Single phonon Fock state ring down spectroscopy in bulk diamond

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We present a new technique combining ultrafast Raman spectroscopy and time-correlated photon counting to measure the ring down time of a single phonon Fock state, as schematized in Fig. 1. Our work is inspired from a theoretical proposal in cavity quantum optomechanics [1], yet it has little in common with the original scheme: we use no cavity, work at room-temperature, and use sub-picosecond optical pulses to create and readout an optical phonon vibrating at ~40 THz in bulk diamond.

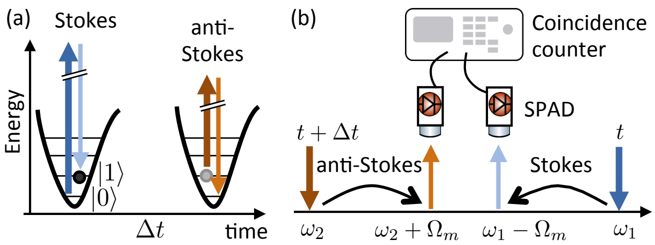

Figure 1: (a) A single phonon is optically created and annihilated, (b) mediating non-classical correlations measured by photon counting.

First, we send a femtosecond laser pulse on a Raman active material, where spontaneous Stokes scattering leads to the creation of an entangled photon-phonon state (a two-mode squeezed state). Performing single-photon detection (a projective measurement) on the Stokes mode permits the probabilistic preparation of the $|n = 1\rangle$ phonon Fock state. Then, after a controlled time delay, we send a second pulse to map the phonon quantum state onto a propagating anti-Stokes photon. The measured photonic Stokes–anti-Stokes correlations can be precisely linked to the prepared phonon quantum state and its dynamics. In particular, we show that the data strongly violate the Cauchy-Schwartz inequality (which bounds classical correlations), and that these non-classical correlations persist for several picoseconds, *evidencing the creation and decay of single phonons*, as shown in Fig. 2.

The experimental setup is based on a femtosecond Ti:Sapph oscillator pumping a tunable, intracavity-doubled optical parametric oscillator (OPO). This enables the generation of two synchronised pulse trains at 80 MHz, each independently tunable within 740 to 860 nm (Ti:Sapph) and 505 to 740 nm (OPO). The main challenge is to work over such a broad wavelength range while maintaining sub-ps time resolution and extremely efficient spectral filtering (pump suppression by at least 100 dB) to isolate the Stokes and anti-Stokes photons. We employ close to a dozen of tunable interference filters over the entire setup, before coupling the signal into optical fibers to an array of 4 single photon detectors.

Previous work on bulk diamond has demonstrated that single optical phonons could be used for THz quantum information processing [2, 3, 4]. However, the technique used in these papers strongly relied on the specific polarization selection rules of Raman scattering by the bulk dimaond optical phonons. On the contrary, our scheme uses spectral multiplexing to distinguish between the *write* and *readout* photons (which are emitted in a time window shorter than any existing detector's response time). It is therefore applicable to a much broader range of material systems. In addition, our setup is broadly tuable (505 nm to 860 nm), which will allow us to perform phonon-sideband excitation of systems with intrinsic (electronic) resonances or placed inside nano-cavities [5].

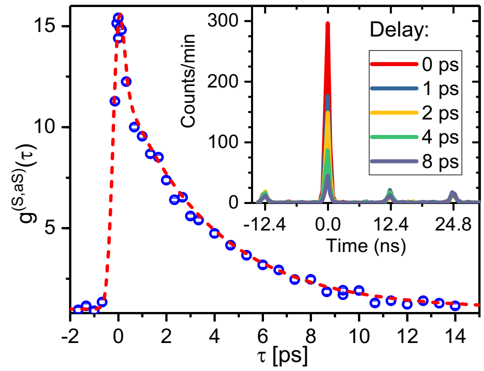

Figure 2: Decay of non-classical correlations mediated by a single phonon in bulk diamond. Inset: coincidence histograms at different delays between the two pulses. The ratio of the central peak to the side peaks measures the Stokes–anti-Stokes correlations, and values above 2 are non-classical.

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