The steady state population inversion of multiple Ξ-type atoms by the squeezed vacuum in a waveguide

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We study the dynamics of multiple Ξ-type atoms driven by a squeezed vacuum reservoir in a quasi-one-dimensional waveguide. We show that the atomic system's steady state is a pure state, and a complete population inversion can occur when the dipole moment of the second transition is almost perpendicular to the polarization of the incident squeezed light. We also prove that the steady state of the system is the direct product of that in the single-atom case with modified squeezed vacuum even when dipole-dipole interaction is involved. This steady state population inversion may be used to study the two-photon laser or collective atomic effect.

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I. INTRODUCTION

The concept of population inversion is of fundamental importance in laser physics because the population inversion is a key step of generating laser. However, the population inversion can never exist for a system at thermal equilibrium because of the spontaneous emission. The achievement of population inversion therefore requires pushing the system into a non-equilibrated state [1]. Thus, the spontaneous emission must be inhibited in order to maintain the population inversion in a steady state. In 1946, Purcell showed that the spontaneous decay rate of an emitter can be modified by engineering the electromagnetic bath environment with which the emitters interact [2]. One famous example of bath engineering is the squeezed vacuum which leads to many novel effects and techniques in quantum optics and atomic spectroscopy. The reduction of quantum fluctuations below vacuum level by the squeezed vacuum yields many interesting phenomenons, for example, the suppression of dephasing rate in one direction and enhancment in the other for a two-level emitter [3–10], the subnatural linewidth of resonance fluorescence [11, 12], and improvement of an atomic clock using squeezed vacuum [13]. The entanglement nature of the squeezed vacuum also leads to interesting results like pairwise excitation of atomic states [14–16]. In 1993, Ficek and Drummond studied the dynamical properties of a single three-level atom in the squeezed vacuum where they showed that a single three-level atom in the cascade configuration coupled to squeezed modes in a cavity can reach steady state with level population inversion relative to the ordinary laser spectroscopy [17–19]. In their model, they found a population inversion of about 78%.

Here, instead of a cavity, we consider that the case in quasi-one-dimensional waveguide. Squeezing all modes in the three-dimensional (3D) space is technically impractical. In contrast, people have made a great achievement in generating the squeezed vacuum reservoir in the quasi-one-dimensional cavity. Suppression of the radiative decay of atomic coherence and the linewidth of the resonance fluorescence have been experimentally demonstrated in a one-dimensional (1D) microwave transmission line coupled to a single artificial atom [12, 20–24]. Recently, photon transport in a 1D waveguide coupled to quantum emitters (well known as "waveguide-QED") has attracted much attention because it can not only enhance the interaction but also transport information by guiding the photon [25–31]. In these studies, the photon modes are usually considered to be ordinary vacuum modes.

In this paper, we consider multiple Ξ-type atoms coupled to a broadband squeezed vacuum in the quasi-1D waveguide where all resonant modes can be technically squeezed. We show that for a single atom, it can always reach a population inversion of almost 100% or any other ratio as long as the direction of its transition dipole moment is properly set. We also mathematically prove that this result can be generalized to arbitrary number of atoms coupled to each other through dipole-dipole interaction, which may be a scenario for studying two-photon laser or collective atomic effect.

This paper is organized as follows. In Sec. II, we derive the general master equation describing the dynamics of multiple three-level atoms coupled to squeezed vacuum in a 1D waveguide. In Sec. III, we consider the single atom case and study the steady state of the atom. In Sec. IV, we consider the multiple-atom case with modified squeezed vacuum and show that population inversion is also possible in the multiple-atom case. Finally, we summarize the results.

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II. MASTER EQUATION OF THREE-LEVEL ATOMS IN THE SQUEEZED VACUUM

In this section, we consider a scenario where N_a Ξ-type atoms are located inside a perfect rectangular waveguide with the squeezed vacuum injected from both ends, as shown in Fig. 1(a). The atomic electronic structure is shown in Fig. 1(b) where the atomic states are labeled as $|a\rangle$, $|b\rangle$, $|c\rangle$ where $|a\rangle$ is the excited state, $|b\rangle$ is the middle state, and $|c\rangle$ is the ground state. Different from the free space, the square waveguide can only support certain photon modes, i.e., TE_{mn} and TM_{mn} modes with cutoff frequency $c\sqrt{(\frac{m\pi}{a})^2 + (\frac{n\pi}{b})^2}$, where $a \times b$ is the dimension of the waveguide's cross section. For the rectangular waveguide, there is no TM_{01} or TM_{10} mode. Assuming $b < a$, TE_{10} is the ground mode with the lowest cutoff frequency. Thus, the atom is only coupled to the TE_{10} mode as long as $\frac{\pi c}{a} < \omega_{ab}, \omega_{bc} < min(\frac{\pi c}{b}, \frac{2\pi c}{a})$. We assume that $\omega_{ac} = 2\omega_0$ where ω_0 is the center frequency of the broadband squeezed vacuum. We also assume that the bandwidth of the squeezed vacuum reservoir is much larger than $|\omega_{ab} - \omega_{bc}|$ so it can couple to both transitions.

The atom-field system is described by the Hamiltonian

$$H = H_A + H_F + H_{AF} \tag{1}$$

where $H_A = \sum_{l=1}^{N_a} \sum_{e=a,b,c} \hbar\omega_{e,l} |e_l\rangle \langle e_l|$ is the atomic Hamiltonian, and $|e_l\rangle$ is the energy state of the lth atom with energy $\hbar\omega_{e,l}$. The Hamiltonian of the EM field is $H_F = \sum_{\boldsymbol{k}s} \hbar\omega_{\boldsymbol{k}s}(\hat{a}_{\boldsymbol{k}s}^\dagger \hat{a}_{\boldsymbol{k}s} + \frac{1}{2})$ where $\hat{a}_{\boldsymbol{k}s}$ and $\hat{a}_{\boldsymbol{k}s}^\dagger$ are the annihilation and creation operators of the filed mode with wavevector $\boldsymbol{k}$, polarization s (in waveguide, it represents TE_{mn} or TM_{mn}), and frequency $\omega_{\boldsymbol{k},s}$. The inter-

action Hamiltonian in the electric-dipole approximation is $H_{AF} = -i\hbar \sum_{\boldsymbol{k}s} \sum_{i=1,2} \sum_{l=1}^{N_a} [\boldsymbol{\mu}_{l,i} \cdot \boldsymbol{u}_{\boldsymbol{k}s}(\boldsymbol{r}_{l,i}) S_{l,i}^+ \hat{a}_{\boldsymbol{k}s} + \boldsymbol{\mu}_{l,i}^* \cdot \boldsymbol{u}_{\boldsymbol{k}s}(\boldsymbol{r}_{l,i}) S_{l,i}^- \hat{a}_{\boldsymbol{k}s} - H.c.]$ where $\boldsymbol{\mu}_{l,i}$ is the electric dipole moment for the ith transition of the lth atom, where $i = 1$ denotes the transition from $|a\rangle$ to $|b\rangle$, and $i = 2$ denotes the transition from $|b\rangle$ to $|c\rangle$. Here, $S_{l,i}^+$ and $S_{l,i}^-$ are the raising and lowering operator for the transition i of the lth atom. The mode function of the squeezed vacuum in the TE_{10} mode is given by

$$\boldsymbol{u}_{\boldsymbol{k}s}(\boldsymbol{r}_i) = \sqrt{\frac{\omega_{\boldsymbol{k}s}}{2\epsilon_0 \hbar V}} \boldsymbol{x} e^{i\boldsymbol{k}\cdot(\boldsymbol{r}_i - \boldsymbol{o}_{\boldsymbol{k}s})} \tag{2}$$

where $\boldsymbol{o}_{\boldsymbol{k}s}$ is a phenomenological parameter which includes the effects of the initial phase and the position of the squeezing source [16]. The correlation functions for the squeezed vacuum are [32]:

$$\begin{aligned}
\left\langle a_{\boldsymbol{k},s}^\dagger a_{\boldsymbol{k}',s'} \right\rangle &= \sinh^2 r \delta_{\boldsymbol{k}'\boldsymbol{k}} \delta_{ss'} \\
\left\langle a_{\boldsymbol{k},s} a_{\boldsymbol{k}',s'}^\dagger \right\rangle &= \cosh^2 r \delta_{\boldsymbol{k}'\boldsymbol{k}} \delta_{ss'} \\
\left\langle a_{\boldsymbol{k},s}^\dagger a_{\boldsymbol{k}',s'}^\dagger \right\rangle &= -e^{-i\theta} M \delta_{\boldsymbol{k}',2\boldsymbol{k}_0 - \boldsymbol{k}} \delta_{ss'} \\
\left\langle a_{\boldsymbol{k},s} a_{\boldsymbol{k}',s'} \right\rangle &= -e^{i\theta} M \delta_{\boldsymbol{k}',2\boldsymbol{k}_0 - \boldsymbol{k}} \delta_{ss'}
\end{aligned} \tag{3}$$

where r is the squeezing parameter and M is bounded by $M \leq \cosh(r)\sinh(r)$. For simplicity, we can set the squeezing parameter $\theta = 0$, and assume that the dipole moments of all atoms are aligned along the same direction. The dynamics of the atomic system can be described by the following master equation (See Appendix A for details of derivation):

$$\begin{aligned}
\frac{d\rho^S}{dt} = &-i\sum_{ijkl} \Lambda_{ijkl}[S_{i,j}^+ S_{k.l}^-, \rho^S] e^{i(\omega_j - \omega_l)t} - \frac{1}{2}\sum_{ijkl} \gamma_{ijkl}(1+N)(\rho^S S_{i,j}^+ S_{k.l}^- + S_{i,j}^+ S_{k,l}^- \rho^S - 2S_{k.l}^- \rho^S S_{i,j}^+) e^{i(\omega_j - \omega_l)t} \\
&- \frac{1}{2}\sum_{ijkl} \gamma_{ijkl} N(\rho^S S_{i,j}^- S_{k.l}^+ + S_{i,j}^- S_{k,l}^+ \rho^S - 2S_{k,l}^+ \rho^S S_{i,j}^-) e^{-i(\omega_j - \omega_l)t} \\
&- \frac{1}{2}\sum_{\alpha=\pm} \sum_{ijkl} \gamma_{ijkl}' M e^{2\alpha i k_{0z} R} e^{i\alpha(\omega_j + \omega_l - 2\omega_0)t} (\rho^S S_{i,j}^\alpha S_{k.l}^\alpha + S_{i,j}^\alpha S_{k,l}^\alpha \rho^S - 2S_{k,l}^\alpha \rho^S S_{i,j}^\alpha)
\end{aligned} \tag{4}$$

where $N = \sinh(r)^2$, $\omega_1 = \omega_{ab}$, $\omega_2 = \omega_{bc}$, and $\gamma_1(\gamma_2)$ is the decay rate for transition $|a\rangle \to |b\rangle (|b\rangle \to |c\rangle)$ in the ordinary vacuum reservoir. The coefficients in Eq. (4) are

$$\begin{aligned}
\gamma_{ijkl} &= \sqrt{\gamma_j \gamma_l} \cos(k_{0z} r_{ik}) \\
\Lambda_{ijkl} &= \frac{\sqrt{\gamma_j \gamma_l}}{2} \sin(k_{0z} r_{ik}) \\
\gamma_{ijkl}' &= \sqrt{\gamma_j \gamma_l} \cos[k_{0z}(r_i + r_k)]
\end{aligned} \tag{5}$$

where subscripts i, k label the atom index, $j(l)$ labels the

transitions of the $ith(kth)$ atom. Contrary to the free space where the decay rate is independent of the dipole direction, the anisotropic character of the waveguide results in the sensitivity of the atomic decay rate to its dipole direction. Thus, $\gamma_j = \gamma_{jmax} \cos\theta_j$ where γ_{jmax} is the value of γ_j when the transition dipole is parallel to the polarization of the waveguide modes, and $\cos\theta_j$ is the angle between them.
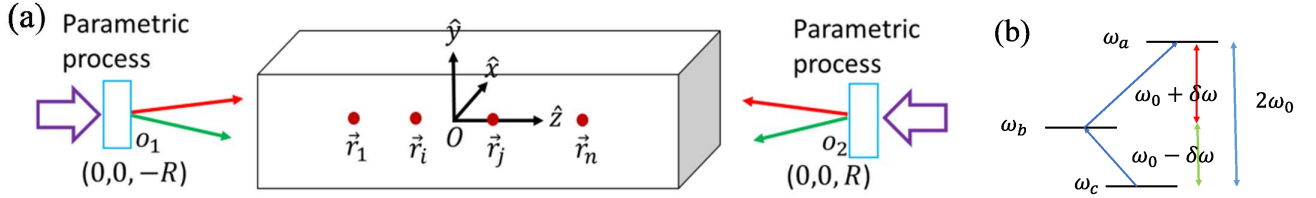
Fig. 1: (a) Schematic setup: A Ξ-type atom is located inside the waveguide with the broadband squeezed vacuum incident from both ends. (b)The energy structure of the three level atom. Transition $|a\rangle \to |c\rangle$ is forbidden and $\omega_{ac} = 2\omega_0$ where ω_0 is the center frequency of the squeezed vacuum. ω_{ab} and ω_{bc} differ by a small amount $2\delta\omega_0$ and they are within the bandwidth of the squeezed vacuum reservoir.

III. STEADY STATE OF A SINGLE ATOM

In this section, we study the steady state of a single atom in the squeezed vacuum reservoir. For a single three level atom, we have $r_i = r_k$, and for simplicity we set $r_i = r_j = 0$ and the resonant condition $\omega_1 + \omega_2 = 2\omega_0$. It follows from Eq. (4) that various matrix elements satisfy the following equation:

$$\dot{\rho}_{aa} = -\gamma_1 ch^2 \rho_{aa} + \gamma_1 sh^2 \rho_{bb} - \frac{1}{2}\sqrt{\gamma_1\gamma_2}M(\rho_{ac} + \rho_{ca})$$

(6a)

$$\dot{\rho}_{bb} = \gamma_1(ch^2\rho_{aa} - sh^2\rho_{bb}) + \gamma_2(sh^2\rho_{cc} - ch^2\rho_{bb}) + \sqrt{\gamma_1\gamma_2}M(\rho_{ac} + \rho_{ca})$$

(6b)

$$\dot{\rho}_{cc} = \gamma_2 ch^2\rho_{bb} - \gamma_2 sh^2\rho_{cc} - \frac{1}{2}\sqrt{\gamma_1\gamma_2}M(\rho_{ac} + \rho_{ca})$$

(6c)

$$\Re[\dot{\rho}_{ac}] = -\frac{1}{2}(\gamma_1 ch^2 + \gamma_2 sh^2)\Re[\rho_{ac}] - \frac{1}{2}\sqrt{\gamma_1\gamma_2}M(\rho_{aa} - 2\rho_{bb} + \rho_{cc})$$

(6d)

$$\Re[e^{i\delta\omega t}\dot{\rho}_{ba}] = -\frac{1}{2}\sqrt{\gamma_1\gamma_2}(M - 2sh^2)sh\Re[e^{i\delta\omega t}\rho_{bc}] - \frac{1}{2}((\gamma_1 + \gamma_2)ch^2 + \gamma_1 sh^2 - \gamma_1 M)\Re[e^{i\delta\omega t}\rho_{ba}]$$

(6e)

$$\Re[e^{i\delta\omega t}\dot{\rho}_{bc}] = \frac{1}{2}\sqrt{\gamma_1\gamma_2}(2ch^2 - M)\Re[(e^{-i\delta\omega t}\rho_{ab}] - \frac{1}{2}((\gamma_1 + \gamma_2)sh^2 + \gamma_2 ch^2 - 2\gamma_2 M)\Re[e^{i\delta\omega t}\rho_{bc}]$$

(6f)

where $\Re$ means real part, $ch = \cosh(r)$, $sh = \sinh(r)$, and $\gamma_1 = \gamma_{ab}(\gamma_2 = \gamma_{bc})$ is the decay rate from $|a\rangle$ to $|b\rangle(|b\rangle$ to $|c\rangle)$ in ordinary vacuum due to the waveguide modes. Equations (6e) and (6f) are for the off-diagonal elements ρ_{ab}, ρ_{bc}. The steady state solution of these two equations is $\rho_{ab} = \rho_{bc} = 0$ because they are homogeneous linear equations. The first four equations Eqs. (6a)-(6d) also have a steady state solution when they are combined with the normalization condition $\rho_{aa} + \rho_{bb} + \rho_{cc} = 1$. It is also

worth noting that Eqs. (6a)-(6d) are independent of $\delta\omega$, so the difference between ω_{ab} and ω_{bc} does not influence the steady state of the single atom case as long as both ω_{ab} and ω_{bc} are within the squeezing bandwidth. Thus, considering the minimum uncertainty squeezed vacuum where $M = \cosh(r)\sinh(r)$, the steady state solution is:

$$\begin{aligned} \rho_{aa} &= \frac{sh^2\gamma_2}{ch^2\gamma_1 + sh^2\gamma_2}, \\ \rho_{cc} &= \frac{ch^2\gamma_1}{ch^2\gamma_1 + sh^2\gamma_2}, \\ \rho_{ac} &= \rho_{ca} = -\frac{chsh\sqrt{\gamma_1\gamma_2}}{ch^2\gamma_1 + sh^2\gamma_2}, \\ \rho_{bb} &= \rho_{ba} = \rho_{bc} = 0, \end{aligned}$$

(7)

which is in fact a pure state of a superposition of $|a\rangle$ and $|c\rangle$

$$|\psi_{ss}\rangle = \frac{sh\sqrt{\gamma_2}}{\sqrt{ch^2\gamma_1 + sh^2\gamma_2}}|a\rangle - \frac{ch\sqrt{\gamma_1}}{\sqrt{ch^2\gamma_1 + sh^2\gamma_2}}|c\rangle. \quad (8)$$

Since there is no population in the state $|b\rangle$, population inversion can always occur between states $|a\rangle$ and $|b\rangle$ in the steady state. If $\tanh r > \sqrt{\frac{\gamma_1}{\gamma_2}}$, population inversion can also occur between the state $|a\rangle$ and $|c\rangle$. This result is similar to the result in Ref. [19]. However, in our scheme, the population inversion can approach 100% with zero population in the ground state and the middile state if $\gamma_2 \gg \gamma_1$. In comparision, the population inversion in the cavity case shown in Ref. [19] is about of 78%.

The steady state population distribution for different ratios of $\frac{\gamma_{ab}}{\gamma_{bc}}$ is shown in Fig. 2(a). The mechanism of this population inversion can be interpreted with the help of Fig. 3. In Fig. 3 we show that the direct transition between $|a\rangle$, $|b\rangle$, and $|c\rangle$ are allowed just like the thermal reservoir case. However, in the squeezed vacuum, there are additional paths for the population flow: atom in any of these three states can evolve into the other two through an intermediate "state" ρ_{ac}. Although ρ_{ac} is an off-diagonal element rather than a state, it can be used to elucidate our idea. When $\gamma_{ab} \ll \gamma_{bc}$, the transition rate for the $|a\rangle \to |b\rangle$ transition is negligible compared to γ_{bc} and $\sqrt{\gamma_{ab}\gamma_{bc}}$. Thus the atom in the state $|c\rangle$ can
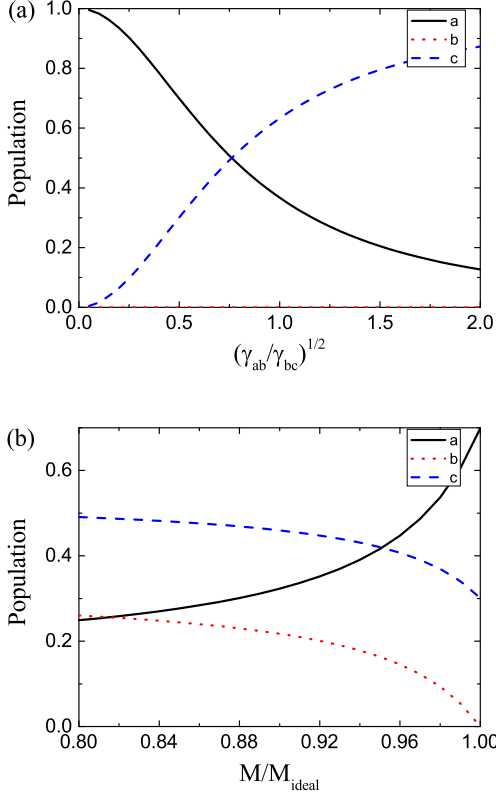
Fig. 3: The allowed population flow in the squeezed vacuum.

Fig. 2: (a) The steady state population distribution for different μ_{ab} and μ_{bc}. The squeezing parameter $r = 1$ and the squeezing is perfect ($M = \sqrt{N(N+1)}$). (b) The steady state population distribution for non-ideal squeezed vacuum which is characterized by the ratio of M and $\sqrt{N(N+1)}$. The squeezing parameter $r = 1$, and $\gamma_{ab} = \frac{1}{4}\gamma_{bc}$.

IV. STEADY STATE OF MULTIPLE ATOMS

In the last section, we demonstrated that arbitrary population inversion can occur for a single Ξ-type atom driven by the squeezed vacuum reservoir. However, with Eq. (7), this result can not be simply generalized to the multi-atom case since $\gamma'_{ijij} = \sqrt{\gamma_j\gamma_j}\cos[2k_{0z}r_i]$, i.e., different atoms have different γ'_{ijij} for the usual case unless all the atoms are perioidically distributed with period $n\lambda/2$. The squeezing term in Eq. (4) vanishes for atoms located around $r_i = \frac{\pi}{4k_{0z}} + \frac{n\pi}{2k_{0z}}$. Thus, for a group of randomly located atoms, if we want to achieve steady state population inversion in the squeezed vacuum, we need to modify our scheme. Here we consider the following correlation functions:

$$\left\langle a^\dagger_{\boldsymbol{k},s} a_{\boldsymbol{k}',s'} \right\rangle = \sinh^2 r \delta_{\boldsymbol{k}'\boldsymbol{k}}\delta_{ss'}$$
$$\left\langle a_{\boldsymbol{k},s} a^\dagger_{\boldsymbol{k}',s'} \right\rangle = \cosh^2 r \delta_{\boldsymbol{k}'\boldsymbol{k}}\delta_{ss'}$$
$$\left\langle a^\dagger_{\boldsymbol{k},s} a^\dagger_{\boldsymbol{k}',s'} \right\rangle = -e^{-i\theta}\cosh(r)\sinh(r)\delta_{\boldsymbol{k}',-(2\boldsymbol{k}_0-\boldsymbol{k})}\delta_{ss'}$$
$$\left\langle a_{\boldsymbol{k},s} a_{\boldsymbol{k}',s'} \right\rangle = -e^{i\theta}\cosh(r)\sinh(r)\delta_{\boldsymbol{k}',-(2\boldsymbol{k}_0-\boldsymbol{k})}\delta_{ss'} \quad (9)$$

which indicates that the photons are entangled with those from the opposite direction. **In principle, we can split the squeezed vacuum into two beams by a trianglar prism and inject them into opposite ends of the waveguide**. Then the coefficients in the master equation shown in Eq. (4) become

$$\gamma_{ijkl} = \sqrt{\gamma_i\gamma_k}\cos(k_{0z}r_{jl}),$$
$$\Lambda_{ijkl} = \frac{\sqrt{\gamma_i\gamma_k}}{2}\sin(k_{0z}r_{jl}), \quad (10)$$
$$\gamma'_{ijkl} = \sqrt{\gamma_i\gamma_k}\cos(k_{0z}r_{jl}).$$

We can see that γ'_{ijij} is now independent of the atomic position because $r_{jj} = 0$. The resulting master equation is the traditionally studied master equation for atoms in squeezed reservoir [14]. The detailed derivation of the coefficients shown in Eq. (9) is given in Appendix B with the phase of the squeezing source included. Based on the

be excited to $|a\rangle$ through $|c\rangle \to |b\rangle \to \rho_{ac} \to |a\rangle$, but $|a\rangle$ can not decay back to $|c\rangle$, which results in the population trapping in the level $|a\rangle$. This phenomenon is similar to the coherent population trapping, but here we achieve the trapping for Ξ structure with the squeezed vacuum reservoir, which cannot be realized with coherent pump due to spontaneous emission. Since it is hard to achieve perfect squeezing with $M = \sqrt{N(N+1)}$ in experiments, we also study the effect of different values of M on the steady state population with parameters $\gamma_{ab} = \frac{1}{4}\gamma_{bc}$ and $r = 1$, which is shown in Fig. 2(b). In general, there is population in all three energy levels. Although the steady state population distribution is very sensitive to the value of M, the population inversion between $|a\rangle$ and $|b\rangle$ still holds for $M = 0.8\sqrt{N(N+1)}$. Only when M is larger than 0.95 can the population inversion occur between the state $|a\rangle$ and the state $|c\rangle$.
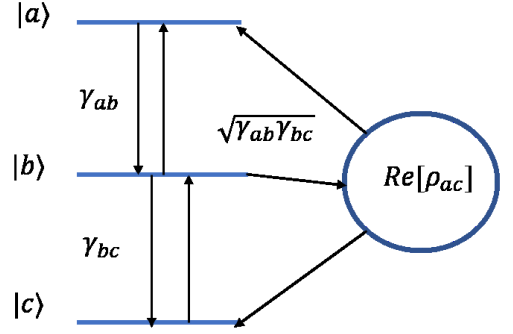
master equation in Eq. (4) with coefficients given by Eq. (9), we can show that a single atom can reach population inversion anywhere in the waveguide, as discussed in Sec. III. When there are multiple atoms in the waveguide where the dipole-dipole interaction should be considered, our calculation shows that the population inversion can still occur for all the atoms. In fact, it is very interesting that the final state of the multiple-atom case is just the direct product of the steady state of independent atoms despite of the dipole-dipole interaction. This result can be proved by the mathematical induction.

Considering the fact that $|\omega_1 - \omega_2| \gg \gamma_i$, it is reasonable to apply the secular approximation on Eq. (4) such that those terms with $e^{\pm i(\omega_1 - \omega_2)t}$ and $e^{\pm i(2\omega_i - 2\omega_0)t}$ are dropped and the master equation is then given by

$$\frac{d\rho^S}{dt} = -i\sum_{i,k,j}\Lambda_{ijkl}[S_{i,j}^+ S_{k,j}^-, \rho^S]$$
$$-\frac{1}{2}\sum_{i,j,k}\gamma_{ijkl}(1+N)\Big(\{\rho^S, S_{i,j}^+ S_{k,j}^-\} - 2S_{k,j}^-\rho^S S_{i,j}^+\Big)$$
$$-\frac{1}{2}\sum_{i,j,k}\gamma_{ijkl}N\Big(\{\rho^S, S_{i,j}^- S_{k,j}^+\} - 2S_{k,j}^+\rho^S S_{i,j}^-\Big)$$
$$-\frac{1}{2}\sum_{\alpha=\pm}\sum_{i,k,j\neq l}\gamma'_{ijkl}M\Big(\{\rho^S, S_{i,j}^\alpha S_{k,l}^\alpha\} - 2S_{k,l}^\alpha\rho^S S_{i,j}^\alpha\Big)$$

$$(11)$$

In the following, we use mathemtics induction to prove that steady state of this system is direct product of the steady state of a single atom. Assume that the steady state of N-atom system is $\rho^S = \rho_1\rho_2...\rho_N$ where $\rho_i = (A|a_i\rangle + C|c_i\rangle)(A\langle a_i| + C\langle c_i|)$ and $A = \frac{sh\sqrt{\gamma_2}}{\sqrt{ch^2\gamma_1 + sh^2\gamma_2}}, C = -\frac{ch\sqrt{\gamma_1}}{\sqrt{ch^2\gamma_1 + sh^2\gamma_2}}$. Then for $(N+1)$-atom case, the extra terms induced by the $(N+1)th$ atom on the right hand side of Eq. (11) are composed of three parts: $i = k = N+1$ terms, $i = N+1, k = 1, 2, \cdots, N$ terms, and $i = 1, 2, \cdots, N, k = N+1$ terms. The $i = k = N+1$ terms are the exact terms for the the $(N+1)th$ atom as a single independent atom, so the net result of this term is 0. The terms with $i = N+1, k = 1, 2, \cdots, N$ are

$$-i\sum_{j,k}\Lambda_{N+1,j,k,j}[S_{N+1,j}^+ S_{k,j}^-, \rho^S]$$
$$-\frac{1}{2}\sum_{j,k}\gamma_{N+1,j,k,j}ch^2\Big(\{\rho^S, S_{N+1,j}^+ S_{k,j}^-\} - 2S_{k,j}^-\rho^S S_{N+1,j}^+\Big)$$
$$-\frac{1}{2}\sum_{j,k}\gamma_{N+1,j,k,j}sh^2\Big(\{\rho^S, S_{N+1,j}^- S_{k,j}^+\} - 2S_{k,j}^+\rho^S S_{N+1,j}^-\Big)$$
$$-\sum_{\alpha=\pm}\sum_{i,k,j\neq l}\gamma'_{N+1,j,k,j}\frac{M}{2}\Big(\{\rho^S, S_{N+1,j}^\alpha S_{k,l}^\alpha\} - 2S_{k,l}^\alpha\rho^S S_{N+1,j}^\alpha\Big).$$

$$(12)$$

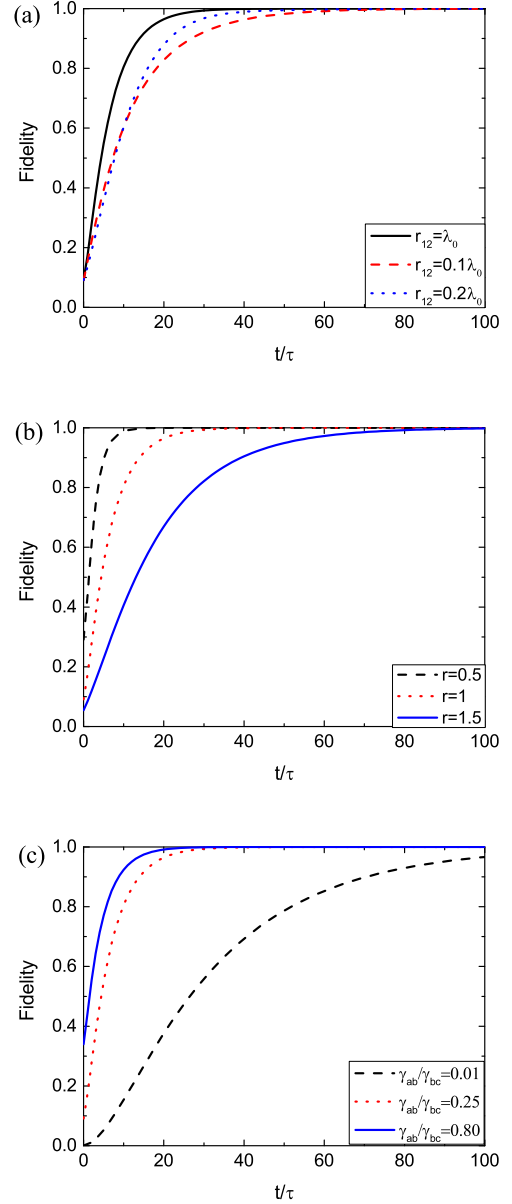For the energy shift term (the first term) in expression

Fig. 4: (a)Fidelity evolution with different atomic separations. The atomic separations of $\lambda_0, 0.1\lambda_0, 0.2\lambda_0$ are plotted. Squeezing parameter $r = 1$, decay rate $\frac{\gamma_1}{\gamma_2} = \frac{1}{4}$ and time unit $\tau = 1/\sqrt{\gamma_{ab}\gamma_{bc}}$ is the geometric mean of the transition $|a\rangle \to |b\rangle$ and $|b\rangle \to |c\rangle$'s spontaneous emission rates in ordinary vacuum. (b) Fidelity evolution with different squeezing parameters. Decay rate $\frac{\gamma_1}{\gamma_2} = \frac{1}{4}$, and atomic separation $r_{12} = \lambda_0$. (c) Fidelity evolution with different decay rates. Squeezing parameter $r = 1$, and atomic separation $r_{12} = \lambda_0$.

(12), we have

$$S_{N+1,j}^+ S_{k,j}^-\rho^S = \rho_1...(S_{k,j}^-\rho_k)...\rho_N(S_{N+1,j}^+\rho_{N+1}) = 0$$
$$\rho^S S_{N+1,j}^+ S_{k,l}^- = \rho_1...(\rho_k S_{k,l}^-)...\rho_N(\rho_{N+1}S_{N+1,j}^+) = 0$$

$$(13)$$

For the thermal terms (the second and third terms) in expression (12), we have

$$\rho^S S^+_{N+1,j} S^-_{k,j} = S^+_{N+1,j} S^-_{k,j} \rho^S = 0$$
$$\rho^S S^-_{N+1,j} S^+_{k,j} = S^-_{N+1,j} S^+_{k,j} \rho^S = 0$$
$$S^-_{k,j} \rho^S S^+_{N+1,j} = \rho_1...(S^-_{k,j}\rho_k)...\rho_N(\rho_{N+1}S^+_{N+1,j})$$
$$= \rho_1...(S^-_{k,1}\rho_k)...\rho_N(\rho_{N+1}S^+_{N+1,1})$$
$$= \rho_1...(A|b_k\rangle)(A\langle a_k| + C\langle c_k|)..\rho_N$$
$$\otimes (A|a_{N+1}\rangle + C|c_{N+1}\rangle)(A\langle b_{N+1}|)$$
$$S^+_{k,j} \rho^S S^-_{N+1,j} = \rho_1...(S^+_{k,j}\rho_k)...\rho_N(\rho_{N+1}S^-_{N+1,j})$$
$$= \rho_1...(S^+_{k,2}\rho_k)...\rho_N(\rho_{N+1}S^-_{N+1,2})$$
$$= \rho_1...(C|b_k\rangle)(A\langle a_k| + C\langle c_k|)...\rho_N$$
$$\otimes (A|a_{N+1}\rangle + C|c_{N+1}\rangle)(C\langle b_{N+1}|) \quad (14)$$

For the squeezed vacuum terms(the fourth term), we

have

$$\rho^S S^\alpha_{N+1,j} S^\alpha_{k,l} = S^\alpha_{N+1,j} S^\alpha_{k,l} \rho^S = 0$$
$$S^+_{k,1} \rho^S S^+_{N+1,2} = \rho_1...(S^+_{k,1}\rho_k)...\rho_N(\rho_{N+1}S^+_{N+1,2}) = 0$$
$$S^+_{k,2} \rho^S S^+_{N+1,1} = \rho_1...(S^+_{k,2}\rho_k)...\rho_N(\rho_{N+1}S^+_{N+1,1})$$
$$= \rho_1...(C|b_k\rangle)(A\langle a_k| + C\langle c_k|)...\rho_N$$
$$\otimes (A|a_{N+1}\rangle + C|c_{N+1}\rangle)(A\langle b_{N+1}|),$$
$$S^-_{k,1} \rho^S S^-_{N+1,2} = \rho_1...(S^-_{k,1}\rho_k)...\rho_N(\rho_{N+1}S^-_{N+1,2})$$
$$= \rho_1...(A|b_k\rangle)(A\langle a_k| + C\langle c_k|)...\rho_N$$
$$\otimes (A|a_{N+1}\rangle + C|c_{N+1}\rangle)(C\langle b_{N+1}|)$$
$$S^-_{k,2} \rho^S S^-_{N+1,1} = \rho_1...(S^-_{k,2}\rho_k)...\rho_N(\rho_{N+1}S^-_{N+1,1}) = 0 \quad (15)$$

On substituting from Eqs. (13)-(15) into expression (12), we have

$$\sum_k \gamma_{N+1,1,k,1}(ch^2)S^-_{k,1}\rho^S S^+_{N+1,1} + \sum_{j,k} \gamma_{N+1,2,k,2}sh^2 S^+_{k,2}\rho^S S^-_{N+1,2} + \sum_{\alpha=\pm}\sum_{i,k,j\neq l} \gamma'_{N+1,j,k,l}chsh S^\alpha_{k,l}\rho^S S^\alpha_{N+1,j}$$
$$=\sum_k \gamma_{N+1,1,k,1}(ch^2)\rho_1...(A|b_k\rangle)(A\langle a_k| + C\langle c_k|)..\rho_N(A|a_{N+1}\rangle + C|c_{N+1}\rangle)(A\langle b_{N+1}|)$$
$$+ \sum_k \gamma_{N+1,2,k,2}sh^2 \rho_1...(C|b_k\rangle)(A\langle a_k| + C\langle c_k|)...\rho_N(A|a_{N+1}\rangle + C|c_{N+1}\rangle)(C\langle b_{N+1}|)$$
$$+ \sum_k \gamma'_{N+1,j,k,l}chsh[\rho_1...(C|b_k\rangle)(A\langle a_k| + C\langle c_k|)...\rho_N(A|a_{N+1}\rangle + C|c_{N+1}\rangle)(A\langle b_{N+1}|)$$
$$+ \rho_1...(A|b_k\rangle)(A\langle a_k| + C\langle c_k|)...\rho_N(A|a_{N+1}\rangle + C|c_{N+1}\rangle)(C\langle b_{N+1}|)]$$
$$=\sum_k (\gamma_{N+1,1,k,1}ch^2 A^2 + \gamma_{N+1,2,k,2}sh^2 C^2 + 2\gamma'_{N+1,j,k,l}chshCA)$$
$$\times \rho_1...(|b_k\rangle)(A\langle a_k| + C\langle c_k|)..\rho_N(A|a_{N+1}\rangle + C|c_{N+1}\rangle)(\langle b_{N+1}|).$$

$$(16)$$

It is not difficutlt to prove that $ch^2 A^2 \gamma_{N+1,1,k,1} + sh^2 C^2 \gamma_{N+1,2,k,2} + 2chshCA\gamma'_{N+1,j,k,l} = 0$ by substituting the expressions of A, B, C and Eq. (10). Hence, the extra terms with the atom index $i = N+1$ and $k = 1 \sim N$ when we add the $N+1$th atom are 0. Similarly, the terms with $i = 1 \sim N$ and $k = N+1$ also vanish. Thus, we prove that the right hand side of Eq. (11) is zero when the state of the system is direct product of the steady state of single atom. This indicates that direct product of the steady state of single atom is the steady state of the multiple atoms driven by the squeezed vacuum shown in Eq. (9). It is interesting to note that while introducing the dipole-dipole interaction between the atoms affects the evolution of the system, the final steady state still remains unaffected. Therefore, for mul-

tiple atoms, a population inversion of almost 100% can also be achieved even the dipole-dipole interaction is considered, under the condition that the dipole direction for all atoms are properly oriented to satisfy $\gamma_{ab} \ll \gamma_{bc}$. Actually, in the normal squeezed vacuum with correlation shown in Eq. (3), the steady state of the atoms can also be direct product of the steady state of a single atom if all the atoms are in the nodes of the standing wave. In this case, the coefficients in the master equation are the same as those shown in Eq. (10).

We also numerically show that the steady state of multiple atoms is indeed the direct product of steady state of single atom. Since the cost for numerical simulation increases exponentially as the number of atoms increases, we only show the fidelity of two-atom state with respect to theoretical steady state as a function

of time in Fig. 4, where the system is initially in the ground state. **From Fig. 4(a), we can see that different atom separations have different evolution dynamics because they have different dipole-dipole interactions.** However, we can see that the system finally evolves into the following equation regardless of the atomic separation, squeezing parameter, and the ratio of decay rate γ_{ab}/γ_{bc}:

$$|\psi_s s\rangle = \left(\frac{sh\sqrt{\gamma_2}}{\sqrt{ch^2\gamma_1 + sh^2\gamma_2}}|a_1\rangle - \frac{ch\sqrt{\gamma_1}}{\sqrt{ch^2\gamma_1 + sh^2\gamma_2}}|c_1\rangle \right)$$
$$\otimes \left(\frac{sh\sqrt{\gamma_2}}{\sqrt{ch^2\gamma_1 + sh^2\gamma_2}}|a_2\rangle - \frac{ch\sqrt{\gamma_1}}{\sqrt{ch^2\gamma_1 + sh^2\gamma_2}}|c_2\rangle \right)$$
$$(17)$$

From Fig. 4(b), we see that the system takes less time to evolve into the steady state for a smaller squeezing parameter. Fig. 4(c) shows that while smaller γ_{ab} results in higher population inversion, it takes much longer for the system to evolve into the steady state.

V. SUMMARY

We studied the Ξ-type atoms coupled to a broadband squeezed vacuum reservoir in a quasi-one-dimensional waveguide, with the overall transition frequency $\omega_{ac} = 2\omega_0$. We showed that a single atom evolves into a steady state which is a superposition of the second excited state and the ground state. If the decay rate from the second excited state to the first excited state is much smaller than that from the first excited state to the ground state, the population can be almost 100% trapped in the second excited state, which is a great improvement compared to the maximum ratio of 78% in Ref. [19]. What is more, we proved that the above result can be generalized to an arbitrary number of atoms interacting with each other via dipole-dipole interaction, and the system's final steady state is a direct product of that in the single-atom case with modified squeezed vacuum shown in Eq. (9). **This is one of the most interesting results here and its physical insight still needs further studies. We also argued that the arbitrary ratio of the two transitions' decay rates can be effectively controlled by different waveguide structure. This population-inversed system is experimentally feasible since the experiments on the broadband squeezed vacuum coupled to the artificial atom in a 1D cavity have been widely conducted[12, 20–24].**

VI. ACKNOWLEDGEMENT

APPENDIX A: DERIVATION OF EQ. (4)

Here we show how to derive the master equation Eq. (4). The interaction Hamiltonian is:

$$V(t) = -i\hbar \sum_{\boldsymbol{k}s} [D(t)a_{\boldsymbol{k}s}(t) - D^+(t)a_{\boldsymbol{k}s}^\dagger(t)], \tag{A1}$$

where

$$D(t) = \sum_{l,i} [\boldsymbol{\mu}_{l,i} \cdot \boldsymbol{u}_{\boldsymbol{k},s}(r_{l,i})S_{l,i}^\dagger(t) + \boldsymbol{\mu}_{l,i}^* \cdot \boldsymbol{u}_{\boldsymbol{k},s}(r_{l,i})S_{l,i}^-(t)] \tag{A2}$$

The reduced master equation of atoms in the reservoir is:

$$\begin{aligned}
\frac{d\rho^S}{dt} = & -\frac{1}{\hbar^2} \int_0^t d\tau Tr_F\{[V(t),[V(t-\tau),\rho^S(t-\tau)\rho^F\} \\
= & -\frac{1}{\hbar^2} \int_0^t d\tau Tr_F\{V(t)V(t-\tau)\rho^S(t-\tau)\rho^F + \rho^S(t-\tau)\rho^F V(t-\tau)V(t) \\
& - V(t)\rho^S(t-\tau)\rho^F V(t-\tau) - V(t-\tau)\rho^S(t-\tau)\rho^F V(t)\}
\end{aligned} \tag{A3}$$

Here we just show how to deal with the first term in Eq. (A3), the remaining terms can be calculated in the same way. For the first term, we have

$$-\frac{1}{\hbar^2}\int_0^t d\tau Tr_F\{V(t)V(t-\tau)\rho^S(t-\tau)\rho^F\}$$

$$=\int_0^t d\tau \sum_{\bm{ks},\bm{k's'}}\{D(t)D(t-\tau)Tr_F[\rho^F a_{ks}(t)a_{k's'}(t-\tau)]-D(t)D^+(t-\tau)Tr_F[\rho^F a_{ks}(t)a_{k's'}^\dagger(t-\tau)]$$

$$-D^+(t)D(t-\tau)Tr_F[\rho^F a_{ks}^\dagger(t)a_{k's'}(t-\tau)]+D^+(t)D^+(t-\tau)Tr_F[\rho^F a_{ks}^\dagger(t)a_{k's'}^\dagger(t-\tau)]\}\rho^S(t-\tau)\}. \quad (A4)$$

Under the rotating wave approximation(RWA), we have

$$-\frac{1}{\hbar^2}\int_0^t d\tau Tr_F\{V(t)V(t-\tau)\rho^S(t-\tau)\rho^F\}$$

$$=\sum_{ijlm}\sum_{\bm{ks},\bm{k's'}}\int_0^t d\tau\{\bm{\mu}_{l,i}\cdot\bm{u}_{\bm{ks}}(r_{l,i})S_{l,i}^+e^{i\omega_i t}\bm{\mu}_{m,j}\cdot\bm{u}_{\bm{k's'}}(r_{m,j})S_{m,j}^+e^{i\omega_j(t-\tau)}e^{-i(\omega_{\bm{ks}}+\omega_{\bm{k's'}})t+i\omega_{\bm{k's'}}\tau}[-M\delta_{\bm{k'},2\bm{k}_0-\bm{k}}\delta_{ss'}]$$

$$-\bm{\mu}_{l,i}\cdot\bm{u}_{\bm{ks}}(r_{l,i})S_{l,i}^+e^{i\omega_i t}\bm{\mu}_{m,j}^*\cdot\bm{u}_{\bm{k's'}}^*(r_{m,j})S_{m,j}^-e^{-i\omega_j(t-\tau)}e^{-i\omega_{\bm{k's'}}\tau}\cosh^2 r\delta_{\bm{kk'}}\delta_{ss'}$$

$$-\bm{\mu}_{l,i}^*\cdot\bm{u}_{\bm{ks}}(r_{l,i})S_{l,i}^-e^{-i\omega_i t}\bm{\mu}_{m,j}\cdot\bm{u}_{\bm{k's'}}^*(r_{m,j})S_{m,j}^+e^{i\omega_j(t-\tau)}e^{-i\omega_{\bm{k's'}}\tau}\cosh^2 r\delta_{\bm{kk'}}\delta_{ss'}$$

$$-\bm{\mu}_{l,i}^*\cdot\bm{u}_{\bm{ks}}^*(r_{l,i})S_{l,i}^-e^{-i\omega_i t}\bm{\mu}_{m,j}\cdot\bm{u}_{\bm{k's'}}(r_{m,j})S_{m,j}^+e^{i\omega_j(t-\tau)}e^{i\omega_{\bm{k's'}}\tau}\sinh^2 r\delta_{\bm{kk'}}\delta_{ss'}$$

$$-\bm{\mu}_{l,i}\cdot\bm{u}_{\bm{ks}}^*(r_{l,i})S_{l,i}^+e^{i\omega_i t}\bm{\mu}_{m,j}^*\cdot\bm{u}_{\bm{k's'}}(r_{m,j})S_{m,j}^-e^{-i\omega_j(t-\tau)}e^{i\omega_{\bm{k's'}}\tau}\sinh^2 r\delta_{\bm{kk'}}\delta_{ss'}$$

$$+\bm{\mu}_{l,i}^*\cdot\bm{u}_{\bm{ks}}^*(r_{l,i})S_{l,i}^-e^{-i\omega_i t}\bm{\mu}_{m,j}^*\cdot\bm{u}_{\bm{k's'}}^*(r_{m,j})S_{m,j}^-e^{-i\omega_j(t-\tau)}e^{i(\omega_{\bm{ks}}+\omega_{\bm{k's'}})t-i\omega_{\bm{k's'}}\tau}[-M\delta_{\bm{k'},2\bm{k}_0-\bm{k}}\delta_{ss'}]\}\rho^S(t-\tau)$$

$$(A5)$$

where l,m are used for labeling different atoms, and i,j are used for transitions within an atom. Here we just calculate the first and second term to show how to get the master equation Eq. (4). Since all atoms are identical, $\omega_{l,i}=\omega_i$, $|\bm{\mu}_{l,i}|=|\bm{\mu}_i|$, and $r_{l,i}=r_l$ can be used to simplify Eq. (A5). For simplicity, we define μ_j to be the projection of $\bm{\mu_j}$ on the x axis. For the second term(thermal term), we have

$$-\sum_{k_z}\int_0^t d\tau\bm{\mu}_{l,i}\cdot\bm{u}_{\bm{ks}}(r_l)S_{l,i}^+e^{i\omega_i t}\bm{\mu}_{m,j}^*\cdot\bm{u}_{\bm{k's'}}^*(r_m)S_{m,j}^-e^{-i\omega_j(t-\tau)}e^{-i\omega_{\bm{k's'}}\tau}\cosh^2 r\rho^S(t-\tau)\delta_{\bm{kk'}}\delta_{ss'}$$

$$=-\frac{L}{2\pi}e^{i(\omega_i-\omega_j)t}\int_{-\infty}^\infty dk_z\int_0^t d\tau e^{i\omega_j\tau}e^{-i\omega_{k_z}\tau}\frac{\omega_k\mu_i\mu_j}{\epsilon_0 LS\hbar}e^{ik_z(r_l-r_m)}\cosh^2 rS_{l,i}^+S_{m,j}^-\rho^S(t-\tau)$$

$$\approx-\frac{L}{2\pi}e^{i(\omega_i-\omega_j)t}\int_0^\infty dk_z\int_0^t d\tau e^{i\omega_j\tau}e^{-i[\omega_j+c^2k_{jz}(k_z-k_{jz})/\omega_j]\tau}\frac{\omega_k\mu_i\mu_j}{\epsilon_0 LS\hbar}[e^{ik_z(r_l-r_m)}+e^{-ik_z(r_l-r_m)}]\cosh^2 rS_{l,i}^+S_{m,j}^-\rho^S(t-\tau)$$

$$\approx-\frac{L}{2\pi}e^{i(\omega_i-\omega_j)t}\int_{-k_{0z}}^\infty d\delta k_z\int_0^t d\tau e^{-i\tau c^2k_{jz}\delta k_z/\omega_j}\frac{\omega_k\mu_i\mu_j}{\epsilon_0 LS\hbar}[e^{i(k_{jz}+\delta k_z)(r_l-r_m)}+e^{-i(k_{jz}+\delta k_z)(r_l-r_m)}]\cosh^2 rS_{l,i}^+S_{m,j}^-\rho^S(t-\tau)$$

$$\approx-\frac{L}{2\pi}e^{i(\omega_i-\omega_j)t}\int_{-\infty}^\infty d\delta k_z\int_0^t d\tau e^{-i(c^2k_{jz}\delta k_z/\omega_j)\tau}\frac{\omega_k\mu_i\mu_j}{\epsilon_0 LS\hbar}[e^{i(k_{jz}+\delta k_z)(r_l-r_m)}+e^{-i(k_{jz}+\delta k_z)(r_l-r_m)}]\cosh^2 rS_{l,i}^+S_{m,j}^-\rho^S(t-\tau)$$

$$\approx-\frac{L}{2\pi}e^{i(\omega_i-\omega_j)t}\int_0^t d\tau\frac{\omega_j\mu_i\mu_j}{\epsilon_0 LS\hbar}2\pi[e^{ik_{jz}(r_l-r_m)}\delta((r_l-r_m)-\frac{c^2k_{jz}}{\omega_0}\tau)+e^{-ik_{jz}(r_l-r_m)}\delta((r_l-r_m)+\frac{c^2k_{jz}}{\omega_0}\tau)]$$

$$\times\cosh^2 rS_{l,i}^+S_{m,j}^-\rho^S(t-\tau)$$

$$\approx-\frac{L}{2\pi}e^{ik_{jz}r_{lm}}\frac{\omega_j\mu_i\mu_j}{\epsilon_0 LS\hbar}2\pi\frac{\omega_j}{c^2k_{0z}}\cosh^2 rS_{l,i}^+S_{m,j}^-\rho^S(t)e^{i(\omega_i-\omega_j)t}$$

$$\approx-[\frac{\sqrt{\gamma_i\gamma_j}}{2}\cos(k_{0z}r_{lm})+i\frac{\sqrt{\gamma_i\gamma_j}}{2}sin(k_{0z}r_{lm})]\cosh^2 rS_{l,i}^+S_{m,j}^-\rho^S(t)e^{i(\omega_i-\omega_j)t}$$

$$(A6)$$

where emitter separation $r_{lm}=|r_l-r_m|$, collective decay rate $\gamma_i=2\mu_i^2\omega_i^2/\hbar\epsilon_0 Sc^2k_{iz}$, and collective energy shift $\Lambda_{ij}=\sqrt{\gamma_i\gamma_j}\sin(k_{0z}r_{ij})/2$. In the third line we expand $\omega_k=c\sqrt{(\frac{\pi}{a})^2+(k_z)^2}$ around $k_z=k_{0z}$ since resonant modes provide dominant contributions. In the fifth line we extend the integration $\int_{-k_{jz}}^\infty dk_z\to\int_{-\infty}^\infty dk_z$ because the main

contribution comes from the components around $\delta k_z = 0$. In the next line, Weisskopf-Wigner approximation is used. Thus, we have obtained γ_{ij} and Λ_{ij} as is shown in Eq. (7).

Next we need to calculate the first term (squeezing term) in Eq. (A5), putting aside the overall factor $e^{i(\omega_i+\omega_j-2\omega_0)t}$, we have:

$$\sum_{k_z}\int_0^t d\tau\{\boldsymbol{\mu}_{l,i}\cdot\boldsymbol{u}_{2\boldsymbol{k}_0-\boldsymbol{k}}(r_l)S_{l,i}^+\boldsymbol{\mu}_{m,j}\cdot\boldsymbol{u}_{\boldsymbol{k}}(r_m)S_{m,j}^+e^{i(\omega_{\boldsymbol{k}}-\omega_j)\tau}(-M)\rho^S(t-\tau)$$

$$=-\frac{L}{2\pi}\int_0^{2k_{0z}}dk_z\int_0^t d\tau e^{i(\omega_{k_z}-\omega_j)\tau}e^{i(2k_{jz}-k_z)(r_l-o_1)}e^{ik_z(r_m-o_1)}\frac{\sqrt{\omega_{k_z}\omega_{2k_{0z}-k_z}}\mu_i\mu_j}{\epsilon_0 LS\hbar}MS_{l,i}^+S_{m,j}^+\rho^S(t-\tau) \qquad (A7)$$

$$-\frac{L}{2\pi}\int_{-2k_{0z}}^0 dk_z\int_0^t d\tau e^{i(\omega_{k_z}-\omega_j)\tau}e^{i(-2k_{jz}-k_z)(r_l-o_2)}e^{ik_z(r_m-o_2)}\frac{\sqrt{\omega_{k_z}\omega_{-2k_{0z}-k_z}}\mu_i\mu_j}{\epsilon_0 LS\hbar}MS_{l,i}^+S_{m,j}^+\rho^S(t-\tau)$$

For terms with $r_l = r_j$, Eq. (A7) reduces to

$$\sum_{k_z}\int_0^t d\tau\{\boldsymbol{\mu}_{l,i}\cdot\boldsymbol{u}_{2\boldsymbol{k}_0-\boldsymbol{k}}(r_l)S_{l,i}^+\boldsymbol{\mu}_{l,j}\cdot\boldsymbol{u}_{\boldsymbol{k}}(r_l)S_{l,j}^+e^{i(\omega_{\boldsymbol{k}}-\omega_j)\tau}(-M)\rho^S(t-\tau)$$

$$=-\frac{L}{2\pi}\int_0^{2k_{0z}}dk_z\int_0^t d\tau e^{i\frac{c^2 k_{jz}}{\omega_j}(k_z-k_{jz})\tau}e^{i2k_{0z}(r_l-o_1)}\frac{\sqrt{\omega_{k_z}\omega_{2k_{0z}-k_z}}\mu_i\mu_j}{\epsilon_0 LS\hbar}MS_{l,i}^+S_{l,j}^+\rho^S(t-\tau)$$

$$-\frac{L}{2\pi}\int_{-2k_{0z}}^0 dk_z\int_0^t d\tau e^{i\frac{c^2 k_{jz}}{\omega_j}(k_z-k_{jz})\tau}e^{-i2k_{0z}(r_l-o_2)}\frac{\sqrt{\omega_{k_z}\omega_{-2k_{0z}-k_z}}\mu_i\mu_j}{\epsilon_0 LS\hbar}MS_{l,i}^+S_{l,j}^+\rho^S(t-\tau) \qquad (A8)$$

$$=-\frac{L}{2\pi}[e^{i2k_{0z}(r_l-o_1)}+e^{-i2k_{0z}(r_l-o_2)}]\frac{\sqrt{\omega_i\omega_j}\mu_i\mu_j}{\epsilon_0 LS\hbar}\int_0^t d\tau 2\pi\delta(\frac{c^2 k_{jz}}{\omega_j}\tau)MS_{l,i}^+S_{l,j}^+\rho^S(t-\tau)$$

$$=-e^{i2k_{jz}R}\frac{\omega_0^2\mu_i\mu_j}{\epsilon_0\hbar Sc^2 k_{0z}}\cos(2k_{0z}r_l)MS_{l,i}^+S_{l,j}^+\rho^S(t)$$

$$=-e^{i2k_{0z}R}\frac{\sqrt{\gamma_i\gamma_j}}{2}\cos(2k_{0z}r_l)MS_{l,i}^+S_{l,j}^+\rho^S(t)$$

where we have used the fact that the origin of coordinate system is at equal distant from two sources(i.e., $o_2 = -o_1 = R$) in the second last line. Incorporating index l into i, we have $\gamma_{ij}' = \sqrt{\gamma_i\gamma_j}\cos(2k_{0z}r_i)$. For $r_i \neq r_j$, Eq. (A7) reduces to

$$\sum_{k_z}\int_0^t d\tau\{\boldsymbol{\mu}_{l,i}\cdot\boldsymbol{u}_{2\boldsymbol{k}_0-\boldsymbol{k}}(r_l)S_{l,i}^+\boldsymbol{\mu}_{m,j}\cdot\boldsymbol{u}_{\boldsymbol{k}}(r_m)S_{m,j}^+e^{i(\omega_{\boldsymbol{k}}-\omega_j)\tau}(-M)\rho^S(t-\tau)$$

$$=-\frac{L}{2\pi}\int_0^{2k_{0z}}dk_z\int_0^t d\tau e^{i\frac{c^2 k_{jz}}{\omega_j}(k_z-k_{jz})\tau}e^{i2k_{0z}(r_c-o_1)}e^{-i(k_z-k_{0z})(r_l-r_m)}\frac{\sqrt{\omega_{k_z}\omega_{2k_{0z}-k_z}}\mu_i\mu_j}{\epsilon_0 LS\hbar}MS_{l,i}^+S_{m,j}^+\rho^S(t-\tau)$$

$$-\frac{L}{2\pi}\int_{-2k_{0z}}^0 dk_z\int_0^t d\tau e^{i\frac{c^2 k_{jz}}{\omega_j}(-k_z-k_{jz})\tau}e^{-i2k_{0z}(r_c-o_2)}e^{-i(k_z+k_{0z})(r_l-r_m)}\frac{\sqrt{\omega_{k_z}\omega_{-2k_{0z}-k_z}}\mu_i\mu_j}{\epsilon_0 LS\hbar}MS_{l,i}^+S_{m,j}^+\rho^S(t-\tau)$$

$$=-\frac{L}{2\pi}e^{i2k_{0z}(r_c-o_1)}\frac{\sqrt{\omega_i\omega_j}\mu_i\mu_j}{\epsilon_0 LS\hbar}\int_{-\infty}^{\infty}dk_z\int_0^t d\tau e^{i\frac{c^2 k_{jz}}{\omega_j}(k_z-k_{jz})\tau}e^{-i(k_z-k_{0z})(r_l-r_m)}MS_{l,i}^+S_{m,j}^+\rho^S(t-\tau)$$

$$-\frac{L}{2\pi}e^{-i2k_{0z}(r_c-o_2)}\frac{\sqrt{\omega_i\omega_j}\mu_i\mu_j}{\epsilon_0 LS\hbar}\int_{-\infty}^{\infty}dk_z\int_0^t d\tau e^{i\frac{c^2 k_{jz}}{\omega_j}(k_z-k_{jz})\tau}e^{i(k_z-k_{0z})(r_l-r_m)}MS_{l,i}^+S_{m,j}^+\rho^S(t-\tau)$$

$$\approx-\frac{L}{2\pi}e^{i2k_{0z}R}\frac{\omega_0^2\mu_i\mu_j}{\epsilon_0 LS\hbar}\int_0^t d\tau 2\pi[e^{i2k_{0z}r_c}\delta(r_l-r_m-\frac{c^2 k_{0z}}{\omega_0}\tau)+e^{-i2k_{0z}r_c}\delta(r_l-r_m+\frac{c^2 k_{0z}}{\omega_0}\tau)]MS_{l,i}^+S_{m,j}^+\rho^S(t-\tau)$$

$$\approx-e^{i2k_{0z}R}\frac{\omega_0^2\mu_i\mu_j}{\epsilon_0\hbar Sc^2 k_{0z}}e^{i2k_{0z}r_c sgn(r_l-r_m)}S_{l,i}^+S_{m,j}^+\rho^S(t)\to-\frac{\sqrt{\gamma_i\gamma_j}}{2}e^{i2k_{0z}R}\cos(k_{0z}(r_l+r_m))S_{l,i}^+S_{m,j}^+\rho^S(t)$$

$$(A9)$$

where $sgn(r_l - r_m)$ is the sign function. The last arrow is because we need to sum over i, j, so the imaginary part of $e^{i2k_{0z}r_c sgn(i-j)}$ vanishes, so the neat result is that $\gamma_{ijkl}' = e^{i2k_{0z}R}\sqrt{\gamma_j\gamma_l}\cos(k_{0z}(r_i+r_k))$. As for $S_i^+\rho^S(t)S_j^+$ terms, the combination of the last two terms in Eq. (A3) makes the imaginary part of $e^{i2k_{0z}r_c sgn(r_l-r_m)}$ vanish. Thus, we have γ_{ijkl}' in Eq. (7).

APPENDIX B: DERIVATION OF EQ. (9)

Here we show how to derive the master equation with coefficients Eq. (B1). The mode function of the squeezed vacuum is given by

$$\boldsymbol{u}_{\boldsymbol{k}s}(\boldsymbol{r}_i) = \sqrt{\frac{\omega_{ks}}{2\epsilon_0 \hbar V}} \boldsymbol{e}_{ks} e^{i\boldsymbol{k}\cdot(\boldsymbol{r}_i - \boldsymbol{o}_{ks})} \tag{B1}$$

where $\boldsymbol{o}_{\boldsymbol{k}s}$ is a phenomenological parameter which includes the effects of the initial phase and the position of the squeezing source [16]. The correlation functions for the squeezed vacuum are [32]:

$$
\begin{aligned}
\left\langle a_{\boldsymbol{k},s}^{\dagger} a_{\boldsymbol{k}',s'} \right\rangle &= \sinh^2 r \delta_{\boldsymbol{k}'\boldsymbol{k}} \delta_{ss'} \\
\left\langle a_{\boldsymbol{k},s} a_{\boldsymbol{k}',s'}^{\dagger} \right\rangle &= \cosh^2 r \delta_{\boldsymbol{k}'\boldsymbol{k}} \delta_{ss'} \\
\left\langle a_{\boldsymbol{k},s}^{\dagger} a_{\boldsymbol{k}',s'}^{\dagger} \right\rangle &= -e^{-i\theta} \cosh(r)\sinh(r)\delta_{\boldsymbol{k}',2\boldsymbol{k}_0-\boldsymbol{k}}\delta_{ss'} \\
\left\langle a_{\boldsymbol{k},s} a_{\boldsymbol{k}',s'} \right\rangle &= -e^{i\theta} \cosh(r)\sinh(r)\delta_{\boldsymbol{k}',2\boldsymbol{k}_0-\boldsymbol{k}}\delta_{ss'}
\end{aligned}
\tag{B2}
$$

For simplicity, we can set the squeezing parameter $\theta = 0$, and all atoms to align along the same direction.

Since the only difference is the squeezing terms $\left\langle a_{\boldsymbol{k},s}^{\dagger} a_{\boldsymbol{k}',s'}^{\dagger} \right\rangle$, $\langle a_{\boldsymbol{k},s} a_{\boldsymbol{k}',s'} \rangle$, we just start from Eq. (A7). Apart from the factor $e^{i(\omega_i + \omega_j - 2\omega_0)t}$, When $r_i \neq r_j$, Eq. (A7) becomes:

$$
\begin{aligned}
&\sum_{k_z} \int_0^t d\tau \{\boldsymbol{\mu}_{l,i} \cdot \boldsymbol{u}_{-2\boldsymbol{k}_0+\boldsymbol{k}}(r_l) S_{l,i}^+ \boldsymbol{\mu}_{m,j} \cdot \boldsymbol{u}_{\boldsymbol{k}}(r_m) S_{m,j}^+ e^{i(\omega_{\boldsymbol{k}}-\omega_0)\tau}(-M)\rho^S(t-\tau) \\
&\approx -\frac{L}{2\pi}\int_0^{2k_0} dk \int_0^t d\tau e^{i\frac{c^2k_0}{\omega_0}(k-k_0)\tau} e^{-i(2k_0-k)(r_l-o_2)+ik(r_m-o_1)} \frac{\sqrt{\omega_{k_z}\omega_{2k_{0z}-k_z}}\mu_i\mu_j}{\epsilon_0 LS\hbar} M S_{l,i}^+ S_{m,j}^+ \rho^S(t-\tau) \\
&\quad - \frac{L}{2\pi}\int_{-2k_0}^{0} dk \int_0^t d\tau e^{i\frac{c^2k_0}{\omega_0}(-k-k_0)\tau} e^{i(2k_0+k)(r_l-o_2)+ik(r_m-o_1)} \frac{\sqrt{\omega_{k_z}\omega_{-2k_{0z}-k_z}}\mu_i\mu_j}{\epsilon_0 LS\hbar} M S_{l,i}^+ S_{m,j}^+ \rho^S(t-\tau) \\
&= -\frac{L}{2\pi}\int_0^{2k_0} dk \int_0^t d\tau e^{i\frac{c^2k_0}{\omega_0}(-k_0)\tau} e^{-i(2k_0)(r_l-o_2)} e^{ik(\frac{c^2k_0}{\omega_0}\tau+r_l-o_1+r_m-o_2)} \frac{\sqrt{\omega_{k_z}\omega_{2k_{0z}-k_z}}\mu_i\mu_j}{\epsilon_0 LS\hbar} M S_{l,i}^+ S_{m,j}^+ \rho^S(t-\tau) \\
&\quad - \frac{L}{2\pi}\int_0^{2k_0} dk \int_0^t d\tau e^{i\frac{c^2k_0}{\omega_0}(-k_0)\tau} e^{i(2k_0)(r_l-o_2)} e^{ik(\frac{c^2k_0}{\omega_0}\tau-r_l+o_1-r_m+o_2)} \frac{\sqrt{\omega_{k_z}\omega_{-2k_{0z}-k_z}}\mu_i\mu_j}{\epsilon_0 LS\hbar} M S_{l,i}^+ S_{m,j}^+ \rho^S(t-\tau) \\
&\approx -\frac{L}{2\pi}\int_0^t d\tau e^{i\frac{c^2k_0}{\omega_0}(-k_0)\tau} e^{-i(2k_0)(r_l)} e^{2ik_0 o_2} 2\pi\delta(\frac{c^2k_0}{\omega_0}\tau+2r_c-o_1-o_2)\frac{\sqrt{\omega_i\omega_j}\mu_i\mu_j}{\epsilon_0 LS\hbar} M S_{l,i}^+ S_{m,j}^+ \rho^S(t-\tau) \\
&\quad - \frac{L}{2\pi}\int_0^t d\tau e^{i\frac{c^2k_0}{\omega_0}(-k_0)\tau} e^{i(2k_0)(r_l)} e^{-2ik_0 o_2} 2\pi\delta(\frac{c^2k_0}{\omega_0}\tau-2r_c+o_1+o_2)\frac{\sqrt{\omega_i\omega_j}\mu_i\mu_j}{\epsilon_0 LS\hbar} M S_{l,i}^+ S_{m,j}^+ \rho^S(t-\tau) \\
&\approx -\frac{L}{2\pi} e^{i(-k_0)|2r_c-o_1-o_2|} e^{sgn(r_c-o_1-o_2)i(2k_0)(r_l)} e^{-2ik_0 o_2} 2\pi \frac{\omega_0^2 \mu_i\mu_j}{\epsilon_0 LS\hbar c^2 k_{0z}} M S_{l,i}^+ S_{m,j}^+ \rho^S(t) \\
&= -\frac{L}{2\pi} e^{sgn(2r_c-o_1-o_2)ik_0(r_l-r_m)} e^{ik_0(o_1-o_2)} 2\pi \frac{\omega_0^2 \mu_i\mu_j}{\epsilon_0 LS\hbar c^2 k_{0z}} M S_{l,i}^+ S_{m,j}^+ \rho^S(t)
\end{aligned}
\tag{B1}
$$

Since we need to sum over l,m,i,j, the imaginary part of $e^{sgn(2r_c-o_1-o_2)ik_0(r_l-r_m)}$ gets canceled, which yields $\gamma'_{ijkl} = \sqrt{\gamma_i\gamma_k}\cos[k_{0z}(r_{jl})]$. The above calculation is also valid when $r_i = r_j$.

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