# SUPPLEMENTAL MATERIAL
# Dressed Atom Revisited: Hamiltonian-Independent Treatment of the Radiative Cascade


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This Supplemental Material contains the detailed derivation of the results reported in the main text.


## DERIVATION OF THE MASTER EQUATION

The dissipative dynamics of the dressed atom is determined by the atom-radiation interaction Hamiltonian

$$V_{\mathrm{AR}} = \hbar \int \mathrm{d}\omega \sqrt{\frac{\Gamma(\omega)}{2\pi}} \left( \mathcal{S}_+ a(\omega) + \mathcal{S}_- a^\dagger(\omega) \right), \tag{S1}$$

where $\Gamma(\omega)$ vanishes if $\omega \leq 0$. Due to the structure of the atom-laser Hamiltonian

$$H_{\mathrm{AL}} = \sum_{j=1,2} \sum_N E_j(N) |j(N)\rangle\langle j(N)|, \quad \text{with } E_j(N) = \hbar \left( N\omega_L - (-1)^j \frac{\Omega}{2} \right), \tag{S2}$$

the atom-radiation dynamics can yield on-shell decay of the dressed eigenstates at the frequencies $q\omega_L$ and $q\omega_L \pm \Omega$.

Using the assumption that, for all practical purposes, the composition of the dressed eigenstates $|j(N)\rangle$ in terms of the bare states $|e, N'\rangle$ and $|g, N'\rangle$ only depends on $N - N'$, the atomic transition operators $\mathcal{S}_+ = |e\rangle\langle g|$ and $\mathcal{S}_- = |g\rangle\langle e|$ can be conveniently expressed as

$$\mathcal{S}_+ = \sum_{i,j\in\{1,2\}} \sum_q A_{ij}^{(q)} S_+^{ij(q)}, \qquad \mathcal{S}_- = \sum_{i,j\in\{1,2\}} \sum_q \big(A_{ij}^{(q)}\big)^* S_-^{ij(q)} \tag{S3}$$

with the coefficients

$$A_{ij}^{(q)} = \sum_p \langle i(N+p+q)|e, N\rangle\langle g, N|j(N+p)\rangle \tag{S4}$$

actually independent of $N$, and

$$S_+^{ij(q)} = \sum_N |i(N)\rangle\langle j(N-q)|, \qquad S_-^{ij(q)} = \sum_N |j(N-q)\rangle\langle i(N)|. \tag{S5}$$

The Lindblad operators associated to the allowed transitions can thus be expressed as

$$L_+^{(q)} = \sum_N \sqrt{\Gamma\left(\frac{E_1(N) - E_2(N-q)}{\hbar}\right)} \big(A_{12}^{(q)}\big)^* |2(N-q)\rangle\langle 1(N)| = \sqrt{\Gamma(q\omega_L + \Omega)} \big(A_{12}^{(q)}\big)^* S_-^{12(q)}, \tag{S6}$$

$$L_-^{(q)} = \sum_N \sqrt{\Gamma\left(\frac{E_2(N) - E_1(N-q)}{\hbar}\right)} \big(A_{21}^{(q)}\big)^* |1(N-q)\rangle\langle 2(N)| = \sqrt{\Gamma(q\omega_L - \Omega)} \big(A_{21}^{(q)}\big)^* S_-^{21(q)}, \tag{S7}$$

$$L_c^{(q)} = \sum_{jN} \sqrt{\Gamma\left(\frac{E_j(N) - E_j(N-q)}{\hbar}\right)} \big(A_{jj}^{(q)}\big)^* |j(N-q)\rangle\langle j(N)| = \sqrt{\Gamma(q\omega_L)} \left[ \big(A_{11}^{(q)}\big)^* S_-^{11(q)} + \big(A_{22}^{(q)}\big)^* S_-^{22(q)} \right]. \tag{S8}$$

Therefore, the dissipative part of the GKLS master equation, applied to the atom-laser density matrix $\sigma$, reads

$$\begin{aligned} \mathcal{L}_{\mathrm{diss}}(\sigma) &= \sum_{n\in\{+,-,c\}} \sum_q \left[ L_n^{(q)} \sigma \big(L_n^{(q)}\big)^\dagger - \frac{1}{2} \left\{ \big(L_n^{(q)}\big)^\dagger L_n^{(q)}, \sigma \right\} \right] \\ &= \sum_q \left[ \sum_{i,j\in\{1,2\}} \Gamma_{ij}^{(q)} \left( S_-^{ij(q)} \sigma S_+^{ij(q)} - \frac{1}{2} \left\{ S_+^{ij(q)} S_-^{ij(q)}, \sigma \right\} \right) + K_{12}^{(q)} S_-^{11(q)} \sigma S_+^{22(q)} + \big(K_{12}^{(q)}\big)^* S_-^{22(q)} \sigma S_+^{11(q)} \right], \end{aligned} \tag{S9}$$

where

$$\Gamma_{11}^{(q)} = \Gamma(q\omega_L)\big|A_{11}^{(q)}\big|^2, \quad \Gamma_{22}^{(q)} = \Gamma(q\omega_L)\big|A_{22}^{(q)}\big|^2, \quad \Gamma_{12}^{(q)} = \Gamma(q\omega_L+\Omega)\big|A_{12}^{(q)}\big|^2, \quad \Gamma_{21}^{(q)} = \Gamma(q\omega_L-\Omega)\big|A_{21}^{(q)}\big|^2, \tag{S10}$$

and

$$K_{12}^{(q)} = \Gamma(q\omega_L)\big|A_{11}^{(q)}A_{22}^{(q)}\big|\mathrm{e}^{\mathrm{i}\varphi(q)} = \sqrt{\Gamma_{11}^{(q)}\Gamma_{22}^{(q)}}\mathrm{e}^{\mathrm{i}\varphi(q)}, \tag{S11}$$

with $\varphi(q)$ the relative phase between $A_{22}^{(q)}$ and $\big(A_{11}^{(q)}\big)^*$ and $\{A,B\} = AB+BA$ the anticommutator in the atom-laser Hilbert space. Expanding the expression (S9) in terms of the transition operators $S_\pm^{(q)}$ provides

$$\begin{aligned}\mathcal{L}_{\mathrm{diss}}(\sigma) = \sum_{qNN'}\Bigg[&\sum_{ij}\Gamma_{ij}^{(q)}\left(\sigma_{i(N),i(N')}|j(N-q)\rangle\langle j(N'-q)| - \frac{\delta_{NN'}}{2}\left\{|i(N)\rangle\langle i(N)|,\sigma\right\}\right)\\ &+ K_{12}^{(q)}\sigma_{1(N),2(N')}|1(N-q)\rangle\langle 2(N'-q)| + \big(K_{12}^{(q)}\big)^*\sigma_{2(N),1(N')}|2(N-q)\rangle\langle 1(N'-q)|\Bigg],\end{aligned} \tag{S12}$$

with

$$\sigma_{i(N),j(N')} = \langle i(N)|\sigma|j(N')\rangle. \tag{S13}$$

## FULL AND REDUCED ATOM-LASER DYNAMICS

The evolution of the atom-laser density matrix $\sigma$ is determined by the combination of the Markovian dissipative dynamics (S12) and the Hamiltonian dynamics yielded by $H_{\mathrm{AL}}$:

$$\frac{\mathrm{d}}{\mathrm{d}t}\sigma(t) = -\frac{\mathrm{i}}{\hbar}\left[H_{\mathrm{AL}},\sigma(t)\right] + \mathcal{L}_{\mathrm{diss}}(\sigma(t)) = \frac{\mathrm{i}}{\hbar}\sum_{j,N}E_j(N)\left[\sigma(t),|j(N)\rangle\langle j(N)|\right] + \mathcal{L}_{\mathrm{diss}}(\sigma(t)). \tag{S14}$$

From the above expression and from (S12), one can obtain the coupled evolution equations for each element:

$$\dot\sigma_{1(N),1(N')} = \mathrm{i}(N'-N)\omega_L\sigma_{1(N),1(N')} + \sum_q\left[\Gamma_{11}^{(q)}\sigma_{1(N+q),1(N'+q)} + \Gamma_{21}^{(q)}\sigma_{2(N+q),2(N'+q)} - \left(\Gamma_{11}^{(q)}+\Gamma_{12}^{(q)}\right)\sigma_{1(N),1(N')}\right], \tag{S15}$$

$$\dot\sigma_{2(N),2(N')} = \mathrm{i}(N'-N)\omega_L\sigma_{2(N),2(N')} + \sum_q\left[\Gamma_{12}^{(q)}\sigma_{1(N+q),1(N'+q)} + \Gamma_{22}^{(q)}\sigma_{2(N+q),2(N'+q)} - \left(\Gamma_{21}^{(q)}+\Gamma_{22}^{(q)}\right)\sigma_{2(N),2(N')}\right], \tag{S16}$$

$$\dot\sigma_{1(N),2(N')} = \mathrm{i}[(N-N')\omega_L-\Omega]\sigma_{1(N),2(N')} + \sum_q\left[-\frac{\Gamma_{11}^{(q)}+\Gamma_{12}^{(q)}+\Gamma_{21}^{(q)}+\Gamma_{22}^{(q)}}{2}\sigma_{1(N),2(N')} + K_{12}^{(q)}\sigma_{1(N+q),2(N'+q)}\right], \tag{S17}$$

$$\dot\sigma_{2(N),1(N')} = \mathrm{i}[(N-N')\omega_L+\Omega]\sigma_{2(N),1(N')} + \sum_q\left[-\frac{\Gamma_{11}^{(q)}+\Gamma_{12}^{(q)}+\Gamma_{21}^{(q)}+\Gamma_{22}^{(q)}}{2}\sigma_{2(N),1(N')} + \big(K_{12}^{(q)}\big)^*\sigma_{2(N+q),1(N'+q)}\right]. \tag{S18}$$

After introducing the reduced density matrix elements

$$\sigma_{ij}^{(\ell)} = \sum_N\sigma_{i(N),j(N+\ell)}, \tag{S19}$$

one can derive their dynamics by summing the above equations on $N$. For the elements with different indices, one gets the decoupled equations

$$\dot\sigma_{12}^{(\ell)} = \left[\mathrm{i}(\ell\omega_L-\Omega)-\Gamma_{\mathrm{coh}}\right]\sigma_{12}^{(\ell)}, \qquad \dot\sigma_{21}^{(\ell)} = \left[\mathrm{i}(\ell\omega_L+\Omega)-\Gamma_{\mathrm{coh}}\right]\sigma_{21}^{(\ell)} \tag{S20}$$

with

$$\Gamma_{\rm coh} = \sum_q \left[ \frac{\Gamma_{11}^{(q)} + \Gamma_{12}^{(q)} + \Gamma_{21}^{(q)} + \Gamma_{22}^{(q)}}{2} - {\rm Re}\left(K_{12}^{(q)}\right) \right], \tag{S21}$$

which can be integrated as

$$\sigma_{12}^{(\ell)}(t+\tau) = \exp\left[{\rm i}(\ell\omega_L - \Omega)\tau - \Gamma_{\rm coh}\tau\right] \sigma_{12}^{(\ell)}(t), \quad \sigma_{21}^{(\ell)}(t+\tau) = \exp\left[{\rm i}(\ell\omega_L + \Omega)\tau - \Gamma_{\rm coh}\tau\right] \sigma_{21}^{(\ell)}(t), \tag{S22}$$

respectively. Notice that the imaginary parts of $K_{12}^{(q)}$ have been consistently neglected with respect to the $O(\ell\omega_L \pm \Omega)$ terms. It is also interesting to observe that, in the standard RWA evaluation, the real part of $K_{12}^{(1)}$ is negative, thus providing an increase in the coherence decay rate. The reason will become clear in the last section.

The equal-index reduced density matrix elements satisfy the coupled equations

$$\dot{\sigma}_{11}^{(\ell)} = \left[{\rm i}\ell\omega_L - \Gamma_{12}\right] \sigma_{11}^{(\ell)} + \Gamma_{21}\sigma_{22}^{(\ell)}, \qquad \dot{\sigma}_{22}^{(\ell)} = \left[{\rm i}\ell\omega_L - \Gamma_{21}\right] \sigma_{22}^{(\ell)} + \Gamma_{12}\sigma_{11}^{(\ell)}. \tag{S23}$$

A specific case is represented by the populations

$$\Pi_j = \sigma_{jj}^{(0)} = \sum_N \langle j(N)|\sigma|j(N)\rangle \qquad \text{with } j = 1, 2, \tag{S24}$$

whose evolution, satisfying $\Pi_1(t) + \Pi_2(t) = 1$ at all times, converges to the steady-state values

$$\Pi_1^{\rm st} = \frac{\Gamma_{21}}{\Gamma_{\rm pop}}, \qquad \Pi_2^{\rm st} = \frac{\Gamma_{12}}{\Gamma_{\rm pop}}, \qquad \text{with } \Gamma_{\rm pop} = \Gamma_{12} + \Gamma_{21}, \tag{S25}$$

while the coherences approach the steady-state behavior

$$\sigma_{jj,{\rm st}}^{(\ell)}(t) = {\rm e}^{{\rm i}\ell\omega_L t}\Pi_j^{\rm st}. \tag{S26}$$

In general, the coupled equations (S23) can be integrated into

$$\sigma_{11}^{(\ell)}(t+\tau) = {\rm e}^{{\rm i}\ell\omega_L\tau} \left[ \left(\Pi_1^{\rm st} + \Pi_2^{\rm st}{\rm e}^{-\Gamma_{\rm pop}\tau}\right) \sigma_{11}^{(\ell)}(t) + \Pi_1^{\rm st}\left(1 - {\rm e}^{-\Gamma_{\rm pop}\tau}\right) \sigma_{22}^{(\ell)}(t) \right], \tag{S27}$$

$$\sigma_{22}^{(\ell)}(t+\tau) = {\rm e}^{{\rm i}\ell\omega_L\tau} \left[ \Pi_2^{\rm st}\left(1 - {\rm e}^{-\Gamma_{\rm pop}\tau}\right) \sigma_{11}^{(\ell)}(t) + \left(\Pi_2^{\rm st} + \Pi_1^{\rm st}{\rm e}^{-\Gamma_{\rm pop}\tau}\right) \sigma_{22}^{(\ell)}(t) \right]. \tag{S28}$$

Therefore, the quantity $\Gamma_{\rm pop}$ defined in Eq. (S25) plays the role of a relaxation rate towards the steady-state regime.

## SPECTRAL DENSITY OF EMITTED PHOTONS

The spectral density of the emitted photons can be obtained starting from the Heisenberg evolution of the field operators [1] entailed by the Hamiltonian

$$H = H_{\rm AL} + \hbar \int {\rm d}\omega\, \omega a^\dagger(\omega)a(\omega) + \hbar \int {\rm d}\omega \sqrt{\frac{\Gamma(\omega)}{2\pi}} \left(\mathcal{S}_+ a(\omega) + \mathcal{S}_- a^\dagger(\omega)\right), \tag{S29}$$

yielding

$$a(\omega, t) = a(\omega){\rm e}^{-{\rm i}\omega t} - {\rm i}\sqrt{\frac{\Gamma(\omega)}{2\pi}} \int_0^t {\rm d}t'\, \mathcal{S}_-(t'){\rm e}^{-{\rm i}\omega(t-t')}, \tag{S30}$$

where the integration constants are set in such a way that at the initial time $t = 0$, when the state of the atom-laser-radiation system is

$$\rho(0) = \sigma(0) \otimes |{\rm vac}\rangle\langle{\rm vac}| \tag{S31}$$

with $a(\omega)|{\rm vac}\rangle = 0$, the Heisenberg operator $a(\omega, 0)$ coincides with the Schrödinger operator $a(\omega)$, acting only on the radiation degrees of freedom.

After an evolution time $T$, the probability density associated to photon frequency reads

$$\mathrm{Tr}\left[a^\dagger(\omega)a(\omega)\rho(T)\right] = \langle a^\dagger(\omega,T)a(\omega,T)\rangle = \frac{\Gamma(\omega)}{2\pi}\int_0^T \mathrm{d}t \int_0^T \mathrm{d}t' \langle \mathcal{S}_+(t')\mathcal{S}_-(t)\rangle \mathrm{e}^{-\mathrm{i}\omega(t'-t)}$$
$$= \frac{\Gamma(\omega)}{\pi}\mathrm{Re}\int_0^T \mathrm{d}t \int_0^{T-t} \mathrm{d}\tau \langle \mathcal{S}_+(t+\tau)\mathcal{S}_-(t)\rangle \mathrm{e}^{-\mathrm{i}\omega\tau} \tag{S32}$$

with $\langle \dots \rangle$ the average on the initial state $\rho(0)$. The evolution towards a steady state entails the presence of constant contributions in $t$, yielding diverging integrals, and oscillating ones, which are not integrable but provide comparatively negligible contributions. Therefore, it is convenient to define the asymptotic spectral density as

$$\mathcal{J}(\omega) = \lim_{T\to\infty} \frac{\langle a^\dagger(\omega,T)a(\omega,T)\rangle}{T} = \lim_{T\to\infty} \frac{\Gamma(\omega)}{\pi}\mathrm{Re}\int_0^T \frac{\mathrm{d}t}{T}\int_0^{T-t} \mathrm{d}\tau \langle \mathcal{S}_+(t+\tau)\mathcal{S}_-(t)\rangle \mathrm{e}^{-\mathrm{i}\omega\tau}. \tag{S33}$$

The correlator in the integral can be evaluated by using the quantum regression theorem (QRT) [1], which is consistent with the applicability condition of the Markovian approximation, namely the fact that the time scale of vacuum correlations (in this case, the width of the Fourier transform of $\Gamma(\omega)$) is much smaller than any other scale of time variation in the open system dynamics. Since $\mathcal{S}_+$ can be written as

$$\mathcal{S}_+ = \sum_{i,j\in\{1,2\}}\sum_q \mathcal{S}_+^{ij(q)}, \qquad \text{with } \mathcal{S}_+^{ij(q)} = A_{ij}^{(q)}\sum_N |i(N)\rangle\langle j(N-q)|, \tag{S34}$$

the spectral density can be accordingly decomposed in the following terms,

$$\mathcal{J}(\omega) = \sum_q \left(\mathcal{J}_+^{(q)}(\omega) + \mathcal{J}_-^{(q)}(\omega) + \mathcal{J}_c^{(q)}(\omega)\right) \tag{S35}$$

which will be now derived separately.

## Sidebands

From the definitions (S34) and (S19), one can easily obtain that

$$\langle \mathcal{S}_+^{ij(q)}\rangle = A_{ij}^{(q)}\sigma_{ji}^{(q)}. \tag{S36}$$

Therefore, the evolution (S22) of the unequal-index coherences determines

$$\langle \mathcal{S}_+^{12(q)}(t+\tau)\rangle = \exp\left[\mathrm{i}(q\omega_L+\Omega)\tau - \Gamma_{\mathrm{coh}}\tau\right]\langle \mathcal{S}_+^{12(q)}(t)\rangle. \tag{S37}$$

From QRT, $\langle \mathcal{S}_+^{ij(q)}(t+\tau)\mathcal{S}_-(t)\rangle$ follows the same evolution in $\tau$ as $\langle \mathcal{S}_+^{ij(q)}(t+\tau)\rangle$, and therefore

$$\langle \mathcal{S}_+^{12(q)}(t+\tau)\mathcal{S}_-(t)\rangle = \exp\left[\mathrm{i}(q\omega_L+\Omega)\tau - \Gamma_{\mathrm{coh}}\tau\right]\langle \mathcal{S}_+^{12(q)}(t)\mathcal{S}_-(t)\rangle. \tag{S38}$$

The expectation value of $\mathcal{S}_+^{12(q)}\mathcal{S}_-$ at time $t$ can be evaluated by using the expression (S3), as

$$\langle \mathcal{S}_+^{12(q)}(t)\mathcal{S}_-(t)\rangle = A_{12}^{(q)}\sum_{j\in\{1,2\}}\sum_\ell \left(A_{j2}^{(\ell)}\right)^*\sigma_{j1}^{(q-\ell)}(t) = A_{12}^{(q)}\sum_\ell \left(A_{12}^{(\ell)}\right)^*\Pi_1^{\mathrm{st}}\mathrm{e}^{\mathrm{i}(q-\ell)\omega_L t} + f_{12}^{(q)}(t), \tag{S39}$$

where $f_{12}^{(q)}(t)$ contains transient contributions in $t$, which certainly do not contribute to the spectral density, as defined in (S33). Combining the above results, one obtains the spectral density contribution

$$\mathcal{J}_+^{(q)}(\omega) = \lim_{T\to\infty}\frac{\Gamma(\omega)}{\pi}\mathrm{Re}\int_0^T \mathrm{d}t\int_0^{T-t}\mathrm{d}\tau\langle \mathcal{S}_+^{12(q)}(t+\tau)\mathcal{S}_-(t)\rangle \mathrm{e}^{-\mathrm{i}\omega\tau}$$
$$= \frac{\Gamma(\omega)}{\pi}\left|A_{12}^{(q)}\right|^2\Pi_1^{\mathrm{st}}\,\mathrm{Re}\int_0^\infty \mathrm{d}\tau\,\mathrm{e}^{-\mathrm{i}(\omega-q\omega_L-\Omega)\tau-\Gamma_{\mathrm{coh}}\tau}$$
$$= \Gamma_{12}^{(q)}\Pi_1^{\mathrm{st}}\frac{1}{\pi}\frac{\Gamma_{\mathrm{coh}}}{[\omega-(q\omega_L+\Omega)]^2+\Gamma_{\mathrm{coh}}^2} \tag{S40}$$

with the last equality holding up to $O(\Gamma_{\rm coh})$ corrections, considering that $\Gamma_{12}^{(q)} = \Gamma(q\omega_L + \Omega)\big|A_{12}^{(q)}\big|^2$. The computation of the lower-energy sideband contribution

$$\mathcal{J}_-^{(q)}(\omega) = \lim_{T\to\infty} \frac{\Gamma(\omega)}{\pi} \mathrm{Re} \int_0^T \mathrm{d}t \int_0^{T-t} \mathrm{d}\tau \langle \mathcal{S}_+^{21(q)}(t+\tau)\mathcal{S}_-(t)\rangle \mathrm{e}^{-\mathrm{i}\omega\tau} = \Gamma_{21}^{(q)} \Pi_2^{\rm st} \frac{1}{\pi} \frac{\Gamma_{\rm coh}}{[\omega - (q\omega_L - \Omega)]^2 + \Gamma_{\rm coh}^2} \tag{S41}$$

proceeds in an entirely analogous way, with the roles of indexes 1 and 2 exchanged, and the sign in front of $\Omega$ reversed.

### Central lines

The evaluation of the central-line contributions

$$\mathcal{J}_c^{(q)}(\omega) = \lim_{T\to\infty} \frac{\Gamma(\omega)}{\pi} \mathrm{Re} \int_0^T \mathrm{d}t \int_0^{T-t} \mathrm{d}\tau \langle \Big[\mathcal{S}_+^{11(q)}(t+\tau) + \mathcal{S}_+^{22(q)}(t+\tau)\Big] \mathcal{S}_-(t)\rangle \mathrm{e}^{-\mathrm{i}\omega\tau} \tag{S42}$$

is complicated by the fact that the evolutions of $\langle \mathcal{S}_+^{jj(q)}\rangle = A_{jj}^{(q)} \sigma_{jj}^{(q)}$ for $j = 1$ and 2 are coupled to each other. Actually, following the evolution (S27)-(S28) of the same-index reduced coherences, one obtains

$$\langle \mathcal{S}_+^{11(q)}(t+\tau) + \mathcal{S}_+^{22(q)}(t+\tau)\rangle = \mathrm{e}^{\mathrm{i}q\omega_L\tau} \left[\left(C_1^{(q)} + D_1^{(q)} \mathrm{e}^{-\Gamma_{\rm pop}\tau}\right) \langle \mathcal{S}_+^{11(q)}(t)\rangle + \left(C_2^{(q)} + D_2^{(q)} \mathrm{e}^{-\Gamma_{\rm pop}\tau}\right) \langle \mathcal{S}_+^{22(q)}(t)\rangle\right] \tag{S43}$$

with

$$C_1^{(q)} = \Pi_1^{\rm st} + \frac{A_{22}^{(q)}}{A_{11}^{(q)}} \Pi_2^{\rm st}, \quad C_2^{(q)} = \Pi_2^{\rm st} + \frac{A_{11}^{(q)}}{A_{22}^{(q)}} \Pi_1^{\rm st}, \quad D_1^{(q)} = \Pi_1^{\rm st} \left(1 - \frac{A_{22}^{(q)}}{A_{11}^{(q)}}\right), \quad D_2^{(q)} = \Pi_2^{\rm st} \left(1 - \frac{A_{11}^{(q)}}{A_{22}^{(q)}}\right). \tag{S44}$$

Notice that in the standard dressed-atom approach, in which RWA is considered in the atom-laser interaction, such a computation is oversimplified by the property $A_{22}^{(1)} = -A_{11}^{(1)}$ [1] (with all the other coefficients for $q \neq 1$ vanishing). Using QRT, we obtain

$$\begin{aligned} &\langle \Big[\mathcal{S}_+^{11(q)}(t+\tau) + \mathcal{S}_+^{22(q)}(t+\tau)\Big] \mathcal{S}_-(t)\rangle \\ &\qquad = \mathrm{e}^{\mathrm{i}q\omega_L\tau} \left[\left(C_1^{(q)} + D_1^{(q)} \mathrm{e}^{-\Gamma_{\rm pop}\tau}\right) \langle \mathcal{S}_+^{11(q)}(t)\mathcal{S}_-(t)\rangle + \left(C_2^{(q)} + D_2^{(q)} \mathrm{e}^{-\Gamma_{\rm pop}\tau}\right) \langle \mathcal{S}_+^{22(q)}(t)\mathcal{S}_-(t)\rangle\right]. \end{aligned} \tag{S45}$$

The expectation values appearing in the above expression can be evaluated as in the case of sidebands, using (S34) and (S3),

$$\langle \mathcal{S}_+^{11(q)}(t)\mathcal{S}_-(t)\rangle = A_{11}^{(q)} \sum_{j\in\{1,2\}} \sum_\ell \big(A_{j1}^{(\ell)}\big)^* \sigma_{j1}^{(q-\ell)}(t) = A_{11}^{(q)} \sum_\ell \big(A_{11}^{(\ell)}\big)^* \Pi_1^{\rm st} \mathrm{e}^{\mathrm{i}(q-\ell)\omega_L t} + f_{11}^{(q)}(t), \tag{S46}$$

$$\langle \mathcal{S}_+^{22(q)}(t)\mathcal{S}_-(t)\rangle = A_{22}^{(q)} \sum_{j\in\{1,2\}} \sum_\ell \big(A_{j2}^{(\ell)}\big)^* \sigma_{j2}^{(q-\ell)}(t) = A_{22}^{(q)} \sum_\ell \big(A_{22}^{(\ell)}\big)^* \Pi_2^{\rm st} \mathrm{e}^{\mathrm{i}(q-\ell)\omega_L t} + f_{22}^{(q)}(t), \tag{S47}$$

where $f_{jj}^{(q)}(t)$ contain transient contributions in $t$. Collecting the above results yields

$$\begin{aligned} \mathcal{J}_c^{(q)}(\omega) &= \frac{\Gamma(\omega)}{\pi} \left[\mathcal{A}^{(q)} \,\mathrm{Re} \int_0^\infty \mathrm{d}\tau\, \mathrm{e}^{-\mathrm{i}(\omega - q\omega_L)\tau} + \mathcal{B}^{(q)} \,\mathrm{Re} \int_0^\infty \mathrm{d}\tau\, \mathrm{e}^{-\mathrm{i}(\omega - q\omega_L)\tau - \Gamma_{\rm pop}\tau}\right] \\ &= \Gamma(q\omega_L)\mathcal{A}^{(q)} \delta(\omega - q\omega_L) + \Gamma(\omega)\mathcal{B}^{(q)} \frac{1}{\pi} \frac{\Gamma_{\rm pop}}{[\omega - (q\omega_L + \Omega)]^2 + \Gamma_{\rm pop}^2}, \end{aligned} \tag{S48}$$

with

$$\mathcal{A}^{(q)} = C_1^q \big|A_{11}^{(q)}\big|^2 \Pi_1^{\rm st} + C_2^q \big|A_{22}^{(q)}\big|^2 \Pi_2^{\rm st} = \left|A_{11}^{(q)} \Pi_1^{\rm st} + A_{22}^{(q)} \Pi_2^{\rm st}\right|^2, \tag{S49}$$

$$\mathcal{B}^{(q)} = D_1^q \big|A_{11}^{(q)}\big|^2 \Pi_1^{\rm st} + D_2^q \big|A_{22}^{(q)}\big|^2 \Pi_2^{\rm st} = \left|A_{11}^{(q)} - A_{22}^{(q)}\right|^2 \Pi_1^{\rm st} \Pi_2^{\rm st}. \tag{S50}$$

After approximating $\Gamma(\omega) \simeq \Gamma(q\omega_L)$, up to $O(\Gamma_{\text{pop}})$ corrections, one can express the spectral density of the $q$-th central line as

$$\mathcal{J}_c^{(q)}(\omega) = \left[\Gamma_{11}^{(q)} \left(\Pi_1^{\text{st}}\right)^2 + \Gamma_{22}^{(q)} \left(\Pi_2^{\text{st}}\right)^2 + 2\,\text{Re}K_{12}^{(q)}\Pi_1^{\text{st}}\Pi_2^{\text{st}}\right] \delta(\omega - q\omega_L) \\ + \left(\Gamma_{11}^{(q)} + \Gamma_{22}^{(q)} - 2\,\text{Re}K_{12}^{(q)}\right) \Pi_1^{\text{st}}\Pi_2^{\text{st}} \frac{1}{\pi} \frac{\Gamma_{\text{pop}}}{(\omega - q\omega_L)^2 + \Gamma_{\text{pop}}^2}. \quad \text{(S51)}$$

It is evident that frequency integration, useful to determine the line weight, cancels the contributions that depend on $K_{12}^{(q)}$.

## PERTURBATIVE CORRECTION OF RWA EIGENSTATES

If the composition of the dressed eigenstates $\{|1(N)\rangle, |2(N)\rangle\}$ in terms of the bare states $|e, N\rangle$ and $|g, N\rangle$ is expressed by the amplitudes

$$\alpha_j^{(p)} = \langle g, N+1-p|j(N)\rangle, \qquad \beta_j^{(p)} = \langle e, N-p|j(N)\rangle, \quad \text{(S52)}$$

then the coefficients in Eq. (S4), which are crucial to determine the features of a specific model, read

$$A_{ij}^{(q)} = \sum_p \left(\beta_i^{(p+q-1)}\right)^* \alpha_j^{(p)}. \quad \text{(S53)}$$

In the case of the standard RWA dressed atom, the eigenstates have the form

$$|1(N)_0\rangle = \sin\theta|g, N+1\rangle + \cos\theta|e, N\rangle, \qquad |2(N)_0\rangle = \cos\theta|g, N+1\rangle - \sin\theta|e, N\rangle, \quad \text{(S54)}$$

implying that the only non-vanishing amplitudes (S52) are those with $p = 0$, and the only non-vanishing coefficients in $\mathcal{S}_+$ are those with $q = 1$:

$$A_{11}^{(1)} = \cos\theta\sin\theta, \quad A_{22}^{(1)} = -\cos\theta\sin\theta, \quad A_{12}^{(1)} = \cos^2\theta, \quad A_{21}^{(1)} = -\sin^2\theta. \quad \text{(S55)}$$

Therefore, only the coefficients

$$A_{ij}^{(0)} = \left(\beta_i^{(0)}\right)^* \alpha_j^{(0)} + \sum_{p\neq 0} \left(\beta_i^{(p)}\right)^* \alpha_j^{(p)} \quad \text{(S56)}$$

contain terms of $O(1)$ in perturbations of the RWA Hamiltonian. Incidentally, notice that the quantity $K_{12}^{(1)}$, as defined in Eq. (S11), is negative, because $A_{11}^{(1)}$ and $A_{22}^{(1)}$ have opposite sign. This is ultimately due to the minus sign appearing in the transformation (S54), that connects two orthonormal bases.

Let us consider the case in which the Hamiltonian

$$H_{\text{AL}} = H_{\text{RW}} + H_{\text{AS}} + H_{\text{CR}} \quad \text{(S57)}$$

contains an "unperturbed" term

$$H_{\text{RW}} = \hbar\omega_0|e\rangle\langle e| \otimes \mathbb{1}_L + \mathbb{1}_A \sum_N N\hbar\omega_L|N\rangle\langle N| + \frac{\hbar\Omega_{\text{R}}}{2} \sum_N \left(|e, N\rangle\langle g, N+1| + |g, N+1\rangle\langle e, N|\right), \quad \text{(S58)}$$

whose eigenstates are of the form (S54), with

$$\tan(2\theta) = \frac{\Omega_{\text{R}}}{\omega_0 - \omega_L}, \quad \text{(S59)}$$

and

$$H_{\text{AS}} = \hbar\Omega_{\text{AS}} \sum_N \left(|e, N\rangle\langle e, N+1| + |e, N+1\rangle\langle e, N|\right), \quad \text{(S60)}$$

$$H_{\text{CR}} = \frac{\hbar\Omega_{\text{R}}}{2} \sum_N \left(|e, N+1\rangle\langle g, N| + |g, N\rangle\langle e, N+1|\right), \quad \text{(S61)}$$

perturbation terms depending on a permanent dipole moment of the atomic state $|e\rangle$ and on the relevance of counter-rotating terms, respectively. Since $H_{\rm AS}$ and $H_{\rm CR}$ do not couple states inside the same doublet, the relevant conditions of perturbation theory applicability are $\Omega_{\rm AS} \ll \omega_L$ and $\Omega_{\rm R} \ll \omega_L$. Moreover, to ensure that RWA is a good zeroth-order approximation, the condition

$$\Omega = \sqrt{\Omega_{\rm R}^2 + (\omega_0 - \omega_L)^2} \ll \omega_L, \tag{S62}$$

stronger than $\Omega_{\rm R} \ll \omega_L$, must hold as well, and $\Omega/\omega_L$ will thus be treated as a small parameter.

Applying perturbation theory at the first order, the RWA eigenstates $|j(N)\rangle$ hybridize with $|j'(N+p)\rangle$ with $p = -1, +1$, due to $H_{\rm AS}$, and $p = -2, +2$, due to $H_{\rm CR}$. The resulting eigenstates read

$$\begin{aligned} |1(N)\rangle = |1(N)_0\rangle &+ \frac{\Omega_{\rm AS}\Omega}{\omega_L(\omega_L+\Omega)}\cos^2\theta\sin\theta|g,N\rangle + \frac{\Omega_{\rm AS}}{\omega_L}\cos\theta\left(1-\frac{\Omega\sin^2\theta}{\omega_L+\Omega}\right)|e,N-1\rangle \\ &+ \frac{\Omega_{\rm AS}\Omega}{\omega_L(\omega_L-\Omega)}\cos^2\theta\sin\theta|g,N+2\rangle - \frac{\Omega_{\rm AS}}{\omega_L}\cos\theta\left(1+\frac{\Omega\sin^2\theta}{\omega_L-\Omega}\right)|e,N+1\rangle \\ &+ \frac{\Omega_{\rm R}}{4\omega_L}\frac{\Omega\sin^2\theta\cos\theta}{2\omega_L-\Omega}|g,N+3\rangle - \frac{\Omega_{\rm R}}{4\omega_L}\sin\theta\left(1+\frac{\Omega\sin^2\theta}{2\omega_L-\Omega}\right)|e,N+2\rangle \\ &+ \frac{\Omega_{\rm R}}{4\omega_L}\cos\theta\left(1-\frac{\Omega\cos^2\theta}{2\omega_L+\Omega}\right)|g,N-1\rangle + \frac{\Omega_{\rm R}}{4\omega_L}\frac{\Omega\sin\theta\cos^2\theta}{2\omega_L+\Omega}|e,N-2\rangle \end{aligned} \tag{S63}$$

and

$$\begin{aligned} |2(N)\rangle = |2(N)_0\rangle &- \frac{\Omega_{\rm AS}\Omega}{\omega_L(\omega_L-\Omega)}\cos\theta\sin^2\theta|g,N\rangle - \frac{\Omega_{\rm AS}}{\omega_L}\sin\theta\left(1+\frac{\Omega\cos^2\theta}{\omega_L-\Omega}\right)|e,N-1\rangle \\ &- \frac{\Omega_{\rm AS}\Omega}{\omega_L(\omega_L+\Omega)}\cos\theta\sin^2\theta|g,N+2\rangle + \frac{\Omega_{\rm AS}}{\omega_L}\sin\theta\left(1-\frac{\Omega\cos^2\theta}{\omega_L+\Omega}\right)|e,N+1\rangle \\ &+ \frac{\Omega_{\rm R}}{4\omega_L}\frac{\Omega\sin\theta\cos^2\theta}{2\omega_L+\Omega}|g,N+3\rangle - \frac{\Omega_{\rm R}}{4\omega_L}\cos\theta\left(1-\frac{\Omega\cos^2\theta}{2\omega_L+\Omega}\right)|e,N+2\rangle \\ &- \frac{\Omega_{\rm R}}{4\omega_L}\cos\theta\left(1+\frac{\Omega\cos^2\theta}{2\omega_L-\Omega}\right)|g,N-1\rangle - \frac{\Omega_{\rm R}}{4\omega_L}\frac{\Omega\sin^2\theta\cos\theta}{2\omega_L-\Omega}|e,N-2\rangle. \end{aligned} \tag{S64}$$

The lowest-order contribution to the coefficients (S4) comes from the terms

$$A_{ij}^{(q)} \simeq \big(\beta_i^{(q-1)}\big)^*\alpha_j^{(0)} + \big(\beta_i^{(0)}\big)^*\alpha_j^{(1-q)} + O\left(\frac{\Omega_{\rm AS}}{\omega_L}\right)^2 + O\left(\frac{\Omega_{\rm R}}{\omega_L}\right)^2. \tag{S65}$$

with corrections of order $(\Omega_{\rm AS}/\omega_L)^2$ and $(\Omega_{\rm R}/\omega_L)^2$. Therefore, the coefficient determining the low-frequency line strength reads

$$A_{12}^{(0)} = -\frac{\Omega_{\rm AS}}{\omega_L}\cos^2\theta\left(1+2\frac{\Omega\sin^2\theta}{\omega_L-\Omega}\right) + O\left[\left(\frac{\Omega_{\rm AS}}{\omega_L}\right)^2\right] = -\frac{\Omega_{\rm AS}}{\omega_L}\cos^2\theta + O\left(\frac{\Omega_{\rm AS}\Omega}{\omega_L^2}\right), \tag{S66}$$

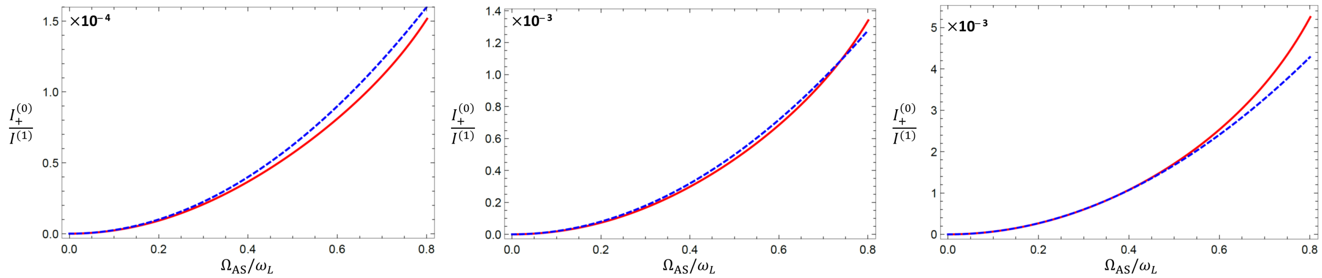
while, within the same approximation order, the coefficients $A_{ij}^{(2)}$ that determine the emission triplet around $2\omega_L$ read

$$A_{11}^{(2)} \simeq \frac{\Omega_{\rm AS}}{\omega_L}\cos\theta\sin\theta, \quad A_{22}^{(2)} \simeq -\frac{\Omega_{\rm AS}}{\omega_L}\cos\theta\sin\theta, \quad A_{12}^{(2)} \simeq \frac{\Omega_{\rm AS}}{\omega_L}\cos^2\theta, \quad A_{21}^{(2)} \simeq -\frac{\Omega_{\rm AS}}{\omega_L}\sin^2\theta. \tag{S67}$$

Notice the proportionality to the unperturbed coefficients $A_{ij}^{(1)}$ in Eq. (S55), implying that the relative weight of the low-energy transition with respect to the $q=1$ and the $q=2$ triplets differs only by a factor $(\Omega_{\rm AS}/\omega_L)^2$ and by the different ratio of form factors. In the case of $q=3$, one can verify that the dominant terms

$$A_{ij}^{(3)} \simeq \langle i(N)|e,N-2\rangle\langle g,N+1|j(N)\rangle + \langle i(N)|e,N\rangle\langle g,N+3|j(N)\rangle = O\left(\frac{\Omega_{\rm R}\Omega}{\omega_L^2}\right) \tag{S68}$$

are suppressed by an additional order $\Omega/\omega_L)$ besides $\Omega_{\rm R}/\omega_L$, leading to the considerations reported in the main text.




(a) $\Omega_{\rm R} = 0.1\omega_L$ (b) $\Omega_{\rm R} = 0.2\omega_L$ (c) $\Omega_{\rm R} = 0.3\omega_L$

Figure S1. Relative weight of the low-frequency emission compared to the Mollow triplet, evaluated in resonance conditions $\omega_0 = \omega_L$ (solid red lines). The quadratic approximation determined by the behavior around $\Omega_{\rm AS} = 0$ is reported for comparison (dashed blue lines). The results are obtained under the assumption that the form factor $\Gamma(\omega)$ of the atomic transition behaves like $\omega^3$ for the transition frequencies involved in the processes.

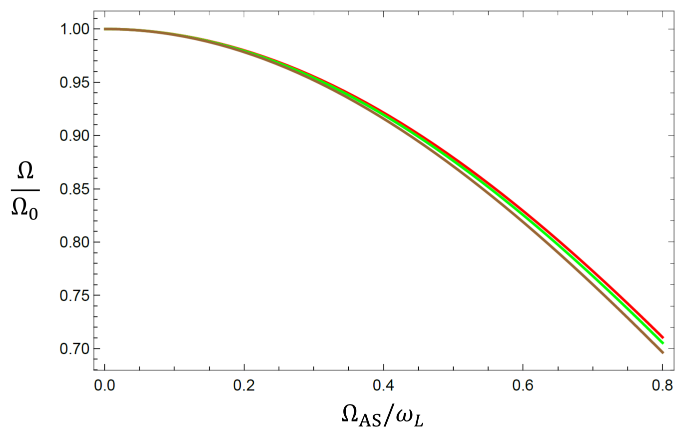



Figure S2. Comparison between the low-frequency gap $\Omega = [E_1(N) - E_2(N)]/\hbar$ obtained with varying $\Omega_{\rm AS}$ and the value $\Omega_0$ obtained for $\Omega_{\rm AS} = 0$. The reported curves are referred to resonance cases ($\omega_0 = \omega_L$) with $\Omega_{\rm R} = 0.1\omega_L$ (red line), $\Omega_{\rm R} = 0.2\omega_L$ (green line), and $\Omega_{\rm R} = 0.3\omega_L$ (brown line).

A non-perturbative numerical diagonalization of the full Hamiltonian $H_{\rm AL}$, truncated to 50 levels, provides the results in Figure S1, which report the relative weight, in terms of photon number, of the low-frequency emission compared to the Mollow triplet, as a function of $\Omega_{\rm AS}$, in resonance conditions for $\Omega_{\rm R} = 0.1\omega_L, 0.2\omega_L, 0.3\omega_L$. Interestingly, as shown in Figure S2, increasing the permanent dipole while keeping $\Omega_{\rm R}$ fixed determines a further reduction of the lowest transition frequency in the system.

[1] C. Cohen-Tannoudji, J. Dupont-Roc, and G. Grynberg, *Atom-Photon Interactions* (John Wiley & Sons, New York, NY, 1998).