Atomic photoionization: When does it actually begin?

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Synopsis We analyze a time delay of one- and two-electron photoemission from an atom after absorption of an attosecond XUV pulse. We establish this delay by solving the time dependent Schrödinger equation and by subsequent tracing of the field-free evolution of the photoelectron wave packet. This delay can also be related to the energy derivative of the phase of the complex photoionization amplitude which makes the time delay a sensitive probe of many-electron correlation.

Among other spectacular applications of the attosecond streaking technique, it has become possible to determine the time delay between subjecting an atom to a short laser pulse and subsequent emission of the photoelectron [1]. This observation opened up a question as to when does atomic photoionization actually begin [2].

We address this question by solving the time dependent Schrödinger equation (TDSE) and by carefully examining the time evolution of the photoelectron wave packet. In this way we establish the apparent "time zero" when the photoelectron leaves the atom. At the same time, we provide a stationary treatment to the photoionization process and connect the observed time delay with the quantum phase of the dipole transition matrix element, the energy dependence of which defines the emission timing.

As an illustration of our approach, we consider the valence shell photoionization of Ne [3] and double photoionization (DPI) of He [4]. In Ne, we relate the opposite signs of the time delays $t_0^{2s} < 0$ and $t_0^{2p} > 0$ (see Fig. 1) with energy dependence of the p and d scattering phases which is governed by the Levinson-Seaton theorem. In He, we demonstrate that an attosecond time delay measurement can distinguish between the two leading mechanisms of DPI. The shake-off (SO) mechanism is driven by a fast rearrangement of the atomic core after departure of the primary photoelectron. The characteristic SO time is inversely proportional to the energy difference of the singly ionized state in the frozen atom and the fully rearranged ion $\tau_{so} \simeq (E_{atom}^+ - E_{ion}^+)^{-1} \simeq 20$ as for the helium atom in its ground state. In contrast, the knock-out (KO) mechanism involves repeated interaction of the primary photoelectron with the remaining electron bound to the singly charged ion. The characteristic time of this process can be roughly estimated from the correlation energy

of the ground state taken as the difference between the exact energy and the non-correlated energy with the corresponding effective charge $\tau_{ko} \simeq (E_{Z^*} - E_{exact})^{-1} \simeq 200$ as. The explicit TDSE calculations confirm that the time delay in DPI of He varies within these limits as the cross-over between the shake-off and knock-out mechanisms takes place.
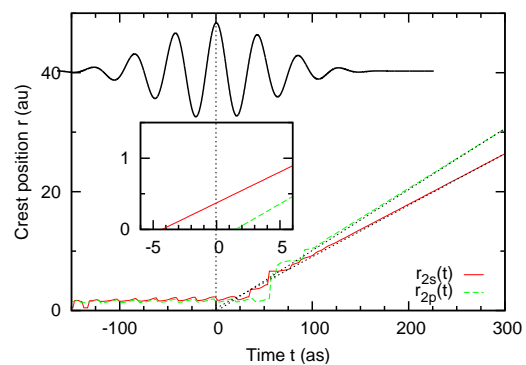

Figure 1. The crest positions of the $2s$ and $2p$ wave packets as functions of time measured from the center of the attosecond XUV pulse ($t = 0$). The crest position after the pulse end is fitted with a straight line to simulate a free propagation. In the inset, extrapolation of the free propagation inside the atom is shown. The intersect of the straight lines with the abscissa gives the corresponding time delays $t_0^{2s} < 0$ and $t_0^{2p} > 0$

References

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