Supplementary Information for

Nonlinear dynamics and quantum entanglement in optomechanical systems

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S1. PHOTON-PHONON COUPLING

The photon-phonon coupling can be transformed to the photon-photon coupling by applying the so-called polaron transformation [1]: $\hat{\hat{H}} = \hat{U}^{\dagger}\hat{H}\hat{U}$, in the absence of any driving and dissipation. The resulting Hamiltonian

$$\hat{\tilde{H}} = \hbar\omega_c \hat{a}^{\dagger} \hat{a} + \hbar\omega_M \hat{b}^{\dagger} \hat{b} - \hbar \frac{g_0^2}{\omega_M} (\hat{a}^{\dagger} \hat{a})^2$$
(S1)

has the eigenvalue $E_{nj} = \hbar \omega_c n - \hbar g_0^2 n^2 / \omega_M + \hbar \omega_M j$, where *n* and *j* are the photon and phonon numbers, respectively. The effective photon-photon coupling term makes the photon level spectrum anharmonic, leading to many interesting physical phenomena such as photon block effect and photon-induced tunneling [1, 2].

S2. RELEVANCE OF WEAK COUPLING REGIME

In order to understand the dynamics of fluctuations in the optomechanical system as described by the quantum Langevin equations, we consider the corresponding variational equations. For example, a dynamical variable x can be written as $\mathbf{x}(t) = \mathbf{x}_0(t) + \delta \mathbf{x}(t)$, where $\mathbf{x}_0(t)$ is the corresponding variable in the zeroth-order system and $\delta \mathbf{x}(t)$ characterizes the fluctuations. For the full system described by Eq. (1) in the main text, the zeroth-order system is given by

$$\ddot{q}_0 = -\omega_m^2 q_0 - \Gamma_M \dot{q}_0 + \sqrt{2} g_0 \omega_m |a_0|^2$$
(S2a)

$$\dot{a}_0 = -(\frac{\kappa}{2} + i\Delta_0)a_0 + i\sqrt{2}g_0a_0q_0 + E.$$
 (S2b)

Letting $x_0 = g_0 q_0$ and $\alpha_0 = \frac{a_0}{E}$, we can write Eq. (S2) as

$$\ddot{x_0} = -\omega_m^2 x_0 - \Gamma_M \dot{x_0} + \sqrt{2}g_0^2 E^2 \omega_m |\alpha_0|^2$$
(S3a)

$$\dot{\alpha_0} = -(\frac{\kappa}{2} + i\Delta_0)\alpha_0 + i\sqrt{2}\alpha_0 x_0 + 1,$$
 (S3b)

where the parameters g_0 and E appear as a product, indicating that the zeroth-order properties of the system are determined by this product as well as other system parameters. To study how the system dynamical behaviors depend on g_0 and E, we can conveniently fix their product $g_0^2 E^2$ and change one of them systematically, say g_0 .

As described in the main text, quasiperiodic motion emerges in the weak optomechanical coupling regime, i.e., $g_0 \ll \kappa$, and it can enhance quantum entanglement. This remarkable phenomenon, however, cannot be guaranteed to occur in the strong coupling regime $g_0 \sim \kappa$, due to the fact that the magnitude of the input noise behaves as $\xi \to g_0 \xi$ and $\sqrt{\kappa} a^{in} \to \sqrt{\kappa} a^{in}/E$. As we increase g_0 towards the strong coupling region, the amplitude of the input noise will be enhanced by a factor of the the same order of magnitude. In this case, noise and the zeroth-order terms will have comparable magnitude. Classically, the system dynamics will then be affected strongly by noise, making it difficult to assess the interplay between nonlinear dynamics and quantum entanglement.

S3. NUMERICAL SIMULATION OF THE QUANTUM LANGEVIN EQUATIONS

In order to gain insights into the dynamics of the optomechanical system and to validate the time-dependent covariant matrix method for quantifying quantum entanglement, we use a stochastic fourth-order Runge-Kutta (RK4) method to simulate the evolution of the quantum Langevin equations [7]. The stochastic RK4 method can yield accurate and stable solutions even when using step size Δ of two orders of magnitude larger than that in the widely used Heun's method [8].

The quantum Langevin equations can generally be written as

$$\frac{d\mathbf{X}}{dt} = \mathbf{F} + \mathbf{G} \cdot \mathbf{z},\tag{S4}$$

where $\mathbf{G} \cdot \mathbf{z} = [G_1 z_1, G_2 z_2, ..., G_n z_n]^T$, G_i is the autocorrelation of the *i*th noise input, and z_i is the Gaussian white noise with the normal distribution N(0, 1). The Stochastic RK4 method can be explicitly expressed as

$$\mathbf{X}(t+\Delta) = \mathbf{X}(t) + \sum_{j=0}^{3} p_j \mathbf{K}_j \Delta + \sum_{j=0}^{3} q_j \mathbf{M}_j \cdot \mathbf{z}$$
(S5)

where

$$\begin{split} \mathbf{K}_{0} &= \mathbf{F}(\mathbf{X}(t), t) \\ \mathbf{M}_{0} &= \mathbf{G}(\mathbf{X}(t), t) \\ \mathbf{X}^{(1)} &= \mathbf{X}(t) + \frac{1}{2}\mathbf{K}_{0}\Delta + \frac{1}{2}\mathbf{M}_{0} \cdot \mathbf{z} \\ \mathbf{K}_{1} &= \mathbf{F}(\mathbf{X}(t)^{(1)}, t + \frac{1}{2}\Delta) \\ \mathbf{M}_{1} &= \mathbf{G}(\mathbf{X}(t)^{(1)}, t + \frac{1}{2}\Delta) \\ \mathbf{X}^{(2)} &= \mathbf{X}(t) + \frac{1}{2}\mathbf{K}_{1}\Delta + \frac{1}{2}\mathbf{M}_{1} \cdot \mathbf{z} \\ \mathbf{K}_{2} &= \mathbf{F}(\mathbf{X}^{(2)}(t), t + \frac{1}{2}\Delta) \\ \mathbf{M}_{2} &= \mathbf{G}(\mathbf{X}^{(2)}(t), t + \frac{1}{2}\Delta) \\ \mathbf{M}_{3} &= \mathbf{K}(\mathbf{X}^{(3)}(t), t + \Delta) \\ \mathbf{M}_{3} &= \mathbf{G}(\mathbf{X}^{(3)}(t), t + \Delta) \end{split}$$

The stochastic RK4 algorithm converges to that based on the Stratonovich calculus [7]. For our optomechanical system, G is a constant, facilitating greatly numerical integration. Note that $M \cdot z$ has the same meaning as $G \cdot z$. We simulate the quantum Langevin equations a large number (e.g., 3000) of times to obtain well converged ensemble-averaged quantities, as exemplified in Fig. 3 in the main text.

S4. TIME EVOLUTION OF FLUCTUATIONS

As described in the main text, the time evolution of the fluctuations in the optomechanical systems is governed by

$$\dot{u}(t) = A(t)u(t) + n(t), \tag{S6}$$

where A(t) is a 4×4 matrix given by

$$A(t) = \begin{bmatrix} 0 & \omega_M & 0 & 0 \\ -\omega_M & -\Gamma_M & g_x(t) & g_y(t) \\ -g_y(t) & 0 & -\kappa & \Delta(t) \\ g_x(t) & 0 & -\Delta(t) & -\kappa \end{bmatrix}.$$
 (S7)

Here we have made the following substitutions: $\delta x = (\delta a + \delta a^{\dagger})/\sqrt{2}$ and $\delta y = -i(\delta a - \delta a^{\dagger})/\sqrt{2}$, so that $u = (\delta q, \delta p, \delta x, \delta y)^T$ and $n = (0, \xi, \sqrt{\kappa} x^{in}, \sqrt{\kappa} y^{in})^T$. Other variables are defined as $g_x(t) = g_0 \langle x(t) \rangle$, $g_y(t) = g_0 \langle y(t) \rangle$ and $\Delta(t) = \Delta_0 - g_0 \langle q(t) \rangle$.

S5. LOGARITHMIC NEGATIVITY

There has been no universal definition of quantum entanglement that can be applied to different situations of physical interest, nor any general quantitative measure that can be used to characterize the degree of quantum entanglement. Only special cases can be dealt with where, for example, the density operators are relatively simple. In a canonical bipartite system described by continuous variables, quantum entanglement can be quantified by the so-called measure of logarithmic negativity, defined for Gaussian quantum state as well as pure and symmetric states. In particular, say the quantum system has the density operator ρ and has a subsystem A. The logarithmic negativity is defined as [3]

$$E_{\mathcal{N}}(\rho) \equiv \log_2 ||\rho^{T_A}||_1, \tag{S8}$$

where ρ^{T_A} is the partial transpose of the bipartite mixed state ρ for its subsystem A, $|| \cdot ||_1$ means its trace norm and is expressed as

$$||\rho^{T_A}||_1 = 1 + 2|\sum_i \mu_i|,\tag{S9}$$

where μ_i 's are the negative eigenvalues of ρ^{T_A} . For a general mixed state of infinite dimension without any symmetry, it is extremely difficult to calculate the trace norm. However, for a Gaussian state, the method of symplectic diagonalization or normal-mode decomposition can be employed, in which the state is transformed into a tensor product of independent thermal oscillator states fully specified by their energies. Note that the quantum properties a Gaussian state can be completely characterized by its covariance matrix. The normal-mode decomposition then enables us to diagonalize the covariance matrix as diag $(c_1, c_1, c_2, c_2, ..., c_n, c_n)$, where c_i is the energy of the *i*th thermal oscillator state. For a state at thermal equilibrium, we have [4]

$$\rho = \frac{e^{-\beta a^{\dagger} a}}{\operatorname{Tr}[e^{-\beta a^{\dagger} a}]} = (1-z) \sum_{n=0} z^n |n\rangle \langle n|, \qquad (S10)$$

where $z = e^{-\beta}$ and $\beta \propto 1/(k_BT)$. In general, for a physical density operator, we have $z \ge 0$. However, the operator under consideration here is a partially transposed operator for which the uncertainty relation is not preserved. There can then be states with -1 < z < 0. Nonetheless, a connection between $||\rho^{T_A}||_1$ and c_i can be established via the quantity z through the definition of trace norm as well as the energy relation. Particularly, for a Gaussian state with diagonal covariance matrix diag(c, c), its trace norm is

$$F(c) = \begin{cases} 0, & \text{for } 2c \ge 1\\ -\log_2(2c), & \text{for } 2c \le 1 \end{cases}$$
(S11)

and the logarithmic negativity is the sum of different trace norms of c. The physical meaning is that, for $2c \ge 1$, i.e., $z \ge 0$, the state corresponds to a normal thermal state, indicating that energy is characterized by c. As the trace norm of this state is 1, it has no contribution to the entanglement of the total system. However, for $2c \le 1$, i.e., -1 < z < 0, a negative density operator arises with a non-trivial trace norm, which contributes to entanglement.

The quantum states in an optomechanical system are naturally bipartite state: any such state is composed of the entangled sub-states associated with the optical and mechanical degrees of freedom, respectively. In this case, the quantities c_1 and c_2 can be calculated from

$$(ic)^{4} + [\det(A) + \det(B) - 2\det(C)](ic)^{2} + \det\gamma = 0,$$
(S12)

where A, B, and C are the 2×2 block matrices comprising the covariant matrix

$$\gamma = \begin{pmatrix} A & C \\ C^T & B \end{pmatrix}.$$
 (S13)

It can be seen that det(A), det(B), det(C) and $det(\gamma)$ are four invariants under the symplectic transformation $S_A \oplus S_B$, where $S_A, S_B \in Sp(2, \mathbb{R})$. The conditions of c for the entangled states can be obtained directly from the PHS criterion [5] associated with the uncertainty relation [6]. We then have

$$E_{\mathcal{N}} = -\sum_{i=1}^{2} \min(0, \log(c_i)).$$
 (S14)

S6. TEMPERATURE DEPENDENCE

How robust is quantum entanglement against thermal fluctuations? To address this question, we calculate the dependence of maximum entanglement measure E_N on temperature for a large number of values of the laser driving power. In the system of quantum Langevin equations, a convenient way to incorporate the temperature effect is through the input noise. There are two sources of noise: vacuum radiation input noise $a^{in}(t)$ for the optical subsystem and the viscous force to the mechanical subsystem through the Brownian stochastic process $\xi(t)$. For the vacuum radiation input noise, at high optical frequency, the equilibrium mean thermal photon number

$$N(\omega_c) = \left[\exp\left(\hbar\omega_c/k_BT\right) - 1\right]^{-1}$$
(S15)



FIG. S1: For three values of the driving power, dependence of the entanglement maximum E_N on the environmental temperature T. In all cases, robust entanglement exists for T in the range of tens of millikelvin. Inset: rescaled average entanglement.

tends to zero [9]. This leads to the correlation functions

$$\langle a^{in}(t)a^{in,\dagger}(t')\rangle = [N(\omega_c) + 1]\delta(t - t') \to \delta(t - t')$$
(S16a)

$$\langle a^{in,\dagger}(t)a^{in}(t')\rangle = N(\omega_c)\delta(t-t') \to 0$$
 (S16b)

which do not depend on the temperature. For the case of Brownian noise, as its frequency has the same order of magnitude as the mechanical frequency, the equilibrium mean thermal phonon number can remain finite even at relative low temperature. As a result, the temperature of the mechanical reservoir can affect the dynamics of the system through

$$\bar{n} = \frac{1}{\exp\left[\hbar\omega_M/(k_B T)\right] - 1}.$$
(S17)

Three representative cases are shown in Fig. S1. We plot the maximum of entanglement as well as the rescaled average entanglement within the stable periodic or quasiperiodic regime after the transient behaviour has died out. In general, we find that, for low mechanical dissipation rate Γ_M and high mechanical frequency, entanglement can persist in the temperature range up to tens of millikevins, which is experimentally readily achievable. This is reasonable as the magnitude of the autocorrelation of the mechanical Brownian noise is $\Gamma_M(2\bar{n} + 1)$. Insofar as the mechanical mode has a high-Q factor, the effect of the stochastic mechanical effect is small. Especially, for high frequency, the thermal occupation number of the mechanical mode is small, leading to a robust entanglement. A rather surprising phenomenon is that quantum entanglement corresponding to classical quasiperiodic motion is more temperature-robust than that associated with periodic motion.

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