Two-Photon Transition Delays Inferred from Interferences with One-Photon Reference Transitions

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Photoemission is one of the most fundamental processes that attosecond science allows revisiting in the time-domain. Over the past 15 years, a variety of pump-probe schemes were developed to access either the photoelectron scattering dynamics upon absorption of light , or the dynamics of the transition itself when more than one photon is involved . Temporal insight on this fundamental quantum process is achieved through interferometric measurements of the photoelectron spectral phase, as notably done in schemes derived from the RABBIT method.

In the present study, we developed an alternative interferometric scheme to access the spectral phase of resonant 2-photon transition amplitudes. It uses single-photon transitions as a reference, rather than 2-photon transitions as in RABBIT-like approaches, leading to a number of advantages. The relevance of our original approach is demonstrated in the case of the two-photon XUV+IR ionization of helium through the intermediate resonant states $1snp$ with $n = 3, 4, 5, 6$.

We use a single harmonic beam (H15) overlapped with its generating IR field to induce a 2-photon transition towards the continuum, creating the photoelectron wavepacket (EWP) of interest. The latter is structured by the transient resonances the intermediate $1snp$ states, leading to a series of π rad spectral phase jumps . Besides, the interaction leads to a significant coherent population of the $1snp$ states through 1-photon transition. After a controlable delay τ, we send an additional IR pulse, that transfers some population from the intermediate bound states to the continuum, again by a 1-photon transition. This creates a reference EWP ending up at the same energy than the probed 2-photon EWP. In contrast to the resonant two-photon transition, the sequence of one-photon reference transitions produces a rigorously unstructured EWP.

Interferences between the probed and reference EWPs encode their relative phase that has two contributions: i) the *known* difference of phases accumulated during the delay τ by the $1snp$ and the continuum states, and ii) the phase of the probed resonant two-photon transition. By measuring the τ- dependent photoelectron spectrum over hundreds of femtoseconds, we can access the latter phase with both high accuracy and high spectral resolution. A significant advantage of this interferometric scheme is that the reference EWP has the same partial wave decomposition as the studied EWP, which makes it an ideal probe maximizing the contrast of the interferences regardless of the electron emission direction.