

MAR17-2016-020035

Abstract for an Invited Paper
for the MAR17 Meeting of
the American Physical Society

Earle K. Plyler Prize Lecture: The Three Pillars of Ultrafast Molecular Science - Time, Phase, Intensity¹

ALBERT STOLOW, University of Ottawa & National Research Council

We discuss the probing and control of molecular wavepacket dynamics in the context of three main ‘pillars’ of light-matter interaction: time, phase, intensity. Time: Using short, coherent laser pulses and perturbative matter-field interactions, we study molecular wavepackets with a focus on the ultrafast non-Born-Oppenheimer dynamics, that is, the coupling of electronic and nuclear motions. Time-Resolved Photoelectron Spectroscopy (TRPES) is a powerful ultrafast probe of these processes in polyatomic molecules because it is sensitive both electronic and vibrational dynamics [1, 2]. Ideally, one would like to observe these ultrafast processes from the molecule’s point of view – the Molecular Frame – thereby avoiding loss of information due to orientational averaging. This can be achieved by Time-Resolved Coincidence Imaging Spectroscopy (TRCIS) which images 3D recoil vectors of both photofragments and photoelectrons, in coincidence and as a function of time, permitting direct Molecular Frame imaging of valence electronic dynamics during a molecular dynamics [3]. Phase: Using intermediate strength non-perturbative interactions, we apply the second order (polarizability) Non-Resonant Dynamic Stark Effect (NRDSE) to control molecular dynamics without any net absorption of light [4]. NRDSE is also the interaction underlying molecular alignment and applies to field-free 1D of linear molecules and field-free 3D alignment of general (asymmetric) molecules [5]. Using laser alignment, we can transiently fix a molecule in space, yielding a more general approach to direct Molecular Frame imaging of valence electronic dynamics during a chemical reaction [6, 7]. Intensity: In strong (ionizing) laser fields, a new laser-matter physics emerges for polyatomic systems [8] wherein both the single active electron picture and the adiabatic electron response, both implicit in the standard 3-step models, can fail dramatically. This has important consequences for all attosecond strong field spectroscopies of polyatomic molecules, including high harmonic generation (HHG) [9]. We discuss an experimental method, Channel-Resolved Above Threshold Ionization (CRATI), which directly unveils the electronic channels participating in the attosecond molecular strong field ionization response [10]. [1] Nature 401, 52, (1999). [2] Chemical Reviews 104, 1719 (2004). [3] Science 311, 219 (2006). [4] Science 314, 278 (2006). [5] Physical Review Letters 94, 143002 (2005); 97, 173001 (2006). [6] Science 323, 1464 (2009). [7] Nature Physics 7, 612 (2011). [8] Physical Review Letters 86, 51 (2001); 93, 203402 (2004); 93, 213003 (2004). [9] Science 322, 1207 (2008). [10] Science 335, 1336 (2012); Physical Review Letters 110, 023004 (2013)

¹This work was supported by the National Research Council of Canada and the Natural Sciences Engineering Research Council.