Phase coherence and decoherence in the correlated-spontaneous-emission laser

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The role of atomic phase coherence in various correlated-emisson laser (CEL) schemes is investigated. A CEL scheme is introduced in which coherence is generated by a Raman-type two-photon process. Ideal CEL action, that is, vanishing phase diffusion in the difference phase of two modes, is predicted even in the presence of nonradiative transverse decay in this scheme and in a previously suggested configuration. The results are applicable to the analysis of dephasing collisions between atoms in a gaseous CEL medium.

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I. INTRODUCTION

The correlated-emission laser (CEL) has been the subject of theoretical [1–19] and experimental [20,21] attention. Nevertheless, certain aspects of CEL action remain unexplained. The aim of the present paper is to point out and analyze the various ways in which atomic coherence leads to the suppression of phase-diffusion noise in the CEL. In particular, we show that even in the presence of certain coherence-depleting processes such as atomic collisions, it is still possible to completely suppress diffusion in the difference phase of two CEL modes.

First of all, we introduce a CEL scheme in which the atomic coherence is generated by a Raman-type two-photon process (see Fig. 1). This atomic configuration has experimental advantages over existing schemes. For example, it avoids the problem of driving a dipole-forbidden transition (which is necessary in a microwave driven CEL; see Fig. 2), and ideal CEL action is possible without the need for strong driving fields. CEL action in this configuration is compared to a previously analyzed scheme (see Fig. 2), where the atomic coherence is generated by directly driving the two levels in question [2].

In order to set the stage for the discussion of the role of atomic coherence in the CEL, let us first consider the conceptually simplest CEL configuration that incorporates the basic ingredients of the problem. At the heart of the matter, CEL action in the atomic level schemes of Figs. 1 and 2 is the suppression of phase diffusion between the two lasing modes a_1 and a_2 due to coherence between the upper atomic levels $|a_1\rangle$ and $|a_2\rangle$. In the simplest case this coherence is provided by preparing atoms with the level scheme of Figs. 1 or 2 (without auxiliary level $|c\rangle$ and in the absence of the driving fields and transverse decay) in a coherent superposition before they are injected into the interaction region. The injected coherence is chosen to be lower than maximum, i.e., we set the initial atomic density matrix for each atom at the injection time t_i to be $\rho_{a_1a_1}(t=t_i)=\rho_{a_2a_2}(t=t_i)=\frac{1}{2}$ and $\rho_{a_1a_2}(t=t_i)=(1-\epsilon)/2$ $(\epsilon>0)$.

If θ_i denotes the phase of the field in mode i $(i=1,2)$, then the linear gain and cross-coupling coefficients—obtained in analogy to Appendixes A and B—yield for the phase-diffusion coefficient of the difference phase $\Psi=\theta_1-\theta_2$ with the help of Eq. (24),

$$D_\Psi=\frac{\alpha}{2\rho^2}[1-(1-\epsilon)\cos\Psi] , \tag{1}$$

where α is the linear gain coefficient and ρ^2 is the mean number of photons. Therefore, one may at best achieve $D_\Psi=\epsilon\alpha/2\rho^2$ (with $\Psi=0$). In other words, if we define the degree of coherence $d_{\mathrm{coh}}=|\rho_{a_1a_2}|/(\rho_{a_1a_1}\rho_{a_2a_2})^{1/2}$, then maximum initial coherence corresponding to $d_{\mathrm{coh}}=1$ (i.e., $\epsilon=0$) leads to complete suppression of phase diffusion. However, any submaximum ratio $d_{\mathrm{coh}}=1-\epsilon$ $(\epsilon>0)$ results in a nonvanishing phase-diffusion constant. Thus we are led to conclude that coherence-depleting processes, such as collisions of the lasing atoms, result in incomplete CEL action.

Consider now the Raman-driven quantum beat CEL as per Fig. 1. Here, coherence is generated by two external resonant driving fields with Rabi frequencies Ω_1 and Ω_2 coupling the two upper levels $|a_1\rangle$ and $|a_2\rangle$ to an auxiliary level $|c\rangle$. In the absence of transverse decay $(\Gamma_p=\Gamma_p'=0)$ and for $\Omega_1\equiv\Omega_2$ the degree of coherence for $\rho_{a_1a_2}$ is easily calculated to be $d_{\mathrm{coh}}=1$ in steady state. Thus we expect ideal CEL action in this scheme. Not surprisingly, this is indeed the case, as shown later in the present paper. If we now include some nonvanishing transverse decay $(\Gamma_p\neq0)$, then the degree of coherence between the upper two levels decreases, $d_{\mathrm{coh}}=\Gamma/(\Gamma+\Gamma_p)<1$ (independently of $\Omega_1=\Omega_2$), but contrary to the above arguments, we nevertheless obtain complete suppression of phase diffusion, i.e., ideal CEL action.

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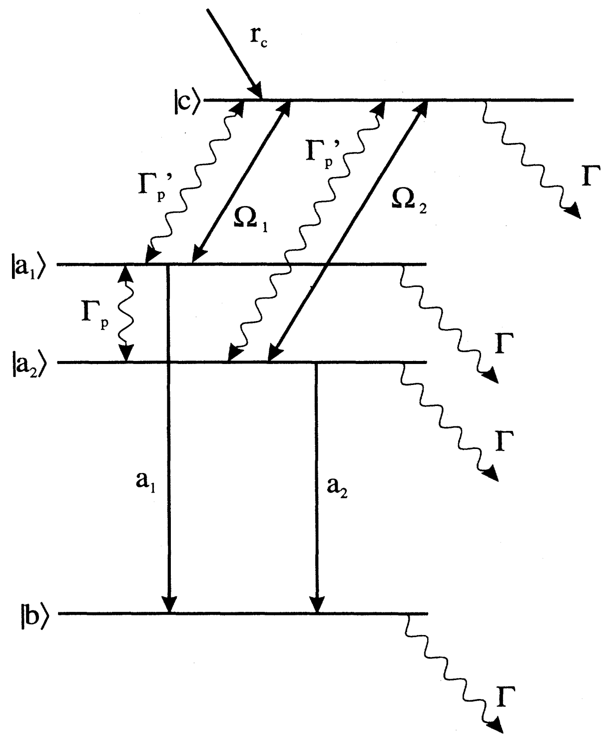

FIG. 1. Atomic level scheme for Raman-driven quantum beat CEL action.

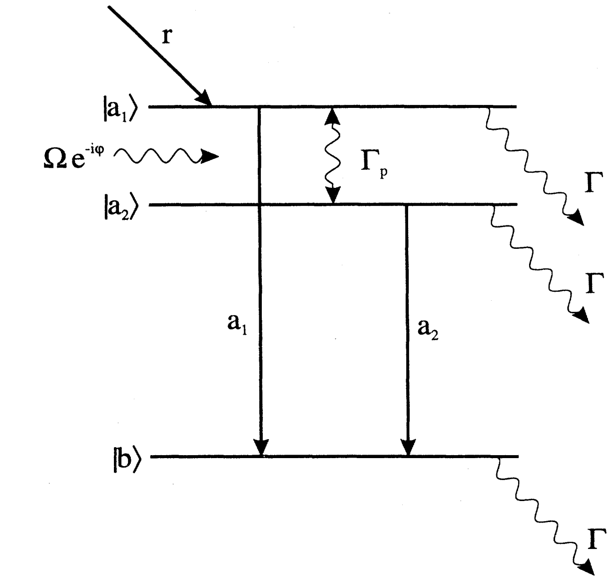

FIG. 2. Atomic level scheme for directly driven quantum beat CEL action.

Note in particular that the transverse decay decreases the degree of coherence irrespective of the strength of the driving fields. Thus it is not possible to wipe out the depletion of the coherences due to the transverse decay by simply turning up the driving field. Indeed, as the results of the present paper show, the transverse decay rates survive in the linear gain and cross-coupling constant, even in the strongly driven limit, i.e., the transverse decay does influence the laser properties (but not the possibility for complete suppression of phase diffusion).

In the directly driven (microwave-driven) quantum beat CEL depicted in Fig. 2, coherence between the two upper levels $|a_1\rangle$ and $|a_2\rangle$ is generated by an external resonant driving field with Rabi frequency Ω. In this scheme the degree of coherence generated by the driving field is not maximum, even in the absence of transverse decay ($\Gamma_p=0$), but instead given by $d_{\text{coh}}=\Gamma/(2\Gamma^2+\Omega^2)^{1/2}<1$. Therefore, we may have at best $d_{\text{coh}}=1/\sqrt{2}$. This is somewhat surprising, since it has been shown [2] that this configuration allows for ideal CEL action.

In technical terms this apparent paradox is easily resolved if one realizes that it is the relative size of the linear gain and cross-coupling coefficients that decides over CEL action, as per Eq. (24). These coefficients α_{ij} $(i,j=1,2)$ are not always simply proportional to the respective atomic population and coherences ρ_{ij}. Instead, they contain contributions from populations and coherences in the externally driven schemes. Therefore, the phase-diffusion constant given by Eq. (24) depends on the steady-state atomic density matrix in a more subtle manner, and the logic cannot be simply transplanted from the CEL with injected coherence (where indeed $\alpha_{ij}\propto\rho_{ij}$) to the driven schemes. In other words, ideal coherence in the sense $d_{\text{coh}}=1$ is *not* a necessary condition for ideal CEL action.

Nevertheless, if an atomic scheme like the directly driven quantum beam CEL displays ideal CEL action in the absence of nonradiative transverse decay, then the inclusion of such transverse decay may be expected to lead to residual phase diffusion. After all, the transverse decay decreases the coherences in the system below the levels at which ideal CEL is initially predicted. However, as is shown in the present paper, ideal CEL action is still possible also in the directly driven quantum beat CEL in the presence of nonradiative transverse decay.

Apart from gaining theoretical insight into the role of atomic coherence in different CEL schemes, the considerations presented here are of relevance to the experimental implementation of CEL schemes. Although experimental results demonstrating CEL action were obtained in gas lasers [20,21] without detailed theoretical investigations of the effect of atomic collisions, the present results show that such collisions are not the limiting factor and that the improvement of these experiments therefore does not depend on the suppression of atomic collisions.

II. LINEAR GAIN AND CROSS-COUPLING COEFFICIENTS

Correlated-spontaneous-emission lasing was originally predicted using a general two-mode field master equation, in which the two oscillator modes are coupled according to [2],

$$\dot{\rho}_f=\sum_{ij}\mathcal{M}_{ij}\rho_f\ , \tag{2}$$

where the operator $\mathcal{M}$ is given by

$$\mathcal{M}_{ii}\rho_f = -\tfrac{1}{2}[\alpha_{ii}\rho_f a_i a_i^\dagger + \alpha_{ii}^* a_i a_i^\dagger \rho_f - (\alpha_{ii}+\alpha_{ii}^*)a_i^\dagger \rho_f a_i] \quad (i=1,2)\,, \tag{3}$$

$$\mathcal{M}_{12}\rho_f = -\tfrac{1}{2}[\alpha_{12}\rho_f a_1^\dagger a_2 + \alpha_{21}^* a_1^\dagger a_2 \rho_f - (\alpha_{12}+\alpha_{21}^*)a_1^\dagger \rho_f a_2]e^{i\Phi(t)}\,, \tag{4}$$

$$\mathcal{M}_{21}\rho_f = -\tfrac{1}{2}[\alpha_{21}\rho_f a_1 a_2^\dagger + \alpha_{12}^* a_1 a_2^\dagger \rho_f - (\alpha_{21}+\alpha_{12}^*)a_2^\dagger \rho_f a_1]e^{-i\Phi(t)}\,. \tag{5}$$

Here, the α_{ij} $(i,j=1,2)$ are the linear gain and cross-coupling coefficients and $\Phi(t)$ is a phase that is determined by the details of the laser medium and is as yet unknown. We have neglected cavity losses and mode pulling terms because they do not influence the noise properties of the CEL.

In the case of the directly driven quantum beat CEL (see Fig. 2), the coefficients α_{ij} have been derived in Ref. [2] without taking into account coherence-destroying collisions. In this system two lasing transitions couple two closely spaced upper levels $|a_1\rangle$ and $|a_2\rangle$ to a common lower level $|b\rangle$. The coherence between $|a_1\rangle$ and $|a_2\rangle$ is generated by an external field with Rabi frequency Ω. The system is pumped by injecting atoms in state $|a_1\rangle$ into the interaction region with a constant rate r, and all states decay with a common rate Γ to some other atomic levels.

We show in the present paper how an additional phase decay Γ_p dephasing the upper two levels influences the CEL properties of the system. In Appendix A we calculate linear gain and cross-coupling coefficients, including such additional phase decay starting from the interaction picture Hamiltonian

$$H=\hbar g(a_1 e^{i\Delta_1 t}|a_1\rangle\langle b| + a_2 e^{i\Delta_2 t}|a_2\rangle\langle b|) - \frac{\hbar\Omega e^{-i\phi}}{2}|a_1\rangle\langle a_2| + \text{H.a.}\,, \tag{6}$$

where we have made the following assumptions: First, the coupling constants between the two transitions $|a_1\rangle \to |b\rangle$ and $|a_2\rangle \to |b\rangle$ and the modes with annihilation operators a_1 and a_2, respectively, are equal and called g. Second, the coherence-inducing driving field with Rabi frequency Ω and phase ϕ is resonant with the transition $|a_1\rangle \to |a_2\rangle$. The detunings Δ_1 and Δ_2 are defined by $\Delta_i = \omega_{a_i b} - \nu_i$ $(i=1,2)$ with the atomic transition frequencies $\omega_{\alpha\beta}$ and the mode oscillation frequencies ν_i. We obtain

$$\alpha_{11} = \frac{rg^2}{2\Gamma D_1}\{(\Gamma+\Gamma_p)[\Omega^2+2\Gamma(2\Gamma+\Gamma_p)] + 2i\Delta_1[\Omega^2+2\Gamma(\Gamma+\Gamma_p)]\}\,, \tag{7}$$

$$\alpha_{22} = \frac{rg^2}{2\Gamma D_2}\{\Omega^2(3\Gamma+\Gamma_p+2i\Delta_2)\}\,, \tag{8}$$

$$\alpha_{12} = \frac{rg^2}{2\Gamma D_2}\{-i\Omega[\Gamma(2\Gamma+\Gamma_p)-\Omega^2+2i\Delta_2\Gamma]\}\,, \tag{9}$$

$$\alpha_{21} = \frac{rg^2}{2\Gamma D_1}\{i\Omega[\Gamma(4\Gamma+3\Gamma_p)+\Omega^2+2i\Delta_1\Gamma]\}\,, \tag{10}$$

where we used

$$D_j = [\Omega^2+\Gamma(\Gamma+\Gamma_p)]\left[\frac{2\Gamma+\Gamma_p}{2}+i\left[\Delta_j+\frac{\Omega}{2}\right]\right] \times \left[\frac{2\Gamma+\Gamma_p}{2}+i\left[\Delta_j-\frac{\Omega}{2}\right]\right] \quad (j=1,2)\,. \tag{11}$$

As in Ref. [2], $\Phi(t)$ is given by $\Phi(t)=(\nu_1-\nu_2-\omega_{a_1a_2})t-\phi$.

In addition to the directly driven quantum beat CEL, we investigate a different kind of coherence-inducing mechanism in the present paper. The atomic level scheme for this case is depicted in Fig. 1. Again, two upper levels $|a_1\rangle$ and $|a_2\rangle$ are coupled to a common lower level $|b\rangle$ via two lasing transitions. But now the coherence between $|a_1\rangle$ and $|a_2\rangle$ is generated by coupling these upper levels to an auxiliary level $|c\rangle$ by fields Ω_1 and Ω_2, respectively, in a Raman-like interaction. The system is pumped by injecting atoms in state $|c\rangle$ at a rate r_c, and all levels decay at the same rate Γ to some other atomic levels outside the system of interest. In addition to this longitudinal decay, we include additional transverse decay between levels $|a_1\rangle$ and $|a_2\rangle$ at rate Γ_p and between $|c\rangle$ and $|a_i\rangle$ $(i=1,2)$ at a rate Γ_p'. This transverse decay may be due to collisions between lasing atoms or other dephasing processes.

For this Raman-driven quantum beat CEL, we start with the interaction picture Hamiltonian

$$H=\hbar g(a_1 e^{i\Delta_1 t}|a_1\rangle\langle b| + a_2 e^{i\Delta_2 t}|a_2\rangle\langle b|) - \frac{\hbar\Omega_1}{2}|c\rangle\langle a_1| - \frac{\hbar\Omega_2}{2}|c\rangle\langle a_2| + \text{H.a.}\,, \tag{12}$$

where again the driving fields are assumed to be resonant. For the calculation of the linear gain and cross-coupling coefficients, we refer to Appendix B. There we obtain in the simplified case when $\Omega_1=\Omega_2=\Omega$ and $\Delta_1=\Delta_2=\Delta$,

$$\alpha_{11}=\alpha_{22}=\frac{r_c g^2}{D}\frac{\Omega^2(2\Gamma\Gamma_{ac}+\Omega^2)}{Z_{ab}(2Z_{ab}Z_{bc}+\Omega^2)} \times \left[\Omega^2\left[\frac{\Gamma_{aa}}{\Gamma}-1\right]+\frac{2\Gamma_{aa}}{\Gamma}(\Gamma+2Z_{bc})Z_{ab}\right]\,, \tag{13}$$

$$\alpha_{12}=\alpha_{21}=\frac{r_c g^2}{D}\frac{\Omega^2(2\Gamma\Gamma_{ac}+\Omega^2)}{Z_{ab}(2Z_{ab}Z_{bc}+\Omega^2)} \times \left[-\Omega^2\left[\frac{\Gamma_{aa}}{\Gamma}-1\right]+2(\Gamma_{aa}+2Z_{bc})Z_{ab}\right]\,, \tag{14}$$

with

$$Z_{\alpha\beta}=\Gamma_{\alpha\beta}+i\Delta\,, \tag{15}$$

$$\Gamma_{aa}=\Gamma+\Gamma_p\,, \tag{16}$$

$$\Gamma_{ab}=\Gamma+\Gamma_p/2\,, \tag{17}$$

$$\Gamma_{ac}=\Gamma+\Gamma'_p\ , \tag{18}$$

$$\Gamma_{bc}=\Gamma+(\Gamma'_p-\Gamma_p/2)\ , \tag{19}$$

$$D=4\Gamma^2\Gamma_{aa}\Gamma_{ac}^2+2\Gamma\Gamma_{ac}\Omega^2(\Gamma+4\Gamma_{aa})+\Omega^4(\Gamma+3\Gamma_{aa})\ , \tag{20}$$

and $\Phi(t)=(\nu_1-\nu_2-\omega_{a_1a_2})t$. For the derivation of Eqs. (16)–(19), see Appendix C. (Note that Γ_p and Γ'_p are not independent quantities and that we always have $\Gamma'_p-\Gamma_p/2>0$.)

III. CEL ACTION

We now show the possibility for CEL action in several cases. Let us first consider the directly driven quantum beat scheme. As a particularly simple case, we set the two detunings equal, $\Delta_1=\Delta_2=\Omega/2$, and assume $\Omega\gg\Gamma$ [cf. Ref. [2], Eq. (28)]. Then Eqs. (7)–(10) reduce to

$$\alpha_{ij}=\alpha=\frac{rg^2}{2\Gamma(\Gamma+\Gamma_p/2)}\quad (i,j=1,2)\ . \tag{21}$$

The presence of Γ_p in the denominator of α shows that the CEL medium is indeed seeing the influence of the collisions, even in the limit of strong driving, as mentioned in the Introduction. This behavior is related to results in the two-photon CEL with phase fluctuations in the injected coherence [22].

The locking equation for the difference phase $\Psi=\theta_1-\theta_2+\phi$ of the respective field mode phases θ_i then reads [2]

$$\dot\Psi=\tfrac{1}{2}\ \mathrm{Im}\left[\alpha_{11}-\alpha_{22}+\alpha_{12}\frac{\rho_2}{\rho_1}e^{-i\Psi}-\alpha_{21}\frac{\rho_1}{\rho_2}e^{i\Psi}\right]\ , \tag{22}$$

$$\dot\Psi=-\alpha\sin\Psi\ , \tag{23}$$

where we have set the mean photon numbers equal, $\rho_1=\rho_2$. From Eq. (23), we see that the difference phase locks to zero. On the other hand, the phase-diffusion coefficient for the beat signal $\langle a_1^\dagger a_2\rangle$ is given by [2]

$$D_\Psi=\tfrac{1}{4}\ \mathrm{Re}\left[\frac{\alpha_{11}}{\rho_1^2}+\frac{\alpha_{22}}{\rho_2^2}-\frac{(\alpha_{12}+\alpha_{21}^*)e^{-i\Psi}}{\rho_1\rho_2}\right]\ , \tag{24}$$

$$D_\Psi=\frac{\alpha}{2\rho^2}(1-\cos\Psi)\ , \tag{25}$$

such that we have vanishing phase diffusion in the difference phase, even for collisional relaxation of the upper two levels. This result holds in the limit of strong driving, $\Omega\gg\Gamma$, and it may be argued that stronger and stronger driving simply compensates the coherence-destroying mechanisms. However, we can also generalize the case of Eq. (27) of Ref. [2] to

$$\Delta_1=\frac{\Gamma+\Gamma_p}{2\Gamma\Omega}[\Omega^2+2\Gamma(2\Gamma+\Gamma_p)]\ , \tag{26}$$

$$\Delta_2=-\frac{3\Gamma+\Gamma_p}{2\Gamma}\Omega\ . \tag{27}$$

For these detunings, we find, after a somewhat lengthy calculation without assumptions about the magnitude of Ω, that indeed $\mathrm{Re}(\alpha_{11}+\alpha_{22}-\alpha_{12}-\alpha_{21})=0$, as required for $D_\Psi|_{\Psi=0}=0$. This somewhat surprising result means that we can compensate for collisional and other phase-destroying mechanisms by adjusting the detunings rather than driving the system harder.

We now switch to the Raman-driven quantum beat CEL. After inserting our result for the linear gain and cross-coupling coefficients Eqs. (13) and (14), D_Ψ reads

$$D_\Psi=\frac{r_cg^2\Omega^2}{2\rho^2D\Gamma_{ab}}\frac{2\Gamma\Gamma_{ac}+\Omega^2}{2\Gamma_{ab}\Gamma_{bc}+\Omega^2}\times\left[2\Gamma_{aa}\Gamma_{ab}(1-\cos\Psi)+4\Gamma_{ab}\Gamma_{bc}\left[\frac{\Gamma_{aa}}{\Gamma}-\cos\Psi\right]+\Omega^2\left[\frac{\Gamma_{aa}}{\Gamma}-1\right](1+\cos\Psi)\right]\ , \tag{28}$$

where the simplification $\Delta=0$ has been made. In the case of vanishing additional transverse decay, i.e., $\Gamma_p=\Gamma'_p=0$, this result simplifies to

$$D_\Psi=\frac{r_cg^2\Omega^2(1-\cos\Psi)}{2\rho^2(2\Gamma^2+\Omega^2)(\Gamma^2+2\Omega^2)}\ . \tag{29}$$

For Ψ locked to 0, we therefore have ideal CEL action, $D_\Psi=0$. This mode of operation is indeed possible, as can be seen from the equation of motion for the phase difference Ψ in the present case,

$$\dot\Psi=-\alpha_{12}\sin\Psi\ . \tag{30}$$

This relation tells us that Ψ locks to 0.

Note that this mode of ideal CEL action was derived without assumptions about the strength of the driving fields. In this sense the present Raman-driven CEL may be advantageous to implement in an experimental situation. Another aspect of the usefulness of this atomic level scheme is the fact that only dipole-allowed transitions are to be driven, in contrast to the directly driven CEL.

In the more general case of nonvanishing Γ_p and Γ'_p, Eq. (30) is still valid, and the phase-diffusion coefficient given by Eq. (28) simplifies with $\Psi=0$ to

$$D_\Psi|_{\Psi=0}=\frac{r_cg^2\Omega^2(2\Gamma\Gamma_{ac}+\Omega^2)(\Gamma_{aa}/\Gamma-1)}{\rho^2D\Gamma_{ab}}\ . \tag{31}$$

Here we clearly see the influence of collisional phase decay: The term $(\Gamma_{aa}/\Gamma-1)=\Gamma_p/\Gamma$ is responsible for the residual phase diffusion due to additional transverse decay and vanishes for $\Gamma_p=0$ such that $D_\Psi|_{\Psi=0}=0$, corresponding to ideal CEL action.

However, the Raman-driven quantum beat CEL also allows for ideal CEL action in the presence of collisional phase decay: In the case of nonvanishing detuning, $\Delta=\Omega/\sqrt{2}$, we obtain in the limit of strong driving, $\Omega\gg\Gamma$,

$$\alpha_{ij}=\alpha=\frac{r_cg^2(\Gamma+\Gamma_{aa})}{\Gamma(\Gamma+3\Gamma_{aa})(\Gamma_{ab}+\Gamma_{bc})}\ . \tag{32}$$

Thus, from Eqs. (22) and (24) we again have ideal CEL action, $D_\Psi|_{\Psi=0}=0$, irrespective of the phase decay.

IV. DISCUSSION

In the present paper we have shown that *ideal* CEL action is possible, even in the presence of nonradiative phase decay induced, for example, by atomic collisions. Ideal CEL action can be achieved by strong driving at certain detunings or, in the case of the directly driven quantum beat CEL, by the choice of the detunings alone. This result is noteworthy insofar as even for strong driving fields the dephasing rate between the upper two lasing levels $|a_1\rangle$ and $|a_2\rangle$ does indeed influence the system behavior described by the linear gain and cross-coupling coefficients α_{ij} $(i,j=1,2)$.

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APPENDIX A: LINEAR GAIN AND CROSS-COUPLING COEFFICIENTS FOR THE DIRECTLY DRIVEN QUANTUM BEAT CEL

With the Hamiltonian Eq. (6), the master equation for the radiation field can be obtained from

$$\begin{aligned}\dot{\rho}_f &= -\frac{i}{\hbar}\,\mathrm{Tr}_{\mathrm{atom}}[H,\rho]\\ &= -\frac{i}{\hbar}(\langle a_1|[H,\rho]|a_1\rangle + \langle a_2|[H,\rho]|a_2\rangle\\ &\quad + \langle b|[H,\rho]|b\rangle)\ ,\end{aligned} \tag{A1}$$

$$\begin{aligned}\dot{\rho}_f &= ige^{-i\Delta_1 t}(\rho_{a_1 b}a_1^\dagger - a_1^\dagger\rho_{a_1 b})\\ &\quad + ige^{-i\Delta_2 t}(\rho_{a_2 b}a_2^\dagger - a_2^\dagger\rho_{a_2 b}) + \text{H.a.}\end{aligned} \tag{A2}$$

Therefore, we have to find $\rho_{a_i b}$ $(i=1,2)$. In order to accomplish this task, we apply a perturbative approach and first calculate the subsequently needed density-matrix elements to zeroth order in the coupling constant g. Note that we do not have a rotating frame for our system as in general $\Delta_1 \neq \Delta_2$ and the driving field is assumed resonant. Therefore, we have to solve for the time-dependent one-atom density matrix first and integrate over the injection times later. The equations of motion for the relevant zeroth-order one-atom matrix elements are

$$\dot{\rho}^{(0)}_{a_1a_1} = -\Gamma\rho^{(0)}_{a_1a_1} + i\frac{\Omega}{2}(e^{-i\phi}\rho^{(0)}_{a_2a_1} - e^{i\phi}\rho^{(0)}_{a_1a_2})\ , \tag{A3}$$

$$\dot{\rho}^{(0)}_{a_2a_2} = -\Gamma\rho^{(0)}_{a_2a_2} - i\frac{\Omega}{2}(e^{-i\phi}\rho^{(0)}_{a_2a_1} - e^{i\phi}\rho^{(0)}_{a_1a_2})\ , \tag{A4}$$

$$\dot{\rho}^{(0)}_{a_1a_2} = -(\Gamma+\Gamma_p)\rho^{(0)}_{a_1a_2} + i\frac{\Omega e^{-i\phi}}{2}(\rho^{(0)}_{a_2a_2} - \rho^{(0)}_{a_1a_1})\ . \tag{A5}$$

The solution to this system subject to the initial condition $\rho^{(0)}_{a_1a_1}(0)=1$ is

$$\rho^{(0)}_{a_1a_1}(t) = \frac{e^{-\Gamma(t-t_0)}}{2}\left\{1 + e^{-\Gamma_p(t-t_0)/2}\left[\cos[\Omega'(t-t_0)] + \frac{\Gamma_p}{2\Omega'}\sin[\Omega'(t-t_0)]\right]\right\}\rho_f(t)\ , \tag{A6}$$

$$\rho^{(0)}_{a_2a_2}(t) = \frac{e^{-\Gamma(t-t_0)}}{2}\left\{1 - e^{-\Gamma_p(t-t_0)/2}\left[\cos[\Omega'(t-t_0)] + \frac{\Gamma_p}{2\Omega'}\sin[\Omega'(t-t_0)]\right]\right\}\rho_f(t)\ , \tag{A7}$$

$$\rho^{(0)}_{a_1a_2}(t) = -\frac{i\Omega'}{2\Omega}e^{-(\Gamma+\Gamma_p/2)(t-t_0)-i\phi}\sin[\Omega'(t-t_0)]\left[1 + \frac{\Gamma_p^2}{4\Omega'^2}\right]\rho_f(t)\ , \tag{A8}$$

where $\Omega' = \sqrt{\Omega^2 - \Gamma_p^2/4}$. The equations of motion for the first-order density-matrix elements are

$$\dot{\rho}^{(1)}_{a_1b} = -\left[\Gamma + \frac{\Gamma_p}{2}\right]\rho^{(1)}_{a_1b} + i\frac{\Omega e^{-i\phi}}{2}\rho^{(1)}_{a_2b} + ig(e^{i\Delta_1 t}\rho^{(0)}_{a_1a_1}a_1 + e^{i\Delta_2 t}\rho^{(0)}_{a_1a_2}a_2)\ , \tag{A9}$$

$$\dot{\rho}^{(1)}_{a_2b} = -\left[\Gamma + \frac{\Gamma_p}{2}\right]\rho^{(1)}_{a_2b} + i\frac{\Omega e^{i\phi}}{2}\rho^{(1)}_{a_1b} + ig(e^{i\Delta_1 t}\rho^{(0)}_{a_2a_1}a_1 + e^{i\Delta_2 t}\rho^{(0)}_{a_2a_2}a_2)\ . \tag{A10}$$

These equations can be integrated and yield

$$\begin{aligned}\rho^{(1)}_{a_1b}(t) = ig\int_{t_0}^{t}dt'e^{-(\Gamma+\Gamma_p/2)(t-t')}\Bigg\{&e^{i\Delta_1 t'}\left[\cos\left[\frac{\Omega}{2}(t-t')\right]\rho^{(0)}_{a_1a_1}(t') + ie^{-i\phi}\sin\left[\frac{\Omega}{2}(t-t')\right]\rho^{(0)}_{a_2a_1}(t')\right]a_1\\ &+ e^{i\Delta_2 t'}\left[\cos\left[\frac{\Omega}{2}(t-t')\right]\rho^{(0)}_{a_1a_2}(t') + ie^{-i\phi}\sin\left[\frac{\Omega}{2}(t-t')\right]\rho^{(0)}_{a_2a_2}(t')\right]a_2\Bigg\}\ ,\end{aligned} \tag{A11}$$

$$\rho^{(1)}_{a_2b}(t)=ig\int_{t_0}^{t}dt'e^{-(\Gamma+\Gamma_p/2)(t-t')}\left\{e^{i\Delta_1t'}\left[\cos\left[\frac{\Omega}{2}(t-t')\right]\rho^{(0)}_{a_2a_1}(t')+ie^{i\phi}\sin\left[\frac{\Omega}{2}(t-t')\right]\rho^{(0)}_{a_1a_1}(t')\right]a_1\right.$$
$$\left.+e^{i\Delta_2t'}\left[\cos\left[\frac{\Omega}{2}(t-t')\right]\rho^{(0)}_{a_2a_2}(t')+ie^{i\phi}\sin\left[\frac{\Omega}{2}(t-t')\right]\rho^{(0)}_{a_1a_2}(t')\right]a_2\right\}. \tag{A12}$$

Inserting the zeroth-order elements from Eqs. (A6)–(A8) and integrating over the injection times according to

$$\rho_{a_ib}(t)=r\int_{-\infty}^{t}dt_0\rho^{(1)}_{a_ib}(t,t_0)\ , \tag{A13}$$

leads, after a lengthy calculation, to

$$\rho_{a_1b}(t)=\frac{i}{2g}e^{i\Delta_1t}\alpha_{11}\rho_f a_1+\frac{i}{2g}e^{i\Delta_2t-i\phi}\alpha_{12}\rho_f a_2\ , \tag{A14}$$

$$\rho_{a_2b}(t)=\frac{i}{2g}e^{i\Delta_1t+i\phi}\alpha_{21}\rho_f a_1+\frac{i}{2g}e^{i\Delta_2t}\alpha_{22}\rho_f a_2\ . \tag{A15}$$

Using Eqs. (2) and (A2), we identify $\Phi(t)=(\nu_1-\nu_2-\omega_{a_1a_2})t-\phi$ and the linear gain and cross-coupling coefficients, as given in Eqs. (7)–(10).

APPENDIX B: LINEAR GAIN AND CROSS-COUPLING COEFFICIENTS FOR THE RAMAN-DRIVEN QUANTUM BEAT CEL

The master equation for the radiation field can be obtained from Eq. (A1). The trace of the atomic parts for the Hamiltonian Eq. (12) is given by

$$\mathrm{Tr}_{\mathrm{atom}}[H,\rho]=\langle a_1|[H,\rho]|a_1\rangle+\langle a_2|[H,\rho]|a_2\rangle$$
$$+\langle b|[H,\rho]|b\rangle+\langle c|[H,\rho]|c\rangle\ , \tag{B1}$$

$$\mathrm{Tr}_{\mathrm{atom}}[H,\rho]=-\hbar g e^{-i\Delta_1t}(\rho_{a_1b}a_1^\dagger-a_1^\dagger\rho_{a_1b})$$
$$-\hbar g e^{-i\Delta_2t}(\rho_{a_2b}a_2^\dagger-a_2^\dagger\rho_{a_2b})-\text{ H.a.} \tag{B2}$$

Therefore, we have to find ρ_{a_ib} $(i=1,2)$. In order to accomplish this task, we could proceed along the lines of Appendix A. However, in this larger atomic level scheme, it is more convenient to first consider the case $\Delta_1=\Delta_2=\Delta$ (then we are able to transform to a rotating frame), and subsequently generalize the result to arbitrary detunings.

In zeroth order, the equations of motion for the relevant population matrix elements are

$$\dot\rho^{(0)}_{a_1c}=-\Gamma_{ac}\rho^{(0)}_{a_1c}+i\frac{\Omega_1}{2}(\rho^{(0)}_{cc}-\rho^{(0)}_{a_1a_1})-i\frac{\Omega_2}{2}\rho^{(0)}_{a_1a_2}\ , \tag{B3}$$

$$\dot\rho^{(0)}_{a_2c}=-\Gamma_{ac}\rho^{(0)}_{a_2c}+i\frac{\Omega_2}{2}(\rho^{(0)}_{cc}-\rho^{(0)}_{a_2a_2})-i\frac{\Omega_1}{2}\rho^{(0)}_{a_2a_1}\ , \tag{B4}$$

$$\dot\rho^{(0)}_{a_1a_1}=-\Gamma\rho^{(0)}_{a_1a_1}+i\frac{\Omega_1}{2}\rho^{(0)}_{ca_1}-i\frac{\Omega_1}{2}\rho^{(0)}_{a_1c}\ , \tag{B5}$$

$$\dot\rho^{(0)}_{a_2a_2}=-\Gamma\rho^{(0)}_{a_2a_2}+i\frac{\Omega_2}{2}\rho^{(0)}_{ca_2}-i\frac{\Omega_2}{2}\rho^{(0)}_{a_2c}\ , \tag{B6}$$

$$\dot\rho^{(0)}_{cc}=-\Gamma\rho^{(0)}_{cc}+i\frac{\Omega_1}{2}\rho^{(0)}_{a_1c}-i\frac{\Omega_1}{2}\rho^{(0)}_{ca_1}+i\frac{\Omega_2}{2}\rho^{(0)}_{a_2c}$$
$$-i\frac{\Omega_2}{2}\rho^{(0)}_{ca_2}+r_c\rho_f\ , \tag{B7}$$

$$\dot\rho^{(0)}_{a_1a_2}=-\Gamma_{aa}\rho^{(0)}_{a_1a_2}+i\frac{\Omega_1}{2}\rho^{(0)}_{ca_2}-i\frac{\Omega_2}{2}\rho^{(0)}_{a_1c}\ , \tag{B8}$$

where we have assumed real Rabi frequencies Ω_1 and Ω_2. Here, $\Gamma_{ac}=\Gamma+\Gamma_p'$ and $\Gamma_{aa}=\Gamma+\Gamma_p$ (see Appendix C). Applying the adiabatic approximation, the time derivatives on the left-hand side are set to zero, and we obtain for the subsequently needed matrix elements

$$\rho^{(0)}_{a_1c}=\frac{ir_c\Omega_1}{2D}G_{21}\rho_f\ , \tag{B9}$$

$$\rho^{(0)}_{a_2c}=\frac{ir_c\Omega_2}{2D}G_{12}\rho_f\ , \tag{B10}$$

$$\rho^{(0)}_{a_1a_1}=\frac{r_c\Omega_1^2}{2\Gamma D}G_{21}\rho_f\ , \tag{B11}$$

$$\rho^{(0)}_{a_2a_2}=\frac{r_c\Omega_2^2}{2\Gamma D}G_{12}\rho_f\ , \tag{B12}$$

$$\rho^{(0)}_{a_1a_2}=\frac{r_c\Omega_1\Omega_2}{2D}\tilde\Omega^2\rho_f\ , \tag{B13}$$

where we have introduced the auxiliary quantities

$$G_{12}=2\Gamma_{aa}(2\Gamma\Gamma_{ac}+\Omega_1^2)+\Gamma(\Omega_2^2-\Omega_1^2)\ , \tag{B14}$$

$$G_{21}=G_{12}|_{1\leftrightarrow2}\ , \tag{B15}$$

$$\tilde\Omega^2=4\Gamma\Gamma_{ac}+\Omega_1^2+\Omega_2^2\ , \tag{B16}$$

$$D=4\Gamma^2\Gamma_{aa}\Gamma_{ac}^2+\Gamma\Gamma_{ac}(\Gamma+4\Gamma_{aa})(\Omega_1^2+\Omega_2^2)$$
$$+\Gamma(\Omega_1^4-\Omega_1^2\Omega_2^2+\Omega_2^4)+3\Gamma_{aa}\Omega_1^2\Omega_2^2\ . \tag{B17}$$

After changing into a rotating frame in which $\rho_{a_ib}\to\rho_{a_ib}e^{-i\Delta t}$ $(i=1,2)$ and $\rho_{bc}\to\rho_{bc}e^{i\Delta t}$, the relevant equations for the first-order matrix elements are

$$\dot{\rho}^{(1)}_{a_1b} = -(\Gamma_{ab}+i\Delta)\rho^{(1)}_{a_1b} + i\frac{\Omega_1}{2}\rho^{(1)}_{cb} + ig\rho^{(0)}_{a_1a_1}a_1 + ig\rho^{(0)}_{a_1a_2}a_2 \ , \tag{B18}$$

$$\dot{\rho}^{(1)}_{a_2b} = -(\Gamma_{ab}+i\Delta)\rho^{(1)}_{a_2b} + i\frac{\Omega_2}{2}\rho^{(1)}_{cb} + ig\rho^{(0)}_{a_2a_2}a_2 + ig\rho^{(0)}_{a_2a_1}a_1 \ , \tag{B19}$$

$$\dot{\rho}^{(1)}_{cb} = -(\Gamma_{bc}+i\Delta)\rho^{(1)}_{cb} + i\frac{\Omega_1}{2}\rho^{(1)}_{a_1b} + i\frac{\Omega_2}{2}\rho^{(1)}_{a_2b} + ig\rho^{(0)}_{ca_1}a_1 + ig\rho^{(0)}_{ca_2}a_2 \ , \tag{B20}$$

where $\Gamma_{ab}=(2\Gamma+\Gamma_p)/2$ and $\Gamma_{bc}=\Gamma+(\Gamma'_p-\Gamma_p/2)$ (see Appendix C). Upon applying the adiabatic approximation once more, we obtain

$$\rho^{(1)}_{a_1b} = \frac{i}{2g}(\alpha_{11}\rho_f a_1 + \alpha_{12}\rho_f a_2) \ , \tag{B21}$$

$$\rho^{(1)}_{a_2b} = \frac{i}{2g}(\alpha_{21}\rho_f a_1 + \alpha_{22}\rho_f a_2) \ , \tag{B22}$$

with the linear gain and cross-coupling coefficients α_{ij} $(i,j=1,2)$

$$\alpha_{11} = \frac{r_c g^2}{DD'}\left\{\frac{1}{\Gamma}G_{21}\Omega_1^2[2(\Gamma+2Z_{bc})Z_{ab}+\Omega_2^2] - \Omega_1^2\Omega_2^2\tilde{\Omega}^2\right\} \ , \tag{B23}$$

$$\alpha_{22} = \alpha_{11}|_{1\leftrightarrow 2} \ , \tag{B24}$$

$$\alpha_{12} = \frac{r_c g^2}{DD'}\left\{\frac{1}{\Gamma}G_{12}\Omega_1\Omega_2[2\Gamma Z_{ab}-\Omega_2^2] + (4Z_{ab}Z_{bc}+\Omega_2^2)\Omega_1\Omega_2\tilde{\Omega}^2\right\} \ , \tag{B25}$$

$$\alpha_{21} = \alpha_{12}|_{1\leftrightarrow 2} \ , \tag{B26}$$

with $Z_{ij}=\Gamma_{ij}+i\Delta$ and

$$D' = Z_{ab}(4Z_{ab}Z_{bc}+\Omega_1^2+\Omega_2^2) \ . \tag{B27}$$

For the simplified case $\Omega_1=\Omega_2=\Omega$, this results in the linear gain and cross-coupling coefficients Eqs. (13) and (14).

Note that we can generalize our results to arbitrary detunings if we write out the first-order equations Eqs. (B18)–(B20), but this time not in a rotating frame,

$$\dot{\rho}^{(1)}_{a_1b} = -\Gamma_{ab}\rho^{(1)}_{a_1b} + i\frac{\Omega_1}{2}\rho^{(1)}_{cb} + ig(e^{i\Delta_1 t}\rho^{(0)}_{a_1a_1}a_1 + e^{i\Delta_2 t}\rho^{(0)}_{a_1a_2}a_2) \ , \tag{B28}$$

$$\rho^{(1)}_{a_2b} = -\Gamma_{ab}\rho^{(1)}_{a_2b} + i\frac{\Omega_2}{2}\rho^{(1)}_{cb} + ig(e^{i\Delta_2 t}\rho^{(0)}_{a_2a_2}a_2 + e^{i\Delta_1 t}\rho^{(0)}_{a_2a_1}a_1) \ , \tag{B29}$$

$$\dot{\rho}^{(1)}_{cb} = -\Gamma_{bc}\rho^{(1)}_{cb} + i\frac{\Omega_1}{2}\rho^{(1)}_{a_1b} + i\frac{\Omega_2}{2}\rho^{(1)}_{a_2b} + ig(e^{i\Delta_1 t}\rho^{(0)}_{ca_1}a_1 + e^{i\Delta_2 t}\rho^{(0)}_{ca_2}a_2) \ . \tag{B30}$$

We see that the time dependences always come in as $e^{i\Delta_j t}a_j$ $(j=1,2)$, just as in Eqs. (A9) and (A10) for the directly driven quantum CEL. Therefore, we can obtain the general result for arbitrary detunings by substituting $\Delta\rightarrow\Delta_1$ in Eqs. (B23) and (B26), and likewise $\Delta\rightarrow\Delta_2$ in Eqs. (B24) and (B25).

APPENDIX C: TRANSVERSE DECAY RATES IN THE RAMAN-DRIVEN QUANTUM BEAT CEL

If we just consider the time dependence of the atomic coherence $\rho_{\alpha\beta}$ $(\alpha,\beta=a_1,a_2,b,c;\ a\neq\beta)$ due to collisions or similar phase-destroying mechanisms, we may write

$$\dot{\rho}_{\alpha\beta}(t) = -i\omega_{\alpha\beta}\rho_{\alpha\beta}(t) = -i[\omega^0_{\alpha\beta}+\delta\omega_\alpha(t)-\delta\omega_\beta(t)]\rho_{\alpha\beta}(t) \ . \tag{C1}$$

Here, $\omega^0_{\alpha\beta}$ is the transition frequency in the absence of perturbations. We treated the collisions by introducing δ-function-correlated independent fluctuations of the atomic energy levels $\delta\omega_\alpha(t)$ and $\delta\omega_\beta(t)$, fulfilling $\langle\delta\omega_i(t)\delta\omega_j(t')\rangle_{\text{coll}}=\delta_{ij}2\gamma_i\delta(t-t')$ $(i,j=\alpha$ or $\beta)$. The subscript coll denotes an average over the stochastic process. Formal integration yields

$$\rho_{\alpha\beta}(t) = e^{-i\omega^0_{\alpha\beta}t}\rho_{\alpha\beta}(0) - i\int_0^t dt' e^{-i\omega^0_{\alpha\beta}(t-t')}[\delta\omega_\alpha(t')-\delta\omega_\beta(t')]\rho_{\alpha\beta}(t') \ . \tag{C2}$$

We insert this result back into Eq. (C1) and carry out the integration. This leads to

$$\langle\dot{\rho}_{\alpha\beta}\rangle_{\text{coll}} = -(i\omega^0_{\alpha\beta}+\gamma_\alpha+\gamma_\beta)\langle\rho_{\alpha\beta}\rangle_{\text{coll}} \ . \tag{C3}$$

We now assume $\gamma_{a_1}=\gamma_{a_2}$ and $\gamma_b=0$, define $\Gamma_p=2\gamma_a$ and $\Gamma'_p=\gamma_a+\gamma_c$, and obtain the decay rates given in Eqs. (16)–(19).

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