NEW TYPE OF CAVITY-QUANTUM ELECTRODYNAMICS TRANSITION IN MULTIMODE CAVITIES UNDER STRONG COUPLING

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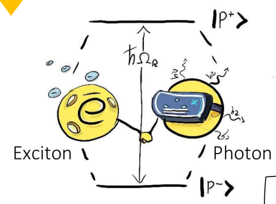
Polaritons



Collective coupling:

$$\Omega_R \sim d\sqrt{\hbar\omega\frac{N_{mol}}{V_{cavity}}} \sim \sqrt{n_{mol}}$$

Molecular transition dipole | Mode volume | Number of molecules | Concentration

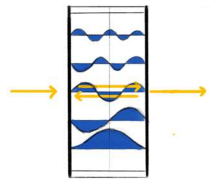
- Strong Coupling : Formation of hybrid (collective) quantum states.
- Determined by the "concentration" of the molecule.
- Light matter interaction is counteracted by dissipation (γ).

Coupling regimes

Cavity QED can be divided into three main regimes

Weak coupling	Strong coupling	Ultrastrong coupling
$\Omega_R < \gamma$	$\Omega_R > \gamma$	$\Omega_R \sim E_x/10$

Multimode cavity



$L \gg \lambda$ - several longitudinal modes close to E_x

E_x - exciton energy

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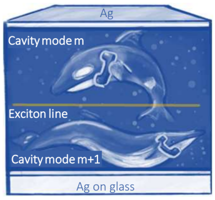
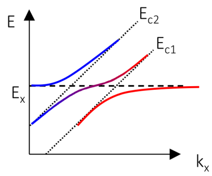
How will several optical resonances interact with the material?



$$H_{N+1} = \begin{pmatrix} E_x & g & g \\ g & E_{c1} & 0 \\ g & 0 & E_{c2} \end{pmatrix}$$

$$H_{2N} = \begin{pmatrix} E_x & g & & 0 \\ g & E_{c1} & & \\ & & E_x & g \\ 0 & & g & E_{c2} \end{pmatrix}$$



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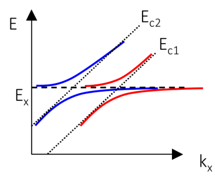

Mid-Polariton branch : material-mediated modal coupling

Band Gap: two pairs of decoupled polaritonic branches

Different Hamiltonians = different physics

WHICH ONE IS CORRECT?

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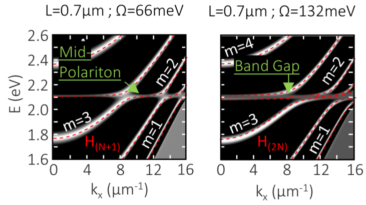
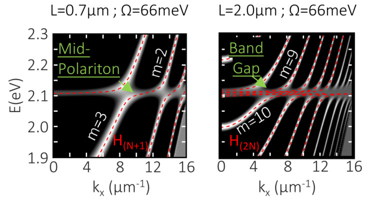
Theoretical results

different coupling strengths



different cavity sizes



- Transition: competition between molecular lifetime and cavity round trip time

$$\tau_c > 2\tau_x$$

exciton lifetime : $\tau_x = \frac{h}{\pi\gamma}$

cavity round trip time : $\tau_c = \frac{L}{|v_g|}$

- Critical thickness for transition

$$L_0 \simeq \frac{hc(n_0\gamma)}{\pi f_c}$$

the oscillator strength : $f = \frac{2NE_xd^2}{n_0^2} = (\hbar\Omega_R n_0)^2.$

Balasubrahmaniyam et al., PRB 103, L241407 (2021)

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Experimental results

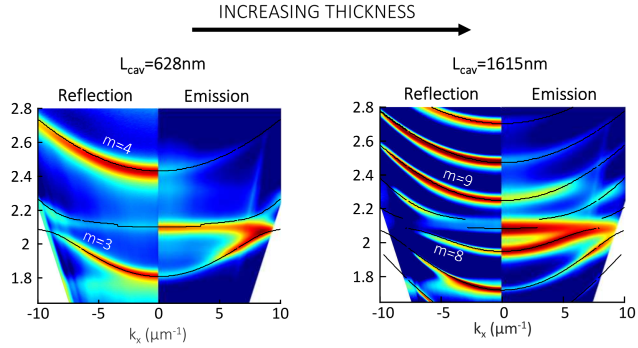
Identical coupling strength, different behavior for different cavity sizes

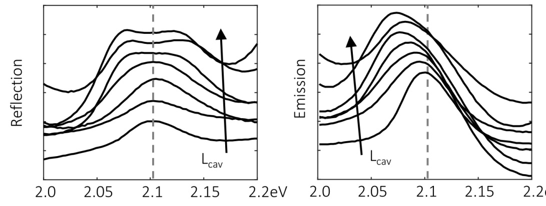
Reflection measurements

- The transition has been experimentally proven.
- Single peak: cavity modes talking through molecules.
- Two peaks: polaritons decoupled.

Emission measurements

- Emission from high energy polaritons, breaking Kasha's rule?





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Conclusions

Weak coupling	Strong coupling	Ultrastrong coupling
$\Omega_R < \gamma$	$\Omega_R > \gamma$	$\Omega_R \sim E_x/10$

Mid-Polariton	Band gap
$\tau_c < 2\tau_x$	$\tau_c > 2\tau_x$

- Strong coupling can occur in two very different manners, depending on coupling strength + cavity size .
- The dimensions of the system do matter.
- In progress: ultrafast spectroscopy for studying the dynamic of the system.