Measurement-induced collective vibrational quantum coherence under spontaneous Raman scattering in a liquid

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Abstract

Spontaneous vibrational Raman scattering is a ubiquitous form of light–matter interaction whose description necessitates quantization of the electromagnetic field. It is classically considered as an incoherent process because the scattered field lacks any predictable phase relationship with the in-coming field [1]. When probing an ensemble of molecules, the question therefore arises: What quantum state should be used to describe the molecular ensemble following spontaneous Stokes scattering? We experimentally address this question by measuring time-resolved Stokes–anti-Stokes two-photon coincidences on a molecular liquid consisting of several sub-ensembles with slightly different vibrational frequencies (Figure 1) [2]. When spontaneously scattered Stokes photons and subsequent anti-Stokes photons are detected into a single spatiotemporal mode, the observed dynamics is inconsistent with a statistical mixture of individually excited molecules. Instead, we show that the data are reproduced if Stokes–anti-Stokes correlations are mediated by a collective vibrational quantum, i.e. a coherent superposition of all molecules interacting with light. Our results demonstrate that the degree of coherence in the vibrational state of the liquid is not an intrinsic property of the material system, but rather depends on the optical excitation and detection geometry. Our work nourishes the debate about the relation between optical coherence, quantum coherence and entanglement [3, 4].

References

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Figures

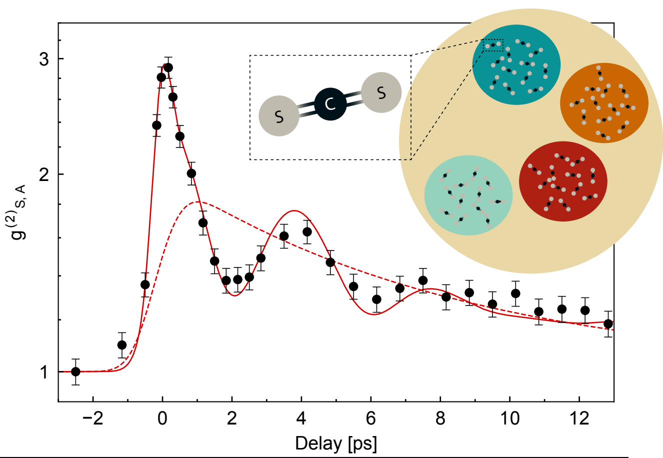

Figure 1: Time-resolved Stokes–anti-Stokes correlations. Full circles represent the measured data points while the solid red line indicates the model prediction. The observed quantum beats are consistent with a macroscopic quantum superposition of four sub-ensembles of CS2 molecules (inset) sharing a single quantum of vibration. The dotted red line represents the multi-exponential decay that would result from a statistical mixture of single vibrating molecules.