Plasmon Exciton-Polariton Lasing

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Acknowledgement

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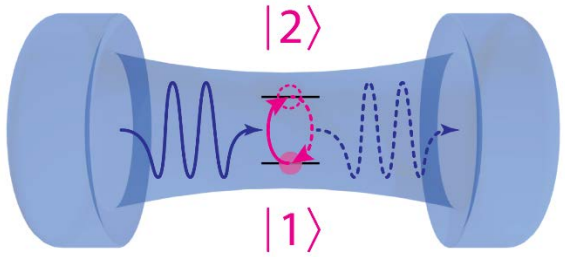
Exciton-Polaritons (Strong light-matter interaction)



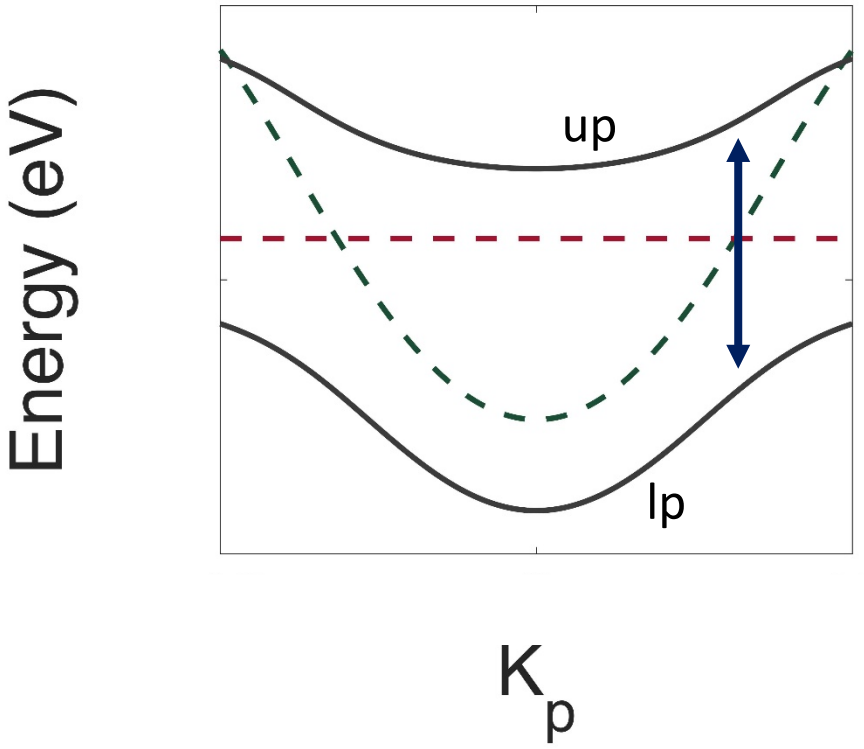
$$H = \begin{pmatrix} E_{cavity} - i\gamma_1 & \Omega \\ \Omega & E_{exciton} - i\gamma_2 \end{pmatrix}$$

$$Strong\ coupling\colon\ \Omega > \gamma_1 + \gamma_2$$

Energy exchange rate exceeds the decoherence

$$\Omega \propto \sqrt{N}$$

(N: Number of the 2-level systems)

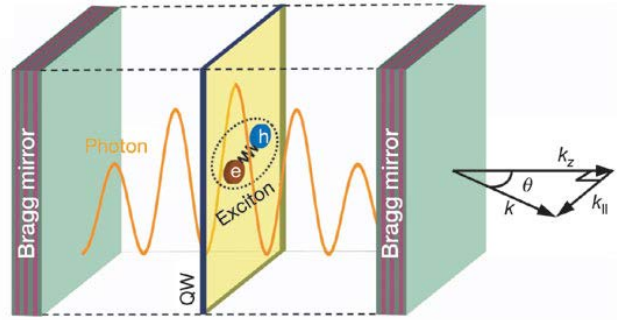


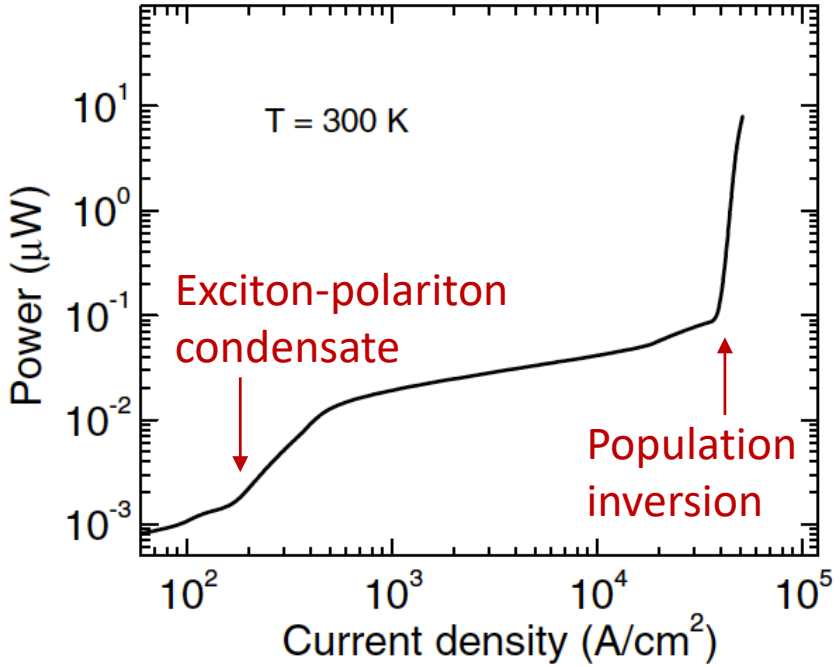
$$|Polariton\rangle = a|Cavity\ photons\rangle + b|Exciton\rangle$$

H. Deng, et al., Rev. Mod. Phys. (2010)

Exciton-Polariton condensation

- Exciton-polaritons are bosons
- Condensation at room temperature ($T_{critical} \propto 1/ m_{eff}$)
- Coherent radiation is the result of the exciton-polariton decay
- Exciton-polariton condensation occurs at lower thresholds than photon (conventional) lasing

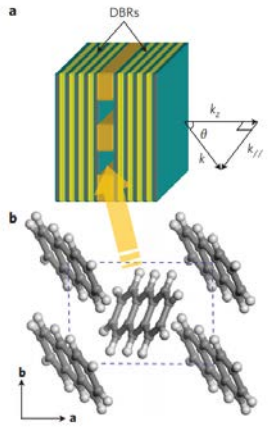




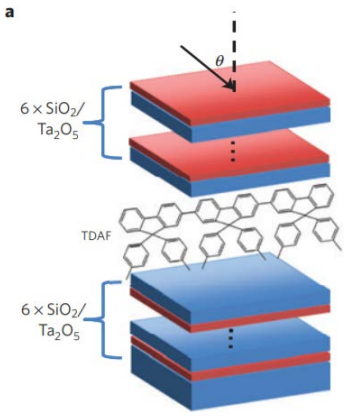
*J. Kasprzak et al., Nature **443**, 409-414 (2006)*
*Bhattacharya et al., PRL **112**, (2014)*

Organic-based exciton-polariton condensation

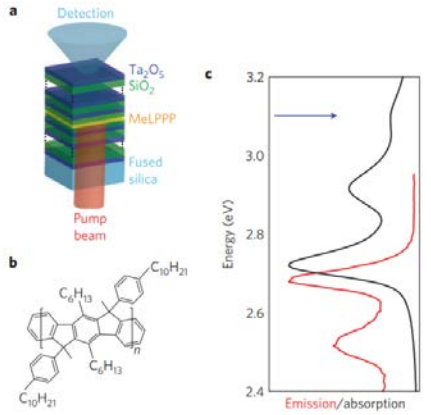
Experiment



S. Kena-Cohen, et. al,
Nature Photonics, 2010



C. Daskalakis, et. al,
Nature Materials, 2013



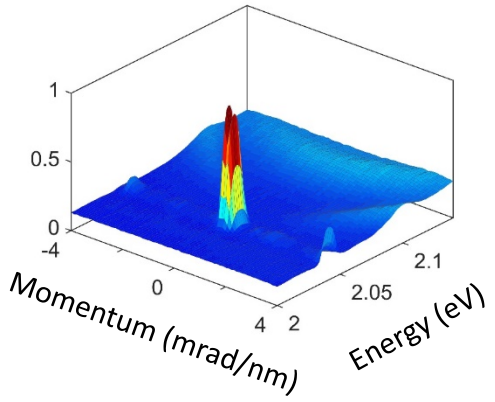
J. Plumhof, et. al,
Nature Materials, 2013

Motivations

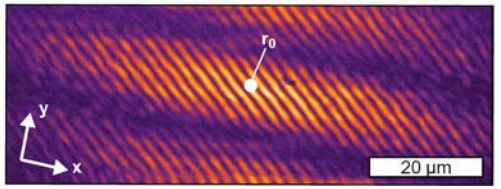
- **Application perspective:** Room Temperature, Compatibility with organic electronics, covering entire visible spectrum, …
- **Fundamental perspective:** Condensation mechanism: Role of molecular vibrations, Interactions, ….

Content

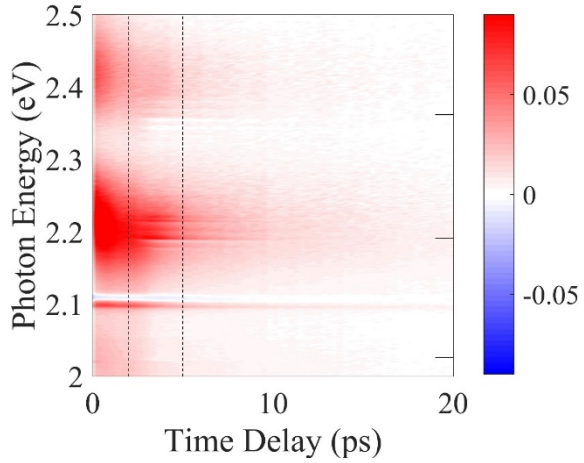
1. Polariton lasing from a plasmonic based grating



2. Long-range Spatial coherence properties

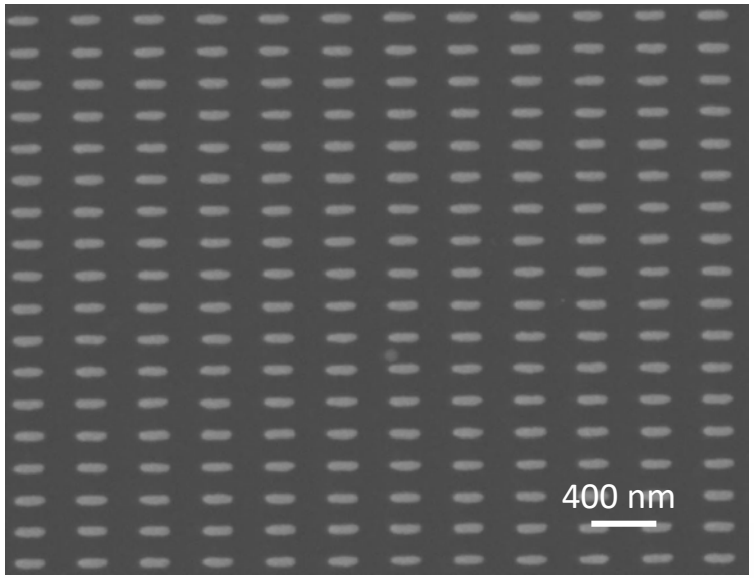


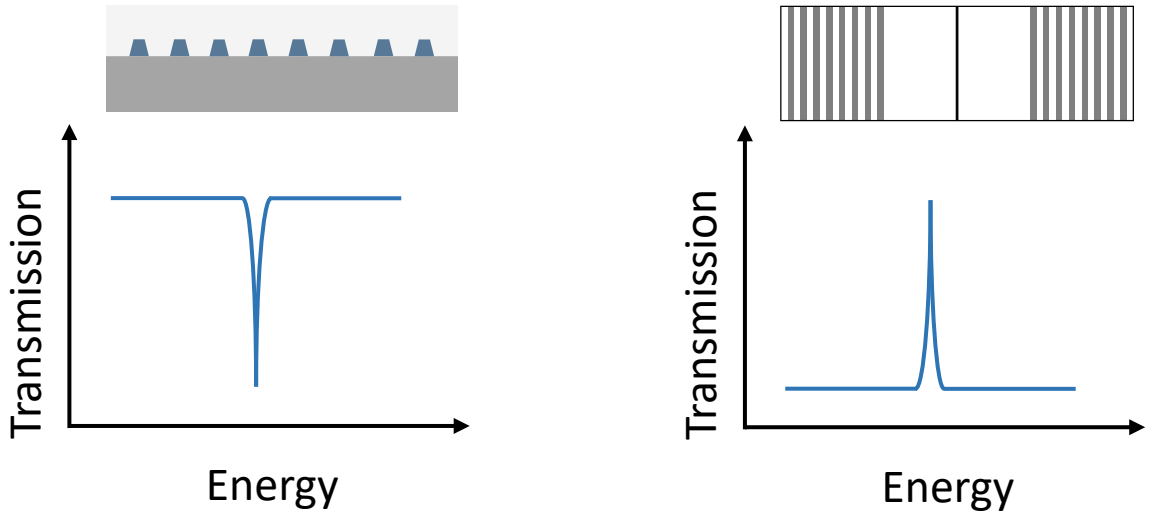
3. Ultrafast spectroscopy of the exciton-polariton condensate

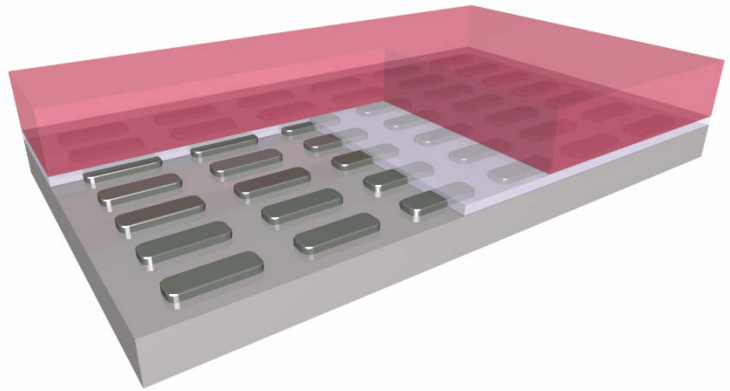


Plamonic cavities

- Simple and easy to fabricate over large scale
- Open cavity , i.e., no restriction on pump and probe





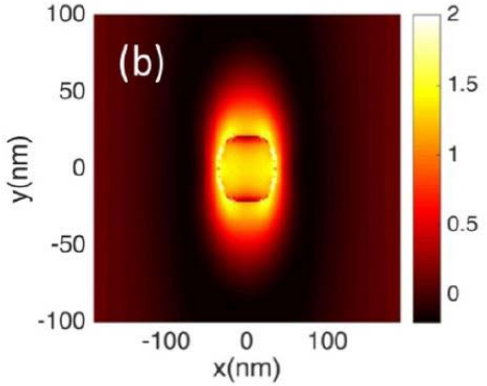


Plasmonic cavities

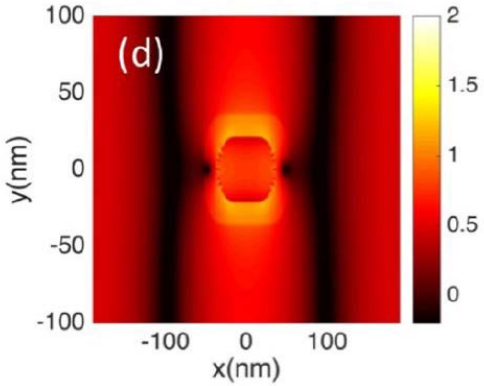
Plasmonic

Strong field enhancement at close proximity of the particles

Single particle



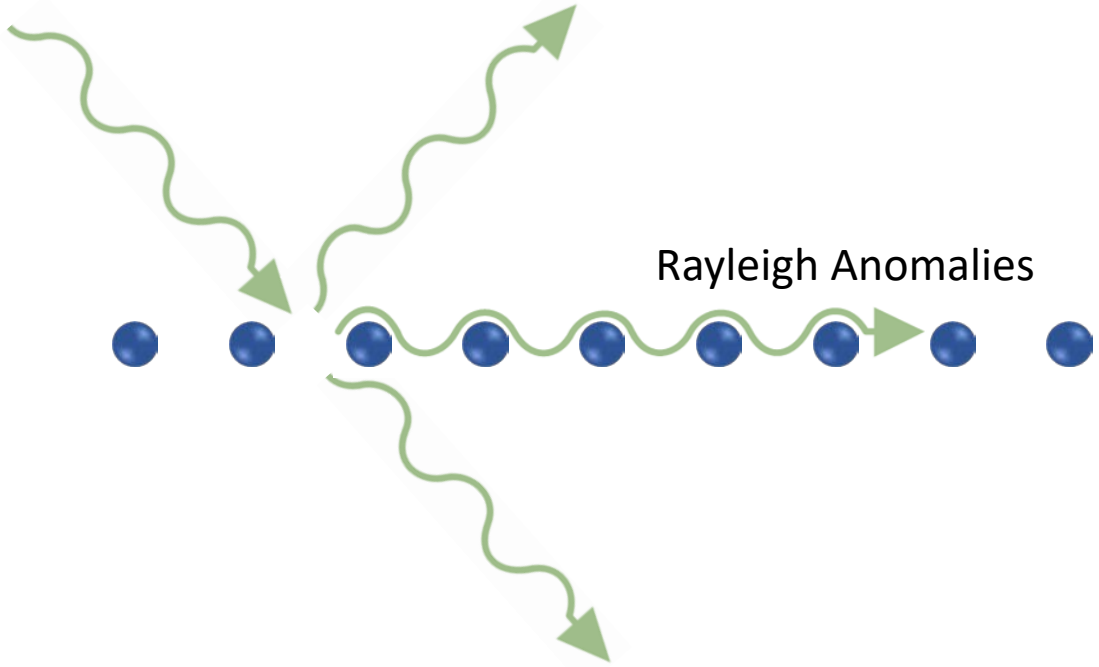
Lattice



Surface Lattice Resonance

Lattice

Delocalized electromagnetic field



M. Ramezani, et. al, ACS Photonics, 2018

Plasmonic cavities

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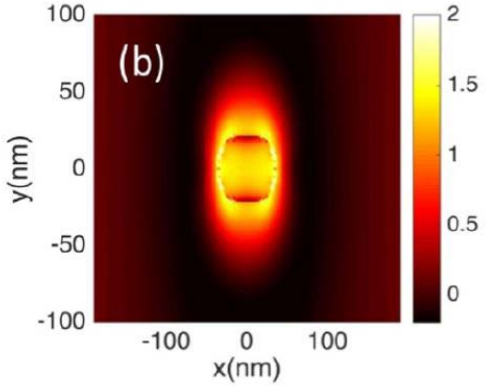
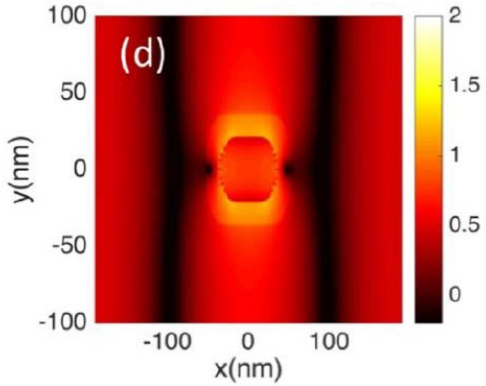


Lattice



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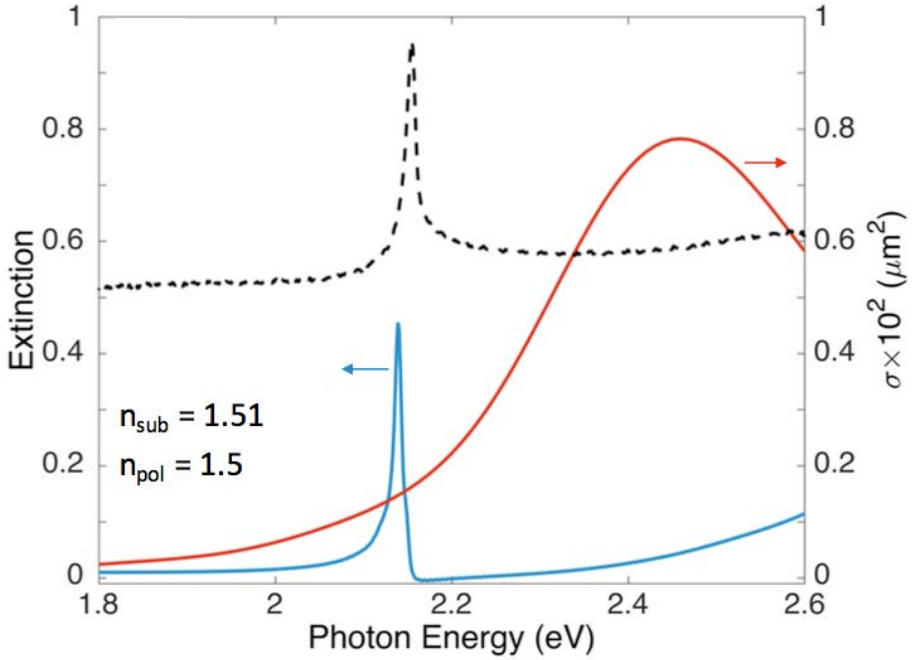
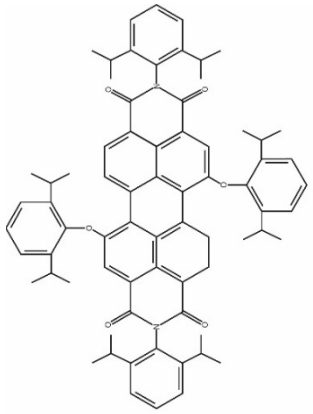
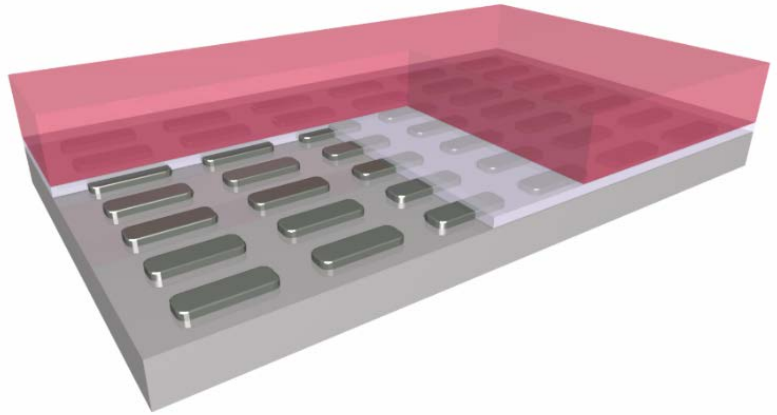


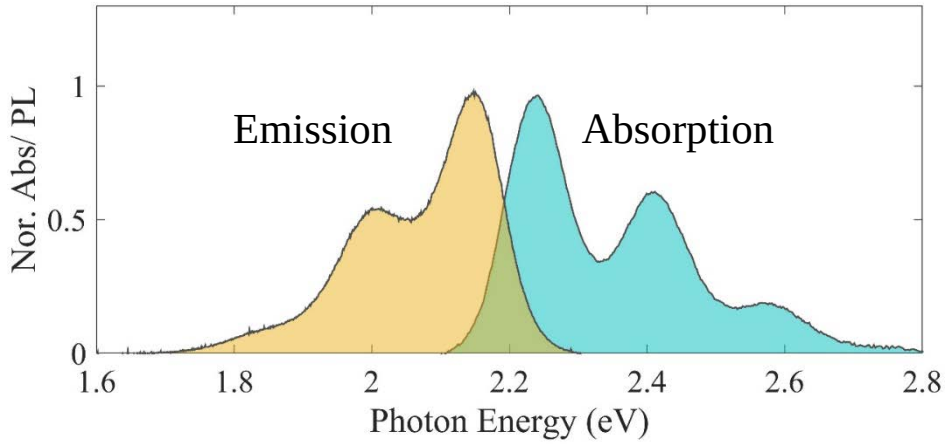
M. Ramezani, et. al, ACS Photonics, 2018

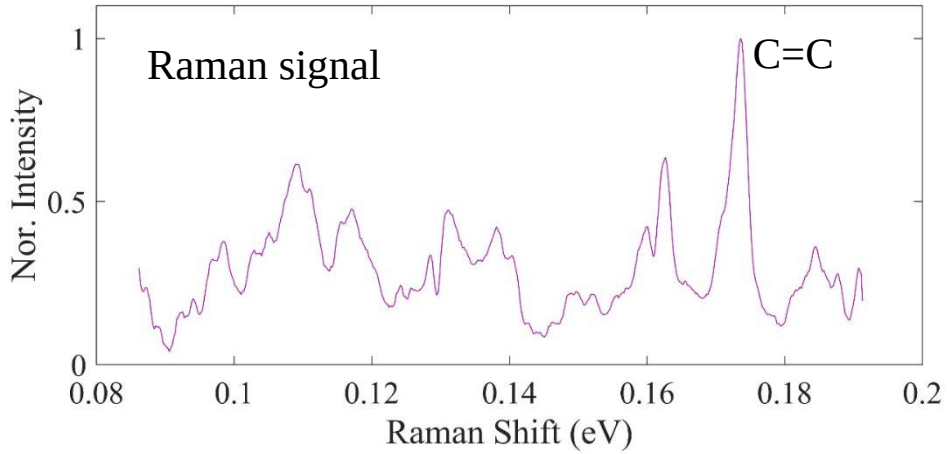
Organic molecules

- Thin layer of polymer doped with organic molecules (Frenkel excitons)
- Multiple vibrational modes in the Raman spectrum





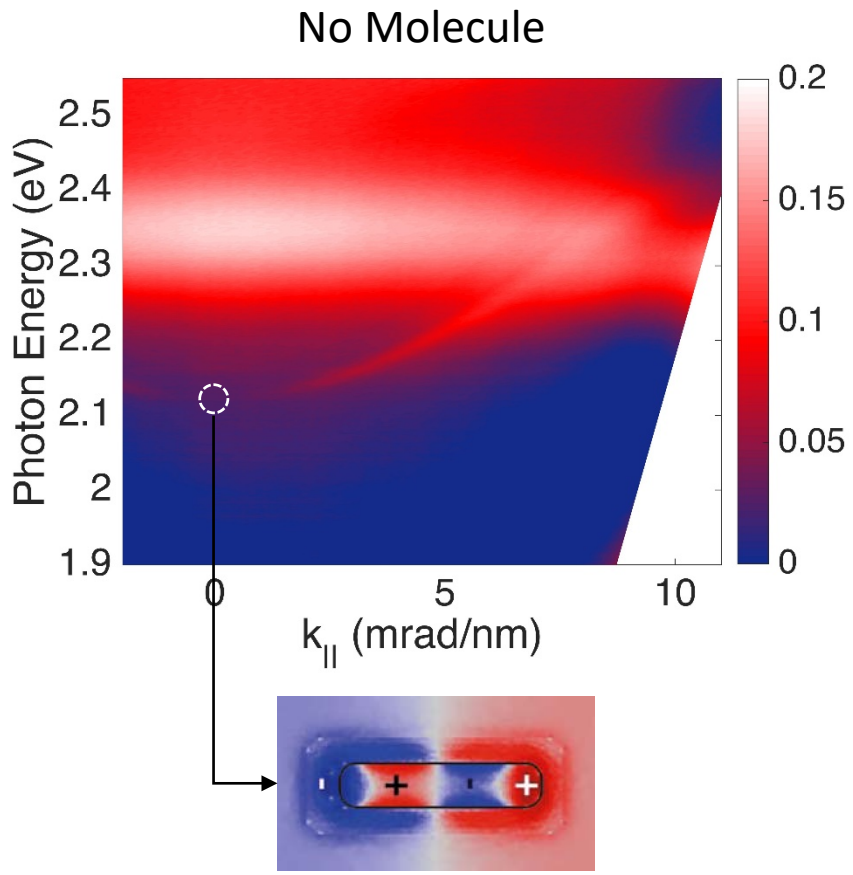




M. Ramezani, et. al, Optica, 2017
M. Ramezani, et. al, Phy. Rev. Let., 2018

Plasmon-Exciton-Polaritons

Dark Mode: Due to the symmetry of the mode the radiative decay is highly suppressed.



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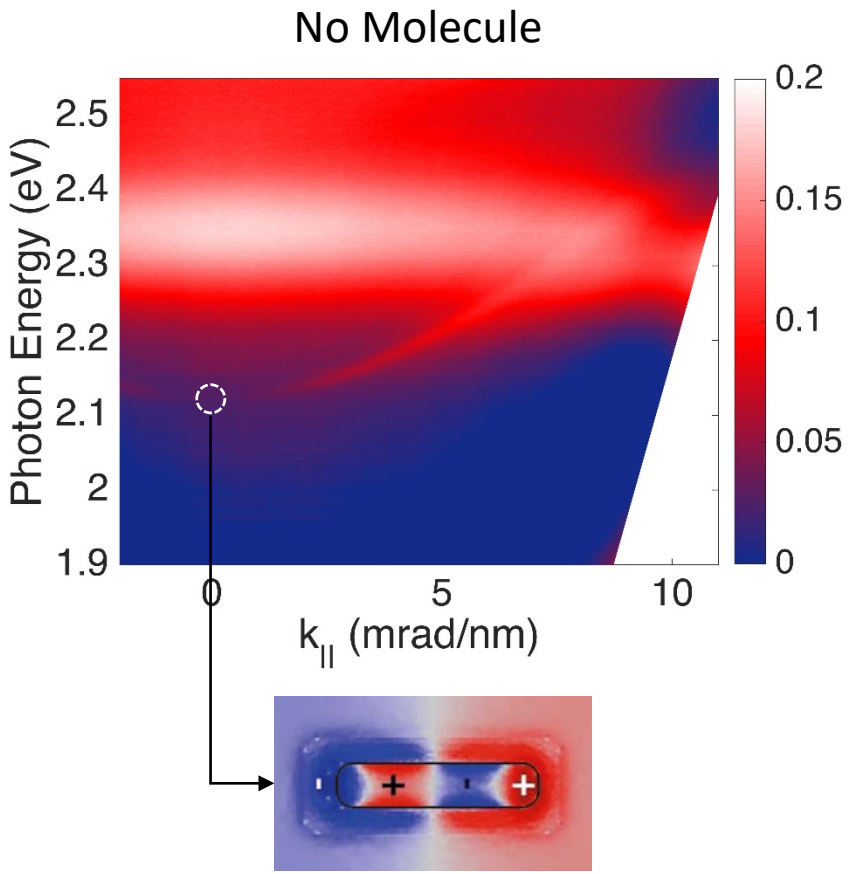
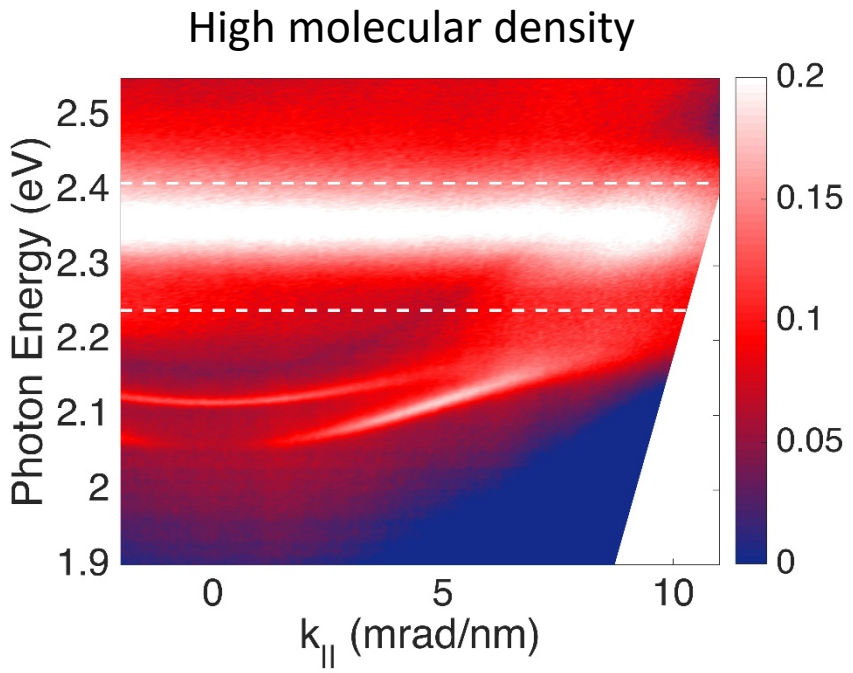
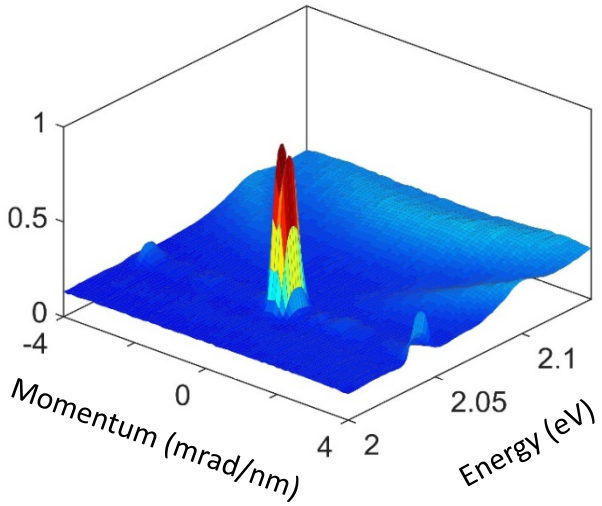


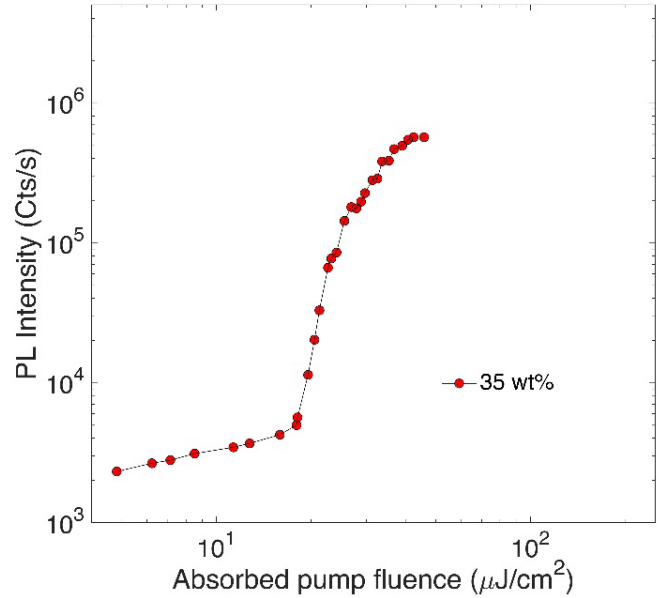


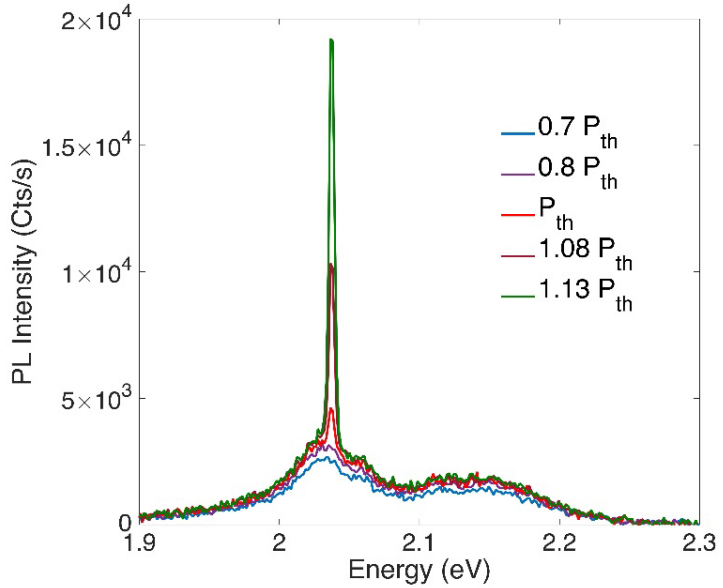
M. Ramezani, et. al, Optica, 2017
M. Ramezani, et. al, Phy. Rev. Let., 2018

Plasmon-Exciton-Polaritons Lasing

- Nonlinear increase of the emission intensity above threshold
- Sharp emission peak with enhanced temporal coherence
- Enhanced directionality toward forward direction





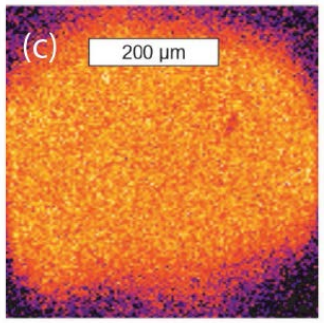


M. Ramezani, et. al, Optica, 2017
M. Ramezani, et. al, Phy. Rev. Let., 2018

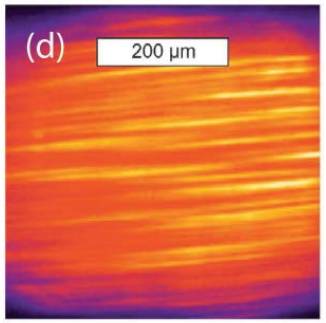
Long-range spatial coherence

- Diffuse emission below threshold
- Elongated patterns above threshold

Below Threshold

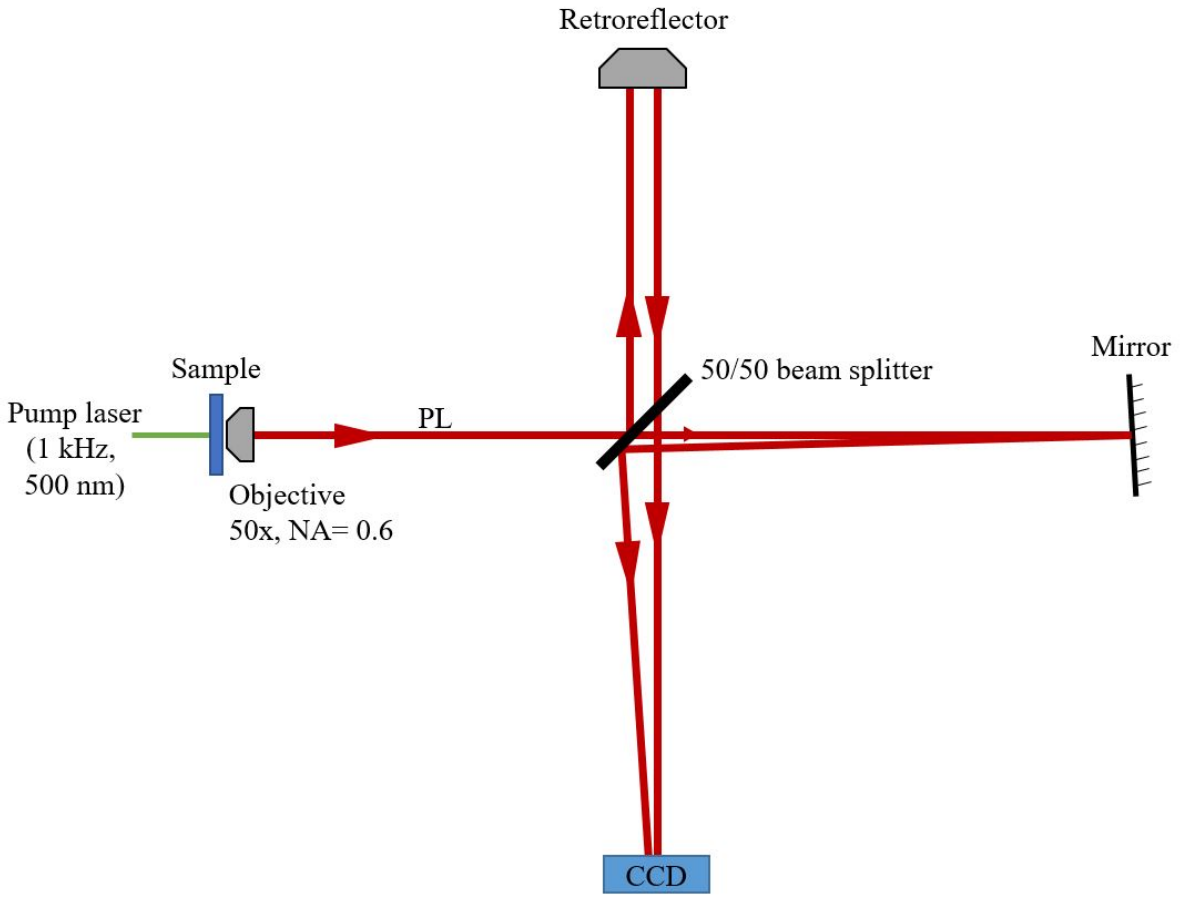


Above Threshold



De Giorgi, et. al, ACS Photonics, 2018

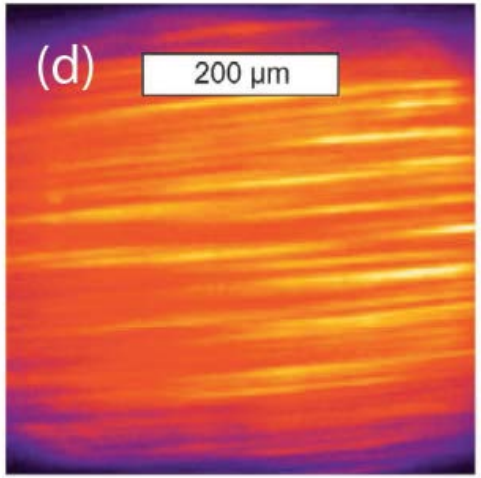
Long-range spatial coherence

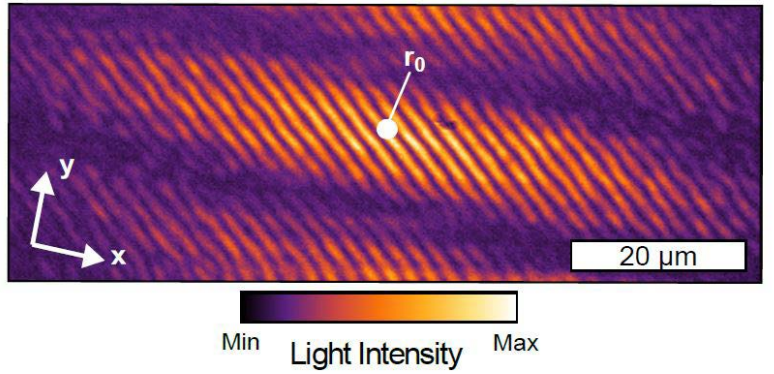


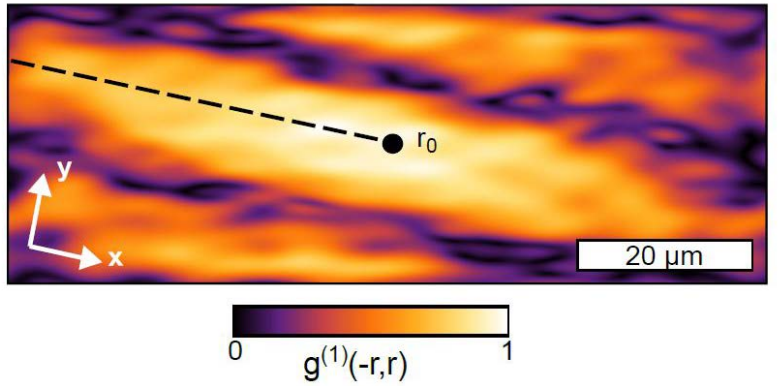
Long-range spatial coherence

- Coherence over 100 µm
  (Pump diameter = 50 µm)

Above Threshold



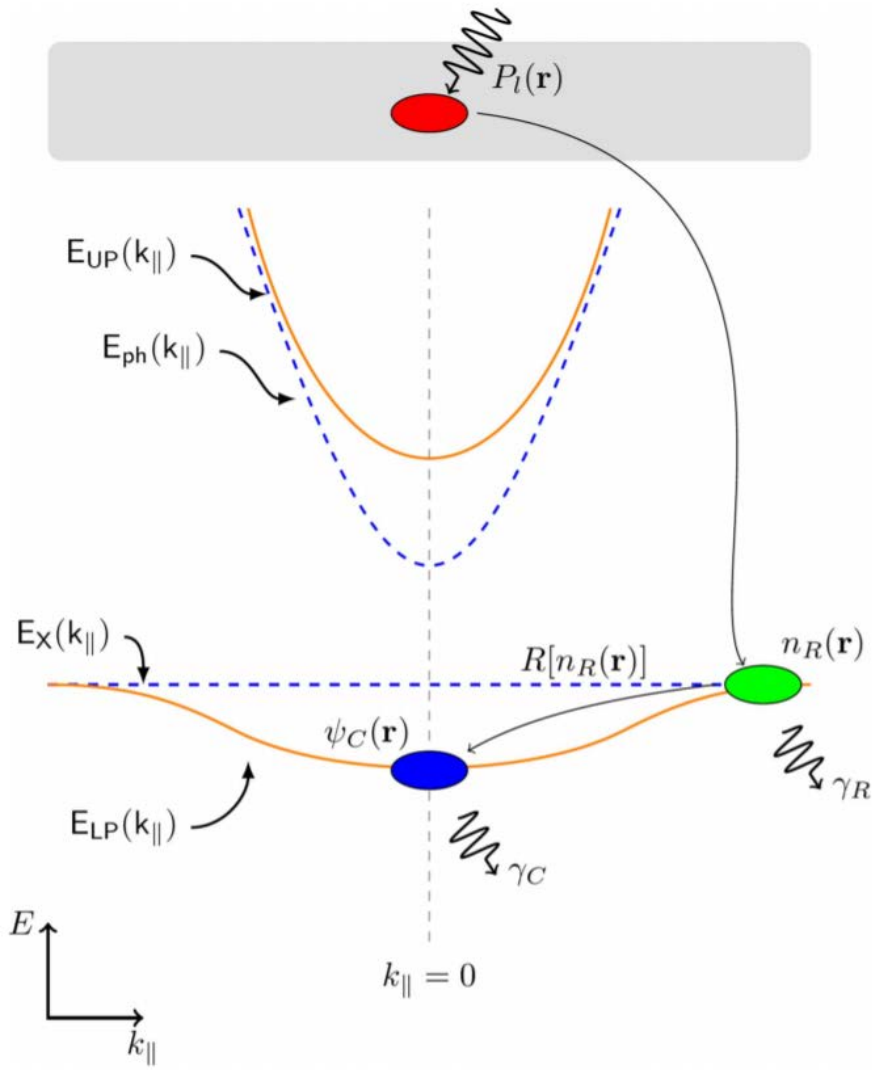




De Giorgi, et. al, ACS Photonics, 2018

Condensation Dynamics

- How does the condensation process occur?
- What are the involved time-scales ?

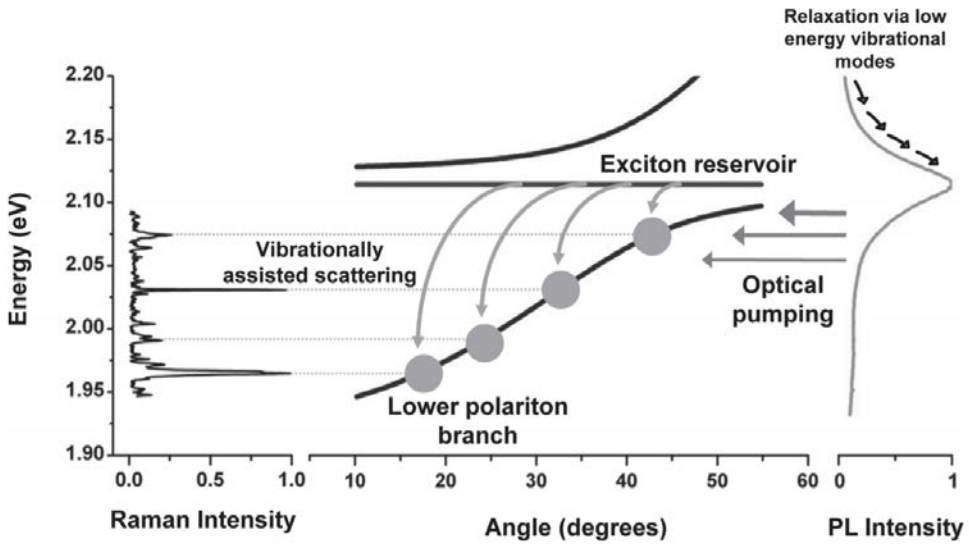


Semicond. Sci. Technol. 32 (2017)

Dynamics of organic-based exciton-polariton

Organic systems:

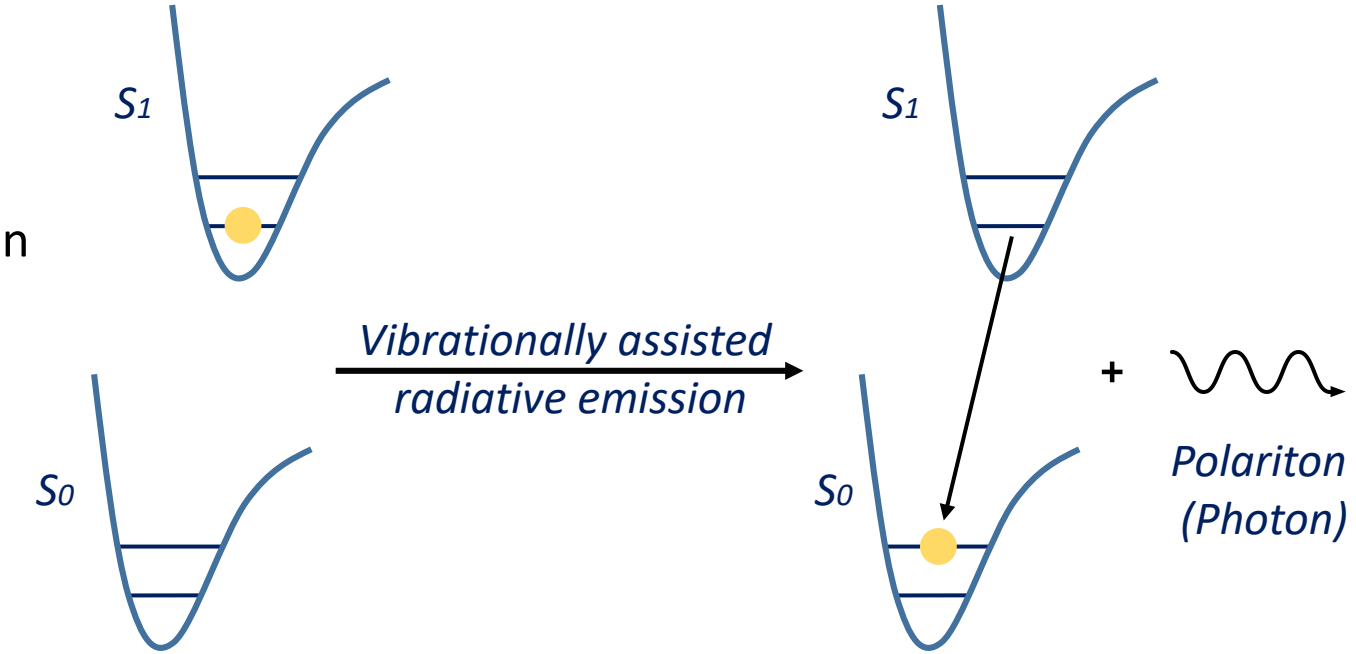
- Molecular vibrations are efficient relaxation channel
- Correlation between enhanced PL from organic exciton-polaritons and discrete vibrational modes



Nomaschi et al., APL **99**, 2011, Cole et. Al., Adv. Funct. Mater. **2011**, 21, 3691–3696

Organic-based polariton lasing

- Room-temperature: Vibrations are relevant for scattering
- Radiative recombination of a molecular exciton assisted by emission of a vibrational quanta in the ground state

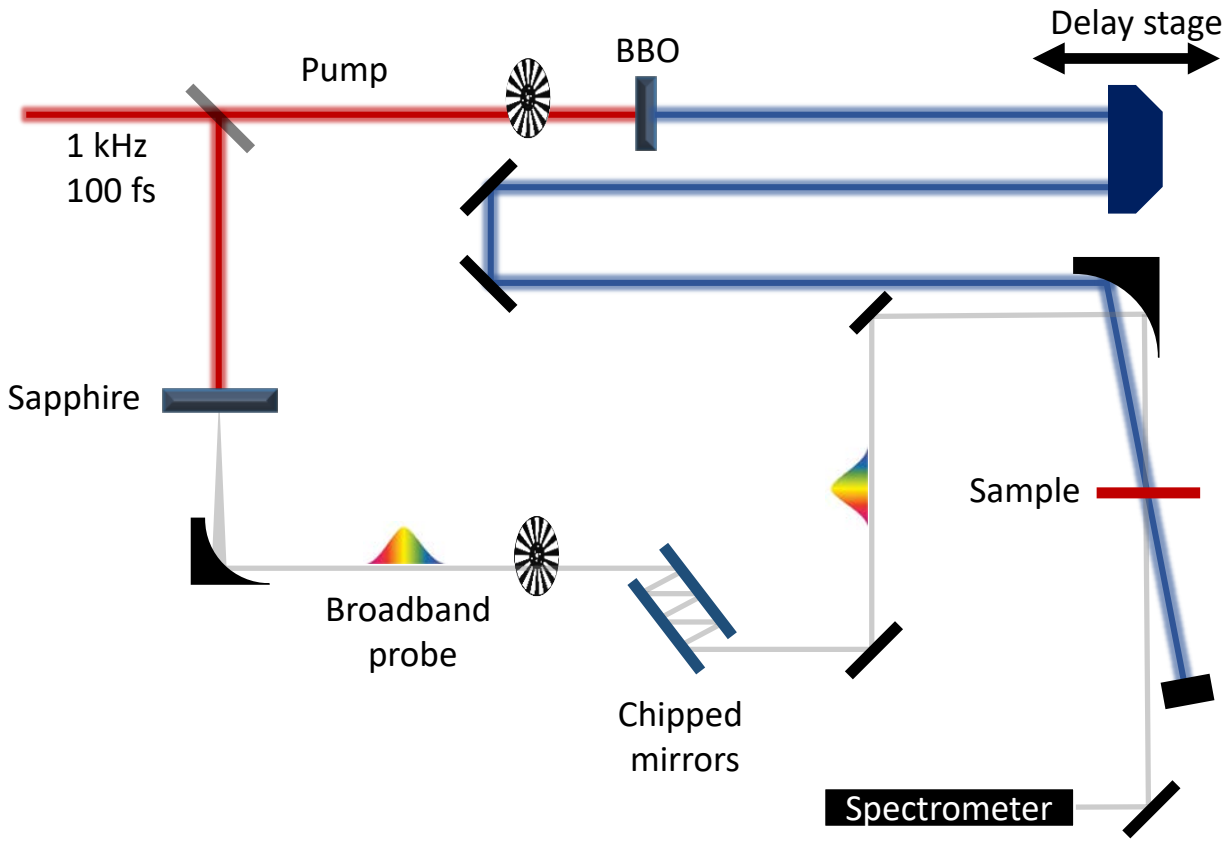


Question:
Can we probe (in real time) these specific vibrations associated with the condensate (in the ground state signal)?

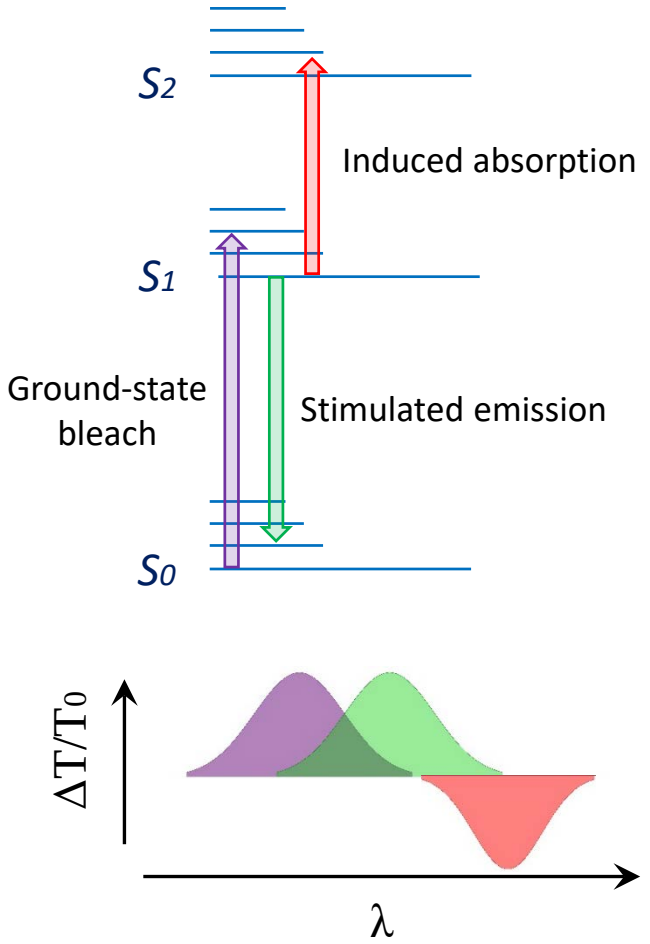
*PRB **80**, 235314 (2009) & PRB **88**, 075321 (2013), PRL **121** (19), 193601 (2018)*

Ultrafast Nonlinear Spectroscopy

Pump-Probe Setup



Transient Absorption Signal



Conclusions

- Organic Plasmon-Exciton-Polariton condensate is demonstrated using open structures

- Role of molecular vibrations in condensation investigated with transient absorption

Thank you