EMBARGO / SPERRFRIST: Donnerstag, 15. Juni 2017, 20:00 Uhr

A perfect attosecond experiment

Attosecond science techniques are currently revolutionizing ultrafast laser physics research, and enable experiments that provide unprecedented insights into the structure and time-dependent dynamics of electrons in atoms, molecules and condensed phase systems. In a new experiment, physicists from Waseda University (Japan), the National Research Council (Canada) and the Max-Born Institute (Germany) have used attosecond science techniques to fully characterize the quantum mechanical wave function of an electron that is formed by photoionization. The work, reported in Science, is the first example of a "perfect" experiment using attosecond technology.

The development of quantum mechanics in the early part of the last century forced scientists to accept that at the microscopic level matter behaves according to physical laws that are altogether different from the physical laws that apply in our macroscopic world. In the microscopic world concepts like the uncertainty principle play a role, posing limits on the precision with which certain properties of tiny particles, such as their position and space, can simultaneously be measured. Quantum mechanics furthermore introduced wave-particle duality, meaning that the behavior of tiny particles can sometimes better be understood by considering the particles as waves.

These counterintuitive manifestations of quantum mechanics are due to the fact that every measurement that is performed on a quantum mechanical system only gives one out of a huge range of possible outcomes. The likelihood to measure a certain outcome is determined by a probability distribution that derives from the fundamental entity in quantum mechanics, the wave function. The wave function itself is not directly measurable, although strategies can be devised whereby multiple measurements performed on a quantum system lead to a complete characterization of the wave function.

In a paper published in Science (Villeneuve et al., "*Coherent Imaging of an Attosecond Electron Wave Packet*"), a novel approach is presented for the complete characterization of an atomic wave function using novel ultrafast lasers that have only been developed in the last few years. In the measurement, the scientists characterize the wave function of an electron that is released from a Neon atom as a result of the interaction of the atom with a series of laser pulses.

Electrons are elementary particles that are responsible for everyday things like electricity. They are characterized by several properties, such as one unit of (negative) charge, and an angular momentum, which is a vector that characterizes the rotation of the electron around the center of the atom. A slow rotation or a rotation close to the positive core of the atom, correspond to a low angular momentum, whereas a fast rotation or a rotation far away from the core imply a high angular momentum. The laws of quantum mechanics dictate that the angular momentum can only have certain distinct magnitudes. Accordingly, angular momentum states are called "s", "p", "d" and "f" for angular momentum quantum numbers l=0-3. In addition to the magnitude of the angular momentum, the length of the projection of the angular momentum vector onto a chosen laboratory frame axis (e.g. the polarization axis of the laser used in the experiment), characterized by the magnetic quantum number m, affects the outcome and interpretation of laboratory experiments.

In their paper, the scientists managed to accomplish a complete characterization of the wave function of the ionized electron, which contains contributions from angular momenta up to a value l=3 (i.e. s, p, d and f-contributions). Each of these angular momentum states is contained in the wave function with a specific amplitude, meaning a magnitude and a phase. In the experiment, these magnitudes and phases are determined by carrying out a series of interference experiments. Interference experiments exploit the

wave-like character of quantum mechanical particles. Just like two water waves that cross each other can extinguish or enhance each other, so too can interference between different parts of a quantum mechanical wave function lead to an enhanced or a reduced probability to detect the particle at a particular place or with a particular speed. By performing a series of interference experiments under different conditions, pairwise interferences could be observed between the s- and d-part of the wave function, between the p- and the f-part, and finally, between all four components combined (see Figure 1). Accordingly, an exact and complete mathematical expression was obtained for the wave function of the ionized electron.

A crucial component in the accomplishment of this unique feat was the use of attosecond laser pulses (1 as = 10^{-18} s). Attosecond pulses are the shortest laser pulses that can be produced in state-of-the-art laser laboratories. They are produced in a process called “high-harmonic generation”. Here, an atomic gas is exposed to an intense infrared laser that typically has a duration in the femtosecond (1 fs = 10^{-15} s) range. If the intensity of the infrared laser is high enough, the laser can pull electrons out of the atoms, which are subsequently accelerated by the oscillatory electric field of the infrared laser. Some of the accelerated electrons collide with the atoms from which they were previously removed. When this happens, the electron may be re-absorbed by the atom. All the energy that has been invested in the ionization and acceleration of the electron is then released in the form of a very energetic light particle (i.e. photon in the extreme ultra-violet (XUV) or soft X-ray part of the wavelength spectrum). Since the different steps in the high-harmonic generation process all occur on a timescale that is short compared to the duration of one optical cycle of the infrared laser (typically, just a few femtoseconds), this XUV/X-ray light appears in the form of a short – i.e. attosecond – pulse.

In the experiment the researchers used attosecond XUV pulses to ionize the Neon atoms. When only the attosecond pulse was fired in the experiment, a combination of s- and d-type electrons were formed, whose amplitude and relative phase could be determined from their angular distribution (see Figure 1A). When the ionization by the attosecond pulse was performed under conditions where beside the attosecond laser a replica of the infrared laser was present in the experiment, the amplitude and relative phase of the p- and f-components could be extracted (see Figure 1B). Finally, when the attosecond pulses were generated using a two-color laser field (both the afore-mentioned infrared laser and a copy of this laser with half the wavelength) the amplitude and relative phase of all four components (s, p, d and f) could be determined. The results of the experiment and the determination of the amplitude and phase of all angular momentum components are shown in Figures 1C and 1D. The clearly visible six-fold structure is caused by the dominant contribution of the f-orbital with m=0, which is produced by XUV+IR ionization. By the coherent addition of a contribution from the totally symmetric s-orbital (produced by XUV-only ionization), and changing the delay between the XUV and the IR pulse, an oscillation up and down along the vertical laser polarization axis is induced, revealing the phase of the f-orbital contribution.

The experiment is what atomic physicists consider a “complete” experiment, yielding a complete mathematical description of the wave function of the ionized electron, and is the latest example of how attosecond science techniques are currently revolutionizing ultrafast laser physics research. With the present work, this research has for the first time reached a state of perfection.

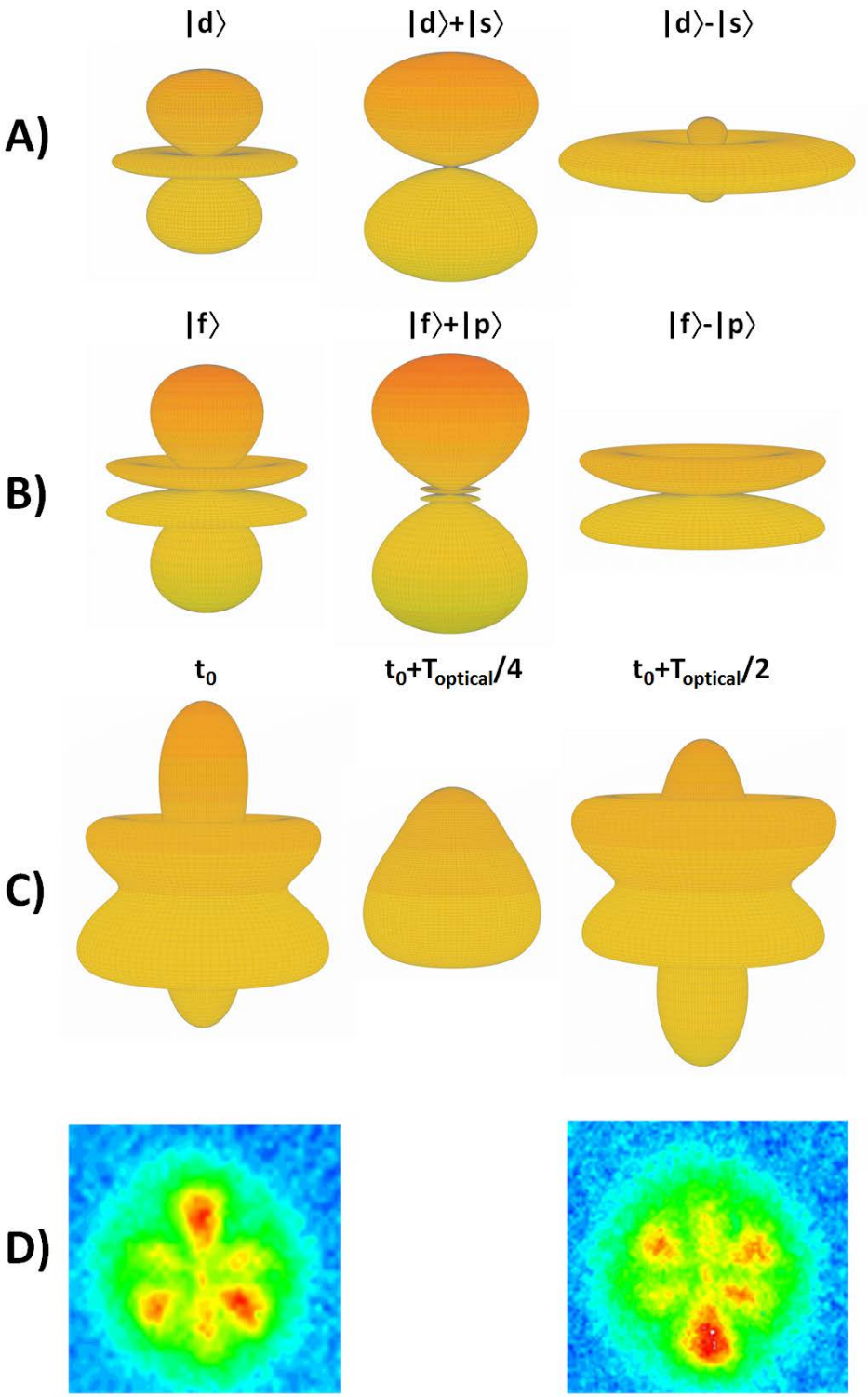

Figure 1: A) XUV-only ionization produces an electron that is in a state characterized by "s" and "d" angular momentum, where a measurement of the electron angular distribution yields a determination of their relative amplitude and phase; B) XUV+IR ionization produces an electron that is in a state characterized by "p" and "f" angular momentum, where a measurement of the electron angular distribution once more yields a determination of their relative amplitude and phase; C) combined XUV-only and XUV+IR ionization produces an electron wave function containing both "s", "p", "d" and "f" contributions. The interference between these angular momentum components evolves with the delay between the XUV pulse and the co-propagating IR pulse. The large contribution of the "f" component is clearly visible in the first and last image; D) Measured electron momentum images at two time delays between the XUV pulse and the co-propagating IR pulse (corresponding to the first and last image in C). The reported experiment provides a complete determination of the relative amplitude and phases of all angular momentum components and thus represents a "perfect" experiment.