Quantum Optics Course Project Report

Molecular Optomechanics in Anharmonic Cavity-QED Regime Using Hybrid Metal-Dielectric Cavity Modes

IIT Kanpur Bhavya Jaiswal 210268

Introduction

Photons interacting with molecules can induce spontaneous scattering, where optical fields couple to the molecular vibrations, and the scatter at phonon-shifted frequencies. This paper was about the study of how vibrational modes and cavity emitted spectrum change with respect to various factors in the Anharmonic Cavity-QED regime, where the system had Hybrid Metal-Dielectric Cavity modes. It explores molecular optomechanics in the strong coupling regimes, which manifests in the anharmonic emission lines we observe in the sidebands of the cavity emitted spectrum.

System and its Properties

In our hybrid cavity system, there are two dominant modes that inherit properties of both dielectric and metallic parent modes. These result in two important properties:
1) Subwavelength spatial localization (or small effective mode volume V_c) and
2) High Quality factor(Q) or small cavity decay rate(κ)
For strong optomechanical coupling we need both the conditions to be true, specifically we want $g^2/\omega_m > \kappa$ and $\omega_m > \kappa$ both to be true. This condition is not feasible with typical plasmonic modes. We want $g^2/\omega_m > \kappa$ to have a strong coupling and $\omega_m > \kappa$ to be in the sideband resolved regime.
For our system, the two hybird modes are:
1) Plasmonic like low Q mode: This mode maintains a small volume, however, it has low quality factor.
2) Dielectric like high Q mode: This mode has a small κ, and at the same time maintains a pretty small mode volume which is good enough for our study. For this mode Q = 3500, $V_c = 5.36\times10^{-6}\lambda^3$. This strong confinement results in the optomechanical coupling factor "g" to be between 0.1-4 meV.
We this use the High Q dielectric like mode of the hybrid system to study the regime of strong optomechanical coupling.

System Hamiltonian, Dressed states, Eigenenergies, and Master Equation

The Hamiltonian of our system looks like:

$$\mathrm{H} = \hbar\Delta a^\dagger a + \hbar\omega_m b^\dagger b - \hbar g a^\dagger a(b^\dagger + b) + \hbar\Omega(a^\dagger + a)$$

where: $a, a^\dagger$ and $b, b^\dagger$ represent the annihilation and creation operators of cavity and vibrational modes respectively, $\Delta = \omega_c - \omega_l$ is the detuning between optical cavity and pump laser, ω_m is the frequency of molecular vibrational mode, and Ω is the Rabi frequency of the optical cavity mode. $\mathrm{g} = (\hbar R_m/2\omega_m)^{-1/2}\omega_c/\epsilon_0 V_c$, is the optomechanical coupling factor, where R_m is the Raman activity associated with the vibration.
The dressed state of the system is of the form:

$$|\psi\rangle_{n,k} = D^\dagger\left(\frac{gn}{\omega_m}\right)|n, k\rangle$$

and the corresponding dressed state energies are:

$$\varepsilon_{n,k} = n\hbar\Delta + k\hbar\omega_m - n^2\frac{\hbar g^2}{\omega_m}$$

with D being the displacement operator. Here, n represents the number of photons and k is the phonon number state.
In our system the ensuing master equation looks like:

$$\frac{d\rho(t)}{dt} = -\frac{i}{\hbar}[H, \rho(t)] + \frac{\kappa}{2}D[a]\rho(t)$$

$$+\frac{\gamma_m(n_{th}+1)}{2}D[b]\rho(t) + \frac{\gamma_m n_{th}}{2}D[b^\dagger]\rho(t)$$

where κ is the cavity decay rate, γ_m is the vibrational decay rate, $D[]$ is the Lindblad Superoperator, and n_{th} = $1/(\exp(\hbar\omega_m/k_bT)$ - 1) is the thermal population of the vibrational mode at temperature T.
Since the phonon states are displaced by b $\longrightarrow$ b - $d_0 a^\dagger a/\omega_m$, the more correct form of the master equation for our system looks like:

$$\frac{d\rho(t)}{dt} = -\frac{i}{\hbar}[H, \rho(t)] + \frac{\kappa}{2}D[a]\rho(t)$$

$$+\ \frac{\gamma_m(n_{th}+1)}{2}D[b - d_0 a^\dagger a]\rho(t) + \frac{\gamma_m n_{th}}{2}D[b\dagger - d_0 a^\dagger a]\rho(t)$$

$$+\ \frac{2\gamma_m k_b T d_0^2}{\hbar\omega_m}D[a^\dagger a]\rho(t)$$

Using the quantum regression theorem along with the master equation given above, I have numerically reproduced the cavity-emitted spectrum of the hybrid device by taking a Fourier transform of the first-order quantum correlation functions:

$$S(\omega) =$$

$$\mathrm{Re}\Big[\int_0^\infty dt e^{(\omega_l-\omega)t} \times [< a^\dagger(t)a(0) >_{ss} - < a^\dagger >_{ss} < a >_{ss}]\Big]$$

where all the expectation values are taken at the steady state of the system and the coherent contribution is removed because it overpowers the incoherent part which is of our interest.

Parameters

The properties of the two hybrid modes are:

Mode	$\hbar\omega_m$	$\hbar g$	Q	V_c
High Q	1.61 eV	0.1 meV	3500	$5.36 \times 10^{-6}\lambda_c^3$
Low Q	1.83 eV	20 meV	17	$4.54 \times 10^{-8}\lambda_c^3$

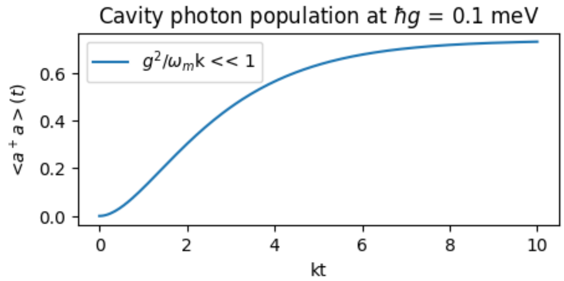
We are working with the high Q mode in the resonant case, where laser pump frequency $\omega_l = \omega_c$, frequency of optical cavity, i.e. $\Delta = 0$. Additionally, we have assumed an intrinsic quality factor, $Q_m = \omega_m/\gamma_m = 100$. The various parameters used in the calculations are listed below:

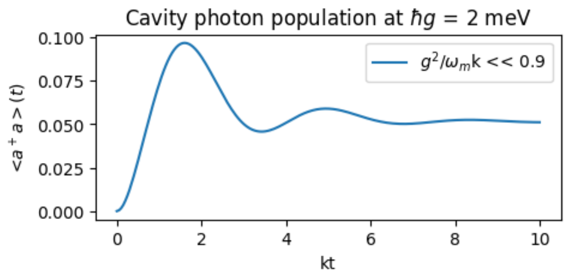
$\hbar\omega_c$	1.61 eV	$\hbar\kappa$	0.46 meV
$\hbar\omega_l$	1.61 eV	$\hbar g$	0.1, 2, 4 meV
$\hbar\omega_m$	10 meV	γ_m	0.1 meV
$\hbar\Omega$	0.1 meV	T	4K, 50K

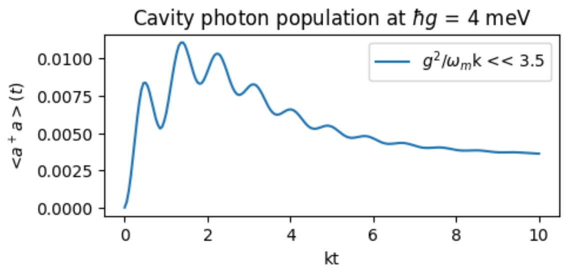
For our calculations, I have assumed that at t = 0, the cavity is in the ground state (n = 0) and the vibrational mode is in a thermal state corresponding to the temperature.

Calculations and Plots

Given below are the plots I obtained numerically using QuTip. These plots differ from the plots given in the paper, due to some issues which I was not able to identify and resolve.

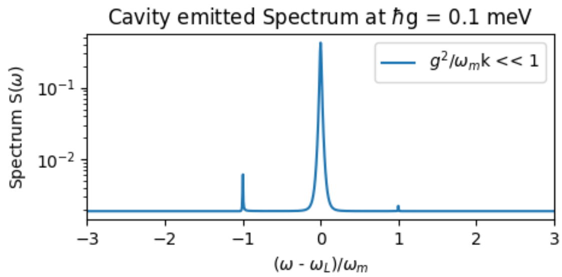


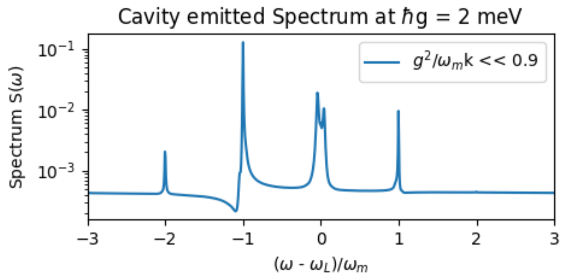


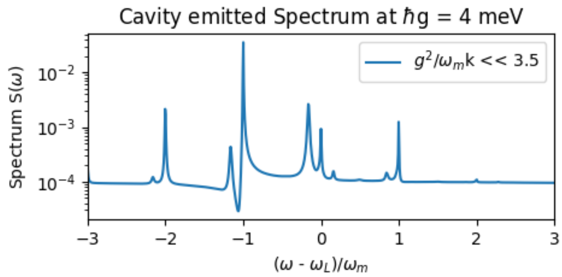


The graph given above are the plots of cavity photon population as a function of time at different values of $\hbar g = 0.1$ meV, 2 meV and 4 meV (top to bottom). The temperature is kept constant at T = 4K. By increasing g, we observe that the corresponding cavity populations become nontrivial with time as we get into the strong coupling regime. The standard master equation has been used to get these plots.

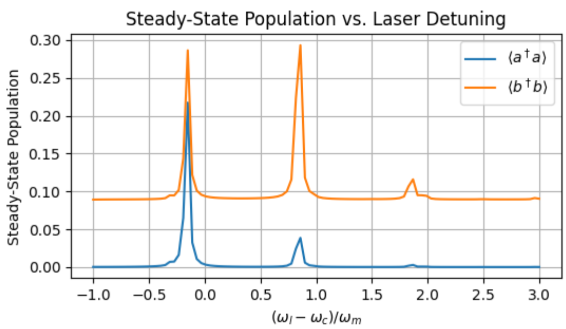
The plots below are the cavity emitted spectra at different values of $\hbar g$ = 0.1 meV, 2 meV and 4 meV. Here, T = 4K, $\hbar\omega_m$ = 10 meV. Again, to get these plots the standard master equation has been used. We observe that as the g-factor increases, we observe nontrivial shifts in the cavity resonance, and as we enter into the strong coupling regime (sideband resolving regime) me observe the emergence of Raman sidepeaks. These additional peaks and shifts arise due to the anharmonicity induced in our system due to strong optomechanical coupling.



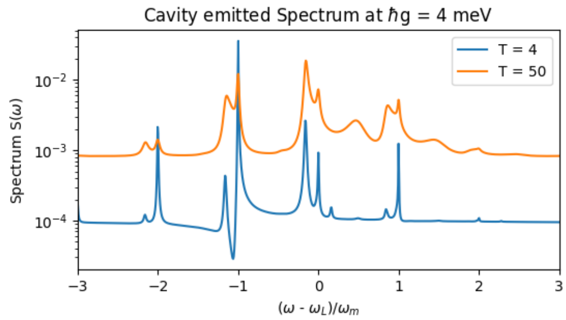




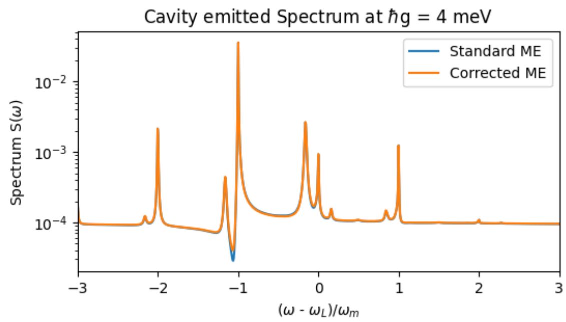
The plot given below, is the plot of steady-state population of photons $< a^\dagger a >$ and phonons $< b^\dagger b >$as a function of laser detuning. In all other cases, we have taken the detuning to be zero, but here we are varying the detuning.



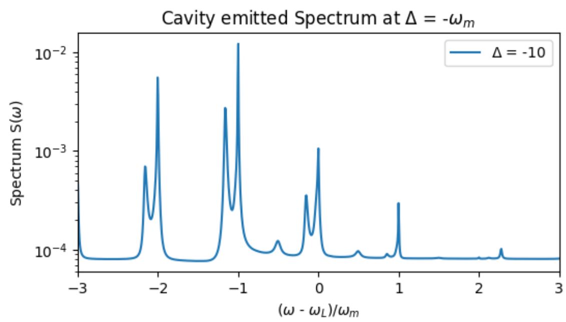
In the plot below, we have explored the effect of temperature on the emission spectrum. This result that I obtained is not similar to the one given in paper. What I do observe is that with increasing temperature, the peaks of the spectrum broaden. There is also a displacement of the spectrum along y-axis, which could be because of some error that I would've made while numerically calculating the spectrum.

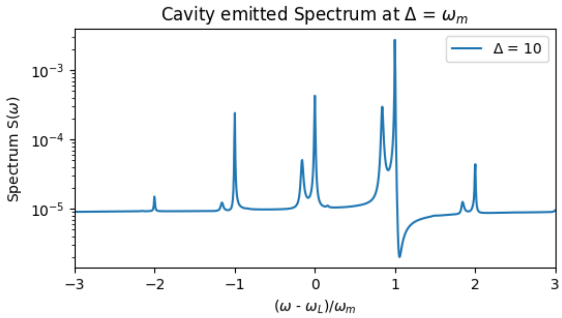


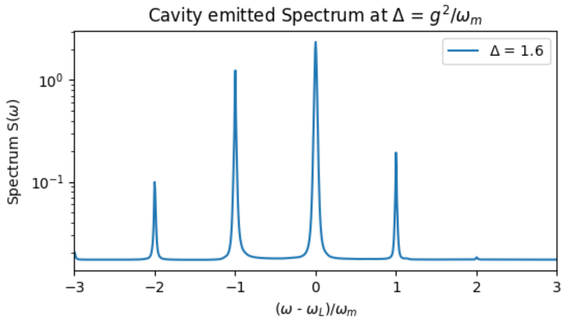
In the next plot, I have plotted the cavity emission spectrum at temperature T = 4 K and g = 4 meV by using the two different master equations, i.e. the standard master equation and the corrected for of the master equation. We see not a big difference due to the effect of modified dissipation terms of the generalised ME.



In the plots below, I have explored how the cavity emitted spectrum varies over time for different detunings of the system. The g is constant as $\hbar g$ = 4 meV, and temparature T = 4K. In the first graph, the anharmonic peaks are visible, with a peak at $\omega = \omega_l + \omega_m$.







Conclusion

The paper was more or less about the study of how the emission lines and Raman peaks of a strongly coupled optomechanical system depend of various parameters. We achieved this strong coupling by using MNP's in the system, which helped in creating a hybrid metal-dielectric system which had modes that provided both a small mode volume and small decay rates. We used the High Q mode in the study, and observed the changes in emitted spectrum by varying different parameters. In practical we can vary the g-factor by varying the size and shape of the MNP's, or by adjusting the gap size between them. These change the effective volume, which in-turn changes the coupling factor.
As we increase the coupling in our system, we see that the energies get shifted by some amount, these result in the anharmonic side-bands that we see in the plots.

Sources of errors

My plots are not exactly the same as the plots given in the paper. This I believe could be because of some errors which unfortunately I was not able to identify and resolve. However, I believe that some reasons for the the difference in my plots and those of the paper could be:
1) In the paper, all the calculations are done in eV unit, but I have done all the calculations in meV units. This should not be causing a problem, but I observed that changing the units did make a difference in my plots, and that is why I had to use the units of meV in my numerical calculations.
2) Cut-off used in calculations: The cut-off I have used in my code, is 5. By increasing the cut-off, my plots got refined and better, But unfortunately, by using high cut-off like 10, my laptop used to freeze, because the computation was too heavy I believe. So, I had to use a smaller cut-off in my code.
I believe that these were the reasons why I did not get the same plots as in the paper.

References

The paper I studied is : https://arxiv.org/pdf/1805.10153.pdf
Reference paper for QuTip :QuTip
The reference books I used are:
1) Quantum Optics by O. Scully and M. Zubairy
2) Quantum Optics by G.S. Agarwal (to study about system Hamiltonian, dressed states and dressed energies)