Abstract Submitted
for the DAMOP17 Meeting of
The American Physical Society

Non-Born-Oppenheimer dynamics in small molecules - connecting experiment and theory KIRK A LARSEN, ELIO G CHAMPENOIS, University of California, Berkeley and Lawrence Berkeley National Laboratory, LOREN GREENMAN, Lawrence Berkeley National Laboratory, C WILLIAM MCCURDY, University of California, Davis and Lawrence Berkeley National Laboratory, THORSTEN WEBER, DANIEL S SLAUGHTER, Lawrence Berkeley National Laboratory — A femtosecond pulse of VUV light can coherently excite a wavepacket in a molecule that then evolves on an excited state potential energy surface (PES). This can lead to non-Born-Oppenheimer dynamics via the coupling of electronic and nuclear degrees of freedom near conical intersections of PESs. Even for molecular systems comprised of just a few atoms, its PESs exist in a highly dimensional space. This can make interpreting time-resolved VUV/XUV pump-probe experiments on molecules very challenging, as it can be difficult to ascertain which dimensions of the PESs play central roles in driving the quantum dynamics. Here, I present preliminary results from time-resolved VUV pump-probe electron and ion momentum imaging experiments on simple polyatomic molecules, such as NH_3, using a high harmonic generation light source, and discuss the use of parallelized ab initio time-independent molecular electronic structure calculations to give insight into the results of these experiments. In certain cases, even for molecules with highly dimensional PESs, time-independent theory can elucidate observed wavepacket motion. I will also present time-dependent dynamics calculations and discuss the strengths and weaknesses of these approaches to understanding the results our experiments.

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Date submitted: 27 Jan 2017 Electronic form version 1.4