Researchers establish ultrafast control of quantum processes at several attoseconds

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A team of physicists including Russian researchers conducted an experiment that established control over the ultrafast motion of electrons at three attoseconds for the first time. (An attosecond is 1×10^{-18} of a second—that's one quintillionth of a second. For context, an attosecond is to a second what a second is to about 31.71 billion years.) This opens new directions of research that previously seemed improbable. The experiment was conducted with the help of the free-electron laser FERMI located at the Elettra Sincrotrone research center in Trieste, Italy.

The speed of chemical, physical and biological processes can be extremely high—for instance, atomic bonds are broken and restored within femtoseconds (one millionth of one billionth of a second). The Egyptian-American chemist Ahmed Zewail was the first to succeed in observing the dynamics of chemical processes, which made him a winner of the 1999 Nobel Prize in Chemistry.

Nevertheless, nature is capable of operating even faster. While atomic motions within a molecule can be measured with femtosecond resolution, the dynamics of electrons, which define the nature of chemical bonds, occur a thousand times faster—within tens and hundreds of attoseconds.

The only tools appropriate for studying such processes are so-called X-ray free-electron lasers. In "conventional" gas, liquid and solid-state lasers, excitation of electrons in the bound atomic state serves as the source of photons. In contrast, free-electron lasers operate with the help of a high-quality electron beam wiggling along a sinusoidal path under the effect of an array of magnets. During that process, electrons lose energy by producing radiation.

X-ray free-electron lasers generate radiation with a unique set of properties: a wavelength in the extreme ultraviolet or soft X-rays, unprecedented luminosity, ultrashort femtosecond pulses, tunable frequency and polarization, and coherence. While the properties of the laser itself did not allow for observations accurate to attoseconds, the researchers found a workaround. In their experiment, they irradiated neon atoms with free-electron laser pulses of two frequencies instead of one, and traced the direction of photoelectrons leaving the atom. They used radiation with the fundamental frequency and its second harmonic (with twice the frequency and hence half the wavelength)—specifically, wavelengths of 63.0 and 31.5 nanometers.

Changing the time delay between the harmonics,the scientists observed the dynamics of the process, measuring changes in the photoelectrons' angular distribution. As a result, they managed to overcome the natural obstacles and observed quantum interference between two channels of atomic photoionization with a precision of three attoseconds—simply speaking, they used indirect indicators to measure the time gap of electrons leaving the atom.

"In this experiment, we managed to carry out a scheme that allows to distinguish relative phases of two free-electron laser harmonics," says Elena Gryzlova, senior researcher at the D.V.Skobeltsyn Institute of Nuclear Physics, Moscow State University. "There are many methods to eliminate, or vice versa, to distinguish extra frequencies in visible radiation. But in high-frequency ranges like extreme ultraviolet or X-ray, all of them are inapplicable, as there are no common mirrors or polarizers. However, the main conclusion we can draw based on this experiment is that control over quantum processes with a precision of several attoseconds is possible."

The contribution of Russian scientists in this work is significant: "Our colleague, Alexei Grum-Grzhimailo, made first derivations and co-authored the very idea of the experiment," says Gryzlova. " Later, together with Prof. Svetlana Strakhova, we succeeded in calculating the scale of that effect, investigating whether it would be detectable at all. We then provided formulas to extract the necessary parameters from the general set of data collected in the experiment."

The authors of the article state that the 'dichromatic' laser measurements open a new horizon for research in physics of ultrafast processes.

According to Gryzlova, an application for beamtime to conduct similar experiments on FERMI using molecules, which comprise more complicated system than the neon atom, was recently submitted. The team is also considering researching complex phenomena related to catalyst processes and atmospheric chemistry.

"We expect that this scientific direction will develop further, as the problem of quantum control is one of the cornerstones of contemporary fundamental physics," says Gryzlova.

More information: K. C. Prince et al. Coherent control with a short-wavelength free-electron laser, *Nature Photonics* (2016). DOI: 10.1038/nphoton.2016.13

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