Time Delay in Photoemission

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One of the projects with which the Atomic and Molecular Physics group (ATMOPH) of our institute has been involved during the past two years has to do with a crucial *attosecond physics* experiment on *time delay in photoemission*, that was developed and carried out in the Max-Planck-Institut für Quantenoptik (MPIQ) (Garching, Germany), under the directorship of F. Krausz. Our work was done in collaboration with Dr. V. Yakovlev of the MPIQ.

The fundamental question in such an experiment is whether, due to the nature of electronic structure and the details of matter-field interaction, the emission of electrons from different (sub)shells upon the absorption of an ultrashort pulse of high-energy photons occurs at different moments. This question implies that both the experiment and the theory must be able to *time-resolve* the photoemission process of, say, two electrons of different energies at the hyper-short scale of attoseconds.

Until recently, the only observable in photoelectron spectroscopy had been the photoionization cross-section measured as a function of photon energy and observation direction. This corresponds to measuring the probability that quantum mechanics assigns to transitions from a bound atomic state to states where a photoelectron propagates toward the detector with certain energy. The quantum phase of the probability amplitude, the energy derivative of which defines, according to theory, the emission timing, had remained hidden from experimental access.

In the absence of resonances, the rate of photoemission is generally believed to instantly follow temporal variations of the incident light intensity. A delay in the formation of an exiting electron wave packet due to interelectronic interactions, if it exists, would change the timing of ejection of the electron pulse with respect to the arrival of the photon pulse. In turn, this would compromise the accuracy of setting the zero of time for clocking microscopic processes on the atomic time scale. This timing is of fundamental importance because – at present – attosecond light pulses [1-5] constitute the most precise trigger for timing electronic processes. Hence, the zero of time in atomic-scale *chronoscopy*, is to coincide with the arrival time of this photon pulse. Any unknown delay Δt in the emission of the photoelectron packet would result in a shift of the origin of the time arrow and would falsify clocking of the microscopic motion under scrutiny.

The measurements in [5] revealed a delay of 21 ± 5 attoseconds in the emission of electrons liberated from the 2*p* orbitals of neon atoms with respect to those released from the 2*s* orbital by the same 100-eV light pulse.
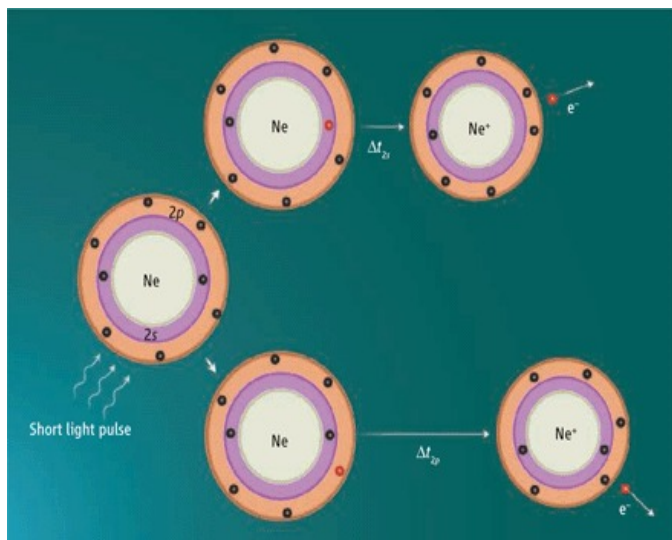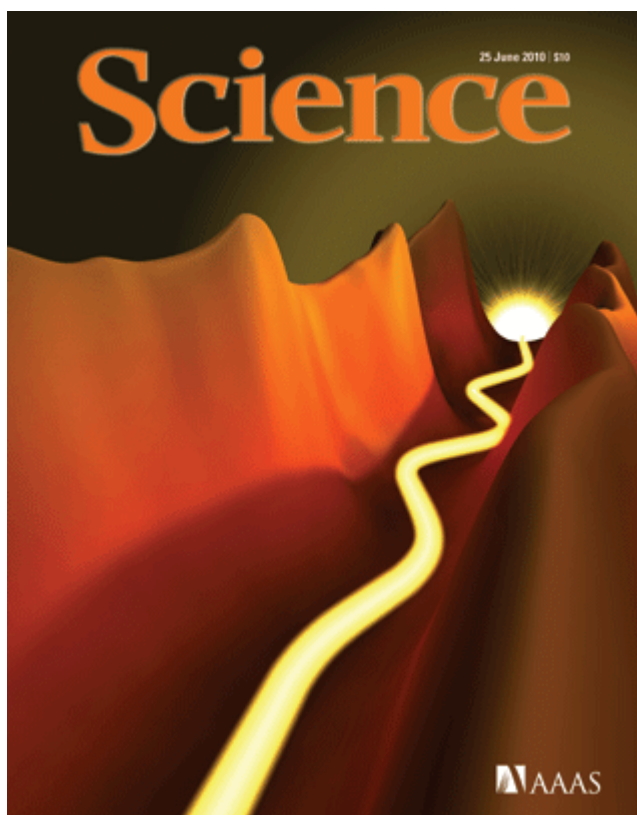
Schematic diagram of a photoemission process for Ne. An incoming photon of an ultrashort light pulse is absorbed by either a 2s (top row) or a 2p (bottom row) electron. After photoabsorption, the electron escapes, while the orbitals of the other electrons adjust to the new surroundings as the atom becomes an ion. This adjustment leads to a time delay Δt in the emission of the electron, which is longer for emission of a 2p electron than for emission of a 2s electron.

In short, ultrafast spectroscopy and multielectron calculations reveal complex electron dynamics occurring just before an atom emits a photoelectron.

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As part of the Athens-Munich cooperation, the Athens ATMOPH group obtained quantitatively absolute and relative time delays from ab initio computations which take into account electron correlations before and after photoionization. The theoretical and computational framework for such many-electron calculations, developed by the group, is known as "***State-specific expansion approach***" (SSEA), [6, 7]. A discrepancy remains between the experimental measurements and the theoretical results, which makes this area of research even more challenging.

The experimental-theoretical results were published in SCIENCE, June 2010 [5].

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