A NOVEL APPROACH IN 2DIR SPECTROSCOPY PERMITS MEASURING LONG-RANGE CORRELATIONS.

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The applicability of two-dimensional infrared (2DIR) spectroscopy methods for accessing molecular structures relies strongly on the range of distances accessible in the measurements. A novel technique, the relaxation-assisted 2DIR (RA 2DIR) method, is discussed that uses molecular scale vibrational energy transport for measuring cross peaks between modes separated by distances larger than 10 A. We show that the characteristic time for the vibrational energy transport between the modes, the arrival time, can be correlated with the intermode distance and reports on the bond connectivity pattern. An analytical power of the RA 2DIR method is demonstrated.

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