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Control of photoemission delay in resonant two-photon transitions

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Synopsis In contrast to one-photon transitions and non-resonant multiphoton transitions, time delay in resonant multi-photon electron emission can exhibit large positive and negative values that have no scattering equivalent, due to the interference of multiple ionization paths.

The photoelectron emission time delay τ associated to one-photon absorption, which coincides with half the Wigner delay τ_{W} [1, 2] experienced by an electron scattered off the ionic potential, is a fundamental descriptor of the photoelectric effect [3]. Although it is hard to access directly from experiment, it is possible to infer it from the time delay of twophoton transitions, $\tau^{(2)}$, measured with attosecond pump-probe schemes [4, 5], provided that the contribution of the probe stage can be factored out [6]. In absence of resonances, τ can be expressed as the energy derivative of the one-photon ionization amplitude phase, $\tau = \partial_E \arg D_{Eg}$, and, to a good approximation, $\tau = \tau^{(2)} + \tau_{cc}$, where τ_{cc} is associated to the dipole transition between Coulomb functions. Here we show that, the presence of a resonance, the correspondence between τ and $\partial_E \arg D_{Eg}$ is lost. Furthermore, while $au^{(2)}$ can still be written as the energy derivative of the two-photon ionization amplitude phase, $\partial_E \arg D_{Eg}^{(2)}$, it does not have any scattering counterpart. Indeed, $\tau^{(2)}$ can be much larger than the lifetime of an intermediate resonance in the two-photon process, or more negative than the lower bound imposed on scattering delays by causality. Finally, we show that $\tau^{(2)}$ is controlled by the frequency of the probe pulse, $\omega_{\rm IR}$, so that by varying $\omega_{\rm IR}$, it is possible to radically alter the photoelectron group delay [7].

To draw our conclusions, we use a general finitepulse analytical model for resonant multi-photon ionization amplitudes [8, 9], which we have developed and applied successfully to a number of attosecond interferometric experiments and simulations [8, 10, 5]. We also offer selected examples in the case of helium, using parameters obtained from accurate ab initio simulations of the photoionization of the helium atom [11].



Figure 1. Sketch of the Wigner time delay. As the electron wave packet (green) scatters off the potential, it acquires a phase shift η as compared to a reference free electron (orange). This phase shift is related to the time lapsed between the detection of the scattered electronic wave packet and the detection of the unscattered free electron, called the Wigner time delay.

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