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Two-dimensional electronic spectroscopy of cold, controlled systems

FRANK STIENKEMEIER, University of Freiburg

The first two-dimensional electronic spectroscopy (2DES) study of cold molecules (sub Kelvin internal temperature) prepared by helium nanodroplet matrix isolation in a molecular beam apparatus is presented. In contrast to experiments in the liquid/solid phase, our approach allows for the preparation of isolated model systems in well-defined initial quantum states and to study their ultrafast dynamics with high (rovibrational) resolution under the influence of a controlled environment. The principle is demonstrated for high-spin Rb_2 and Rb_3 molecules synthesized on the surface of superfluid helium nanodroplets which are studied with phase-modulated 2DES combined with photoionization. Our 2DES spectra exhibit unprecedented high resolution: molecular spin-orbit states are clearly distinguished and a striking asymmetry between absorption and emission is observed due to the cold initial molecular states. As an intriguing aspect of system-bath interactions, we observe coherent spin dynamics in Rb_2 (symmetry-forbidden in the gas phase) and evidence for a spin-driven chemical reaction in Rb_3 . Both processes have not been reported for these systems before, which confirms the potential of our unique experimental approach. In general, the established combination of 2DES with well-defined model systems exhibiting a controllable coupling to the environment and limited complexity, will allow studying fundamental principles in physics and chemistry and will help to advance theoretical descriptions of multidimensional spectroscopy. Considering the low density in our molecular beam ($\leq 10^7$ cm^{-3}), our experimental setup furthermore opens the possibility to expand 2DES to new fields, e.g. ultracold atoms and molecules, trapped ions or size-selected molecular clusters.