Bell correlations between light and vibrations at ambient conditions

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Abstract. Mechanical oscillators have been identified as new resources for quantum optics and its applications in metrology, sensing and information processing. Developing new techniques to prepare non-classical states of mechanical oscillators and engineer their quantum correlations with light fields also promises new insights into the dynamics and decoherence of vibrations in the quantum regime. This talk describes a new ultrafast pump-probe Raman spectroscopy technique that harness time-correlated single-photon counting to prepare and characterise non-classical states of Raman-active vibrations in crystals and molecules.

Keywords: Quantum Optics, Optomechanics, Raman Spectroscopy

Time-resolved Raman spectroscopy techniques offer various ways to study the dynamics of molecular vibrations in liquids or gases and optical phonons in crystals. While these techniques give access to the coherence time of the vibrational modes, they are not able to reveal the fragile quantum correlations that are spontaneously created between light and vibration during the Raman interaction.

Building on previous techniques [1,2], we present a new scheme leveraging universal properties of spontaneous Raman scattering to demonstrate for the first time Bell correlations between light and a collective molecular vibration (i.e. an optical phonon)[3]. We measure the decay of these hybrid photonphonon Bell correlations with sub-picosecond time-resolution and find that they survive over several hundred oscillations at ambient conditions. Our method offers a universal approach to generate entanglement between light and molecular vibrations, which could be extended to ultra-cold molecules. Moreover, our results pave the way for the study of quantum correlations in highly complex solidstate and molecular systems in their natural state, i.e. without any engineering of their coherence properties or their interaction with light.



Figure 1 Time-resolved photon-phonon Bell correlations. The CHSH parameter S is plotted as a function of delay between Stokes and anti-Stokes scattering, with error bars computed from a Monte Carlo simulation. The blue region, demarcated by $2 < |S| \le 2\sqrt{2}$, certifies Bell correlations.

References

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