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Photoionization dynamics: Transition and scattering delays

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Synopsis The advent of attosecond science, with main objectives to probe and to control ultrafast dynamics in quantum systems, brought in the past few years a fresh look on photoemission. This very fundamental process was indeed recently revisited in the time domain, in a series of pioneering experiments [1] that were soon accompanied by an intense theoretical activity devoted to the interpretation of the reported "photoemission delays". At the conference, we will present some contributions of our group to this topic [2], with a focus on the characterization of photoemission dynamics in terms of "scattering delay" and "transition delay" as evidenced in numerical experiments performed on simple model systems.

Resolving electron motion in atoms and molecules on its natural attosecond (as) scale is way beyond the temporal resolution of available detection devices. The techniques developed to achieve such sub-fs resolution thus rely on interferometric setups, and the reported times are actually group delays derived from phase measurements, involving coherent photoemission processes. Therefore, the analysis of the experimental data and the related theoretical development ask for rigorous and unambiguous definitions and interpretations of these phases, and of the inferred group delays.

It is now accepted that a "scattering delay" [4] affects the dynamics of any photoemission process. However, the simplicity of the underlying physics is not fully recognized yet. Formally, such delays are imprinted in the phase shifts of the photoelectron wave-functions, which are commonly expressed on the basis of incoming waves. In this framework, the "scattering phase" associated to photoemission appears as the argument of the transition amplitude, thus obscuring the significance of the delay – which may be misinterpreted for example as a transition duration. Here, we will present the benefits of working with the continuum wave-functions selected by the transitions (SCWF) [3], which (i) carry all the information related to the continuum reached by photoabsorption, (ii) are defined independently of the arbitrary basis one chooses to work with and (iii) are real valued for single-photon transitions. They provide a clear-cut interpretation of the scattering delays.

In higher order processes, the SCWF comes with an additional complex phase, as soon as the transition is resonant. We will show that the group delay associated with that phase can now be interpreted as a "transition delay", and how it can be accessed experimentally in a straightforward reinterpretation of the RABBIT interferometric technique, initially designed for the characterization of coherent XUV pulses [5].

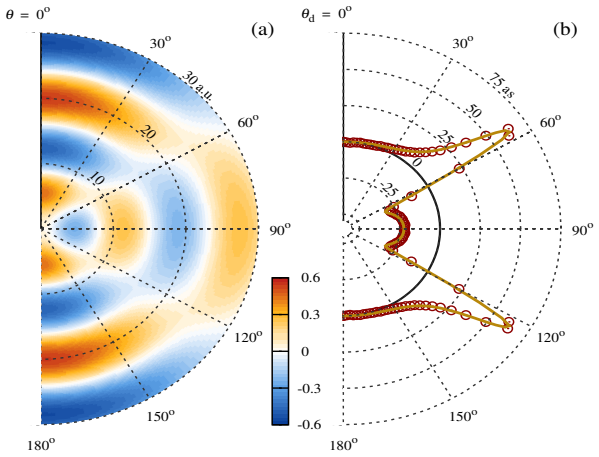

Figure 1. $He^+(2p_0)$ @ 17.06 eV. (a) Real valued SCWF . (b) Ionization delays against the detection direction θ_d: time-dependent simulations (circles); scattering group delays (full line). The delays are defined and computed with $\theta_d = 0$ as reference.

References

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