Single-Phonon Addition and Subtraction to a Mechanical Thermal State

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Adding or subtracting a single quantum of excitation to a thermal state of a bosonic system has the counter-intuitive effect of approximately doubling its mean occupation. We perform the first experimental demonstration of this effect outside optics by implementing single-phonon addition and subtraction to a thermal state of a mechanical oscillator via Brillouin optomechanics in an optical whispering-gallery microresonator. Using a detection scheme that combines single-photon counting and optical heterodyne detection, we observe this doubling of the mechanical thermal fluctuations to a high precision. The capabilities of this joint click-dyne detection scheme adds a significant new dimension for optomechanical quantum science and applications.

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Introduction.—Performing single-quantum-level operations to bosonic quantum systems provides a rich avenue for quantum-state engineering, quantum-information and communication applications, as well as exploring the foundations of physics. Prominent examples of quantum-state engineering using such operations include the generation of nonclassical states of motion of trapped ions [1], microwave fields inside superconducting resonators [2,3], and high-frequency phonons coupled to superconducting qubits [4,5]. This type of control is also central to many quantum technologies such as quantum communications with quantum repeaters [6,7], on-demand single-photon preparation [8,9], and continuous-variable entanglement distillation [10]. Moreover, these operations allow for studies of nonclassicality [11,12] and exploring the interface between quantum information and quantum thermodynamics [13].

A practical and powerful way to achieve single-quantum addition or subtraction is to use an interaction with light followed by single-photon detection. This approach has been applied in optics to create "kitten" states by single-photon subtraction from squeezed vacuum [14,15] and to explore the properties of single-photon-added coherent states [11]. Single-quantum addition via single-photon detection has also been recently applied to atomic-spin ensembles to create spin-ensemble states [16] exhibiting significant nonclassicality [17]. These non-Gaussian operations can be used to create highly nonclassical states, and it has been theoretically shown that the addition operation creates nonclassicality for any initial mean thermal occupation [18–20].

Cavity quantum optomechanics now provides a means for performing single-phonon addition and subtraction to macroscopic mechanical resonators [21]. These operations were first demonstrated experimentally using optical phonon modes in bulk diamond and then using silicon photonic-crystal structures, where nonclassical light-matter correlations [22,23] and entangled states of two mechanical resonators [24,25] were generated. There is significant scope for further exploration of single-phonon addition and subtraction within optomechanics to, e.g., generate a wide range of macroscopic quantum states that are yet to be experimentally realized and for new studies of quantum thermodynamics.

Curiously, when single-quantum addition or subtraction is applied to a thermal state, the mean number of quanta actually increases in both cases. Indeed, for a thermal state of mean occupation $\bar{n}$, when applying an addition (subtraction) operation, the mean occupation undergoes the transformation $\bar{n} \rightarrow 2\bar{n} + 1$ ($\bar{n} \rightarrow 2\bar{n}$). Though this increase appears counterintuitive, an understanding of this effect can be obtained by considering the Bayesian inference with the information gained by the measurement that heralds this nonunitary operation. This behavior has been observed for thermal optical fields by performing heralded single-photon addition or subtraction followed by homodyne detection [26,27], and the approximate doubling of the mean occupation was utilized for work extraction in Ref. [13]. Though these operations are now well studied for optical fields, they remain far less explored for other bosonic systems. In particular, the approximate doubling of the mean occupation by these operations to a thermal state

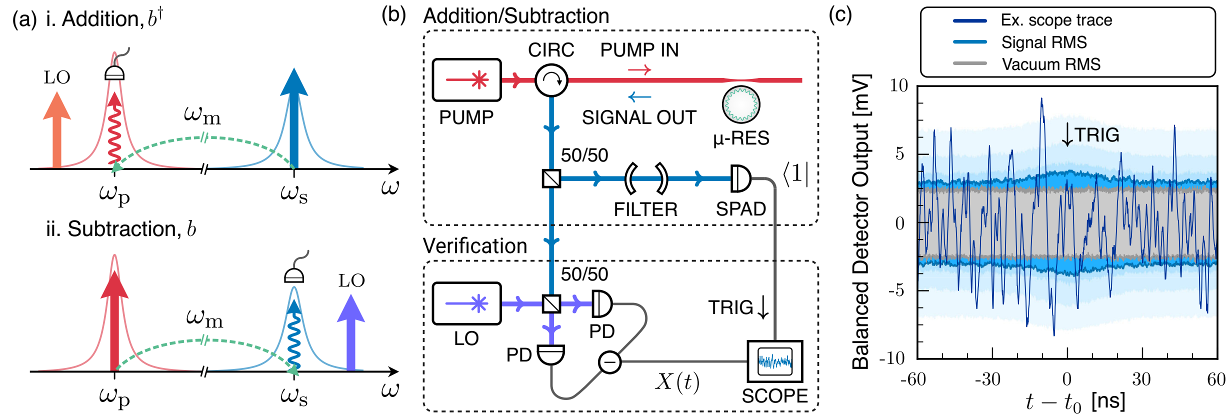

FIG. 1. Scheme and setup. (a) Optical pumping and detection configuration comprising a pair of optical resonances spaced by approximately the mechanical frequency. For single-phonon addition, the higher-frequency mode of the pair is pumped and the down-shifted signal field is filtered and then detected by a single-photon counter to herald the operation. The frequencies of pumping and detection are reversed for the subtraction operation. (b) Schematic of the experimental setup that uses a pump laser and single-photon detection for the single-phonon addition or subtraction operations, and a separate laser for verification via heterodyne detection. (LO: local oscillator laser, PD: photodiode, SPAD: single photon avalanche photodiode.) (c) An example recorded heterodyne time trace about a subtraction event (dark blue curve). In the background, the optical vacuum, and signal rms levels are plotted (grey, and blue, respectively), and a color gradient is used to show the statistics of the signal feature above the optical vacuum noise.

is yet to be demonstrated for any system other than traveling light fields.

In this Letter, we report the observation of doubling of the mean occupation of a mechanical oscillator via heralded single-phonon addition and subtraction in a Brillouin-optomechanical system. We measure the temporal dynamics of the resulting increase in the mechanical excitation via a heterodyne detection scheme and observe the aforementioned doubling with a high signal-to-noise ratio. This work combines both photon counting and optical-dyne detection in a single experiment, thus, taking a step toward the realization of hybrid quantum protocols that exploit both discrete and continuous variables [28]. Moreover, the strong optomechanical coupling achievable in this geometry [29] and the excellent mechanical coherence times achievable with crystalline materials [30,31] provides a promising experimental path for the development of quantum memories and repeaters.

Brillouin single-phonon addition and subtraction scheme.—Brillouin scattering is a nonlinear-optical process involving the interaction between two optical fields and one high-frequency acoustic wave. In this experiment, an optical pump field interacts with thermally excited phonons to create frequency down-shifted (Stokes) or up-shifted (anti-Stokes) fields of light. For the anti-Stokes case, the pump interacts with counter-propagating phonons, and for the Stokes case, the pump interacts with copropagating phonons. In both cases, these new optical fields are back-scattered with respect to the pump field. In bulk materials, both the Stokes and anti-Stokes processes occur simultaneously and with similar strengths. To break this symmetry and select only one of these processes, we utilize two optical modes of a cavity approximately spaced by the mechanical frequency, see Fig. 1(a). By pumping the higher-frequency mode of the pair, anti-Stokes scattering can be strongly suppressed, allowing the Stokes scattering to be selectively driven. Similarly, by pumping the lower-frequency mode of the pair, the anti-Stokes process can be selectively driven. In this pumped regime, the interaction can be linearized and accurately described by quadratic interaction Hamiltonians. For the Stokes-scattering case, the interaction is modeled by a photon-phonon two-mode-squeezing Hamiltonian $H/\hbar = G(ab + a^\dagger b^\dagger)$, while for the anti-Stokes case, the interaction is modeled by a photon-phonon beam-splitter Hamiltonian $H/\hbar = G(a^\dagger b + ab^\dagger)$. Here, a is the annihilation operator for the optical cavity mode supporting the scattered field, b is the mechanical annihilation operator, and G is the interaction rate for these two separate cases, which is proportional to the intrinsic coupling rate and the intracavity pump amplitude.

In cavity optomechanics, performing single-phonon addition or subtraction by driving the blue or red sideband followed by single-photon counting of the scattered signal was first considered theoretically in Ref. [21], and we have adapted that approach for the Brillouin-scattering-based experiment we perform here. To implement a single-phonon addition operation, the higher-frequency optical cavity mode of the pair is weakly pumped by a coherent state [cf. Fig. 1(a)] to drive the two-mode-squeezing interaction, and the resulting counter-propagating Stokes signal is separated from the pump and detected by a single-photon counter to herald the addition operation. For this operation, the angular frequency of the signal field is $\omega_s = \omega_p - \omega_m$, where ω_p refers to the pump laser angular frequency, and ω_m is the mechanical angular frequency.

Implementing a single-phonon subtraction operation is performed in a similar manner where the lower-frequency mode of the pair is pumped to bring the beam-splitter interaction into resonance prior to single-photon detection. The signal field for this operation has the angular frequency $\omega_s = \omega_p + \omega_m$. In this weak-pump regime, the heating or cooling of the mechanical mode via the optomechanical interaction with the pump field is negligible. Moreover, the probability of two or more photons being detected is also negligible, ensuring that the measurement heralds a single-phonon operation. Under these conditions, we model these nonunitary operations to the input mechanical state with the mechanical creation and annihilation operators $b^\dagger$ and b, respectively, and, in this weak-drive regime, these operations have a heralding probability proportional to the mechanical occupation $\bar{n}$. Brillouin optomechanics is very well suited to implementing single-phonon addition and subtraction owing to the high mechanical frequencies available in the back-scattering configuration and the ability to be readily implemented in ultralow optical loss materials.

Also, note that as we are pumping only in the forward direction and observing scattered light in the backward direction, due to the Brillouin phase-matching conditions, we are then applying the addition operation to a forward acoustic wave and the subtraction operation to a backward wave. Should it be desired to perform addition or subtraction to the same mechanical mode, the microresonator can be pumped in the reverse direction, as the system is symmetric under inversion of the propagation directions of all three waves.

Following an addition or subtraction operation, we use a heterodyne detection scheme to measure the variance of the mechanical amplitude fluctuations to verify the effect of the operation on the mechanical state. For experimental simplicity, we use the same continuous weak drive for the addition and subtraction operations and for the verification. For weak optical drive and $\bar{n} > 1$, the optical amplitude on top of the vacuum noise on the Stokes or anti-Stokes scattered light serves as a proxy for the mechanical amplitude, thus, enabling the dynamics of the mechanical fluctuations about the herald event to be characterized.

Experimental setup.—To experimentally implement this scheme, a BaF_2 optical microresonator, fabricated using a diamond nanolathe, is used. The device has a micro-rod-resonator geometry [32] with a diameter of approximately 1.5 mm, and a lateral confinement region with a radius of curvature of approximately 40 μm. An optical microscope image of the microresonator is provided in the Supplemental Material [33].

We employ an all fiber-based optical setup and use a continuous-wave pump laser running at 1550 nm. A silica tapered optical fiber is used to evanescently couple to the microresonator, and despite the small difference in refractive index between the fiber and the resonator, up to and beyond critical coupling to the optical cavity modes can be achieved. By recording the optical transmission spectra with a weak field, we observe optical whispering-gallery-mode resonances with an intrinsic quality factor of $Q \simeq 10^8$ and identify a pair of optical resonances that have a frequency separation approximately equal to the Brillouin shift of $\omega_m/2\pi = 8.21$ GHz. These optical modes are not spaced by the free-spectral range of the microresonator but have different transverse spatial profiles while still providing significant Brillouin optomechanical coupling. For the taper-coupling position used, the optical amplitude damping rates of the lower- and higher-frequency cavity modes are 6.8 and 7.8 MHz, respectively. The mechanical mode is a pseudolongitudinal elastic whispering-gallery wave [34] that fulfills the Brillouin phase matching conditions [33]. We perform the experiment at 300 K (stabilized), which corresponds to a mechanical mean occupation of $\bar{n} \simeq 760$. Based on the experimental observations described below, we estimate the mechanical amplitude decay to be $\gamma/2\pi = (17.0 \pm 3.2)$ MHz, which is expected to be largely from intrinsic material damping owing to the room-temperature operation.

We lock to either the higher- or lower-frequency optical mode by Pound-Drever-Hall laser-frequency stabilization. The laser input-pump power is on the order of 1 mW, leading to intracavity powers of 0.7 W, and optomechanical coupling strengths $G/2\pi$ of typically 2 MHz. We intentionally perform this particular experiment in the weak optomechanical coupling regime, where the coupling rate G is much smaller than the optical amplitude decay rate κ and the mechanical amplitude decay rate γ. For these parameters, the steady-state optomechanical cooling or heating to the mechanical oscillator is less than 1% [33]. Brillouin Stokes and anti-Stokes light backscattered from the cavity is coupled out through the taper, is separated from pump light via an optical circulator, and is then detected via a single-photon avalanche photodiode (SPAD) to herald the single-phonon addition and subtraction operations, see Fig. 1(b). Prior to single-photon detection, spurious residual pump photons are filtered using two fiber-based Fabry-Perot filters that have an intensity FWHM linewidth of 120 MHz and a free-spectral range of 25 GHz. The SPAD was operated with a quantum efficiency of ~12.5%, a gate rate of 50 kHz, a detector recovery time of 20 μs between gates to avoid afterpulsing, and an 8 ns effective gate duration. Note that the optical losses in our setup are large, however, this only affects the heralding probability and not the fidelity of the operation performed. This resilience to optical loss is because the dark-count rate 3 s^{-1} is much lower than the signal rate 500 s^{-1} and the probability of multiphoton detection is negligible, thus, a detector click corresponds to high-fidelity addition or subtraction of a single phonon.

A detection event by the SPAD at time t_0 heralds the single-phonon operation and the SPAD output signal triggers a digital storage oscilloscope to record a heterodyne time trace. An example heterodyne trace and the rms noise is plotted in Fig. 1(c). We repeat this measurement approximately 20 000 times for both single-phonon addition and subtraction, which enables us to easily observe the increase in the mean occupation and the nonequilibrium dynamics around the herald event. We implement both single photon detection and heterodyne detection by splitting the signal field with a 50:50 beamsplitter, which simplifies the setup at the cost of a reduced heralding rate. To implement the heterodyne detection, a balanced detector and a local oscillator shifted by approximately 150 MHz with respect to the scattered light frequency is used.

Results and discussion.—In Fig. 2, the ensemble average of the square of the heterodyne signal X is plotted with time about the herald event for both the addition and subtraction operations. This heterodyne signal is normalized such that $\langle X^2 \rangle = 1/2$ when just optical vacuum is measured, which is shown as the grey lines in the two plots, and was experimentally verified to be 20 dB above the electronic noise of the detector. For both interactions used here, we experimentally verified that the scattered optical signal fields are rotationally invariant in phase space and, hence, did not phase-lock the local oscillator for the heterodyne

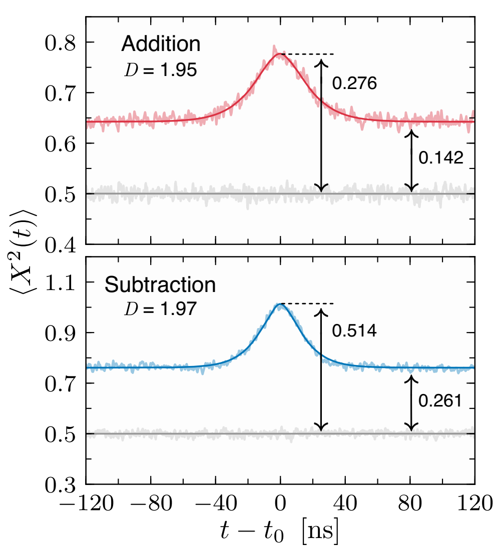

FIG. 2. Variance of the heterodyne signal as a function of time about the heralding event for the addition operation (red trace, above) and the subtraction operation (blue trace, below). The variance of the optical vacuum, measured separately, is normalized to $1/2$ and is shown in grey. The variance is determined from 20 000 runs of the experiment for each plot. About the heralding event $(t - t_0 = 0)$, a clear feature is observed indicating that the mechanical fluctuations have increased. Excellent agreement between the experimental data (pale lines) and theoretical fits (dark lines) is obtained [33]. The ratio of the variance above the vacuum noise at the center of the feature to far from the feature we label D, which is very close to two in both of these cases, indicating that the mechanical mean occupation has doubled to a high precision.

verification measurements. For the single-phonon addition operation, we observe a variance 0.142 above the vacuum level at a time far from the herald event, owing to the mechanical thermal fluctuations being mapped onto the optical field. Then, at the time of the herald event, we observe a feature in time that peaks at 0.276 above the vacuum level. These levels above the vacuum are proportional to the mechanical mean occupation, and taking the ratio of the two indicates that the mean phonon number has increased by a factor of $D = 1.95 \pm 0.02$. For the single-phonon subtraction case, we observed 0.261 above the vacuum away from the herald event and 0.514 above the vacuum level at the herald event, indicating that the mean occupation has increased by a factor of $D = 1.97 \pm 0.02$. These factors are very close to the theoretical prediction of a doubling of the mechanical contribution to the quadrature variance [33]. Note that the variances observed are different for the single-phonon addition and subtraction cases as the pumping is swapped between the two optical resonances, which possess different linewidths, and the input pump power was also adjusted. Also, note that the peak of the feature is reduced by approximately 1% due to a small amount of dark counts in the SPAD.

The temporal dynamics of the "doubling feature" are governed by the interplay between the optical and mechanical damping rates, κ and γ, respectively. To quantitatively describe these dynamics we developed a model using Langevin equations for Brillouin optomechanical interactions, cavity input-output, and quantum measurement theory [33]. We find excellent agreement between the experimental data and the theoretical prediction with the only free fitting parameters in this model being the factor D and the mechanical damping rate, with the latter being consistent with fits of the anti-Stokes heterodyne spectra. The estimates of these fitting parameters can be improved by taking into account the response of the optical filter prior to the SPAD and the spectral response of the balanced detector [33].

A further contribution made by this work is that the measurement configuration introduced here can also be used to examine the degree to which multiple mechanical modes couple. This is possible as the presence of multiple mechanical sources will give rise to temporal interference fringes in the heterodyne signal around the herald event. A mathematical model for the time evolution of the quadrature variance in the presence of multiple mechanical modes is given in the Supplemental Material [33]. In this experimental work, as no temporal interference fringes are observed, we infer that our coupling is predominantly to a single mechanical mode.

Outlook.—Brillouin optomechanics with high-frequency phonons [29,35] is emerging as a powerful new platform for quantum and classical optomechanics applications. Owing to the favorable properties of crystalline materials, such systems now provide excellent mechanical Qf

products [30,31], where $Qf > 10^{17}$ Hz, hence, enabling ultralow mechanical decoherence even for modest cryogenic temperatures (~4 K). The BaF_2 crystalline resonators used here provide a promising path for accessing this regime for the whispering-gallery-mode geometry. Moreover, BaF_2 is an ultra-low-loss optical material well suited to these studies [36], which enables optically induced heating to be minimized and Brillouin optomechanical strong coupling to be readily achieved [29]. These advantages, combined with the techniques demonstrated here, open a rich avenue for further studies including quantum memories and repeaters [6,7], which can even be combined with the backscatter operation of Brillouin scattering for (quantum) routing and nonreciprocity applications.

To the best of our knowledge, this is the first optomechanics experiment that combines photon counting and optical dyne detection, which lays further groundwork for mechanical quantum-state engineering applications, such as mechanical superposition-state preparation via mechanical squeezing and then single-phonon addition or subtraction [28]. Following a state-preparation protocol, one can then switch to a stronger anti-Stokes interaction and utilize the strong coupling achievable with these systems [29] for mechanical state transfer to light for mechanical quantum-state reconstruction [37]. Further exciting lines of study opened by this work, applicable to Brillouin-based and other optomechanical systems, include continuous-variable quantum-state orthogonalization [21], precision thermometry in Brillouin optomechanical systems when at low temperature, and utilizing the dramatic change in the mean occupation for measurement-based quantum thermodynamics applications with phonons.

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