

Tabletop laser-like device creates coherent multicolor beams of ultraviolet, T- and Xrays

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This art represents an electron being ripped from an atom by a strong laser field, which stretches its quantum wave function over hundreds of atomic sizes. Just as electrons accelerated in an X-ray tube emit bremsstrahlung radiation, electrons accelerated by a laser can emit rainbows of coherent X-rays in a laser-like beam. X-ray light is "invisible to the human eye but is important for being able to 'see' the fine details and fastest motions of the nanoworld." Credit: Tenio Popmintchev, JILA and University of Colorado at Boulder

For the first time, researchers have produced a coherent, laser-like, directed beam of light that simultaneously streams ultraviolet light, X-rays, and all wavelengths in between.

One of the few light sources to successfully produce a <u>coherent beam</u> that includes X-rays, this new technology is the first to do so using a



setup that fits on a laboratory table.

An international team of researchers, led by engineers from the NSF Engineering Research Center (ERC) for EUV Science and Technology, reports their findings in the June 8, 2012, issue of *Science*.

By focusing intense pulses of infrared light--each just a few optical cycles in duration--into a high-pressure gas cell, the researchers converted part of the original laser energy into a coherent super-continuum of light that extends well into the X-ray region of the spectrum.

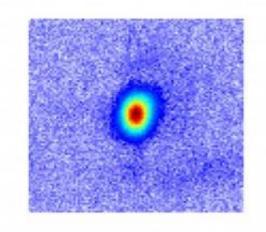
The X-ray burst that emerges has much shorter wavelengths than the original laser pulse, which will make it possible to follow the tiniest, fastest physical processes in nature, including the coupled dance of electrons and ions in molecules as they undergo chemical reactions, or the flow of charges and spins in materials.

"This is the broadest spectral-bandwidth, coherent-light source ever generated," says engineering and physics professor Henry Kapteyn of JILA at the University of Colorado at Boulder, who led the study with fellow JILA professor Margaret Murnane and research scientist Tenio Popmintchev, in collaboration with researchers from the Vienna University of Technology, Cornell University and the University of Salamanca.

"It definitely opens up the possibility to probe the shortest space and time scales relevant to any process in our natural world other than nuclear or fundamental particle interactions," Kapteyn adds. The breakthrough builds upon earlier discoveries from Murnane, Kapteyn and their colleagues to generate laser-like <u>beams of light</u> across a broad spectrum of wavelengths.



The researchers use a technique called high-harmonic generation (HHG). HHG was first discovered in the late 1980s, when researchers focused a powerful, ultra-short laser beam into a spray of gas. The researchers were surprised to find that the output beam contained a small amount of many different wavelengths in the ultraviolet region of the spectrum, as well as the original laser wavelength. The new ultraviolet wavelengths were created as the gas atoms were ionized by the laser.



This picture shows an actual image of a coherent (laser-like) X-ray beam. In contrast to the incoherent (light-bulb-like) light emitted in all directions from a Roentgen X-ray tube, the X-rays produced by high harmonic generation emerge as well-directed, laser-like, beams. Credit: Tenio Popmintchev, JILA and University of Colorado at Boulder

"Just as a violin or guitar string will emit harmonics of its fundamental sound tone when plucked strongly, an atom can also emit harmonics of light when plucked violently by a laser pulse," adds Murnane. "The laser pulse first plucks electrons from the atoms, before driving them back again where they can collide with the atoms from which they came. Any excess energy is emitted as high-energy ultraviolet photons."



Like many phenomena, when HHG was first discovered, there was little science to explain it, and it was considered more a curious phenomenon than a potentially useful light source. After years of work, scientists eventually understood how very high harmonics were emitted, however there was one major challenge that most researchers gave up on--for most wavelengths in the X-ray region, the output HHG beams were extremely weak.

Murnane, Kapteyn and their students realized that there might be a chance to overcome that challenge and turn HHG into a useful X-ray light source--the tabletop-scale X-ray laser that has been a goal for laser science since shortly after the laser was first demonstrated in 1960.

"This was not an easy task," says Murnane. "Unlike a laser--which gets more intense as more energy is pumped into the system--in HHG, if the laser hits the atoms too hard, too many electrons are liberated from the gas atoms, and those electrons cause the laser light to speed up. If the speed of the laser and X-rays do not match, there is no way to combine the many X-ray waves together to create a bright output beam, since the X-ray waves from different gas atoms will interfere destructively."

Popmintchev and JILA graduate student Ming-Chang Chen worked out conditions that enable X-ray waves from many atoms in the gas to interfere constructively. The key was to use a relatively long-wavelength, mid-infrared laser and a high pressure gas cell that also guides the laser light. The resulting bright, X-ray beams maintain the coherent, directed beam qualities of the laser that drives the process.

The HHG process is effective only when the atoms are hit "hard and fast" by the laser pulses, with durations nearing 10^{-14} seconds--a fundamental limit representing just a few oscillations of the electromagnetic fields. Murnane and Kapteyn pioneered the technology for generating such light pulses in the 1990s, and used those lasers to



develop and utilize HHG-based light sources in the extreme-ultraviolet (EUV) region of the spectrum in the 2000s. However, while researchers were using those lasers and the HHG technique to measure ever-shorter duration light pulses, they were stymied in how to make coherent light at shorter wavelengths in the more penetrating X-ray region of the spectrum.



This image depicts the experimental setup used to create a coherent version of the Roentgen tube in the soft X-ray region of the spectrum. When a longwavelength, femtosecond laser is focused into this hollow waveguide filled with high-pressure helium gas, part of the laser is converted into an ultrafast, laserlike, X-ray beam. Credit: Tenio Popmintchev, JILA and University of Colorado at Boulder

The new paper in *Science*, under lead author and senior research associate Popmintchev, demonstrates that breakthrough, showing that the understanding of the HHG process the researchers developed is broadly valid.

"We would have never found this if we hadn't sat down and thought about what happens overall during HHG, when we change the wavelength of the laser driving it, what parameters have to be changed to



make it work," added Kapteyn. "The amazing thing is that the physics seem to be panning out even over a very broad range of parameters. Usually in science you find a scaling rule that prevents you from making a dramatic jump, but in this case, we were able to generate 1.6 keV each X-ray photon was generated from more than 5,000 infrared photons."

When the researchers first started to work with ultrafast, mid-infrared lasers just a few years ago, they actually made a step backwards and generated bright extreme-ultraviolet light of longer wavelengths than they used to achieve in the lab.

"However, we discovered a new regime that helped us to realize, just on paper, that we could make this giant step forward towards much shorter electromagnetic wavelengths and generate bright, laser-like, soft and hard X-rays," adds Popmintchev. "What the experiments were suggesting back then looked too good to be true! It seemed that Mother Nature has combined together, in the most simple and beautiful way, all the microscopic and macroscopic physics. Now, we are already at X-ray wavelengths as short as roughly 7.7 angstroms, and we do not know the limit."

To truly control the beam of photons, the researchers needed to understand the HHG process at the atomic level and how X-rays emitted from individual atoms combine to form a coherent beam of light.

That understanding combines microscopic and macroscopic models of the HHG process with the fact that those interactions occur at very high intensity in a dynamically changing medium. The development of such a conceptual understanding took the last decade to develop.

The result was the realization that there is no fundamental limit to the energy of the photons that can be generated using the HHG process. To



obtain higher-energy photons, the system paradoxically begins with laser light using lower energy photons--specifically, mid-infrared lasers.

The JILA researchers demonstrated the validity of that principle in their labs in Colorado, but to achieve their breakthrough, the researchers traveled to Vienna with their beam-generating setup. There, they used a laser developed by co-author Andrius Baltuška and colleagues at the Vienna University of Technology--the world's most-intense ultrashort-pulse laser operating in the mid-infrared, with a wavelength of four microns.

"Thirty years ago, people were saying we could make a coherent X-ray source, but it would have to be an X-ray <u>laser</u>, and we'd need an atomic bomb as the energy source to pump it," said Deborah Jackson, the program officer who oversees the ERC's grant. "Now, we have these guys who understand the science fundamentals well enough to introduce new tricks for efficiently extracting energetic photons, pulling them out at X-ray wavelengths . . . and it's all done on a table-top!"

In addition to achieving the high energy, the increasingly broad spectrum opens a range of new applications.

"In an experiment using such a source, one energy region from the beam will correspond with one element, another with another element, and so on to simultaneously look at atoms across entire molecules, and that will allow us to see how charge moves from one part of a molecule to another as a chemical reaction is happening," adds Kapteyn. "It'll take us awhile to learn how to use this, but it's very exciting."

More information: "Bright Coherent Ultrahigh Harmonics in the keV X-ray Regime from Mid-Infrared Femtosecond Lasers," by T. Popmintchev et al, *Science*, 2012.



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