Phase squeezing in two-photon correlated-spontaneous-emission lasers

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We show that in a two-photon correlated-spontaneous-emission laser with a fast-decaying intermediate relay level, it is possible to obtain the same degree of phase squeezing as when all levels have comparable lifetimes, but with a much higher gain coefficient.

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I. INTRODUCTION

Lasers and masers based on two-photon transitions have been the object of a multitude of theoretical work since the first suggestions, made by Sorokin, Braslau, and Prokhorov, that this system would allow for easier tunability and faster growth of the field density than in the case of usual lasers [1]. These studies, complemented by the proposal that two-photon oscillators could be used to generate squeezed light [2], lead to a strong motivation to build these devices, in spite of the enormous difficulties associated with the second-order character of the underlying gain process.

The recent experimental realization of two-photon micromasers [3] and lasers [4] has revived the interest in this problem. However, the spontaneous-emission noise associated with an inverted system inhibits any possibility of obtaining squeezing at steady state [5]. Only transient squeezing becomes then possible [6]. An alternative approach would be to pump the active atoms into a coherent superposition of the states involved in the lasing process, thus providing some control on the relative phase and therefore on the spontaneous emission transitions between those states [7–9]. In this way, it has been shown that generation of squeezed light by two-photon lasers is compatible with gain [8] and even with atomic inversion [9], thus providing bright sources of squeezed light. This is in contrast with the usual situation involving parametric amplifiers, where the active atoms are far from saturation, due to the fact that the fields are of low intensity and off-resonance with respect to the atomic transitions. Under these conditions, spontaneous-emission noise is negligible [10]. The counterpart is however that usually only feeble light is obtained from these devices [11].

For homogeneously-broadened one-photon lasers, injection of the atoms in a coherent superposition of states does not lead to phase squeezing, but only to sub-Poissonian photon statistics [12]. Therefore, phase squeezing is a specific property of two-photon correlated-spontaneous-emission devices, so that this phenomenon constitutes yet another motivation for the experimental investigation of these systems.

Work on two-photon correlated-spontaneous-emission lasers usually assumes that all the states involved in the process have identical lifetimes. This assumption has a technical origin: it makes it easier to derive the corresponding master equation. We show here however that, by allowing different lifetimes, one can substantially increase the gain, while keeping the squeezing the same as before.

In Sec. II, we describe our model and write down the corresponding master equation (which is derived in the Appendix), and Fokker-Planck equation, obtaining from it the equation of motion for the average phase, as well as the phase diffusion coefficient. In Sec. III, we discuss the conditions under which phase squeezing is compatible with maximum gain, and derive an expression for the phase uncertainty, for the field inside the laser cavity. We show that it is possible to get up to 50% of phase squeezing (implying up to 100% squeezing for the field outside the cavity), with a gain which can be much higher than the one obtained before. This conclusion holds both for the three-level model discussed in Ref. [8] and the four-level model of Ref. [9]. In Sec. IV, we summarize our conclusions.

II. THE MODEL

We consider a system of three-level atoms interacting with a single-mode field in a cavity. The corresponding level scheme is displayed in Fig. 1. Two-photon resonance is assumed, but the intermediate level is taken to be detuned with respect to the one-photon transition. The cavity is assumed to have sufficiently high finesse so as to prevent one-photon emission. At the same time, the intermediate level b acts as a relay level, enhancing the transition probability from a to c. Precisely this configuration was involved in the two-photon micromaser experiment [3,6]. Since we are here interested in the laser case,

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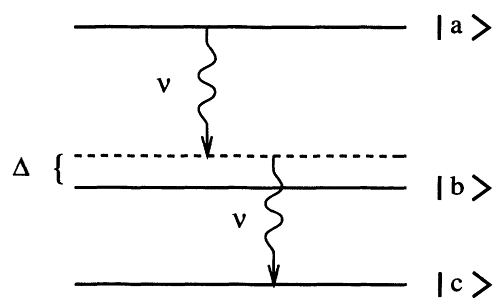

FIG. 1. Level scheme for the two-photon correlated-spontaneous-emission laser. The atoms are pumped into a coherent superposition of states $|a\rangle$ and $|c\rangle$, with no population in the relay level b.

we add to this model the assumption that each level has its own decaying reservoir, that is, one or more lower-lying levels to which it eventually decays. We assume furthermore that all atoms are pumped into a superposition of the states $|a\rangle$ and $|c\rangle$, with no initial population in state $|b\rangle$. That is, the atoms have initial populations ρ_{aa} and ρ_{cc}, and initial coherence $\rho_{ca} = \rho_{ac}^*$. The Hamiltonian in the interaction picture, within the dipole and rotating wave approximations, is $\hbar V$, where

$$V = \left\{ e^{i\Delta_1 t} a |a\rangle\langle b| + e^{i\Delta_2 t} a |b\rangle\langle c| + \text{H.c.} \right\} g + (\Omega - \nu) a^\dagger a \,, \tag{2.1}$$

and

$$\Delta_1 = \omega_a - \omega_b - \nu, \quad \Delta_2 = \omega_b - \omega_c - \nu \,.$$

In these equations, $\hbar\omega_i$ is the energy of level $|i\rangle$, Ω is the resonance frequency of the empty cavity, ν is the laser operating frequency, and g is the coupling constant between the field resonant mode and the atoms of the amplifying medium (assumed for simplicity to be the same for all the relevant levels). As usual in two-photon lasers, the states $|a\rangle$ and $|c\rangle$ are assumed to have the same parity, opposite to that of state $|b\rangle$, to which they are connected through dipole transitions. The inclusion in (2.1) of the term proportional to the photon number operator allows one to write the unperturbed Hamiltonian in terms of the operating frequency of the laser.

In the Appendix, we derive the following master equation for the field reduced density matrix:

$$\begin{aligned} \dot{\rho}(t) = -i(\Omega - \nu) a^\dagger a \rho(t) - \tfrac{1}{2} \Big\{ & \alpha_{a,ab} \mathcal{L}_1^{ab} \, \rho_{aa} [a a^\dagger \rho(t) - a^\dagger \rho(t) a] \\ & + \alpha_{c,cb} \mathcal{L}_2^{cb} \, \rho_{cc} [\rho(t) \, a^\dagger a - a \rho(t) \, a^\dagger] + \alpha_{ac,ab} \, \mathcal{L}_1^{ab} \mathcal{L}_4^{ac} \, \rho_{ca} [a a \rho(t) - a \rho(t) a] \\ & + \alpha_{ac,cb} \mathcal{L}_2^{cb} \mathcal{L}_4^{ac} \, \rho_{ca} [\rho(t) \, a a - a \rho(t) \, a] \Big\} - \frac{\gamma}{2} \, [a^\dagger a \rho(t) - a \rho(t) \, a^\dagger] + \text{H.c.} \,, \end{aligned} \tag{2.2}$$

where a and $a^\dagger$ are, respectively, the annihilation and creation operators for the field mode, γ is the decay constant for the field in the cavity, Γ_k is the decay rate of level $|k\rangle$, r is the average pumping rate (Poissonian pumping is assumed), and

$$\Delta_4 = \omega_a - \omega_c - 2\nu \,, \quad \alpha_{\gamma\delta,\beta\eta} = \frac{2g^2 r}{\Gamma_{\gamma\delta} \Gamma_{\beta\eta}},$$

$$\alpha_{\delta,\beta\eta} = \frac{2g^2 r}{\Gamma_\delta \Gamma_{\beta\eta}} \,, \quad \mathcal{L}_k^{\beta\eta} = \frac{\Gamma_{\beta\eta}}{\Gamma_{\beta\eta} - i\Delta_k} \,, \quad \Gamma_{\alpha\beta} = \frac{\Gamma_\alpha + \Gamma_\beta}{2} \,.$$

A Fokker-Planck equation for the Glauber-Sudarshan P representation of the density operator is derived from this master equation in the usual way [13,14] ($a|\epsilon\rangle = \epsilon|\epsilon\rangle$):

$$\begin{aligned} \frac{\partial P}{\partial t} = & -\frac{\partial}{\partial \varepsilon} (d_\varepsilon P) - \frac{\partial}{\partial \varepsilon^*} (d_{\varepsilon^*} P) + 2 \frac{\partial^2}{\partial \varepsilon \partial \varepsilon^*} (D_{\varepsilon^* \varepsilon} P) \\ & + \frac{\partial^2}{\partial \varepsilon^2} (D_{\varepsilon\varepsilon} P) + \frac{\partial^2}{\partial \varepsilon^{*2}} (D_{\varepsilon^* \varepsilon^*} P) \,, \end{aligned} \tag{2.3}$$

where

$$d_\varepsilon = (d_{\varepsilon^*})^* = R\varepsilon + G^* \varepsilon^* \,, \tag{2.4}$$

$$D_{\varepsilon\varepsilon} = (D_{\varepsilon^*\varepsilon^*})^* = -\tfrac{1}{2} \alpha_{ac,cb} \, \mathcal{L}_2^{cb^*} \mathcal{L}_4^{ac^*} \, \rho_{ac} \,, \tag{2.5}$$

$$R = \tfrac{1}{2} (\alpha_{a,ab} \, \mathcal{L}_1^{ab} \, \rho_{aa} - \alpha_{c,cb} \, \mathcal{L}_2^{cb^*} \, \rho_{cc}) - i(\Omega - \nu) - \frac{\gamma}{2} \,, \tag{2.6}$$

$$G = \tfrac{1}{2} (\alpha_{ac,ab} \, \mathcal{L}_1^{ab} - \alpha_{ac,cb} \mathcal{L}_2^{cb}) \, \mathcal{L}_4^{ac} \, \rho_{ca} \,, \tag{2.7}$$

$$D_{\varepsilon^*\varepsilon} = \tfrac{1}{4} (\mathcal{L}_1^{ab} + \mathcal{L}_1^{ab^*}) \, \alpha_{a,ab} \, \rho_{aa} \,. \tag{2.8}$$

Polar coordinates ($\epsilon = re^{i\phi}$) yield the following phase-diffusion coefficient [14] (assuming two-photon resonance, $\Delta_1 = \Delta = -\Delta_2$, and far-above-the-threshold operation)

$$\begin{aligned} D_{\phi\phi} = \frac{1}{4\tilde{n}} \{ & \alpha_{a,ab} \, \rho_{aa} \cos^2 \mu_{ab} \\ & + \alpha_{ac,cb} |\rho_{ca}| \cos \mu_{cb} \cos(2\phi + \theta_{ca} - \mu_{cb}) \} \,, \end{aligned} \tag{2.9}$$

where $\tilde{n}$ is the mean photon number and

$$\rho_{ca} = |\rho_{ca}| \, e^{i\theta_{ca}} \,, \tag{2.10}$$

$$\tan \mu_{ab} = \frac{\Delta}{\Gamma_{ab}} \,. \tag{2.11}$$

For the average phase, one gets the following equation of motion:

$$\dot{\phi} = -|\rho_{ca}|\alpha_{ac,\Delta}\{\sin\mu_{cb}\sin(2\phi+\theta_{ca}-\mu_{cb}) - \sin\mu_{ab}\sin(2\phi+\theta_{ca}+\mu_{ab})\} + \nu - \Omega - \tfrac{1}{2}\{\alpha_{a,ab}\rho_{aa}\cos\mu_{ab}\sin\mu_{ab} - \alpha_{c,cb}\rho_{cc}\cos\mu_{cb}\sin\mu_{cb}\}\ , \tag{2.12}$$

where

$$\alpha_{ac,\Delta} = \alpha_{ac,cb}\cot\mu_{cb} = \alpha_{ac,ab}\ \cot\mu_{ab}\ . \tag{2.13}$$

From (2.12), we get both a frequency-pulling equation,

$$\nu = \Omega + \tfrac{1}{2}\{\alpha_{a,ab}\rho_{aa}\cos\mu_{ab}\sin\mu_{ab} - \alpha_{c,cb}\rho_{cc}\cos\mu_{cb}\sin\mu_{cb}\}\ , \tag{2.14}$$

and a phase-locking-like equation,

$$\dot{\phi} = -|\rho_{ca}|\alpha_{ac,\Delta}\{\sin\mu_{cb}\sin(2\phi+\theta_{ca}-\mu_{cb}) - \sin\mu_{ab}\sin(2\phi+\theta_{ca}+\mu_{ab})\}\ . \tag{2.15}$$

In the following section, we discuss conditions under which phase squeezing can be obtained, with a gain appreciably larger than the one corresponding to the model with equal lifetimes.

III. PHASE SQUEEZING AND GAIN

In order to have true two-photon transitions, the detuning Δ must be much larger than the linewidth Γ_b of state $|b\rangle$. Under this condition, the time it takes for the two-photon transition to occur can be estimated as $1/\Delta$, and therefore one should have $\Delta \gg \Gamma_a, \Gamma_c$, otherwise the atomic coherence ρ_{ac} would decay before the transition takes place. This would destroy any squeezing, because spontaneous-emission noise reduction depends on the atomic coherence between levels $|a\rangle$ and $|c\rangle$. Now, let us consider the gain. One way of increasing it is to slow down the rate in which $|a\rangle$ gets deexcited. This is the case when Γ_a (and consequently Γ_c, which we assume to have the same order of magnitude as Γ_a), is much smaller than Γ_b.

These arguments suggest that one should look at the following limit: $\Delta \gg \Gamma_b \gg \Gamma_a, \Gamma_c$. Under these conditions, Eq. (2.15) becomes

$$\dot{\phi} = -|\rho_{ca}|\alpha_{ac,\Delta}\sin^2\mu\cos(2\phi+\theta_{ca})\ , \tag{3.1}$$

where $\Gamma_b \equiv 2\Gamma$, $\tan\mu \equiv \frac{\Delta}{\Gamma}$, and $\alpha_{ac,\Delta} \equiv \frac{2g^2r}{\Gamma_{ac}\Delta}$.

Phase locking is obtained for

$$\phi_0 = \tfrac{1}{2}\theta_{ac} - \left(\tfrac{3}{4} \pm \tfrac{1}{2}\right)\pi\,\mathrm{sgn}\,\Delta\,,$$

which coincides with the expression found in Ref. [8], in spite of the fact that here all lifetimes are different. The existence of two stable solutions for the locked phase is a peculiar feature of two-photon devices, as discussed in Ref. [9]: the two-photon polarization ρ_{ac} is a source for the *square* of the electric field, so that the usual locking equation is now satisfied by 2ϕ, which can have only one stable value. This value is defined, however, modulo 2π, so ϕ is given modulo π, thus yielding two stable values.

When the average phase locks, the phase diffusion coefficient becomes

$$D_{\phi\phi} = \frac{1}{4\tilde{n}}\ \alpha_{a,ab}\cos^2\mu\left\{\frac{\Gamma_{ac}}{\Gamma_a}\ \rho_{aa} - \frac{|\rho_{ca}\Delta|}{\Gamma}\right\}\ . \tag{3.2}$$

This expression closely resembles its counterpart from Ref. [8]. The differences between them arise from the presence of Γ_{ac}/Γ_a multiplying ρ_{aa} and from $\alpha_{a,ab}$ being substituted for α.

Unless $D_{\phi\phi}$ becomes negative there is no squeezing. From Eq. (3.2) we see that squeezing requires

$$\frac{\rho_{aa}}{|\rho_{ca}|} < \frac{|\Delta|}{\Gamma}\ \frac{\Gamma_a}{\Gamma_{ac}}\ . \tag{3.3}$$

The phase uncertainty at steady state is given by

$$\langle(\Delta\phi)^2\rangle = \frac{1}{8\tilde{n}}\left\{1 + \frac{\Gamma_{ac}}{\Gamma_a}\ \frac{\Gamma\rho_{aa}}{|\rho_{ca}\Delta|}\right\}\ , \tag{3.4}$$

and is indeed seen to be smaller than the coherent-state value $1/4\tilde{n}$ when condition (3.3) is satisfied.

On the other hand, the linear gain coefficient, obtained from the Fokker-Planck equation (2.3), is given by

$$G = \alpha_{ac,b}\left\{\frac{\Gamma_{ac}}{\Gamma_a}\ \rho_{aa} - \frac{\Gamma_{ac}}{\Gamma_c}\ \rho_{cc} + 2\ \frac{|\rho_{ca}\Delta|}{\Gamma}\right\}\cos^2\mu. \tag{3.5}$$

The oscillation threshold is given by $G > \gamma$. We identify two contributions to (3.5), corresponding to two distinct physical effects. The one dependent on the populations is associated with stimulated processes (emission for the term proportional to ρ_{aa}, absorption for the ρ_{cc} contribution), while the other term, proportional to the coherence, corresponds to a Klystron-like gain, generated by the injected atomic polarization.

For $\Gamma_{ac} = \Gamma_a = \Gamma_c \equiv \Lambda$, these expressions become

$$\langle(\Delta\phi)^2\rangle = \frac{1}{8\tilde{n}}\ \left\{1 + \frac{\Gamma\rho_{aa}}{|\rho_{ca}\Delta|}\right\}\ , \tag{3.6}$$

$$G = (1+\delta)\ \alpha\left\{\rho_{aa} - \rho_{cc} + 2\ \frac{|\rho_{ca}\Delta|}{\Gamma}\right\}\cos^2\mu, \tag{3.7}$$

where $1+\delta \equiv \Gamma/\Lambda$ and $\alpha \equiv 2g^2r/\Gamma^2$.

For $\Delta \gg \Gamma$, we see that for perfect coherence [$|\rho_{ac}| = (\rho_{aa}\rho_{cc})^{1/2}$] it is possible to get up to 50% squeezing for the field inside the cavity—and therefore up to 100% squeezing for the output field spectrum [15]—while still having inversion and stimulated gain, that is $\rho_{aa} > \rho_{cc}$. On the other hand, the linear gain given by (3.7) differs from the corresponding expression of Ref. [8] by the factor $1+\delta$. Thus the condition $\Gamma/\Lambda \gg 1$ greatly increases the gain, for the same squeezing as in the equal-lifetime situation.

Analogous considerations can be applied to the four level model suggested in Ref. [9], where the single level c is replaced by a duplet of almost degenerate levels c and d, and the possibility of initial coherences ρ_{ac}, ρ_{ad}, and ρ_{cd} is taken into account. The corresponding results are obtained in the same way as before from the master equation obtained in the Appendix. For $\Gamma_b = 2\Gamma$ and $\Gamma_a = \Gamma_c = \Gamma_d = \Lambda$ with $\Delta \gg \Gamma \gg \Lambda$, one gets the same phase-locking condition as in Ref. [9], and the same amount of squeezing, but the gain increases by the same factor as in the three-level model.

IV. CONCLUSIONS

The compatibility of squeezing with inversion and stimulated emission gain has been demonstrated before in the framework of two-photon correlated spontaneous-emission lasers. However, previous treatments suffered from the restriction that all relevant lifetimes were assumed to be identical.

We have shown here that, by assuming that the relay level in the two-photon transition decays much faster than the other lasing levels, it is possible to greatly increase the gain, while at the same time keeping phase squeezing close to the ideal 50%. Since coherent atomic injection does not lead to phase squeezing in one-photon lasers, our result provides yet another motivation for building two-photon oscillators, since it reinforces the idea of using these systems as bright sources of squeezed light.

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APPENDIX

We derive now the master equation for a four-level system with different decay times. The corresponding equation for a three level system is then obtained as a special case. We follow the general procedure of Appendix I of Ref. [13].

For each atomic state $|j\rangle$, let us introduce a fictitious state $|j'\rangle$ to account for spontaneous decay. The reduced density matrix for the field is obtained from the total density matrix by tracing over all atomic states—$|a\rangle, |b\rangle, |c\rangle, |d\rangle, |a'\rangle, |b'\rangle, |c'\rangle, |d'\rangle$. However, after a time τ, large compared to the atomic lifetimes, the atom must have decayed to the primed states. In the direct product atom-field basis, one writes

$$\rho^1_{nm}(t+\tau) \approx \varrho^1_{a'n;a'm}(t+\tau) + \varrho^1_{b'n;b'm}(t+\tau) + \varrho^1_{c'n;c'm}(t+\tau) + \varrho^1_{d'n;d'm}(t+\tau), \quad \text{(A1)}$$

where ρ designates the reduced density matrix for the field and ϱ the total density matrix. The superscript one reminds us that we are dealing with a single atom.

Considering the eight-level atom plus the field as a closed system, we write its state ket as

$$|\psi_{af}(t')\rangle = \sum_n \Big([C^{\psi}_{an\{0\}}(t')|a\rangle + C^{\psi}_{bn\{0\}}(t')|b\rangle + C^{\psi}_{cn\{0\}}(t')|c\rangle + C^{\psi}_{dn\{0\}}(t')|d\rangle] + \sum_r [C^{\psi}_{a'n\{1_r\}}(t')|a'\rangle + C^{\psi}_{b'n\{1_r\}}(t')|b'\rangle + C^{\psi}_{c'n\{1_r\}}(t')|c'\rangle + C^{\psi}_{d'n\{1_r\}}(t')|d'\rangle]|\{1_r\}\rangle\Big)|n\rangle, \quad \text{(A2)}$$

where $\{0\}$ and $\{1_r\}$ refer to all modes except the lasing one. $\{0\}$ stands for all of them in the vacuum state and $\{1_r\}$ for one photon in mode r and none in the rest. In terms of these coefficients, the total density-matrix elements in Eq. (A1) are

$$\varrho^1_{a'n;a'm}(t+\tau) = \sum_\psi P_\psi \sum_r C^{\psi}_{a'n\{1_r\}}(t+\tau) C^{\psi^*}_{a'm\{1_r\}}(t+\tau), \quad \text{(A3a)}$$

$$\varrho^1_{b'n;b'm}(t+\tau) = \sum_\psi P_\psi \sum_r C^{\psi}_{b'n\{1_r\}}(t+\tau) C^{\psi^*}_{b'm\{1_r\}}(t+\tau), \quad \text{(A3b)}$$

$$\varrho^1_{c'n;c'm}(t+\tau) = \sum_\psi P_\psi \sum_r C^{\psi}_{c'n\{1_r\}}(t+\tau) C^{\psi^*}_{c'm\{1_r\}}(t+\tau), \quad \text{(A3c)}$$

$$\varrho^1_{d'n;b'm}(t+\tau) = \sum_\psi P_\psi \sum_r C^{\psi}_{d'n\{1_r\}}(t+\tau) C^{\psi^*}_{d'm\{1_r\}}(t+\tau), \quad \text{(A3d)}$$

where P_ψ is the probability of finding state $|\psi_{af}\rangle$ in the statistical mixture.

Now for each decay $|\alpha\rangle \to |\alpha'\rangle$ we neglect the other atomic levels and treat the problem as a two level atom interacting with the field. This is a good approximation as long as the coupling between the atom and the background modes—i.e., all modes except the lasing one—is weak. Assuming a typical dipole interaction Hamiltonian in the rotating wave approximation, one can write equations of motion for the probability amplitudes C in the interaction picture. Integrating the equation for $C^{\psi}_{\alpha'n\{1_r\}}$, we get

$$C^{\psi}_{\alpha'n\{1_r\}}(t+\tau) = -ig_{r\alpha\alpha'} \int_0^\tau d\tau' e^{i(\Omega_r - \omega_{\alpha'\alpha})\tau'} C^{\psi}_{\alpha n\{0\}}(t+\tau'), \quad \text{(A4)}$$

where $g_{r\alpha\alpha'}$ is the coupling between $|\alpha\rangle \to |\alpha'\rangle$ and the rth field mode, Ω_r is the mode frequency, and $\omega_{\alpha'\alpha} = \omega_\alpha - \omega_{\alpha'}$, with $|\alpha\rangle$ being any of the four levels.

From Eqs. (A3) and (A4), we get an equation for $\varrho^1_{\alpha' n;\alpha' m}$. Performing the Weisskopf-Wigner approximation, we end up with

$$\varrho^1_{\alpha' n;\alpha' m}(t+\tau) = \Gamma_\alpha \int_0^\tau d\tau' \varrho^1_{\alpha n;\alpha m}(t+\tau'), \tag{A5}$$

where

$$\Gamma_\alpha = 2\pi g^2_{\alpha\alpha'}(\omega_{\alpha'\alpha})\wp(\omega_{\alpha'\alpha}),$$

$$g_{r\alpha\alpha'} \to g_{\alpha\alpha'}(\Omega_r),$$

$$\sum_r \to \int_0^\infty \wp(\Omega)d\Omega,$$

with $\wp(\Omega)$ being the mode density.

Plugging (A5) into (A1), we get

$$\begin{aligned}\rho^1_{nm}(t+\tau) &= \int_0^\tau d\tau' \{\Gamma_a \varrho^1_{an;am}(t+\tau') + \Gamma_b \varrho^1_{bn;bm}(t+\tau') \\ &\quad + \Gamma_c \varrho^1_{cn;cm}(t+\tau') + \Gamma_d \varrho^1_{dn;dm}(t+\tau')\}.\end{aligned} \tag{A6}$$

In order to carry out the integration required in (A6), one must know how these matrix elements evolve in time. If there were no spontaneous decay to primed states, their time evolution would be given by the laser Hamiltonian

$$\begin{aligned}V = &\{e^{i\Delta_1 t} a|a\rangle\langle b| + e^{i\Delta_2 t} a|b\rangle\langle c| + e^{i\Delta_3 t} a|b\rangle\langle d| + \text{H.c.}\} \\ &\times g + (\Omega - \nu)a^\dagger a.\end{aligned}$$

In the Weisskopf-Wigner approximation, spontaneous emission leads to exponential decay of upper level populations. So we introduce the corresponding decay terms in the equations of motion obtained from the interaction V:

$$\dot\varrho^1_{a;a}(t+\tau') = -\Gamma_a \varrho^1_{a;a}(t+\tau') - \{i(\Omega-\nu)a^\dagger a \varrho^1_{a;a}(t+\tau') + ige^{i\Delta_1\tau'} a\varrho^1_{b;a}(t+\tau') + \text{H.c.}\}, \tag{A7a}$$

$$\begin{aligned}\dot\varrho^1_{b;b}(t+\tau') = &-\Gamma_b \varrho^1_{b;b}(t+\tau') - \{i(\Omega-\nu)a^\dagger a \varrho^1_{b;b}(t+\tau') + ig[e^{-i\Delta_1\tau'} a^\dagger \varrho^1_{a;b}(t+\tau') + e^{i\Delta_2\tau'} a\varrho^1_{c;b}(t+\tau') \\ &+ e^{i\Delta_3\tau'} a\varrho^1_{d;b}(t+\tau')] + \text{H.c.}\},\end{aligned} \tag{A7b}$$

$$\dot\varrho^1_{c;c}(t+\tau') = -\Gamma_c \varrho^1_{c;c}(t+\tau') - \{i(\Omega-\nu)a^\dagger a \varrho^1_{c;c}(t+\tau') + ige^{-i\Delta_2\tau'} a^\dagger \varrho^1_{b;c}(t+\tau') + \text{H.c.}\}, \tag{A7c}$$

$$\dot\varrho^1_{d;d}(t+\tau') = -\Gamma_d \varrho^1_{d;d}(t+\tau') - \{i(\Omega-\nu)a^\dagger a \varrho^1_{d;d}(t+\tau') + ige^{-i\Delta_3\tau'} a^\dagger \varrho^1_{b;d}(t+\tau') + \text{H.c.}\}, \tag{A7d}$$

where $\Delta_1 = \omega_a - \omega_b$, $\Delta_2 = \omega_b - \omega_c - \nu$, and $\Delta_3 = \omega_b - \omega_d - \nu$.

Now, the right-hand side of (A6) is a sum of terms of the form

$$\int_0^\tau d\tau' \Gamma_\alpha \varrho^1_{\alpha;\alpha}(t+\tau'),$$

which can be written as

$$-\int_0^\tau d\tau' (-\Gamma_\alpha e^{-\Gamma_\alpha \tau'}) \{\varrho^1_{\alpha;\alpha}(t+\tau') e^{\Gamma_\alpha \tau'}\},$$

so that, integrating by parts, we get

$$\varrho^1_{\alpha;\alpha}(t) + \int_0^\tau d\tau' e^{-\Gamma_\alpha\tau'} \frac{d}{d\tau'}\{\varrho^1_{\alpha;\alpha}(t+\tau') e^{\Gamma_\alpha\tau'}\},$$

because $\varrho^1_{\alpha;\alpha}(t+\tau) \approx 0$ due to the decay to $|\alpha'\rangle$.

We can now write (A6) in the following form:

$$\begin{aligned}\rho^1(t+\tau) - \rho^1(t) = \int_0^\tau d\tau' \{&\dot\varrho^1_{a;a}(t+\tau') + \Gamma_a \varrho^1_{a;a}(t+\tau') + \dot\varrho^1_{b;b}(t+\tau') \\ &+ \Gamma_b \varrho^1_{b;b}(t+\tau') + \dot\varrho^1_{c;c}(t+\tau') + \Gamma_c \varrho^1_{c;c}(t+\tau') + \dot\varrho^1_{d;d}(t+\tau') + \Gamma_d \varrho^1_{d;d}(t+\tau')\},\end{aligned} \tag{A8}$$

where we have used that $\varrho^1_{a;a}(t) + \varrho^1_{b;b}(t) + \varrho^1_{c;c}(t) + \varrho^1_{d;d}(t) = \rho^1(t)$, because the primed states are not populated until the unprimed states population starts decaying.

Plugging (A7) into (A8), we obtain

$$\rho^1(t+\tau)-\rho^1(t)=\int_0^\tau d\tau'\{-i(\Omega-\nu)a^\dagger a[\varrho^1_{a;a}(t+\tau')+\varrho^1_{b;b}(t+\tau')+\varrho^1_{c;c}(t+\tau')+\varrho^1_{d;d}(t+\tau')]$$
$$-ig([e^{i\Delta_1\tau'}a,\varrho^1_{b;a}(t+\tau')]+[e^{i\Delta_2\tau'}a,\varrho^1_{c;b}(t+\tau')]+[e^{i\Delta_3\tau'}a,\varrho^1_{d;b}(t+\tau')])\}+\text{H.c.} \tag{A9}$$

The coarse-grained [13] time derivative of ρ is obtained by multiplying the change in ρ due to a single atom by the average pumping rate r. This procedure, justifiable if each atom makes only a small contribution to the field, or under the assumption of Poissonian pumping statistics [16], yields a master equation for the field reduced density operator.

Since $\varrho^1_{\alpha;\alpha}$ is nonvanishing only for a short time, $\Delta\tau \ll \tau$, we can approximate the integral

$$r\int_0^\tau d\tau'[\varrho^1_{a;a}(t+\tau')+\varrho^1_{b;b}(t+\tau')+\varrho^1_{c;c}(t+\tau')+\varrho^1_{d;d}(t+\tau')]$$

by $r\Delta\tau\rho^1(t)$.

Note that $r\Delta\tau$ is the number of atoms pumped into the lasing levels before the first atom has time to decay appreciably. Since $\rho^1(t)$ is the contribution of a single atom to the density matrix, we can approximate $r\Delta\tau\rho^1(t)$ by $\rho(t)$ which includes the contribution of all the atoms pumped during the time interval $\Delta\tau$. When we do so, we are just adding the contributions of each individual atom to $\rho(t)$. Again this is a good approximation if each atom makes only a small contribution to the field, which is consistent with our perturbative approach. So the coarse-grained time derivative of ρ is given by

$$\dot\rho(t)=-i(\Omega-\nu)a^\dagger a\rho(t)-igr\int_0^\tau d\tau'\{[e^{i\Delta_3\tau'}a,\varrho^1_{b;a}(t+\tau')]+[e^{i\Delta_2\tau'}a,\varrho^1_{c;b}(t+\tau')]+[e^{i\Delta_3\tau'}a,\varrho^1_{d;b}(t+\tau')]\}+\text{H.c.} \tag{A10}$$

In order to calculate the integral in the above expression, one has to know the time evolution of $\varrho^1_{b;a}(t+\tau')$, $\varrho^1_{d;b}(t+\tau')$, $\varrho^1_{c;b}(t+\tau')$. Proceeding as before, we get the following equations of motion for these matrix elements:

$$\dot\varrho^1_{b;a}(t+\tau')=-\Gamma_{ab}\varrho^1_{b;a}(t+\tau')-i(\Omega-\nu)[a^\dagger a,\varrho^1_{b;a}(t+\tau')]-ig\{e^{-i\Delta_1\tau'}a^\dagger\varrho^1_{a;a}(t+\tau')+e^{i\Delta_2\tau'}a\varrho^1_{c;a}(t+\tau')$$
$$+e^{i\Delta_3\tau'}a\varrho^1_{d;a}(t+\tau')-e^{-i\Delta_1\tau'}\varrho^1_{b;b}(t+\tau')a^\dagger\}, \tag{A11a}$$

$$\dot\varrho^1_{c;b}(t+\tau')=-\Gamma_{cb}\varrho^1_{c;b}(t+\tau')-i(\Omega-\nu)[a^\dagger a,\varrho^1_{c;b}(t+\tau')]-ig\{e^{-i\Delta_2\tau'}a^\dagger\varrho^1_{b;b}(t+\tau')-\varrho^1_{c;a}(t+\tau')ae^{i\Delta_1\tau'}$$
$$-\varrho^1_{c;c}(t+\tau')a^\dagger e^{-i\Delta_2\tau'}-\varrho^1_{c;d}(t+\tau')a^\dagger e^{-i\Delta_3\tau'}\}, \tag{A11b}$$

$$\dot\varrho^1_{d;b}(t+\tau')=-\Gamma_{db}\varrho^1_{d;b}(t+\tau')-i(\Omega-\nu)[a^\dagger a,\varrho^1_{d;b}(t+\tau')]-ig\{e^{-i\Delta_3\tau'}a^\dagger\varrho^1_{b;b}(t+\tau')-\varrho^1_{d;a}(t+\tau')ae^{i\Delta_1\tau'}$$
$$-\varrho^1_{d;c}(t+\tau')a^\dagger e^{-i\Delta_2\tau'}-\varrho^1_{d;d}(t+\tau')a^\dagger e^{-i\Delta_3\tau'}\}, \tag{A11c}$$

where

$$\Gamma_{\alpha\beta}=\frac{\Gamma_\alpha+\Gamma_\beta}{2}.$$

We solve equations (A11) perturbatively up to first order in the coupling constant g and plug the result into Eq. (A10). After having performed all the integrations required, we get

$$\dot\rho(t)=-i(\Omega-\nu)a^\dagger a\rho(t)-\tfrac{1}{2}\{\alpha_{a,ab}\mathcal{L}_1^{ab}\rho_{aa}[aa^\dagger\rho(t)-a^\dagger\rho(t)a]+(\alpha_{c,cb}\mathcal{L}_2^{cb}\rho_{cc}$$
$$+\alpha_{cd,cb}\mathcal{L}_2^{cb}\mathcal{L}_6^{cd*}\rho_{cd}+\alpha_{cd,db}\mathcal{L}_3^{db}\mathcal{L}_6^{cd}\rho_{dc}+\alpha_{d,db}\mathcal{L}_3^{db}\rho_{dd})[\rho(t)a^\dagger a-a\rho(t)a^\dagger]$$
$$+\mathcal{L}_1^{ab}(\alpha_{ac,ab}\mathcal{L}_4^{ac}\rho_{ca}+\alpha_{ad,ab}\mathcal{L}_5^{ad}\rho_{da})[aa\rho(t)-a\rho(t)a]$$
$$+(\alpha_{ac,cb}\mathcal{L}_2^{cb}\mathcal{L}_4^{ac}\rho_{ca}+\alpha_{ad,db}\mathcal{L}_3^{db}\mathcal{L}_5^{ad}\rho_{da})[\rho(t)aa-a\rho(t)a]\}-\frac{\gamma}{2}[a^\dagger a\rho(t)-a\rho(t)a^\dagger]+\text{H.c.}, \tag{A12}$$

where $\rho_{\alpha\beta}$ are the atomic density matrix elements before the atom enters the cavity,

$$\Delta_4=\Delta_1+\Delta_2,$$
$$\Delta_5=\Delta_1+\Delta_3,$$
$$\Delta_6=\Delta_3-\Delta_2,$$
$$\alpha_{\gamma\delta,\beta\eta}=\frac{2g^2r}{\Gamma_{\gamma\delta}\Gamma_{\beta\eta}},$$
$$\alpha_{\gamma,\beta\eta}=\frac{2g^2r}{\Gamma_\gamma\Gamma_{\beta\eta}},$$

and

$$\mathcal{L}_k^{\beta\eta} = \frac{\Gamma_{\beta\eta}}{\Gamma_{\beta\eta} - i\Delta_k}.$$

Of course, the term proportional to γ does not come out of these calculations. It was added to account for cavity decay.

The master equation for the three-level model is obtained from the above one by dropping the terms involving the state d. One gets then

$$\begin{aligned}\dot{\rho}(t) = -i(\Omega - \nu)a^\dagger a\rho(t) - \tfrac{1}{2}\{&\alpha_{a,ab}\mathcal{L}_1^{ab}\rho_{aa}[aa^\dagger\rho(t) - a^\dagger\rho(t)a] \\ &+\alpha_{c,cb}\mathcal{L}_2^{cb}\rho_{cc}[\rho(t)a^\dagger a - a\rho(t)a^\dagger] + \alpha_{ac,ab}\mathcal{L}_1^{ab}\mathcal{L}_4^{ac}\rho_{ca}[aa\rho(t) - a\rho(t)a] \\ &+\alpha_{ac,cb}\mathcal{L}_2^{cb}\mathcal{L}_4^{ac}\rho_{ca}[\rho(t)aa - a\rho(t)a]\} - \frac{\gamma}{2}[a^\dagger a\rho(t) - a\rho(t)a^\dagger] + \text{H.c.}\end{aligned} \tag{A13}$$

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