TRANSIENT TWO-DIMENSIONAL INFRARED SPECTROSCOPY OF METAL CARBONYL PHOTODISSOCIATION DYNAMICS

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The ultrafast photodisocciation of $Mn_2(CO)_{10}$ has been studied using triggered-exchange multidimensional infrared and UV-pump-2DIR-probe methods. The present study is the first to embed non-equilibrium electronic excitation within the three-puslse Fourier transform approach to 2DIR spectroscopy. These new techniques allow for the direct correlation between reactant and product vibrational modes and yield information such as excited state dynamics, quantum yields, and vibrational relaxation pathways. Our results indicate that the reactant vibrational mode at 2013 cm^{-1} gives rise to the photoproduct band centered at 1985 cm^{-1}. Furthermore, strong anharmonic signatures in the two-dimensional data indicate excess excitation energy is dissipated into the product vibrational modes.