IMAGING PAIR-CORRELATED PREDISSOCIATION OF HYDROGEN-BONDED DIMERS

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Photofragment imaging is used to interrogate state-specific energy flow patterns in predissociation of hydrogen-bonded mixed dimers of polyatomic species. Imaging-based dynamic spectroscopy provides action spectra of photofragments following infrared excitation of selected vibrational transitions in the dimer. Pair-correlated energy distributions are inferred from velocity map images of specific product rovibronic states. Predissociation of dimers of acetylene with hydrogen chloride and ammonia will be discussed, as well as the ammonia-water dimer. Dissociation energies are determined and compared with theory and energy flow patterns are analyzed. The experiments demonstrate that while general propensity rules exist on how energy is coupled to the dissociation coordinate, it is the fine details of how excitation is deposited in the dimer and its subsequent propagation through the two dimer moieties that give rise to state-specific effects. Calculations based on the hard-ellipsoid model illustrate how kinematic constraints can determine points of impact and the ensuing transformation of linear to angular momentum.