PHYSICS

# Taking an Attosecond Pulse Of Subatomic Behavior

If a handful of theoretical physicists are correct in their calculations, researchers may be on the verge of one of the scientific world's shortest lived achievements: making a pulse of light that disappears a few hundred billionths of a billionth of a second after its creation.

That's a few hundred attoseconds, and physicists, chemists, and laser researchers think this short-term feat could have long-lasting effects. A laser firing attosecond light pulses can function as a high-speed camera flash, allowing scientists to get their first-ever looks at fleeting phenomena such as electrons whirling about a single atom. "This will open up the possibility of looking at the motion of electrons within atoms on their natural time scale," says Paul Corkum, a physicist at Canada's National Research Council (NRC) in Ottawa.

Rather than deducing subatomic behavior from its aftermath—reaction products, molecules formed or broken—scientists will be able to get snapshots of atoms being ionized or electron bonds forming as they happen. "It's a completely new window on atomic physics," says Anne L'Huillier, a physicist at the Lund Institute of Technology in Sweden.

Right now, an attosecond pulse is just a little ahead of its time. The fastest current laser, invented in 1987, flashes in femtoseconds (a femtosecond is a thousand times longer than an attosecond). Revving one up to an attosecond pace is a daunting task, requiring a multitude of light frequencies to be synchronized in a manner and at a speed never before attempted.

But in recent months physicists such as Corkum and his NRC colleague Misha Ivanov, as well as Alexander Kaplan of Johns Hopkins University, have come up with schemes for breaking the attosecond barrier. The ideas are generating optimism. "There are enough good ideas out there that someone will be able to make pulses significantly shorter than they are now, maybe even in the attosecond range," says Phil Bucksbaum, a physicist at the University of Michigan, Ann Arbor (see box).

To break the attosecond barrier, the current crop of ideas must meet four challenges. First, an attosecond pulse must be composed of many different—and high—frequencies of light. If too few frequencies are involved, or if their frequencies are too low, the overall pulse will last too long. Second, even though these light waves have different frequencies, they must be coherent: Their peaks and troughs must align periodically—a task akin to swinging the pendulums of a dozen grandfather clocks at different rates, yet making them all hit the right extreme of their arc once every minute. Third, a pulse must be strong, generating enough photons to illuminate its subatomic subjects. And finally, the attosecond flashes must be separated by at least 1 millionth of a second in order to work with the "low-speed" electronics that will record the results.

None of these challenges will be easy to meet. Corkum's plan is generally regarded as having the best chance, as it is based on well-understood—although difficult to apply—principles of optics. "It's clearly the one that most people talk about," says Chris Barty, a physicist at the University of California, San Diego (UCSD). Adds Ken Schafer, also a UCSD physicist: "I think he can get it."

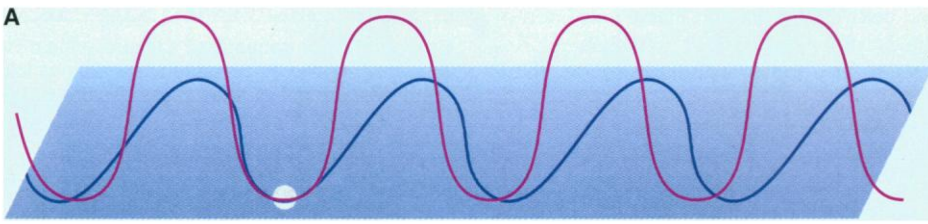
**Playing with a broad band.** The first challenge—a broad spectrum—is posed by quantum mechanics, which states that there is a mathematical trade-off between the duration of a light pulse and the range of frequencies that make it up: As one shrinks, the other grows. So as a light pulse is squeezed into a shorter time period, the range of frequencies in the pulse becomes broader. Because lasers generally pump out light at one or a few closely related frequencies, using one to create a quick broad-spectrum pulse can be difficult. But in recent years, researchers around the world have devised a way to use laser pulses of visible light to generate additional frequencies at regular multiples of the original in the short, ultraviolet frequencies. And it's these "harmonics" that Corkum and his NRC colleagues propose to combine into attosecond pulses.

In constructing a functional attosecond laser, Corkum and his colleagues plan to use a pulse from a titanium sapphire laser to ionize a gas of xenon or argon atoms. The pulse would generate an oscillating electric field which first drives the freed electrons in the gas away from their parent atoms, then reverses course and drives them back at near light speed. A small percentage of these energy-rich electrons actually collide with their parents, and the energy of the collision is released as a short-wavelength photon. Most of them cancel one another out—the peak of one coincides with the trough of another—so only the harmonics survive.

These harmonics are much like the overtones of musical notes. And their mathematical relationship will help the NRC team meet the second challenge of coherence. As they travel together, the regular relationship among harmonic wave peaks and troughs will align them at regular intervals. Without this relationship, waves would rise and fall at different times, canceling each other out.

But the gas that generates the harmonics in the first place also makes this second challenge more difficult. Although all the harmonics begin as coherent waves, some travel more slowly through the gas, so over distance they fall completely out of step. To prevent this, Corkum plans to limit the number of atoms in the gas. That should reduce the interaction between light waves and atoms in the gas, keeping different frequencies of light in step. But it comes with a cost: reducing the number of harmonic photons generated in the first place.

Corkum also plans to screen out low-frequency harmonic photons with a special metallic filter to ensure the pulse stays tightly bunched: A mixture of low and high frequencies would spread out the pulse duration past the attosecond limit. The remaining high-frequency harmonic photons will, Corkum hopes, combine to produce pulses of ultraviolet light lasting a mere 400 attoseconds. "It's a sound idea," says Charles Rhodes, a physicist at the University of Illinois, Chicago.




**Shrinking a pulse down to size. (A)** Two intersecting electromagnetic fields of different frequencies, generated by a laser pulse, periodically coincide *(white circle).* **(B)** This interaction, in a xenon gas, slams electrons (e–) into their parent atoms, producing a series of photons. **(C)** These photons have a broad range of frequencies, which add up to an attosecond pulse.

**Avoiding an electron blur.** Sound it may be, says Alexander Kaplan, a theoretical physicist at Johns Hopkins, but he argues that it's weak when it comes to the third

## Other Paths to Faster Pulses

Harmonic-derived attosecond pulses (see main text) may not be bright enough for subatomic spectroscopy, some researchers say. They've got other ideas.

One, proposed by Alexander Kaplan of Johns Hopkins University, is to use another method for generating a broad bandwidth of light, known as cascaded stimulated Raman scattering. The scattering scheme begins when a pulse of visible laser light is fired into a gas containing hydrogen molecules, which absorb photons from the pulse. This briefly excites electrons in these molecules, and in order to return to lower energy levels they must give off their excess energy. Much of it is given up as a photon with a slightly lower frequency than the laser light.

This is the beginning of the cascade effect: The new photons are then absorbed by other gas molecules, again stimulating the release of photons with still lower frequencies. At the same time, photons at several higher frequencies are being made when a gas molecule absorbs two or more photons from the original laser pulse simultaneously—the energy of the photons has an additive effect, prompting electrons in the molecule to release high-energy (and thus high-frequency) photons. This too has a cascade effect.

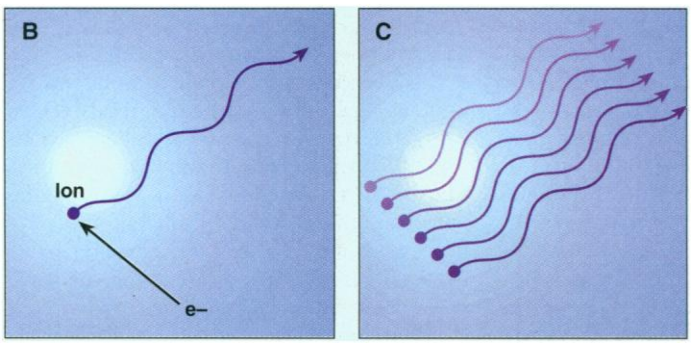
The end result is a pulse containing as many as 15 different light frequencies, ranging from the near-infrared to the ultraviolet, with about 40% of the strength of the original laser pulse. As these photons propagate, Kaplan predicts, they should line up to produce 200-attosecond pulses. But in this case, it would produce a train of them—one every 8 femtoseconds by his calculations. That's too fast to make snapshots of electronic motion: The pulses will flash many times on a single electron, rendering it a blur, just like a strobing flash will blur a photo of a runner if the camera shutter is open too long. But Kaplan and some others believe a train of attosecond pulses with frequencies in the infrared and visible light ranges could be useful for other applications, such as understanding the ultrafast reactions taking place in dense plasmas created by plasma fusion reactors.

Kaplan has other attosecond schemes as well, as do other researchers. At the University of Illinois, Chicago, Charles Rhodes is trying to make short pulses by generating a broad band of x-ray frequencies. And at Washington State University in Pullman, Henry Kapteyn and Margaret Murnane have suggested generating a broad range of ultraviolet frequencies by reflecting photons from an infrared laser off an electron-rich plasma. If any of these researchers can meet all the challenges of making an attosecond pulse, the results could be highly illuminating.

–R.F.S.

challenge: a strong pulse. Not only is harmonic generation an inefficient process—transforming just a tiny fraction of the laser's original photons into harmonic photons—but Corkum's plan limits the number of harmonic-generating atoms in the gas and screens out all but the high-frequency harmonics, which have the lowest cumulative intensity. Corkum admits this is a problem. "You would like to have more light," he concedes. "But you just have to start with what you have and see if it's enough."

Most observers feel that Corkum's plan gets much stronger when it comes to the final challenge of producing intermittent pulses. Corkum needs to do this to accommodate the low-speed electronics that will record an electron's position. These detectors will act like a camera shutter, and if two or more attosecond flashes go off while the shutter is open, the electron they illuminate will be seen at several positions, not just one, and look like a blur.




To avoid this, Corkum has a "very exciting" solution, says L'Huillier. The idea, published in the 10 April *Physical Review Letters*, is to ensure that bursts of photons are made only at (relatively) widely spaced intervals by changing the oscillation of the electromagnetic field in the original laser pulse that creates them. Essentially, such oscillations can be described as "linear" and "circular." The linear type smashes the electrons back into their parent atoms to make photons, but Corkum realized that if he switched the oscillations to a circular pattern, he could build a pause into his stream of attosecond pulses.

Corkum proposes starting with a conventional 50-femtosecond laser pulse of light, which he would then split into two beams with different frequencies. One beam would pass through a filter to change the direction of its oscillation, or "polarization," 90 degrees. He will then recombine the beams, in effect creating two beams in perpendicular but intersecting planes (see diagram). Because the light waves in the two beams have different frequencies, they will be oscillating at slightly different rates. As a result, the relationship of their peaks and troughs will change over time. Most of the time they will add up to give circularly polarized light. But he calculates that at just one point during each pulse, the waves will add up to produce linear polarization, driving electrons into parent atoms and creating an attosecond pulse.

**Seeing is believing.** If this scheme works, Corkum and his colleagues plan to use attosecond pulses in so-called "pump-probe" experiments to detect electrons in an atom. These experiments are currently done with femtosecond lasers to observe interactions between atoms. They start by dividing the short pulses into two beams and delaying one beam slightly by routing it through an extra series of mirrors. Energy from the first beam, the pump, triggers a reaction, such as splitting a molecule into its component atoms. The probe is then routed in just behind the pump beam to illuminate the atoms as they fly apart. By repeating the experiment many times and changing the delay of the probe beam, researchers can take a series of snapshots of the interaction.

With attosecond pulses, the movie will focus on the motion of an electron around an atom or perhaps its motion during ionization. But the kickoff date for such a production, Corkum says, is still at least a couple of years away, as no group yet has all the equipment for both generating single attosecond pulses and measuring the results. Corkum's group has applied for a $1 million, 5-year grant to begin this work; they're awaiting a decision, which they expect by the end of the year. "This will be done," Corkum says, "if not by us then by other people." And no matter who gets final production credit, researchers anticipate this attosecond short film will have a very long run.

–**Robert F. Service**