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Harnessing Attosecond Science in the Quest for Coherent X-rays

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Science **317**, 775 (2007);
DOI: 10.1126/science.1143679

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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 lightwave 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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Figs. S1 and S2
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10.1126/science.1142855

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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(2). Nevertheless, the generation of coherent hard x-rays from x-ray lasers remains a daunting prospect.

Fortunately, in recent years scientists have found a way to make rapid progress toward generating coherent light at very short wavelengths by using alternative techniques. Large-scale free-electron lasers promise to produce high-energy pulses of coherent x-rays using, for example, the 2-km-long electron accelerator at the Stanford Linear Accelerator Center. Extreme nonlinear optical techniques have succeeded in upconverting visible laser light into x-rays, making a tabletop source of coherent x-rays possible. This ability has given us a new coherent light source that spans such a large region of the spectrum that we can now access processes that occur on subfemtosecond or attosecond (1 as = 10^{-18} s) time scales. Equally intriguing is the fact that we have learned how to use femtosecond laser light to coherently manipulate electrons in atoms and molecules on their fundamental attosecond time scales. The richness and complexity of attosecond science and technology are only just beginning to be uncovered. As we discuss here, attosecond science can capture the complex interwoven dance of electrons in molecules and materials. Attosecond science also shows great promise for developing new ultrasensitive molecular imaging and spectroscopic techniques. Finally, attosecond science represents the most promising avenue to achieve what had seemed hopelessly impractical until now: the generation of bright, coherent, hard x-ray beams using a tabletop-scale apparatus.

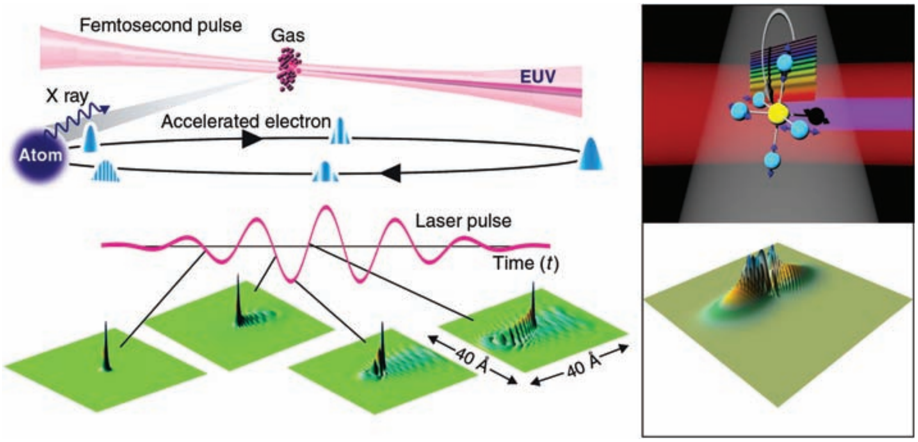

Fig. 1. Attosecond coherent electron rescattering from atoms and molecules. Classical (**top**) and quantum (**bottom**) pictures of high harmonic generation. A strong laser field plucks an electron from an atom (**left**) or molecule (**right**). After evolving as a free electron for a fraction of a femtosecond, the electron can recombine with the ion, emitting a coherent x-ray. The quantum wave functions of the ionizing electrons (shown in green) have many modulations that rapidly change in time as the electron is accelerated by the laser field.

The Birth of Attosecond Science

Attosecond science began with the discovery of high harmonic generation: an extreme nonlinear process that remains to date perhaps the best example of complex attosecond dynamics. When driven by a strong femtosecond laser, atoms can emit coherent light at frequencies much higher than that of the incident laser. This high harmonic generation process is a result of an electron rescattering process (Fig. 1). When an atom or molecule is exposed to a strong electric field that is comparable to the field binding the electron to the nucleus, the most loosely bound electron can be ripped from the atom. The laser intensities required—10^{13} to 10^{15} W cm^{-2}—are easily accessible with tabletop femtosecond lasers. Once free, the electron will follow a trajectory controlled by the laser field, first moving away from the parent ion and then reversing its motion as the laser field oscillates in time.

Upon returning to the vicinity of its parent ion, this electron has some probability of recombining with it and giving up its excess kinetic energy as a photon, with energy corresponding to dozens or hundreds of visible laser photons. Each time this recollision happens, a burst of attosecond-duration x-rays is emitted. This burst generally occurs twice during each cycle of the driving laser field, or every 1.3 fs for a driving laser at 800 nm wavelength (because the ionizing laser field peaks twice each optical cycle). A classical picture of high harmonic generation (Fig. 1, top left) gives a simple expression for the maximum energy of the photons that can be generated ($h\nu_{max}$), which is simply the energy that the electron possesses when it recollides with the ion (Eq. 1)

$$h\nu_{max} = I_p + 3.2U_p \propto I_{laser}\lambda^2 \qquad (1)$$

where I_p is the ionization potential of the atom, U_p is the ponderomotive potential (or the average kinetic energy of an electron oscillating in response to the driving laser field), and I_{laser} and λ are the intensity and wavelength of the driving laser at the time of the ionization (*3*, *4*). From this simple picture, one can see that high harmonic generation corresponds to the coherent version of the x-ray tube first demonstrated by Roentgen, where electrons accelerated by an electric field collide with atoms in a target. In high harmonic generation, the randomly phased electron impact that occurs in an x-ray tube is replaced by the coherent impact of an electron driven by a coherent laser field. Each electron starts in a bound state of the atom and is ripped from the atom and accelerated by the same coherent laser field. Therefore, under the correct conditions, the re-radiated x-ray waves are identical for each atom and add together to generate a fully coherent laserlike x-ray beam (*5*). Moreover, the linear intensity scaling law given by the cutoff relation of Eq. 1 (for high recollision energies $\gg I_p$) is remarkably favorable; the intensities required to generate hard x-rays of ~1 to 10 keV are readily accessible using current tabletop lasers.

Much of the beauty and complexity that have sparked long-term interest in high harmonic generation originate because of its quantum or wavelike properties (*3*, *6*). During its trajectory as a free particle, the electron wave evolves with a deBroglie wavelength corresponding to $\lambda = h/p$ (where p is the electron momentum and h is Planck's constant) (Fig. 1, bottom). The total quantum phase advance of the electron during its free trajectory between ionization and recombination is quite large, corresponding to dozens to hundreds of radians. Moreover, the quantum phase accumulated by the electron depends on the exact shape of the electromagnetic field of the laser that guides it during the suboptical cycle time interval between ionization and rescattering. This same quantum phase accumulated by the electron is then acquired by the high harmonic x-ray wave that is emitted when the electron recombines with the ion. The surprising end result is that the phase of the x-ray waves is thus not rigidly related to the phase of the driving laser, but rather depends on the light field that guides the electron in its boomerang-like journey first away from, and then back to, the ion. This property is very different from those pertaining to any other type of nonlinear optical process and opens up many exciting possibilities. For example, during the attosecond time interval between when the electron is ejected and when it recombines with the ion, its wave function can be manipulated in useful ways (*7*–*9*). This can be accomplished by shaping the driving laser field itself or by imposing another external light field.

Several remarkable scientific and technological opportunities have emerged as a result of the interaction of strong light fields with matter—a property of matter that is intimately associated with attosecond science and was discovered only 20 years ago (*10*, *11*). Most current research in attosecond science falls into three broad categories: (i) understanding how to control electron rescattering in order to manipulate electrons on attosecond time scales in useful ways, (ii) learning how to use the rescattering electrons as a probe of molecular dynamics, and (iii) using the attosecond time

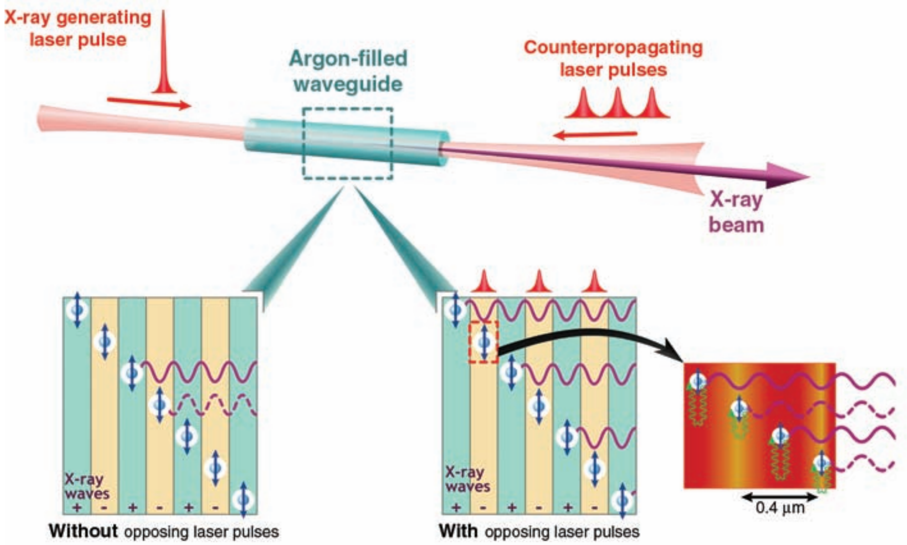

Fig. 2. Crystals made from light by manipulating electron rescattering. Shining a light pattern on a gas can control where in the gas the coherent x-rays are generated (bottom left). X-rays are generated throughout the medium, but emissions from the orange regions cancel out emissions from the blue regions, because the driving laser field and x-ray waves slip in phase by π after traveling through each region (corresponding to one L_c or a few hundred micrometers at 70-eV photon energies). This results in a weak x-ray output from the medium. (Bottom right) A light pattern is used to scramble and disrupt the electron and x-ray phases in regions where the forward- and backward-propagating laser beams overlap. In these overlapping regions (shown in the inset, and indicated by the horizontal black arrow), the laser intensity is modulated on short distances (0.4 μm) that are much shorter than L_c. The modulated laser intensity modulates and scrambles the electron phase, shown in green in the inset. This eliminates x-ray emission from the orange regions, so that emission from the blue regions can now add constructively to generate a bright x-ray output.

structure of x-rays generated by high harmonic generation as a probe of complex electron dynamics in molecules and materials. We briefly discuss the exciting scientific opportunities in each area.

Manipulating Electrons on Attosecond Time Scales

In exciting recent experimental and theoretical developments (*9*, *12*), the concept of manipulating coherent electrons on attosecond time scales has been used to overcome one of the major outstanding problems in nonlinear optics: the efficient generation of coherent high-energy x-rays from lasers. The major challenge in generating a usable flux of x-rays from high-order harmonic generation is in phase-matching the process. As the driving laser beam propagates in the gas, the harmonic signal wave will build up constructively over a long distance only if the driving laser wave and the generated harmonic wave travel with the same crest or phase velocity throughout the medium. In conventional nonlinear optics, this is achieved using birefringence; that is, a nonlinear crystal structure is chosen specifically to equalize the propagation phase velocities of the two disparate colors. This approach of structuring the medium will not work for high harmonic generation, where a gas must be used as the medium because it is literally ripped apart by the strong laser field. Moreover, free electrons act like highly dispersive prisms, causing a severe phase-velocity slip between the laser and the x-ray waves. This effect greatly reduces the distance over which the x-ray waves build up constructively [called the coherence length (L_C)] to millimeters or micrometers for photon energies between 150 eV and 1 keV. (Photon energies below 130 eV can be generated before the gas is fully ionized. Under these conditions, the dispersion of the neutral gas and the free-electron plasma can balance, resulting in perfect phase-velocity matching of the laser and x-ray waves.)

The propagating phase-velocity slip that occurs as the laser and harmonic x-ray waves travel through the gas will add to any quantum phase accumulated by the electron during its trajectory away from the ion. So an intriguing question arises: Is there a way to compensate for the propagation phase slip using the quantum phase accumulated by the electron during its attosecond excursion from the ion? Fortunately, the answer is yes. The phase-matching challenge for high harmonic generation at high energies can be overcome by patterning the laser field driving the process rather than by structuring the medium, as is the case in visible nonlinear optics. This is because high harmonic generation is a purely electronic process that nevertheless does not respond instantly but rather has a subfemtosecond response time (*13*). The quantum phase resulting from the recollision process can be influenced by the driving laser field or by any other field that is simultaneously applied to the atom or molecule (*14*). This provides a way to control electrons at angstrom spatial dimensions and on attosecond time scales (*15*).

Two approaches for using a patterned light field to correct for the propagation phase-velocity slip between the laser and x-rays have been demonstrated experimentally to date. In one approach, the driving laser propagates through a periodically modulated gas-filled waveguide (*16*). The x-ray emission is brightest where the waveguide diameter is smallest and the laser intensity is highest. By limiting the x-ray generation to certain regions in the waveguide where the laser intensity is highest, the laser and x-ray waves can slip back into perfect phase alignment in the regions in between, where no x-rays are generated. This quasi–phase-matching (QPM) scheme therefore automatically ensures that in-phase spatial regions contribute most to the x-ray signal, while the out-of-phase regions are suppressed.

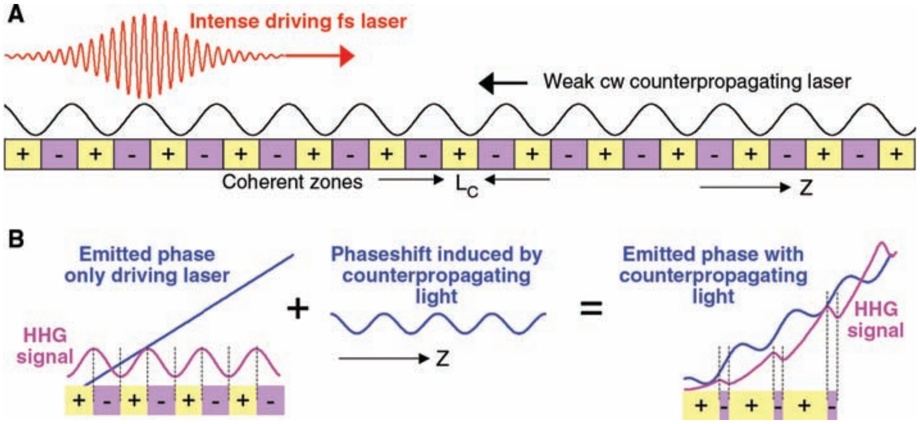

Fig. 3. Coherent hard x-rays produced by manipulating electron rescattering. (**A**) An intense laser pulse (red) generates harmonic x-rays as it travels through a gas. Most of the emitted x-rays interfere with each other from the different out-of-phase zones shown in yellow and purple, because the driving laser and generated x-ray waves slip in phase by π over each zone. (**B**) Shining a continuous wave laser, whose wavelength matches the coherent zone widths, can manipulate the phase of the recolliding electron on attosecond time scales, to adjust its phase so that x-ray emissions from the entire medium can add constructively. This scheme is ideally suited for hard x-rays around 1 keV and higher, where the coherent zone widths are in the micrometer range.

In more recent experiments, the quantum phase of the recolliding electron was manipulated using a second, independent, patterned light field (*9*). While the electron is away from the ion, it is essentially free of its Coulomb field. Remarkably, very small changes in the light field can result in large changes in the phase of the recolliding electron, because this phase is approximately proportional to the applied laser intensity. Any change in the laser intensity will therefore change the quantum phase of the electron, and this phase shift is then directly mapped into the phase of the x-ray emission as a result of quantum mechanics. As illustrated in Fig. 2, a sequence of weak counterpropagating pulses can interfere with the driving laser pulse that generates the coherent x-rays. In any spatial region where the two pulses collide, an interference pattern is created between the forward- and backward-going pulses. This modulated laser intensity scrambles the recolliding electron phase on very short spatial lengths (≈400 nm), and as a result, will also scramble the x-ray phase, effectively preventing any x-ray wave buildup. In regions where the forward- and backward-propagating pulses do not intersect, the x-ray signal grows over distances as great as L_c, and the x-ray signal from the different regions can add in-phase under the correct conditions. Such light sequences were used recently to selectively enhance a single harmonic order by almost three orders of magnitude (*9*).

How far can we go with these attosecond manipulation techniques? Is it possible to generate bright, coherent, hard x-rays using high harmonic generation for applications in crystallography, biology, materials science, and medical imaging? In theory, the answer is yes. As shown in Fig. 3, instead of using sequences of pulses to eliminate harmonic emission from wide (a fraction of a millimeter) regions of the medium that would otherwise contribute destructively, the oscillating field of a continuous-wave laser can be used to continually adjust the phase of the recolliding electron and x-rays (*12*). This approach shows great promise for generating bright coherent beams at very high photon energies well above 1 keV, where the phase-slip distance is extremely short, on the order of micrometers, and is well matched to infrared wavelengths. Even more complicated light patterns could be used to manipulate x-ray wave fronts; for example, to focus them.

Attosecond Electron Recollisions with Molecules

Another exciting frontier of attosecond science is to exploit attosecond electron recollisions with molecules. In these experiments, an electron is plucked from a molecule and returns to the same molecule a fraction of an optical cycle later (Fig. 1) while emitting a coherent x-ray. As the electron accelerates in the laser field, it gains energy ranging from tens to hundreds of electron volts, corresponding to a characteristic electron deBroglie wavelength of ~1 Å. This electron wavelength is well matched to the spacing between atoms in a molecule. What is intriguing about this electron recollision is that the electron is coherent with its parent ion and can be used as an in situ probe of molecular dynamics—essentially using a molecule's own electrons for a new type of electron diffraction experiment. X-ray harmonics generated from molecules are very sensitive to the orientation (*17*), structure (*18*), and dynamic motion (*19*, *20*) of the electrons and atoms in a molecule. To date, this technique has been used to map the valence electron orbital in a diatomic molecule and to observe simple shape changes and vibrational dynamics in molecules.

As a method for observing molecular dynamics, high harmonic generation also conveniently probes motions of the molecules in their ground electronic state, which is particularly relevant to chemistry. Moreover, the time resolution is high enough to decouple the electronic and nuclear motions. In the future, high harmonic generation from molecules could become a broadly applicable probe of chemical dynamics, combining ultrahigh time resolution with the potential for obtaining structural information, complementary to techniques such as femtosecond electron diffraction.

Probing Attosecond Electron Dynamics in Atoms, Molecules, and Materials

The x-ray high harmonic bursts generated by recolliding electrons represent the fastest strobe light in existence (*21*–*23*), fast enough to capture the fleeting motion of electrons in atoms, molecules, and solids. No other probe has succeeded in this endeavor to date. Moreover, their average brightness compares well with bending magnet synchrotron sources and will increase further as more powerful lasers are developed. Single, isolated, attosecond x-ray bursts can be produced with a laser pulse lasting only a few optical periods (~5 fs) (*22*, *23*). In this case, the time-varying few-cycle field ensures that the highest harmonics are emitted only during one half-cycle of the laser field. Trains of attosecond bursts of x-rays are generated if a longer driving laser pulse is used. These attosecond bursts of x-rays (whether isolated or in a train of pulses, depending on the experiment) are ideal probes of complex correlated electron dynamics in atoms, molecules, and materials.

To date, attosecond pulses have been used to follow some of the fastest electron dynamics, such as Auger decay in atoms (*24*) or laser-assisted photoemission from solids (*25*). In some cases, these experiments have confirmed information already available from spectral studies. More sophisticated current experiments are just beginning to probe electron dynamics in molecules and solids that cannot be examined in other ways, such as the dynamics of multi-electron processes, highly excited and strong field processes, and correlated electron dynamics at surfaces or in nanomaterials.

Looking Forward

The attosecond physics of high-order harmonic generation is one of the great success stories of nonlinear optics in the past 20 years. The first experiments were little more than a physics curiosity, with limited apparent use. Since that time, many exciting applications of attosecond bursts of coherent x-rays have been demonstrated, including high-resolution coherent x-ray imaging, femtosecond holography for studying nanothermal transport, real-time observation of molecular motion on surfaces, ultrasensitive molecular spectroscopies and imaging, and capturing the motion of electrons in atoms, molecules, and materials (*26*–*29*). As we look forward, attosecond science is poised to revolutionize how we understand and control electron dynamics in matter, whereas attosecond technology may revolutionize crystallography, x-ray spectroscopy, and biological, materials, and medical imaging.

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32. The authors gratefully acknowledge support for their research from NSF and the U.S. Department of Energy.

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