Unconditional Accumulation of Nonclassicality in a Single-Atom Mechanical Oscillator

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The robust experimental accumulation of nonclassicality of motion of a single trapped ion is reported. The nonclassicality stems from deterministic incoherent modulation of thermal phonon number distribution implemented by a laser excitation of nonlinear coupling between the ion’s internal—electronic levels and external—motional states. It is demonstrated that the repetitive application of this nonlinear process monotonically accumulates the observable state nonclassicality. The output states converge to a phonon number distribution with high overlap with a particular Fock state and visible quantum non-Gaussian aspects including corresponding negative Wigner function. The resulting oscillator states prove deterministic transition in the hierarchy of quantum non-Gaussianity up to four phonons. This transition is very robust against experimental imperfections and produces increasing entanglement potential.

1. Introduction

Quantum nonclassical states represent a directly observable product of nonlinear quantum processes and are a paramount resource for studies of light and matter,1–6 processing of quantum information,7–9 and have proven to be beneficial for a broad range of metrological and sensing applications.10–14 One of the obstacles for their full utilization in mechanical oscillators is oftentimes severely limited effective creation probability for optical or microwavemeasurements and still large energy of thermal environment for solid-state experiments at cryogenic temperatures. In a vast majority of experimental demonstrations, generation of nonclassical phonon-number states utilizes an initial ground state of the system with very low entropy combined with strong nonlinear interaction, high quality projection measurement, or both.15–23 In many mechanical systems the available level of control is mostly insufficient for the initial step of entropy minimization and, at the same time, nonlinear couplings are weak to be employed on the relevant timescales and efficient noiseless projective measurement is not available.

To overcome these natural limitations, we report on the experimental demonstration of unconditional accumulation of nonclassicality for a single-atom mechanical oscillator prepared initially in thermal state with energy of several motional quanta by employing the scheme presented in the seminal paper by Blatt et al.24 We use the basic Rabi interaction corresponding to a coupling between the mechanical oscillator states of a single atom and its electronic spin.19,37–40 Such nonlinear interaction is thus applicable to a broad range of experimental systems which allow for a direct implementation of a blue detuned Rabi interaction, including trapped atoms, superconducting qubits coupled to a microwave radiation, or increasing variety of solid-state systems and optomechanical platforms.29–36 For oscillator’s thermal state and ground state electronic spin the Rabi coupling already realizes a complex deterministic modulation of a phonon number distribution. This modulation brings nonclassicality in phonon-number distribution of the oscillator as a counterpart of a fundamental collapse and revival effect in the electronic spin.19,37–40 We experimentally test an accumulation of the nonclassicality through repetitive application of the blue-detuned Rabi interaction with reinitialized electronic state which results in a monotonous accumulation of the generated nonclassicality manifested in several witnesses of nonclassicality. For any temperature of the oscillator, accumulated nonclassicality becomes apparent in the phonon number distribution converging to a dominant Fock states with corresponding negative Wigner function and level of quantum non-Gaussian hierarchy.41 The quantum non-Gaussian properties provably enhance for each of the consecutively repeated interactions, while each of these interactions is already theoretically provably sufficient for nonclassicality generation. While the presented nonclassicality accumulation shares some phenomenological similarities with the conventional nonclassicality distillation,42,43 it crucially differs in the fact, that it is unconditional. Moreover, we do not dynamically engineer neither state of the atoms nor coupling strength to reach accumulation, as in the reservoir engineering44,45 or in methods exploiting adiabatic passage.46,47 The accumulation of nonclassical properties can be viewed as quantum non-Gaussian mechanical counterpart.
of nonlinear optical parametric processes in a cavity where the initial weak nonclassical effect generated by a single implementation of the nonlinear interaction is gradually accumulated. 

2. Nonlinear Interaction with Atomic Mechanical Oscillator

The presented experimental demonstration of deterministic nonclassicality accumulation utilizes a high degree of control of k-times repeated coupling between the mechanical motion of a single trapped ion and its internal electronic state. The experimental setup comprises of a linear Paul trap for spatial localization of single $^{40}\text{Ca}^+$ ion. A simplified excitation geometry is shown in Figure 1a. To realize the deterministic nonclassicality accumulation, we employ ion’s axial motion with secular frequency set to $\omega_{ax} = 2\pi \times 1.2$ MHz. Excitation laser beams at 397, 866, and 729 nm are propagating under angle 45° with respect to trap axial direction. The 397 nm optical pumping and 854 nm repumping beams are propagating along the direction of the applied magnetic field $B$ with circular and elliptical polarizations, respectively. The experimental sequence begins with a 1 ms period of Doppler cooling using the 397 nm laser and the 866 nm beam is used for reshuffling the atomic population from initially classical thermal population of atomic motion. The nonlinear AJC coupling is followed by a reinitialization of internal atomic population to the $|S\rangle$ level. This process is repeated k-times.

is implemented using the 729 nm laser tuned to the first red motional sideband of the $4S_{1/2}(m = -1/2) \leftrightarrow 3D_{5/2}(m = -5/2)$ transition together with a weak 854 nm beam which reshuffles the $3D_{5/2}(m = -5/2)$ population to the ground state $4S_{1/2}(m = -1/2)$.

For any step $k$ of the accumulation, the interaction between ion’s axial motional mode with an input state $\rho_k = \sum_{n=0}^{\infty} P_k(n) |n\rangle \langle n|$ and internal quasi-two level system corresponding to the transition $|S\rangle = 4S_{1/2}(m = -1/2) \leftrightarrow |D\rangle = 3D_{5/2}(m = -1/2)$ is realized by the excitation of the first blue motional sideband using the 729 nm laser. An observable large number of Rabi oscillations with close to full contrast with a Rabi frequency on a blue motional sideband corresponding to $g = 2\pi \times 5.8$ kHz in our setup allows for implementation of coherent blue-detuned Rabi (anti-Jaynes–Cummings) interaction between spin and motion with high fidelity, see Supporting Information S2. However we note that successful implementation of the nonclassicality accumulation only requires small coherence of the anti-Jaynes–Cummings (AJC) interaction and is thus applicable to systems with much smaller coherence of nonlinear coupling and with higher presence of noise. The accessible high control of trapped ion states should be viewed as a feasible tool for the realization of proof of principle characterization of the scheme rather than demanding condition.

In the Lamb–Dicke regime, the AJC interaction corresponding to the excitation of a blue motional sideband of trapped ions can be well approximated by effective Hamiltonian:

$$\hat{H}_{\text{blue}} = g/2 (\hat{a}^\dagger \hat{\sigma}_+ + \hat{a} \hat{\sigma}_-)$$

(1)
where $g$ is the coupling strength, $\hat{a}$ is a bosonic operator acting on the axial harmonic motional mode, and $\hat{\sigma}_z, \hat{\sigma}_-$ are two-level raising and lowering operators of electronic spin, respectively. For oscillator with input phonon number distribution $P_k(n)$ in $k$th step of the procedure and the electronic spin prepared in the ground state $|S\rangle$, the AJC interaction results in the state with modulated phonon populations,

$$P_{k+1} = \sum_{n=0}^{\infty} P_k(n) \left( \cos \left( \frac{gt}{2 \sqrt{n+1}} \right) \right)^2 |n\rangle \langle n| + \sum_{n=0}^{\infty} P_k(n) \left( \sin \left( \frac{gt}{2 \sqrt{n+1}} \right) \right)^2 |n+1\rangle \langle n+1|$$

where $t$ stands for the laser excitation time and $gt$ therefore stands for the effective area of the driving pulse. The output state $P_{k+1} = \sum_{n=0}^{\infty} P_{k+1}(n)|n\rangle \langle n|$ is fully defined by its phonon population $P_{k+1}(n)$ and is separated from the state of the electronic spin. It can be therefore directly used in the next iteration step, which is again represented by (2). Note that the accumulation process is the same in all $k$ steps; the pulse area $gt$ is not changed.

Already for an initial thermal state with $P_0(n) = \sum_n \frac{n^k}{(\sqrt{\pi} n_0)^{k+1}}^\infty$, the incoherent modulation (2) can deterministically result in a nonclassical state for a broad range of electronic and motional thermal states and excitation pulse lengths. The nature of the operation depends on the pulse area. For our proof-of-principle demonstration of the concept we choose $gt = \hbar \Omega_0 t \approx \pi$ and the initial internal state corresponding to $|S\rangle$, because it corresponds to an addition of one phonon to a ground state of the oscillator. Here $\eta_{229}$ is 0.063 is the Lamb–Dicke parameter for the interaction with the 729 nm beam, $\Omega_0 = 2 \pi \times (92 \pm 1)$ kHz is the Rabi frequency on the corresponding carrier transition and the length of the laser pulse is set to 91 $\mu$s. Beyond initial motional ground state, single step $k = 1$ deterministically shifts the phonon number distribution, as depicted in Figure 1c.

We test the nonclassical properties of the shifted phonon-number distributions $P_k(n)$ resulting from the first round of iteration (2) for input thermal phonon populations $P_0(n)$ with various energies. This is followed by reshuffling of the excited $|D\rangle$ level population back to the initial ground state $|S\rangle$ using the excitation by 854 nm laser to the $4P_{1/2}$ manifold followed by the emission of a single 393 nm photon. In addition, a short optical pumping 397 nm pulse ensures that atomic population is pumped to the initial Zeeman sublevel $|S\rangle \approx 4S_{1/2}, m = -1/2$. The reshuffling effectively corresponds to resetting the internal electronic state of the ion and makes the operation unconditional. However, at the same time, the random recoils from the resonant 854 nm laser excitation and 393 nm emission result in a small heating of mechanical populations with the total weight given by the probability of finding the ion in the $|D\rangle$-level after the interaction. In the Lamb–Dicke regime, such redistribution effectively corresponds to the interaction of mechanical mode with a thermal reservoir and it happens only with the probability of finding the ion in the excited state $|D\rangle$ after the process (2). A detailed model used for employed process simulations can be found in Supporting Information S1.

Let us first analyze this single step of the procedure. We have applied nonlinear AJC (2) with $gt = \pi$ to the oscillators initially in thermal distribution $P_0(n)$ with different mean energies. The phonon number distributions $P_k(n)$ were then obtained from fits of measured Rabi oscillations on the blue motional sideband. The initial states after sideband cooling correspond very well to the states with close to ideal Bose–Einstein statistics. The initial states after sideband cooling correspond very well to the states with close to ideal Bose–Einstein statistics. The initial states after sideband cooling correspond very well to the states with close to ideal Bose–Einstein statistics.

![Figure 2](image-url) Figure 2. The results of evaluation of nonclassicality for the measured phonon number distributions $P_k(n)$ after single nonlinear AJC interaction of atomic mechanical oscillator prepared in thermal state. The Fano factors evaluated for initial and generated phonon populations demonstrate the dependence of non-Gaussian states and excitation pulse lengths. [24,49,50] The nature of the operation depends on the pulse area. For our proof-of-principle demonstration of the concept we choose $gt = \hbar \Omega_0 t \approx \pi$ and the initial internal state corresponding to $|S\rangle$, because it corresponds to an addition of one phonon to a ground state of the oscillator. Here $\eta_{229}$ is 0.063 is the Lamb–Dicke parameter for the interaction with the 729 nm beam, $\Omega_0 = 2 \pi \times (92 \pm 1)$ kHz is the Rabi frequency on the corresponding carrier transition and the length of the laser pulse is set to 91 $\mu$s. Beyond initial motional ground state, single step $k = 1$ deterministically shifts the phonon number distribution, as depicted in Figure 1c.

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3. Unconditional Accumulation of Nonclassicality

Even a single operation (2) can significantly transform the state of a mechanical oscillator, a repetition of the same interaction may significantly increase the observable nonclassicality and quantum non-Gaussianity.[23,24] During the repetitions the operation remains constant; there is no optimization qubit state, interaction strength $g$, time duration $t$, or any other interaction characteristics. The process bears similarities to evolution of light inside a single resonant cavity with a nonlinear medium where nonclassicality is enhanced by a cyclic nonlinear interaction of the resonant optical mode with an off-resonant pump.[48] However, the accumulation has to not only overcome initial thermal occupation to the state $|S\rangle$. The key aspect of the considered AJC (2) that is responsible for its properties is the nonclassical nonlinear modulation of all terms $P_{\ell}(n)$ of the oscillator phonon population. This effect is independent of the initial population of the motional state and is enhanced by repeated application. It can no longer be understood just as repeated addition of single phonon, because the harmonic modulation terms of (2) are merged and this leads to constructive enhancement of nonclassical aspects of output phonon populations $P_{\ell}(n)$ which can be observed by several important metrics of nonclassicality.

Figure 3 shows the results of reconstruction of output phonon number distributions $P_{\ell}(n)$ for up to $k = 20$ repetitions of the AJC process for an initial thermal state with a mean phonon number $\bar{n} = 1.19 \pm 0.04$. They have been reconstructed from the measured Rabi oscillations after each displayed number of interaction repetitions $k$ for up to $n = 7$. This dimension has been chosen so that the displayed $P_{\ell}(n)$ of the initial thermal state includes 99 % of its population. At the same time, this basis suffices for the observation of phenomena underlaying the accumulation process and allows for relatively small errors on $P_{\ell}(n)$ when employing solely measurements on first blue motional sideband, see Supporting Information S2 for more details. The initial thermal energy has been chosen so that it allows for clear illustration of important manifestations of accumulation dynamics for interaction (2), which include depopulation of motional ground state, population accumulation in phonon basis $|n\rangle$ corresponding to close to an integer multiples of pulse area of $gt\sqrt{n + 1} \approx 2\pi$,[24] and effect of heating corresponding to random photon recoils accompanying the reset of internal state. The initial thermal population $P_{0}(n)$ is again transformed into a nonclassical phonon number distribution $P_{1}(n)$ after first step mainly due to the population shift from $P_{0}(0) \leftrightarrow P_{0}(1)$. However, its further repetitions accumulate other nonclassical aspects of modulation (2).
The first qualitative feature can be seen from the evolution of truncated phonon number distributions $P_l(n)$, which are depicted in Figure 3a–c for three different pulse areas $gt = 0.9\pi; \pi$ and $1.1\pi$. We test these slightly different gt to demonstrate the feasibility of control of the accumulation process. We can see that in all scenarios the repetition of the procedure transforms the initial thermal state into a state closely resembling Fock state. The particular created Fock state depends on the pulse area and it is, respectively, for the three pumping areas, [4], [3], and [2]. This transition can be also seen from the monotonous increase of the states’ average phonon number plotted in Figure 3d).

Insight into these features of nonclassicality accumulation can be gained by considering its asymptotic properties, similarly to refs. [49,50]. When the pulse area satisfies condition

$$gt\sqrt{n} + 1 = 2l\pi$$ (3)

for any natural numbers $n$ and fixed $l$, sufficiently high number of perfect operations (2) transforms any state with initial phonon number distribution $P_0(n)$ into an asymptotic mixture of Fock states for large $k$:

$$P_\infty = \sum_{j=1}^{\infty} P_\infty(n_j)\langle n_j|n_j\rangle$$ (4)

where $n_i$ are the phonon numbers satisfying the condition (3) for given $l$. The probabilities of the mixture can be obtained from the initial phonon number distribution as $P_\infty(n_j) = \sum_{m=n_j-l+1}^{\infty} P_0(m)$ where, for the sake of notation, we assume $n_0 = -1$. For example, repeating operations with pulse area $gt = \pi$ presented in Figure 3b produces mixture with Fock states $n_j = 3, 15, 35, …$ operations with pulse area $gt = 2\pi/\sqrt{3}$ produce mixture with $n_j = 2, 11, 47, …$ and operations with pulse area $gt = 2\pi/\sqrt{5}$ mixture with $n_j = 4, 19, 79, …$. This behavior also well explains the phonon number distributions in Figure 3a,c, because the chosen pulse areas $gt = 0.9\pi \approx 2\pi/\sqrt{5}$ and $gt = 1.1\pi \approx 2\pi/\sqrt{3}$ are close enough to the theoretical values. The fit for $gt = 1.1\pi$ is slightly worse which manifests as visible degradation of the Fock state for higher number of repetitions. This also demonstrates the need for high precision in practical setting of the pulse area $gt$.

The fundamental limitation on the achievable population of the Fock state $P_k(n)$ in the accumulation procedure is given by the sum of populations $\sum_{n=0}^{n_k} P_0(n)$ of the initial thermal state. However, the practical limit for Fock states with high $n_k$ will be mostly set by the requirement of high number of iterations $k$, the ability to control the applied pulse area $gt$ with a very high precision, and by the effective thermalization probability in the repumping process. In the example presented in Figure 3b corresponding to $gt = \pi$, the population $P(3)$ reaches $P^{rep}_\infty(3) = 0.52 \pm 0.01$ after 20 accumulation steps. The discrepancy with the theoretical prediction $P^{th}_\infty(3) = 0.63$ evaluated from the model presented in Supporting Information S1 can be attributed to a residual offset in an experimental setting of the pulse area $gt$. Its estimation from the fit of the measured photon number distribution $P_\infty(n)$ results in $gt = 1.026\pi$ corresponding to $P^{th}_\infty(3) = 0.54$, which is in a very good agreement with the measured value. While the fundamental limit on the achievable population $P(3)$ corresponding to the initial mean thermal phonon number $n_\text{th} = 1.19 \pm 0.04$ is $P_\infty(n_\text{th} = 3) \approx 0.91$, the measured value is further limited by the finite contrast of the applied $\pi$-pulse $\kappa = 97 \%$ and by the effective thermalization probability given by $\eta_{eff} = 0.17 \pm 0.04$. The thermalization cannot be fully avoided in most experimental scenarios; however, the close-to-ideal contrast $\kappa \approx 100 \%$ is feasible and would result in $P^{th}_\infty(3) = 0.86$ after 40 repetition steps. A direct reduction of the thermalization rate could be achieved by utilizing of higher trapping frequency with prospects of asymptotically reaching $P^{th}_\infty(3) = 0.84$ for the presented experimental parameters and for $\omega ax \approx 2\pi \times 5$ MHz. Theoretical estimation with an ideal $\pi$-pulse contrast and no thermalization would result in $P_\infty(3) = 0.88$, and reach $P_{11}(3) = 0.91$ after reasonable 43 repetition steps.

Initial thermal state with higher energy leads to lower purity of the produced state. It can, however, lead to states with greater weight of higher Fock states. We can also see that for any energy of the input thermal state we can, in principle, design a pulse that eventually produces a Fock state with purity that is arbitrarily close to one. Such pulse would have low area $gt$ that would need to be set with very high precision. In theory, a laser pulse with an area $gt = 2\pi/\sqrt{21}$ would asymptotically produce Fock state |20⟩ with element $P_{20}(20) > 0.9$ for any input thermal state with $\langle n \rangle < 9$.

The highly quantum non-Gaussian aspects of the states resulting from the accumulation process can be further evidenced in the reconstructed Wigner function $W(x,0)$, which has been evaluated from the measured $P_l(n)$ as an incoherent sum of Wigner quasi-distributions functions for $|n\rangle$ corresponding to state $P_l = \sum_{n=0}^{\infty} P_l(n)|n\rangle\langle n|$. The resulting Wigner functions effectively illustrate the state with population $P_l(n)$ and with randomized phase, which can be always implemented by random phase shift of the local oscillator in the reconstruction process. The corresponding data shown in Figure 3e point to several crucial aspects of the phonon distributions resulting from the accumulation process. The $P_l(n)$ converges to distribution with two unambiguously negative concentric annuli in phase space which are directly observable, that is, without any correction for noise contribution or phonon detection efficiency. The number of observable negative regions increases correspondingly as the phonon population traverses to higher phonon numbers and, at the same time, concentrates in particular number state $|n\rangle$. This is further manifested by a graduate transition in a faithful hierarchy of non-Gaussianity criteria up to four phonons in increasing order shown in the inset. It shows that, indeed, accumulated phonon number states and their quantum non-Gaussian aspects remain well limited to maximally 4 phonons.

We use the measured phonon number distributions to evaluate entanglement potential (EP) of quantum non-Gaussian states for future applications. EP, which is defined as logarithmic negativity of entangled state created from the studied state by energetically passive coupling between two mechanical oscillators, is plotted in Figure 3f. We can see that even though the increase in nonclassicality is best visible in the first step in which the oscillator state goes from vacuum to mostly single phonon state, it still monotonously increases with the number of repetitions. Importantly, this effect can be seen even though the measurements include a random and unavoidable diffusion
of phonon number statistics due to the excitation and decay on the reshuffling transition with finite Lamb–Dicke parameters. The accumulation process is apparently robust against experimental imperfections and can be applied also to states with high thermal energy resulting from a simple Doppler cooling process, irrespective of additional heating caused by the resetting of electronic state.

4. Conclusions

We have experimentally verified that nonclassicality of the generated phonon number distributions can be unconditionally accumulated. It is achievable by the modulation of thermal phonon number distributions using a natural Rabi interaction with a two-level system.\textsuperscript{[24,52,57]} It represents a highly nonlinear extension of nonclassicality accumulation from single-resonant optical parametric oscillators to the platforms which allow for a direct implementation of Rabi interaction.\textsuperscript{[4–6,10–36]} The realized experiment demonstrates an unprecedented possibility of deterministic generation of quantum non-Gaussian properties for controllable nonlinear interactions and promises a feasible bypass for no-go theorems for Fock state processing.\textsuperscript{[58–59]} The presented nonlinear interaction can be directly extended to nonlinear couplings in a solid-state mechanical oscillators\textsuperscript{[22,60–62]} and generation of nonclassicality in experimental systems of several coupled oscillators and spins.\textsuperscript{[63–65]}

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

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