Polarization-Induced Molecular Photoionization Delays and Equivalence of RABITT and Streaking Dipole-Laser Coupling

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Photoionization delays provide an intriguing observable that delivers information about the photoionization event, sometimes with greater sensitivity than the photoionization cross sections. Direct measurement of these delays is experimentally inaccessible; however, several methods that employ absorption of further photons have been devised to overcome this limitation. These either make use of classical addition of momenta with the electromagnetic vector potential (attosecond streaking), or take advantage of subtle multi-pathway interference (reconstruction of attosecond beating by interference of two-photon transitions, RABITT). We show that the addition of the second (IR) field allows to study laser-induced electron correlation effects. Theoretical analysis of above-threshold multi-photon ionization has been notoriously difficult and this process is typically solved either in fully time-dependent way or using asymptotic approximation.

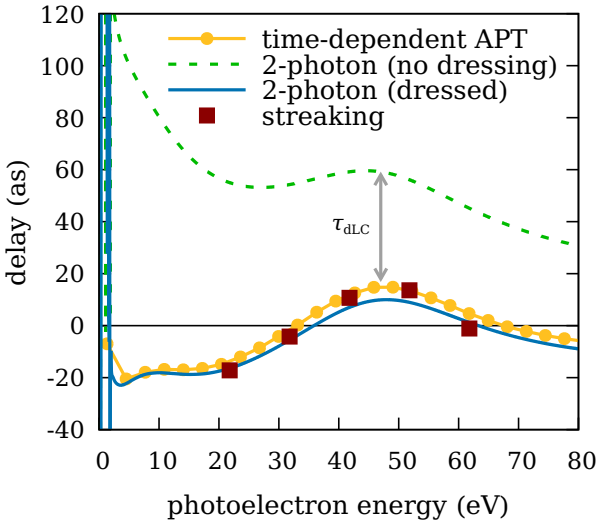

Figure 1: Calculated RABITT and streaking delays in LiH. Parallel configuration, emission from Li end, static exchange model, 780-nm IR field

As a computationally very efficient alternative, a comprehensive time-independent molecular multiphoton theory [1] has been developed recently, which allows one to directly calculate multi-photon ionization amplitudes resolved in ion states and partial waves without the asymptotic approximation.

In this contribution we leverage the strength of the multi-photon method to discover an overlooked contribution to the RABITT sideband delay [2] that arises in the "ion-ion" transition during absorption of the probe IR photon. This delay is significant for residual cation states that are separated by energies comparable to the IR photon energy, and which have non-zero transition dipole.

Additionally, we analyze dynamics of the initial state of strongly polar molecules in the IR field, observing that the IR field dresses the initial state and gives rise to new emission/absorption pathways in RABITT on top of the standard XUV±IR pair [3]. Inclusion of these pathways results in appearance of the dipole-laser coupling delay known from attosecond streaking. Its effect is particularly strong in the molecular frame.

References

[1] J Benda and Z Mašín, Sci. Rep. **11**, 11686 (2021)

[2] J Benda, Z Mašín and J D Gorfinkiel, Phys. Rev. A **105**, 053101 (2022)

[3] J Benda and Z Mašín, arXiv 2209.06676 (2023)