Atom billiards with X-rays: A new approach to look inside molecules

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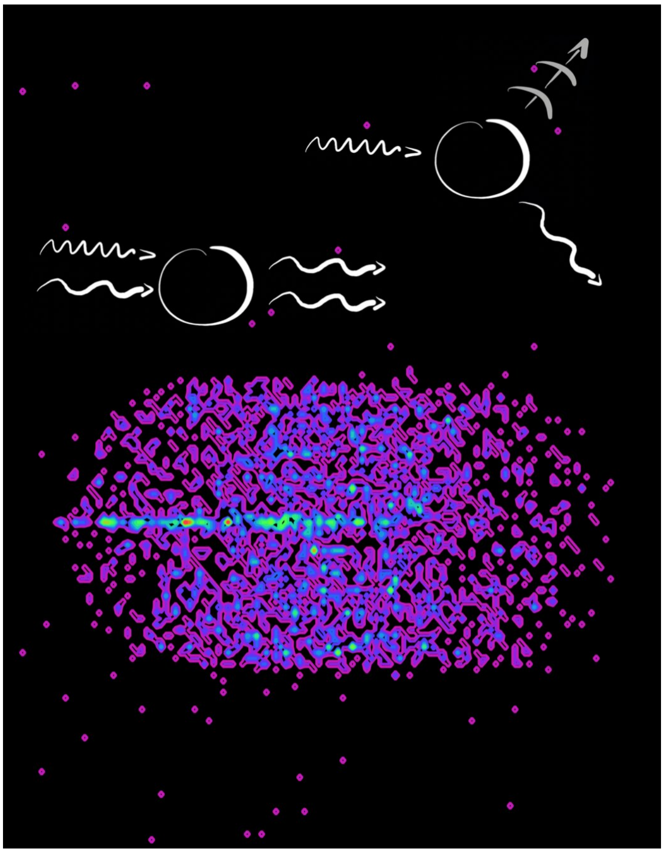

Those atoms, which have undergone interaction with two photons simultaneously hit the detector in a line (horizontal, green). In contrast, atoms, which have absorbed one photon and subsequently emitted another photon, are scattered over a larger area. Credit: Stefan Eisebitt/Max Born Institute

In 1921, Albert Einstein received the Nobel Prize in physics for the discovery that light is quantized, interacting with matter as a stream of particles called photons. Since these early days of quantum mechanics, physicists have known that photons also possess momentum. The photon's ability to transfer momentum was used in a novel approach by scientists of the Max Born Institute, Uppsala University and the European X-ray Free-Electron Laser Facility to observe a fundamental process in the interaction of X-rays with atoms. The detailed experimental and theoretical results are reported in the journal *Science*.

Absorption and emission of photons by atoms are fundamental processes of the interaction of light with matter. Much rarer are processes in which several photons simultaneously interact with one atom. The availability of intense laser beams since the 1960s has led to the development of nonlinear optics for observing and exploiting such processes.

Entirely new possibilities emerge if it is possible to use nonlinear optics with X-rays instead of visible light. The use of ultrashort flashes of X-rays allows for detailed insight into the motion of electrons and atomic nuclei in molecules and solids. This perspective was one of the drivers leading to the construction of X-ray lasers based on particle accelerators in several countries. When the European XFEL X-ray free-electron laser started operation in 2017, the scientific community made an important step in that direction. Nevertheless, progress in the use of nonlinear X-ray processes to study fundamental interaction with matter has been slower than expected. "Typically, the much stronger linear processes occlude the interesting nonlinear processes," says Prof. Ulli Eichmann from Max Born Institute for nonlinear optics and short pulse spectroscopy in Berlin.

The German-Swedish research team has now demonstrated a new method to observe the nonlinear processes without being disturbed by the linear processes. To this end, the team made use of the momentum that is transferred between X-rays and atoms. When crossing a supersonic atomic beam with the X-ray beam, they can identify those atoms that have undergone the so-called stimulated Raman scattering process—a fundamental nonlinear process whereby two photons of different wavelengths hit an atom and two photons of the longer wavelength leave the atom. The results were reported in the journal *Science*.

"Photons transfer momentum to an atom—completely analogous to a billiard ball hitting another one," explains Eichmann. In the stimulated Raman process, both photons leave the atom in the exact same direction as the two incident photons, hence the momentum of the atom and its direction of flight remain essentially unchanged. The much more frequent linear processes, where one photon is absorbed followed by the emission of another photon, have a different signature: As the emitted photon is typically emitted in a different direction, the atom will be deflected. Observing the direction of the atoms, the scientists could thus clearly discriminate the stimulated Raman process from other processes.

"The new method opens unique possibilities when combined in the future with two time-delayed X-ray pulses of different wavelengths. Such pulse patterns have recently become available at X-ray lasers like the European XFEL," explains Dr. Michael Meyer, researcher at the European XFEL.

As X-ray pulses with different wavelengths allow researchers to specifically address particular atoms in a molecule, it is possible to observe in detail how the wavefunctions of electrons in molecules evolve over time. In the long run, the scientists hope not only to observe this evolution, but to influence it via tailored laser pulses. "Our approach allows for a better understanding of chemical reactions on the atomic scale and may help to steer the reactions in a desired direction. As the movement of electrons is the essential step in chemical and photochemical reactions occurring e.g. in batteries and solar cells, our approach may give new insight in such processes as well," says Jan-Erik Rubensson, professor at Uppsala University.

More information: Eichmann et al., Photon-recoil imaging: Expanding the view of nonlinear x-ray physics. *Science* (2020). science.sciencemag.org/cgi/doi … 1126/science.abc2622

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