CFEL SCIENCE

Colloquium

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FLASH HALL, Seminar Room (28c)

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Imaging the Ultrafast Dynamics of Polyatomic Molecules: Molecular Frame Photoelectron Imaging, Quantum Control, Attosecond Strong Field Physics

The most general molecular dynamic processes involve the coupled flow of both valence electronic charge and vibrational energy. Time-Resolved Photoelectron Spectroscopy (TRPES) is a powerful probe of ultrafast non-adiabatic dynamics in polyatomic molecules, as it simultaneously observes both electronic and vibrational dynamics [1]. Time-Resolved Coincidence Imaging Spectroscopy (TRCIS) images the kinematically complete 3D momentum vectors of both photoions and photoelectrons in coincidence and as a function of time. This allows for study of the time evolution of both scalar and vector correlations during molecular processes such as chemical reaction. One important Molecular Frame vector correlation permits dynamical imaging of electronic wavefunction evolution [2]. Another general method, based on nonresonant laser pre-alignment, also permits direct time-resolved imaging of electronic dynamics in the Molecular Frame during a chemical reaction [3].

As electric forces underlie all of solid state and molecular physics, controlling material processes with electric fields is a natural approach. We discuss Quantum Control using the non-resonant Dynamic Stark Effect as a new and powerful tool. Dynamic Stark Control (DSC), which uses the electric field intensity envelope of a laser pulse rather than its frequency content, can be used to control molecular dynamics without any net absorption of light [4]. We also illustrate the use of DSC in creating molecular frame alignment. Using a novel pulse sequence, we demonstrate full 3D, field-free, axis alignment of an asymmetric top rotor – transiently 'fixing the molecule in space' in all three dimensions in order to make a field-free measurement [5].

As laser fields get stronger still, a new laser-matter physics emerges for polyatomic systems wherein both the single active electron picture and the adiabatic electron response, both implicit in the standard atomic models, can fail dramatically. A new Nonadiabatic Multi-Electron (NME) dynamics emerges [6] and has important consequences for all strong field processes in polyatomic molecules, including high harmonic generation (HHG) and attosecond spectroscopy. Multiple electronic continua do participate in the HHG spectroscopy of polyatomic molecules [7]. We present a new experimental method, Channel-Resolved Above Threshold Ionization (CRATI), which directly unveils the electronic continua participating in the sub-cycle molecular strong field response.

[1] Nature 401, 52, (1999). **[2]** Science 311, 219 (2006). **[3]** Science 323, 1464 (2009). **[4]** Science 314, 278 (2006). **[5]** Phys.Rev.Lett. 97, 173001 (2006); 97, 173001 (2006). **[6]** Physical Review Letters 86, 51 (2001); 93, 203402 (2004); 93, 213003 (2004). **[7]**. Science 322, 1207 (2008)

Host: Oriol Vendrell, CFEL