Spatial phase effects in Raman coherence

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Femtosecond Raman coherence is traditionally regarded as a time-resolved pump-probe technique [1]. However, coherence extends beyond the temporal domain, encompassing spatial degrees of freedom as well. In this contribution, we explore the intrinsic connection between spatial and temporal components of Raman coherence by employing an approach that maps temporal information onto a spatial image of the Raman signal across the transverse plane, orthogonal to the laser pulse propagation.

Our theoretical calculations for rotational and vibrational Raman coherence in gas molecules reveal how conventional time-resolved measurements might exhibit apparent temporal shifts and signal distortions. Furthermore, we demonstrate that spatial phase coherence complements temporal phase coherence in view of higher sensitivity to temperature variations. An example is illustrated in the figure where the simulated homodyne signal, captured by an imaging device at the fixed delay of 4.19 ps, reveals the details of the half revival of the N_2 rotational wave packet. This demonstrates the potential for single-shot thermometry in place of a delay scan needed for the reconstruction of the time-resolved signal.

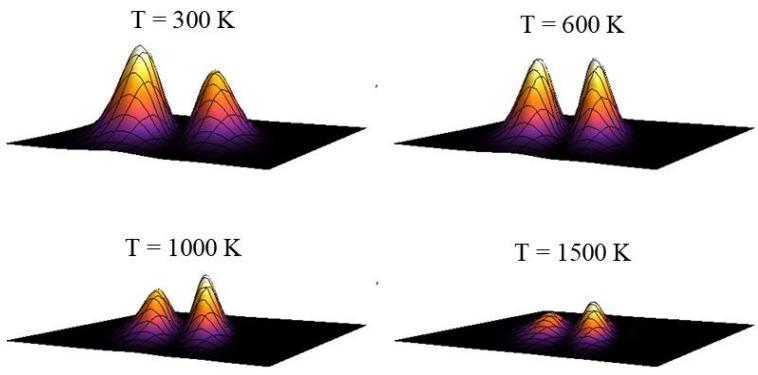

Figure 1. Simulated homodyne signal of the N_2 half revival of the rotational wave packet detected at an imaging device for the fixed delay of 4.19 ps.

1. S. Mukamel, *Principles of Nonlinear Optical Spectroscopy*, (Oxford University Press, New York, 1995)