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Control of Electron Localization in Molecular Dissociation by a Midinfrared Two-Color Laser Field

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Intense femtosecond (10-15s) laser pulses can induce molecular dissociation via a broad range of pathways involving different electronic states. Furthermore, breaking the symmetry of a laser field allows control of these processes via the steering of electron motion, which is accomplished by superimposing electronic states of different parity. The symmetry break can be obtained, for example, by controlling the carrier envelope phase (CEP) of a few-cycle laser pulse, or by combining two laser beams of different wavelength, whose relative phase is adjusted. Both techniques are compared for mid-infrared (MIR) laser pulses (1.8 μm) generated from a laser source that was developed at the Advanced Laser Light Source (ALLS). They lead in many cases to a significant control gain compared to an excitation wavelength of 800 nm.
We used a MIR two-color (1.8 μm and 900 nm) laser field to ionize and control H2 and D2 dissociations, thus providing an alternative to CEP stable laser source. By varying the relative phase of both colors, asymmetries (i.e., electron localization selectivity) of up to 15% were calculated directly from a time of flight (TOF) mass analysis. By retrieving fragments position and flight time from their initial spatial location, kinetic energy release (KER) asymmetries up to 30% were also evaluated. Dissociation channels such as the bond softening (BS) channel and the recollision excitation (RCE) channel were identified according to pathways suggested in literature. The results demonstrate a higher level of control compared to previous CEP control experiments and suggest the possibility to shape the proper asymmetric electric field for a specific dissociation channel. The method was further used to ionize and control the deprotonation of C2D2 and C2H2.

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