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CFEL-bldg. 99, seminar room IV

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The role of the carrier envelope phase for molecular dynamics: A theoretical perspective

Control of chemical reactions is in the very heart of chemistry. The increasing ability to control laser light nearly on demand adds a new dimension for this endeavour. Light sources in the few femtosecond regime, which can be modulated in frequency, phase and polarization offer the possibility of coherent interaction with the quantum nature of the molecular system to steer chemical reactions. The resulting signals and optimized fields are often not easy to interpret and theoretical support or guidance is vital for the success and understanding of the mechanism.

Chemical reactions are intrinsically accompanied by electron motion, which can, in principal, be traced by strong few-cycle pulses in the (sub-) femtosecond regime. The additional control parameter is the adaptable carrier envelope phase (CEP), which basically controls the sign of the superposition in the created electronic wave packet. From our analysis, we extract the systems requirements defining the time window for control via electronic coherence. We mapped out a photoreaction scenario in which the guidance of an electronic wavepacket through a conical intersection allows to control the reaction outcome [1,2]. However, in polyatomic molecules the situation becomes much more complex. The CEP not only affects the electronic but also the vibrational motion. We could demonstrate this for unsaturated hydrocarbons. Here the CEP controls the superposition sign in the vibrational wave packets and allows to differentiate between the terminal left and right carbon-hydrogen bond of these symmetric molecules [3,4]. The same mechanism can explain even more complex reactions like isomerizations and seems to be a common feature.

1. P. von den Hoff et al., IEEE Journal of Selected Topics in Quantum Electronics 18, 119-129 (2012).
2. M. Kling et al., Phys. Chem. Chem. Phys. 15, 9448-9467 (2013).
3. A. S. Alnaser et al., Nat. Commun. 5, 3800 (2014).
4. M. Kübel et al., Phys. Rev. Lett. 116, 193001 (2016).

