New Contributions to the Molecular Attosecond Delays Revealed by Multi-Photon R-Matrix Theory

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Perturbative multi-photon processes are a frequently used probe of atomic and molecular continua. This includes the Reconstruction of Attosecond Beating by Interference of two-photon Transitions (RABITT) [1] which has become the dominant tool for investigation of attosecond time-delays in atoms and molecules, see *e.g.* [2]. The rapid development of X-ray free electron lasers can be expected to increase the range of phenomena studied by multi-photon above- and below-threshold ionization. This goes in hand with demands on the theory to provide the corresponding photoionization amplitudes. Nevertheless, up to recently, direct calculation of the involved multi-photon matrix elements has been a challenge, especially for complex multi-electron atoms and molecules.

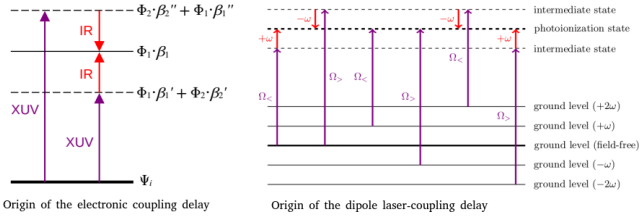

Figure 1: Multiple pathways contributing to the RABITT signal

The commonly used approach is to study multi-photon processes using explicitly time-dependent calculations, see *e.g.* [3]. Nevertheless, those calculations are naturally highly computationally demanding and don't allow us to use the state of art models available for single-photon ionization, see *e.g.* [4]. To overcome this limitation we have recently developed a new time-independent (stationary) approach, based on the R-matrix method, for direct calculation of multi-photon matrix elements for complex molecules [5]. While allowing to employ highly sophisticated multi-channel models (including hundreds of ionic states), it expands the range of tools available for analysis of various multi-photon phenomena.

Applied to RABITT, the stationary method led to the discovery of two new contributions to the RABITT delay: the electronic coupling delay and the dipole laser coupling delay [6,7], see Fig.1. Typically, RABITT delays are used to provide information only about the field-free scattering delay but the new contributions show the potential of the method to study laser-driven electron-molecule interactions too.

In this contribution we will discuss our results for non-polar and polar molecules, including a comparison with recent experiments [8], where we showed that the RABITT delays can be represented using complex β parameters, similarly to the atomic case [9].

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