Modeling Dye-Mediated Contribution to Photon-Photon Interaction in Condensates of Light

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Theory of Condensed Matter and Many Body Systems

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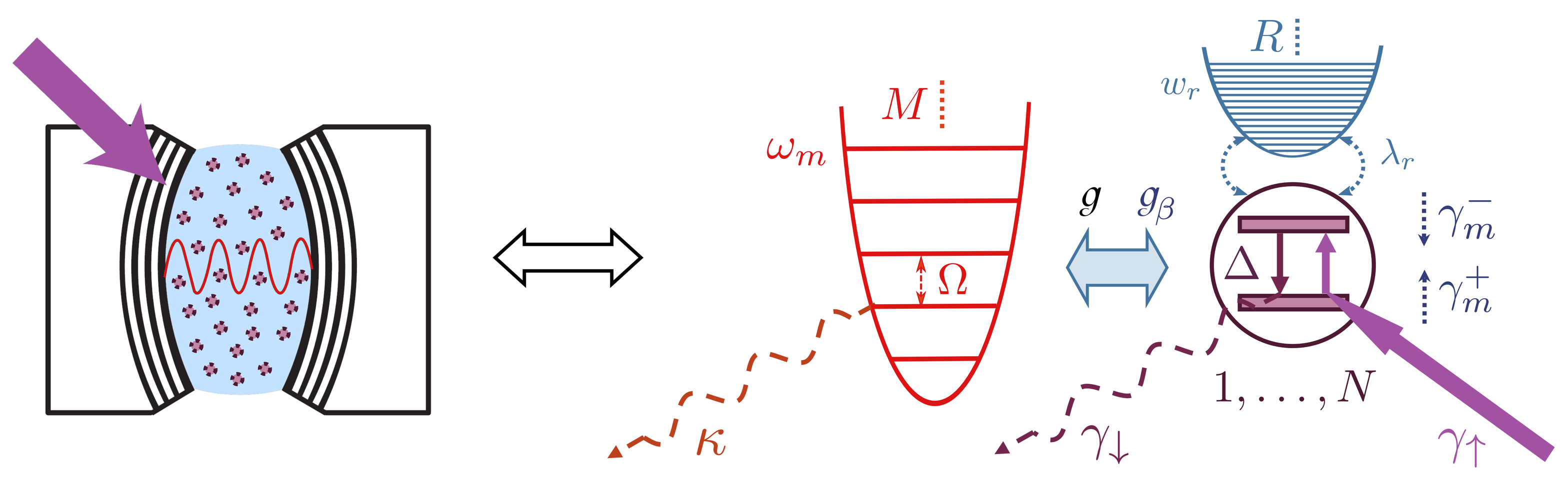
Abstract Based entirely on the Lindblad master equation approach we obtain a microscopic description of photons in a dye-filled cavity [1]. This system features condensation of light [2–4]. The model is a generalization of the known nonequilibrium one [5] and gives us the possibility to determine the dye-mediated contribution to photon-photon interaction in the light condensate. We describe the dynamics of the system using the equations of motion approach [6] and discuss the existence of two limiting cases: photon BEC and laser-like. In the former case, we determine the corresponding dimensionless interaction strength relying on realistic experimental data and find a good agreement with the previous theoretical estimate [7].

Microscopic model [1, 5]



✗ System Hamiltonian:

$$H = \sum_{m=1}^{M} \omega_m a_m^\dagger a_m + \underbrace{\sum_{j=1}^{N} \left(\frac{\Delta}{2}\sigma_j^z + \sum_{r=1}^{R} \left[w_r b_{j,r}^\dagger b_{j,r} + \lambda_r (b_{j,r}^\dagger + b_{j,r}) \sigma_j^z \right] \right)}_{j\text{-th dye molecule + its reservoir of oscillators}} + \underbrace{g \sum_{m=1}^{M} \sum_{j=1}^{N} \left(a_m^\dagger \sigma_j^- + a_m \sigma_j^+ \right)}_{\text{dye-cavity interaction } V_{\rm int}}$$

✗ Polaron transformation

$$\tilde{H} = U H U^\dagger, \quad \text{with} \quad U = \exp\left[\sum_{j=1}^{N} \sigma_j^z \sum_{r=1}^{R} \frac{\lambda_r}{w_r} (b_{j,r}^\dagger - b_{j,r}) \right]$$

leads to

$$\tilde{H} = \sum_{m=1}^{M} \omega_m a_m^\dagger a_m + \sum_{j=1}^{N} \left(\frac{\Delta}{2}\sigma_j^z + \underbrace{\sum_{r=1}^{R} w_r b_{j,r}^\dagger b_{j,r}}_{\text{free part } H_{\rm R}^{(j)}} \right) + \underbrace{g \sum_{m=1}^{M} \sum_{j=1}^{N} \left(a_m^\dagger \sigma_j^- D_j^- + a_m \sigma_j^+ D_j^+ \right)}_{\text{reservoir-dressed interaction } \tilde{V}_{\rm int}}$$

$D_j^\pm = \otimes_{r=1}^{R} \exp\left[\pm \frac{2\lambda_r}{w_r} (b_{j,r}^\dagger - b_{j,r}) \right]$ – polaron displacement operators of j-th dye molecule

Master equation approach

✗ Oscillators in polaron frame represent bath in thermal state

$$\rho_\beta = Z_\beta^{-1} \otimes_{j=1}^{N} \exp\left[-\beta H_{\rm R}^{(j)} \right], \quad \beta = 1/(k_{\rm B}T)$$

which is initially uncorrelated with dye-cavity subsystem $\rho_{\rm total}(0) = \rho_{\rm D,C}(0) \otimes \rho_\beta$

✗ First order bath effect is to reduce coherent dye-cavity coupling

$$\langle \tilde{V}_{\rm int} \rangle_\beta = g_\beta \sum_{m=1}^{M} \sum_{j=1}^{N} \left(a_m^\dagger \sigma_j^- + a_m \sigma_j^+ \right), \quad g_\beta = g \langle D_j^\pm \rangle_\beta < g, \quad \langle D_j^\pm \rangle_\beta = \exp\left[-2 \sum_{r=1}^{R} \frac{\lambda_r^2}{w_r^2} \coth \frac{\beta w_r}{2} \right]$$

$\langle X \rangle_\beta \equiv {\rm Tr}[X \rho_\beta]$ – bath thermal average

✗ Second order bath contribution is dissipative

$$\dot{\rho}_{\rm D,C} = -i \left[\sum_{m=1}^{M} \delta_m a_m^\dagger a_m + g_\beta \sum_{m=1}^{M} \sum_{j=1}^{N} \left(a_m^\dagger \sigma_j^- + a_m \sigma_j^+ \right), \rho_{\rm D,C} \right] - \left\{ \sum_{m=1}^{M} \frac{\kappa}{2} \mathcal{L}[a_m] + \sum_{j=1}^{N} \left(\frac{\gamma_\uparrow}{2} \mathcal{L}[\sigma_j^+] + \frac{\gamma_\downarrow}{2} \mathcal{L}[\sigma_j^-] \right) + \sum_{m=1}^{M} \sum_{j=1}^{N} \left(\frac{\gamma_m^+}{2} \mathcal{L}[a_m \sigma_j^+] + \frac{\gamma_m^-}{2} \mathcal{L}[a_m^\dagger \sigma_j^-] \right) \right\} \rho_{\rm D,C}$$

where $\delta_m = \omega_m - \Delta$, $\mathcal{L}[X]\rho = \{X^\dagger X, \rho\} - 2X\rho X^\dagger$ and

$$\gamma_m^\pm = \gamma(\pm\delta_m), \qquad \gamma(\delta) = 2g^2 {\rm Re} \int_0^\infty e^{-\frac{1}{2}(\gamma_\uparrow + \gamma_\downarrow)t} \left(\langle D_j^-(t) D_j^+ \rangle_\beta - \langle D_j^-(t) \rangle_\beta \langle D_j^+ \rangle_\beta \right) e^{i\delta t} dt$$

References

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Equations-of-motion approach

Equations of motion for expectation values of system observables $\langle X \rangle \equiv {\rm Tr}[X\rho_{\rm D,C}]$ after using cumulant expansion method [6] and keeping cumulants up to second order:

$$\frac{d}{dt}\langle n_m \rangle = -\kappa \langle n_m \rangle + iNg_\beta \left(\langle a_m \sigma_1^+ \rangle - \langle a_m^\dagger \sigma_1^- \rangle \right) - \frac{N}{2}\gamma_m^+ \langle n_m \rangle (1 - \langle \sigma_1^z \rangle) + \frac{N}{2}\gamma_m^- (\langle n_m \rangle + 1)(1 + \langle \sigma_1^z \rangle)$$

$$\frac{d}{dt}\langle \sigma_1^z \rangle = \gamma_\uparrow (1 - \langle \sigma_1^z \rangle) - \gamma_\downarrow (1 + \langle \sigma_1^z \rangle) + 2ig_\beta \sum_{m=1}^{M} \left(\langle a_m^\dagger \sigma_1^- \rangle - \langle a_m \sigma_1^+ \rangle \right) + \sum_{m=1}^{M} \left[\gamma_m^+ \langle n_m \rangle (1 - \langle \sigma_1^z \rangle) - \gamma_m^- (\langle n_m \rangle + 1)(1 + \langle \sigma_1^z \rangle) \right]$$

$$\frac{d}{dt}\langle a_m \sigma_1^+ \rangle = \left(-i\delta_m - \frac{\gamma_\uparrow + \gamma_\downarrow + \kappa}{2} \right) \langle a_m \sigma_1^+ \rangle - ig_\beta \left[\frac{1 + \langle \sigma_1^z \rangle}{2} + \langle \sigma_1^z \rangle \sum_{k=1}^{M} \langle a_k^\dagger a_m \rangle + (N-1) \langle \sigma_1^+ \sigma_2^- \rangle \right] - \frac{\gamma_m^+}{4}(N-1) \langle a_m \sigma_1^+ \rangle (1 - \langle \sigma_1^z \rangle) + \frac{\gamma_m^-}{4}(N-1) \langle a_m \sigma_1^+ \rangle (1 + \langle \sigma_1^z \rangle) - \sum_{k=1}^{M} \left\{ \frac{\gamma_k^+}{2} \left[\langle a_m \sigma_1^+ \rangle \langle n_k \rangle + \langle a_k \sigma_1^+ \rangle \langle a_k^\dagger a_m \rangle \right] + \frac{\gamma_k^-}{2} \left[\langle a_m \sigma_1^+ \rangle (\langle n_k \rangle + 1) + \langle a_k \sigma_1^+ \rangle \langle a_k^\dagger a_m \rangle \right] \right\}$$

$$\frac{d}{dt}\langle a_k^\dagger a_m \rangle = \left[-\kappa + i(\delta_k - \delta_m) \right] \langle a_k^\dagger a_m \rangle + iNg_\beta \left(\langle a_m \sigma_1^+ \rangle - \langle a_k^\dagger \sigma_1^- \rangle \right) - \frac{N}{4} (\gamma_k^+ + \gamma_m^+) \langle a_k^\dagger a_m \rangle (1 - \langle \sigma_1^z \rangle) + \frac{N}{4} (\gamma_k^- + \gamma_m^-) \langle a_m a_k^\dagger \rangle (1 + \langle \sigma_1^z \rangle)$$

$$\frac{d}{dt}\langle \sigma_1^+ \sigma_2^- \rangle = -(\gamma_\uparrow + \gamma_\downarrow) \langle \sigma_1^+ \sigma_2^- \rangle + ig_\beta \langle \sigma_1^z \rangle \sum_{m=1}^{M} \left(\langle a_m \sigma_1^+ \rangle - \langle a_m^\dagger \sigma_1^- \rangle \right) - \sum_{m=1}^{M} \left\{ \gamma_m^+ \left[\langle \sigma_1^+ \sigma_2^- \rangle \langle n_m \rangle + \langle a_m^\dagger \sigma_1^- \rangle \langle a_m \sigma_1^+ \rangle \right] + \gamma_m^- \left[\langle \sigma_1^+ \sigma_2^- \rangle (\langle n_m \rangle + 1) + \langle a_m \sigma_1^+ \rangle \langle a_m^\dagger \sigma_1^- \rangle \right] \right\}$$

Two regimes: photon BEC and laser-like state

✗ Choosing bath spectral density: $J(w) = \sum_{r=1}^{R} \lambda_r^2 \delta(w - w_r) \implies J(w) = \frac{\eta}{w_c^2} w^3 \exp\left(-\frac{w}{w_c} \right)$

$$\langle D_j^\pm \rangle_\beta = \exp\left\{ 2\eta \left[1 - \frac{2\psi'(\frac{1}{\beta w_c})}{(\beta w_c)^2} \right] \right\}, \quad \langle D_j^-(t) D_j^+ \rangle_\beta = \exp\left\{ 4\eta \left[1 - \frac{1}{(1 - iw_c t)^2} + \frac{\psi'(\frac{1 - iw_c t}{\beta w_c}) + \psi'(\frac{1 + iw_c t}{\beta w_c}) - 2\psi'(\frac{1}{\beta w_c})}{(\beta w_c)^2} \right] \right\}$$

where $\psi(z) = \Gamma'(z)/\Gamma(z)$. Note that $\lim_{t\to\infty} \langle D_j^-(t) D_j^+ \rangle_\beta = \langle D_j^- \rangle_\beta \langle D_j^+ \rangle_\beta$ (compare with [5])

✗ Correlations between cavity modes of different energies $\omega_\ell = \omega_1 + (\ell - 1)\Omega$

$$c_{\ell,\ell'} = \frac{|\langle a_\ell^\dagger a_{\ell'} \rangle|}{(\langle n_\ell \rangle \langle n_{\ell'} \rangle)^{1/2}}, \quad 1 \le \ell < \ell' \le L$$

✗ Cavity level ω_ℓ has degeneracy $d_\ell = 2\ell$ and for any arbitrary function f holds

$$\sum_{m=1}^{M} f(a_m, a_m^\dagger, \ldots) = \sum_{\ell=1}^{L} d_\ell\, f(a_\ell, a_\ell^\dagger, \ldots)$$

✗ Two limiting regimes:

(a) $\eta \gtrsim 1 \implies g_\beta/g = \langle D_j^\pm \rangle_\beta \ll 1$

⇒ coherent bath contribution becomes highly suppressed

⇒ evolution is dominated by dissipative effects

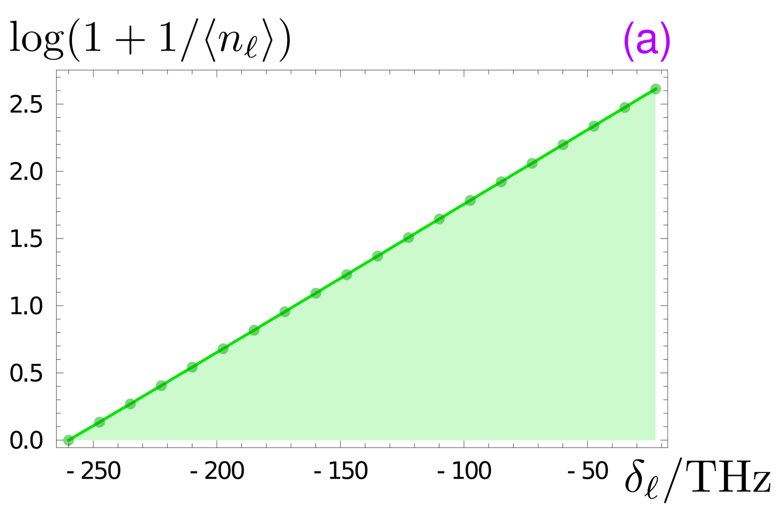
⇒ thermalization of light and emergence of *photon BEC*

(b) $\eta \ll 1 \implies g_\beta/g \approx 1$

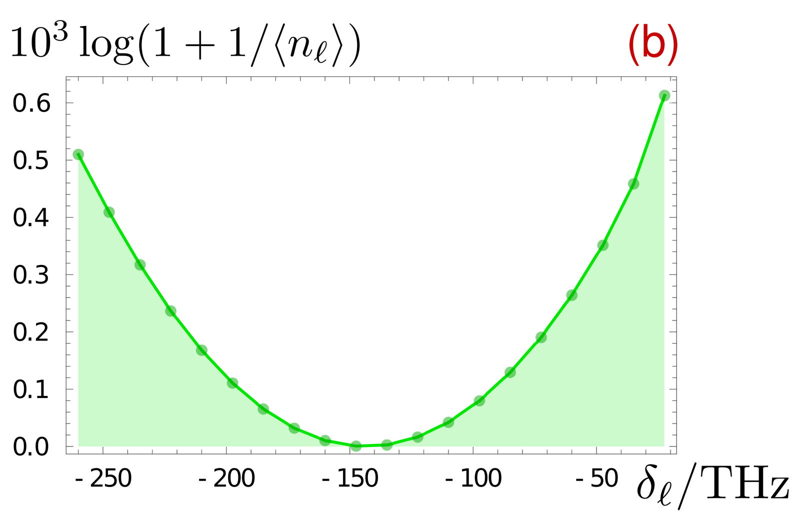
⇒ bath has pronounced coherent influence that builds-up correlations

⇒ dissipative influence is overwhelmed, but still relevant

⇒ non-equilibrium stationary state is highly coherent and *laser-like*



$\eta = 1.5, \quad c_{\ell,\ell'} < 5 \times 10^{-7}$



$\eta = 0.05, \quad c_{\ell,\ell'} > 0.993$

Stationary populations of cavity modes for $\Omega = 12.5\,$THz and $\ell = 1, \ldots, L = 20$

Realistic parameters: $N = 10^9$, $w_c = 20.5\,$THz, $T = 300\,$K, $g = 2.3\,$GHz, $\gamma_\downarrow = 0.25\,$GHz, $\gamma_\uparrow = 0.1\,$GHz, $\kappa = 3.5\,$GHz, and $\delta_1 = -260\,$THz

Conclusions

✗ In condensates of light both coherent and dissipative influence of environment have to be taken into account

✗ Interplay between the two determines system's behavior

✗ Dominance of dissipative contribution leads to emergence of photon BEC

✗ Coherent influence promotes build-up of correlations and laser-like states

✗ Dye-mediated photon-photon interaction is consequence of coherent processes

✗ Beyond mean-field approach is necessary for additional characterization of stationary states

✗ Various model extensions are possible