Origins of attosecond delays

Hakon Volkmann[1], Vinay P. Majety[1,2] and Armin Scrinzi[1]

[1] *Ludwig Maximilian University, Munich, Germany*
[2] *Indian Institute of Technology Tirupati, Tirupati, India*

E-mail: armin.scrinzi@lmu.de

ABSTRACT Using the comparatively simple example of photoemission from the CO_2 molecule, we identify various sources of photo-emission delays. We find that long- and short-range scattering, non-perturbative effects, and dynamical correlation all contribute on the level of 10 or more attoseconds. Consequently, any interpretion of attosecond precision measurements in any material mandates careful consideration of these effects.

The impressive precision of attosecond measurement needs to be matched by an unambiguous interpretation and use of the results. The enticing promise of imaging internal electronic motion faces serious challenges when observations are to be related to specific internal mechanisms. We explore the relation between the *ab initio* based simulation of single-electron emission from an isolated CO_2 molecule to various simplifying models. Fundamentally, on the 10 as level, no manageable model appears to reproduce the correlated multi-channel calculations using accurate quantum chemical wave functions (hybrid antisymmetrized Coupled Channels, haCC [1]).

Some reasons for this can be identified and partially remedied by refining the single-elecron models. The well-known "universal" Coulomb-delay can be extended to a more general "long-range"-delay that accounts for the rotational asymmetry of the system and that is accessible to efficient simulation. In contrast, "short-range"-delays by the local environment prior to emission are in general determined by correlations for which no unambiguous description other than large scale simulation is known at present. Also, in molecules laser intensities need to be kept low, as non-perturbative effects set in early and add to the delays on the >10 as level. For CO_2 such effects appear at intensities from $10^{12} – 10^{13}$ W/cm^2.

The haCC method will be introduced and the above observations will be demonstrated in relation to a recent experiment [2].

References:

[1] V. P. Majety, A. Zielinski and A. Scrinzi. N. J. Physics, New J. Phys. 17:063002, (2015).
[2] M. Lucchini *et al.*, submitted to Physical Review A.