FROM MOLECULES TO CRYSTALS: COHERENT ROTATIONAL, PHONON AND VIBRONIC DYNAMICS

Nikolaus Schwentner

Department of Experimental Physics, Free University Berlin, 14195 Berlin, Germany

The coupling of coherent quantum systems to the environment and the correlation of decoherence with the number of degrees of freedom is an important topic. We pick up and trace coherences in free molecules, matrix-isolated molecules and molecular crystals with delocalized excitations up to a mesoscopic scale.

We demonstrate, that collinear pump-probe type measurements of birefringence (Optical Kerr Effect) allow a very sensitive detection of induced anisotropies. An alignment $<\cos^2\theta>$ of the molecular axis to the pump-field's linear polarization of the order of 10^{-5} can still be resolved. We derived detailed information for the J-dependence of dephasing, depopulation and thermalization cross-sections for N_2-molecules at ambient conditions. Specific rotational wavepackets from selected J sub-ensembles can be prepared by tuning the pulse-length for a nonresonant pump between adiabatic and nonadiabatic with respect to the J-state transition energies and by adding appropriately timed second pulses (C5).

Scaling to a molecular quantum-crystal with cryogenic pH_2, kept at a temperature below the rotational energies, yields to coherent oscillations in the 94fs range, which last for about 1000 periods. They originate from delocalized rotons with subtle 0.5fs splitting into three |M| sub-states, which can be resolved and is also visible in a beat pattern of 17ps in time frame. These rotons interact with a single, delocalized lattice phonon with 900fs period, which follows the Raman-selection rule.

With phase-stabilized pulse trains from a pulse shaper unit we imprint frequency combs into isolated Br_2-molecules embedded in a classical Ar-matrix. In this rational control scheme the combs are derived from the spectroscopic information on vibronic zero-phonon line and phonon-sideband progressions. The combs become active in the molecules only if the pulse sequence is absorbed coherently. We demonstrate by the phonon-sideband response, that a coherent, coupled vibron-phonon wavepacket is prepared in a volume containing the chromophore and of the order of 1000 matrix-atoms. Coherent control of predissociation via selective excitation of relevant phonon-modes according to the theoretical prediction (C1) is pursued.