Fourier–limited XUV pulse of a duration of 250 attoseconds. arb. u., arbitrary units.

pulse is both versatile and sufficiently compact and affordable to allow proliferation in small laboratories. The core infrastructure required is sketched in Fig. 2. The scope of the technology largely depends on the characteristics and flexibility of its key tools: synthesized fields of laser light and attosecond pulses synchronized to them.

From Controlled Light Waveforms Toward Optical Waveform Synthesis

Lightwave electronics benefits from ever-broader optical bandwidth in several ways. Superposition of spectral components beyond an octave permits subcycle shaping of the light waveform and hence sculpting of the electric force on the electronic time scale for steering electrons in atomic systems. The increased bandwidth and improved dispersion control also lead to shorter optical pulses, which allow advancing subfemtosecond XUV pulse generation in terms of all relevant parameters: duration, intensity, and photon energy.

Intense few-cycle optical pulses (~5 fs) were first generated (*40*) by spectral broadening of femtosecond pulses (initially ~25 fs) in a hollow-core waveguide, and the resultant supercontinuum was compressed by reflection off of chirped multilayer dielectric mirrors (*41*) (Fig. 3A). Combined with waveform control (*4*), the technology matured to become a workhorse for attosecond science. Its current state is represented by the results summarized in Fig. 3, B and C. The spectrum of near–20-fs, 800-nm pulses is first broadened by self-phase modulation in a gas-filled capillary to a supercontinuum reaching from 400 to 1000 nm, followed by compression of the ~400-μJ pulses to ~3.5 fs with octave-spanning chirped mirrors (*42*). These pulses constitute the shortest controlled optical waveforms demonstrated to date.

Figure 3D shows how a single subfemtosecond XUV pulse can emerge from an atom ionized by a linearly polarized few-cycle light field. The green lines depict the kinetic energy, W_{kin}, with which different portions of the freed electron wave packet (ψ_{free} in Fig. 1B) return to the core as a function of return time, determining the energy of the emitted harmonic photon, $\hbar\omega = W_{kin} + W_b$, where W_b is the binding energy of the electron. For the optimum (near-cosine) waveform (Fig. 3D), we can estimate—in the limit of $\hbar\omega_X >> W_b$—the bandwidth over which photons of the highest energy (near the cutoff of the emitted spectrum depicted in violet) are emitted from one recollision only as

$$\frac{\Delta W_{\text{single-pulse}}}{\hbar\omega_X} \approx 0.6\left(\frac{T_{osc}}{T_{FWHM}}\right)^2 \qquad (1)$$

where T_{osc} and T_{FWHM} stand for the oscillation period (osc) of the carrier wave and the full width at half maximum (FWHM) duration of the laser pulse ionizing the atoms, respectively (*43*). Equation 1 underscores the importance of minimizing the number of cycles within the driver pulse. The intense sub–1.5 cycle ($T_{FWHM} < 1.5T_{osc}$) laser pulses that can now be produced with commercially available ingredients of ultrafast laser technology (Fig. 3, A to C) constitute a promising tool for pushing the frontiers of attosecond-pulse technology.

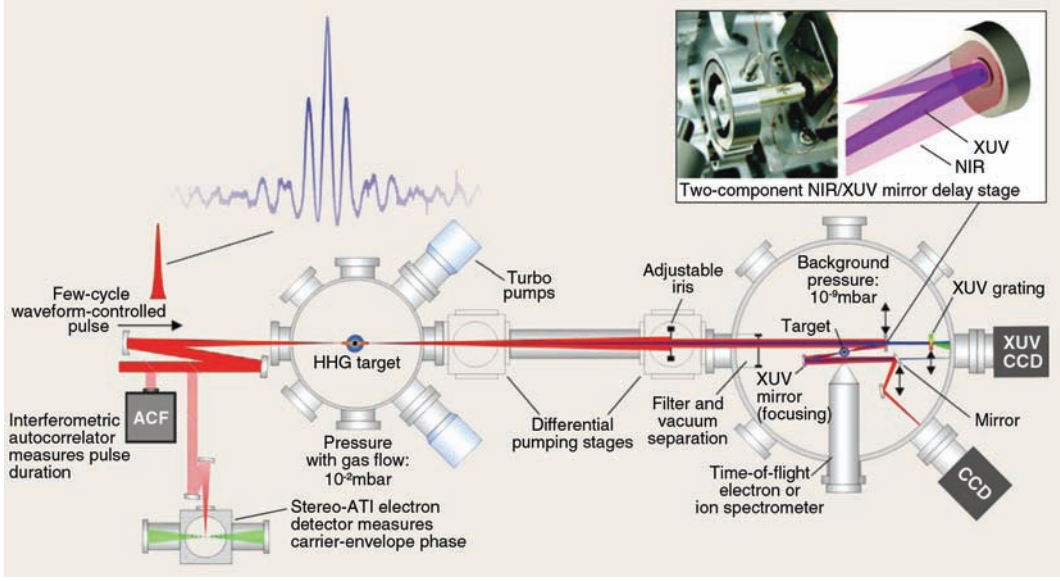


Fig. 2. Schematic of an experimental setup for attosecond-pulse generation and attosecond metrology as well as spectroscopy: the AS-1 attosecond beamline at MPQ. The intense, waveform-controlled few-cycle NIR laser pulse generates a subfemtosecond XUV pulse in the first interaction medium (jet of noble gas). The collinear XUV and NIR beams then propagate into a second vacuum chamber, where they are focused by a two-component XUV multilayer mirror into a gas target. The inner and outer part of the two-component mirror reflects and focuses the XUV and the (more divergent) NIR beam, respectively. By positioning the internal mirror with a nanometer-precision piezotranslator, the XUV pulse can be delayed with respect to the NIR pulse with attosecond accuracy. Analysis of the generated electrons and/or ions as a function of delay between the subfemtosecond XUV and the waveform-controlled NIR pulse permits characterization of the attosecond tools (Fig. 1) as well as real-time observation of atomic-scale electron dynamics in all forms of matter by means of attosecond streaking spectroscopy (*3*) and attosecond tunneling spectroscopy (*13*) with the same apparatus. In the former case, the final energy distribution of the outgoing electron is analyzed with a time-of-flight spectrometer; in the latter case, the observables measured as a function of delay are multiple-charged ions. ACF, auto-correlation function; CCD, charged-coupled device.

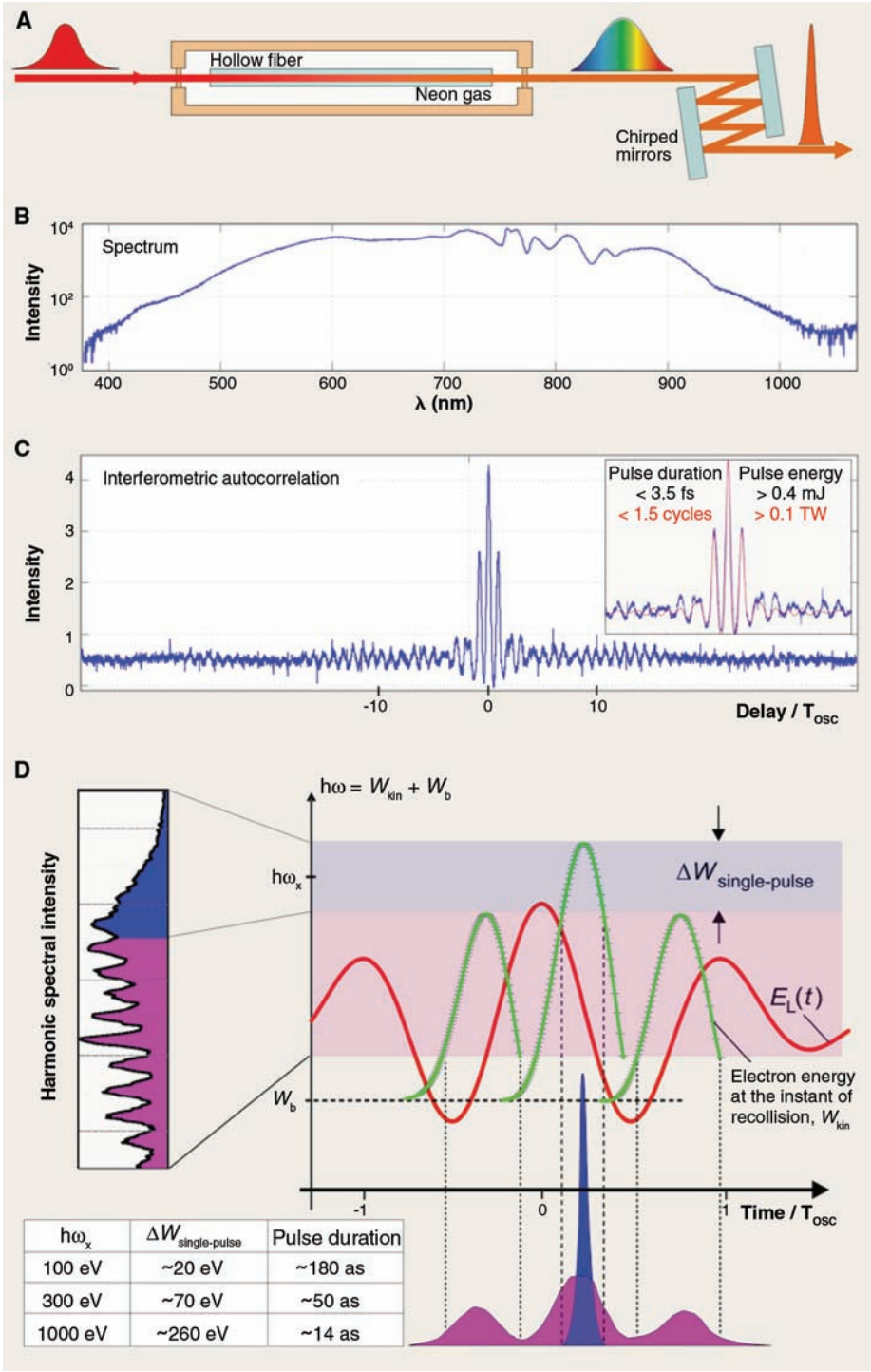
Advancing femtosecond technology to its ultimate limit is the key to extending the frontiers of both measuring and controlling dynamics with attosecond precision. Metrology benefits from broader spectral coverage and shorter duration of subfemtosecond pulses. The scope of control critically depends on the variety of waveforms, which is determined by the available bandwidth of intense coherent radiation. The shortest intense laser pulses also provide ideal conditions for generating the broadest supercontinua of coherent light, extending over more then three octaves (*44*). Advanced chirped multilayer mirror technology holds promise for versatile and scalable (multichannel) implementation of optical waveform synthesis. A prototypical three-channel synthesizer operational over the 1- to 3-eV band indicates the vast variety of possibilities for steering electrons in atomic systems with synthesized optical waves (fig. S2).

From Subfemtosecond Toward Attosecond Pulses

Equation 1 suggests that femtosecond technology must approach the monocycle limit to push the frontiers of XUV pulse generation toward and below 100 as. However, there are other options. Modulation of the polarization state of the ionizing NIR pulse (*33*, *37*, *45*) or of its amplitude by coherent addition of second harmonic radiation (*46*, *47*) can efficiently increase $\Delta W_{\text{single-pulse}}$ even by using several-cycle NIR driver pulses. Implementation of polarization gating with waveform-controlled, approximately two-cycle NIR pulses (*37*) has recently led to a spectacular achievement: the generation of isolated XUV

pulses at $\hbar\omega_{carrier} \approx 36$ eV with a bandwidth of $\Delta W_{single\text{-}pulse} \approx 15$ eV. Along with dispersion control and trajectory selection (*48*), these isolated pulses resulted in near–single-cycle XUV pulses that had a duration of 130 as (*33*) (see the low-energy streaking spectrogram in Fig. 4A).

The increased $\Delta W_{single\text{-}pulse}$ with several-cycle pulses comes, however, at the expense of efficiency because (i) only a small temporal fraction of the driver pulse participates in the generation process and (ii) the cycles preceding the generation moment pre-ionize the atoms, causing substantial depletion of the ground state before the XUV pulse can be emitted. The unprecedented confinement of electromagnetic energy into a single well-controlled oscillation of light in sub–1.5-cycle NIR pulses (Fig. 3) avoids these shortcomings and offers several more benefits. Indeed, the high contrast between the wave crests at $t = -T_{osc}/2$ and earlier ones in Fig. 3D permits—thanks to the exponential scaling of ionization probability with the field strength—adjustment of the pulse intensity so as to allow the electron to survive with a high probability in its ground state until $t = -T_{osc}/2$ (in Fig. 3D) and then to be set free with a high probability near this instant through tunneling. The resulting wave packet, ψ_{free} (Fig. 1A), is launched with unprecedented amplitude. Upon its return to the core at about $t \approx T_{osc}/4$, it interferes with the ground state portion of the wave function causing



$\hbar\omega_x$	$\Delta W_{single\text{-}pulse}$	Pulse duration
100 eV	~20 eV	~180 as
300 eV	~70 eV	~50 as
1000 eV	~260 eV	~14 as

Fig. 3. Toward intense, monocycle optical pulses. (**A**) Hollow-fiber/chirped-mirror optical pulse compressor for the generation of intense, few-cycle laser pulses. (**B**) The spectrum of millijoule-energy, near–20-fs, phase-controlled near-infrared laser pulses delivered at a repetition rate of 3 kHz from a Ti:sapphire laser (Femtopower, Femtolasers GmbH) is first broadened to more than an octave (~450 to 950 nm) in a neon-filled capillary (pressure ~ 2 bar, length ~ 1 m, bore diameter 250 μm). (**C**) The spectrally broadened pulses are compressed by octave-spanning chirped multilayer mirrors. The interferometric autocorrelation trace indicates a near–bandwidth-limited ~3.5-fs pulse carried at $\lambda_L \approx 720$ nm. (**D**) Energies of XUV photons emitted from an atom exposed to a linearly polarized 3.5-fs, 720-nm Gaussian laser pulse that is sufficiently intense for efficient tunnel ionization. The photon energies (green lines) are depicted as a function of the return time of the electron to the atomic core and have been obtained by analyzing classical electron trajectories (*43*). The width of the energy band within which only one recollision contributes to XUV emission determines the bandwidth $\Delta W_{single\text{-}pulse}$ available for the generation of an isolated attosecond pulse. Based on the classical trajectory analysis, $\Delta W_{single\text{-}pulse}$ is found to be maximum for a near–cosine-shaped waveform, $E_L(t) = E_0\varepsilon(t)\cos(\omega_L t + \varphi)$, with the carrier-envelope phase φ varying between $\pi/12$ and $\pi/6$ depending on the pulse shape and intensity. Here, E_0 and ω_L stand for the amplitude and angular frequency of the oscillations of the laser electric field, with $\varepsilon(t)$ being the amplitude envelope function. The table summarizes bandwidths over which the sub–1.5-cycle pulses are predicted to support the emergence of an isolated attosecond pulse at different XUV photon energies. The estimated pulse durations derive from the conservative assumption that no spectral components are available for the pulse synthesis outside the given spectral window.

high-frequency (soft–x-ray) dipole oscillations with unprecedented amplitude. Hence, the intense near–single-cycle waveforms appear to constitute ideal tools for the pursuit of powerful soft–x-ray pulses with pulse durations approaching the atomic unit of time (see table in Fig. 3D) and photon energies approaching the kilo–electron volt frontier.

A first indication of the potential that near–single-cycle NIR pulses offer for attosecond-pulse generation is provided by the high-energy streaking spectrogram in Fig. 4A. It has been recorded with sub–two-cycle (near–4-fs) NIR (750-nm) pulses and XUV pulses filtered by a molybdenum-silicon chirped multilayer mirror with a high-reflectivity band of ~16 eV (FWHM) centered at ~93 eV (Fig. 4B). The bandwidth (FWHM) of the bandpass filtered XUV light amounts to ~13 eV. The streaking spectrogram indicates isolated sub–170-as pulses that are near bandwidth-limited (*49*). The pulses contain more than 10^6 photons, which are delivered at a repetition rate of 3 kHz, implying a photon flux of $>3 \times 10^9$ photons per second transported in a near–diffraction-limited beam. With the Mo/Si mirror, which has a 10-cm focal length, used in the MPQ attosecond beamline AS-1 (Fig. 2), this beam can be focused to a spot diameter of several micrometers.

The recently demonstrated, waveform-controlled sub–1.5-cycle NIR pulses are capable of producing $\Delta W_{\text{single-pulse}} > 30$ eV at $\hbar\omega_X \approx 120$ eV (Fig. 4D), offering the potential for generating sub–100-as pulses at a wavelength of ~10 nm (*42*). To exploit this potential, researchers must develop ultrabroadband soft–x-ray multilayer mirrors with precisely controlled group-delay (GD) dispersion. Figure 4C plots the reflectivity and GD of the first chirped multilayer mirror design that supports sub–100-as pulses. Chirped multilayer mirror technology (both metallic and dielectric) is likely to play as important a role in advancing attosecond technology as it has played in advancing femtosecond technology to its ultimate limits.

Prospects

New research tools allow scientists to seek answers to questions that, in the absence of a means of addressing them, have never been posed before. Grand questions serve as a compass providing directions for research that promises the most extensive return. We pose a few questions across disparate areas of science, connected by the fundamental role that atomic-scale electronic motion plays in physical, chemical, and biological processes.

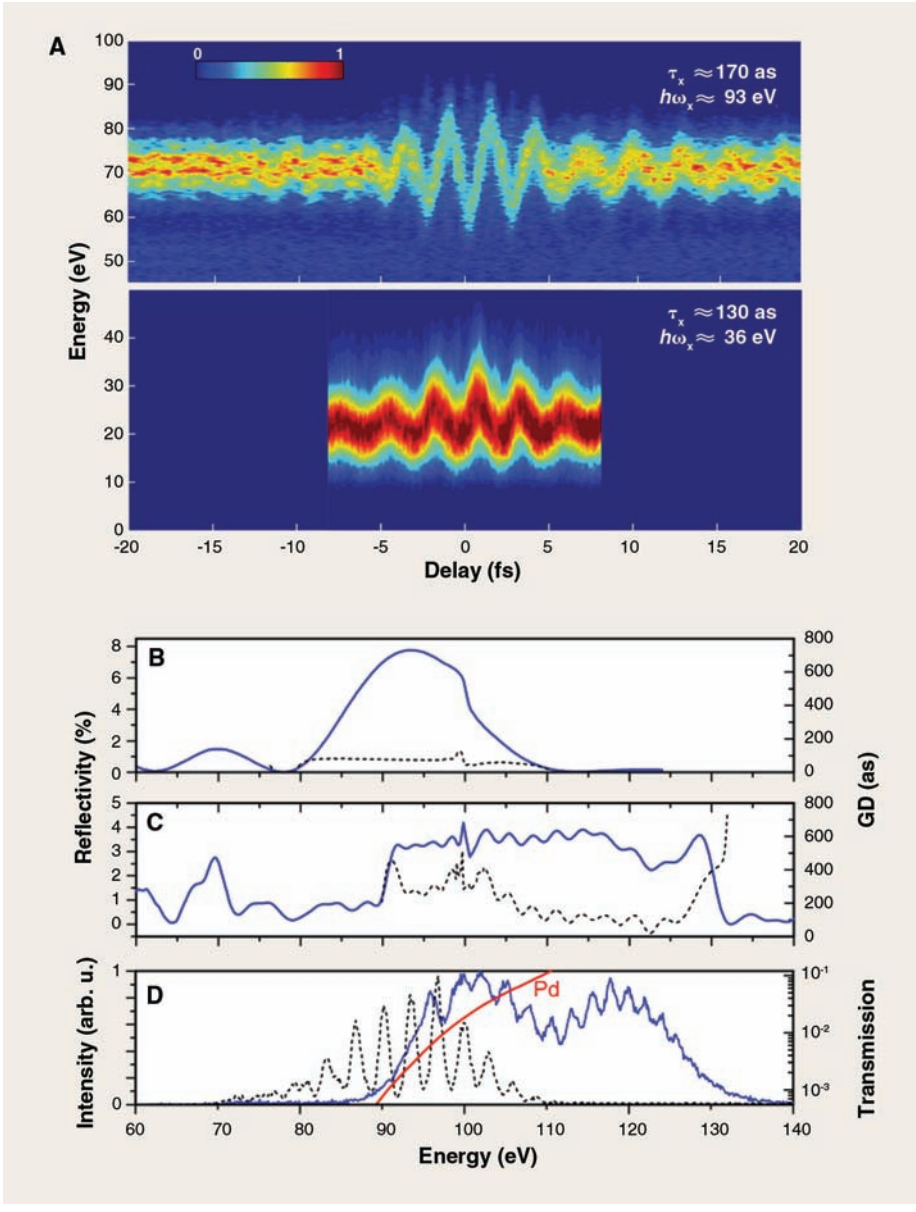


Fig. 4. Current frontiers of attosecond technology. (**A**) Attosecond streaking spectrograms recorded with few-cycle NIR pulses ($\lambda_L \approx 750$ nm). The low-energy spectrogram (bottom) shows the carrier photon energy of an XUV pulse, $\hbar\omega_X \approx 36$ eV, with argon target atoms (courtesy of M. Nisoli). The high-energy spectrogram (top) shows $\hbar\omega_X \approx 93$ eV, with neon target atoms (*49*). (**B**) Computed reflectivity and group delay of the chirped Mo/Si multilayer mirror used for filtering and focusing the isolated sub–170-as, 93-eV pulses. (**C**) Computed reflectivity and group delay of an ultrabroadband chirped multilayer Mo/Si mirror designed for compensating the chirp carried by an attosecond pulse filtered near cutoff from short-trajectory emission (group delay dispersion ~ –0.007 fs^2) and for reflecting a band sufficient for sub–100-as pulse generation (bandwidth > 35 eV). (**D**) Spectrum of high-order harmonics emitted from neon ionized by ~4-fs–duration NIR (~720-nm) pulses near cutoff as transmitted through a high-pass filter (150-nm palladium). The transmission of the Pd foil is shown by the red curve. The spectrum has been recorded with the carrier-envelope phase φ of the waveform-controlled NIR pulse set to provide the broadest continuum. Variation of φ reshapes the overall distribution of the continuum (*42*) rather than introducing a pronounced harmonic-like structure, as observed with several-cycle driver pulses for appropriate phase setting (dashed line). The smooth continuum with a bandwidth of >30 eV suggests the feasibility of sub–100-as XUV pulse generation without manipulation of the driving pulse (e.g., superimposing its second harmonic or gating its polarization).

Atomic physics and x-ray science. How is the energy of an x-ray photon distributed between electrons upon its absorption by an atom? Can electronic transitions deep inside atoms be affected by controlled ultrastrong external fields rivaling in strength the internal Coulomb fields, e.g., for opening up novel routes to efficient, compact x-ray lasers?

Physical chemistry, molecular biology, bioinformatics, and photovoltaics. Can controlled light fields offer a fundamentally new way of modifying the structure and/or composition of molecules by driving electron wave packets across molecules with synthesized optical fields? What are the microscopic mechanisms underlying biological information transport? Can charge-transfer in host-guest systems (e.g., dye-semiconductor assemblies) be exploited for developing solar cells with unprecedented efficiency?

Information technology. Can electron-based information processing and storage be downscaled to atomic dimensions and sped up to the atomic time scale (i.e., to optical frequencies)? Can these ultimate limits be reached by exploiting electric interactions (electronics) or magnetic interactions (spintronics) or collective electron motion (plasmonics)? Which incarnation of light-wave electronics will be the ultimate electron-based information technology?

The answers to these questions will rely on exploring and controlling the microscopic motion of electrons, on atomic scales in space and time. Attosecond technology now offers the tools for tackling these and many other exciting questions. The importance of the answers being sought will drive its proliferation.

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Supporting Online Material

www.sciencemag.org/cgi/content/full/317/5839/769/DC1
Figs. S1 and S2
References

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REVIEW

Harnessing Attosecond Science in the Quest for Coherent X-rays

Henry Kapteyn, Oren Cohen, Ivan Christov, Margaret Murnane*

Modern laser technology has revolutionized the sensitivity and precision of spectroscopy by providing coherent light in a spectrum spanning the infrared, visible, and ultraviolet wavelength regimes. However, the generation of shorter-wavelength coherent pulses in the x-ray region has proven much more challenging. The recent emergence of high harmonic generation techniques opens the door to this possibility. Here we review the new science that is enabled by an ability to manipulate and control electrons on attosecond time scales, ranging from new tabletop sources of coherent x-rays to an ability to follow complex electron dynamics in molecules and materials. We also explore the implications of these advances for the future of molecular structural characterization schemes that currently rely so heavily on scattering from incoherent x-ray sources.

Next year, 2008, will mark the 50th anniversary of the revolutionary paper by Schawlow and Townes that proposed the laser (*1*). This paper extended concepts first used to demonstrate the maser in the microwave region of the spectrum into the visible spectrum. Soon after the laser was demonstrated, scientists discovered how to control laser light to generate extremely short nanosecond, picosecond, and even femtosecond pulses. Given the origin of the laser, it was also natural to attempt to generate coherent light at shorter and shorter wavelengths. However, this effort proved very challenging because of the punishing power scaling inherent in lasers. Basic physics dictates that the energy required to implement a laser scales roughly as $1/\lambda^5$; that is, a laser at a 10 times shorter wavelength (λ) requires ~100,000 times the input power. Thus, the first x-ray lasers implemented in the 1980s used the building-sized Nova fusion laser at Lawrence Livermore National Laboratory as a power source to generate soft (relatively long-wavelength) x-rays. Since that initial x-ray laser, considerable progress has been made in downscaling the laser needed as the power source

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