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Coherent Dynamics Following Strong Field Ionization of Polyatomic Molecules ARKAPRABHA KONAR, YINAN SHU, VADIM LOZOVOY, JAMES JACKSON, BENJAMIN LEVINE, MARCOS DANTUS, Michigan State University — Molecules, as opposed to atoms, present confounding possibilities of nuclear and electronic motion upon strong field ionization. The dynamics and fragmentation patterns in response to the laser field are structure sensitive; therefore, a molecule cannot simply be treated as a "bag of atoms" during field induced ionization. We consider here to what extent molecules retain their molecular identity and properties under strong laser fields. Using time-of-flight mass spectrometry in conjunction with pump-probe techniques we study the dynamical behavior of these molecules, monitoring ion yield modulation caused by intramolecular motions post ionization. The delay scans show that among positional isomers the variations in relative energies, amounting to only a few hundred meVs, influence the dynamical behavior of the molecules despite their having experienced such high fields (V/Å). Ab initio calculations were performed to predict dynamics along with single and multiphoton resonances in the neutral and ionic states. We propose that single electron ionization occurs within an optical cycle with the electron carrying away essentially all of the energy, leaving behind little internal energy in the cation. Evidence for this observation comes from coherent vibrational motion governed by the potential energy surface of the ground state of the cation. Subsequent fragmentation of the cation takes place as a result of further photon absorption modulated by one- and two-photon resonances, which provide sufficient energy to overcome the dissociation energy.

Arkaprabha Konar
Michigan State University

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