Stripping atoms

Researchers can now remove 36 electrons from a xenon atom using a pulse of high-energy photons from a free-electron laser. *Nature Photonics* spoke with Daniel Rolles to learn about the process and why the ionization is stronger than expected.

What level of ionization did you achieve?

Prior experiments on light atoms such as neon had shown that the highest charge state created by the interaction with X-ray free-electron laser pulses was the last charge state that could be reached by direct sequential photoionization. That is, the atomic ions would simply keep absorbing photons until their ionization potential exceeded the photon energy, after which the sequential ionization would stop. We found that this was also true for xenon at a photon energy of 2 keV, whereas at 1.5 keV much higher charge states were created, although both photon energies primarily ionize the M-shell in xenon. At 1.5 keV, the direct sequential limit would be Xe^{26+}, but we saw charge states up to Xe^{36+}, which is, as far as we know, the highest charge state ever observed in the photoionization of atoms using a single photon pulse.

LAUREN KNOCHE, SLAC

Daniel Rolles (fifth from left, green shirt) and part of the team at the SLAC Linac Coherent Light Source. The group has demonstrated stronger than expected ionization states of xenon (up to Xe^{36+}) by tearing electrons from the atoms using free-electron laser pulses.

How does the strong ionization occur?

When an X-ray photon is absorbed by an atom or molecule, it usually knocks out one of the electrons from deep inside the atom through a process called photoionization or, more specifically, inner-shell photoionization. This process leaves behind a core-excited atom or molecule that will decay further. Depending on the atom and the electronic shell from which the electron is knocked out, the remaining hole left behind by the electron can either undergo fluorescence decay or Auger decay. In Auger decay, an electron from an outer shell fills the hole and another electron is emitted. Sometimes, several Auger processes can happen in what is referred to as an 'Auger cascade'.

Despite the low probability that a given X-ray photon is absorbed by a given atom (typical cross-sections are around 10^{-19} cm^{-2}), a single X-ray pulse from a free-electron laser such as the Linac Coherent Light Source contains enough photons to allow more than one X-ray photon to be absorbed by the same atom. In our case, the xenon atoms absorbed 20 photons or more in a single pulse, which is more than twice the number we had expected from looking at the known photoabsorption cross-sections. We realized that the reason for this discrepancy was the large number of 'transient' resonances, which appear only during the ionization process. As the xenon atoms are ionized by absorbing one photon after another (and, in most cases, further decay via Auger cascades), their ionization potential increases until it reaches the photon energy. Rather than stopping the photoabsorption as we expected, broad bands of resonances appeared in certain photon energy ranges, which allow for inner-shell electrons to be excited to bound atomic states with very high cross-sections. Because the cross-sections are so high, and because these are no longer individual resonances that are merely spaced far apart but are rather bands of dozens or even hundreds of resonances, many electrons can be excited by the same broadband free-electron laser pulse, thus allowing the atom to decay via autoionization to charge states that are much higher than those allowed in a model of direct sequential photoionization.

What is next?

Going to higher photon energies and/or to the regime of two-photon one-electron ionization would be very interesting in order to investigate the underlying resonance processes further. In terms of practical applications, for example, to assess the extent to which radiation damage in large biomolecules or nanoparticles is influenced by our finding, the most important next step would be to investigate the resonance enabled (or enhanced) X-ray multiple ionization mechanism in molecules, in order to see how a molecular environment affects the processes. We have performed some experiments on small molecules in this respect, but this work needs to be pushed to larger systems.

A significant practical challenge for these studies is the very small amount of beam time available at free-electron laser facilities and the fierce competition in the proposal process. This is especially true at beamlines with higher photon energies, where our experiments are in direct competition with X-ray diffraction experiments. As an atomic, molecular and optical scientist, my view on this question is biased, and I strongly believe that microscopic studies must go hand-in-hand with more applied fields such as X-ray diffraction. Because the beam time at large-scale facilities is allocated by a competitive proposal process, we rely on the experts in the proposal review panels to share and support this view by granting beam time for further study of the remaining questions at the microscopic level.

INTERVIEW BY DAVID PILE

Daniel Rolles and colleagues have a Letter about the strong ionization of xenon using X-ray pulses on page 858 of this issue.