Dissipation and detection of polaritons in the ultrastrong-coupling regime

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We have investigated theoretically a dissipative polariton system in the ultrastrong light-matter coupling regime without using the rotating-wave approximation on system-reservoir coupling. Photons in a cavity and excitations in matter respectively couple two large ensembles of harmonic oscillators (photonic and excitonic reservoirs). Inheriting the quantum statistics of polaritons in the ultrastrong coupling regime, in the ground state of the whole system, the two reservoirs are not in the vacuum states but they are squeezed and correlated. We presume the reservoirs to be in this nonvacuum state in the master equation and in the input-output formalism with Langevin equations. Both approaches consistently guarantee the decay of the polariton system to its ground state; no photon detection is also obtained when the polariton system is in the ground state.

I. INTRODUCTION

Light-matter ultrastrong coupling [1–28] means that the coupling strength g is comparable to or larger than the transition frequency ω_x of excitations in matter ($g \gtrsim \omega_x$), and it shows a variety of peculiar properties, such as virtual photons in the ground state [1,5], squeezed eigenstates [1,19], nearly degenerate ground states [11,17,18], quantum phase transitions [12,29], and so on. The ultrastrong coupling has been realized experimentally by intersubband transitions in semiconductor quantum wells [6–9,26,27], artificial atoms in superconducting circuits [14–16], and cyclotron transition in two-dimensional electron gas [25]. In most cases, photons with THz or microwave frequency are confined in a cavity, and the cavity photons are coupled with external photonic field (outside the cavity) by an unignorable dissipation rate (but small compared to the light-matter coupling). In other words, when we neglect the coupling with matter, the real eigenmodes of photons are represented as coupled fields of cavity mode and external fields [30]. As we discuss in the present paper by the Fano-type diagonalization technique [31–33], when the cavity mode is squeezed in the ground state due to the ultrastrong light-matter coupling, the external photonic field is also squeezed in the ground state of the whole system. However, we cannot observe the squeezing or the energy flow by photon detectors if the system is in the ground state. In this paper, we have developed two frameworks, the master equation and Langevin equations with input-output relation. They are derived by assuming squeezed external fields and they both consistently show no photon detection.

In the standard theory of quantum optics [2,24,34–42], in order to introduce a dissipation of cavity mode or of excitations in matters, we consider coupling with an ensemble of harmonic oscillators with continuous frequencies, and the oscillators are presumed to be in the vacuum state. The dissipation of the relevant system has been successfully described by such a treatment in both the master equation and the input-output formalism, at least in the weak and (normally) strong light-matter coupling regimes ($g \lesssim \Gamma$ and

$\Gamma \lesssim g \ll \omega_x$, respectively, for dissipation rate Γ). As pointed out in some papers [34,39–43], the master equation should be derived by considering the eigenstates of the relevant system, and the rotating-wave approximation (RWA) should be performed carefully on the system-reservoir coupling even if the system-reservoir coupling is weak compared to the light-matter coupling (in the strong light-matter coupling regime). In the ultrastrong coupling regime, such treatment has been performed by Beaudoin, Gambetta, and Blais [24]. The dissipation of the ultrastrong coupling systems can be successfully described under the RWA on system-reservoir coupling (both pretrace and post-trace RWAs are used in terms of Ref. [41]) by considering the eigenstates of the cavity system (there are squeezed virtual photons in the ground state). Furthermore, the photon detection has also been discussed in Ref. [28], and virtual photons in the cavity are not counted by normal and time ordering the operators of cavity system. Under the pretrace RWA on system-reservoir coupling, counter-rotating terms (lowering or raising both the relevant system and the reservoir) are neglected. If the dissipation is weak enough, such a treatment is appropriate and simple for discussing the dynamics of the relevant system. However, even under the weak dissipation, it is not clear whether the RWA is valid for discussing the statistics of output photons emitted from the cavity, because the output statistics are strongly affected by the modification of the system-reservoir coupling. There are actually virtual photons not only in the cavity but also in the photonic reservoir even if the whole system is in the ground state, which is naturally derived by the Fano-type diagonalization, although the virtual photons are not counted by detectors. Of course, when the cavity system is excited, we can detect photons emitted from the cavity. Whereas antibunching of emission can survive even under the RWA on system-reservoir coupling [28], the quantum fluctuation (squeezing) of the emission is easily diminished in such treatment, because the interference between the reservoir free field and the cavity contribution is important for squeezing [35,36,38]. Therefore, in order to fully discuss the quantum statistics of emission in the ultrastrong light-matter coupling regime, we have to develop a comprehensive framework describing the dissipation and emission without using the RWA on light-matter coupling or on system-reservoir coupling.

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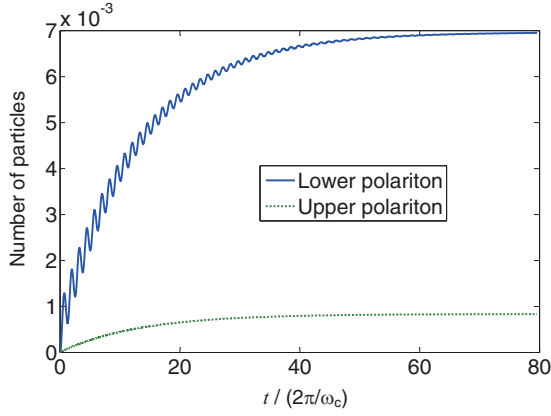*bamba@acty.phys.sci.osaka-u.ac.jp

FIG. 1. (Color online) Numbers of lower and upper polaritons are plotted as a function of dimensionless time $t/(2\pi/\omega_c)$. The initial state is given as $\hat{\rho}(0) = |\text{g.s.}\rangle\langle\text{g.s.}|$, and the reservoirs are presumed to be in the vacuum state. The time development is calculated by master equation (34), correlation (40), and memory kernel (41). Parameters: $\omega_x = \omega_c$, $g = \omega_c$, $D = g^2/\omega_x$, $\Gamma_c = \Gamma_x = 10^{-2}\omega_c$, and $\Omega_c^{\text{cutoff}} = \Omega_x^{\text{cutoff}} = 10^3\omega_c$.

In the weak and normally strong light-matter coupling regimes, the ground state of the whole system is approximately represented by the vacuum states of photonic and excitonic reservoirs (exactly the vacuum states under the RWA on light-matter coupling). In both the master equation and the input-output formalism, the vacuum reservoirs are usually considered for describing the dissipation, and no photon detection is naturally obtained in the ground state. Then, these three approaches (analysis of ground state, master equation, and input-output formalism) are consistent in the standard dissipation theory in the weak and strong light-matter coupling regimes. However, if we simply presume the reservoirs to be in the vacuum state, in the ultrastrong light-matter coupling regime, the master equation and input-output formalism give different results. As we demonstrate in Fig. 1, the relevant system is excited by the vacuum reservoirs, and real photons are emitted from the system to the photonic reservoir according to the master-equation formalism. However, as discussed in Ref. [2], the vacuum output is obtained by the vacuum input according to the input-output formalism. This is because the photonic and excitonic reservoirs are not in the vacuum states in the ground state of the whole system, but they are actually squeezed and correlated. We have to presume the reservoirs to be in the nonvacuum state in order to remove the discrepancy between the results of master equation and of input-output formalism.

In the present paper, we discuss a polariton system consisting of two bosonic modes, photons in a cavity and excitations in matter, each of which couples with an ensemble of harmonic oscillators (photonic and excitonic reservoirs). Diagonalizing the whole system by the Fano-type technique [31–33], we find the squeezed and correlated reservoirs in the ground state. By supposing this reservoir state, the master equation certainly guarantees the decay of the polariton system to its original ground state in the closed case. We also check that, when the polariton system is in the ground state, the virtual photons in the photonic reservoir are not counted by normal and time

ordering the operators in a polariton base. In the input-output formalism, we also obtain no photon detection by supposing the squeezed and correlated reservoirs and by normal- and time-ordering the operators. Then we achieve the consistency of the three approaches (diagonalization, master equation, and input-output formalism) even in the ultrastrong light-matter coupling regime.

This paper is organized as follows. The Hamiltonian is shown in Sec. II, and basic features of the ultrastrong coupling regime are also discussed. The Fano-type diagonalization of the photonic part is performed in Appendix A . In Sec. III, we diagonalize the whole Hamiltonian and show that the reservoirs are squeezed and correlated even in the ground state of the whole system. The master-equation approach is discussed in Sec. IV, where we demonstrate the decay of a polariton system to its original ground state. Correlation functions of reservoir fields are calculated in Appendix B, and the detailed calculation of master equation and photon detection is shown and Appendix C. The input-output approach is discussed in Sec. V, and the detailed calculation of photon detection in this approach is shown in Appendix D. Finally, we discuss the comparison with previous theories in Sec. VI, and the summary is in Sec. VII.

II. HAMILTONIAN

The Hamiltonian describing cavity photons and excitations in matter is written as

$$
\begin{aligned}
\hat{H}_{\text{S}} = {} & \hbar\omega_c \hat{a}^\dagger \hat{a} + \hbar\omega_x \hat{b}^\dagger \hat{b} \\
& + i\hbar g(\hat{a} + \hat{a}^\dagger)(\hat{b} - \hat{b}^\dagger) + \hbar D(\hat{a} + \hat{a}^\dagger)^2.
\end{aligned}
\tag{1}
$$

Here, $\hat{a}$ and $\hat{b}$ are annihilation operators of the photon and excitation, respectively, satisfying the bosonic commutation relations $[\hat{a},\hat{a}^\dagger] = [\hat{b},\hat{b}^\dagger] = 1$ and $[\hat{a},\hat{a}] = [\hat{b},\hat{b}] = [\hat{a},\hat{b}] = [\hat{a},\hat{b}^\dagger] = 0$. ω_c and ω_x are their eigenfrequencies, and g is the coupling strength. The ultrastrong coupling means $g \gtrsim \omega_x$. The last term is the so-called diamagnetic term naturally derived in the minimal coupling scheme [1], and the coefficient is normally $D \geqslant g^2/\omega_x$, by which we cannot expect the quantum phase transition [12,29]. If the polariton system is isolated from the environment, as discussed in Ref. [1], this Hamiltonian can be diagonalized as

$$
\hat{H}_{\text{S}} = \sum_{j=L,U} \hbar\omega_j \hat{p}_j^\dagger \hat{p}_j + \text{const.}
\tag{2}
$$

Here, $\hat{p}_L$ and $\hat{p}_U$ are annihilation operators of lower and upper polaritons, respectively. They are represented as a combination of annihilation and creation operators of photon and excitation:

$$
\hat{p}_j = w_j \hat{a} + x_j \hat{b} + y_j \hat{a}^\dagger + z_j \hat{b}^\dagger.
\tag{3}
$$

These coefficients and eigenfrequencies ω_j are determined by solving

$$
\begin{pmatrix}
\omega_c + 2D & -ig & -2D & -ig \\
ig & \omega_x & -ig & 0 \\
2D & -ig & -\omega_c - 2D & -ig \\
-ig & 0 & ig & -\omega_x
\end{pmatrix}
\begin{pmatrix}
w_j \\ x_j \\ y_j \\ z_j
\end{pmatrix}
= \omega_j
\begin{pmatrix}
w_j \\ x_j \\ y_j \\ z_j
\end{pmatrix}.
\tag{4}
$$

From this eigenvalue problem, we get four eigenvalues $\{\omega_L, \omega_U, -\omega_L, -\omega_U\}$, whose eigenvectors correspond to $\{\hat{p}_L, \hat{p}_U, \hat{p}_L^\dagger, \hat{p}_U^\dagger\}$, respectively. The coefficients are normalized for satisfying $[\hat{p}_j, \hat{p}_k^\dagger] = \delta_{j,k}$ for $j, k = L, U$ and we also get $[\hat{p}_j, \hat{p}_k] = 0$. Inversely, the photon and excitation operators are represented by the polariton operators as

$$\begin{pmatrix} \hat{a} \\ \hat{b} \\ \hat{a}^\dagger \\ \hat{b}^\dagger \end{pmatrix} = \begin{pmatrix} w_L^* & w_U^* & -y_L & -y_U \\ x_L^* & x_U^* & -z_L & -z_U \\ -y_L^* & -y_U^* & w_L & w_U \\ -z_L^* & -z_U^* & x_L & x_U \end{pmatrix} \begin{pmatrix} \hat{p}_L \\ \hat{p}_U \\ \hat{p}_L^\dagger \\ \hat{p}_U^\dagger \end{pmatrix}. \quad (5)$$

The photonic and excitonic reservoirs are individually represented as ensembles of harmonic oscillators as

$$\hat{H}_R = \sum_m \hbar \Omega_m^c \hat{\alpha}_m^\dagger \hat{\alpha}_m + \sum_m \hbar \Omega_m^x \hat{\beta}_m^\dagger \hat{\beta}_m, \quad (6)$$

where $\hat{\alpha}_m$ and $\hat{\beta}_m$ are annihilation operators of oscillators in photonic and excitonic reservoirs, respectively, and $\Omega_m^{c,x}$ is the oscillating frequency. The ensembles show nearly continuous spectra. These operators satisfy $[\hat{\alpha}_m, \hat{\alpha}_n^\dagger] = [\hat{\beta}_m, \hat{\beta}_n^\dagger] = \delta_{m,n}$ and $[\hat{\alpha}_m, \hat{\alpha}_n] = [\hat{\beta}_m, \hat{\beta}_n] = [\hat{\alpha}_m, \hat{\beta}_n] = [\hat{\alpha}_m, \hat{\beta}_n^\dagger] = 0$. The system-reservoir coupling is represented as

$$\hat{H}_{\text{S-R}} = i\hbar (\hat{F}_c^\dagger \hat{a} - \hat{a}^\dagger \hat{F}_c) + i\hbar (\hat{F}_x^\dagger \hat{b} - \hat{b}^\dagger \hat{F}_x). \quad (7)$$

Here, $\hat{F}_c$ and $\hat{F}_x$ are photonic and excitonic reservoir fields, respectively, and they are expressed by the annihilation operators $\hat{\alpha}_m$ and $\hat{\beta}_m$ and coupling strengths κ_m and γ_m as

$$\hat{F}_c = \sum_m \kappa_m \hat{\alpha}_m, \quad (8a)$$

$$\hat{F}_x = \sum_m \gamma_m \hat{\beta}_m. \quad (8b)$$

It is worth noting that Eq. (7) is not the result of RWA, but this expression is naturally derived considering the transmission and reflection of particles between the inside and the outside of the cavity [30]. In other words, concerning the coupling between cavity photons and the external photonic field, they are coupled through the electric field and also through the magnetic field. By summing these two interactions, $(i\hbar/2)(\hat{a} - \hat{a}^\dagger)(\hat{F}_c^\dagger + \hat{F}_c)$ and $(i\hbar/2)(\hat{a} + \hat{a}^\dagger)(\hat{F}_c^\dagger - \hat{F}_c)$, we can derive the first term in Eq. (7). For simplicity, we also presume a similar situation concerning the coupling with an excitonic reservoir. Whereas the Hermitian expressions have been presumed for describing the dissipation in some works [24,28,39,40], Eq. (7) can be considered as the standard expression, because there is no ambiguity as to whether the system-reservoir coupling is electric or magnetic. Of course, when we presume specific systems, the expression of system-reservoir coupling is automatically determined. In terms of the polariton operators, the system-reservoir coupling is rewritten as

$$\hat{H}_{\text{S-R}} = i\hbar (\hat{F}_L^\dagger \hat{p}_L - \hat{p}_L^\dagger \hat{F}_L) + i\hbar (\hat{F}_U^\dagger \hat{p}_U - \hat{p}_U^\dagger \hat{F}_U), \quad (9)$$

where

$$\hat{F}_j = w_j \hat{F}_c + x_j \hat{F}_x + y_j \hat{F}_c^\dagger + z_j \hat{F}_x^\dagger \quad (10)$$

is the external field that couples with the lower ($j = L$) or upper ($j = U$) polariton inside the cavity.

As discussed in Ref. [1], when we consider only the $\hat{H}_S$ system isolated from the reservoirs, there are virtual photons and virtual excitations even in the ground state. This is because the ground state should satisfy $\hat{p}_j |\text{g.s.}\rangle = 0$, and then the cavity mode is represented by a squeezed vacuum state in the ground state as

$$\langle \hat{a}^\dagger \hat{a} \rangle_{\text{g.s.}} = \sum_{j=L,U} |y_j|^2, \quad (11a)$$

$$\langle \hat{a}\hat{a} \rangle_{\text{g.s.}} = - \sum_{j=L,U} w_j^* y_j, \quad (11b)$$

where $\langle \cdots \rangle_{\text{g.s.}}$ means an expectation value in the ground state $|\text{g.s.}\rangle$. The excitations in matter are also expressed as a squeezed vacuum state, and the photons and excitations are correlated in the ground state, as discussed in Ref. [1]. If the system-reservoir coupling is weak enough compared to the light-matter coupling g, the squeezing and correlation of cavity photon and excitation should be maintained.

III. DIAGONALIZATION OF THE WHOLE SYSTEM

First of all, we diagonalize the whole system $\hat{H} = \hat{H}_S + \hat{H}_{\text{S-R}} + \hat{H}_R$ by using the Fano-type technique [31–33]. As shown in Appendix A , the photonic part consisting of cavity mode and photonic reservoir is diagonalized as

$$\hat{H}_{\text{ph}} = \hbar \omega_c \hat{a}^\dagger \hat{a} + \sum_m \hbar \Omega_m^c \hat{\alpha}_m^\dagger \hat{\alpha}_m + i\hbar (\hat{F}_c^\dagger \hat{a} - \hat{a}^\dagger \hat{F}_c) \quad (12a)$$

$$= \int_0^\infty d\omega \, \hbar \omega \hat{A}^\dagger(\omega) \hat{A}(\omega) + \text{const.} \quad (12b)$$

Here, the partially diagonalized operator $\hat{A}(\omega)$ is defined in Eq. (A3) and satisfies

$$[\hat{A}(\omega), \hat{H}_{\text{ph}}] = \hbar \omega \hat{A}(\omega) \quad (13)$$

and

$$[\hat{A}(\omega), \hat{A}^\dagger(\omega')] = \delta(\omega - \omega'). \quad (14)$$

Similarly, the excitonic part is diagonalized as

$$\hat{H}_{\text{ex}} = \hbar \omega_x \hat{b}^\dagger \hat{b} + \sum_j \hbar \Omega_j^x \hat{\beta}_j^\dagger \hat{\beta}_j + i\hbar (\hat{F}_x^\dagger \hat{b} - \hat{b}^\dagger \hat{F}_x) \quad (15a)$$

$$= \int_0^\infty d\omega \, \hbar \omega \hat{B}^\dagger(\omega) \hat{B}(\omega) + \text{const.} \quad (15b)$$

The operator $\hat{B}(\omega)$ is represented in Eq. (A12). Using these partially diagonalized operators, the whole Hamiltonian is represented as

$$\hat{H} = \int_0^\infty d\omega \, [\hbar \omega \hat{A}^\dagger(\omega) \hat{A}(\omega) + \hbar \omega \hat{B}^\dagger(\omega) \hat{B}(\omega)]$$
$$+ i\hbar g(\hat{a} + \hat{a}^\dagger)(\hat{b} - \hat{b}^\dagger) + \hbar D(\hat{a} + \hat{a}^\dagger)^2 + \text{const.} \quad (16)$$

The light-matter coupling and diamagnetic terms are also expressed in terms of $\hat{A}(\omega)$ and $\hat{B}(\omega)$ by using Eqs. (A11) and (A13). For diagonalizing the whole Hamiltonian $\hat{H}$, we presume a new operator

$$\hat{P}(\omega) = \int_0^\infty d\omega' \, [W(\omega,\omega') \hat{A}(\omega') + X(\omega,\omega') \hat{B}(\omega')$$
$$+ Y(\omega,\omega') \hat{A}^\dagger(\omega') + Z(\omega,\omega') \hat{B}^\dagger(\omega')]. \quad (17)$$

The coefficient functions are determined for satisfying

$$[\hat{P}(\omega),\hat{H}] = \hbar\omega\hat{P}(\omega) \tag{18}$$

and

$$[\hat{P}(\omega),\hat{P}^\dagger(\omega)] = \delta(\omega - \omega'). \tag{19}$$

Then, $\hat{H}$ can be diagonalized as

$$\hat{H} = \int_0^\infty d\omega\,\hbar\omega\hat{P}^\dagger(\omega)\hat{P}(\omega) + \text{const.} \tag{20}$$

From Eq. (18), the coefficient functions are determined by the eigenvalue problem

$$\begin{pmatrix} \omega' + 2D - \omega & -ig & -2D & -ig \\ ig & \omega' - \omega & -ig & 0 \\ 2D & -ig & -\omega' - 2D - \omega & -ig \\ -ig & 0 & ig & -\omega' - \omega \end{pmatrix}$$
$$\times \begin{pmatrix} u_c(\omega')W(\omega,\omega') \\ u_x(\omega')X(\omega,\omega') \\ u_c(\omega')^*Y(\omega,\omega') \\ u_x(\omega')^*Z(\omega,\omega') \end{pmatrix} = 0, \tag{21}$$

where the coefficient $u_{c,x}(\omega)$ is represented in Eq. (A9). This eigenvalue problem is equivalent to Eq. (5) by replacing ω_c and ω_x with ω', and the eigenfrequencies $\omega_{L,U}(\omega')$ must be equal to ω. Then, operator $\hat{P}(\omega)$ is represented as

$$\hat{P}(\omega) = \sum_{j=L,U} \left\{ \frac{w_j(\omega'_j)}{u_c(\omega'_j)}\hat{A}(\omega'_j) + \frac{x_j(\omega'_j)}{u_x(\omega'_j)}\hat{B}(\omega'_j) \right.$$
$$\left. + \frac{y_j(\omega'_j)}{u_c(\omega'_j)^*}\hat{A}^\dagger(\omega'_j) + \frac{z_j(\omega'_j)}{u_x(\omega'_j)^*}\hat{B}^\dagger(\omega'_j) \right\}. \tag{22}$$

Here, ω'_j is the frequency satisfying $\omega = \omega_j(\omega'_j)$, and $w_j(\omega)$, $x_j(\omega)$, $y_j(\omega)$, and $z_j(\omega)$ are the coefficients when we solve Eq. (5) by replacing ω_c and ω_x with ω. They are normalized for satisfying Eq. (19). Inversely, we can rewrite $\hat{A}(\omega)$ and $\hat{B}(\omega)$ as

$$\hat{A}(\omega) = u_c(\omega)\sum_{j=L,U}[w_j(\omega)^*\hat{P}(\omega_j(\omega)) - y_j(\omega)\hat{P}^\dagger(\omega_j(\omega))], \tag{23a}$$

$$\hat{B}(\omega) = u_x(\omega)\sum_{j=L,U}[x_j(\omega)^*\hat{P}(\omega_j(\omega)) - z_j(\omega)\hat{P}^\dagger(\omega_j(\omega))]. \tag{23b}$$

Then, the original photon, excitation, and reservoir operators are also expressed in terms of $\hat{P}(\omega)$ by using Eqs. (A11) and (A13).

The ground state $|\tilde{g.s.}\rangle$ of the whole system $\hat{H}$ is determined for satisfying $\hat{P}(\omega)|\tilde{g.s.}\rangle = 0$ for $0 < \omega < \infty$. Here, we define photonic and excitonic free fields $\hat{F}_{c,x}^{(0)}(\tau)$ as

$$\hat{F}_\mu^{(0)}(\tau) = e^{i\hat{H}_R t}\hat{F}_\mu^{(0)}e^{-i\hat{H}_R t}, \tag{24}$$

which appears in the master equation and input-output formalism in the following sections. They satisfy $(\mu,\nu = c,x)$

$$[\hat{F}_\mu^{(0)}(t),\hat{F}_\nu^{(0)\dagger}(t')] = \delta_{\mu,\nu}G_\mu(t - t'), \tag{25a}$$

$$[\hat{F}_\mu^{(0)}(t),\hat{F}_\nu^{(0)}(t')] = 0, \tag{25b}$$

where $G_c(\tau)$ and $G_x(\tau)$ are memory functions of the reservoirs expressed as

$$G_c(\tau) = \sum_m |\kappa_m|^2 e^{-i\Omega_m^c\tau}, \tag{26a}$$

$$G_x(\tau) = \sum_m |\gamma_m|^2 e^{-i\Omega_m^x\tau}. \tag{26b}$$

When we apply the photonic free field onto the ground state, it is represented as

$$\hat{F}_c^{(0)}(\tau)|\tilde{g.s.}\rangle = -\int_0^\infty d\omega \int_0^\infty d\omega'\,e^{-i\omega\tau}\kappa(\omega)v_c(\omega',\omega)^*u_c(\omega')$$
$$\times \sum_{j=L,U} y_j(\omega')\hat{P}^\dagger(\omega_j(\omega'))|\tilde{g.s.}\rangle, \tag{27}$$

where coefficient $v_c(\omega',\omega)$ is expressed in Eq. (A6). The phase-independent correlation function is written as

$$\langle\tilde{g.s.}|\hat{F}_c^{(0)\dagger}\hat{F}_c^{(0)}(\tau)|\tilde{g.s.}\rangle$$
$$= \int_0^\infty d\omega \int_0^\infty d\omega' \int_0^\infty d\omega''\,e^{-i\omega'\tau}\kappa(\omega)^*\kappa(\omega')$$
$$\times v_c(\omega'',\omega)v_c(\omega'',\omega')^*|u_c(\omega'')|^2 \sum_{j=L,U}|y_j(\omega'')|^2. \tag{28}$$

Here, the coefficients $u_c(\omega)$ and $v_c(\omega,\omega')$ are singular at $\omega = \omega_c$ as seen in Eqs. (A9) and (A6). Since the coefficients are normalized as

$$\int_0^\infty d\omega\,|u_c(\omega)|^2 = 1, \tag{29}$$

$$\int_0^\infty d\omega\,v_c(\omega,\omega')v_c(\omega,\omega'') = \delta(\omega' - \omega''), \tag{30}$$

if the dissipation of photons is weak enough compared to the characteristic frequencies of the $\hat{H}_S$ system (g, ω_c, and ω_x), the correlation function is approximately represented in the ground state as

$$\langle\tilde{g.s.}|\hat{F}_c^{(0)\dagger}\hat{F}_c^{(0)}(\tau)|\tilde{g.s.}\rangle$$
$$\simeq \int_0^\infty d\omega\,e^{-i\omega\tau}|\kappa(\omega)|^2 \sum_{j=L,U}|y_j(\omega_c)|^2$$
$$= G_c(\tau)\langle\hat{a}^\dagger\hat{a}\rangle_{\text{g.s.}}. \tag{31}$$

In the same manner, the phase-sensitive correlation is expressed in the ground state as

$$\langle\tilde{g.s.}|\hat{F}_c^{(0)}\hat{F}_c^{(0)}(\tau)|\tilde{g.s.}\rangle$$
$$= -\int_0^\infty d\omega \int_0^\infty d\omega' \int_0^\infty d\omega''\,e^{-i\omega'\tau}\kappa(\omega)\kappa(\omega')$$
$$\times v_c(\omega'',\omega)^*v_c(\omega'',\omega')^*u_c(\omega'')^2 \sum_{j=L,U}w_j(\omega'')^*y_j(\omega'')$$
$$\simeq -\int_0^\infty d\omega\,e^{-i\omega\tau}|\kappa(\omega)|^2 \sum_{j=L,U}w_j(\omega_c)^*y_j(\omega_c)$$
$$= G_c(\tau)\langle\hat{a}\hat{a}\rangle_{\text{g.s.}}. \tag{32}$$

Since the photonic reservoir field is normalized as Eq. (25), the degree of squeezing is obtained as

$$1 + 2\frac{\langle \widetilde{g.s.}|\hat{F}_c^\dagger \hat{F}_c|\widetilde{g.s.}\rangle - |\langle \widetilde{g.s.}|\hat{F}_c \hat{F}_c|\widetilde{g.s.}\rangle|}{[\hat{F}_c, \hat{F}_c^\dagger]}$$
$$= 1 + 2\langle \hat{a}^\dagger \hat{a}\rangle_{\text{g.s.}} - 2|\langle \hat{a}\hat{a}\rangle_{\text{g.s.}}|. \qquad (33)$$

Therefore, even in the ground state, the photonic reservoir field $\hat{F}_c$ is also squeezed by the same degree as the cavity mode, and it is determined by the parameters of the relevant system (especially g and D). In the same manner, the correlation of the two reservoir fields are also the same as internal ones if the dissipation is weak enough. Since the reservoir fields $\hat{F}_{c,x}$ are expressed as the sum of the reservoir modes as in Eq. (8), the expectation values of each mode, such as $\langle \hat{\alpha}_m^\dagger \hat{\alpha}_m\rangle_{\text{g.s.}}$, $\langle \hat{\alpha}_m \hat{\alpha}_m\rangle_{\text{g.s.}}$, ..., are quite small (inversely proportional to the number of reservoir modes) compared to the internal ones $\langle \hat{a}^\dagger \hat{a}\rangle_{\text{g.s.}}$, $\langle \hat{a}\hat{a}\rangle_{\text{g.s.}}$, ... However, as seen in Eqs. (31) and (32), correlation functions of the total reservoir fields $\hat{F}_c$ and $\hat{F}_x$ are not diminished by the huge numbers of reservoir modes; instead, the factor $G_{c,x}(\tau)$ is multiplied. In the frequency domain, the correlation functions are expressed such as $\langle \widetilde{g.s.}|\hat{F}_c^{(0)\dagger}\hat{F}_c^{(0)}(\omega)|\widetilde{g.s.}\rangle = G_c(\omega)\langle \hat{a}^\dagger \hat{a}\rangle_{\text{g.s.}}$, then the system-reservoir coupling strength $|\kappa_m|^2$ or $|\gamma_m|^2$ is multiplied. In this way, the internal modes $\hat{a}$ and $\hat{b}$ are balanced with $\hat{F}_c$ and $\hat{F}_x$, respectively, in the ground state of the whole system.

When we initially presume a squeezed cavity mode and a big reservoir in the vacuum state, of course the reservoir does not become equally squeezed but it almost remains in the vacuum state after switching on the coupling between them. This is a nonequilibrium problem. However, when we consider the equilibrium of the cavity mode and the reservoir, if the ground state of the cavity mode is squeezed, the reservoir field is also squeezed in the ground state of the whole system. Therefore, when we consider the dissipation of the ultrastrong light-matter coupling system to its original ground state, we should presume the squeezed and correlated reservoirs as is discussed in the following two sections and also in Sec. VI.

IV. MASTER-EQUATION APPROACH

In this section, we derive a master equation for describing the dissipation of polariton system $\hat{H}_S$ by considering the coupling with reservoirs. Obeying the standard derivation of master equations [24,37,39–42], from the expression (9) of system-reservoir coupling $\hat{H}_{\text{S-R}}$, the master equation for a reduced density operator $\hat{\rho}(t)$ describing $\hat{H}_S$ system is derived under the Born approximation as

$$\frac{\partial}{\partial t}\hat{\rho}(t) = \hat{\mathcal{L}}[\hat{\rho}], \qquad (34)$$

where $\hat{\mathcal{L}} = \hat{\mathcal{L}}_0 + \hat{\mathcal{L}}_{\text{diss}}$ and

$$\hat{\mathcal{L}}_0[\hat{\rho}] = \frac{1}{i\hbar}[\hat{H}_S, \hat{\rho}(t)], \qquad (35)$$

$$\hat{\mathcal{L}}_{\text{diss}}[\hat{\rho}] = \sum_{j,k=L,U} \{[\hat{\mathcal{D}}_{jk}^{\text{L}}[\hat{\rho}], \hat{p}_j^\dagger] + [\hat{p}_j, \hat{\mathcal{D}}_{jk}^{\text{R}}[\hat{\rho}]] + [\hat{\mathcal{C}}_{jk}^{\text{L}}[\hat{\rho}], \hat{p}_j]$$
$$+ [\hat{p}_j^\dagger, \hat{\mathcal{C}}_{jk}^{\text{R}}[\hat{\rho}]] + [\hat{p}_j^\dagger, \hat{\mathcal{B}}_{jk}^{\text{L}}[\hat{\rho}]] + [\hat{\mathcal{B}}_{jk}^{\text{R}}[\hat{\rho}], \hat{p}_j^\dagger]$$
$$+ [\hat{p}_j, \hat{\mathcal{A}}_{jk}^{\text{L}}[\hat{\rho}]] + [\hat{\mathcal{A}}_{jk}^{\text{R}}[\hat{\rho}], \hat{p}_j]\}, \qquad (36)$$

$$\hat{\mathcal{D}}_{jk}^{\text{L}}[\hat{\rho}] = \int_{t_0}^t dt' \, \hat{U}_{\text{S}}(t-t')[\hat{p}_k \hat{\rho}(t')]\langle \hat{F}_j^{(0)}(t)\hat{F}_k^{(0)\dagger}(t')\rangle_{\text{in}}, \qquad (37a)$$

$$\hat{\mathcal{D}}_{jk}^{\text{R}}[\hat{\rho}] = \int_{t_0}^t dt' \, \hat{U}_{\text{S}}(t-t')[\hat{\rho}(t')\hat{p}_k^\dagger]\langle \hat{F}_k^{(0)}(t')\hat{F}_j^{(0)\dagger}(t)\rangle_{\text{in}}, \qquad (37b)$$

$$\hat{\mathcal{C}}_{jk}^{\text{L}}[\hat{\rho}] = \int_{t_0}^t dt' \, \hat{U}_{\text{S}}(t-t')[\hat{p}_k^\dagger \hat{\rho}(t')]\langle \hat{F}_j^{(0)\dagger}(t)\hat{F}_k^{(0)}(t')\rangle_{\text{in}}, \qquad (37c)$$

$$\hat{\mathcal{C}}_{jk}^{\text{R}}[\hat{\rho}] = \int_{t_0}^t dt' \, \hat{U}_{\text{S}}(t-t')[\hat{\rho}(t')\hat{p}_k]\langle \hat{F}_k^{(0)\dagger}(t')\hat{F}_j^{(0)}(t)\rangle_{\text{in}}, \qquad (37d)$$

$$\hat{\mathcal{B}}_{jk}^{\text{L}}[\hat{\rho}] = \int_{t_0}^t dt' \, \hat{U}_{\text{S}}(t-t')[\hat{p}_k^\dagger \hat{\rho}(t')]\langle \hat{F}_j^{(0)}(t)\hat{F}_k^{(0)}(t')\rangle_{\text{in}}, \qquad (37e)$$

$$\hat{\mathcal{B}}_{jk}^{\text{R}}[\hat{\rho}] = \int_{t_0}^t dt' \, \hat{U}_{\text{S}}(t-t')[\hat{\rho}(t')\hat{p}_k^\dagger]\langle \hat{F}_k^{(0)}(t')\hat{F}_j^{(0)}(t)\rangle_{\text{in}}, \qquad (37f)$$

$$\hat{\mathcal{A}}_{jk}^{\text{L}}[\hat{\rho}] = \int_{t_0}^t dt' \, \hat{U}_{\text{S}}(t-t')[\hat{p}_k \hat{\rho}(t')]\langle \hat{F}_j^{(0)\dagger}(t)\hat{F}_k^{(0)\dagger}(t')\rangle_{\text{in}}, \qquad (37g)$$

$$\hat{\mathcal{A}}_{jk}^{\text{R}}[\hat{\rho}] = \int_{t_0}^t dt' \, \hat{U}_{\text{S}}(t-t')[\hat{\rho}(t')\hat{p}_k]\langle \hat{F}_k^{(0)\dagger}(t')\hat{F}_j^{(0)\dagger}(t)\rangle_{\text{in}}. \qquad (37h)$$

Here, $t_0 \to -\infty$ is the switch-on time of system-reservoir coupling. $\hat{U}_{\text{S}}(\tau)[\hat{O}]$ is the propagator in $\hat{H}_{\text{S}}$ system for arbitrary operator $\hat{O}$ as

$$\hat{U}_{\text{S}}(\tau)[\hat{O}] = e^{-i\hat{H}_{\text{S}}\tau/\hbar}\hat{O}e^{i\hat{H}_{\text{S}}\tau/\hbar}, \qquad (38)$$

and $\hat{F}_{L,U}^{(0)}$ is the reservoir field in the interaction picture (free field):

$$\hat{F}_{L,U}^{(0)}(t) = e^{i\hat{H}_{\text{R}}t}\hat{F}_{L,U}e^{-i\hat{H}_{\text{R}}t}. \qquad (39)$$

These free fields are in the polariton base, and it is represented by the ones $\hat{F}_{c,x}^{(0)}$ in the excitation-photon base as in Eq. (10). From the master equation (34), the dynamics in the $\hat{H}_{\text{S}}$ system are determined by supposing correlation functions of the free fields, which are considered as an input from the reservoirs to the $\hat{H}_{\text{S}}$ system.

First of all, we presume that the photonic and excitonic reservoirs are in the vacuum state and the correlation functions are given as ($\mu, \nu = c, x$)

$$\langle \hat{F}_\mu^{(0)}(t)\hat{F}_\nu^{(0)\dagger}(t')\rangle_{\text{in}} = \delta_{\mu,\nu} G_\mu(t-t'), \qquad (40a)$$

$$\langle \hat{F}_\mu^{(0)\dagger}(t)\hat{F}_\nu^{(0)}(t')\rangle_{\text{in}} = \langle \hat{F}_\mu^{(0)}(t)\hat{F}_\nu^{(0)}(t')\rangle_{\text{in}} = 0. \qquad (40b)$$

In Fig. 1, presuming the ground state $\hat{\rho}(0) = |\text{g.s.}\rangle\langle\text{g.s.}|$ at the initial time $t = 0$, we plot the development of numbers of lower and upper polaritons calculated by the master equation (34) and the correlation (40). The memory kernels are simply presumed

as

$$G_c(\tau) = \int_0^{\Omega_c^{\text{cutoff}}} d\Omega \, \frac{\Gamma_c}{2\pi} e^{-i\Omega\tau}, \tag{41a}$$

$$G_x(\tau) = \int_0^{\Omega_x^{\text{cutoff}}} d\Omega \, \frac{\Gamma_x}{2\pi} e^{-i\Omega\tau}, \tag{41b}$$

where the cutoff frequency $\Omega_{c,x}^{\text{cutoff}}$ governs the memory time of the reservoirs as $\sim 1/\Omega_{c,x}^{\text{cutoff}}$. In a similar way as in Ref. [5], the density operator $\hat{\rho}(t)$ is moved outside the time integral in the master equation (34). This treatment is valid if the memory time $1/\Omega_{c,x}^{\text{cutoff}}$ is short enough compared to the specific oscillation periods ($1/\omega_c$, $1/\omega_x$, and $1/g$) of $\hat{H}_S$ system.

As seen in Fig. 1, the polaritons are excited by the vacuum reservoirs (at zero temperature). The periods of the oscillation are approximately $\pi/\omega_{L,U}$ ($\omega_L/\omega_c = 0.414$ and $\omega_U/\omega_c = 2.414$), whereas they are slightly modified by the Lamb shifts. After a long time compared to $1/\Gamma_c = 1/\Gamma_x = (100/2\pi) \times (2\pi/\omega_c)$, the numbers of polaritons reach certain values, which depend on the system-reservoir coupling strengths $\Gamma_{c,x}$.

The polariton system is excited by the vacuum reservoirs, because it is excited when the virtual photons in the ground state escape to the reservoirs. In other words, the ground state of the polariton system is modified by the coupling with vacuum reservoirs. This result can also be understood by Eq. (36). In order to guarantee the decay of the $\hat{H}_S$ system to its original ground state $|g.s.\rangle$, the photonic and excitonic reservoirs should not be in the vacuum state in the excitation-photon base (in terms of $\hat{F}_{c,x}^{(0)}$), but the free fields $\hat{F}_{L,U}^{(0)}$ in the polariton base should be in the vacuum state. In Refs. [24,28], owing to the RWA on system-reservoir coupling, the decay to the ground state $|g.s.\rangle$ is guaranteed by simply considering the vacuum reservoirs in the excitation-photon base. However, in the present paper, we do not use the RWA to maintain the information of quantum fluctuation of the reservoirs. Instead, we presume that the reservoirs are in the vacuum state in polariton base (squeezed and correlated in excitation-photon base).

Let us derive the correlation of reservoir free fields $\hat{F}_{L,U}^{(0)}$ that guarantees the decay to the ground state $|g.s.\rangle$ of the $\hat{H}_S$ system and is simultaneously appropriate to the analysis of Fano-type diagonalization discussed in Sec. III. We assume that the $\hat{H}_S$ system is in the ground state as $\hat{\rho} = |g.s.\rangle\langle g.s.|$. Under this assumption, let us inversely consider how the reservoirs are modified by coupling with the $\hat{H}_S$ system. As discussed in Ref. [35] and in Appendix B of this paper, we can derive the correlation of free fields $\hat{F}_{L,U}^{(0)}$ (on output side) from the density operator $\hat{\rho}(t)$ of the $\hat{H}_S$ system. The equations of motion (Langevin equations) of cavity photons and excitations are derived as

$$\frac{\partial}{\partial t}\hat{a}(t) = \frac{1}{i\hbar}[\hat{a}, \hat{H}_S](t) - \int_{t_0}^t dt' \, G_c(t-t')\hat{a}(t') - \hat{F}_c^{(0)}(t), \tag{42a}$$

$$\frac{\partial}{\partial t}\hat{b}(t) = \frac{1}{i\hbar}[\hat{b}, \hat{H}_S](t) - \int_{t_0}^t dt' \, G_x(t-t')\hat{b}(t') - \hat{F}_x^{(0)}(t). \tag{42b}$$

In the standard theory of quantum optics [36,38], the memory kernels $G_{c,x}(\tau)$ are approximately described by the Dirac's δ function by elongating the frequency range of reservoirs to $-\infty$ and ∞. Since the escaped photons do not reenter into a cavity, we can consider that the photonic reservoir has a quite small coherence time, and this approximation seems valid in most cases. However, in the ultrastrong coupling regime, when we do not use the RWA on system-reservoir coupling, we must keep the reservoir frequencies positive [2], and the Langevin and master equations are written in the time-nonlocal forms in general. Although in the case of time-local equations we usually use the standard input-output relation [36,38], we calculate the correlation between the free fields and internal ones by the formalism of Ref. [35]. First, we define the propagator $\hat{U}(\tau)[\cdots]$ satisfying

$$\hat{\rho}(t+\tau) = \hat{U}(\tau)[\hat{\rho}(t)], \tag{43}$$

$$\frac{\partial}{\partial\tau}\hat{U}(\tau) = \hat{\mathcal{L}}[\hat{U}(\tau)], \tag{44}$$

and the quantum regression theorem [35–38,44,45] is written for $\tau > 0$ as

$$\langle \hat{O}_1(t+\tau)\hat{O}_2(t)\rangle = \text{Tr}\{\hat{O}_1\hat{U}(\tau)[\hat{O}_2\hat{\rho}(t)]\}, \tag{45a}$$

$$\langle \hat{O}_1(t)\hat{O}_2(t+\tau)\rangle = \text{Tr}\{\hat{O}_2\hat{U}(\tau)[\hat{\rho}(t)\hat{O}_1]\}. \tag{45b}$$

As discussed in detail in Appendix B , by using this and Eq. (42), the correlation between $\hat{F}_\mu^{(0)}(t)$ and arbitrary operator $\hat{S}(t)$ in the $\hat{H}_S$ system is derived as ($\mu = c,x$)

$$\langle \hat{S}(t)\hat{F}_\mu^{(0)}(t+\tau)\rangle = -\int_{t_0}^t dt' \, G_\mu(t+\tau-t')\langle \hat{S}(t)\hat{s}_\mu(t')\rangle, \tag{46a}$$

$$\langle \hat{F}_\mu^{(0)}(t+\tau)\hat{S}(t)\rangle = -\int_{t_0}^t dt' \, G_\mu(t+\tau-t')\langle \hat{s}_\mu(t')\hat{S}(t)\rangle, \tag{46b}$$

$$\begin{aligned}&\langle \hat{S}(t+\tau)\hat{F}_\mu^{(0)}(t)\rangle\\&= -\int_{t_0}^t dt' \, G_\mu(t-t')\langle \hat{S}(t+\tau)\hat{s}_\mu(t')\rangle\\&\quad - \text{Tr}\{\hat{S}\hat{U}(\tau)[\hat{s}_\mu\hat{\mathcal{L}}_{\text{diss}}[\hat{\rho}(t)] - \hat{\mathcal{L}}_{\text{diss}}[\hat{s}_\mu\hat{\rho}(t)]]\},\end{aligned} \tag{47a}$$

$$\begin{aligned}&\langle \hat{F}_\mu^{(0)}(t)\hat{S}(t+\tau)\rangle\\&= -\int_{t_0}^t dt' \, G_\mu(t-t')\langle \hat{s}_\mu(t')\hat{S}(t+\tau)\rangle\\&\quad - \text{Tr}\{\hat{S}\hat{U}(\tau)[\hat{\mathcal{L}}_{\text{diss}}[\hat{\rho}(t)]\hat{s}_\mu - \hat{\mathcal{L}}_{\text{diss}}[\hat{\rho}(t)\hat{s}_\mu]]\},\end{aligned} \tag{47b}$$

where

$$\hat{s}_\mu = \begin{cases} \hat{a} & \text{for} \quad \mu = c, \\ \hat{b} & \text{for} \quad \mu = x. \end{cases} \tag{48}$$

Whereas Eqs. (46) are zero in the limit of the time-local case $G_\mu(\tau) \propto \delta(\tau)$ [35], they are in general nonzero in the present nonlocal situation. Of course, if τ is large enough compared to the memory time of the reservoirs, Eqs. (46) is negligible compared to Eqs. (47). Furthermore, the self-correlation of free fields $\hat{F}_{c,x}^{(0)}$ is obtained in a steady state (the ground state

in the present case) as

$$\langle \hat{F}_{\mu}^{(0)\dagger}(\tau)\hat{F}_{\nu}^{(0)}\rangle = G_{\mu}^{*}(\tau)\langle\hat{s}_{\mu}^{\dagger}\hat{s}_{\nu}\rangle_{\text{g.s.}}, \tag{49a}$$

$$\langle \hat{F}_{\mu}^{(0)}(\tau)\hat{F}_{\nu}^{(0)\dagger}\rangle = G_{\mu}(\tau)\langle\hat{s}_{\mu}\hat{s}_{\nu}^{\dagger}\rangle_{\text{g.s.}}, \tag{49b}$$

$$\langle \hat{F}_{\mu}^{(0)}(\tau)\hat{F}_{\nu}^{(0)}\rangle = \begin{cases} G_{\mu}(|\tau|)\langle\hat{s}_{\mu}\hat{s}_{\nu}\rangle_{\text{g.s.}} & \text{for} \quad \tau > 0, \\ G_{\nu}(|\tau|)\langle\hat{s}_{\mu}\hat{s}_{\nu}\rangle_{\text{g.s.}} & \text{for} \quad \tau < 0, \end{cases} \tag{49c}$$

where $\langle\cdots\rangle_{\text{g.s.}}$ means an expectation value in the steady state (ground state). This correlation certainly satisfies Eqs. (25) and is equivalent to the ones derived in Sec. III.

In the sense of perturbation theory, they are the correlation on output side, that is, the modification of the reservoirs due to the coupling with the $\hat{H}_{\text{S}}$ system. The free-field correlation appearing in the master equation (36) is the one on the input side (effect from reservoirs to $\hat{H}_{\text{S}}$ system). Here, under the equilibrium between $\hat{H}_{\text{S}}$ and reservoirs, the correlation of $\hat{F}_{c,x}^{(0)}$ should be equivalent on both the input and the output side. Let us substitute Eqs. (49) to the master equation (36). The free-field correlation in the polariton base can be derived by using Eq. (10). From Eqs. (49), we can easily get for $j,k = L,U$

$$\langle \hat{F}_{j}^{(0)}(\tau > 0)\hat{F}_{k}^{(0)}\rangle_{\text{in}} = \langle \hat{F}_{j}^{(0)\dagger}(\tau > 0)\hat{F}_{k}^{(0)}\rangle_{\text{in}} = 0, \tag{50}$$

and then the master equation is reduced to

$$\frac{\partial}{\partial t}\hat{\rho}(t) = \hat{\mathcal{L}}_{0}[\hat{\rho}] + \sum_{j,k=L,U}\left\{\left[\hat{\mathcal{D}}_{jk}^{\text{L}}[\hat{\rho}] + \hat{\mathcal{B}}_{jk}^{\text{R}}[\hat{\rho}], \hat{p}_{j}^{\dagger}\right]\right.$$
$$\left. + \left[\hat{p}_{j}, \hat{\mathcal{D}}_{jk}^{\text{R}}[\hat{\rho}] + \hat{\mathcal{A}}_{jk}^{\text{L}}[\hat{\rho}]\right]\right\}. \tag{51}$$

The steady state obtained from this equation is certainly the ground state of the closed case $\hat{\rho}_{\text{ss}} = |\text{g.s.}\rangle\langle\text{g.s.}|$, then the decay of the polariton system to its original ground state $|\text{g.s.}\rangle$ is guaranteed by presuming the squeezed and correlated reservoirs in Eqs. (49).

By using this master equation (51), we have calculated the dynamics of the $\hat{H}_{\text{S}}$ system. In Fig. 2, presuming the vacuum state (no photon and no excitation) at the initial time $t = 0$, the numbers of lower and upper polaritons are plotted as a function
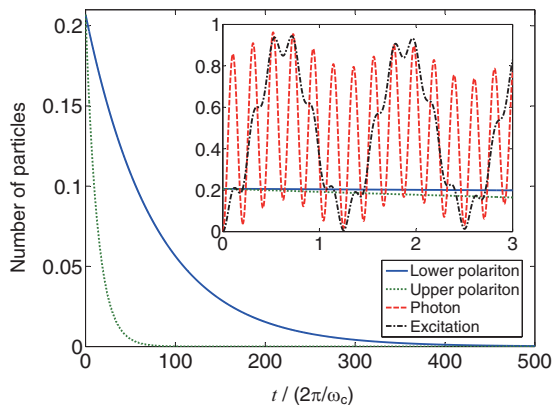

FIG. 2. (Color online) Starting from the vacuum state of photons and excitations, the numbers of polaritons are calculated as a function of dimensionless time $t/(2\pi/\omega_{\text{c}})$ by master equation (51), which are derived by the correlated and squeezed reservoirs as in Eqs. (49). In the inset, the numbers of photons and excitations are also plotted in the early stage. Parameters: $\omega_{\text{x}} = \omega_{\text{c}}$, $g = \omega_{\text{c}}$, $D = g^{2}/\omega_{\text{x}}$, $|\kappa|^{2} = |\gamma|^{2} = 10^{-2}\omega_{\text{c}}/2\pi$, and $\Omega_{c}^{\text{cutoff}} = \Omega_{x}^{\text{cutoff}} = 10^{3}\omega_{\text{c}}$.

of time. In the numerical simulation, the density operator $\hat{\rho}(t)$ is moved outside the time integral, and the memory kernels are also given in Eq. (41). While there are nonzero polaritons at the initial time, the numbers of polaritons decrease and finally go to zero; that is, the $\hat{H}_{\text{S}}$ system decays to its ground state $|\text{g.s.}\rangle$. In the inset of Fig. 2, we also plot the numbers of photons and excitations in the early stage. Whereas both of them are zero at the initial time $t = 0$, they are oscillated with two periods π/ω_{L} and π/ω_{U} (slightly modified by the Lamb shifts), but finally they reach $\langle\hat{a}^{\dagger}\hat{a}\rangle_{\text{g.s.}} = \langle\hat{b}^{\dagger}\hat{b}\rangle_{\text{g.s.}} = 0.207$ after a long time (not shown in the figure).

Under the Born approximation, the total density operator $\hat{\rho}_{\text{tot}}$ is approximately represented by the product of the density operator $\hat{\rho}$ of the $\hat{H}_{\text{S}}$ system and the one $\hat{\rho}_{\text{R}}$ of reservoirs as $\hat{\rho}_{\text{tot}} = \hat{\rho} \otimes \hat{\rho}_{\text{R}}$. If the system-reservoir coupling is weak enough for the Born approximation, in the ground state $|\tilde{\text{g.s.}}\rangle$ of the whole system, the state of $\hat{H}_{\text{S}}$ system is approximately equivalent to the ground state $|\text{g.s.}\rangle$ of the closed case. On the other hand, the free-field correlation (49) approximately reflects the reservoir state that is obtained by tracing over the $\hat{H}_{\text{S}}$ variables on the ground state $|\tilde{\text{g.s.}}\rangle$ as $\hat{\rho}_{\text{R}} \simeq \text{Tr}_{S}\{|\tilde{\text{g.s.}}\rangle\langle\tilde{\text{g.s.}}|\}$, which was verified in Sec. III. This reservoir state is not the ground state of $\hat{H}_{\text{R}}$, but it certainly guarantees the decay of $\hat{H}_{\text{S}}$ system to its original ground state $|\text{g.s.}\rangle$ as seen in Fig. 2. If we presume the ground state of the $\hat{H}_{\text{R}}$ system, in which photonic and excitonic reservoirs are in vacuum (at zero temperature), the $\hat{H}_{\text{S}}$ system does not decay to its ground state $|\text{g.s.}\rangle$, as seen in Fig. 1. However, the obtained steady state is approximately equivalent to the ground state, if the system-reservoir coupling is weak enough. Next, let us calculate the output from the cavity in the formalism of master equation. If the $\hat{H}_{\text{S}}$ system is in the ground state, we cannot detect anything outside the cavity.

As seen in Fig. 3, we consider a ring-shaped cavity embedding a matter interacting with photons inside the cavity, as discussed in Ref. [35]. We assume that backscattering of photons does not occur during the light-matter interaction, and the clockwise and counterclockwise fields are separated. Concerning the external field, we consider a one-dimensional system with length L, and the field is continuous at the two
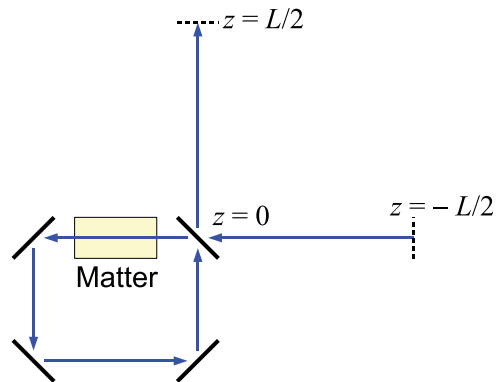

FIG. 3. (Color online) Sketch of the ring-cavity system. Inside the cavity, photons interact with matter, but backscattering of photons does not occur. The external field is defined in the one-dimensional system with length L, and the field is continuously connected at the boundaries $z = \pm L/2$.

ends $z = \pm L/2$. Whereas the external photonic modes are characterized by wave number $k_j = 2\pi j/L$ for $j = 0, \pm 1, \pm 2, \ldots$, the forward field $j > 0$ and backward $j < 0$ fields can be separated into independent subspaces. Here, we focus on the forward field $j > 0$, and its frequency is represented as $\Omega_j^c = ck_j$, where c is the speed of light. The density of states (DOS) is $n_{\text{DOS}}^c = L/2\pi c$. This forward field couples with the counterclockwise intracavity photons. We define the propagating field in the forward direction at position z in the external system as

$$\hat{F}_c^{\text{fwd}}(z,t) = \sum_m \kappa_m \hat{\alpha}_m(t) e^{ik_m z}. \tag{52}$$

From the equation of motion of $\hat{\alpha}_m(t)$, this field is rewritten as

$$\hat{F}_c^{\text{fwd}}(z,t) = \sum_m \kappa_m \hat{\alpha}_m(t_0) e^{-i\Omega_m(t-z/c-t_0)}$$
$$+ \int_{t_0}^t dt' \, G_c(t - t' - z/c) \hat{a}(t'). \tag{53}$$

As discussed in Ref. [35], by choosing a position of observation $z_0 > 0$, we define the output field as

$$\hat{F}_c^{\text{out}}(t) = \hat{F}_c^{\text{fwd}}(z_0, t + z_0/c)$$
$$= \hat{F}_c^{(0)}(t) + \int_{t_0}^{t+z_0/c} dt' \, G_c(t - t') \hat{a}(t'). \tag{54}$$

Here, $\hat{F}_c^{(0)}(t)$ is the free field appearing in the Langevin equation (42) and also in the master equation (34). The second term is the contribution from the cavity. Whereas this term includes the information of cavity photons at time $t + z_0/c > t$, the causality is not violated, because the output field $\hat{F}_c^{\text{out}}(t)$ is actually the propagating field at position z_0 and at time $t + z_0/c$. In the time-local limit $G_c(\tau) = \Gamma_c \delta(\tau)$, Eq. (54) is correctly reduced to the well-known input-output relation [35,36,38]. Further, in the limit of $z_0 \to \infty$ and $t_0 \to -\infty$, Eq. (54) is reduced to the input-output relation (61) in the time-nonlocal case, which is derived in Sec. V.

When we evaluate the output measured by photon detectors, the expectation values should be normal-ordered and time-ordered (expressed as $\langle : \cdots : \rangle$) in terms of polariton operators (not of photon and excitation). The correlation between cavity

photons and the free field of photonic reservoir can be evaluated by Eqs. (46) and (47), and the self-correlation of the free field is also given by Eqs. (49). The detail of the calculation is shown in Appendix C. When we presume that the $\hat{H}_S$ system is in the ground state $|\text{g.s.}\rangle$ by considering the reservoir correlation (49) in the master equation, we have numerically checked that the emission spectrum $\langle : \hat{F}_c^{\text{out}}(\omega) \hat{F}_c^{\text{out}\dagger} : \rangle$ and phase-sensitive correlation $\langle : \hat{F}_c^{\text{out}}(\omega) \hat{F}_c^{\text{out}} : \rangle$ are approximately zero. The deviation is due to the approximation that we used in the numerical calculation [density operator $\hat{\rho}(t)$ is moved outside the integral], and it is not caused by the presumed correlation [Eqs. (49)]. On the other hand, if we presume the vacuum photonic and excitonic reservoirs, we cannot find a policy which guarantees no photon detection, although the vacuum output is obtained for vacuum input in the input-output formalism [2]. This is because of the perturbation treatment in the formalism of the master equation, as we discuss in Sec. VI.

In this way, when we presume the squeezed and correlated reservoir fields as in Eqs. (49), we have successfully obtained the natural result: The $\hat{H}_S$ system decays to its ground state $|\text{g.s.}\rangle$, and the photon emission is not detectable if the system is in the ground state. Furthermore, it is also consistent with the analysis of Fano-type diagonalization (there are virtual photons and excitations in the reservoirs, and photonic and excitonic reservoirs are correlated with each other and also squeezed).

V. INPUT-OUTPUT APPROACH

Another approach for describing the dissipation and emission of photons is the formalism of Langevin equations with input-output relation. As discussed in Ref. [2], the Langevin equations of cavity photons and excitations are derived in the frequency domain as

$$[\mathbf{M}(\omega) - \omega\mathbf{1}] \begin{bmatrix} \hat{a}(\omega) \\ \hat{b}(\omega) \\ \hat{a}(-\omega)^\dagger \\ \hat{b}(-\omega)^\dagger \end{bmatrix} = i \begin{bmatrix} \hat{F}_c^{\text{in}}(\omega) \\ \hat{F}_x^{\text{in}}(\omega) \\ \hat{F}_c^{\text{in}}(-\omega)^\dagger \\ \hat{F}_x^{\text{in}}(-\omega)^\dagger \end{bmatrix}. \tag{55}$$

Here, the coefficient matrix is written as

$$\mathbf{M}(\omega) = \begin{bmatrix} \omega_c + 2D - iG_c(\omega)_+ & ig & 2D & -ig \\ -ig & \omega_x - iG_x(\omega)_+ & -ig & 0 \\ -2D & -ig & -\omega_c - 2D - iG_c(-\omega)_+^* & ig \\ -ig & 0 & -ig & -\omega_x - iG_x(-\omega)_+^* \end{bmatrix}, \tag{56}$$

and the memory kernels $G_{c,x}(\tau)$ are Fourier-transformed for positive time as

$$G_{c,x}(\omega)_+ = \int_0^\infty d\tau \, e^{i\omega\tau} G_{c,x}(\tau). \tag{57}$$

The Langevin (fluctuation) operators are expressed as

$$\hat{F}_c^{\text{in}}(t) = \sum_m \kappa_m \hat{\alpha}_m(t_0) e^{-i\Omega_m^c(t-t_0)} = \sum_m \kappa_m \hat{\alpha}_m^{\text{in}} e^{-i\Omega_m^c t}, \tag{58a}$$

$$\hat{F}_x^{\text{in}}(\omega) = \sum_m \gamma_m \hat{\beta}_m(t_0) e^{-i\Omega_m^x(t-t_0)} = \sum_m \gamma_m \hat{\beta}_m^{\text{in}} e^{-i\Omega_m^x t}, \tag{58b}$$

where $t_0 \to -\infty$ is the switch-on time of system-reservoir interaction, and $\hat{\alpha}_m^{\text{in}}$ and $\hat{\beta}_m^{\text{in}}$ are the input operators. Their Fourier transforms are derived as

$$\hat{F}_c^{\text{in}}(\omega) = \int_{-\infty}^{\infty} dt \, e^{i\omega t} \hat{F}_c^{\text{in}}(t) = 2\pi\theta(\omega)\kappa(\omega)\hat{\alpha}^{\text{in}}(\omega), \quad (59a)$$

$$\hat{F}_x^{\text{in}}(\omega) = \int_{-\infty}^{\infty} dt \, e^{i\omega t} \hat{F}_x^{\text{in}}(t) = 2\pi\theta(\omega)\gamma(\omega)\hat{\beta}^{\text{in}}(\omega). \quad (59b)$$

Here, the reservoir states are rewritten in continuous form as in Eqs. (A1). These fields $\hat{F}_{c,x}(\omega)$ are interpreted as the input fields, and they cannot be defined for negative frequency $\omega < 0$, because the reservoir states are distributed only for positive frequencies $\Omega_j^{c,x} > 0$.

According to the input-output formalism [2], the output photonic field (photonic reservoir field at time $t_1 \to \infty$) is represented as

$$\hat{\alpha}^{\text{out}}(\omega > 0) = \hat{\alpha}^{\text{in}}(\omega) + \kappa(\omega)^* \hat{a}(\omega). \quad (60)$$

As discussed by Ciuti and Carusotto [2], we get the vacuum output for vacuum input. However, the $\hat{H}_S$ system is actually excited by the vacuum reservoirs as $\langle \hat{p}_j^\dagger(\omega)\hat{p}_k \rangle \neq 0$ and $\langle \hat{p}_j(\omega)\hat{p}_k \rangle \neq 0$, which can be easily verified from the Langevin

equations (55). In the master-equation formalism discussed in the previous section, the $\hat{H}_S$ system is also excited, but the vacuum output is not obtained for the vacuum input. Then, there is a discrepancy between the two approaches at least under the Born approximation. Instead, in the input-output formalism, we also presume the squeezed and correlated reservoirs discussed in Secs. III and IV.

According to the standard input-output formalism, the output photonic field is represented as

$$\hat{F}_c^{\text{out}}(t) = \hat{F}_c^{\text{in}}(t) + \int_{-\infty}^{\infty} dt' \, G_c(t - t')\hat{a}(t'), \quad (61a)$$

$$\hat{F}_c^{\text{out}}(\omega) = \hat{F}_c^{\text{in}}(\omega) + G_c(\omega)\hat{a}(\omega). \quad (61b)$$

This expression does not violate the causality [$\hat{F}_c^{\text{out}}(t)$ can be affected by $\hat{a}(t' > t)$] as discussed in Sec. IV. From this input-output relation and the Langevin equations, the output photonic field is eventually represented by the input fields $\hat{F}_{c,x}^{\text{in}}(\omega)$. For discussing the output from the cavity, we have to presume the correlation of input operators $\{\hat{F}_{c,x}^{\text{in}}(\omega)\}$. Here, we consider that the $\hat{H}_S$ system is in the ground state, and the correlation of input operators are also presumed as shown in Eq. (49):

$$\langle \hat{\boldsymbol{F}}_{cx}^{\text{in}}(\omega) \hat{\boldsymbol{F}}_{cx}^{\text{in T}} \rangle_{\text{R}} = \langle \hat{\boldsymbol{F}}_{cx}^{\text{in}}(\omega)_+ \hat{\boldsymbol{F}}_{cx}^{\text{in T}} \rangle_{\text{R}} + \langle \hat{\boldsymbol{F}}_{cx}^{\text{in}}(\omega)_- \hat{\boldsymbol{F}}_{cx}^{\text{in T}} \rangle_{\text{R}}, \tag{62}$$

$$\langle \hat{\boldsymbol{F}}_{cx}^{\text{in}}(\omega)_+ \hat{\boldsymbol{F}}_{cx}^{\text{in T}} \rangle_{\text{R}} = \begin{pmatrix} G_c(\omega)_+ \langle \hat{a}\hat{a}^\dagger \rangle_{\text{g.s.}} & G_c(\omega)_+ \langle \hat{a}\hat{b}^\dagger \rangle_{\text{g.s.}} & G_c(\omega)_+ \langle \hat{a}\hat{a} \rangle_{\text{g.s.}} & G_c(\omega)_+ \langle \hat{a}\hat{b} \rangle_{\text{g.s.}} \\ G_x(\omega)_+ \langle \hat{b}\hat{a}^\dagger \rangle_{\text{g.s.}} & G_x(\omega)_+ \langle \hat{b}\hat{b}^\dagger \rangle_{\text{g.s.}} & G_x(\omega)_+ \langle \hat{b}\hat{a} \rangle_{\text{g.s.}} & G_x(\omega)_+ \langle \hat{b}\hat{b} \rangle_{\text{g.s.}} \\ G_c(-\omega)_+^* \langle \hat{a}^\dagger \hat{a}^\dagger \rangle_{\text{g.s.}} & G_c(-\omega)_+^* \langle \hat{a}^\dagger \hat{b}^\dagger \rangle_{\text{g.s.}} & G_c(-\omega)_+^* \langle \hat{a}^\dagger \hat{a} \rangle_{\text{g.s.}} & G_c(-\omega)_+^* \langle \hat{a}^\dagger \hat{b} \rangle_{\text{g.s.}} \\ G_x(-\omega)_+^* \langle \hat{b}^\dagger \hat{a}^\dagger \rangle_{\text{g.s.}} & G_x(-\omega)_+^* \langle \hat{b}^\dagger \hat{b}^\dagger \rangle_{\text{g.s.}} & G_x(-\omega)_+^* \langle \hat{b}^\dagger \hat{a} \rangle_{\text{g.s.}} & G_x(-\omega)_+^* \langle \hat{b}^\dagger \hat{b} \rangle_{\text{g.s.}} \end{pmatrix}, \tag{63}$$

$$\langle \hat{\boldsymbol{F}}_{cx}^{\text{in}}(\omega)_- \hat{\boldsymbol{F}}_{cx}^{\text{in T}} \rangle_{\text{R}} = \begin{pmatrix} G_c(\omega)_+^* \langle \hat{a}\hat{a}^\dagger \rangle_{\text{g.s.}} & G_x(\omega)_+^* \langle \hat{a}\hat{b}^\dagger \rangle_{\text{g.s.}} & G_c(-\omega)_+ \langle \hat{a}\hat{a} \rangle_{\text{g.s.}} & G_x(-\omega)_+ \langle \hat{a}\hat{b} \rangle_{\text{g.s.}} \\ G_c(\omega)_+^* \langle \hat{b}\hat{a}^\dagger \rangle_{\text{g.s.}} & G_x(\omega)_+^* \langle \hat{b}\hat{b}^\dagger \rangle_{\text{g.s.}} & G_c(-\omega)_+ \langle \hat{b}\hat{a} \rangle_{\text{g.s.}} & G_x(-\omega)_+ \langle \hat{b}\hat{b} \rangle_{\text{g.s.}} \\ G_c(\omega)_+^* \langle \hat{a}^\dagger \hat{a}^\dagger \rangle_{\text{g.s.}} & G_x(\omega)_+^* \langle \hat{a}^\dagger \hat{b}^\dagger \rangle_{\text{g.s.}} & G_c(-\omega)_+ \langle \hat{a}^\dagger \hat{a} \rangle_{\text{g.s.}} & G_x(-\omega)_+ \langle \hat{a}^\dagger \hat{b} \rangle_{\text{g.s.}} \\ G_c(\omega)_+^* \langle \hat{b}^\dagger \hat{a}^\dagger \rangle_{\text{g.s.}} & G_x(\omega)_+^* \langle \hat{b}^\dagger \hat{b}^\dagger \rangle_{\text{g.s.}} & G_c(-\omega)_+ \langle \hat{b}^\dagger \hat{a} \rangle_{\text{g.s.}} & G_x(-\omega)_+ \langle \hat{b}^\dagger \hat{b} \rangle_{\text{g.s.}} \end{pmatrix}, \tag{64}$$

where $\hat{\boldsymbol{F}}_{cx}^{\text{in}}(\omega) = [\hat{F}_c^{\text{in}}(\omega), \hat{F}_x^{\text{in}}(\omega), \hat{F}_c^{\text{in}}(-\omega)^\dagger, \hat{F}_x^{\text{in}}(-\omega)^\dagger]^{\text{T}}$. Precisely speaking the expectation values such as $\langle \hat{a}^\dagger \hat{a} \rangle_{\text{g.s.}}$ should be slightly modified depending on ω as discussed in Appendix D. Assuming this input correlation, the system is certainly in the ground state $\langle \hat{p}_j^\dagger(\omega)\hat{p}_k \rangle = \langle \hat{p}_j(\omega)\hat{p}_k \rangle = 0$ $(j,k = L,U)$, and we also obtain no photon detection $\langle : \hat{F}_c^{\text{out}}(\omega)\hat{F}_c^{\text{out}\dagger} : \rangle = 0$ and $\langle : \hat{F}_c^{\text{out}}(\omega)\hat{F}_c^{\text{out}} : \rangle = 0$ by normal- and time-ordering the operators in the polariton base. The detailed calculation is shown in Appendix D.

In this way, when we presume the squeezed and correlated reservoirs represented in Eqs. (49) and (62), the $\hat{H}_S$ system certainly decays to its ground state $|\text{g.s.}\rangle$ and no photon is detected outside the cavity in both formalisms of master equation and input-output relation. In contrast, when we presume the vacuum reservoirs, different results are obtained in the two formalisms.

VI. DISCUSSION

As discussed in the previous sections, when we consider the squeezed and correlated reservoirs instead of the vacuum ones, both master-equation and input-output formalisms certainly guarantee the decay of the $\hat{H}_S$ system to its ground state and show no photon detection, even though we do not use the RWA on system-reservoir coupling. The presumed reservoir state is approximately equivalent to the one realized in the ground state of the whole system: $\hat{\rho}_R \simeq \text{Tr}_R\{|\tilde{\text{g.s.}}\rangle\langle\tilde{\text{g.s.}}|\}$. On the other hand, if we presume the vacuum reservoirs in excitation-photon base, in the absence of the RWA on system-reservoir coupling, the $\hat{H}_S$ system is excited by the coupling with the reservoirs as seen in Fig. 1.

We have to pay attention to this kind of delicate problem when we discuss the quantum statistics of emission from the system in the ultrastrong light-matter coupling regime, which

has been experimentally realized recently by intersubband transitions [6–9,26,27], superconducting circuits [14–16], and cyclotron transition [25], although only the splitting of eigenmodes has been mainly discussed. In these systems, the dissipation rate is not larger than the light-matter coupling strength g. Then, the squeezing of internal cavity mode could sustain, and the above problem should be, in principle, considered for discussing the emission statistics. Even for the microwave frequencies, the quantum statistics of emission can be measured experimentally in superconducting circuits [46–48].

In the weak or normally strong coupling regime, since the number of virtual photons is quite small and the RWA can be applied on the light-matter coupling, we need not consider the ground state of the whole system, and the relevant system is not excited by the vacuum reservoirs. However, In the ultrastrong coupling regime, we have to determine the reservoir state presumed in the master equation and input-output formalism, according to the situation of how the system and reservoirs start to couple. If we initially prepare the vacuum reservoirs and switch on the system-reservoir coupling, the reservoirs approximately remain in the vacuum state even after the switch on, and the system does not decay to its ground state but to a steady state excited by the vacuum reservoirs as seen in Fig. 1. This is because the $\hat{H}_S$ system is excited when virtual photons escape to the vacuum reservoirs. In order to avoid it, the $\hat{H}_S$ system and the reservoirs should be balanced, as realized in the ground state $|\tilde{g.s.}\rangle$ of the whole system, and we should presume $\hat{\rho}_R \simeq \mathrm{Tr}_R\{|\tilde{g.s.}\rangle\langle\tilde{g.s.}|\}$ in such situation. If we consider that the system and reservoirs are already coupled and the whole system is in the ground state, when we excite the $\hat{H}_S$ system to an excited state, the system certainly decays to its ground state as seen in Fig. 2.

If the reservoirs are quite large and the whole system cannot be in a steady state, we should presume the former situation. When the temperatures of the reservoirs are low enough and the vacuum input from the reservoirs to the system is presumed, the $\hat{H}_S$ system, in principle, does not decay to its ground state. Although the vacuum output is obtained according to the input-output formalism [2], it is not by the master equation. The energy is conserved in the input-output formalism, but it seems not in the master equation. This is because the dynamics of relevant system and the output are discussed in the sense of perturbation theory in the formalism of master equation. In this way, when we presume the vacuum input, we should pay attention to the difference of the two formalisms (at least under the Born approximation). In order to avoid this discrepancy, we should use the RWA on system-reservoir coupling, although the quantum fluctuation of reservoirs is diminished in such a treatment.

On the other hand, if we can define relatively small reservoirs which weakly couple with a large external system with low enough temperature, the small reservoirs and the $\hat{H}_S$ system can decay to the ground state $|\tilde{g.s.}\rangle$ of the coupled system. In such situation, we can presume $\hat{\rho}_R \simeq \mathrm{Tr}_R\{|\tilde{g.s.}\rangle\langle\tilde{g.s.}|\}$, and it guarantees the decay of the $\hat{H}_S$ system to its ground state $|g.s.\rangle$ and gives no photon detection in the small reservoir, as discussed in the previous sections. This result is obtained in both formalisms of master equation and of input-output relation in contrast to supposing the vacuum reservoirs in the excitation-photon base.

As discussed in Ref. [24], by performing the RWA on the system-reservoir coupling, we can simply presume the vacuum reservoirs in excitation-photon base, and the master equation is reduced to the standard Lindblad form. The simplified master equation is derived as follows. Whereas the system-reservoir coupling is originally represented as Eq. (9), here we perform the pretrace RWA [41] as

$$\hat{H}_{\text{S-R}} \simeq i\hbar \sum_{j=L,U} (w_j^* \hat{F}_c^\dagger \hat{p}_j - w_j \hat{p}_j^\dagger \hat{F}_c + x_j^* \hat{F}_x^\dagger \hat{p}_j - x_j \hat{p}_j^\dagger \hat{F}_x),$$
(65)

where the counter-rotating terms are neglected in the polariton base but not in the excitation-photon base. Then, when we presume the vacuum reservoirs in the excitation-photon base as in Eqs. (40), the master equation is derived under the Born approximation as

$$\frac{\partial}{\partial t}\hat{\rho} = \frac{1}{i\hbar}[\hat{H}_S, \hat{\rho}] + \sum_{j,k=L,U} \int_{t_0}^t dt' \{G_c(t-t')w_j w_k^* $$
$$+ G_x(t-t')x_j x_k^*\}[\hat{U}_S(t-t')[\hat{p}_k\hat{\rho}(t')], \hat{p}_j^\dagger] + \text{H.c.}$$
(66)

Further, by neglecting the fast oscillating terms $\hat{p}_k\hat{p}_j^\dagger e^{-i(\omega_k-\omega_j)t}$ for $j \neq k$ (called the post-trace RWA [41]), we finally get the simplified master equation under the Markov approximation as

$$\frac{\partial}{\partial t}\hat{\rho} = \frac{1}{i\hbar}[\hat{H}_S, \hat{\rho}] + \sum_{j=L,U} \frac{\Gamma_c|w_j|^2 + \Gamma_x|x_j|^2}{2}$$
$$\times (2\hat{p}_j\hat{\rho}\hat{p}_j^\dagger - \hat{p}_j^\dagger\hat{p}_j\hat{\rho} - \hat{\rho}\hat{p}_j^\dagger\hat{p}_j),$$
(67)

where the memory kernels are approximated as $G_\mu(t) = \Gamma_\mu\delta(t)$ for simplicity (there remains the Lamb-shift terms in general [24]). If the system-reservoir coupling is expressed in the Hermitian form as $i\hbar(\hat{a} \pm \hat{a}^\dagger)(\hat{F}_c^\dagger \mp \hat{F}_c)$ $[i\hbar(\hat{b} \pm \hat{b}^\dagger)(\hat{F}_x^\dagger \mp \hat{F}_x)]$, the above master equation is simplify rewritten by replacing w_j $[x_j]$ by $w_j \mp y_j$ $[x_j \mp z_j]$. Even in such case, the simplified master equation is represented in the Lindblad form. From Eq. (65), the input-output relation is obtained as

$$\hat{F}_c^{\text{out}} = \hat{F}_c^{\text{in}} + \Gamma_c \sum_j w_j^* \hat{p}_j.$$
(68)

Since the above master equation is reduced to the standard form owing to the pretrace and post-trace RWAs, we consider that the correlation of input operator $\hat{F}_c^{\text{in}}$ is equivalent to that of $\hat{F}_c^{(0)}$ presumed in the master equation: $\langle \hat{F}_c^{\text{in}}(t)\hat{F}_c^{\text{in}\dagger}\rangle_R = G_c(t)$ and $\langle \hat{F}_c^{\text{in}\dagger}(t)\hat{F}_c^{\text{in}}\rangle_R = \langle \hat{F}_c^{\text{in}}(t)\hat{F}_c^{\text{in}}\rangle_R = 0$. Then, the correlation of the output can be calculated as discussed in Ref. [28].

However, in this approach, the photonic and excitonic reservoirs are presumed to be in the vacuum state under the RWA on system-reservoir coupling, although the polariton system does not decay to its ground state $|g.s.\rangle$ in general without the RWA. In other words, the quantum statistics of reservoirs fields are diminished by the RWA, although the reservoirs are originally squeezed and correlated. In contrast, in the present paper, the master equation and input-output formalism are discussed based on the squeezed and correlated reservoirs. The master equation certainly guarantees the decay

of the $\hat{H}_S$ system to its ground state, and in both formalisms no photon is detected when the $\hat{H}_S$ system is in the ground state. Under the Markov approximation the master equation (51) is reduced to

$$\frac{\partial}{\partial t}\hat{\rho} = \frac{1}{i\hbar}[\hat{H}_S,\hat{\rho}] + \sum_{j,k=L,U}\frac{\Gamma_{j,k}}{2}(2\hat{p}_j\hat{\rho}\hat{p}_k^\dagger - \hat{p}_k^\dagger\hat{p}_j\hat{\rho} - \hat{\rho}\hat{p}_k^\dagger\hat{p}_j)$$
$$+ \sum_{j,k=L,U}\left\{\frac{K_{j,k}}{2}(\hat{p}_j\hat{\rho}\hat{p}_k - \hat{p}_k\hat{p}_j\hat{\rho}) + \text{H.c.}\right\}. \quad (69)$$

The coefficients $\Gamma_{j,k}$ and $K_{j,k}$ can be calculated from the presumed free field correlation in Eqs. (49). This does not have the Lindblad form, but certainly guarantees the decay to the ground state |g.s.⟩, as seen in Fig. 2.

In the standard theory [36–38] and also in the discussion of Refs. [24,28], the master equation and the input-output relation are sometimes used together and the correlation of input $\hat{F}_c^{\text{in}}$ is presumed to be equal to that of free field $\hat{F}_c^{(0)}$ given in the master equation. However, the formalism of the master equation is discussed in the sense of perturbation theory. Since the reservoirs are large enough compared to the $\hat{H}_S$ system, the input correlation is not strongly modified and constantly given in the master equation. On the other hand, the output is a perturbation of the reservoirs as a result of the system-reservoir coupling. The correlation of $\hat{F}_c^{(0)}$ can be, in general, different on input and output sides. Actually, when we presume the vacuum reservoirs in excitation-photon base, the self-correlation of $\hat{F}_c^{(0)}$ on output side is not in vacuum, which is calculated by Eq. (49). In order to get the same correlation for input and output sides, we have to consider the squeezed and correlated input $\hat{\rho}_R \simeq \text{Tr}_R\{|\tilde{g}.\text{s.}\rangle\langle\tilde{g}.\text{s.}|\}$. If we want to reduce this complicated formalism into the standard one, we have to perform the RWA on system-reservoir coupling [24,28].

If we already know that the free field $\hat{F}_c^{(0)}$ does not contribute to the observables, we can simply use the RWA on system-reservoir coupling [24,28]. For example, the second-order correlation functions under a resonant excitation can be calculated as discussed in Ref. [28]. However, when we discuss squeezing of the emission, the interference between free field $\hat{F}_c^{(0)}$ and cavity contribution is important, and the quantum fluctuation of $\hat{F}_c^{(0)}$ should not be destroyed by the RWA on system-reservoir coupling. If the cavity system has an optical nonlinearity or embeds ensemble of atoms, we have to treat the Langevin equations perturbatively or the master equation might be appropriate to treat such systems. When we discuss the emission (or lasing) from such complex systems under incoherent excitation, it is difficult to evaluate the validity of the RWA on system-reservoir coupling, and we should presume the squeezed and correlated reservoirs realized in the ground state of the whole system. This kind of approach should give us natural results in the calculation of dissipation and detection of output.

VII. SUMMARY

We have derived the master equation, Langevin equations, and input-output relation for dissipative polariton system in the ultrastrong light-matter coupling regime. The correlation of reservoir free fields are required for calculating not only

the dynamics of the system but also the photon emission from the polariton system. When we presume the vacuum reservoirs, the polariton system is excited in general. Although the vacuum output is obtained for the vacuum input in the input-output formalism, it is not obtained in the master-equation approach under the Born approximation. In order to avoid this discrepancy, we have to perform the RWA on system-reservoir coupling, although it diminishes the quantum statistics of the reservoirs. In order to describe the dissipation in the ultrastrong coupling regime without the RWA on system-reservoir coupling, we have considered the correlation functions of the photonic and excitonic free fields that are squeezed and correlated with each other and realized in the ground state |g̃.s.⟩ of the whole system: $\hat{\rho}_R \simeq \text{Tr}_S\{|\tilde{g}.\text{s.}\rangle\langle\tilde{g}.\text{s.}|\}$. In the formalism of the master equation, the presumed correlation certainly guarantees the decay of the polariton system to its original ground state |g.s.⟩. In the ground state, we have also verified no photon detection as the output from the cavity. Even in the formalism of Langevin equations and input-output relation, we also get no photon detection by considering the squeezed and correlated reservoirs. This reservoir state is also consistent to the analysis of the ground state of the whole system by the Fano-type diagonalization technique. At least when the polariton system is dissipative and is in the ground state, the three approaches, master equation, input-output formalism, and Fano-type diagonalization, give the same result, in contrast to supposing the vacuum reservoirs. The case in the presence of excitation to the system will be discussed in the future. Further, the model of system-reservoir coupling should be determined by first principles, although it was given in Eq. (7) in this paper. Our result suggests that some parts of the large environment are squeezed and correlated by the coupling with a system in the ultrastrong coupling regime, while the rest remains in the vacuum or thermal state. This issue should also be discussed by different models and approaches in the future.

APPENDIX A: DIAGONALIZATION OF PHOTONIC AND EXCITONIC PARTS

In order to diagonalize the whole Hamiltonian $\hat{H} = \hat{H}_S + \hat{H}_{S\text{-}R} + \hat{H}_R$, first of all, we diagonalize the photonic part, Eq. (12). Here, we rewrite the reservoir fields from discrete to continuous form as

$$\hat{\alpha}_m \rightarrow \hat{\alpha}(\Omega_m^c)/\sqrt{n_{\text{DOS}}^c(\Omega_m^c)}, \quad (\text{A1a})$$

$$\hat{\beta}_m \rightarrow \hat{\beta}(\Omega_m^x)/\sqrt{n_{\text{DOS}}^x(\Omega_m^x)}, \quad (\text{A1b})$$

$$\kappa_m \rightarrow \kappa(\Omega_m^c)/\sqrt{n_{\text{DOS}}^c(\Omega_m^c)}, \quad (\text{A1c})$$

$$\gamma_m \rightarrow \gamma(\Omega_m^x)/\sqrt{n_{\text{DOS}}^x(\Omega_m^x)}, \quad (\text{A1d})$$

where $n_{\text{DOS}}^c(\omega)$ and $n_{\text{DOS}}^x(\omega)$ are densities of states of photonic and excitonic reservoirs, respectively. The new reservoir operators satisfy $[\hat{\alpha}(\omega),\hat{\alpha}^\dagger(\omega')] = [\hat{\beta}(\omega),\hat{\beta}^\dagger(\omega')] = \delta(\omega - \omega')$. The photonic Hamiltonian is rewritten as

$$\hat{H}_{\text{ph}} = \hbar\omega_c\hat{a}^\dagger\hat{a} + \int_0^\infty d\omega\, \hbar\omega\hat{a}^\dagger(\omega)\hat{\alpha}(\omega)$$
$$+ i\hbar\int_0^\infty d\omega[\kappa(\omega)^*\hat{\alpha}^\dagger(\omega)\hat{a} - \hat{a}^\dagger\hat{\alpha}(\omega)\kappa(\omega)]. \quad \text{(A2)}$$

This kind of Hamiltonian can be diagonalized by the Fano-type technique [31–33] by introducing an operator for eigenfrequency ω as

$$\hat{A}(\omega) = u_c(\omega)\hat{a} + \int_0^\infty d\omega'\, v_c(\omega,\omega')\hat{\alpha}(\omega'). \quad \text{(A3)}$$

Once this operator satisfies Eq. (13), we can diagonalize the photonic Hamiltonian as in Eq. (12). Further, $\hat{A}(\omega)$ should be normalized as

$$[\hat{A}(\omega),\hat{A}^\dagger(\omega')]$$
$$= u_c(\omega)u_c(\omega')^* + \int_0^\infty d\omega''\, v_c(\omega,\omega'')v_c(\omega',\omega'')^*$$
$$= \delta(\omega - \omega'). \quad \text{(A4)}$$

The coefficient functions $u_c(\omega)$ and $v_c(\omega,\omega')$ are determined as follows. From Eq. (13), we get

$$\omega u_c(\omega) = \omega_c u_c(\omega) + i\int_0^\infty d\omega'\, v_c(\omega,\omega')\kappa(\omega'), \quad \text{(A5a)}$$

$$\omega v_c(\omega,\omega') = \omega' v_c(\omega,\omega') - i\kappa(\omega')u_c(\omega). \quad \text{(A5b)}$$

From the second equation, $v_c(\omega,\omega')$ is expressed as

$$v_c(\omega,\omega') = -i\kappa(\omega')\frac{u_c(\omega)}{\omega - \omega'} \quad \text{(A6a)}$$

$$= -i\kappa(\omega')\left\{\frac{\text{P}}{\omega - \omega'} + \psi(\omega)\delta(\omega - \omega')\right\}u_c(\omega) \quad \text{(A6b)}$$

$$= -i\kappa(\omega')\left\{\frac{1}{\omega - \omega' - i0^+}\right.$$
$$\left. + [\psi(\omega) - i\pi]\delta(\omega - \omega')\right\}u_c(\omega), \quad \text{(A6c)}$$

where P means the principal value integral and function $\psi(\omega)$ is introduced for the following calculation. The expression of $\psi(\omega)$ is determined by substituting the second or third equation into Eq. (A5a) as

$$\psi(\omega) = \frac{1}{|\kappa(\omega)|^2}\left\{\omega - \omega_c - \text{P}\int_0^\infty d\omega'\, \frac{|\kappa(\omega')|^2}{\omega - \omega'}\right\}, \quad \text{(A7a)}$$

$$\psi(\omega) - i\pi = \frac{1}{|\kappa(\omega)|^2}\left\{\omega - \omega_c - \int_0^\infty d\omega'\, \frac{|\kappa(\omega')|^2}{\omega - \omega' - i0^+}\right\}. \quad \text{(A7b)}$$

On the other hand, the expression of $u_c(\omega)$ is determined by the normalization condition, Eq. (14). The commutator is derived

as

$$[\hat{A}(\omega),\hat{A}^\dagger(\omega')] = u_c(\omega)u_c^*(\omega')[\psi(\omega) - i\pi][\psi(\omega) + i\pi]$$
$$\times \delta(\omega - \omega'). \quad \text{(A8)}$$

Then we get

$$u_c(\omega) = \frac{1}{\psi(\omega) - i\pi} = \frac{|\kappa(\omega)|^2}{\omega - \omega_c\zeta(\omega)}, \quad \text{(A9)}$$

where

$$\zeta(\omega) = 1 - \frac{1}{\omega_c}\int_0^\infty d\omega'\, \frac{|\kappa(\omega')|^2}{\omega' - \omega + i0^+}. \quad \text{(A10)}$$

Using the diagonalized operator $\hat{A}(\omega)$, the original ones are represented as

$$\hat{a} = \int_0^\infty d\omega\, u_c(\omega)^*\hat{A}(\omega), \quad \text{(A11a)}$$

$$\hat{\alpha}(\omega) = \int_0^\infty d\omega'\, v_c(\omega',\omega)^*\hat{A}(\omega'). \quad \text{(A11b)}$$

In the same manner, we can also diagonalize the excitonic Hamiltonian as in Eq. (15). The eigenoperator is represented as

$$\hat{B}(\omega) = u_x(\omega)\hat{b} + \int_0^\infty d\omega'\, v_x(\omega,\omega')\hat{\beta}(\omega'). \quad \text{(A12)}$$

The coefficient functions are determined in the same manner by replacing ω_c and $\kappa(\omega)$ with ω_x and $\gamma(\omega)$, respectively. The excitations and excitonic reservoir field are represented as

$$\hat{b} = \int_0^\infty d\omega\, u_x(\omega)^*\hat{B}(\omega), \quad \text{(A13a)}$$

$$\hat{\beta}(\omega) = \int_0^\infty d\omega'\, v_x(\omega',\omega)^*\hat{B}(\omega'). \quad \text{(A13b)}$$

APPENDIX B: CORRELATION OF FREE FIELD

By using the technique in Ref. [35], here we calculate the correlation between the free field $\hat{F}_{c,x}^{(0)}(t)$ and system operators $\hat{a}$ and $\hat{b}$. Further, we also derive the self-correlation of $\hat{F}_{c,x}^{(0)}(t)$.

First, let us calculate $\langle\hat{S}(t)\hat{F}_c^{(0)}(t + \tau)\rangle$ for arbitrary system operator $\hat{S}$ and $\tau > 0$. From the Langevin equation (42), the free field is represented as

$$\hat{F}_c^{(0)}(t) = -\frac{\partial}{\partial t}\hat{a}(t) + \frac{1}{i\hbar}[\hat{a},\hat{H}_{\text{S}}] - \int_{t_0}^t dt'\, G_c(t - t')\hat{a}(t'), \quad \text{(B1)}$$

then we get

$$\langle\hat{S}(t)\hat{F}_c^{(0)}(t + \tau)\rangle$$
$$= -\frac{\partial}{\partial\tau}\langle\hat{S}(t)\hat{a}(t + \tau)\rangle + \frac{1}{i\hbar}\langle\hat{S}(t)[\hat{a},\hat{H}_{\text{S}}](t + \tau)\rangle$$
$$- \int_{t_0}^{t+\tau} dt'\, G_c(t + \tau - t')\langle\hat{S}(t)\hat{a}(t')\rangle. \quad \text{(B2)}$$

The correlation functions of system operators appearing on the right-hand side can be calculated by the master equation (34). By using the quantum regression theorem (45), the first term

of Eq. (B2) is rewritten as

$$
\frac{\partial}{\partial \tau} \langle \hat{S}(t) \hat{a}(t+\tau) \rangle
$$

$$
= \mathrm{Tr}\{\hat{a}\hat{\mathcal{L}}[\hat{U}(\tau)[\hat{\rho}(t)\hat{S}]]\}
$$

$$
= \frac{1}{i\hbar} \langle \hat{S}(t)[\hat{a}, \hat{H}_\mathrm{S}](t+\tau) \rangle + \mathrm{Tr}\{\hat{a}\hat{\mathcal{L}}_\mathrm{diss}[\hat{U}(\tau)[\hat{\rho}(t)\hat{S}]]\},
$$

$$(B3)$$

and its last term is also written as

$$
\mathrm{Tr}\{\hat{a}\hat{\mathcal{L}}_\mathrm{diss}[\hat{U}(\tau)[\hat{\rho}(t)\hat{S}]]\} = -\int_t^{t+\tau} dt'\, \langle \hat{S}(t)\hat{a}(t') \rangle G(t-t').
$$

$$(B4)$$

Substituting these two equations into Eq. (B2), we get Eq. (46a).

Next, let us consider $\langle \hat{F}_c^{(0)}(t+\tau)\hat{S}(t) \rangle$ for $\tau > 0$. From Eq. (B1), we get

$$
\langle \hat{F}_c^{(0)}(t+\tau)\hat{S}(t) \rangle = -\frac{\partial}{\partial \tau} \langle \hat{a}(t+\tau)\hat{S}(t) \rangle + \frac{1}{i\hbar} \langle [\hat{a}(t+\tau), \hat{H}_\mathrm{S}]\hat{S}(t) \rangle - \int_{t_0}^{t+\tau} dt'\, G_c(t+\tau-t')\langle \hat{a}(t')\hat{S}(t) \rangle. \tag{B5}
$$

In the same manner as the above calculation, the first term is rewritten as

$$
\frac{\partial}{\partial \tau} \langle \hat{a}(t+\tau)\hat{S}(t) \rangle = \frac{1}{i\hbar} \langle [\hat{a}, \hat{H}_\mathrm{S}](t+\tau)\hat{S}(t) \rangle - \int_{t_0}^{t+\tau} dt'\, G_c(t+\tau-t')\langle \hat{a}(t')\hat{S}(t) \rangle, \tag{B6}
$$

and we get Eq. (46b).

The next is $\langle \hat{S}(t+\tau)\hat{F}_c^{(0)}(t) \rangle$ for $\tau > 0$. From Eq. (B1), we get

$$
\langle \hat{S}(t+\tau)\hat{F}_c^{(0)}(t) \rangle = -\langle \hat{S}(t+\tau)\frac{\partial}{\partial t}\hat{a}(t) \rangle + \frac{1}{i\hbar} \langle \hat{S}(t+\tau)[\hat{a}, \hat{H}_\mathrm{S}](t) \rangle - \int_{t_0}^t dt'\, G_c(t-t')\langle \hat{S}(t+\tau)\hat{a}(t') \rangle. \tag{B7}
$$

As shown in Ref. [35], the first term is represented as

$$
\left\langle \hat{S}(t+\tau)\frac{\partial}{\partial t}\hat{a}(t) \right\rangle = \frac{1}{i\hbar} \langle \hat{S}(t+\tau)[\hat{a}, \hat{H}_\mathrm{S}](t) \rangle + \mathrm{Tr}\{\hat{S}\hat{U}(\tau)[\hat{a}\hat{\mathcal{L}}_\mathrm{diss}[\hat{\rho}(t)] - \hat{\mathcal{L}}_\mathrm{diss}[\hat{a}\hat{\rho}(t)]]\}, \tag{B8}
$$

and then we get Eqs. (47). The second equation is also derived in the same manner, and similar expressions are obtained also for $\hat{F}_x^{(0)}$.

Since the free field $\hat{F}_c^{(0)}(t)$ is expressed as in Eq. (B1), the self-correlation is represented as

$$
\langle \hat{F}_c^{(0)}(t)\hat{F}_c^{(0)\dagger}(\tau) \rangle = \frac{\partial}{\partial t}\frac{\partial}{\partial \tau} \langle \hat{a}(t)\hat{a}^\dagger(\tau) \rangle - \frac{1}{i\hbar}\frac{\partial}{\partial t} \langle \hat{a}(t)[\hat{a}^\dagger, \hat{H}_\mathrm{S}](\tau) \rangle + \int_{t_0}^\tau dt'\, G_c^*(\tau-t')\frac{\partial}{\partial t} \langle \hat{a}(t)\hat{a}^\dagger(t') \rangle
$$

$$
- \frac{1}{i\hbar}\frac{\partial}{\partial \tau} \langle [\hat{a}, \hat{H}_\mathrm{S}](t)\hat{a}^\dagger(\tau) \rangle + \frac{1}{(i\hbar)^2} \langle [\hat{a}, \hat{H}_\mathrm{S}](t)[\hat{a}^\dagger, \hat{H}_\mathrm{S}](\tau) \rangle - \frac{1}{i\hbar}\int_{t_0}^\tau dt'\, G_c^*(\tau-t')\langle [\hat{a}, \hat{H}_\mathrm{S}](t)\hat{a}^\dagger(t') \rangle
$$

$$
+ \int_{t_0}^t dt''\, G_c(t-t'')\frac{\partial}{\partial \tau} \langle \hat{a}(t'')\hat{a}^\dagger(\tau) \rangle - \frac{1}{i\hbar}\int_{t_0}^t dt''\, G_c(t-t'')\langle \hat{a}(t'')[\hat{a}^\dagger, \hat{H}_\mathrm{S}](\tau) \rangle
$$

$$
+ \int_{t_0}^t dt'' \int_{t_0}^\tau dt'\, G_c(t-t'')G_c^*(\tau-t')\langle \hat{a}(t'')\hat{a}^\dagger(t') \rangle. \tag{B9}
$$

In the same manner as discussed above, the first term is rewritten for $\tau > t$ as

$$
\frac{\partial}{\partial t}\frac{\partial}{\partial \tau} \langle \hat{a}(t)\hat{a}^\dagger(\tau) \rangle = \frac{\partial}{\partial t}\frac{\partial}{\partial \tau}\mathrm{Tr}\{\hat{a}^\dagger\hat{U}(\tau-t)[\hat{\rho}(t)\hat{a}]\}
$$

$$
= \frac{1}{i\hbar}\frac{\partial}{\partial t} \langle \hat{a}(t)[\hat{a}^\dagger, \hat{H}_\mathrm{S}](\tau) \rangle - \frac{\partial}{\partial t}\int_t^\tau dt'\, \langle \hat{a}(t)\hat{a}^\dagger(t') \rangle G_c^*(\tau-t')
$$

$$
= \frac{1}{i\hbar}\frac{\partial}{\partial t} \langle \hat{a}(t)[\hat{a}^\dagger, \hat{H}_\mathrm{S}](\tau) \rangle - \int_t^\tau dt'\, \frac{\partial}{\partial t} \langle \hat{a}(t)\hat{a}^\dagger(t') \rangle G_c^*(\tau-t') + G_c^*(\tau-t)\langle \hat{a}(t)\hat{a}^\dagger(t) \rangle. \tag{B10}
$$

Rewriting the fourth and seventh terms in Eq. (B9), the self-correlation is reduced to

$$
\langle \hat{F}_c^{(0)}(t)\hat{F}_c^{(0)\dagger}(\tau) \rangle = G_c^*(\tau-t)\langle \hat{a}(t)\hat{a}^\dagger(t) \rangle + \int_{t_0}^t dt'\, G_c^*(\tau-t')\frac{\partial}{\partial t} \langle \hat{a}(t)\hat{a}^\dagger(t') \rangle - \frac{1}{i\hbar}\int_{t_0}^t dt'\, G_c^*(\tau-t')\langle [\hat{a}, \hat{H}_\mathrm{S}](t)\hat{a}^\dagger(t') \rangle
$$

$$
+ \int_{t_0}^t dt'' \int_{t_0}^{t''} dt'\, G_c(t-t'')G_c^*(\tau-t')\langle \hat{a}(t'')\hat{a}^\dagger(t') \rangle. \tag{B11}
$$

Rewriting the second term, we finally get

$$
\begin{aligned}
\langle \hat{F}_c^{(0)}(t) \hat{F}_c^{(0)\dagger}(\tau) \rangle &= G_c^*(\tau - t) \langle \hat{a}(t) \hat{a}^\dagger(t) \rangle - \int_{t_0}^{t} dt' \int_{t'}^{t} dt'' \\
&\quad \times G_c(t - t'') G_c^*(\tau - t') \langle \hat{a}(t'') \hat{a}^\dagger(t') \rangle \\
&\quad + \int_{t_0}^{t} dt'' \int_{t_0}^{t''} dt' \, G_c(t - t'') G_c^*(\tau - t') \\
&\quad \times \langle \hat{a}(t'') \hat{a}^\dagger(t') \rangle \\
&= G_c^*(\tau - t) \langle \hat{a}(t) \hat{a}^\dagger(t) \rangle.
\end{aligned}
\tag{B12}
$$

In the same manner, we finally get Eqs. (49).

APPENDIX C: CALCULATION OF OBSERVABLES IN THE MASTER-EQUATION APPROACH

When we detect photons emitted from the cavity, the observables by photon detectors should be calculated by normal- and time-ordering the photon operators [35,36,38]. In the present case, the ordering should be performed in the polariton basis, which really represents the eigenstates of the system. Here, we divide the photon operator $\hat{a}$ into the lowering parts $\hat{a}_\downarrow$ and raising part $\hat{a}_\uparrow$ as

$$
\hat{a}_\downarrow = w_L^* \hat{p}_L + w_U^* \hat{p}_U, \tag{C1a}
$$

$$
\hat{a}_\uparrow = -y_L \hat{p}_L^\dagger - y_U \hat{p}_U^\dagger. \tag{C1b}
$$

Since the system-reservoir coupling is expressed as in Eq. (9), the photonic free field $\hat{F}_c^{(0)}$ is also divided as

$$
\hat{F}_{c\downarrow}^{(0)} = w_L^* \hat{F}_L^{(0)} + w_U^* \hat{F}_U^{(0)}, \tag{C2a}
$$

$$
\hat{F}_{c\uparrow}^{(0)} = -y_L \hat{F}_L^{(0)\dagger} - y_U \hat{F}_U^{(0)\dagger}, \tag{C2b}
$$

and then the output field (54) is rewritten as

$$
\hat{F}_c^{\text{out}} = \hat{F}_{c\downarrow}^{\text{out}} + \hat{F}_{c\uparrow}^{\text{out}}, \tag{C3}
$$

$$
\hat{F}_{c\downarrow}^{\text{out}}(t) = \hat{F}_{c\downarrow}^{(0)}(t) + \int_{t_0}^{t+z_0/c} dt' \, G_c(t - t') \hat{a}_\downarrow(t'), \tag{C4a}
$$

$$
\hat{F}_{c\uparrow}^{\text{out}}(t) = \hat{F}_{c\uparrow}^{(0)}(t) + \int_{t_0}^{t+z_0/c} dt' \, G_c(t - t') \hat{a}_\uparrow(t'). \tag{C4b}
$$

The equation of motion of $\hat{a}_{\downarrow\uparrow}$ is derived as

$$
\begin{aligned}
\frac{\partial}{\partial t} \hat{a}_{\downarrow\uparrow}(t) &= \frac{1}{i\hbar} [\hat{a}_{\downarrow\uparrow}, \hat{H}_S](t) \\
&\quad - \int_{t_0}^{t} dt' \, G_c(t - t') \hat{a}_{\downarrow\uparrow}(t') - \hat{F}_{c\downarrow\uparrow}^{(0)}(t).
\end{aligned}
\tag{C5}
$$

Then, the correlation between $\hat{a}_{\downarrow\uparrow}$ and $\hat{F}_{c\downarrow\uparrow}^{(0)}$ is also derived in the same form as Eqs. (46) and (47), and the self-correlation of $\hat{F}_{c\downarrow\uparrow}^{(0)}$ also has the same form as Eq. (49).

The output field measured by photon detectors outside the cavity is calculated by normal- and time-ordering the operators. The emission spectrum (number of photons) in a steady state is expressed as

$$
\begin{aligned}
\langle :\hat{F}_c^{\text{out}}(\omega) \hat{F}_c^{\text{out}\dagger}: \rangle &= \langle :\hat{F}_c^{(0)}(\omega) \hat{F}_c^{(0)\dagger}: \rangle + G_c(\omega) \langle :\hat{a}(\omega) \hat{F}_c^{(0)\dagger}: \rangle \\
&\quad + G_c(\omega)^* \langle :\hat{F}_c^{(0)}(\omega) \hat{a}^\dagger: \rangle + |G_c(\omega)|^2 \langle :\hat{a}(\omega) \hat{a}^\dagger: \rangle,
\end{aligned}
\tag{C6}
$$

where

$$
\begin{aligned}
&\langle :\hat{F}_c^{(0)}(\omega) \hat{F}_c^{(0)\dagger}: \rangle \\
&= G_c(\omega) \{ \langle \hat{a}_\downarrow^\dagger \hat{a}_\downarrow \rangle + \langle \hat{a}_\uparrow \hat{a}_\uparrow^\dagger \rangle \} + G_c(\omega)_+ \{ \langle \hat{a}_\downarrow^\dagger \hat{a}_\uparrow \rangle \\
&\quad + \langle \hat{a}_\downarrow \hat{a}_\uparrow^\dagger \rangle \} + G_c(\omega)_+^* \{ \langle \hat{a}_\downarrow \hat{a}_\uparrow^\dagger \rangle + \langle \hat{a}_\uparrow^\dagger \hat{a}_\downarrow \rangle \},
\end{aligned}
\tag{C7a}
$$

$$
\begin{aligned}
&\langle :\hat{a}(\omega) \hat{F}_c^{(0)\dagger}: \rangle \\
&= \langle \hat{F}_{c\downarrow}^{(0)}(\omega)^\dagger \hat{a}_\downarrow \rangle + \langle \hat{F}_{c\downarrow}^{(0)}(\omega)_-^\dagger \hat{a}_\uparrow \rangle + \langle \hat{a}_\uparrow(\omega)_- \hat{F}_{c\downarrow}^{(0)\dagger} \rangle \\
&\quad + \langle \hat{a}_\downarrow(\omega)_+ \hat{F}_{c\uparrow}^{(0)\dagger} \rangle + \langle \hat{F}_{c\uparrow}^{(0)}(\omega)_+^\dagger \hat{a}_\downarrow \rangle + \langle \hat{a}_\uparrow(\omega) \hat{F}_{c\uparrow}^{(0)\dagger} \rangle,
\end{aligned}
\tag{C7b}
$$

$$
\begin{aligned}
&\langle :\hat{F}_c^{(0)}(\omega) \hat{a}^\dagger: \rangle \\
&= \langle \hat{a}_\downarrow(\omega)^\dagger \hat{F}_{c\downarrow}^{(0)} \rangle + \langle \hat{a}_\downarrow(\omega)_-^\dagger \hat{F}_{c\uparrow}^{(0)} \rangle + \langle \hat{F}_{c\uparrow}^{(0)}(\omega)_- \hat{a}_\downarrow^\dagger \rangle \\
&\quad + \langle \hat{F}_{c\downarrow}^{(0)}(\omega)_+ \hat{a}_\uparrow^\dagger \rangle + \langle \hat{a}_\uparrow(\omega)_+^\dagger \hat{F}_{c\downarrow}^{(0)} \rangle + \langle \hat{F}_{c\uparrow}^{(0)}(\omega) \hat{a}_\uparrow^\dagger \rangle,
\end{aligned}
\tag{C7c}
$$

$$
\begin{aligned}
&\langle :\hat{a}(\omega) \hat{a}^\dagger: \rangle \\
&= \langle \hat{a}_\downarrow(\omega)^\dagger \hat{a}_\downarrow \rangle + \langle \hat{a}_\downarrow(\omega)_-^\dagger \hat{a}_\uparrow \rangle + \langle \hat{a}_\uparrow(\omega)_- \hat{a}_\downarrow^\dagger \rangle \\
&\quad + \langle \hat{a}_\downarrow(\omega)_+ \hat{a}_\uparrow^\dagger \rangle + \langle \hat{a}_\uparrow(\omega)_+^\dagger \hat{a}_\downarrow \rangle + \langle \hat{a}_\uparrow(\omega) \hat{a}_\uparrow^\dagger \rangle.
\end{aligned}
\tag{C7d}
$$

On the other hand, the phase-sensitive correlation is expressed as

$$
\begin{aligned}
&\langle :\hat{F}_c^{\text{out}}(\omega) \hat{F}_c^{\text{out}}: \rangle \\
&= \langle :\hat{F}_c^{(0)}(\omega) \hat{F}_c^{(0)}: \rangle + G_c(\omega) \langle :\hat{a}(\omega) \hat{F}_c^{(0)}: \rangle \\
&\quad + G_c(-\omega) \langle :\hat{F}_c^{(0)}(\omega) \hat{a}: \rangle + G_c(\omega) G_c(-\omega) \langle :\hat{a}(\omega) \hat{a}: \rangle,
\end{aligned}
\tag{C8}
$$

$$
\begin{aligned}
&\langle :\hat{F}_c^{(0)}(\omega) \hat{F}_c^{(0)}: \rangle \\
&= \{ G_c(\omega)_+ + G_c(-\omega)_+ \} \{ 2\langle \hat{a}_\uparrow \hat{a}_\downarrow \rangle + \langle \hat{a}_\uparrow \hat{a}_\uparrow \rangle + \langle \hat{a}_\downarrow \hat{a}_\downarrow \rangle \},
\end{aligned}
\tag{C9a}
$$

$$
\begin{aligned}
&\langle :\hat{a}(\omega) \hat{F}_c^{(0)}: \rangle \\
&= \langle \hat{a}_\downarrow(\omega)_+ \hat{F}_{c\downarrow}^{(0)} \rangle + \langle \hat{F}_{c\downarrow}^{(0)}(-\omega)_+ \hat{a}_\downarrow \rangle + \langle \hat{F}_{c\uparrow}^{(0)} \hat{a}_\downarrow(\omega) \rangle \\
&\quad + \langle \hat{a}_\uparrow(\omega) \hat{F}_{c\downarrow}^{(0)} \rangle + \langle \hat{F}_{c\uparrow}^{(0)}(-\omega)_- \hat{a}_\uparrow \rangle + \langle \hat{a}_\uparrow(\omega)_- \hat{F}_{c\uparrow}^{(0)} \rangle,
\end{aligned}
\tag{C9b}
$$

$$
\begin{aligned}
&\langle :\hat{F}_c^{(0)}(\omega) \hat{a}: \rangle \\
&= \langle \hat{F}_{c\downarrow}^{(0)}(\omega)_+ \hat{a}_\downarrow \rangle + \langle \hat{a}_\downarrow(-\omega)_+ \hat{F}_{c\downarrow}^{(0)} \rangle + \langle \hat{a}_\uparrow \hat{F}_{c\downarrow}^{(0)}(\omega) \rangle \\
&\quad + \langle \hat{F}_{c\uparrow}^{(0)}(\omega) \hat{a}_\downarrow \rangle + \langle \hat{a}_\uparrow(-\omega)_- \hat{F}_{c\uparrow}^{(0)} \rangle + \langle \hat{F}_{c\uparrow}^{(0)}(\omega)_- \hat{a}_\uparrow \rangle,
\end{aligned}
\tag{C9c}
$$

$$
\begin{aligned}
&\langle :\hat{a}(\omega) \hat{a}: \rangle \\
&= \langle \hat{a}_\downarrow(\omega)_+ \hat{a}_\downarrow \rangle + \langle \hat{a}_\downarrow(-\omega)_+ \hat{a}_\downarrow \rangle + \langle \hat{a}_\uparrow(-\omega) \hat{a}_\downarrow \rangle \\
&\quad + \langle \hat{a}_\uparrow(\omega) \hat{a}_\downarrow \rangle + \langle \hat{a}_\uparrow(-\omega)_- \hat{a}_\uparrow \rangle + \langle \hat{a}_\uparrow(\omega)_- \hat{a}_\uparrow \rangle.
\end{aligned}
\tag{C9d}
$$

We have numerically checked that the correlation functions $\langle :\hat{F}_c^{\text{out}}(\omega) \hat{F}_c^{\text{out}\dagger}: \rangle$ and $\langle :\hat{F}_c^{\text{out}}(\omega) \hat{F}_c^{\text{out}}: \rangle$ are approximately zero

if the $\hat{H}_S$ system is in the ground state $\hat{\rho}_{ss} = |\text{g.s.}\rangle\langle\text{g.s.}|$. The small deviation comes from the approximation in which the density operator $\hat{\rho}(t)$ is moved outside the time integral in the master equation (34).

APPENDIX D: CALCULATION OF OBSERVABLES IN INPUT-OUTPUT FORMALISM

Let us calculate the emission and squeezing of photonic output from the cavity in the input-output formalism. The coefficient matrix (56) of the Langevin equations is diagonalized as

$$\mathbf{M}(\omega) = \mathbf{V}(\omega)\mathbf{D}\begin{bmatrix} \tilde{\omega}_L(\omega) \\ \tilde{\omega}_U(\omega) \\ -\tilde{\omega}_L(-\omega)^* \\ -\tilde{\omega}_U(-\omega)^* \end{bmatrix}\mathbf{V}(\omega)^{-1}, \quad \text{(D1)}$$

where $\mathbf{D}[\cdots]$ represents an diagonal matrix with elements $\cdots$. Due to the coupling with reservoirs, the eigenvalues $\{\tilde{\omega}_{L,U}(\omega)\}$ depend on frequency ω and are modified from the original eigenfrequencies $\{\omega_{L,U}\}$ derived from Eq. (4). The modification depends on the strengths κ_j and γ_j of system-reservoir coupling. We redefine Langevin (fluctuation) operators in the polariton basis as

$$\check{\boldsymbol{F}}_{LU}^{(0)}(\omega) = \begin{bmatrix} \check{F}_L^{(0)}(\omega) \\ \check{F}_U^{(0)}(\omega) \\ \check{F}_L^{(0)}(-\omega)^{\dagger} \\ \check{F}_U^{(0)}(-\omega)^{\dagger} \end{bmatrix} = \mathbf{V}(\omega)^{-1}\begin{bmatrix} \hat{F}_c^{(0)}(\omega) \\ \hat{F}_x^{(0)}(\omega) \\ \hat{F}_c^{(0)}(-\omega)^{\dagger} \\ \hat{F}_x^{(0)}(-\omega)^{\dagger} \end{bmatrix}. \quad \text{(D2)}$$

Because of the modification of the coefficients, these operators are, in general, different from the Fourier transform of free field of the reservoir field $\hat{F}_{L,U}$ [Eq. (10)] in the polariton basis. However, if the system-reservoir coupling is weak

enough compared to the characteristic frequency of the polariton system, the redefined operators are approximately equal to the Fourier transform of Eq. (39). At the same time, the Born approximation used in the master equation is also valid. The photon and excitation operators are then represented in the frequency domain as

$$\begin{bmatrix} \hat{a}(\omega) \\ \hat{b}(\omega) \\ \hat{a}(-\omega)^{\dagger} \\ \hat{b}(-\omega)^{\dagger} \end{bmatrix} = \mathbf{L}(\omega)\check{\boldsymbol{F}}_{LU}^{(0)}(\omega), \quad \text{(D3)}$$

where

$$\mathbf{L}(\omega) = [\mathbf{M}(\omega) - \omega\mathbf{1}]^{-1}\mathbf{V}(\omega). \quad \text{(D4)}$$

Substituting Eqs. (D2) and (D3) into the input-output relation (61), the photonic output operator is represented as

$$\hat{F}_c^{\text{out}}(\omega) = T_L(\omega)\check{F}_L^{(0)}(\omega) + T_U(\omega)\check{F}_U^{(0)}(\omega)$$
$$+ S_L(\omega)\check{F}_L^{(0)}(-\omega)^{\dagger} + S_U(\omega)\check{F}_U^{(0)}(-\omega)^{\dagger}, \quad \text{(D5)}$$

where

$$T_L(\omega) = V_{11}(\omega) + G_c(\omega)L_{11}(\omega), \quad \text{(D6a)}$$

$$T_U(\omega) = V_{12}(\omega) + G_c(\omega)L_{12}(\omega), \quad \text{(D6b)}$$

$$S_L(\omega) = V_{13}(\omega) + G_c(\omega)L_{13}(\omega), \quad \text{(D6c)}$$

$$S_U(\omega) = V_{14}(\omega) + G_c(\omega)L_{14}(\omega). \quad \text{(D6d)}$$

As discussed in Sec. V, for describing the dissipation of $\hat{H}_S$ system, we consider the correlation of free fields as in Eq. (62). Precisely speaking, in order to guarantee no photon detection, the expectation values in the ground state should be replaced by

$$\begin{pmatrix} \langle\hat{a}\hat{a}^{\dagger}\rangle_{\text{g.s.}} & \langle\hat{a}\hat{b}^{\dagger}\rangle_{\text{g.s.}} & \langle\hat{a}\hat{a}\rangle_{\text{g.s.}} & \langle\hat{a}\hat{b}\rangle_{\text{g.s.}} \\ \langle\hat{b}\hat{a}^{\dagger}\rangle_{\text{g.s.}} & \langle\hat{b}\hat{b}^{\dagger}\rangle_{\text{g.s.}} & \langle\hat{b}\hat{a}\rangle_{\text{g.s.}} & \langle\hat{b}\hat{b}\rangle_{\text{g.s.}} \\ \langle\hat{a}^{\dagger}\hat{a}^{\dagger}\rangle_{\text{g.s.}} & \langle\hat{a}^{\dagger}\hat{b}^{\dagger}\rangle_{\text{g.s.}} & \langle\hat{a}^{\dagger}\hat{a}\rangle_{\text{g.s.}} & \langle\hat{a}^{\dagger}\hat{b}\rangle_{\text{g.s.}} \\ \langle\hat{b}^{\dagger}\hat{a}^{\dagger}\rangle_{\text{g.s.}} & \langle\hat{b}^{\dagger}\hat{b}^{\dagger}\rangle_{\text{g.s.}} & \langle\hat{b}^{\dagger}\hat{a}\rangle_{\text{g.s.}} & \langle\hat{b}^{\dagger}\hat{b}\rangle_{\text{g.s.}} \end{pmatrix} \rightarrow \mathbf{V}(\omega)\begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix}\mathbf{V}(\omega)^{*\text{T}}, \quad \text{(D7)}$$

because of the modification of coefficients $\mathbf{V}(\omega)$. If we detect the output photons outside the cavity, the detection process should be dissipative for the whole system, and virtual photons should not be counted. Then, when we calculate emission spectrum $\langle:\hat{F}_c^{\text{out}}(\omega)\hat{F}_c^{\text{out}\dagger}:\rangle$ and phase-sensitive correlation $\langle:\hat{F}_c^{\text{out}}(\omega)\hat{F}_c^{\text{out}}:\rangle$, the fluctuation (Langevin) operators $\{\hat{F}_{L,U}^{(0)}(\omega)\}$ should be normal- and time-ordered, obeying the theory of measurement [35,36,38]. Then, the correlation functions of output photonic field (D5) are represented as

$$\langle:\hat{F}_c^{\text{out}}(\omega)\hat{F}_c^{\text{out}\dagger}:\rangle = \sum_{j,k=L,U}\left\{ T_j(\omega)\langle\check{F}_k^{(0)}(\omega)^{\dagger}\check{F}_j^{(0)}\rangle T_k(\omega)^* + T_j(\omega)\left[\langle\check{F}_j^{(0)}(\omega)_+\check{F}_k^{(0)}\rangle + \langle\check{F}_k^{(0)}(-\omega)_+\check{F}_j^{(0)}\rangle\right]S_k(\omega)^* \right.$$

$$\left. + S_j(\omega)\left[\langle\check{F}_k^{(0)}(-\omega)_-^{\dagger}\check{F}_j^{(0)\dagger}\rangle + \langle\check{F}_j^{(0)}(\omega)_-^{\dagger}\check{F}_k^{(0)\dagger}\rangle\right]T_k(\omega)^* + S_j(\omega)\langle\check{F}_j^{(0)}(-\omega)^{\dagger}\check{F}_k^{(0)}\rangle S_k(\omega)^* \right\}, \quad \text{(D8)}$$

$$\langle:\hat{F}_c^{\text{out}}(\omega)\hat{F}_c^{\text{out}}:\rangle = \sum_{j,k=L,U}\left\{ T_j(\omega)\left[\langle\check{F}_j^{(0)}(\omega)_+\check{F}_k^{(0)}\rangle + \langle\check{F}_k^{(0)}(-\omega)_+\check{F}_j^{(0)}\rangle\right]T_k(-\omega) + T_j(\omega)\langle\check{F}_k^{(0)}(\omega)^{\dagger}\check{F}_j^{(0)}\rangle S_k(-\omega) \right.$$

$$\left. + S_j(\omega)\langle\check{F}_j^{(0)}(-\omega)^{\dagger}\check{F}_k^{(0)}\rangle T_k(-\omega) + S_j(\omega)\left[\langle\check{F}_k^{(0)}(-\omega)_-^{\dagger}\check{F}_j^{(0)\dagger}\rangle + \langle\check{F}_j^{(0)}(\omega)_-^{\dagger}\check{F}_k^{(0)\dagger}\rangle\right]S_k(-\omega) \right\}. \quad \text{(D9)}$$

If we consider the fluctuation correlation as shown in Eq. (62), we can numerically verify that both of them are completely zero.

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