TWO-COLOR, LASER-INDUCED GRATING SPECTROSCOPY: A MULTIDIMENSIONAL PROBE OF EXCITED STATE DYNAMICS

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The population of vibrationally excited levels prior to photolysis alters the excited state dissociation dynamics through nuclear motion that is initiated on a ground electronic surface. Accordingly, we perform vibrationally mediated photodissocation to study novel excited state geometries and reaction pathways of small gas phase molecules. In a technique known as two-color laser-induced grating spectroscopy (TC-LIGS), a grating is formed by the interference of two degenerate "pump" beams tuned to an overtone of a light atom oscillator such as a O–H, N–H, or C–H bond. A third photolysis beam crosses the interference region and is diffracted from this grating, giving rise to a zero-background signal beam. Comparison of photoacoustic (one photon) and two-color grating (two photon) spectra, reveals the influence vibrational excitation has on the behavior of an electronically excited molecule.