INFRARED-INFRARED DOUBLE RESONANCE SPECTROSCOPY OF HCCCN SOLVATED IN SUPERFLUID HELIUM DROPLETS

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We report high-resolution IR-IR double resonance spectra of the ν_1 fundamental (C-H stretch) of HCCCN solvated in superfluid helium droplets. Double resonance schemes of this type are highly effective in "tagging" molecules whose spectral broadening is due in part to static inhomogeneous broadening. The timescale of pump and probe is long compared to vibrational relaxation to the droplet, so that a hole can be burned in the helium droplet size distribution. Experiments of this type remove the contribution to the linewidth from the droplet size distribution and allow for a more in-depth analysis of a solvated molecules' local environment.