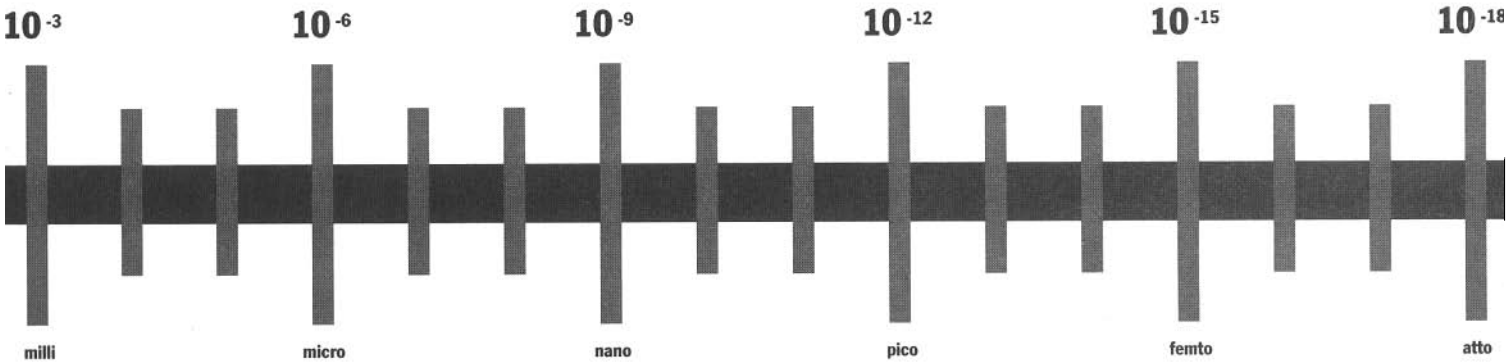


Breaking the Attosecond Barrier

By Paul Corkum

Snapshot: Very short pulses of light enable researchers to probe never-before-observed phenomenon. Corkum describes how attosecond light pulses can be achieved.

Almost immediately after the realization of the laser in 1960, researchers began their inevitable quest for shorter and shorter pulse durations that would allow them to probe previously immeasurable phenomena in chemistry, physics, and biology.

The first milestone was relatively easy. Scientists quickly surpassed the speeds available through electronics and then, using visible and near-visible lasers, went on to set pulse-duration records almost yearly. But once they reached pulses lasting only a few cycles[1] (six femtoseconds) they came up against a fundamental barrier. New records would only be achieved by switching the quest to ultraviolet (UV) or shorter wavelength radiation.

Although this region of the spectrum contains many sources of coherent radiation, only one stands out for the beauty and regularity of its emission: the high harmonics of visible or near-visible light that are produced when rare gases are irradiated by femtosecond high-intensity pulses. Within these high harmonics resides the potential for breaking the attosecond barrier.

The potential is implicit in a figure published in 1992 by Macklin *et al.*[2] Illustrating the harmonic emission that stretches from 30 nm to 7.5 nm, this spectrum has a bandwidth capable of supporting a 10 attosecond (as) pulse. Particularly striking is the parallel between the regularly spaced harmonics and the longitudinal modes of a laser. If the horizontal axis in Figure 1 were not labeled, Figure 1 could easily be mistaken for the output of a mode-locked laser. However, the figure was obtained by illuminating neon with the 125 fs, 10^{15} W/cm^2 pulse of a Ti:sapphire laser.

To exploit the promise hidden in this spectral representation, the physics behind the origin of these harmonics

must be understood. The figure reveals three clues. First, where there is a source of 100 eV photons there must be an electron with at least that much energy. Second, since a 100 eV electron has an energy greater than the ionization potential of any atom, this electron can be thought of as having ionized before it emitted harmonic radiation. Third, to emit a 100 eV photon, the electron must interact with something. The only possibility is its parent ion.

There is additional evidence that ionization and harmonic generation must go hand-in-hand. At 10^{15} W/cm^2 (an intensity typical of a high-harmonics experiment) the peak laser electric field is approximately 10 V/Å. This means that an electron must move considerably more than 10 Å in this laser field to accumulate 150 eV of energy.

Given these clues, there are three areas to investigate before the attosecond barrier can be shattered: the electron's release from the atom as a result of ionization; its behavior in the strong laser field; and the collision of the newly freed electron with its parent ion.

Ionization: when opportunity first knocks

Ionization is the most logical starting point from which to attack the attosecond barrier be-cause it is just after ionization—when the electron experiences the very strong force of the laser field—that the harmonic generation process is most sensitive to experimental control.

Before ionization, the electron is held in place by the Coulomb field of the ion. The laser field provides an opportunity for the electron to tunnel from the vicinity of the ion. Although scientists have understood tunneling[3] since the 1930s, few realize that it accurately describes the very modern problem of atomic ionization by state-of-the-art femtosecond lasers. Tunneling, which ignores the excited states, accurately describes the ionization process because intense short pulses lead to large Stark shifts which smear the atom's electronic structure.[4]

A particularly useful tunneling equation that is applicable to a variety of atoms or atomic ions is the so-called ADK (Ammosov, Delone, and Krainov—the authors of Ref. 5) tunneling,[5] but even simpler equations found in standard quantum mechanics texts[3] can be used for any hydrogen-like ion.

The electron: key to the harmonics

Once freed, the electron's motion in the laser field is far from subtle. The force that the electron experiences is extremely strong and the electron must respond. The newly freed electrons are "buffeted" by the strong force of the laser electric field. Initially, they are pulled away from their parent ions; later—as the laser electric field changes direction—they are driven back. Those that return may "slam" into their parent ion, a collision that forces them to emit radiation.

Electrons are key to harmonic generation because the kinetic energy they gain from the laser field is ultimately transformed into the photon energy of high harmonics if and when the electrons re-en-counter the ion. They are also key to sub-femtosecond pulse generation because it is during the time between ionization and collision that the electron (and consequently the harmonic emission) is most susceptible to control.

Classical physics does an excellent job of describing the electron's motion[6] during this period. Classical trajectories are launched from the position of the ion with zero initial velocities. Since the laser field is much stronger than the field of the ion over most of the electron trajectory, for approximate calculations the ion field can be ignored.

The most important electrons are those that tunnel during the quarter period after the laser field peaks, because these electrons follow trajectories that bring them back past the parent ion. Figure 2 shows the velocities of such electrons as they pass the ion during the first period after their birth, plotted as a function of the field's phase at the moment of tunneling. It is those electrons that produce the high harmonic emission shown in Figure 1. When their trajectories in linearly and elliptically polarized light are compared, they foreshadow a method for generating sub-femtosecond pulses.

In linearly polarized light, any motion perpendicular to the laser electric field must be due to quantum effects. The electron motion is a bit like water in a

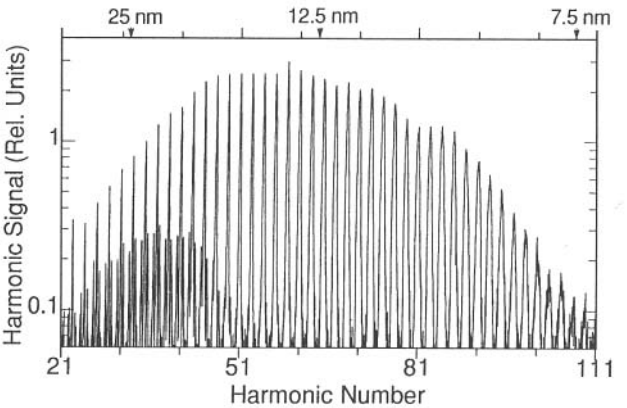


Figure 1. Harmonic signal obtained by the Stanford group[1] by illuminating neon gas with 0.8 mm radiation at an intensity of 10^{15} W/cm^2.

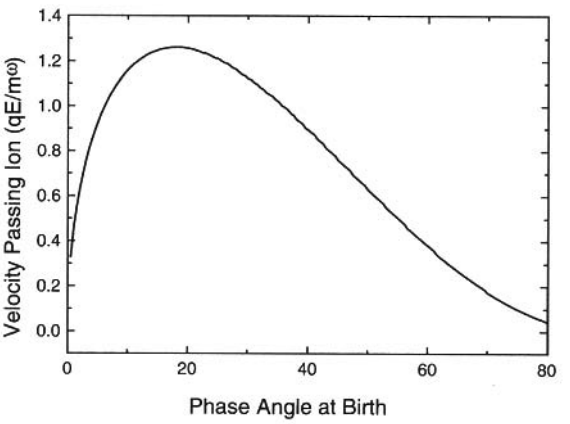


Figure 2. Velocities of the just-ionized electron as it passes the position of the ion the first time after its birth, plotted as a function of the field's phase at the moment of tunneling.

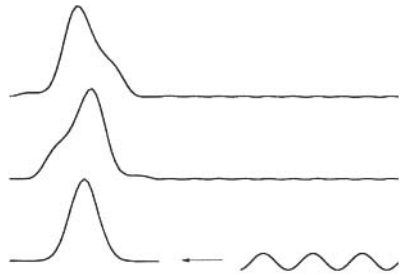
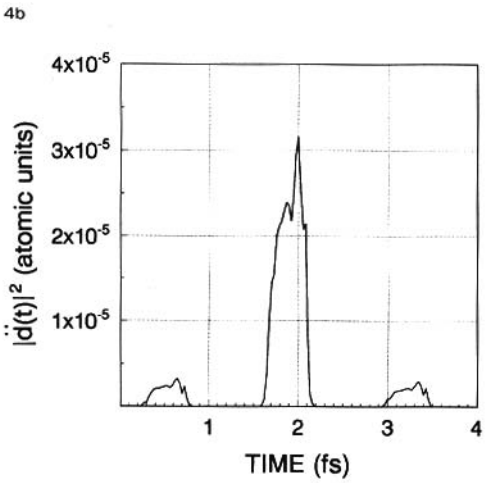


Figure 3. Sketch of the electron wave function before collision (bottom trace) and then the square of the wave function at two moments during the collision (middle and upper trace). Note how quantum interference leads to oscillation of the electron's probability distribution in the vicinity of the nucleus.



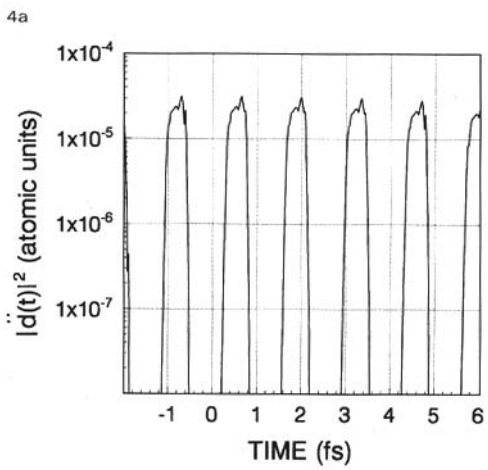


Figure 4. Time dependence of that portion of the harmonic radiation having wavelengths shorter than 100 Å when light from a Ti:sapphire laser irradiates neon at an intensity of 6×10^{14}W/cm^2. (a) Using linearly polarized light and (b) a pulse with time-dependent polarization given by $\omega_1 = 0.93\ \omega_2$.

fountain; it rises from the spout (ion), and as it falls, spreads in the lateral direction. In contrast, when dealing with elliptically polarized light, the classical electron orbit never closes. That is, classically, the electron always misses the parent ion. Only quantum mechanical effects can overcome this. Returning to the fountain analogy, the electron motion resembles a water fountain in a wind storm. As the water follows its vertical trajectory it is blown aside. Depending on the wind velocity (ellipticity), the water may or may not overlap the spout (ion) and therefore may or may not emit harmonics.

The lateral spread is caused by the electron's confinement by the walls of the tunnel from which it has just emerged. The uncertainty principle ensures that its initial transverse momentum cannot be zero. In typical experimental conditions this quantum mechanical effect can be approximated by assuming the electron has an uncertain transverse momentum[7] with a velocity of about 7 Å/fs.

High-harmonic emission: comprehensible from many perspectives

Although there are many ways to qualitatively understand how an electron-ion collision leads to high-harmonic emission, two are particularly helpful.

From the classical perspective, the natural sinusoidal motion of the electron in the laser field is modified by collision with the parent ion, resulting in high frequency emission. In atomic gases, this emission is repeated over many half cycles since every half-period new trajectories are launched. Regularly repeated emission means harmonics.

However, since photo-emission is a quantum mechanical effect, a quantum perspective gives a more complete picture. High-harmonic emission can be thought of as an electron interference effect (as illustrated in Fig. 3) much like interference phenomena in optics. The two critical components of the electron wave function shown in Figure 3(a) are the un-ionized portion remaining in the ground state of the atom and the portion of the wave function representing the electron that is heading for collision. On impact, these two important components of the wave function overlap. The sketches in Figure 3(b) and 3(c) show the sum at two different instants. Interference causes the electron to oscillate, thereby generating high harmonics.

Harmonics from a time perspective: mode-locking the atom

When dealing with ultrafast phenomena, it is often useful to understand the process not only in the frequency domain but also in the time domain. Since high-energy electrons collide with the ion during very limited times within the laser period, the highest energy radiation in the harmonics must be emitted at those times.

Figure 4(a) shows the time dependence of that portion of the harmonic radiation having wavelengths shorter than 100 Å when light from a Ti:sapphire laser irradiates neon at an intensity of 6×10^{14}W/cm^2. This could be achieved with a 2000 Å silver film blocking radiation with wavelengths greater than 100 Å. Silver has very little absorption or dispersion for wavelengths shorter than 100 Å. Figure 4(a) shows that the strongly driven atom emits the high frequency components of the harmonics as if the atom were mode-locked. Individual pulses in the train have a duration of only about 400 as.

Reality check: confirming the model experimentally

One of the model's most dramatic predictions is the exquisite sensitivity of the high-harmonics to the polarization of the fundamental wavelength.[6] In the past two years, at least six groups worldwide have checked how strongly the harmonics depend on the ellipticity of the fundamental light. Figure 5, taken from the work of Dietrich *et al.*,[8] shows that only a slight ellipticity is needed to reduce emission. Just over 4% ellipticity in the intensity of the incident radiation quenches the high-harmonics by two orders of magnitude. (The sensitivity is less for low harmonics.) The solid curve in Figure 5 is calculated using a fully quantum mechanical generalization[9] of the semi-classical model described earlier.

Given the semi-classical and quantum models and the experimental verification just described, it would be irresponsible to ignore the implications. One

major implication is the potential to produce sub-femtosecond pulses.

An "atomic Pockels' cell": a single attosecond pulse

Imagine an ultrashort pulse that begins as circularly polarized, transforms itself to linear polarization at the peak, and then returns to its circularly polarized state: efficient high frequency emission could occur only over a brief interval when the polarization was very close to linear (see Fig. 4). If this interval lasts only a fraction of the period of the fundamental field, a sub-femtosecond short wavelength pulse results.

The equation for the fundamental pulse shows that fabricating such a pulse would not be difficult:

$$E(t)=A(t)[\cos(\omega_1 t)x+\cos(\omega_2 t)y]$$

where A(t) describes the pulse envelope and ω_1 and ω_2 are chosen to produce the necessary rate change of the polarization.

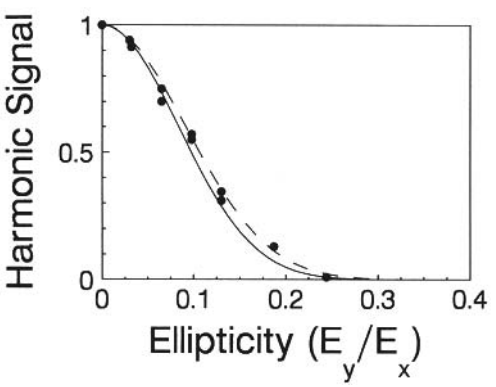


Figure 5. Ellipticity dependence of the 41st harmonic emission from Dietrich *et al.*[8] observed from neon using 800 nm light. The data points plot the experimental results, while the solid curve plots the upper and lower bound to the theoretical results of the quantum theory.

Figure 4(b) shows the calculated output[7] near the time of linear polarization when neon is illuminated with a pulse having a peak intensity of 6×10^{14} ω/cm^2 and $\omega_1 = 0.93\ \omega_2$. The pulse duration is approximately 400 as—more than one order of magnitude shorter than the current experimental record.

New measurement tools

Attosecond science's first hurdle is to develop a method for measuring such short optical pulses or events. Today's techniques were developed for picosecond and femtosecond pulses and for visible and near-visible wavelengths. In the 100 eV region, optics will be difficult and nonlinear media have yet to be identified, so autocorrelation is probably impractical. The most promising approach[10] seems to be a "sub-femtosecond streak camera."

If, at first glance, a sub-femtosecond streak camera seems far-fetched, a closer look shows that, once again, optics can overcome the limitations of electronics. The basic principle of a streak camera is to create a perfect electron replica of the initial light pulse using a photocathode. The resulting electron pulse must then be trans-

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ported without deformation to the region where a time-dependent electric field is applied by rapidly charging deflection plates. The fast-changing electric field deflects the electron from its original trajectory by an angle that depends on when the electrons enter the field.

If an atomic gas were used as a photocathode, the transport problem would be overcome, because the electrons could be produced in the deflection region. A time-dependent field could be applied very rapidly by using an intense laser pulse. The photo-electrons would then be deflected in the electric field of the short laser pulse according to when they were released from the atom.[11] This arrangement would allow much higher sweep speeds than are possible in traditional streak cameras and would provide flexibility for changing the sweep rate, polarization, or intensity of the deflecting field.

A sub-femtosecond streak camera would be very similar to experiments already performed on two-color multiphoton ionization by a group at the University of Virginia.[12]

New scientific challenges

As ultrafast science stands poised on the threshold of a new time scale, it is natural to ask "what does the future hold?" Some insight can be obtained by looking at the characteristic times for electron and nuclear motion. Thermal atomic or molecular collisions occur on the >100 fs time scale, so sub-femtosecond science will consist of studying energetic ions and thermal (or higher) energy electrons, because they move atomic dimensions on the attosecond time scale. Examples from current research illustrates these two possible research directions.

Electron motion can be slowed to below thermal speeds by exciting an electron from a well-defined initial state to a superposition of Rydberg states, enabling researchers[13] to observe the near-classical motion of the electron around an atom. With sub-femtosecond pulses this work could be extended to wave packets in the valence states of molecules, allowing such electronic processes as charge transfer in complex molecules to be time resolved.

Although experiments resolving attosecond electron motion have not yet been performed, effectively sub-femtosecond experiments are already emerging in molecular physics[14] to measure molecular structure. In this case, sub-femtosecond timing arises because the molecule being studied is accelerated to a near relativistic velocity (approximately 0.02c) before colliding with a 30 Å foil. The ions pass the foil in a few hundred attoseconds, during which many electrons are stripped away. The emerging ions immediately repel each other. By observing the direction of the fragments, the geometry of the molecule can be reconstructed.

The stage for attosecond science is now being set. As researchers develop methods to combine the time resolution of ultrafast science and the spatial resolution that is possible with energetic ions, it will at last become possible to realize the dream of both spatial and temporal resolution on the atomic scale.

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