Supplemental Material for "Nonreciprocal Bundle Emissions of Quantum Entangled Pairs"

Qian Bin,[1,2] Hui Jing,[3] Ying Wu,[1] Franco Nori,[4,5,6] and Xin-You Lü[1,*]

[1] *School of Physics and Institute for Quantum Science and Engineering, Huazhong University of Science and Technology, and Wuhan Institute of Quantum Technology, Wuhan 430074, China*
[2] *College of Physics, Sichuan University, Chengdu 610065, China*
[3] *Key Laboratory of Low-Dimensional Quantum Structures and Quantum Control of Ministry of Education, Department of Physics and Synergetic Innovation Center for Quantum Effects and Applications, Hunan Normal University, Changsha 410081, China*
[4] *Theoretical Quantum Physics Laboratory, Cluster for Pioneering Research, RIKEN, Wakoshi, Saitama 351-0198, Japan*
[5] *Center for Quantum Computing, RIKEN, Wakoshi, Saitama 351-0198, Japan*
[6] *Physics Department, The University of Michigan, Ann Arbor, Michigan 48109-1040, USA*

(Dated: July 19, 2024)

In this Supplemental Material, we present the technical details of the effective Hamiltonian and multiquanta resonances discussed in the main text. In Sec. I, we derive the system's effective Hamiltonian using the standard linearization procedure. In Sec. II, we provide a detailed derivation of multiquanta resonance conditions within the Mollow regime. In Sec. III, we discuss the entanglement witness based on quantum Fisher information and covariance. In Sec. IV, we discuss the experimental feasibility of our theoretical model in detail.

I. DERIVATION OF THE EFFECTIVE HAMILTONIAN VIA THE QUANTUM MASTER EQUATION

The Hamiltonian of the system we consider is

$$\begin{aligned} H_t =&(\omega_a + \Delta_F)a^\dagger a + \omega_b b^\dagger b + \omega_m m^\dagger m + \omega_\sigma \sigma^\dagger \sigma + \lambda'_{ab} a^\dagger a(b^\dagger + b) + \lambda'_{am} a^\dagger a(m^\dagger + m) + \lambda'_{a\sigma}(a^\dagger \sigma + a\sigma^\dagger) \\ &+ \xi_d(a^\dagger e^{-i\omega_d t} + a e^{i\omega_d t}) + \xi_p(\sigma^\dagger e^{-i\omega_d t} + \sigma e^{i\omega_d t}), \end{aligned} \tag{S1}$$

where a (b, m) is the annihilation operator of the optical (mechanical, magnon) mode with corresponding frequency ω_a (ω_b, ω_m), $\sigma = |g\rangle\langle e|$ is the lowering operator of the atom with frequency ω_σ, Δ_F is the Fizeau shift, $\lambda'_{a\sigma}$ (λ'_{ab}, λ'_{am}) is the coupling strength between the optical mode and atom (mechanical mode, magnon mode), ξ_d (ξ_p) and ω_d are the amplitude and frequency of the driving at the optical mode (atom), respectively. In the frame rotating with frequency ω_d, Eq. (S1) becomes

$$\begin{aligned} H_t =&(\omega_a - \omega_d + \Delta_F)a^\dagger a + \omega_b b^\dagger b + \omega_m m^\dagger m + (\omega_\sigma - \omega_d)\sigma^\dagger \sigma + \lambda'_{ab} a^\dagger a(b^\dagger + b) + \lambda'_{am} a^\dagger a(m^\dagger + m) + \lambda'_{a\sigma}(a^\dagger \sigma + a\sigma^\dagger) \\ &+ \xi_d(a^\dagger + a) + \xi_p(\sigma^\dagger + \sigma). \end{aligned} \tag{S2}$$

Taking dissipations into consideration, the dissipative dynamics of the system can be described by the quantum master equation

$$\begin{aligned} \frac{d}{dt}\rho =& -i[H_t, \rho] + \frac{\kappa_a}{2}(2a\rho a^\dagger - a^\dagger a\rho - \rho a^\dagger a) + \frac{\kappa_b}{2}(2b\rho b^\dagger - b^\dagger b\rho - \rho b^\dagger b) \\ &+ \frac{\kappa_m}{2}(2m\rho m^\dagger - m^\dagger m\rho - \rho m^\dagger m) + \frac{\gamma}{2}(2\sigma\rho\sigma^\dagger - \sigma^\dagger\sigma\rho - \rho\sigma^\dagger\sigma). \end{aligned} \tag{S3}$$

We shift the optical, phonon, and mechanical modes with their mean values α, β, and μ, i.e., $a = \alpha + \delta a$, $b = \beta + \delta b$, and $m = \mu + \delta m$, Eq. (S3) becomes

$$\begin{aligned}
\frac{d}{dt}\rho =& -i[H_t,\rho] + \frac{\kappa_a}{2}[2(\alpha+\delta a)\rho(\alpha^*+\delta a^\dagger) - (\alpha^*+\delta a^\dagger)(\alpha+\delta a)\rho - \rho(\alpha^*+\delta a^\dagger)(\alpha+\delta a)] \\
&+ \frac{\kappa_b}{2}[2(\beta+\delta b)\rho(\beta^*+\delta b^\dagger) - (\beta^*+\delta b^\dagger)(\beta+\delta b)\rho - \rho(\beta^*+\delta b^\dagger)(\beta+\delta b)] \\
&+ \frac{\kappa_m}{2}[2(\mu+\delta m)\rho(\mu^*+\delta m^\dagger) - (\mu^*+\delta m^\dagger)(\mu+\delta m)\rho - \rho(\mu^*+\delta m^\dagger)(\mu+\delta m)] \\
&+ \frac{\gamma}{2}[2\sigma\rho\sigma^\dagger - \sigma^\dagger\sigma\rho - \rho\sigma^\dagger\sigma] \\
=& -i[H_t,\rho] + \frac{\kappa_a}{2}(\alpha^*\delta a\rho - \alpha^*\rho\delta a - \alpha\delta a^\dagger\rho + \alpha\rho\delta a^\dagger) \\
&+ \frac{\kappa_b}{2}(\beta^*\delta b\rho - \beta^*\rho\delta b - \beta\delta b^\dagger\rho + \beta\rho\delta b^\dagger) + \frac{\kappa_m}{2}(\mu^*\delta m\rho - \mu^*\rho\delta m - \mu\delta m^\dagger\rho + \mu\rho\delta m^\dagger) \\
&+ \frac{\kappa_a}{2}(2\delta a\rho\delta a^\dagger - \delta a^\dagger\delta a\rho - \rho\delta a^\dagger\delta a) + \frac{\kappa_b}{2}(2\delta b\rho\delta b^\dagger - \delta b^\dagger\delta b\rho - \rho\delta b^\dagger\delta b) \\
&+ \frac{\kappa_m}{2}(2\delta m\rho\delta m^\dagger - \delta m^\dagger\delta m\rho - \rho\delta m^\dagger\delta m) + \frac{\gamma}{2}(2\sigma\rho\sigma^\dagger - \sigma^\dagger\sigma\rho - \rho\sigma^\dagger\sigma) \\
=& -i[H_1+H_2,\rho] + \frac{\kappa_a}{2}(2\delta a\rho\delta a^\dagger - \delta a^\dagger\delta a\rho - \rho\delta a^\dagger\delta a) + \frac{\kappa_b}{2}(2\delta b\rho\delta b^\dagger - \delta b^\dagger\delta b\rho - \rho\delta b^\dagger\delta b) \\
&+ \frac{\kappa_m}{2}(2\delta m\rho\delta m^\dagger - \delta m^\dagger\delta m\rho - \rho\delta m^\dagger\delta m) + \frac{\gamma}{2}(2\sigma\rho\sigma^\dagger - \sigma^\dagger\sigma\rho - \rho\sigma^\dagger\sigma),
\end{aligned} \tag{S4}$$

where the Hamiltonians H_1 and H_2 are, respectively,

$$\begin{aligned}
H_1 =& (\omega_a - \omega_d + \Delta_F)|\alpha|^2 + \omega_b|\beta|^2 + \omega_m|\mu|^2 + \lambda'_{ab}|\alpha|^2(\beta^*+\beta) + \lambda'_{am}|\alpha|^2(\mu^*+\mu) + \xi_d(\alpha^*+\alpha) \\
&+ [(\omega_a - \omega_d + \Delta_F)\alpha^* + \lambda'_{ab}\alpha^*(\beta^*+\beta) + \lambda'_{am}\alpha^*(\mu^*+\mu) + \xi_d + \frac{i}{2}\alpha^*\kappa_a]\delta a \\
&+ [(\omega_a - \omega_d + \Delta_F)\alpha + \lambda'_{ab}\alpha(\beta^*+\beta) + \lambda'_{am}\alpha(\mu^*+\mu) + \xi_d - \frac{i}{2}\alpha\kappa_a]\delta a^\dagger \\
&+ (\omega_b\beta^* + \lambda'_{ab}|\alpha|^2 + \frac{i}{2}\beta^*\kappa_b)\delta b + (\omega_b\beta + \lambda'_{ab}|\alpha|^2 - \frac{i}{2}\beta\kappa_b)\delta b^\dagger \\
&+ (\omega_m\mu^* + \lambda'_{am}|\alpha|^2 + \frac{i}{2}\mu^*\kappa_m)\delta m + (\omega_m\mu + \lambda'_{am}|\alpha|^2 - \frac{i}{2}\mu\kappa_m)\delta m^\dagger,
\end{aligned} \tag{S5}$$

and

$$\begin{aligned}
H_2 =& [\omega_a - \omega_d + \Delta_F + \lambda'_{ab}(\beta^*+\beta) + \lambda'_{am}(\mu^*+\mu)]\delta a^\dagger\delta a + \omega_b\delta b^\dagger\delta b + \omega_m\delta m^\dagger\delta m + (\omega_\sigma - \omega_d)\sigma^\dagger\sigma \\
&+ \lambda'_{ab}(\alpha\delta a^\dagger + \alpha^*\delta a)(\delta b^\dagger + \delta b) + \lambda'_{am}(\alpha\delta a^\dagger + \alpha^*\delta a)(\delta m^\dagger + \delta m) + \lambda'_{a\sigma}(\delta a\sigma^\dagger + \delta a^\dagger\sigma) + \lambda'_{a\sigma}(\alpha\sigma^\dagger + \alpha^*\sigma) \\
&+ \lambda'_{ab}\delta a^\dagger\delta a(\delta b^\dagger + \delta b) + \lambda'_{am}\delta a^\dagger\delta a(\delta m^\dagger + \delta m) + \xi_p(\sigma^\dagger + \sigma).
\end{aligned} \tag{S6}$$

Setting $\lambda_{ab} = \lambda'_{ab}|\alpha|$, $\lambda_{am} = \lambda'_{am}|\alpha|$, $\lambda_{a\sigma} = \lambda'_{a\sigma}$, and $\xi = \lambda'_{a\sigma}|\alpha| + \xi_p$, the above equation Eq. (S6) becomes

$$\begin{aligned}
H_2 =& [\omega_a - \omega_d + \Delta_F + \lambda'_{ab}(\beta^*+\beta) + \lambda'_{am}(\mu^*+\mu)]\delta a^\dagger\delta a + \omega_b\delta b^\dagger\delta b + \omega_m\delta m^\dagger\delta m + (\omega_\sigma - \omega_d)\sigma^\dagger\sigma \\
&+ \lambda_{ab}(\delta a^\dagger + \delta a)(\delta b^\dagger + \delta b) + \lambda_{am}(\delta a^\dagger + \delta a)(\delta m^\dagger + \delta m) + \lambda_{a\sigma}(\delta a\sigma^\dagger + \delta a^\dagger\sigma) + \xi(\sigma^\dagger + \sigma) \\
&+ \lambda'_{ab}\delta a^\dagger\delta a(\delta b^\dagger + \delta b) + \lambda'_{am}\delta a^\dagger\delta a(\delta m^\dagger + \delta m).
\end{aligned} \tag{S7}$$

The steady-state mean values α, β, and μ satisfy

$$\begin{aligned}
&(\omega_a - \omega_d + \Delta_F)\alpha + \lambda'_{ab}\alpha(\beta^*+\beta) + \lambda'_{am}\alpha(\mu^*+\mu) + \xi_d - \frac{i}{2}\alpha\kappa = 0, \\
&\omega_b\beta + \lambda'_{ab}|\alpha|^2 - \frac{i}{2}\beta\kappa_b = 0, \\
&\omega_m\mu + \lambda'_{am}|\alpha|^2 - \frac{i}{2}\mu\kappa_m = 0,
\end{aligned} \tag{S8}$$

the system dynamics is then decided by

$$\begin{aligned}
\frac{d}{dt}\rho =& -i[H_2,\rho] + \frac{\kappa_a}{2}(2\delta a\rho\delta a^\dagger - \delta a^\dagger\delta a\rho - \rho\delta a^\dagger\delta a) + \frac{\kappa_b}{2}(2\delta b\rho\delta b^\dagger - \delta b^\dagger\delta b\rho - \rho\delta b^\dagger\delta b) \\
&+ \frac{\kappa_m}{2}(2\delta m\rho\delta m^\dagger - \delta m^\dagger\delta m\rho - \rho\delta m^\dagger\delta m) + \frac{\gamma}{2}(2\sigma\rho\sigma^\dagger - \sigma^\dagger\sigma\rho - \rho\sigma^\dagger\sigma).
\end{aligned} \tag{S9}$$

Under the condition of strong driving at the optical mode, the higher-order terms $\lambda'_{ab}\delta a^\dagger\delta a(\delta b^\dagger+\delta b)$ and $\lambda'_{am}\delta a^\dagger\delta a(\delta m^\dagger+\delta m)$ can be neglected safely because of $|\alpha|\gg 1$ and $\lambda'_{ab},\lambda'_{am}\ll\lambda_{ab},\lambda_{am}$. Then the system Hamiltonian H_2 can be further simplified as

$$\begin{aligned}H_{\text{eff}}=&[\omega_a-\omega_d+\Delta_F+\lambda'_{ab}(\beta^*+\beta)+\lambda'_{am}(\mu^*+\mu)]\delta a^\dagger\delta a+\omega_b\delta b^\dagger\delta b+\omega_m\delta m^\dagger\delta m+(\omega_\sigma-\omega_d)\sigma^\dagger\sigma\\&+\lambda_{ab}(\delta a^\dagger+\delta a)(\delta b^\dagger+\delta b)+\lambda_{am}(\delta a^\dagger+\delta a)(\delta m^\dagger+\delta m)+\lambda_{a\sigma}(\delta a\sigma^\dagger+\delta a^\dagger\sigma)+\xi(\sigma^\dagger+\sigma).\end{aligned}\tag{S10}$$

Assuming $\delta a\to a$, $\delta b\to b$, and $\delta m\to m$, the above equation becomes

$$\begin{aligned}H=&[\omega_a-\omega_d+\Delta_F+\lambda'_{ab}(\beta^*+\beta)+\lambda'_{am}(\mu^*+\mu)]a^\dagger a+\omega_b b^\dagger b+\omega_m m^\dagger m+(\omega_\sigma-\omega_d)\sigma^\dagger\sigma\\&+\lambda_{ab}(a^\dagger+a)(b^\dagger+b)+\lambda_{am}(a^\dagger+a)(m^\dagger+m)+\lambda_{a\sigma}(a\sigma^\dagger+a^\dagger\sigma)+\xi(\sigma^\dagger+\sigma)\\=&[\omega_a-\omega_d+\Delta_F+\lambda'_{ab}(\beta^*+\beta)+\lambda'_{am}(\mu^*+\mu)]a^\dagger a+\omega_b b^\dagger b+\omega_m m^\dagger m\\&+[\omega_\sigma-\omega_a+\omega_a-\omega_d+\lambda'_{ab}(\beta^*+\beta)+\lambda'_{am}(\mu^*+\mu)-\lambda'_{ab}(\beta^*+\beta)-\lambda'_{am}(\mu^*+\mu)]\sigma^\dagger\sigma\\&+\lambda_{ab}(a^\dagger+a)(b^\dagger+b)+\lambda_{am}(a^\dagger+a)(m^\dagger+m)+\lambda_{a\sigma}(a\sigma^\dagger+a^\dagger\sigma)+\xi(\sigma^\dagger+\sigma)\\=&(\Delta_{ad}+\Delta_F)a^\dagger a+\omega_b b^\dagger b+\omega_m m^\dagger m+(\Delta_{ad}+\Delta_{\sigma a})\sigma^\dagger\sigma\\&+\lambda_{ab}(a^\dagger+a)(b^\dagger+b)+\lambda_{am}(a^\dagger+a)(m^\dagger+m)+\lambda_{a\sigma}(a\sigma^\dagger+a^\dagger\sigma)+\xi(\sigma^\dagger+\sigma),\end{aligned}\tag{S11}$$

where $\Delta_{ad}=\omega_a-\omega_d+\lambda'_{ab}(\beta^*+\beta)+\lambda'_{am}(\mu^*+\mu)$ and $\Delta_{\sigma a}=\omega_\sigma-\omega_a-\lambda'_{ab}(\beta^*+\beta)-\lambda'_{am}(\mu^*+\mu)$.

II. DERIVATION OF MULTIQUANTA RESONANCES CONDITIONS

In the large pumping regime, the strong driving laser can dress the atom, and the system forms new eigenstates that are a quantum superposition of the bare states $\{|e\rangle,|g\rangle\}$. The subsystem Hamiltonian for the strongly driven atom is

$$H_\sigma=(\Delta_{\sigma a}+\Delta_{ad})\sigma^\dagger\sigma+\xi(\sigma^\dagger+\sigma),\tag{S12}$$

with the eigenvalues

$$E_{|\pm\rangle}=\frac{\Delta_{\sigma a}+\Delta_{ad}\pm\sqrt{(\Delta_{\sigma a}+\Delta_{ad})^2+4\xi^2}}{2},\tag{S13}$$

and corresponding eigenstates

$$|+\rangle=c_+|g\rangle+c_-|e\rangle,\quad |-\rangle=c_-|g\rangle-c_+|e\rangle,\tag{S14}$$

where

$$c_\pm=\sqrt{\frac{2\xi^2}{(\Delta_{\sigma a}+\Delta_{ad})^2+4\xi^2\pm(\Delta_{\sigma a}+\Delta_{ad})\sqrt{(\Delta_{\sigma a}+\Delta_{ad})^2+4\xi^2}}}\tag{S15}$$

and $c_+^2+c_-^2=1$. Together with the photon, phonon, and magnon modes, and ignoring the influences of the JC interaction and the linear interactions in the optomagnetic system on the energy structure, the eigenvalues of the system Hamiltonian become

$$E_{|n_an_bn_m\pm\rangle}=n_a(\Delta_{ad}+\Delta_F)+n_b\omega_b+n_m\omega_m+\frac{\Delta_{\sigma a}+\Delta_{ad}\pm\sqrt{(\Delta_{\sigma a}+\Delta_{ad})^2+4\xi^2}}{2}.\tag{S16}$$

In the following, we discuss the resonance conditions considered in the main text. For convenience, we always fix the resonator spinning along the counterclockwise direction. The Fizeau shifts $\Delta_F>0$ and $\Delta_F<0$ correspond to the situations of driving the resonator from the left and right, respectively.

First, when the spinning resonator is driven from the left, and the total energy of a single clockwise photon and a single magnon matches the transition between the states $|+\rangle$ and $|-\rangle$, i.e., $E_{|000+\rangle}=E_{|101-\rangle}$, we have

$$(\Delta_{ad}+|\Delta_F|)+\omega_m-\sqrt{(\Delta_{\sigma a}+\Delta_{ad})^2+4\xi^2}=0,\tag{S17}$$

and

$$\Delta_{ad}=\frac{\Delta_{\sigma a}^2+4\xi^2-(\omega_m+|\Delta_F|)^2}{2(\omega_m+|\Delta_F|-\Delta_{\sigma a})}. \tag{S18}$$

In other words, these two states $|000+\rangle$ and $|101-\rangle$ are degenerate when the atom is driven at the photon-magnon resonance. When the spinning resonator is driven from the right, and the total energy of a single counterclockwise photon and a single magnon matches with the transition between the states $|+\rangle$ and $|-\rangle$, the photon-magnon resonance condition becomes

$$\Delta_{ad}=\frac{\Delta_{\sigma a}^2+4\xi^2-(\omega_m-|\Delta_F|)^2}{2(\omega_m-|\Delta_F|-\Delta_{\sigma a})}. \tag{S19}$$

The above results correspond to the excitation of the photon-magnon state $|101-\rangle$. The parameter conditions are in agreement with the results shown in the blue area of Fig.1(c) and the orange area of Fig.1(c) in the main text, respectively.

Second, when the spinning resonator is driven from the left, and the total energy of a single clockwise photon and a single phonon matches the transition between the states $|+\rangle$ and $|-\rangle$, i.e., $E_{|000+\rangle}=E_{|110-\rangle}$ (the states $|000+\rangle$ and $|110-\rangle$ are degenerate), we have

$$(\Delta_{ad}+|\Delta_F|)+\omega_b-\sqrt{(\Delta_{\sigma a}+\Delta_{ad})^2+4\xi^2}=0, \tag{S20}$$

and

$$\Delta_{ad}=\frac{\Delta_{\sigma a}^2+4\xi^2-(\omega_b+|\Delta_F|)^2}{2(\omega_b+|\Delta_F|-\Delta_{\sigma a})}. \tag{S21}$$

When the spinning resonator is driven from the right, and the total energy of a single counterclockwise photon and a single phonon matches with the transition between the states $|+\rangle$ and $|-\rangle$, the photon-phonon resonance condition becomes

$$\Delta_{ad}=\frac{\Delta_{\sigma a}^2+4\xi^2-(\omega_b-|\Delta_F|)^2}{2(\omega_b-|\Delta_F|-\Delta_{\sigma a})}. \tag{S22}$$

The above results correspond to the excitation of the photon-phonon state $|110-\rangle$. The parameter conditions are in agreement with those displayed in the orange area of Fig.1(c) and the pink area of Fig.1(d) in the main text, respectively.

Lastly, because of the system parameters chosen in the main text, the results from the right sides of Eq.(S19) and Eq.(S21) are the same, as shown in the orange areas of Figs.1(c) and 1(d). This demonstrates that, in this parameter regime, the photon-phonon state is resonantly excited when the resonator is driven from the left, while the photon-magnon state is also resonantly excited when the resonator is driven from the right.

III. THE OPTIMAL LOCAL OPERATORS IN QUANTUM FISHER INFORMATION

Multipartite entanglement can be characterized based on quantum Fisher information (QFI) and covariance. For an arbitrary separable quantum states of N particles, the QFI and covariance must satisfy the inequality [1, 2]

$$F_Q[\rho_{\rm sep},\sum_{j=1}^{N}A_j]\ \le 4\sum_{j=1}^{N}{\rm Var}(A_j)_{\rho_{\rm sep}}\equiv B_n, \tag{S23}$$

where A_j is a local observable for the jth particle, and ${\rm Var}(A_j)_\rho=\langle A_j^2\rangle_\rho-\langle A_j\rangle_\rho^2$. The violation of Eq. (S23) for any choice of A_j is a sufficient criterion for entanglement. However, certain choices of operators A_j may be better suited than others to detect the entanglement of a given state ρ. The optimal local operator can be given by a combination of accessible operators $A_j=\sum_m c_j^{(m)}A_j^{(m)}=\mathbf{c}_j\cdot\mathbf{A}_j$, where $\mathbf{A}_j=(A_j^{(1)},A_j^{(2)},\dots)^T$ and $\mathbf{c}_j=(c_j^{(1)},c_j^{(2)},\dots)$ are the set of operators for jth particle and corresponding coefficients. The full operator is given by $A(\mathbf{c})=\sum_{j=1}^{N}\mathbf{c}_j\cdot\mathbf{A}_j=\mathbf{c}\cdot\mathbf{A}$, with the combined vectors $\mathbf{c}=(\mathbf{c}_1,\mathbf{c}_2,\dots,\mathbf{c}_N)$ and the operators $\mathbf{A}=[\mathbf{A}_1,\dots,\mathbf{A}_N]$. To detect the multipartite entanglement in our model, we choose the family of accessible operators $\mathbf{A}_j=(\sigma_j^x,\sigma_j^y,\sigma_j^z)^T$

for the atom and $\mathbf{A}_j = (x_j, p_j, x_j^2, p_j^2, (x_j p_j + p_j x_j)/2)^T$ for the mode o ($o = \{a, b, m\}$), where $x = o + o^\dagger$ and $p = -i(o - o^\dagger)$ [2]. Then the optimal local operators can be given by finding the optimal combined vectors $\mathbf{c}$. According to Eq. (S23), $F_Q[\rho, A(\mathbf{c})] - 4\sum_{j=1}^{N} \mathrm{Var}(\mathbf{c}_j \cdot \mathbf{A}_j)$ must be nonpositive for arbitrary choices of $\mathbf{c}$ whenever ρ is separable. To find the optimal $\mathbf{c}$, we expand the QFI and covariance in their matrix forms. The QFI can be written as $F_Q[\rho, A(\mathbf{c})] = \mathbf{c}Q_\rho^{\mathcal{A}}\mathbf{c}^T$ [3], where $Q_\rho^{\mathcal{A}}$ is the QFI matrix with elements

$$(Q_\rho^{\mathcal{A}})_{jj'}^{mm'} = 2\sum_{k,k'} \frac{(p_k - p_{k'})^2}{p_k + p_{k'}} \langle\psi_k|A_j^{(m)}|\psi_{k'}\rangle\langle\psi_{k'}|A_{j'}^{(m')}|\psi_k\rangle \tag{S24}$$

on the basis of spectral decomposition $\rho = \sum_k p_k|\psi_k\rangle\langle\psi_k|$ [4]. The covariance can be written as $\sum_{j=1}^{N} \mathrm{Var}(\mathbf{c}_j \cdot \mathbf{A}_j) = \mathbf{c}\Gamma_\rho^{\mathcal{A}}\mathbf{c}^T$, where the covariance matrix $\Gamma_\rho^{\mathcal{A}}$ has elements $(\Gamma_\rho^{\mathcal{A}})_{jj'}^{mm'} = \mathrm{Cov}(A_j^{(m)}, A_{j'}^{(m')})_\rho$ and $(\Gamma_\rho^{\mathcal{A}})_{jj}^{mm'} = 0$. Combining the quantum Fisher matrix $Q_\rho^{\mathcal{A}}$ with the covariance matrix $\Gamma_\rho^{\mathcal{A}}$, the separability criterion reads

$$\mathbf{c}(Q_\rho^{\mathcal{A}} - \Gamma_\rho^{\mathcal{A}})\mathbf{c}^T < 0. \tag{S25}$$

The optimal $\mathbf{c}$ can be given by finding the optimal eigenvalue and corresponding eigenstate of the matrix $Q_\rho^{\mathcal{A}} - \Gamma_\rho^{\mathcal{A}}$. Further, we obtain the optimal local operators A_j for the entanglement witness.

A photon-phonon separable state can be described as $\rho_{ab} = \sum_k p_k\rho_a^k \otimes \rho_b^k$ (ρ_j is the reduced density operator). The right-hand side of of Eq. (S23) characterizes the bounds of a given state and local operator A_j. In our case, we have

$$\begin{aligned} B_1(\rho_{ab}) &= 4[\mathrm{Var}(A_a)\rho_a + \mathrm{Var}(A_b)_{\rho_b}], \\ B_2(\rho_{ab}) &= 4[\mathrm{Var}(A_a + A_b)_{\rho_{ab}}]. \end{aligned} \tag{S26}$$

If the quantity $W_1^{ab} = F_Q[\rho_{ab}, A(\mathbf{c})] - 4\sum_{j=a,b} \mathrm{Var}(A_j)_{\rho_j} = F_Q[\rho_{ab}, A(\mathbf{c})] - B_1(\rho_{ab}) > 0$, the photon-phonon state is not separable, i.e., there is entanglement between the photon mode and phonon mode. The inequality $F_Q[\rho_{ab}, A(\mathbf{c})] \leq B_2(\rho_{ab})$ is a bound valid for all physical states ρ_{ab}.

A photon-phonon-atom fully separable state can be described as $\rho_{ab\sigma} = \sum_{k'} p_{k'}\rho_a^{k'} \otimes \rho_b^{k'} \otimes \rho_\sigma^{k'}$. In this case, the bounds become

$$\begin{aligned} B_1(\rho_{ab\sigma}) &= 4[\mathrm{Var}(A_a)\rho_a + \mathrm{Var}(A_b)_{\rho_b} + \mathrm{Var}(A_\sigma)_{\rho_\sigma}], \\ B_2(\rho_{ab\sigma}) &= 4\max\{\mathrm{Var}(A_a + A_b)_{\rho_{ab}} + \mathrm{Var}(A_\sigma)_{\rho_\sigma}, \mathrm{Var}(A_a + A_\sigma)_{\rho_{a\sigma}} + \mathrm{Var}(A_b)_{\rho_b}, \mathrm{Var}(A_b + A_\sigma)_{\rho_{b\sigma}} + \mathrm{Var}(A_a)_{\rho_a}\}, \\ B_3(\rho_{ab\sigma}) &= 4[\mathrm{Var}(A_a + A_b + A_\sigma)\rho_{ab\sigma}]. \end{aligned} \tag{S27}$$

If $W_1^{ab\sigma} = F_Q[\rho_{ab\sigma}, A(\mathbf{c})] - 4\sum_{j=a,b,\sigma} \mathrm{Var}(A_j)_{\rho_j} = F_Q[\rho_{ab\sigma}, A(\mathbf{c})] - B_1(\rho_{ab\sigma}) > 0$, there is entanglement between the atom, photon mode, and phonon mode. Furthermore, if $W_2^{ab\sigma} = F_Q[\rho_{ab\sigma}, A(\mathbf{c})] - B_2(\rho_{ab\sigma}) > 0$, the state is fully inseparable, i.e., there is a fully inseparable photon-magnon-atom tripartite entanglement [2]. The inequality $F_Q[\rho_{ab\sigma}, A(\mathbf{c})] \leq B_3(\rho_{ab\sigma})$ is a bound valid for all physical states $\rho_{ab\sigma}$. Here it is only necessary to replace mode b with mode m to characterize the entanglement between the magnon mode and others.

IV. DISCUSSION OF EXPERIMENTAL FEASIBILITY

Regarding experimental implementations, our proposal could be implemented in a hybrid setup [5, 6], where a coherently driven two-level atom is coupled to a spinning a spinning yttrium iron garnet (YIG) microsphere fixed on a rotating platform [7]. The microsphere supports a mechanical breathing mode and two countercirculating optical whispering gallery modes (WGMs), with the WGM being strongly driven by the input light [5]. Access to the input and output light fields of the microresonator is provided by a tapered optical fiber coupler interfaced with the resonator. The WGMs are modulated by the mechanical breathing mode when the input light pumps at the WGM. Moreover, because of the spin-orbit coupling of light, the WGMs have a spin along the z direction. By applying a magnetic field parallel to the equator to the microsphere and exciting the magnon mode with an antenna coupling to a microwave field, the WGMs could also be modulated by the dynamic magnetic field via the Faraday effect [6]. Here, scaling down the microsphere size to reduce the mode volumes of both magnon and optical fields, or engineering the microresonator structure such as microrings to increase the overlap between optical and magnon fields, can enhance the coupling between the optical mode and magnon mode. The effective linear coupling strength between the optical and magnon (mechanical) mode that is proportional to the classical cavity amplitude can also be adjusting by the strong driving on the optical mode.

Moreover, the selection of microresonator is not limited to a microsphere and can also be replaced with a toroidal or bottle microresonator [8]. The two-level atom of choice can be a cesium atom (e.g., the $6S_{1/2}, F = 4 \rightarrow 6P_{3/2}, F' = 5$ transition in cesium atom) [9–11] or rubidium atom (e.g., the $5S_{1/2}, F = 3 \rightarrow 5P_{3/2}, F' = 4$ transition in rubidium atom) [12–15]. The atom can be coupled with the microresonator by trapping the atom in the vicinity of the microresonator. However, trapping a single atom in the vicinity of a microresonator is indeed a daunting task due to several technical and physical complexities. First, precisely positioning and maintaining a single atom near a microresonator requires extremely high stability and accuracy in spatial control, since the optical fields around microresonators can vary significantly over nanometer scales. The use of optical tweezers or other trapping methods to hold a single atom in place involves complex manipulations of light fields. The intensity, phase, and polarization of these light fields must be precisely controlled to create stable trapping potentials without causing excessive heating or ionization of the atom. Second, efficient coupling of the atom to the microresonator's evanescent field requires the atom to be placed within a fraction of the wavelength of the light used, further complicating the spatial control challenge. Lastly, atoms must typically be cooled to very low temperatures to reduce thermal motion, which otherwise could lead to loss of control over the atom's position relative to the microsphere. Any perturbations from the environment (like mechanical vibrations, electromagnetic noise, etc.) can displace the atom from its optimal position, making sustained interaction with the microresonator difficult.

In recent years, several experiments have successfully achieved coupling between individual cesium atoms (or rubidium atoms) and various types of microresonators, including microspheres, microtoroidal resonators, and bottle microresonators [9–15]. Initially, these experiments utilize a magneto-optical trap to trap and cool atomic clouds in a 'magneto-optical trap chamber'. Subsequently, the trapping and cooling beams are switched off, allowing the atoms to fall onto the microresonator, thus facilitating the coupling of the individual atoms with the evanescent field of the microresonator's WGMs. Furthermore, the addition of an optical dipole trap—created by the interference of the incoming laser beam with its reflection from the microresonator surface—is more likely to trap the falling atoms, thereby increasing the interaction time between the atom and the microresonator [15, 16]. Based on the experimental groundwork, our model can also employ the techniques described in these studies to achieve the trapping of a single atom and its coupling with a microresonator. In our approach, the atom is initially cooled and trapped within a chamber using a magneto-optical trap, then transported to the vicinity of the microresonator. Here, the microresonator's WGM evanescent field, which decays exponentially outside the resonator's surface, interacts with the atom [9–16]. An optical dipole trap may be introduced to enhance the atom's trapping and prolong its interaction time with the microresonator [15, 16]. The decay rate of the cesium or rubidium atom used in trapping near a microresonator has linewidth of tens of MHz. The coupling strength between the selected atom and the microresonator depends on the properties of the microresonator and the distance between the atom and the microresonator's surface. Placing the atom closer to the surface of the microresonator [11, 15], or reducing the size of the microresonator to reduce the mode volume [17, 18], can enhance this coupling strength. H. J. Kimble and his collaborators, using finite element models, have given the atom-microresonator coupling rate can exceed $2\pi \times 700$ MHz in microresonator structure given suitable experimental parameters [17, 18]. In addition to these microresonator-specific techniques, the atom-microresonator coupling can also be enhanced by other physical methods. For instance, both theoretical and experimental evidence suggests that a quantum squeezing method can enhance interactions between quantum systems, even without precise knowledge of the system parameters [19–23]. Additionally, the counting of phonons (magnons) and other types of statistical processing of photons and phonons (magnons) can be measured by applying an auxiliary system to convert the mechanical (magnon) signals [24, 25]. The multipartite entanglement can be characterized by a class of nonlinear squeezing parameters [2, 26]. For this special design, we theoretically predicts that the nonreciprocal entangled photon-phonon pairs and photon-magnon pairs with corresponding antibunching $g^{(2)}_{2,ab} = 0.29$ and $g^{(2)}_{2,am} = 0.49$ could be achieved with the parameters $\omega_b = 2\pi \times 2$ GHz, $\omega_m = 2\pi \times 2.1$ GHz, $\Delta_{\sigma a} = -2\pi \times 6.2$ GHz, $|\Delta_F| = 2\pi \times 50$ MHz, $\lambda_{a\sigma} = 2\pi \times 600$ MHz, $\lambda_{ab} = \lambda_{am} = 2\pi \times 44$ MHz, $\xi = 2\pi \times 1.6$ GHz, $\gamma = 2\pi \times 2$ MHz, and $\kappa = 2\pi \times 16$ MHz [5, 6, 8–18].

* Electronic address: xinyoulu@hust.edu.cn

[1] M. Gessner, L. Pezzè, and A. Smerzi, Phys. Rev. A **94**, 020101(R) (2016).
[2] M.S. Tian, Y. Xiang, F.-X. Sun, M. Fadel, and Q.Y. He, Characterizing Multipartite non-Gaussian Entanglement for a Three-Mode Spontaneous Parametric Down-Conversion Process, Phys. Rev. Applied. **18**, 024065 (2022).
[3] P. Hyllus, O. Gühne, and A. Smerzi, Phys. Rev. A 82, 012337 (2010).
[4] S. L. Braunstein and C. M. Caves, Phys. Rev. Lett. **72**, 3439 (1994).
[5] M. Aspelmeyer, T. J. Kippenberg, and F. Marquardt, Cavity optomechanics, Rev. Mod. Phys. **86**, 1391 (2014).
[6] C.-Z. Chai, Z. Shen, Y.-L. Zhang, H.-Q. Zhao, G.-C. Guo, C.-L. Zou, and C.-H. Dong, Single-sideband microwave-to-optical

conversion in high-Q ferrimagnetic microspheres, Photon. Res. **10**, 820 (2022).
[7] S. Maayani, R. Dahan, Y. Kligerman, E. Moses, A. U. Hassan, H. Jing, F. Nori, D. N. Christodoulides, and T. Carmon, Flying couplers above spinning resonators generate irreversible refraction, Nature **558**, 569 (2018).
[8] X. Zhang, N. Zhu, C.-L. Zou, and H. X. Tang, Optomagnonic Whispering Gallery Microresonators, Phys. Rev. Lett. **117**, 123605 (2016)
[9] T. Aoki, B. Dayan, E. Wilcut, W. P. Bowen, A. S. Parkins, T. J. Kippenberg, K. J. Vahala, and H. J. Kimble, Observation of strong coupling between one atom and a monolithic microresonator, Nature **443**, 671 (2006).
[10] B. Dayan, A. S. Parkins, T. Aoki, E. P. Ostby, K. J. Vahala, and H. J. Kimble, A Photon Turnstile Dynamically Regulated by One Atom, Science **319**, 1062 (2008).
[11] D. J. Alton, N. P. Stern, T. Aoki, H. Lee, E. Ostby, K. J. Vahala, and H. J. Kimble, Strong interactions of single atoms and photons near a dielectric boundary, Nat. Phys. **7**, 159 (2011).
[12] C. Junge, D. O'Shea, J. Volz, and A. Rauschenbeutel, Strong Coupling between Single Atoms and Nontransversal Photons, Phys. Rev. Lett. **110**, 213604 (2013).
[13] D. O'Shea, C. Junge, J. Volz, and A. Rauschenbeutel, Fiber-Optical Switch Controlled by a Single Atom, Phys. Rev. Lett. **111**, 193601 (2013).
[14] I. Shomroni, S. Rosenblum, Y. Lovsky, O. Bechler, G. Guendelman, and B. Dayan, All-optical routing of single photons by a one-atom switch controlled by a single photon, Science **345**, 903 (2014).
[15] E. Will ,L. Masters, A. Rauschenbeutel, M. Scheucher, and J. Volz, Coupling a Single Trapped Atom to a Whispering-Gallery-Mode Microresonator, Phys. Rev. Lett. **126**, 233602 (2021).
[16] J. D. Thompson, T. G. Tiecke, N. P. de Leon, J. Feist, A. V. Akimov, M. Gullans, A. S. Zibrov, V. Vuletić, M. D. Lukin, Coupling a Single Trapped Atom to a Nanoscale Optical Cavity, Science **340**, 1202 (2013).
[17] J. R. Buck and H. J. Kimble, Optimal sizes of dielectric microspheres for cavity QED with strong coupling, Phys. Rev. A **67**, 033806 (2003).
[18] S. M. Spillane, T. J. Kippenberg, K. J. Vahala, K. W. Goh, E. Wilcut, and H. J. Kimble, Ultrahigh-Q toroidal microresonators for cavity quantum electrodynamics, Phys. Rev. A **71** 013817 (2005).
[19] X.-Y. Lü, Y. Wu, J. R. Johansson, H. Jing, J. Zhang, and F. Nori, Squeezed Optomechanics with Phase-Matched Amplification and Dissipation, Phys. Rev. Lett. **114**, 093602 (2015).
[20] W. Qin, A. Miranowicz, P.-B. Li, X.-Y. Lü, J. Q. You, and F. Nori, Exponentially Enhanced Light-Matter Interaction, Cooperativities, and Steady-State Entanglement Using Parametric Amplification, Phys. Rev. Lett. **120**, 093601 (2018).
[21] S. C. Burd, R. Srinivas, H. M. Knaack, W. Ge, A. C. Wilson, D. J. Wineland, D. Leibfried, J. J. Bollinger, D. T. C. Allcock, D. H. Slichter, Quantum amplification of boson-mediated interactions, Nat. Phys. **17**, 898 (2021).
[22] S. C. Burd, H. M. Knaack, R. Srinivas, C. Arenz, A. L. Collopy, L. J. Stephenson, A. C. Wilson, D. J. Wineland, D. Leibfried, J. J. Bollinger, D. T. C. Allcock, and D. H. Slichter, Experimental Speedup of Quantum Dynamics through Squeezing, PRX Quantum **5**, 020314 (2024).
[23] W. Qin and F. Nori, Enhanced Interactions Using Quantum Squeezing, Physics **17**, 64 (2024).
[24] J. D. Cohen, S. M. Meenehan, G. S. MacCabe, S. Groblacher, A. H. Safavi-Naeini, F. Marsili, M. D. Shaw, and O. Painter, Phonon counting and intensity interferometry of a nanomechanical resonator, Nature **520**, 522 (2015).
[25] S. A. Bender, A. Kamra, W. Belzig, and R. A. Duine, Spin Current Cross-Correlations as a Probe of Magnon Coherence, Phys. Rev. Lett. **122**, 187701 (2019).
[26] M. Gessner, A. Smerzi, and L. Pezzè, Metrological Nonlinear Squeezing Parameter, Phys. Rev. Lett. **122**, 090503 (2019).