Supplemental Material for
Exponentially-Enhanced Light-Matter Interaction, Cooperativities, and Steady-State Entanglement Using Parametric Amplification

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In this Supplemental Material to the article on "Exponentially-Enhanced Light-Matter Interaction, Cooperativities, and Steady-State Entanglement Using Parametric Amplification", we first present more details of the elimination of squeezing-induced noises to show an exponential enhancement of the light-matter interaction, as well as of the cooperativity. Then, we derive an effective master equation including an effective Hamiltonian and effective Lindblad operators, and also give a detailed description of our entanglement preparation method. Finally, we discuss, in detail, the effects of counter-rotating terms and show how to remove them.

S1. Elimination of squeezing-induced fluctuation noise

To demonstrate more explicitly the elimination of the squeezing-induced noise, we now derive the Lindblad master equation for our atom-cavity system. In addition to an exponential enhancement of the atom-cavity coupling, the squeezing can introduce undesired noise, including thermal noise and two-photon correlations, into the cavity mode. In order to avoid such noises, our approach employs an auxiliary, high-bandwidth squeezed-vacuum field, which can be experimentally generated, e.g., via optical parametric amplification [S1, S2]. Owing to the bandwidth of the squeezed-vacuum field of up to $\sim$ GHz, the auxiliary field can be thought of as a squeezed-vacuum reservoir for a typical cavity mode with its bandwidth of order of MHz. When being coupled to the cavity mode, the auxiliary field can suppress or even completely eliminate these undesired types of noise of the squeezed-cavity mode.

The Hamiltonian determining the unitary dynamics of our atom-cavity system, as shown in Fig. 1, is given by Eq. (1) and, for convenience, is recalled here

$$H(t) = \sum_k \left[\Delta_e |e\rangle_k\langle e| + \Delta_f |f\rangle_k\langle f|\right] + H_{\rm AC} + H_{\rm NL} + \frac{1}{2}\Omega_{\rm MW}\sum_k \left(|f\rangle_k\langle g| + {\rm H.c.}\right) + V(t), \tag{S1}$$

$$H_{\rm NL} = \Delta_c a^\dagger a + \frac{1}{2}\Omega_p\left[\exp(i\theta_p)\, a^2 + {\rm H.c.}\right], \tag{S2}$$

$$H_{\rm AC} = g\sum_k \left(a|e\rangle_k\langle f| + {\rm H.c.}\right), \tag{S3}$$

$$V(t) = \frac{1}{2}\Omega\exp(i\beta t)\sum_k\left[(-1)^{k-1}|g\rangle_k\langle e| + {\rm H.c.}\right]. \tag{S4}$$

Here $k = 1, 2$ labels the atoms, g is the atom-cavity coupling, the annihilation operator a corresponds to the cavity mode, Ω ($\Omega_{\rm MW}$) is the Rabi frequency of the laser (microwave) drive applied to the atoms, and Ω_p (θ_p) is the amplitude (phase) of the strong pump applied to the nonlinear medium. We have defined the following detunings:

$$\Delta_c = \omega_c - \omega_p/2, \tag{S5}$$

$$\Delta_e = \omega_e - \omega_g - \omega_{\rm MW} - \omega_p/2, \tag{S6}$$

$$\Delta_f = \omega_f - \omega_g - \omega_{\rm MW}, \tag{S7}$$

$$\beta = \omega_L - \omega_{\rm MW} - \omega_p/2, \tag{S8}$$

where ω_c is the cavity frequency, ω_L ($\omega_{\rm MW}$) is the frequency of the laser (microwave) drive applied to the atoms, ω_p is the frequency of the strong pump applied to the nonlinear medium, and ω_z is the frequency associated with level $|z\rangle$ ($z = g, f, e$). When the cavity mode is coupled to the squeezed-vacuum reservoir with a squeezing parameter r_e and a reference phase θ_e, the dynamics of the atom-cavity system is described by the following master equation [S3]:

$$\begin{aligned}\dot{\rho}(t) =& i\left[\rho(t), H(t)\right] - \frac{1}{2}\Bigg\{\sum_{x'}\mathcal{L}(L_{x'})\rho(t) + (N+1)\mathcal{L}(L_a)\rho(t) \\ &+ N\mathcal{L}\left(L_a^\dagger\right)\rho(t) - M\mathcal{L}'(L_a)\rho(t) - M^*\mathcal{L}'\left(L_a^\dagger\right)\rho(t)\Bigg\},\end{aligned} \tag{S9}$$

where $\rho(t)$ is the density operator of the system, a Lindblad operator $L_a = \sqrt{\kappa}a$ describes the cavity decay with a rate κ, and

$$N = \sinh^2(r_e) \quad \text{and} \quad M = \cosh(r_e)\sinh(r_e)e^{-i\theta_e} \tag{S10}$$

describe thermal noise and two-photon correlations caused by the squeezed-vacuum reservoir, respectively. Moreover,

$$\mathcal{L}(o)\rho(t) = o^\dagger o\rho(t) - 2o\rho(t)o^\dagger + \rho(t)o^\dagger o, \tag{S11}$$

$$\mathcal{L}'(o)\rho(t) = oo\rho(t) - 2o\rho(t)o + \rho(t)oo \tag{S12}$$

and the sum runs over all atomic spontaneous emissions, including the Lindblad operators

$$L_{g1} = \sqrt{\gamma_g}|g\rangle_1\langle e|, \quad L_{f1} = \sqrt{\gamma_f}|f\rangle_1\langle e|, \quad L_{g2} = \sqrt{\gamma_g}|g\rangle_2\langle e|, \quad L_{f2} = \sqrt{\gamma_f}|f\rangle_2\langle e|. \tag{S13}$$

Note that, here, we have assumed that the atoms are coupled to a thermal reservoir and that in each atom, $|e\rangle$ decays to $|g\rangle$ and $|f\rangle$, respectively, with rates γ_g and γ_f.

When pumped, the nonlinear medium can squeeze the cavity mode along the axis rotated at an angle $(\pi - \theta_p)/2$, with a squeezing parameter $r_p = (1/4)\ln[(1+\alpha)/(1-\alpha)]$, where $\alpha = \Omega_p/\Delta_c$. This results in a squeezed-cavity mode, as described by the Bogoliubov transformation $a_s = \cosh(r_p)a + \exp(-i\theta_p)\sinh(r_p)a^\dagger$ [S3], such that

$$H_{\rm NL} = \omega_s a_s^\dagger a_s, \tag{S14}$$

where $\omega_s = \Delta_c\sqrt{1-\alpha^2}$ is the squeezed-cavity frequency. In terms of the mode a_s, the atom-cavity interaction Hamiltonian $H_{\rm AC}$ in Eq. (S3) is reexpressed as

$$H_{\rm AC} = \sum_k \left[\left(g_s a_s - g_s' a_s^\dagger\right)|e\rangle_k\langle f| + \text{H.c.}\right], \tag{S15}$$

where $g_s = g\cosh(r_p)$ and $g_s' = \exp(-i\theta_p)g\sinh(r_p)$. Under the assumption that $|g_s'|/(\omega_s + \Delta_e - \Delta_f) \ll 1$, we can make the rotating-wave approximation to neglect the counter-rotating terms, which results in a standard Jaynes-Cummings Hamiltonian

$$H_{\rm ASC} = g_s\sum_k\left(a_s|e\rangle_k\langle f| + \text{H.c.}\right). \tag{S16}$$

This Hamiltonian describes an interaction between the atoms and the squeezed-cavity mode, and demonstrate that as long as $r_p \geq 1$, there is an exponential enhancement in the atom-cavity coupling,

$$\frac{g_s}{g} \sim \frac{1}{2}\exp(r_p). \tag{S17}$$

Furthermore, the master equation in Eq. (S9) can accordingly be reexpressed as

$$\begin{aligned}\dot{\rho}(t) =& i\left[\rho(t), H_s(t)\right] \\ &- \frac{1}{2}\Bigg\{\sum_{x'}\mathcal{L}(L_{x'})\rho(t) + (N_s+1)\mathcal{L}(L_{as})\rho(t) \\ &+ N_s\mathcal{L}\left(L_{as}^\dagger\right)\rho(t) - M_s\mathcal{L}'(L_{as})\rho(t) - M_s^*\mathcal{L}'\left(L_{as}^\dagger\right)\rho(t)\Bigg\},\end{aligned} \tag{S18}$$

$$\begin{aligned}H_s(t) =& \sum_k\left[\Delta_e|e\rangle_k\langle e| + \Delta_f|f\rangle_k\langle f|\right] + \omega_s a_s^\dagger a_s + H_{\rm ASC} \\ &+ \frac{1}{2}\Omega_{\rm MW}\sum_k\left(|f\rangle_k\langle g| + \text{H.c.}\right) + V(t),\end{aligned} \tag{S19}$$

where N_s and M_s are given, respectively, by

$$N_s = \cosh^2(r_p)\sinh^2(r_e) + \sinh^2(r_p)\cosh^2(r_e) + \frac{1}{2}\sinh(2r_p)\sinh(2r_e)\cos(\theta_e+\theta_p), \tag{S20}$$

$$M_s = \exp(i\theta_p)\left[\sinh(r_p)\cosh(r_e) + \exp[-i(\theta_e+\theta_p)]\cosh(r_p)\sinh(r_e)\right] \times \left[\cosh(r_p)\cosh(r_e) + \exp[i(\theta_p+\theta_e)]\sinh(r_e)\sinh(r_p)\right], \tag{S21}$$

corresponding to an effective thermal noise and two-photon correlations of the squeezed-cavity mode, and where $L_{\rm as} = \sqrt{\kappa}a_s$ is a Lindblad operator corresponding to the decay of the squeezed-cavity mode, $g_s = g\cosh(r_p)$ is the enhanced, controllable atom-cavity coupling. We have neglected the counter-rotating terms to obtain the Hamiltonian H_s. From Eqs. (S20) and (S21), we can, as $r_e = 0$, observe the noise caused only by squeezing the cavity mode. However, when choosing $r_e = r_p$ and $\theta_e + \theta_p = \pm n\pi$ $(n = 1, 3, 5, \cdots)$, we have

$$N_s = M_s = 0, \tag{S22}$$

so that the master equation is simplified to a Lindblad form,

$$\dot{\rho}(t) = i\left[\rho(t), H_s(t)\right] - \frac{1}{2}\sum_x \mathcal{L}(L_x)\rho(t). \tag{S23}$$

Here, the sum runs over all dissipative processes, including atomic spontaneous emission and squeezed-cavity decay. From Eq. (S23), we find that the squeezed-cavity mode is equivalently coupled to a thermal reservoir, and the squeezing-induced noises are completely removed as desired. Therefore, we can define the effective cooperativity $C_s = g_s^2/(\kappa\gamma)$, and obtain an exponential enhancement in the atom-cavity cooperativity $C = g^2/(\kappa\gamma)$, that is,

$$\frac{C_s}{C} = \cosh^2(r_p) \sim \frac{1}{4}\exp(2r_p). \tag{S24}$$

This can be used to improve the quality of dissipative entanglement preparation. The resulting entanglement infidelity is no longer lower-bounded by the cooperativity C of the atom-cavity system and could be, in principle, made very close to zero.

Our method is to use a squeezed-vacuum field to suppress the noise of the squeezed-cavity mode, including thermal noise and two-photon correlations. This makes the squeezed-cavity mode equivalently coupled to a thermal-vacuum reservoir. Therefore, this method only changes the environment of the squeezed-cavity mode, and cannot cause the cavity mode to violate the Heisenberg uncertainty principle. To elucidate more explicitly the physics underlying this effect and to obtain an analytical understanding, we consider a simple case when the cavity mode is decoupled from the atoms. In this case, the Hamiltonian only includes the nonlinear term given in Eq. (S2). The cavity mode is then coupled to the squeezed-vacuum reservoir. Following the same method as before, we can find that the squeezed-cavity mode is equivalently coupled to a thermal vacuum reservoir. The corresponding master equation is

$$\dot{\rho}(t) = i\left[\rho(t), \omega_s a_s^\dagger a_s\right] - \frac{\kappa}{2}\left[a_s^\dagger a_s\rho(t) - 2a_s\rho(t)a_s^\dagger + \rho(t)a_s^\dagger a_s\right]. \tag{S25}$$

We now calculate the Heisenberg uncertainty relation of the cavity mode a evolving according to the master equation given in Eq. (S25). To start, we define two rotated quadratures at an angle $(\pi-\theta_p)/2$,

$$X_1 = \frac{1}{2}\left\{a\exp[-i(\pi-\theta_p)/2] + a^\dagger\exp[i(\pi-\theta_p)/2]\right\}, \tag{S26}$$

$$X_2 = \frac{1}{2i}\left\{a\exp[-i(\pi-\theta_p)/2] - a^\dagger\exp[i(\pi-\theta_p)/2]\right\}. \tag{S27}$$

In terms of the a_s mode, X_1 and X_2 can be reexpressed as

$$X_1 = x_1 a_s + x_1^* a_s^\dagger, \tag{S28}$$

$$X_2 = -i\left(x_2 a_s - x_2^* a_s^\dagger\right). \tag{S29}$$

Here,

$$x_1 = \frac{1}{2}\left\{\exp[-i(\pi-\theta_p)/2]\cosh(r_p) - \exp[i(\pi+\theta_p)/2]\sinh(r_p)\right\}, \tag{S30}$$

$$x_2 = \frac{1}{2}\left\{\exp[-i(\pi-\theta_p)/2]\cosh(r_p) + \exp[i(\pi+\theta_p)/2]\sinh(r_p)\right\}. \tag{S31}$$

According to the master equation in Eq. (S25), a straightforward calculation gives

$$
\begin{aligned}
\left(\Delta X_1\right)^2 &= \langle X_1^2\rangle - \langle X_1\rangle^2 \\
&= \Big\{ y_1^2 \exp\left(-i2\omega_s t\right)\left[\langle a_s a_s\rangle(0) - \langle a_s\rangle^2(0)\right] \\
&+ 2|y_1|^2\left[\langle a_s^\dagger a_s\rangle(0) - \langle a_s^\dagger\rangle(0)\,\langle a_s\rangle(0)\right] \\
&+ y_1^{*2}\exp\left(i2\omega_s t\right)\left[\langle a_s^\dagger a_s^\dagger\rangle(0) - \langle a_s^\dagger\rangle^2(0)\right]\Big\}\exp\left(-\kappa t\right) + \frac{1}{4}\exp\left(2r_p\right), \qquad \text{(S32)}\\
\left(\Delta X_2\right)^2 &= \langle X_2^2\rangle - \langle X_2\rangle^2 \\
&= \Big\{ y_2^2 \exp\left(-i2\omega_s t\right)\left[\langle a_s\rangle^2(0) - \langle a_s a_s\rangle(0)\right] \\
&+ 2|y_2|^2\left[\langle a_s^\dagger a_s\rangle(0) - \langle a_s^\dagger\rangle(0)\,\langle a_s\rangle(0)\right] \\
&+ y_2^{*2}\exp\left(i2\omega_s t\right)\left[\langle a_s^\dagger\rangle^2(0)\right] - \langle a_s^\dagger a_s^\dagger\rangle(0)\Big\}\exp\left(-\kappa t\right) + \frac{1}{4}\exp\left(-2r_p\right), \qquad \text{(S33)}
\end{aligned}
$$

where $\langle O\rangle(t)$ represents the expectation value of the operator O at the evolution time t. For simplicity, and without loss of generality, we assume that the squeezed-cavity mode is initially in a Fock state $|n_s\rangle$, with n_s being the squeezed-cavity photon number. In this case, we have

$$
\begin{aligned}
\left(\Delta X_1\right)^2 =& \frac{1}{4}\left[2n_s\exp\left(-\kappa t\right)+1\right]\exp\left(2r_p\right), \qquad \text{(S34)}\\
\left(\Delta X_2\right)^2 =& \frac{1}{4}\left[2n_s\exp\left(-\kappa t\right)+1\right]\exp\left(-2r_p\right), \qquad \text{(S35)}
\end{aligned}
$$

and then

$$
\left(\Delta X_1\right)\left(\Delta X_2\right) = \frac{1}{4}\left[2n_s\exp\left(-\kappa t\right)+1\right] \geq \frac{1}{4}. \qquad \text{(S36)}
$$

It is found, from Eq. (S36), that the Heisenberg uncertainty relation holds, as expected.

We now turn to the discussion of the squeezed vacuum drive. The squeezing strength r_e and squeezing phase θ_e are experimentally adjustable quantities. In optics, the squeezed vacuum can be produced by a pumped $\chi^{(2)}$ nonlinear medium (e.g., a periodically-poled KTiOPO4 (PPKTP) crystal) placed in an optical cavity [S1, S2, S4, S5]. This method is similar to generating cavity-field squeezing of a atom-cavity system. The parameters r_e and θ_e can be controlled by the amplitude and phase of the laser, which pumps the crystal. To confirm the values of the parameters, one can further measure these by using balanced homodyne detection [S6]. The parameters r_p and θ_p can be controlled analogously in such a way to fulfill the conditions $r_e = r_p$ and $\theta_e + \theta_p = \pm n\pi$ $(n = 1, 3, 5, \cdots)$. We note that optical squeezing has also been experimentally implemented utilizing a waveguide cavity [S7].

Superconducting quantum circuits, due to their tunable nonlinearity and low losses for microwave fields, are other promising devices for producing squeezed states. The most popular method to generate microwave squeezing is to use a Josephson parametric amplifier (JPA) [S8–S12]. The JPA is a superconducting LC resonator, which consists of a superconducting quantum interference device (SQUID). This resonator can be pumped not only through the resonator, but also by modulating the magnetic flux in the SQUID. In this case, the parameters r_e and θ_e can be controlled by the amplitude and phase of a pump tone used to modulate the magnetic flux. Recent experiments have shown that the squeezed vacuum, generated by a JPA, can be used to reduce the radiative decay of superconducting qubits [S10] and to modify resonance fluorescence [S13]. The squeezing of quantum noise has also been demonstrated with tunable Josephson metamaterials [S14].

S2. Perturbative treatment and maximizing steady-state entanglement

For the preparation of a steady entangled state, e.g., the singlet state $|S\rangle = \left(|gf\rangle - |fg\rangle\right)/\sqrt{2}$, the key element is that the system dynamics cannot only drive the population into $|\psi_-\rangle$, but also prevent the population from moving out of $|\psi_-\rangle$. In our approach, when we choose $\Delta_e = \beta = \omega_s + \Delta_f$, the coherent couplings mediated by the laser drive and by the squeezed-cavity mode are resonant. In addition, the microwave field also resonantly drives the transition

$$
|\phi_-\rangle \leftrightarrow |\phi_+\rangle \leftrightarrow |\psi_+\rangle. \qquad \text{(S37)}
$$

The proposed entanglement preparation can, therefore, be understood via a hopping-like model, as illustrated in Fig. S1(a). Note that, here, Δ_f is required to be nonzero, or $|\phi_-\rangle$ becomes a dark state of the microwave drive, whose population is trapped and cannot be transferred to $|\psi_+\rangle$. In the preparation process, the populations initially in the states $|\phi_-\rangle$, $|\phi_+\rangle$, and $|\psi_+\rangle$ can be coherently driven to the dark state $|D\rangle$ through the microwave and laser drives and, then, decay to the desired state $|\psi_-\rangle$ through two atomic decays, respectively, with rates γ_{g1} and γ_{g2}. Indeed, such atomic decays originate, respectively, from the spontaneous emissions, $|e\rangle \rightarrow |g\rangle$, of the two atoms, so we have $\gamma_{g1} = \gamma_{g2} = \gamma_g/4$. Furthermore, owing to the laser drive, the state $|\psi_-\rangle$ is resonantly excited to $|\varphi_e\rangle$. This state is then resonantly coupled to $|ff\rangle|1\rangle_s$ by the squeezed-cavity mode. The cavity loss causes the latter state to decay to $|ff\rangle|0\rangle_s$, thus giving rise to population leakage from $|\psi_-\rangle$. However, because of the exponential enhancement in the atom-cavity coupling [i.e., $g_s \sim g\exp(r_p)/2$ in Eq. (S17)], the state $|\varphi_e\rangle$ is split into a doublet of dressed states, $|e_\pm\rangle = (|\varphi_e\rangle \pm |ff\rangle|1\rangle_s)/\sqrt{2}$, exponentially separated by

$$2\sqrt{2}g_s \sim \sqrt{2}g\exp(r_p), \tag{S38}$$

which is much larger than the couplings strength $\Omega_\pm = \Omega/\left(2\sqrt{2}\right)$, as shown in Fig. S1(b). Hence, the population leakage from $|\psi_-\rangle$ is exponentially suppressed, and we can make the effective decay rate, $\Gamma_{\rm out}$, out of $|\psi_-\rangle$, exponentially smaller than the effective decay rate, $\Gamma_{\rm in}$, into $|\psi_-\rangle$. To discuss these decay rates more specifically, we need to give an effective master equation of the system, when the laser drive Ω is assumed to be much smaller than the interactions inside the excited-state subspace. In this case, the coupling between the ground- and excited-state subspaces is treated as a perturbation, so that both cavity mode and excited states of the atoms can be adiabatically eliminated.

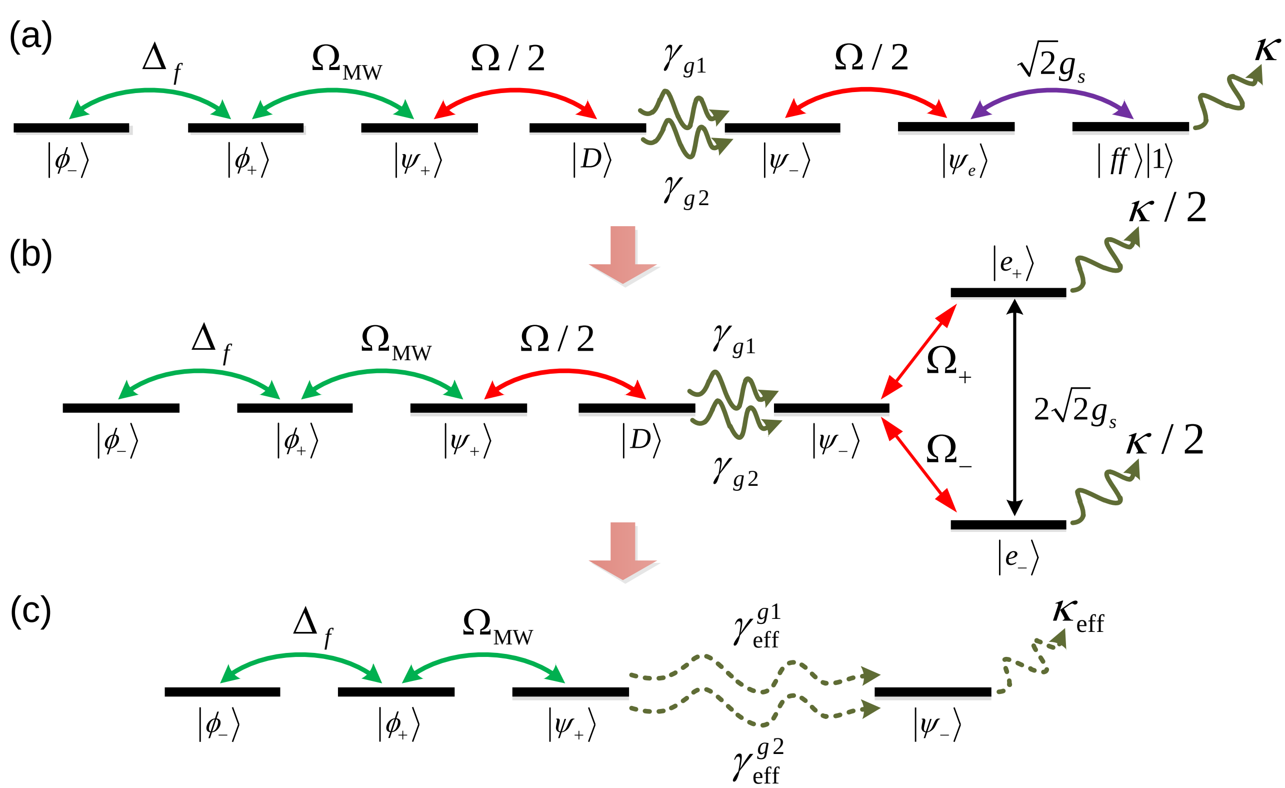

FIG. S1. (Color online) (a) Hopping-like model for the proposed steady-state nearly-maximal entanglement preparation. (b) Exponential suppression in the leakage of the population in $|\psi_-\rangle$. (c) Effective dynamics after adiabatically eliminating the states $|D\rangle$, $|e_+\rangle$, and $|e_-\rangle$.

Specifically, we follow the procedure in Ref. [S15], and begin by considering the Lindblad master equation in Eq. (S23). For convenience, we rewrite the Hamiltonian $H_s(t)$ as

$$H_s(t) = H_g + H_e + v(t) + v^\dagger(t), \tag{S39}$$

with

$$H_g = \sum_{k=1,2} \left[\Delta_f |f\rangle_k \langle f| + \frac{\Omega_{\text{MW}}}{2} \left(|f\rangle_k \langle g| + \text{H.c.} \right) \right], \tag{S40}$$

$$H_e = \sum_{k=1,2} |e\rangle_k \langle e| + \omega_s a_s^\dagger a_s + H_{\text{ASC}}, \tag{S41}$$

representing the interactions, respectively, inside the ground- and excited-state subspaces, and

$$v(t) = \frac{1}{2} \exp\left(i\beta t\right) \Omega \sum_{k=1,2} \exp\left[i\left(k-1\right)\pi\right] |g\rangle_k \langle e| \tag{S42}$$

being the deexcitation from the excited-state subspace to the ground-states subspace. Under the assumption that the laser drive Ω is sufficiently weak compared to the coupling g_s, the effective Hamiltonian and Lindblad operators read:

$$H_{\text{eff}} = -\frac{1}{2} \left[v(t) \left(H_{\text{NH}} - \beta \right)^{-1} v^\dagger(t) \right] + H_g, \tag{S43}$$

$$L_{x,\text{eff}} = L_x \left(H_{\text{NH}} - \beta \right)^{-1} v^\dagger(t), \tag{S44}$$

where

$$H_{\text{NH}} = H_e - \frac{i}{2} \sum_x L_x^\dagger L_x \tag{S45}$$

is the no-jump Hamiltonian. The system dynamics is, therefore, determined by an effective master equation

$$\dot{\rho}_g(t) = i\left[\rho_g(t), H_{\text{eff}}\right] - \frac{1}{2} \sum_x \mathcal{L}\left(L_{x,\text{eff}}\right) \rho_g(t), \tag{S46}$$

where $\rho_g(t)$ is the reduced density operator associated only with the ground states of the atoms. After a straightforward calculation restricted in the Hilbert space having at most one excitation, we have:

$$H_{\text{eff}} = \Delta_f \left(\mathcal{I}/2 - |\phi_+\rangle\langle\phi_-| + \text{H.c.} \right) + \Omega_{\text{MW}} \left(|\psi_+\rangle\langle\phi_+| + \text{H.c.} \right), \tag{S47}$$

$$L_{g1,\text{eff}} = r_g \left[\left(|\psi_+\rangle + |\psi_-\rangle \right) \left(\gamma_{\text{eff},0} \langle\psi_+| + \gamma_{\text{eff},2} \langle\psi_-| \right) + \gamma_{\text{eff},1} \left(|\phi_+\rangle + |\phi_-\rangle \right) \left(\langle\phi_+ + \langle\phi_-| \right) \right], \tag{S48}$$

$$L_{g2,\text{eff}} = -r_g \left[\left(|\psi_+\rangle - |\psi_-\rangle \right) \left(\gamma_{\text{eff},0} \langle\psi_+| - \gamma_{\text{eff},2} \langle\psi_-| \right) + \gamma_{\text{eff},1} \left(|\phi_+\rangle + |\phi_-\rangle \right) \left(\langle\phi_+ + \langle\phi_-| \right) \right], \tag{S49}$$

$$L_{f1,\text{eff}} = r_f \left[\left(|\phi_+\rangle - |\phi_-\rangle \right) \left(\gamma_{\text{eff},0} \langle\psi_+| + \gamma_{\text{eff},2} \langle\psi_-| \right) + \gamma_{\text{eff},1} \left(|\psi_+\rangle - |\psi_-\rangle \right) \left(\langle\phi_+| + \langle\phi_-| \right) \right], \tag{S50}$$

$$L_{f2,\text{eff}} = -r_f \left[\left(|\phi_+\rangle - |\phi_-\rangle \right) \left(\gamma_{\text{eff},0} \langle\psi_+| - \gamma_{\text{eff},2} \langle\psi_-| \right) + \gamma_{\text{eff},1} \left(|\psi_+\rangle + |\psi_-\rangle \right) \left(\langle\phi_+| + \langle\phi_-| \right) \right], \tag{S51}$$

$$L_{\text{as,eff}} = r_{\text{as}} \left[\kappa_{\text{eff},1} |\psi_-\rangle \left(\langle\phi_+| + \langle\phi_-| \right) - \frac{1}{\sqrt{2}} \kappa_{\text{eff},2} \left(|\phi_+\rangle - |\phi_-\rangle \right) \langle\psi_-| \right]. \tag{S52}$$

Here,

$$\mathcal{I} = |\phi_+\rangle\langle\phi_+| + |\phi_-\rangle\langle\phi_-| + |\psi_+\rangle\langle\psi_+| + |\psi_-\rangle\langle\psi_-|, \tag{S53}$$

$$|\phi_\pm\rangle = \frac{1}{\sqrt{2}} \left(|gg\rangle \pm |ff\rangle \right), \tag{S54}$$

$$|\psi_\pm\rangle = \frac{1}{\sqrt{2}} \left(|gf\rangle \pm |fg\rangle \right), \tag{S55}$$

and

$$r_{g(f)} = \exp\left(-i\beta t\right) \frac{\Omega\sqrt{\gamma_{g(f)}}}{4\gamma}, \tag{S56}$$

$$r_{\text{as}} = \exp\left(-i\beta t\right) \frac{\Omega}{2\sqrt{\gamma}}, \tag{S57}$$

$$\gamma_{\text{eff},0} = \frac{1}{\widetilde{\Delta}_{e,1}}, \tag{S58}$$

$$\gamma_{\text{eff},m} = \frac{\widetilde{\omega}_{s,m}}{\widetilde{\omega}_{s,m} \widetilde{\Delta}_{e,m-1} - mC_s}, \tag{S59}$$

$$\kappa_{\text{eff},m} = \frac{\sqrt{mC_s}}{\widetilde{\omega}_{s,m} \widetilde{\Delta}_{e,m-1} - mC_s}, \tag{S60}$$

where

$$\widetilde{\omega}_{s,m} = \frac{1}{\kappa}\left(\omega_s + m\Delta_f - \beta\right) - \frac{i}{2}, \tag{S61}$$

$$\widetilde{\Delta}_{e,m-1} = \frac{1}{\gamma}\left[\Delta_e + (m-1)\,\Delta_f - \beta\right] - \frac{i}{2}, \tag{S62}$$

for $m = 1, 2$, and where $\gamma = \gamma_g + \gamma_f$ is the total atomic decay rate.

Having obtained the effective master equation, let us now consider the decay rates $\Gamma_{\rm in}$ and $\Gamma_{\rm out}$. According to the effective Lindblad operators in Eqs. (S48)-(S52), the decay rates of moving into and out of the singlet state $|\psi_-\rangle$ are given, respectively, by

$$\Gamma_{\rm in} = \frac{\Omega^2}{4\gamma^2}\left(\gamma_g|\gamma_{\rm eff,0}|^2 + 2\gamma_f|\gamma_{\rm eff,1}|^2 + 4\gamma|\kappa_{\rm eff,1}|^2\right), \tag{S63}$$

$$\Gamma_{\rm out} = \frac{\Omega^2}{4\gamma^2}\left(\gamma_g|\gamma_{\rm eff,2}|^2 + 2\gamma_f|\gamma_{\rm eff,2}|^2 + 2\gamma|\kappa_{\rm eff,2}|^2\right). \tag{S64}$$

Let us define the entanglement fidelity as $F = \langle\psi_-|\rho_g\,(t)\,|\psi_-\rangle$ (that is, the probability of the atoms being in $|\psi_-\rangle$) and, then, the entanglement infidelity as $\delta = 1 - F$. In the steady state ($t \to +\infty$), the entanglement infidelity is found

$$\delta \sim \frac{1}{1 + \Gamma_{\rm in}/\left(3\Gamma_{\rm out}\right)}. \tag{S65}$$

Note that, here, we have assumed that $|\phi_+\rangle$, $|\phi_-\rangle$, and $|\psi_+\rangle$ have the same population in a steady state. In order to prepare nearly-maximal steady-state entanglement, we choose the detunings to be

$$\Delta_e = \beta = \omega_s + \Delta_f, \tag{S66}$$

such that $\widetilde{\omega}_{s,m} \sim \widetilde{\Delta}_{e,m-1} \sim -i/2$, yielding

$$\frac{\Gamma_{\rm in}}{\Gamma_{\rm out}} \sim \frac{4\gamma_g}{\gamma}C_s \gg 1, \tag{S67}$$

for $C_s \gg 1$. As shown in Fig. S1(c), the underlying dynamics is as follows: after adiabatically eliminating the excited states $|D\rangle$, $|e_+\rangle$, and $|e_-\rangle$, the states $|\psi_+\rangle$ and $|\psi_-\rangle$ are directly connected by two effective spontaneous emission processes with rates $\gamma_{\rm eff}^{g1}$ and $\gamma_{\rm eff}^{g2}$,

$$\gamma_{\rm eff}^{g1} = \gamma_{\rm eff}^{g2} = |r_g\gamma_{\rm eff,0}|^2 \sim \frac{\gamma_g}{4\gamma^2}\Omega^2, \tag{S68}$$

and at the same time, the desired state $|\psi_-\rangle$ leaks the population through an effective cavity decay with a rate $\kappa_{\rm eff}$,

$$\kappa_{\rm eff} = |r_{\rm as}\kappa_{\rm eff,2}|^2/2 \sim \frac{\Omega^2}{16\gamma C_s}. \tag{S69}$$

Therefore, together with the effective Hamiltonian $H_{\rm eff}$ driving the populations from both $|\phi_+\rangle$ and $|\phi_-\rangle$ to $|\psi_+\rangle$, the initial populations in the ground-states subspace of the atoms can be transferred to $|\psi_-\rangle$ and trapped in this state. By substituting Eq. (S67) into Eq. (S65), we can straightforwardly have

$$\delta \sim \frac{3\gamma}{4\gamma_g C_s}. \tag{S70}$$

As long as $r_p \geq 1$, an exponential enhancement of the cooperativity, $C_s/C \sim \exp\left(2r_p\right)/4$, is obtained, leading to

$$\delta \sim \frac{3\gamma}{\gamma_g \exp\left(2r_p\right) C}. \tag{S71}$$

This equation shows that we can increase the squeezing parameter r_p, so as to exponentially decrease the entanglement infidelity, as seen in Fig. S2. Moreover, the result in this figure also reveals that, by decreasing Ω, one can suppress non-adiabatic errors and, thus, can cause the steady-state infidelity to approach a theoretical value, as expected. Hence, as opposed to prior entanglement preparation protocols, which relied on controlled unitary dynamics or engineered dissipation, such an infidelity is no longer lower bounded by the cooperativity C and, in principle, can be made very close to zero.

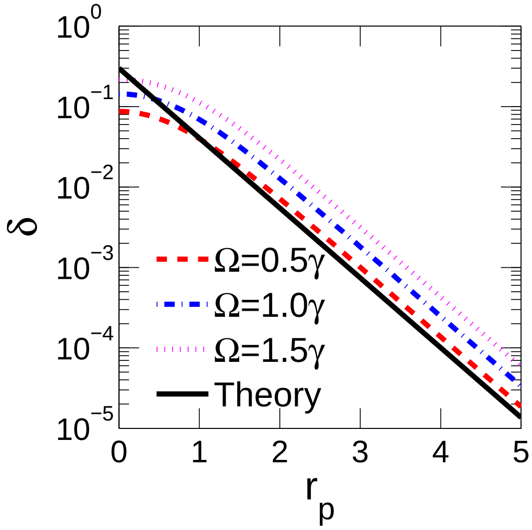

FIG. S2. (Color online) Steady-state entanglement infidelity versus the squeezing parameter r_p. We have plotted the numerical infidelity for $\Omega = 0.5\gamma$ (dashed curve), $\Omega = 1.0\gamma$ (dashed-dotted curve), and $\Omega = 1.5\gamma$ (dotted curve) by calculating the effective master equation, and also plotted the theoretical prediction (solid curve). Here, we have assumed that $\gamma_g = \gamma/2$, $\kappa = 2\gamma/3$, $C = 20$, $\Delta_f = \Omega/2^{7/4}$, $\Omega_{\rm MW} = \sqrt{2}\Delta_f$, and that with the vacuum cavity, the initial state of the atoms is $\left(\mathcal{I} - |\psi_-\rangle\langle\psi_-|\right)/3$.

S3. Effects of the counter-rotating terms

The counter-rotating terms of the form $a_s^\dagger \sum_k |e\rangle_k\langle f|$ and $a_s \sum_k |f\rangle_k\langle e|$, which result from optical parametric amplification, do not conserve the excitation number, and can couple the ground- and double-excited states subspaces. Thus, this would give rise to an additional leakage of the population in the desired state $|\psi_-\rangle$, and decrease the entanglement fidelity. For example, in the presence of the counter-rotating terms, the state $|\psi_-\rangle$ can be excited to a double-excitation state $\left(|ge\rangle - |eg\rangle\right)|1\rangle_s/\sqrt{2}$, which, then, de-excites to the ground state $|gg\rangle|0\rangle$ through cavity decay and spontaneous emission. In general, we can decrease the ratio $|g_s'|/\left(2\Delta_e\right)$ to reduce errors induced by these excitation-number-nonconserving processes. However, to reduce such errors more efficiently in the limit of $|g_s'|/\left(2\Delta_e\right)$, we analyze effects of counter-rotating terms, in detail, in this section, and demonstrate that by modifying external parameters, we can remove these terms and the full system can be mapped to a simplified system described above.

According to Eqs. (S14) and (S15), the full Hamiltonian of the system in the terms of the squeezed mode a_s is

$$
\begin{aligned}
H(t) = &\sum_k \left[\Delta_e|e\rangle_k\langle e| + \Delta_f|f\rangle_k\langle f|\right] + \omega_s a_s^\dagger a_s \\
&+ \sum_k \left[\left(g_s a_s - g_s' a_s^\dagger\right)|e\rangle_k\langle f| + \text{H.c.}\right], \\
&+ \frac{1}{2}\Omega_{\rm MW}\sum_k \left(|f\rangle_k\langle g| + \text{H.c.}\right) + V(t), \qquad \text{(S72)}
\end{aligned}
$$

$$
V(t) = \frac{1}{2}\Omega\exp(i\beta t)\sum_k \left[(-1)^{k-1}|g\rangle_k\langle e| + \text{H.c.}\right]. \qquad \text{(S73)}
$$

Indeed, the counter-rotating terms can be treated as the high-frequency components of the full Hamiltonian. In order to explicitly show these high-frequency components, we can express $H(t)$ into a rotating frame at

$$
H_0 = \Delta_e \sum_k |e\rangle_k\langle e| + \left(\omega_s + \Delta_f\right) a_s^\dagger a_s. \qquad \text{(S74)}
$$

Thus, $H\left(t\right)$ is transformed to

$$\begin{aligned}
\mathcal{H}\left(t\right) = \Delta_f \left(\sum_k |f\rangle_k\langle f| - a_s^\dagger a_s\right) \\
&+ \sum_k \left(g_s a_s |e\rangle_k\langle f| - e^{i2\Delta_e t} g_s' a_s^\dagger |e\rangle_k\langle f| + \text{H.c.}\right) \\
&+ \frac{1}{2}\Omega_{\text{MW}} \sum_k \left(|f\rangle_k\langle g| + \text{H.c.}\right) + \mathcal{V}, \qquad \text{(S75)}\\
\mathcal{V} = \frac{1}{2}\Omega \sum_k \left[(-1)^{k-1} |g\rangle_k\langle e| + \text{H.c.}\right]. \qquad \text{(S76)}
\end{aligned}$$

Here, we have chosen $\Delta_e = \beta = \omega_s + \Delta_f$. Because Δ_f is required to be much smaller than Δ_e, $\mathcal{H}\left(t\right)$ can be divided into two parts, $\mathcal{H}\left(t\right) = H_{\text{low}} + H_{\text{high}}$, where

$$\begin{aligned}
H_{\text{low}} &= \Delta_f \left(\sum_k |f\rangle_k\langle f| - a_s^\dagger a_s\right) + g_s \sum_k \left(a_s |e\rangle_k\langle f| + \text{H.c.}\right) \\
&+ \frac{1}{2}\Omega_{\text{MW}} \sum_k \left(|f\rangle_k\langle g| + \text{H.c.}\right) + \mathcal{V}, \qquad \text{(S77)}\\
H_{\text{high}} &= \sum_k \left(-e^{i2\Delta_e t} g_s' a_s^\dagger |e\rangle_k\langle f| + \text{H.c.}\right), \qquad \text{(S78)}
\end{aligned}$$

represent the low- and high- frequency components, respectively. Here, we consider the limit $|g_s'|/\Delta_e \ll 1$. By using a time-averaging treatment [S16], the behavior of H_{high} can be approximated by a time-averaged Hamiltonian,

$$\begin{aligned}
H_{\text{TA}} &= \frac{|g_s'|^2}{2\Delta_e} \sum_k a_s^\dagger a_s \left(|e\rangle_k\langle e| - |f\rangle_k\langle f|\right) \\
&- \frac{|g_s'|^2}{2\Delta_e} \sum_{k,k'} \left(|f\rangle_k\langle e|\right)\left(|e\rangle_{k'}\langle f|\right). \qquad \text{(S79)}
\end{aligned}$$

The first term describes an energy shift depending on the photon number of the squeezed-cavity mode, and the second term describes a direct coupling between the two atoms. Accordingly, $\mathcal{H}\left(t\right)$ becomes $\mathcal{H}\left(t\right) \simeq H_{\text{low}} + H_{\text{TA}}$, and after transforming back to the original frame, we obtain

$$\begin{aligned}
H\left(t\right) \simeq & \sum_k \left[\Delta_e |e\rangle_k\langle e| + \Delta_f |f\rangle_k\langle f|\right] + \omega_s a_s^\dagger a_s \\
&+ g_s \sum_k \left(a_s |e\rangle_k\langle f| + \text{H.c.}\right), \\
&+ \frac{1}{2}\Omega_{\text{MW}} \sum_k \left(|f\rangle_k\langle g| + \text{H.c.}\right) + V\left(t\right) + H_{\text{TA}}. \qquad \text{(S80)}
\end{aligned}$$

We find, from Eq. (S79), that the counter-rotating terms are able to conserve the excitation number as long as $|g_s'|/\Delta_e \ll 1$. Therefore, we can restrict our discussion in a subspace having at most one excitation, as discussed above. In this subspace, H_{TA} is expanded as

$$\begin{aligned}
H_{\text{TA}} &= -\frac{|g_s'|^2}{2\Delta_e} \left(\mathcal{I}/2 + |\varphi_e\rangle\langle\varphi_e| - |\phi_+\rangle\langle\phi_-| + \text{H.c.}\right) \\
&- \frac{|g_s'|^2}{\Delta_e} \left(\mathcal{I}^{(1)}/2 - |\phi_+^{(1)}\rangle\langle\phi_-^{(1)}| + \text{H.c.}\right), \qquad \text{(S81)}
\end{aligned}$$

where

$$\begin{aligned}
\mathcal{I}^{(1)} &= |\phi_+^{(1)}\rangle\langle\phi_+^{(1)}| + |\phi_-^{(1)}\rangle\langle\phi_-^{(1)}| + |\psi_+^{(1)}\rangle\langle\psi_+^{(1)}| + |\psi_-^{(1)}\rangle\langle\psi_-^{(1)}|, \\
|\phi_\pm^{(1)}\rangle &= \left(|gg\rangle \pm |ff\rangle\right)|1\rangle_s/\sqrt{2}, \\
|\psi_\pm^{(1)}\rangle &= \left(|gf\rangle \pm |fg\rangle\right)|1\rangle_s/\sqrt{2}. \qquad \text{(S82)}
\end{aligned}$$

Equation (S81) indicates that the counter-rotating terms introduce an energy shift of $|g_s'|^2/(2\Delta_e)$ imposed upon the ground states, and a coherent coupling, of strength $|g_s'|^2/(2\Delta_e)$, between the states $|\phi_+\rangle$ and $|\phi_-\rangle$. From Fig. S1(a), we find that in the regime, where $\Omega/|g_s'|$ is comparable to $|g_s'|/\Delta_e$, such an energy shift can cause the $|\psi_+\rangle \to |D\rangle$ transition to become far off-resonant and, thus, suppress the population into the desired state $|\psi_-\rangle$. Meanwhile, this introduced coupling may increase the entanglement error originating from the microwave dressing of the ground states. For example, if $\Delta_f = |g_s'|^2/(2\Delta_e)$, then the state $|\phi_-\rangle$ becomes a dark state of the microwave drive. In this case, the population in $|\phi_-\rangle$ is trapped and cannot be transferred to $|\psi_-\rangle$. To remove these detrimental effects, it is essential to compensate this energy shift. According to the above analysis, the detunings in Eq. (S66) need to be modified as

$$\Delta_e = \beta - \frac{|g_s'|^2}{2\Delta_e} = \omega_s + \Delta_f - \frac{|g_s'|^2}{\Delta_e}. \tag{S83}$$

This modification simplifies the full dynamics to the same hopping-like model, as shown in Fig. S1(a) with $\Delta_f \to \Delta_f' = \Delta_f - |g_s|^2/(2\Delta_e)$. Therefore, we can map the full system to a simple system that excludes the counter-rotating terms and has been discussed above.

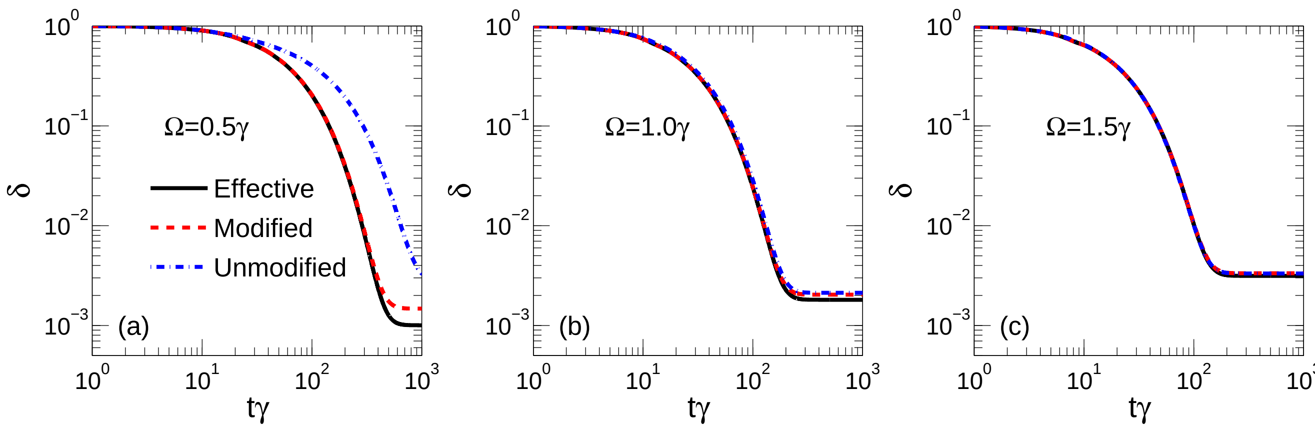

FIG. S3. (Color online) Entanglement infidelity δ as a function of time $t\gamma$ for (a) $\Omega = 0.5\gamma$, (b) $\Omega = 1.0\gamma$, and (c) $\Omega = 1.5\gamma$, assuming a cooperativity of $C = 20$. Solid and dashed-dotted curves are obtained, respectively, from integrations of the effective and full master equations, both with detunings $\Delta_f = \Omega/2^{7/4}$ and $\Delta_e = \beta = \omega_s + \Delta_f$. Dashed curves are also given by calculating the full master equation but with modified detunings $\Delta_f = \Omega/2^{7/4} + |g_s'|^2/(2\Delta_e)$ and $\Delta_e = \beta - |g_s'|^2/(2\Delta_e) = \omega_s + \Delta_f - |g_s'|^2/\Delta_e$. For both full cases, we have assumed $\Delta_e = 200 g_s'$. In all plots, we have assumed that $\gamma_g = \gamma/2$, $\kappa = 2\gamma/3$, $\Omega_{\rm MW} = \sqrt{2}\Delta_f$, $r_p = 3$, and $\theta_p = \pi$. Moreover, the initial state of the atoms is $(\mathcal{I} - |\psi_-\rangle\langle\psi_-|)/3$ and the cavity was initially in the vacuum.

To understand this process better, we can follow the same method as above, but now with the Hamiltonian in Eq. (S80). Thus, we find the effective Hamiltonian and Lindblad operators as follows:

$$H_{\rm eff}' = \Delta_f'(\mathcal{I}/2 - |\phi_+\rangle\langle\phi_-| + {\rm H.c.}) + \Omega_{\rm MW}(|\psi_+\rangle\langle\phi_+| + {\rm H.c.}), \tag{S84}$$

$$L_{g1,\rm eff}' = r_g'\left[(|\psi_+\rangle + |\psi_-\rangle)\left(\gamma_{\rm eff,0}'\langle\psi_+| + \gamma_{\rm eff,2}'\langle\psi_-|\right) + \gamma_{\rm eff,1}'(|\phi_+\rangle + |\phi_-\rangle)(\langle\phi_+ + \langle\phi_-|)\right], \tag{S85}$$

$$L_{g2,\rm eff}' = -r_g'\left[(|\psi_+\rangle - |\psi_-\rangle)\left(\gamma_{\rm eff,0}'\langle\psi_+| - \gamma_{\rm eff,2}'\langle\psi_-|\right) + \gamma_{\rm eff,1}'(|\phi_+\rangle + |\phi_-\rangle)(\langle\phi_+ + \langle\phi_-|)\right], \tag{S86}$$

$$L_{f1,\rm eff}' = r_f'\left[(|\phi_+\rangle - |\phi_-\rangle)\left(\gamma_{\rm eff,0}'\langle\psi_+| + \gamma_{\rm eff,2}'\langle\psi_-|\right) + \gamma_{\rm eff,1}'(|\psi_+\rangle - |\psi_-\rangle)(\langle\phi_+| + \langle\phi_-|)\right], \tag{S87}$$

$$L_{f2,\rm eff}' = -r_f'\left[(|\phi_+\rangle - |\phi_-\rangle)\left(\gamma_{\rm eff,0}'\langle\psi_+| - \gamma_{\rm eff,2}'\langle\psi_-|\right) + \gamma_{\rm eff,1}'(|\psi_+\rangle + |\psi_-\rangle)(\langle\phi_+| + \langle\phi_-|)\right], \tag{S88}$$

$$L_{\rm as,eff}' = r_{\rm as}'\left[\kappa_{\rm eff,1}'|\psi_-\rangle(\langle\phi_+| + \langle\phi_-|) - \frac{1}{\sqrt{2}}\kappa_{\rm eff,2}'(|\phi_+\rangle - |\phi_-\rangle)\langle\psi_-|\right]. \tag{S89}$$

Here,

$$\Delta_f' = \Delta_f - \frac{|g_s|^2}{2\Delta_e}, \tag{S90}$$

$$r_{g(f)}' = \exp(-i\beta t)\frac{\Omega\sqrt{\gamma_{g(f)}}}{4\gamma}, \tag{S91}$$

$$r_{\rm as}' = \exp(-i\beta t)\frac{\Omega}{2\sqrt{\gamma}}, \tag{S92}$$

and

$$\gamma'_{\text{eff},0} = \frac{1}{\widetilde{\Delta}'_e}, \tag{S93}$$

$$\gamma'_{\text{eff},m} = \frac{\widetilde{\omega}'_{s,m}}{\widetilde{\omega}'_{s,m}\widetilde{\Delta}'_{e,m-1} - mC_s}, \tag{S94}$$

$$\kappa'_{\text{eff},m} = \frac{\sqrt{mC_s}}{\widetilde{\omega}'_{s,m}\widetilde{\Delta}'_{e,m-1} - mC_s} \tag{S95}$$

where

$$\widetilde{\Delta}'_e = (\Delta_e + \Delta_f - \beta)/\gamma - i/2, \tag{S96}$$

$$\widetilde{\omega}'_{s,m} = \left[\omega_s + m\left(\Delta_f - \frac{|g'_s|^2}{\Delta_e}\right) - \beta\right]/\kappa - i/2, \tag{S97}$$

$$\widetilde{\Delta}'_{e,m-1} = \left[\Delta_e - \beta + (m-1)\left(\Delta_f - \frac{|g'_s|^2}{\Delta_e}\right)\right]/\gamma - i/2, \tag{S98}$$

for $m = 1, 2$. Upon using the modified parameter, given in Eq. (S83), we obtain $\widetilde{\Delta}'_e \sim \widetilde{\omega}'_{s,m} \sim \widetilde{\Delta}'_{e,m-1} \sim -i/2$. This implies that the dynamics is the same as what we have already described for the simplified system without the counter-rotating terms, thereby leading to the same entanglement infidelity. To confirm this, we perform numerical calculations, as shown in Fig. S3. Specifically, we plot the entanglement infidelity as a function of rescaled time. Solid curves indicate the results obtained by integrating the effective master equation, whereas dashed and dashed-dotted curves reveal the predictions of the full master equation, respectively, with modified and unmodified detunings. These results demonstrate that the detrimental effects of the counter-rotating terms can be strongly suppressed by modifying external parameters, in particular, as what we have discussed above, for the case of weak Ω driving strengths, which are necessary for the validity of the perturbative treatment used in our approach.

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