Partial-wave transitions in attosecond time delays

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In our recent work [1] we tackle the challenge of accurately interpreting photoionization delays in atoms and molecules measured using multiphoton interference techniques, with a particular focus on the Reconstruction of Attosecond Beating by Interference of Two-photon Transitions (RABITT). Specifically, we investigate implications of a "two-harmonic RABITT" experiment performed in argon, featuring a single central sideband, two mainbands and two outer sidebands, each alone well usable for extraction of compatible timing information.

Current methods, which rely on separation of the measurable delays into Wigner-like delay and universal continuum-continuum delay τ_{cc}, fail at low photoelectron kinetic energies and fail to capture the correct angular dependencies. To address this, we have developed an analytical approach that not only predicts correction terms accurately even at very low energies but also incorporates the angular dependence of the continuum-continuum delay. Our method is computationally efficient and its limits have been validated against state-of-the-art ab initio calculations using the stationary multiphoton R-matrix method [2] and the R-matrix with time-dependence (RMT) [3].

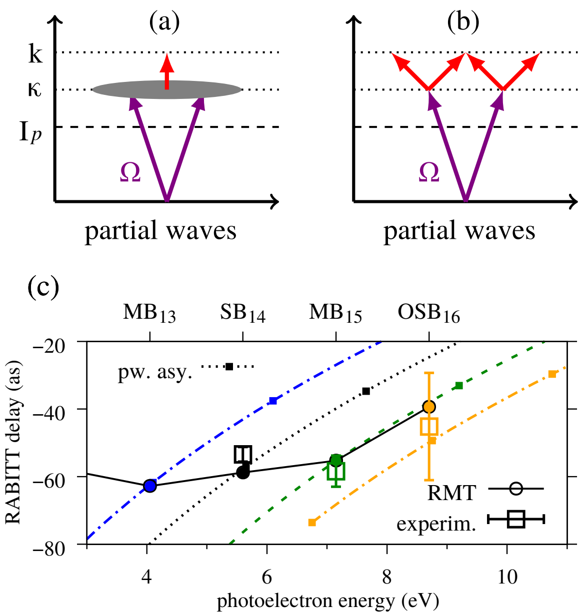

Figure 1: (a) Universal τ_{cc}. (b) Partial-wave resolution of amplitudes. (c) Emission-integrated 2H-RABITT delays in argon.

By introducing a partial-wave-resolved continuum-continuum contribution, we achieve a precise modeling of the infrared field's influence on different photoelectron partial waves. This advancement allows for a clearer interpretation of both angularly resolved and angle-integrated RABITT delays, demonstrating excellent agreement with the full above-threshold perturbation theory. Our experimental setup, utilizing a simplified RABITT configuration, effectively isolates higher-order multiphoton pathways, offering a promising method for analyzing complex molecular photoionization spectra.

[1] J Benda *et al.*, Phys. Rev. A **111**, 013110 (2025).
[2] J Benda, Z Mašín, Sci. Rep. **11**, 11686 (2021).
[3] A C Brown *et al.*, Comput. Phys. Commun. **250**, 107062 (2020).