Technische Universiteit
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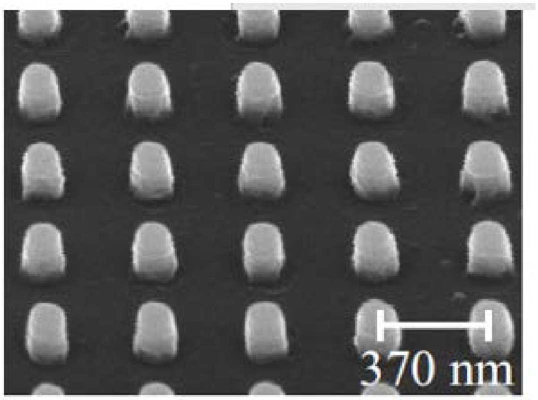
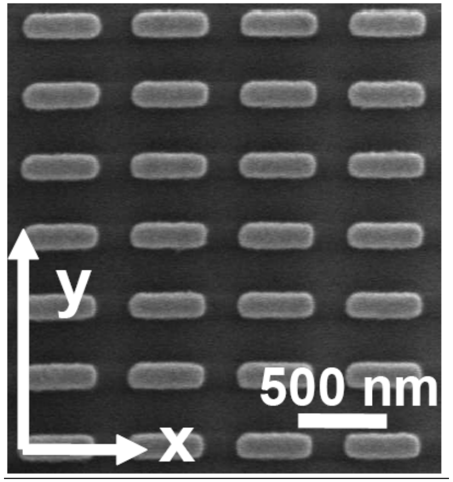
University of Technology

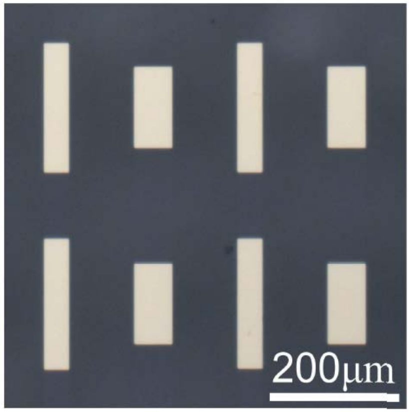
Strong Light-Matter Coupling and Polariton Lasing in Metallic and Dielectric Metasurfaces

Jaime Gómez Rivas

www.surfacephotonics.org

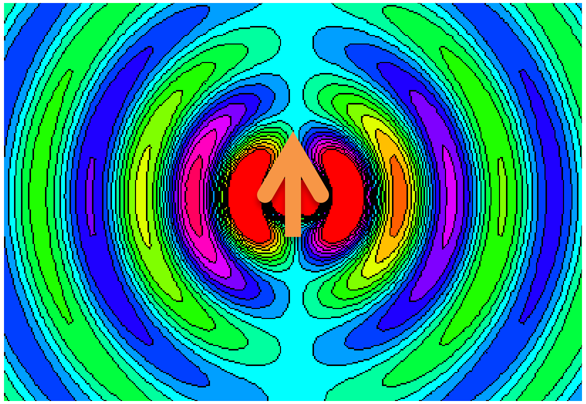






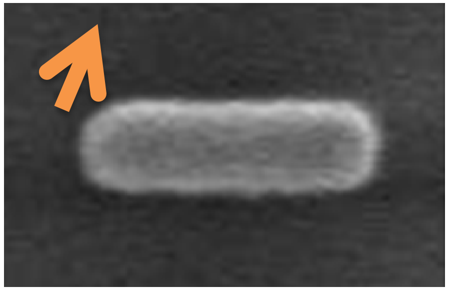
Motivation

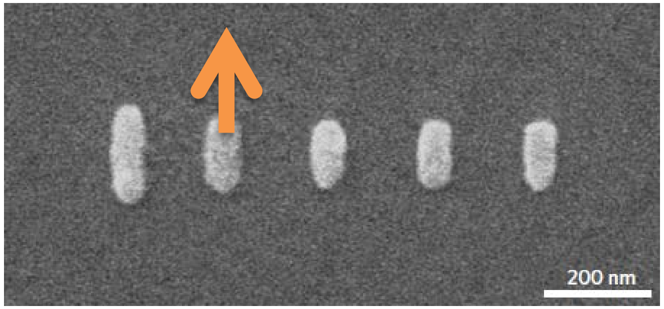
Dipole :
Highly localized source of polarized electromagnetic radiation → Highly non-directional source $\delta\vec{r}\,\delta\vec{p} \geq \hbar / 2$

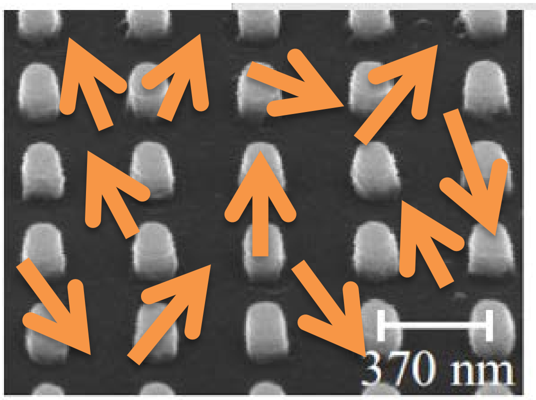


Full control on the emission characteristics of optical sources: Spectrum, efficiency, directionality and polarization

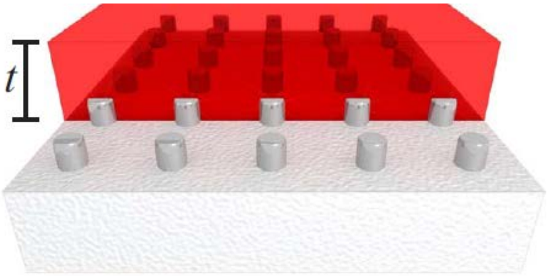
Couple emission to resonant structures ⇒ Optical antennas
Antenna + emitter = structure with designed properties

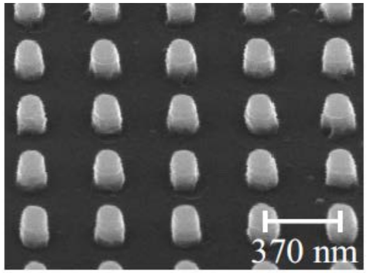






Nanoantenna phased arrays





Consider, for example, a piece of material in which we make little coils and condensers (or their solid state analogs) 1,000 or 10,000 angstroms in a circuit, one right next to the other, over a large area, with little antennas sticking out at the other end—a whole series of circuits. Is it possible, for example, to emit light from a whole set of antennas, like we emit radio waves from an organized set of antennas to beam the radio programs to Europe? The same thing would be to beam the light out in a definite direction with very high intensity. (Perhaps such a beam is not very useful technically or economically.)

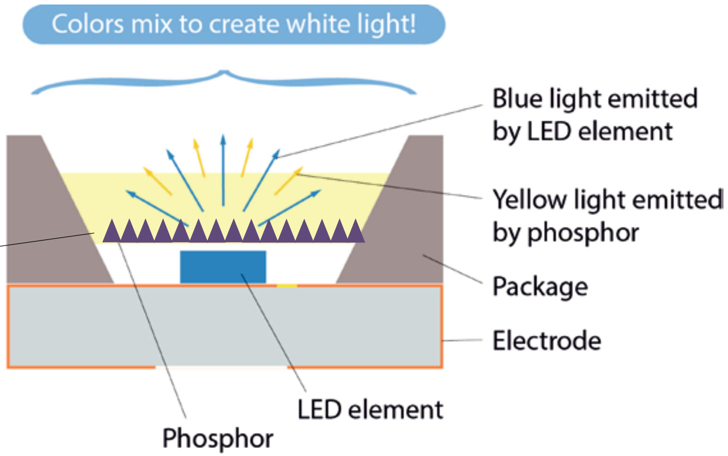
Plenty of Room at the Bottom (Feynman, 1959)

Optical nanoantennas for SSL

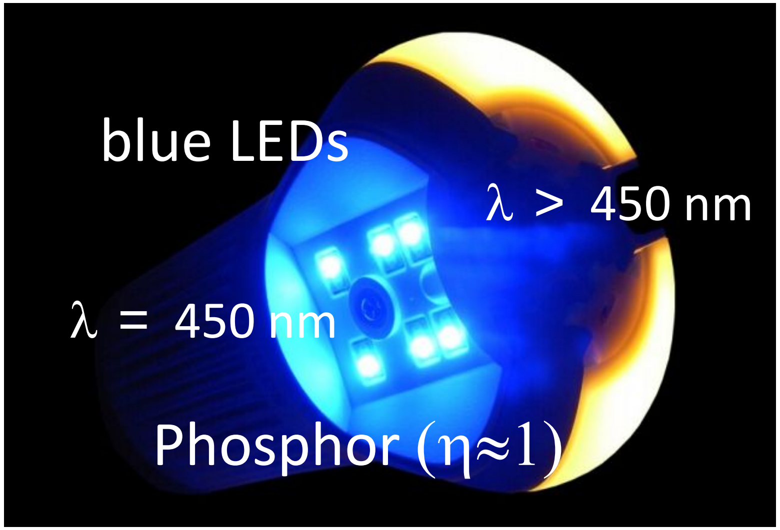
Phosphor-based LEDs

PHILIPS

LUMILEDS



Nanostructures



blue LEDs

$\lambda > 450$ nm

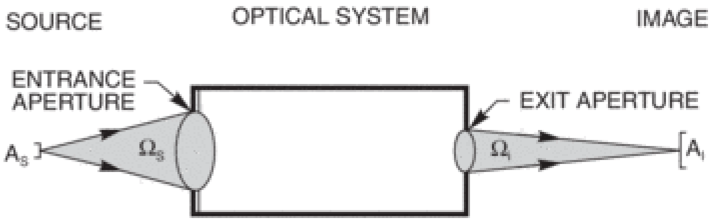
$\lambda = 450$ nm

Phosphor ($\eta \approx 1$)

S. Pimputkar et al. . Prospects for LED lighting. *Nat. Photon.* 2009; **3: 180-182.**

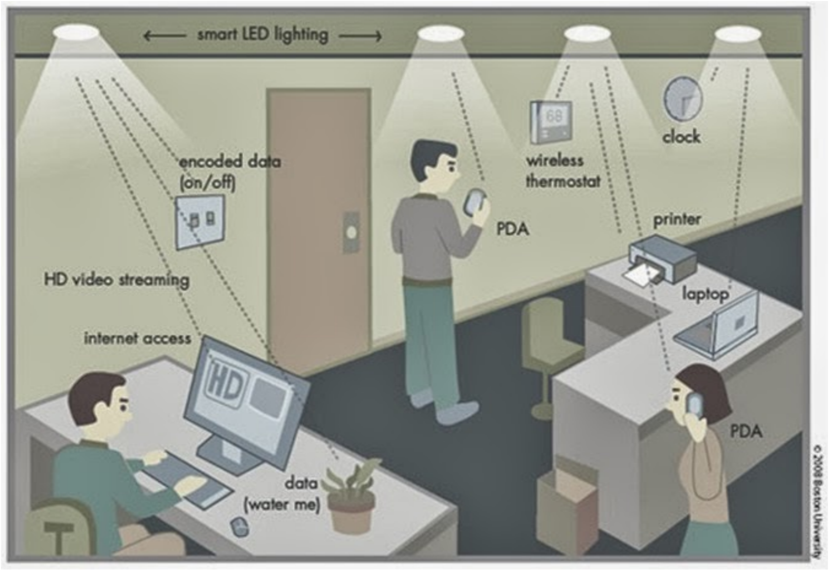
Etendue reduction for long range optical communication

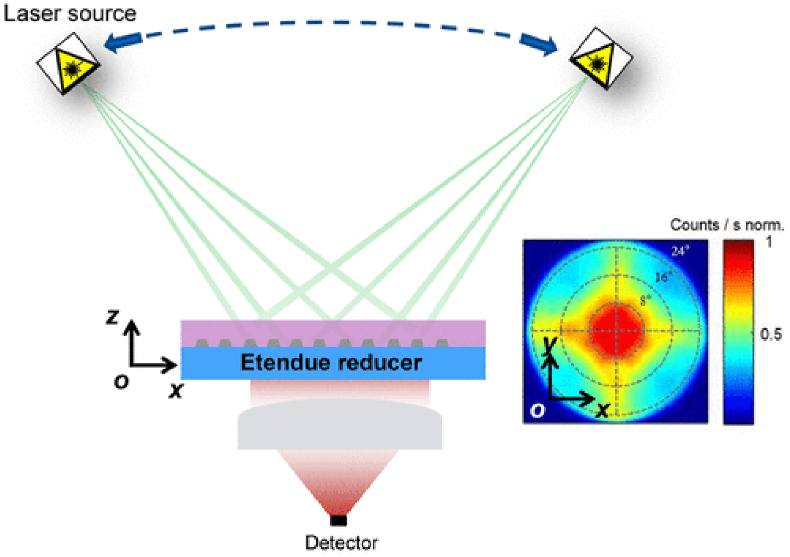
Signify



$$A_s\Omega_s \leq A_i\Omega_i$$

The large étendue in wireless optical systems limits a high-bandwidth and sensitivity detection

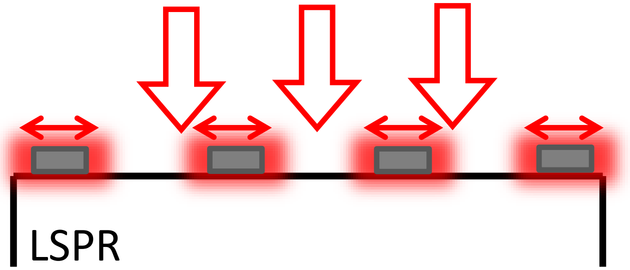




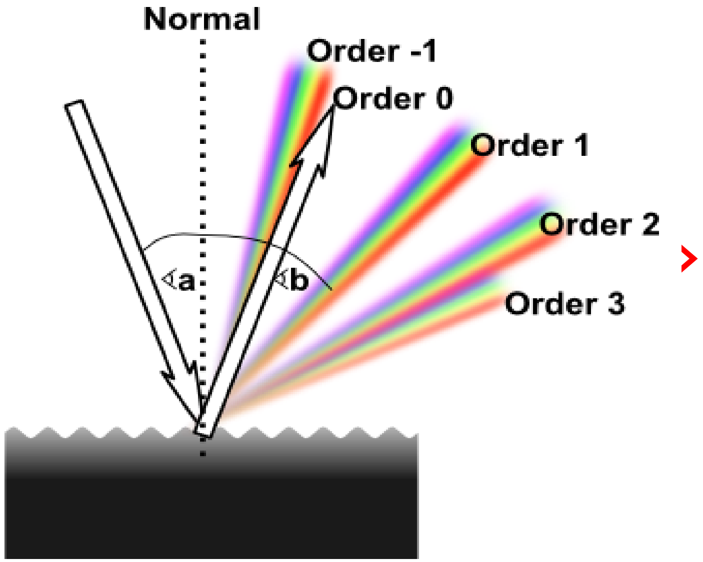
Wang et al. ACS Photonics (2018)

Collective resonances: surface lattice resonances

Localized resonance



- Localized mode
- High local fields
- High losses: radiative and material (if plasmonic)

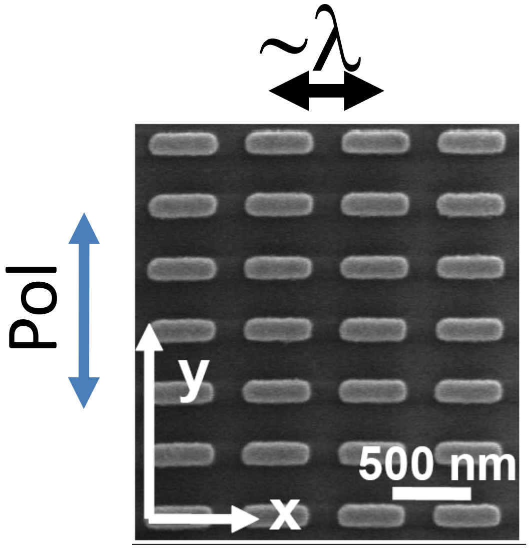


Collective resonances (non-local metasurface): Surface Lattice Resonances

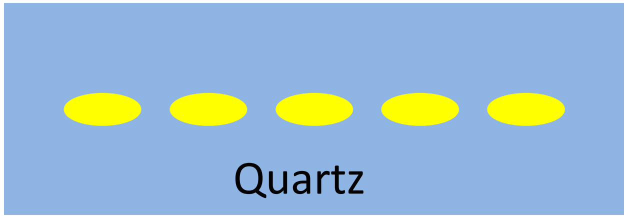
- Hybrid modes
- High local fields, small mode volume
- Low losses

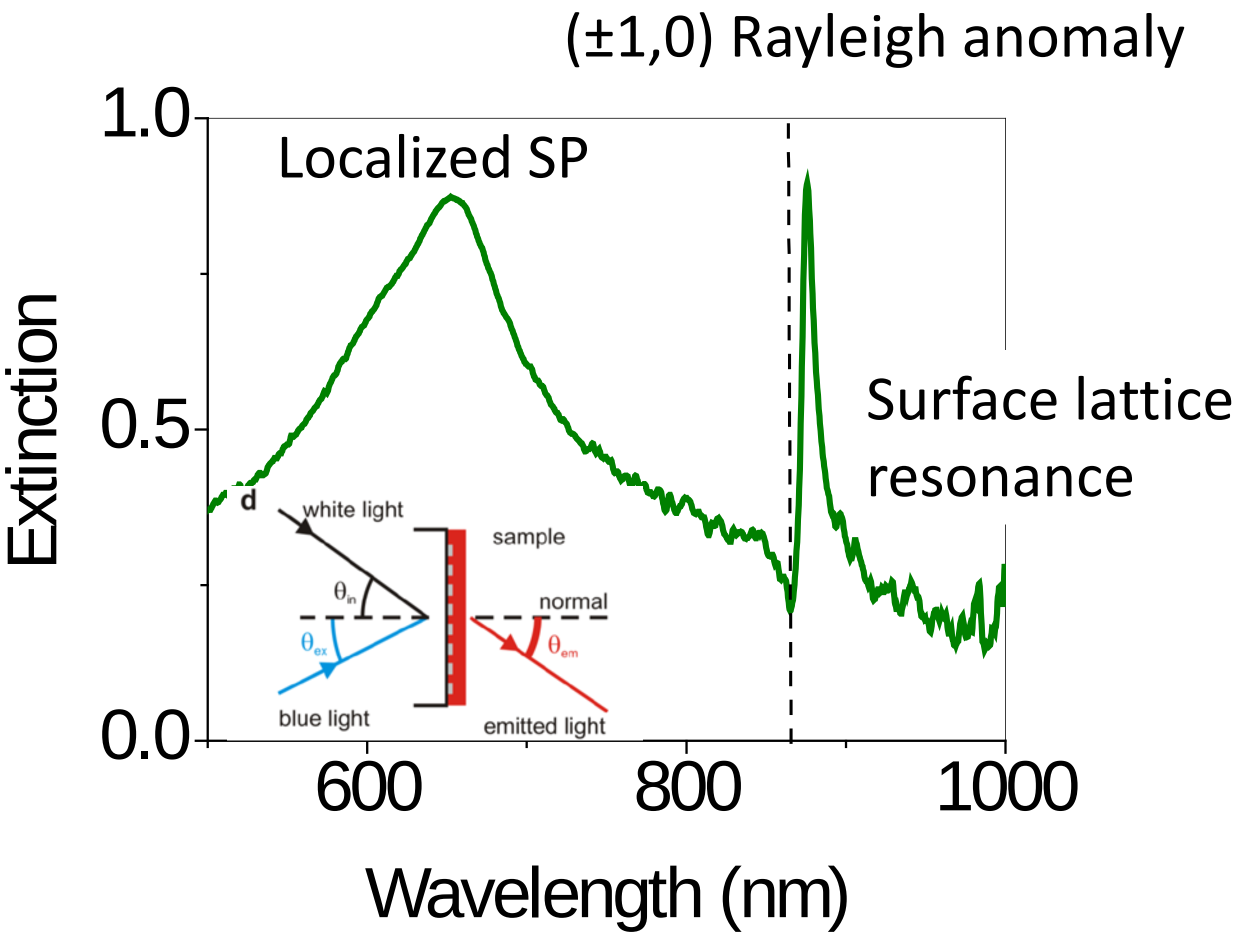
Wang, Ramezani, Väkeväinen, Törma, Gómez Rivas, Odom, Materials Today (2018)
Kravets, Kabashin, Barnes, and Grigorenko, Chem Rev. (2018)
Utyushev, Zakomirnyi, Rasskazov, Rev. Phys. (2021)

Surface Lattice Resonances (SLRs)



Gold particles
W= 85 nm, *L*= 415 nm
a_x = 500 nm, a_y = 300 nm



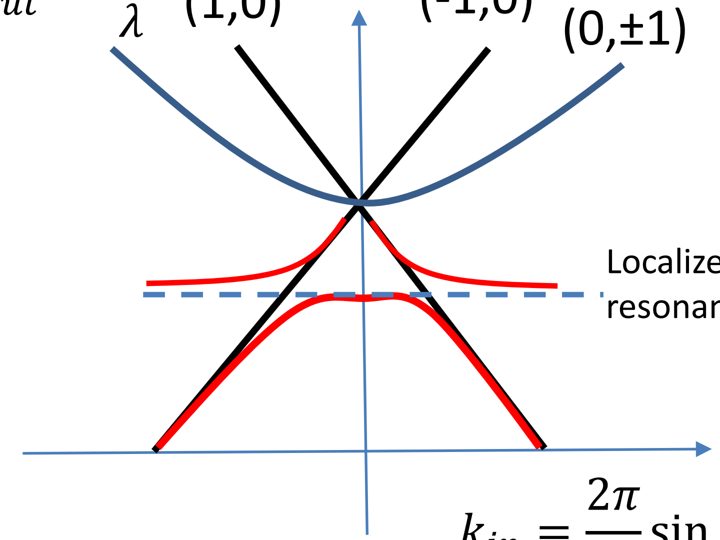


SLRs and in-plane diffraction

Grating equation: $\vec{k}_{\text{out}} = \vec{k}_{\text{in}} + m\vec{G}_x + n\vec{G}_y$

$|G_x| = \frac{2\pi}{a}$ $|G_y| = \frac{2\pi}{b}$ $(m,n) \equiv$ diffraction order

$a, b \equiv$ lattice constants



$k_{out} = \frac{2\pi}{\lambda}$

(1,0)

ω

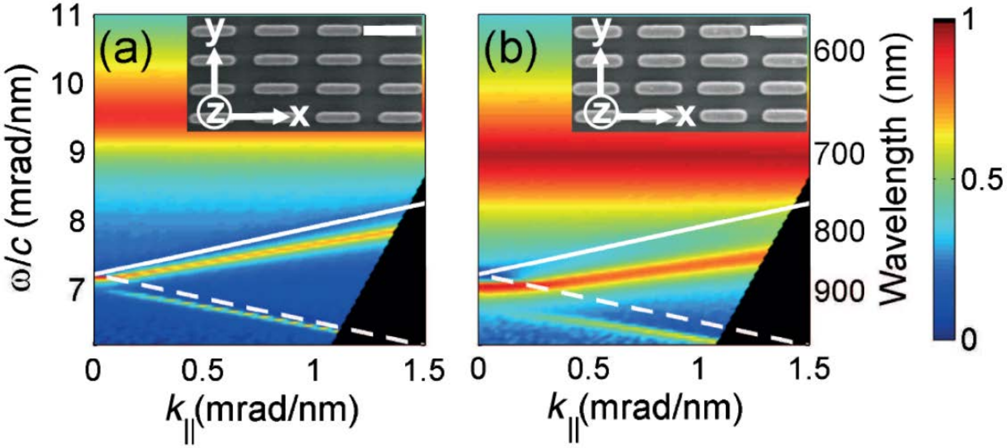
(-1,0)

(0,±1)

Localized resonance

$k_{in} = \frac{2\pi}{\lambda}\sin\theta_{in}$

Gold nanorod arrays

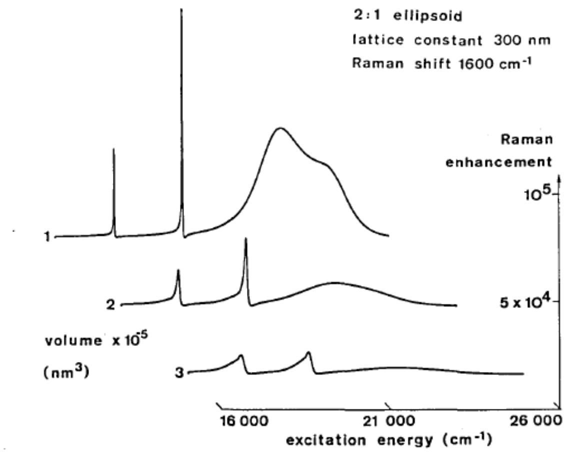


S.R.K. Rodriguez *et al.*, PRX, 1, 021019 (2011);

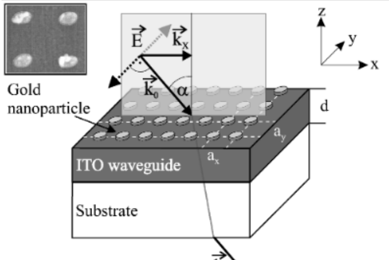
N. Meinzer *et al.*, Nature Photon. 8, 889 (2014)

Surface Lattice Resonances (SLRs)

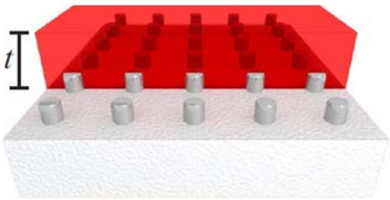
- SERS and SLRs: Carron et al. (JOSA B, 1986), Schatz et al. (J. Chem Phys, 2004)
- SLRs and extinction: Kravets (PRL 2008), Crozier et al. (APL 2008), Barnes et al. (PRL 2008)
- SLRs and spontaneous emission: Vecchi(PRL 2009), Giannini (PRL 2010), Rodriguez (PRL 2012)
- SLRs and stimulated emission: Schatz (Nat. NanoTech. 2013), Schokker (PRB2014)
- SLRs and strong coupling: Rodriguez (Opt. Exp. 2013), Torma (Nano Letters 2014)
- SLRs and polariton condensation: Ramezani (Optica 2017).
- SLRs and photon condensation: Torma (Nature Physics 2018)
- SLRs and sensing, non-linear optics, detectors, etc.



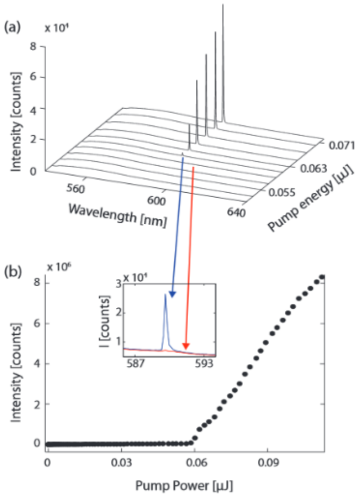
Carron et al. JOSA B, 1986



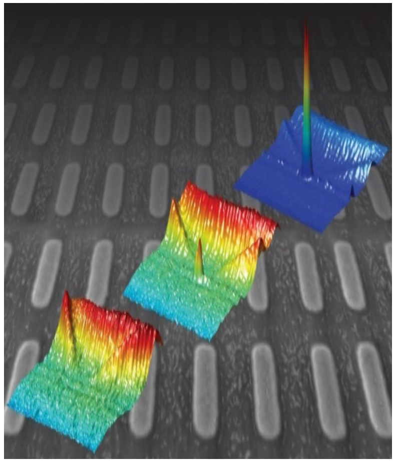
Giessen et al. PRL, 2001



Lozano et al. Nanoscale, 2014



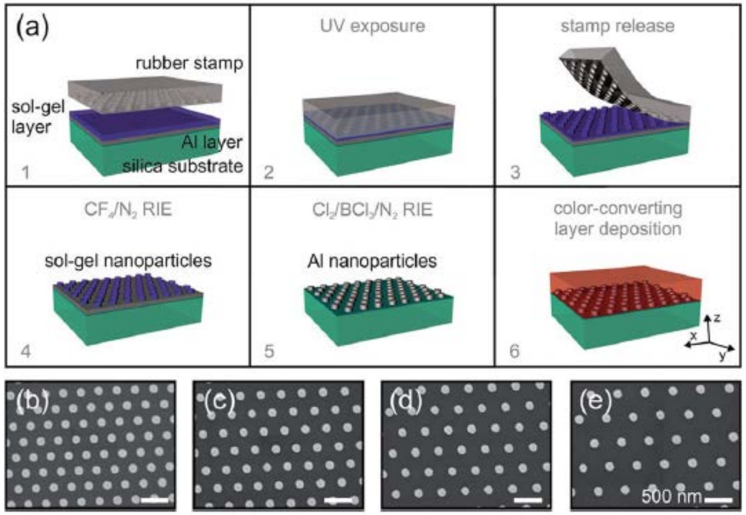
Schokker, Koenderink PRB, 2014

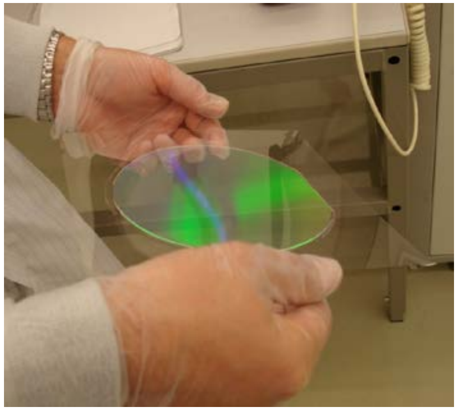


Ramezani, Optica 2017

Surface Lattice Resonances (SLRs)

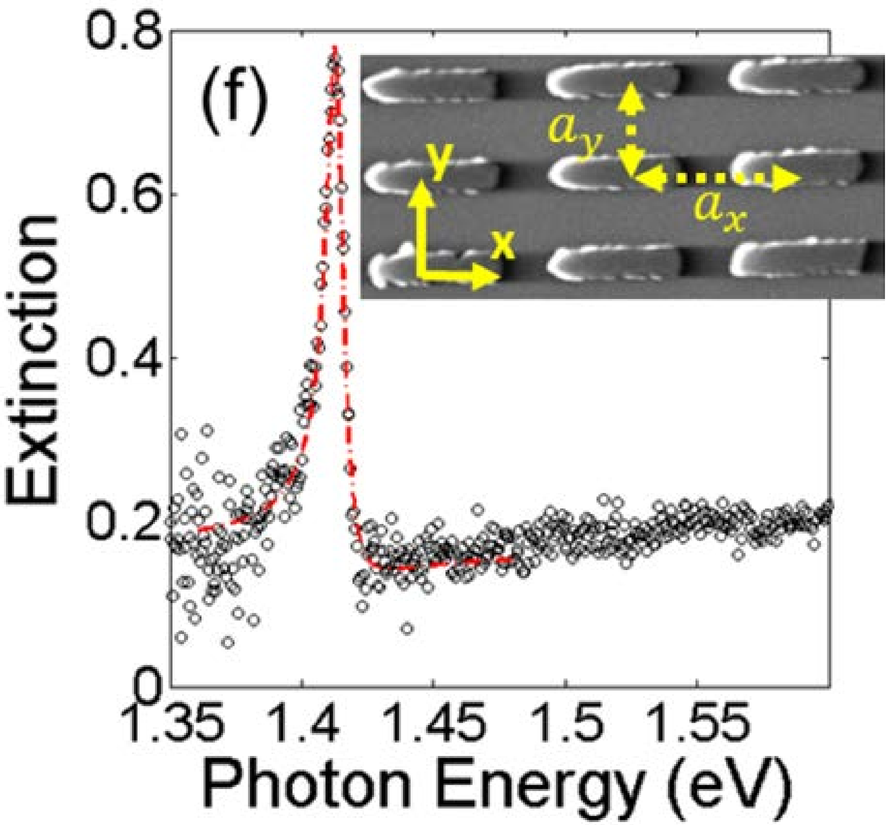
Large array fabrication:
Surface Conformal Imprint Lithography





SCIL
Nanoimprint solutions

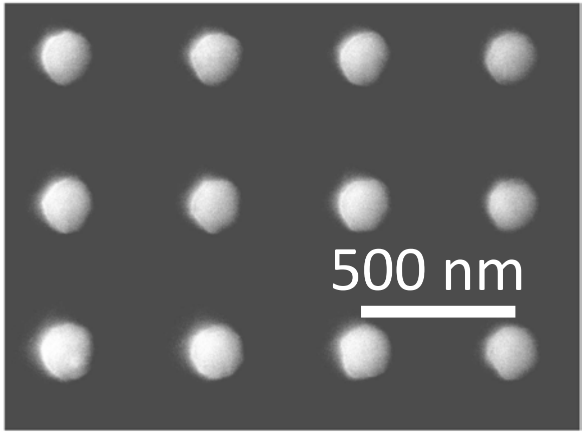
~1 nm linewidth Fano resonance
in Au nanorod array



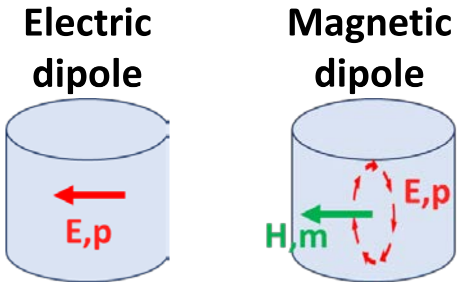
Lozano et al. Nanoscale (2014); A. Abass *et al.,* ACS Photonics (2014).

Dielectric surface lattice resonances (Mie-SLRs)

Si arrays

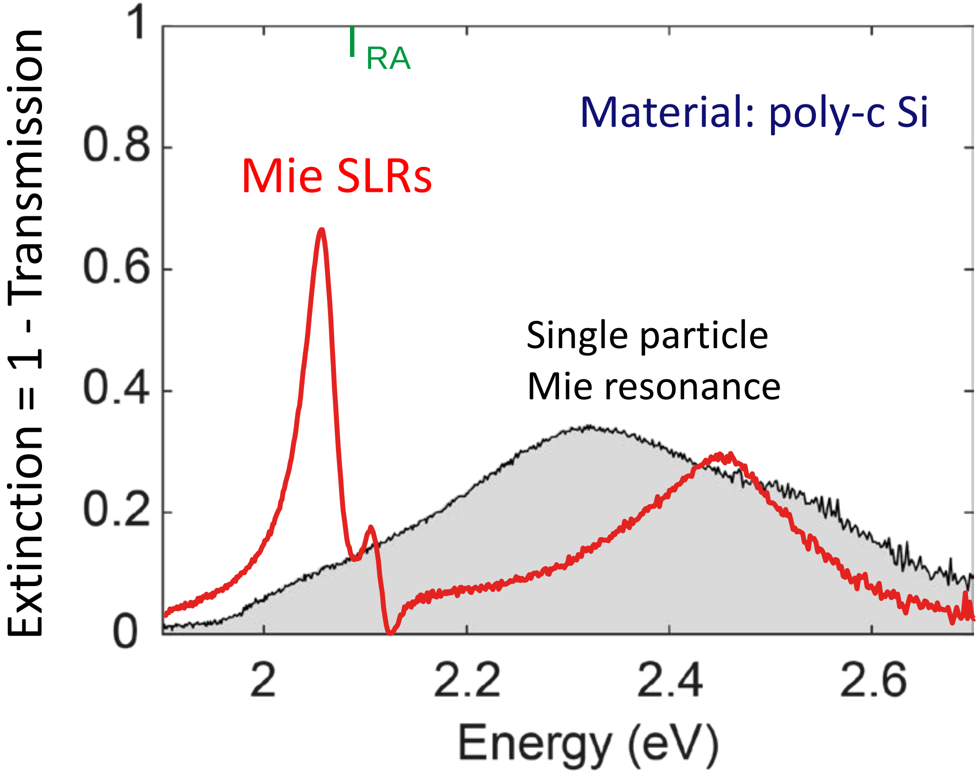


Diameter = 120 nm
Height = 90 nm



Reduce or suppress material losses

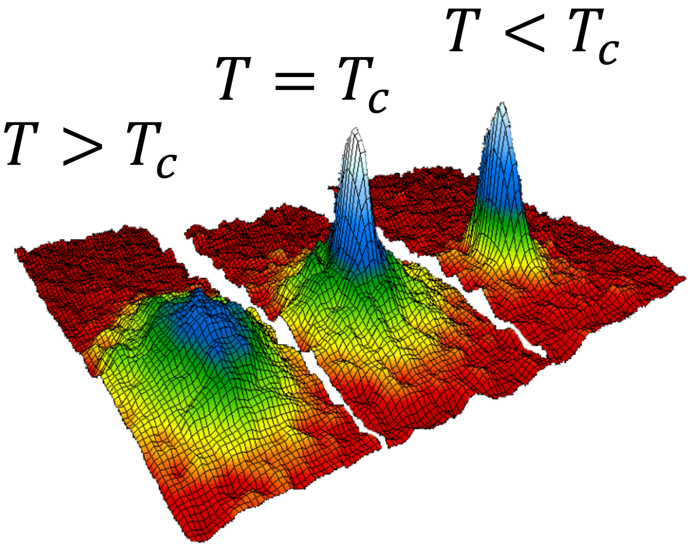
Cavities for strong light-matter coupling and polariton condensation



***G. Castellanos...JGR, ACS Photonics* 7, 5, 1226 (2020);**
Heilmann...Törma, Nanophot. 9, 267 (2020) ; Todisco...Tserkezis, Nanophot. 9, 803 (2020)

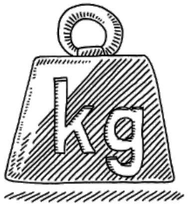
Bose Einstein condensates (polariton lasing)

Ground-state accumulation of bosons at high n & low T



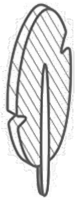
$T_c \propto 1/m$

Large m
Low T_c



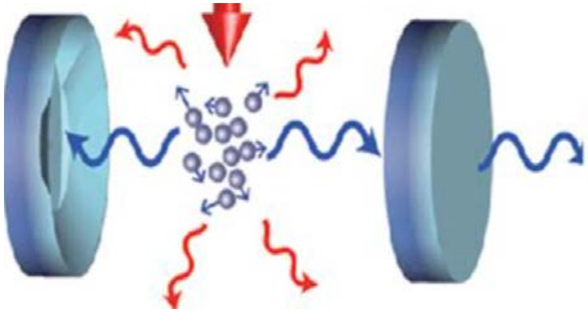
rubidium-87, sodium-23

Small m
High T_c



Excitons – polaritons

- Science 269, 198 (1995)
- PRL 75, 3969 (1995)

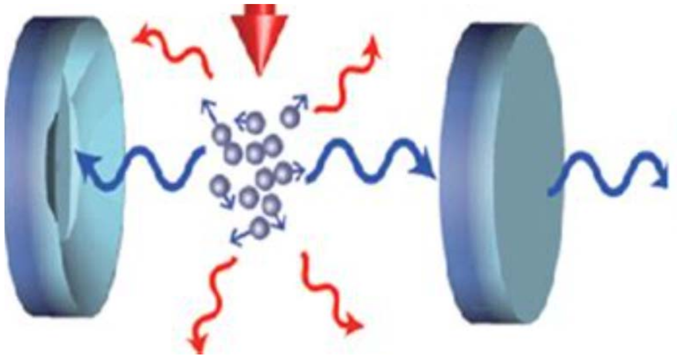


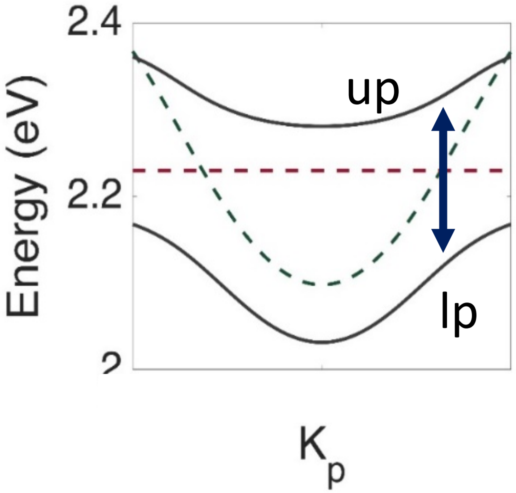
2001
Cornell, Ketterle, Wieman

- Imamoglu et al., PRA **53**, 4250 (1996)
- Deng et al., Science **298**, 199 (2002)
- Kasprzak et al., Nature **443**, 409 (2006)

Exciton-Polaritons (Strong light-matter coupling)

Nanophotonic cavities & organic molecules





Rabi energy ($\hbar\,\boldsymbol{\Omega}$)

Collective coupling st n:

$$g = \frac{\hbar\Omega}{2} = \sqrt{\frac{ncN}{\lambda\varepsilon\varepsilon_o V}}$$

μ_m ≡ Tr on dipole moment

N ≡ N er of excitons

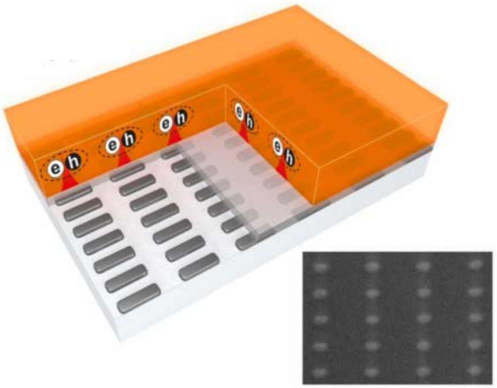
V ode volume

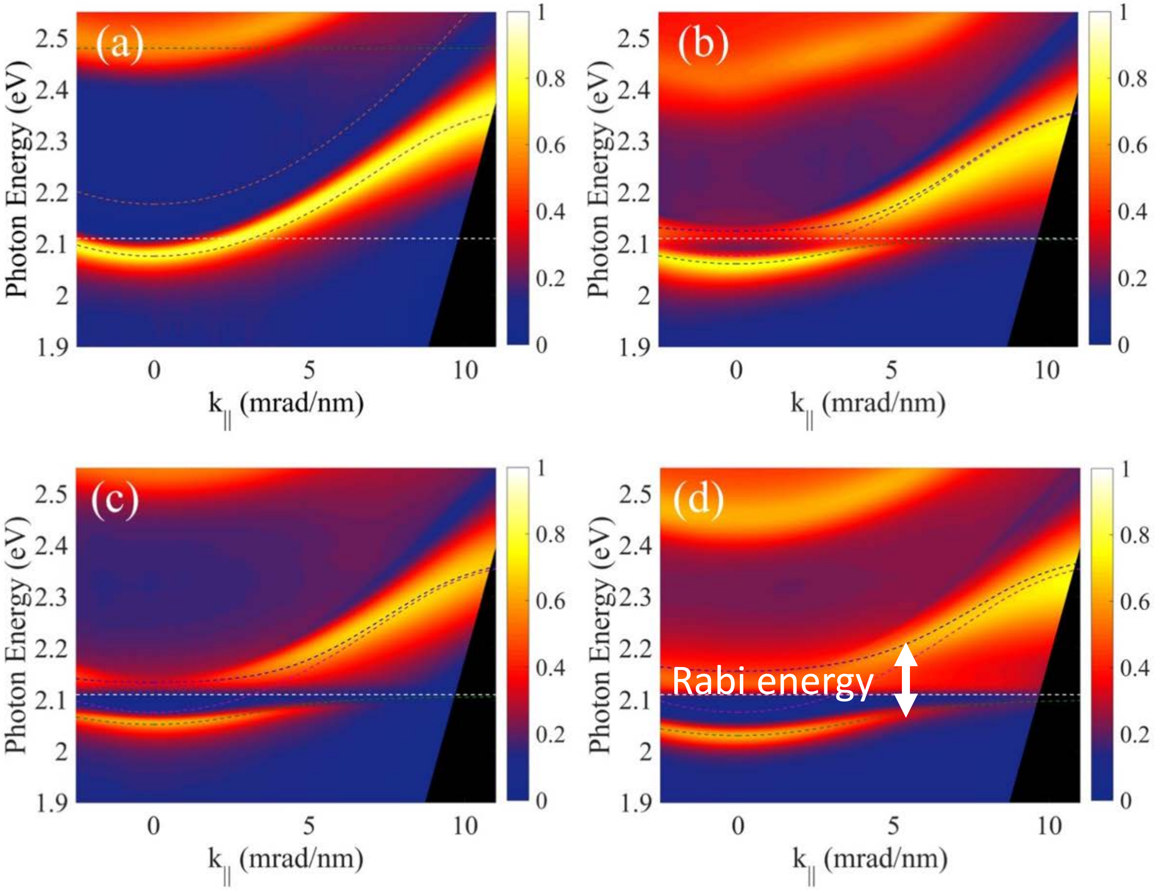
Strong coupling when Rabi frequency is larger than the cavity loss rate and the exciton decoherence rate

J. Kasprzak, et.al., Nature 443, 409-414 (2006)

- Kéna-Cohen, Forrest, Nat. Photonics 4, 371 (2010)
- K. S. Daskalakis et al., Nat. Materials 13, 271 (2014)
- Plumhof et al., Nat. Materials 13, 247 (2014)

Exciton-Polaritons (Strong light-matter coupling)




Bare array

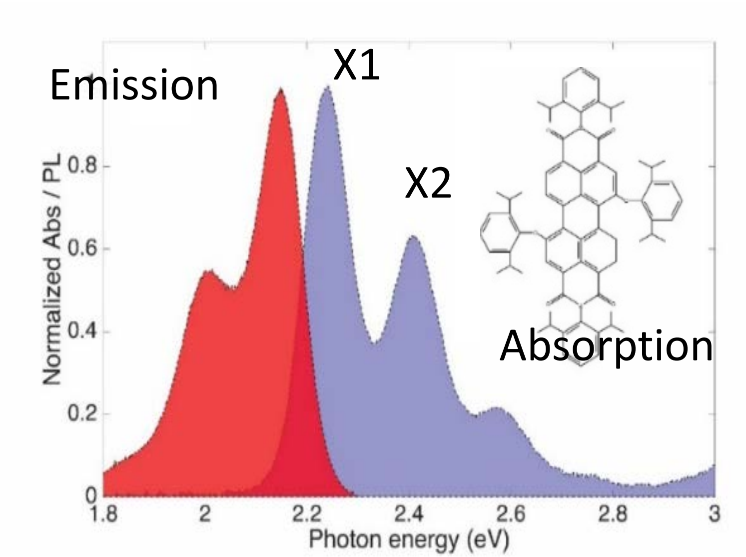
1 layer - TDBC

3 layers - TDBC

6 layers - TDBC

Exciton-Polaritons (Strong light-matter coupling)

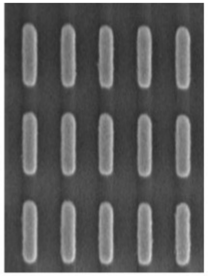
Rylene dye



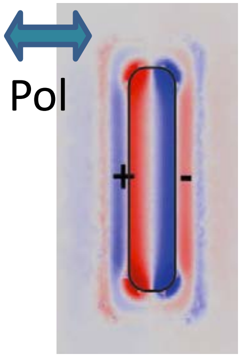
- Plasmon-exciton-polariton condensation:
  Plasmonic (Ag) metasurface
- Mie-exciton-polariton condensation:
  Dielectric (pc-Si) metasurface
- BIC-exciton-polariton condensate

Plasmon-Exciton-Polariton condensation

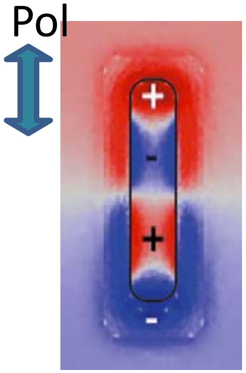
Ag nanoparticles



Dipole

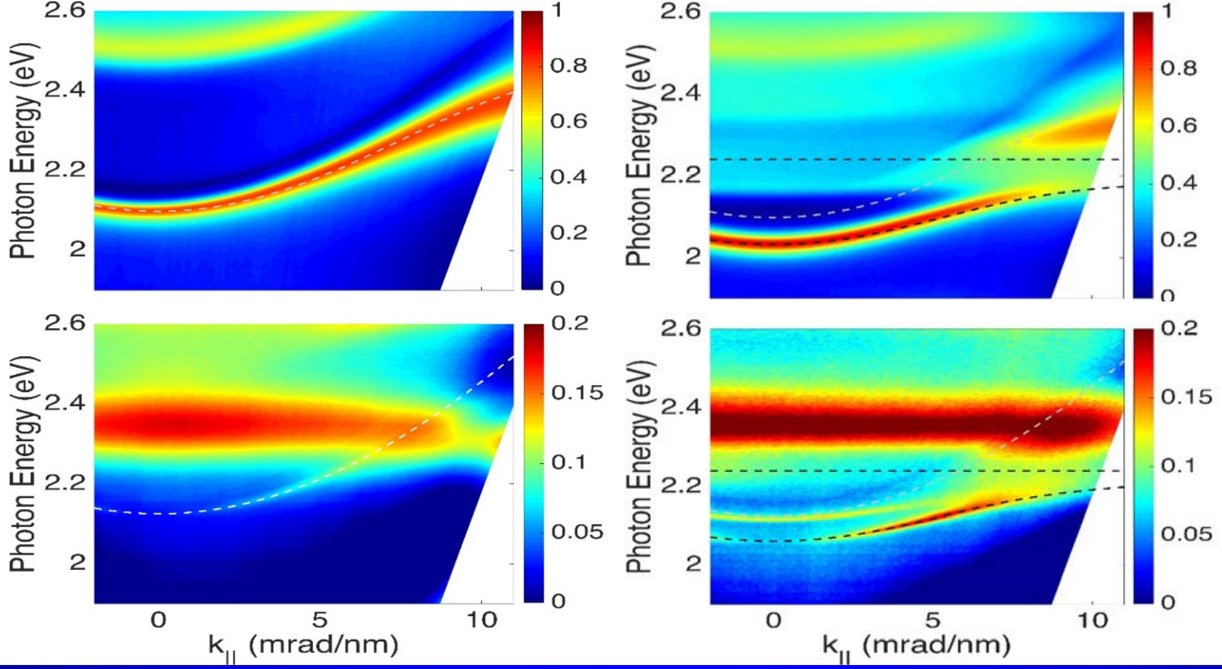


Multipole



No molecules (Bare modes)

35 wt% Molecules



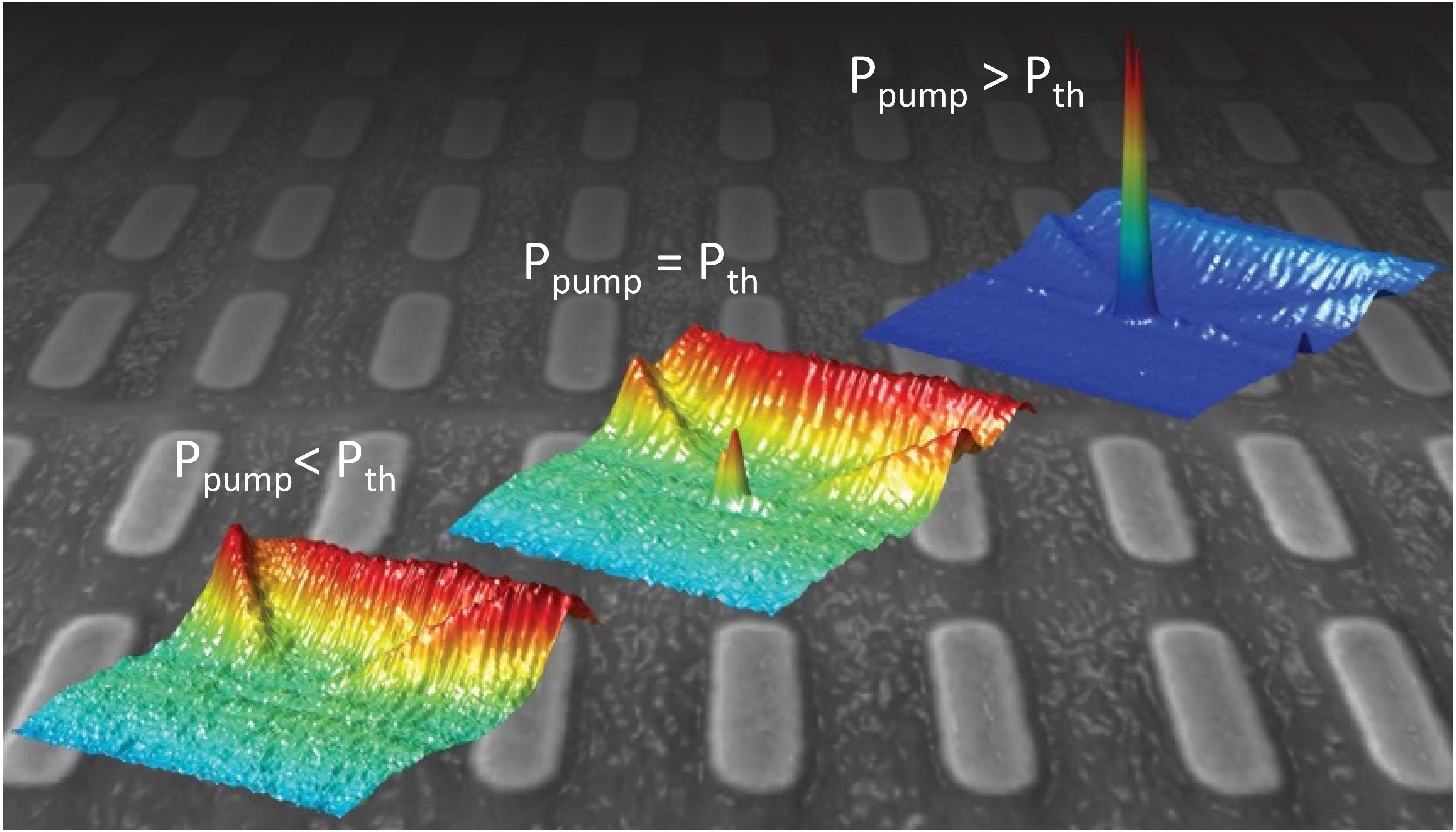
Polaritons with strong radiative decay (bright)

Polaritons with reduced radiative decay (dark)

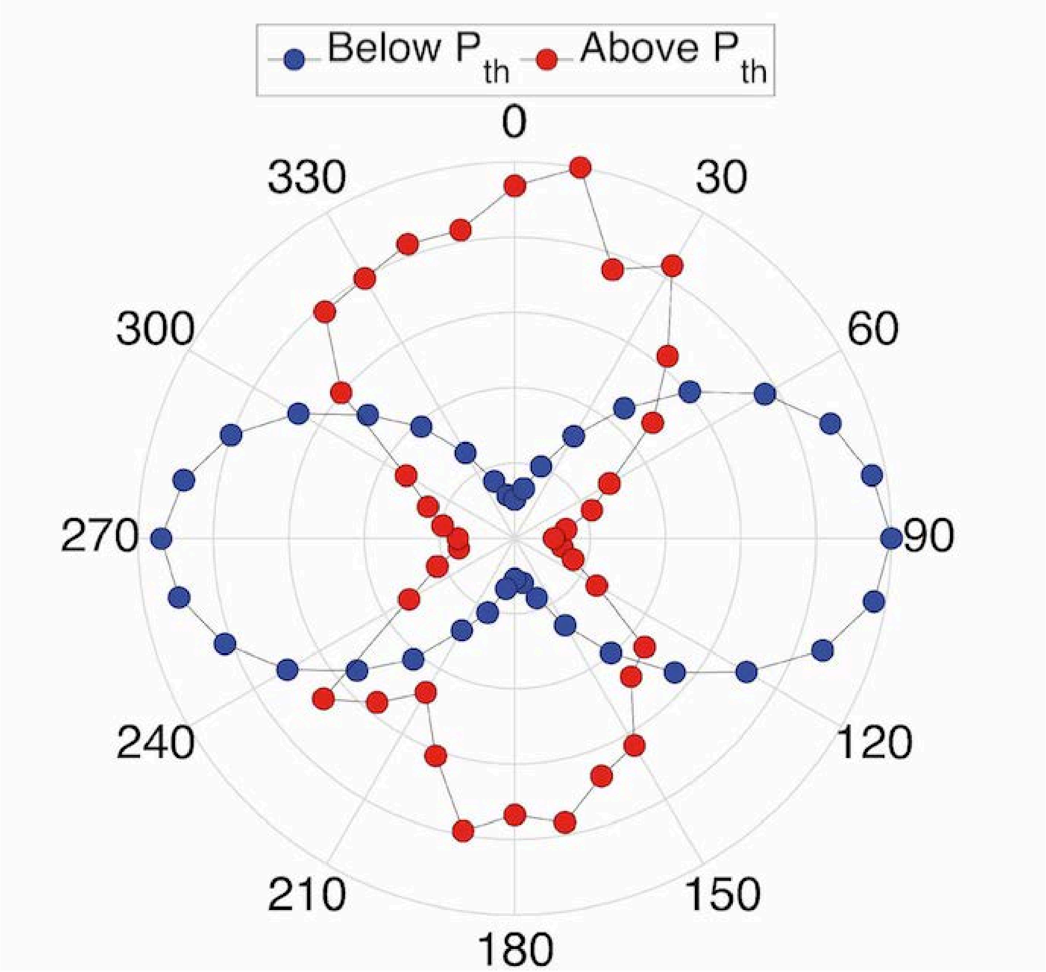
S.R.K. Rodriguez et al., Opt. Express Optics Express 21, 27411 (2013).

M. Ramezani, et. al., Optica 4, 31 (2017).

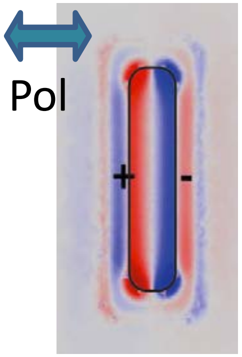
Plasmon-Exciton-Polariton condensation



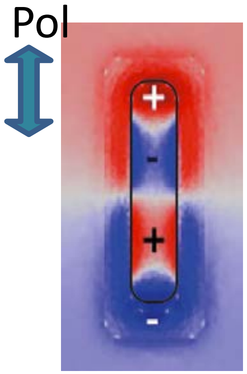
Plasmon-Exciton-Polariton condensation: polarization



Dipole

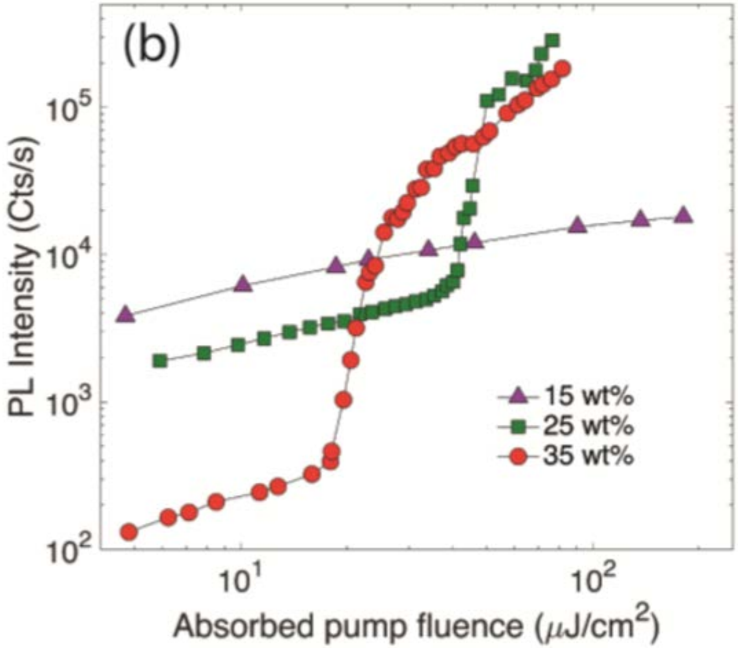


Multipole

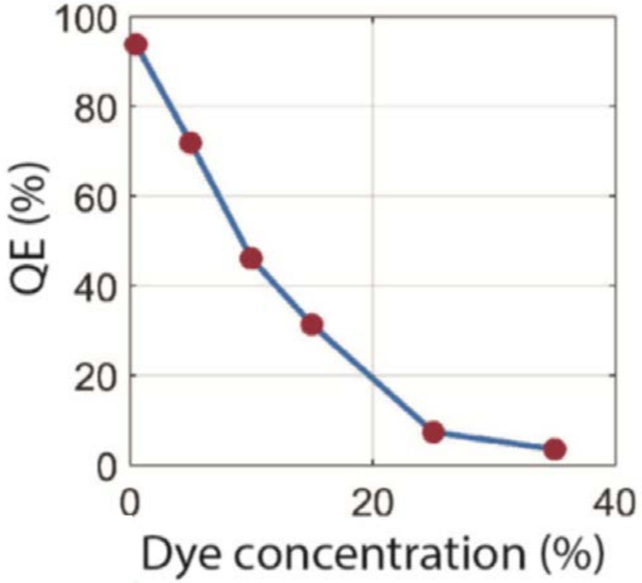


M. Ramezani, et. al., Optica, 4(1), ***2017***

Plasmon-Exciton-Polariton condensation: threshold



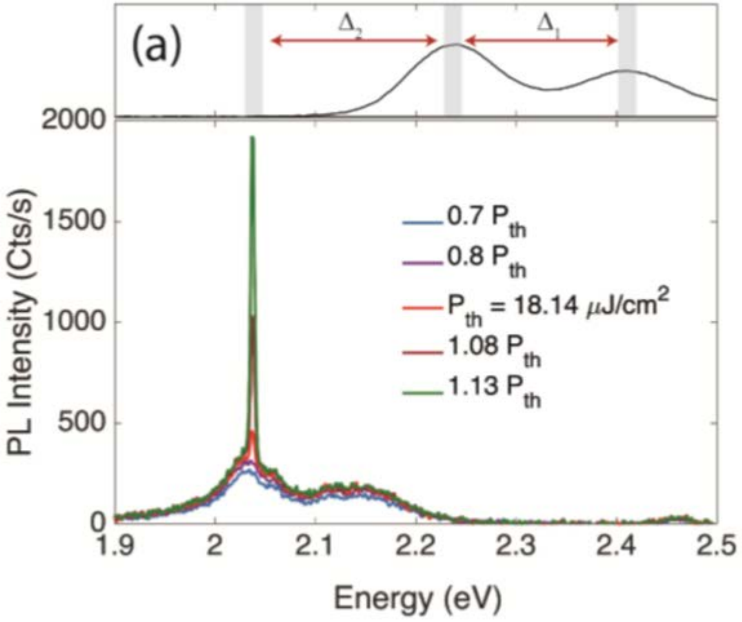
Emission quantum efficiency of dye in PMMA

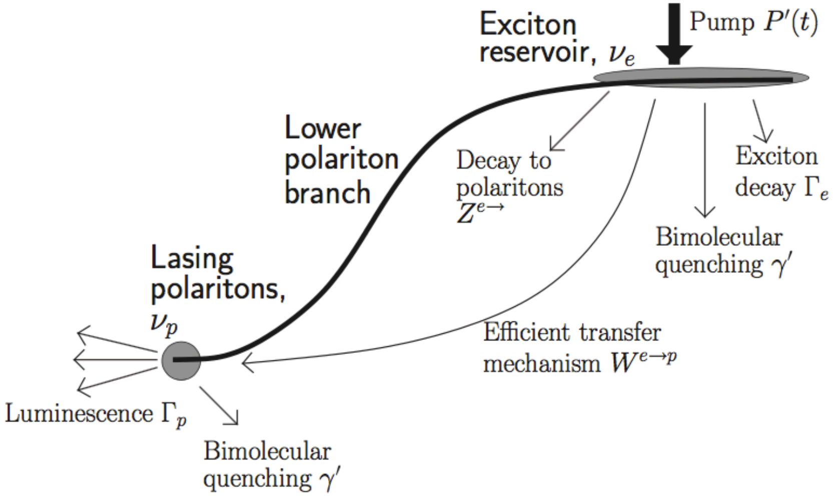


Stimulated scattering of polaritons responsible for condensation in contrast to stimulated emission responsible for lasing

M. Ramezani, et. al., Optica, 4(1), ***2017***

Plasmon-Exciton-Polariton lasing





Transfer mechanism ⇒ Vibronic assisted relaxation

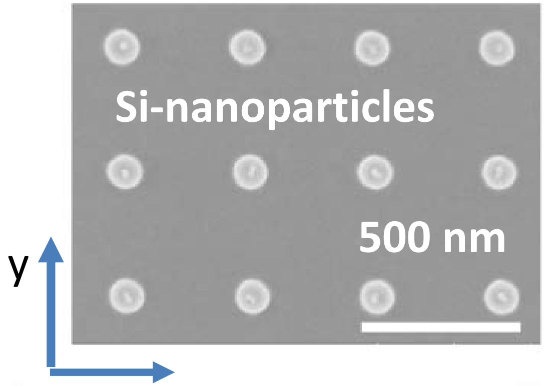
Mazza et al., PRB **88**, 2013

Nomaschi et al., APL **99**, 2011

M. Ramezani, et. al., Optica, 4(1), 2017

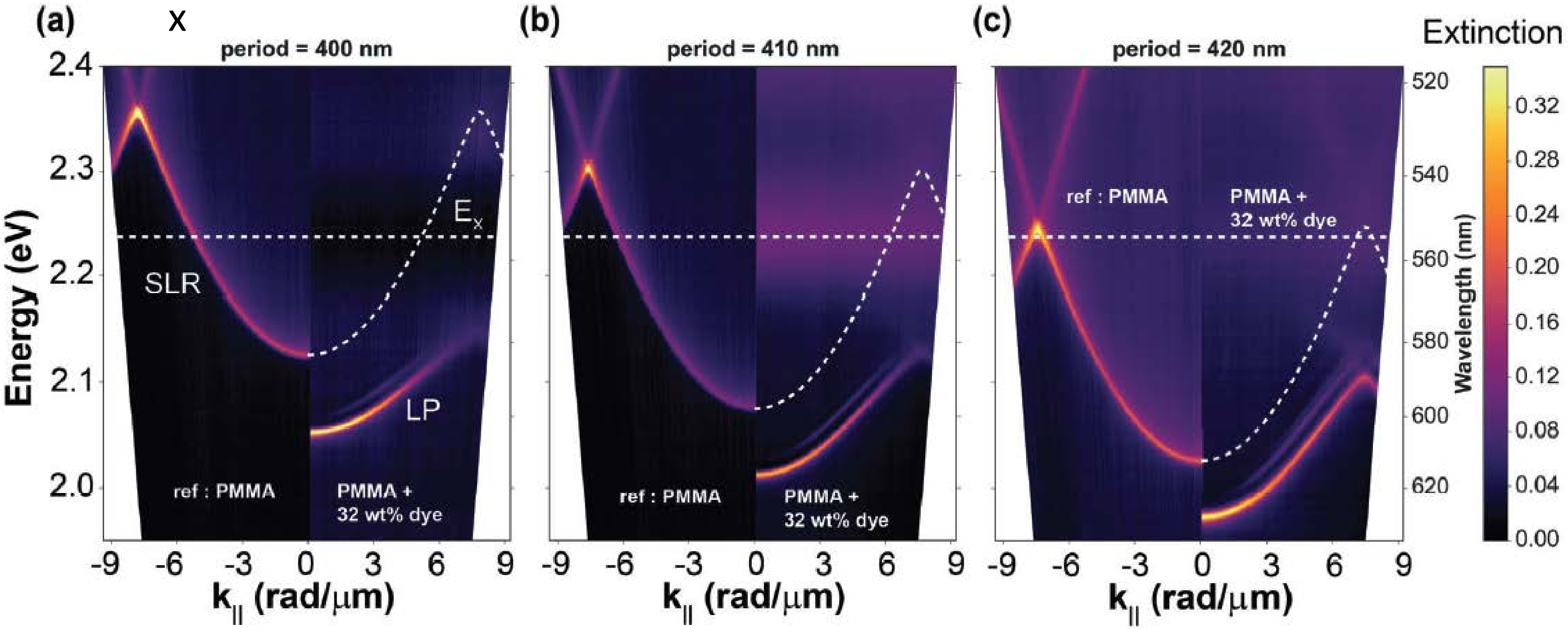
M. Ramezani, et al., Nano Letters 19, 8590 (2019)

Mie-Exciton-Polariton condensation



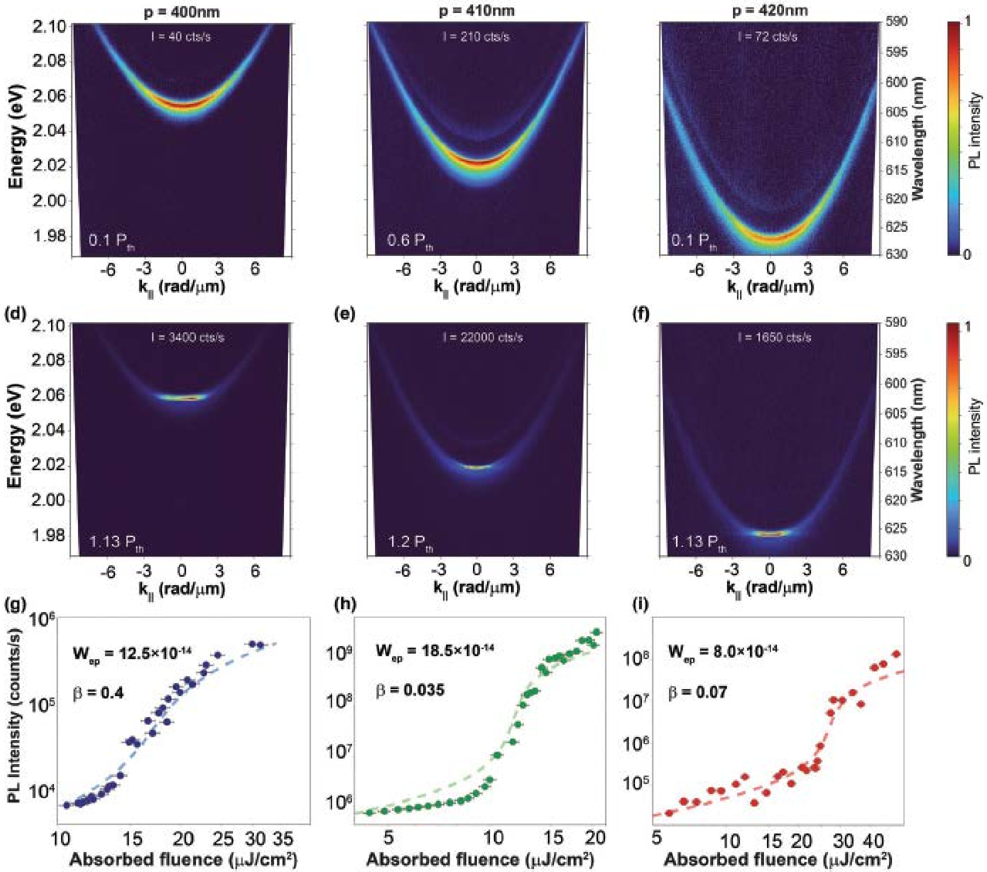
x-polarization

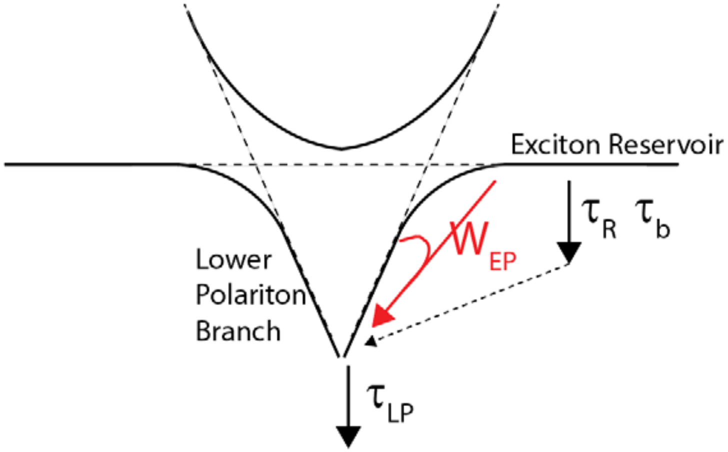
$$k_{||} = k_x$$



G. Castellanos et al., Adv. Opt. Mat. 11, 2202305 (2023)

Mie-Exciton-Polariton condensation





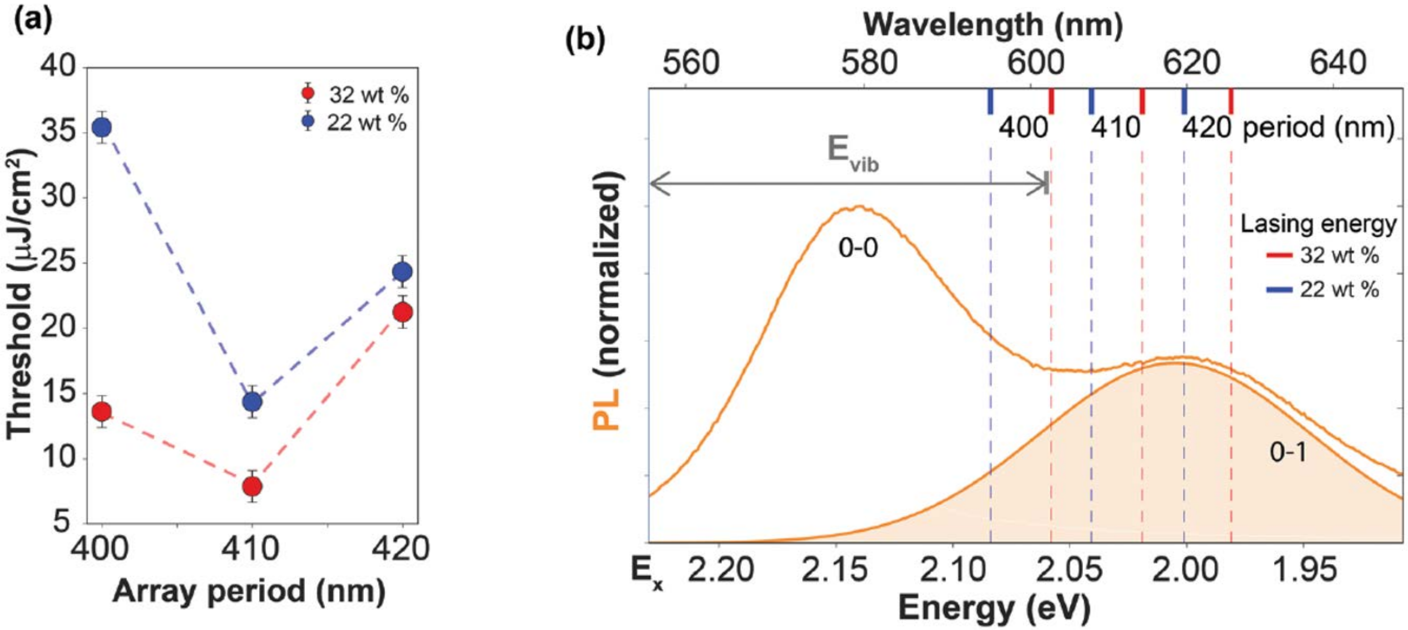
$$\frac{dn_R}{dt} = \left(1 - \frac{n_R}{N_0}\right) P(t) - \frac{n_R}{\tau_R} - \frac{n_R^2}{\tau_b} - W_{ep} n_R n_{LP}$$

$$\frac{dn_{LP}}{dt} = W_{ep} n_R n_{LP} + \beta \frac{n_R}{\tau_R} - \frac{n_{LP}}{\tau_{LP}}$$

- **Radiative relaxation**
- **Vibronic assisted relaxation**

G. Castellanos et al., Adv. Opt. Mat. 11, 2202305 (2023)

Mie-Exciton-Polariton condensation



- For short periods radiative relaxation is less important
- For long periods vibronic assisted relaxation is less important
- Lowest threshold for intermediate periods

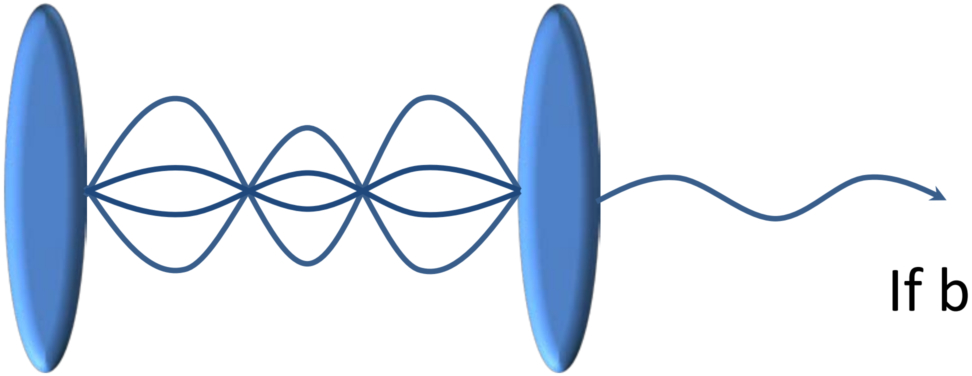
$$\frac{dn_{LP}}{dt} = W_{ep} n_R n_{LP} + \beta \frac{n_R}{\tau_R} - \frac{n_{LP}}{\tau_{LP}}$$

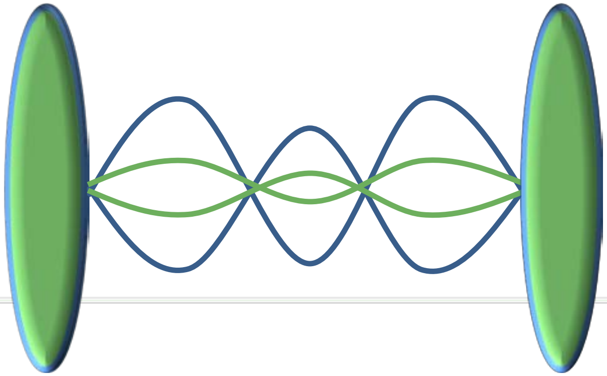
τ_{LP} is limited by the cavity lifetime

G. Castellanos et al., Adv. Opt. Mat. 11, 2202305 (2023)
Hulkko et al., J. Chem. Phys. 154, 154303 (2021); Michetti & Rocca, PRB 79, 035325 (2009)

How good can be a cavity?

- Radiation losses
- Material losses





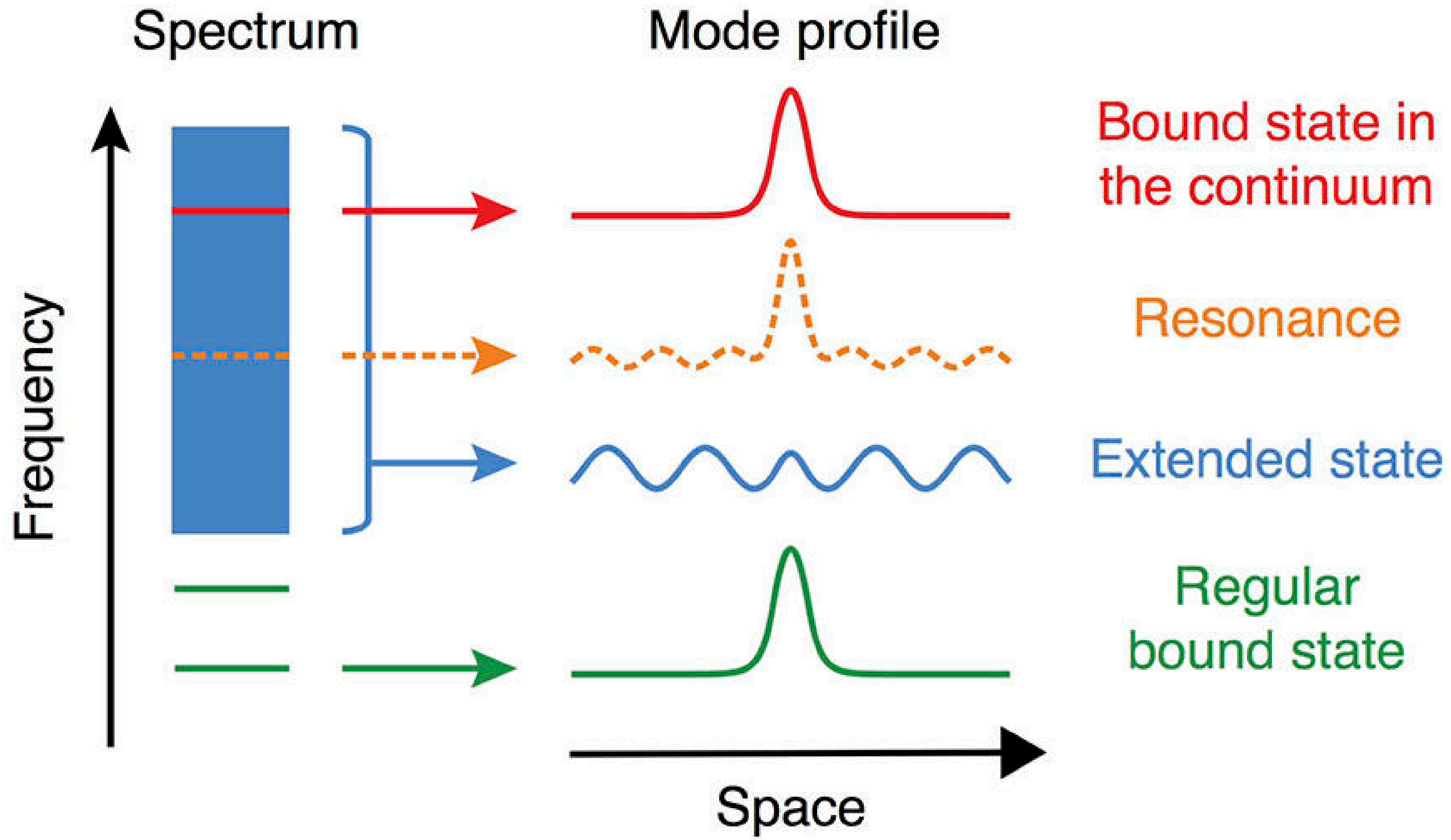
If both losses are suppressed

Lifetime: $\tau_{cav} = \infty$

Linewidth: $\Delta\omega = 0$

Q-factor: $Q = \frac{\omega}{\Delta\omega} = \infty$

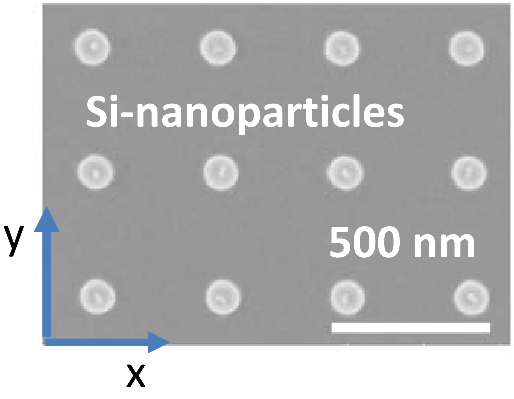
Bound states in the continuum (BICs)



Hsu, Zhen, Stone, Joannopoulos and Soljacic, Nature Reviews (2016)

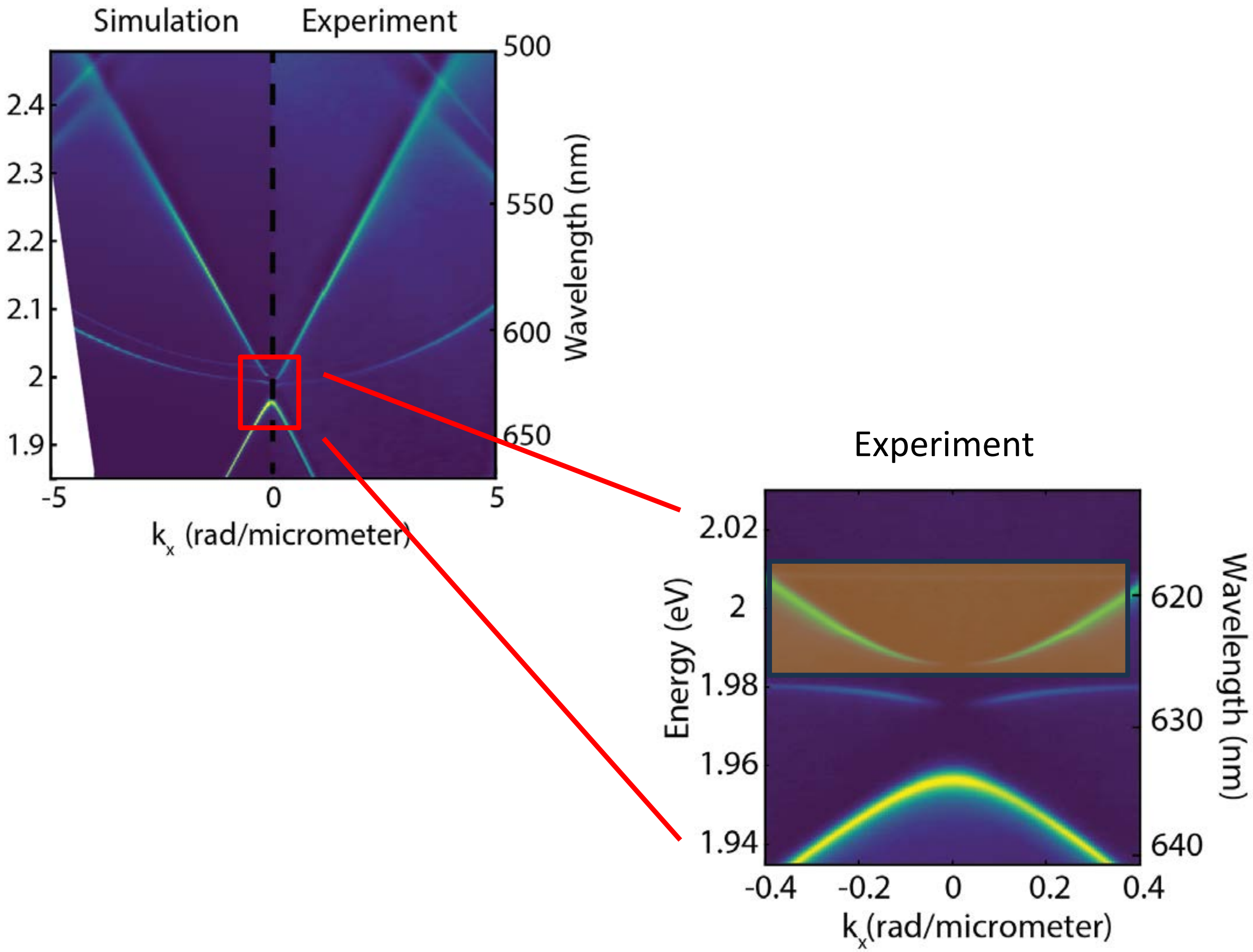
BICs in arrays of Si nanoparticles

a= 420 nm



y-polarization

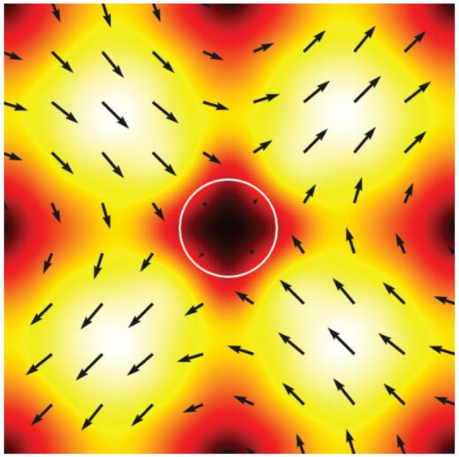
$$k_{||} = k_x$$

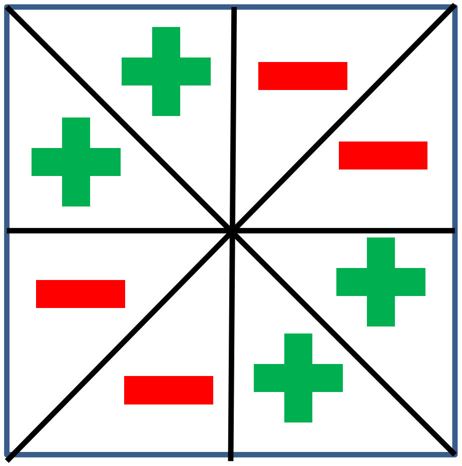


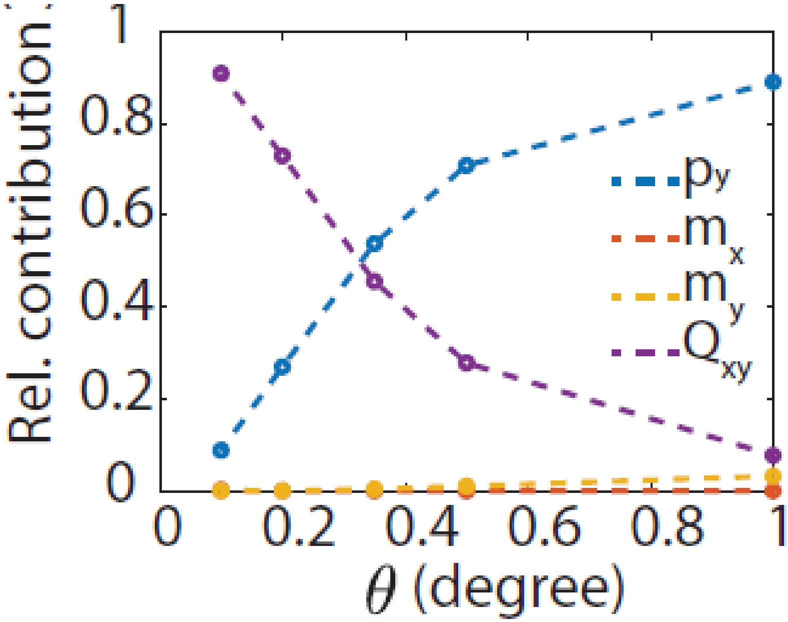
Ohtaka, J. Phys. Soc. Jpn. ***65****, 2670 (1996).*
Mohamed, Laser Photonics Rev. 16, 2100574 (2022)

BICs in arrays of Si nanoparticles

BICs arise due to a symmetry mismatch between free space radiation and the modes in the metasurface



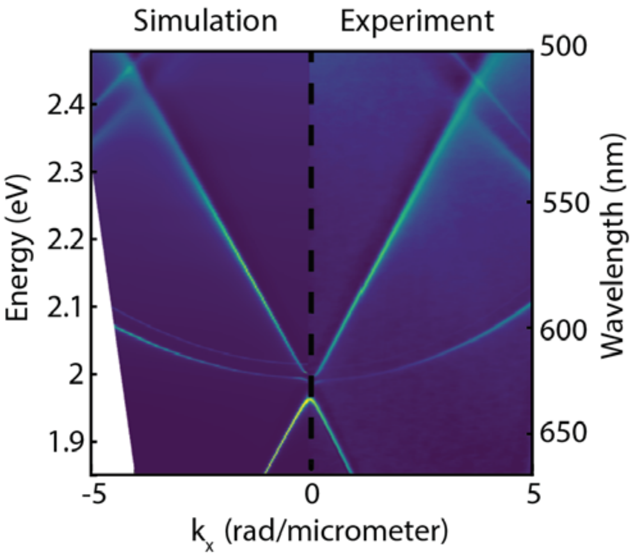




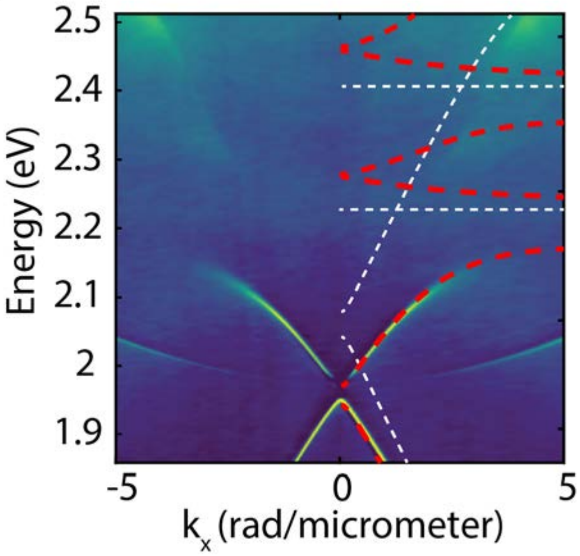
Ohtaka, J. Phys. Soc. Jpn. ***65****, 2670 (1996).*
Mohamed, Laser Photonics Rev. 16, 2100574 (2022)

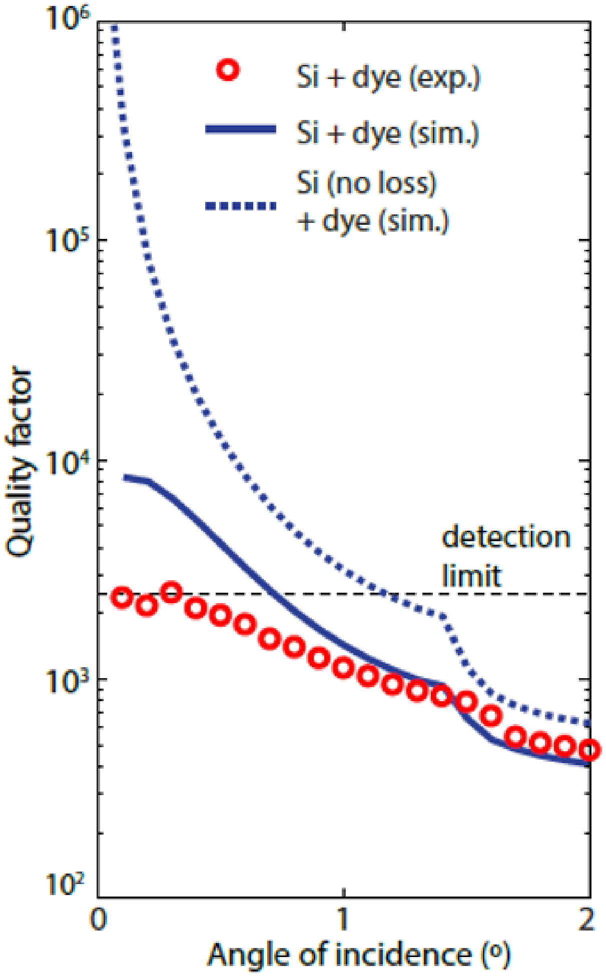
BIC- Exciton-Polaritons

Si and no dye



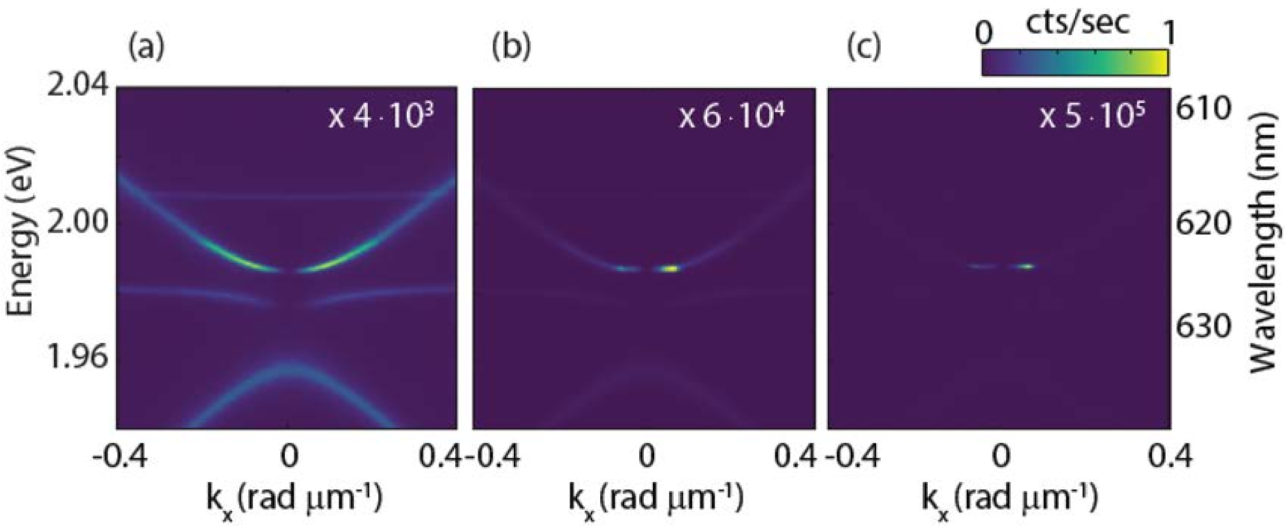
Si + dye (32 %wt)



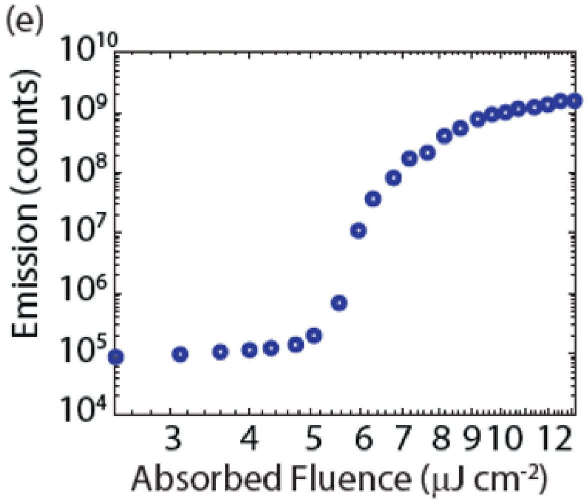


M. Berghuis et al., Nano Lett. 23, 5603 (2023)

BIC- Exciton-Polariton condensation

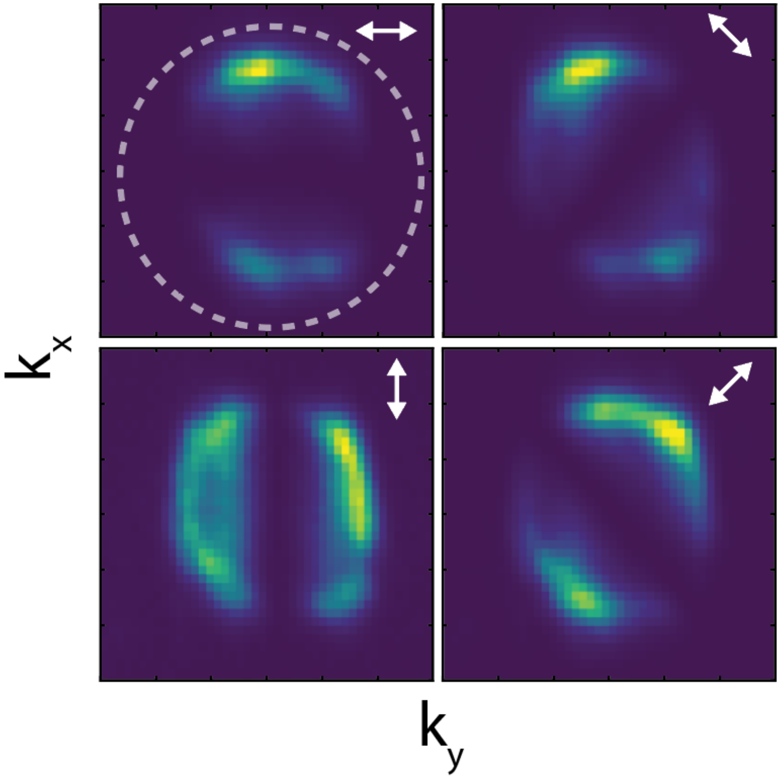


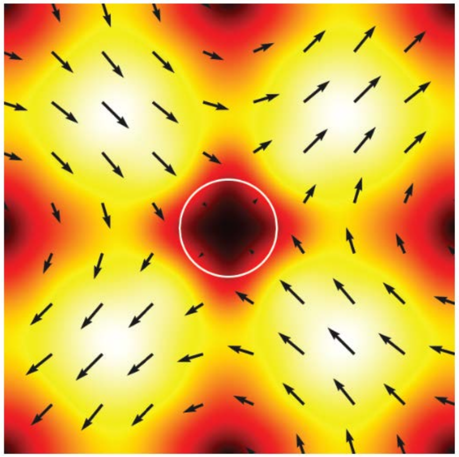
Low threshold of 5 μJ cm^{-2}



M. Berghuis et al., Nano Lett. 23, 5603 (2023)

BIC- Exciton-Polariton condensation

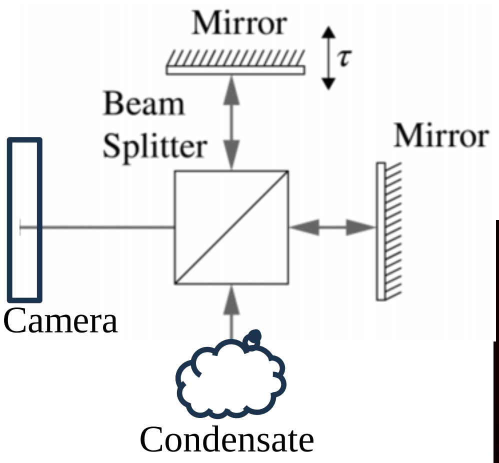




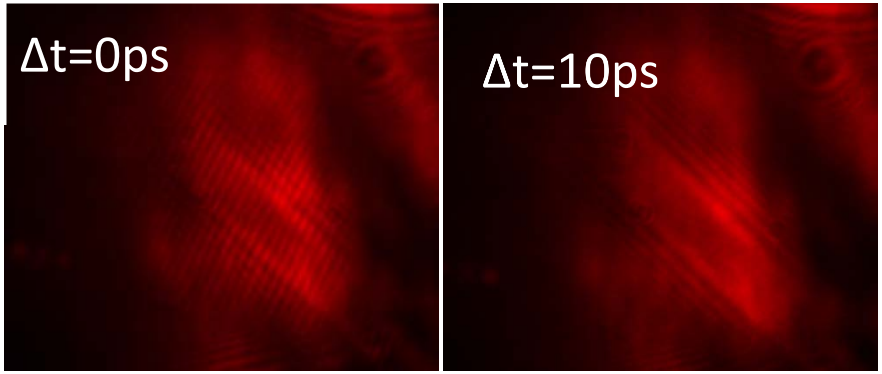
The vortex emission from the condensate shows the quadrupolar character of the mode

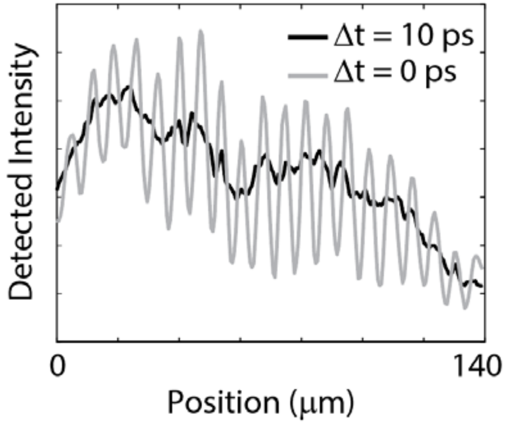
M. Berghuis et al., Nano Lett. 23, 5603 (2023)

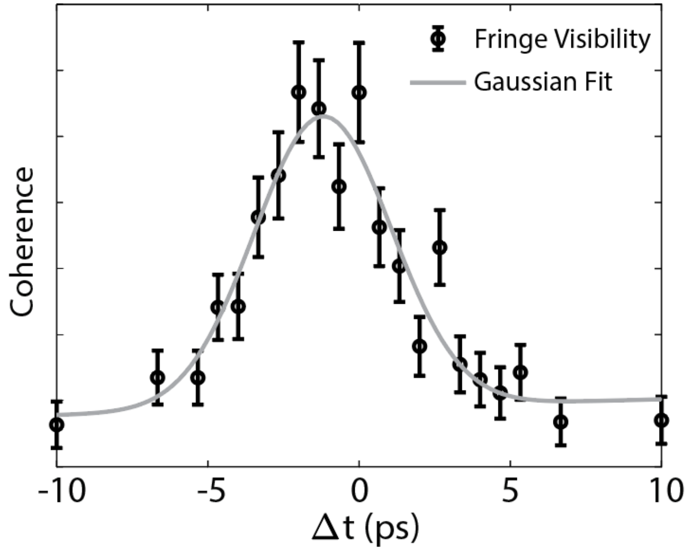
BIC- Exciton-Polariton condensate: coherence time



$$\frac{dn_{LP}}{dt} = W_{ep} n_R n_{LP} + \beta \frac{n_R}{\tau_R} - \boxed{\frac{n_{LP}}{\tau_{LP}}}$$







~5 ps coherence time:

$$Q = 2\pi\,\tau\,\nu_0 \sim 15000$$

Acknowledgements

TU/e

DIFFER
Dutch Institute for
Fundamental Energy Research

PHILIPS

FOM Institute
AMOLF

KYOTO UNIVERSITY
KYOTO JAPAN
FOUNDED 1897

SCIL
Nanoimprint solutions

UAM
Universidad Autónoma
de Madrid

CSIC
CONSEJO SUPERIOR DE INVESTIGACIONES CIENTÍFICAS

- Matthijs Berghuis
- Gabriel Castellanos
- Mohammad Ramezani
- Saad Abdelkhalik
- Aleks Vaskin
- Alexei Halpin
- Said Rodriguez
- Gabriel Lozano

- Shunsuke Murai
- Marc Verschuuren
- Thibault Peyronel
- Tobias G. Tiecke

- Jose A. Sanchez-Gil
- Jose Luis Pura
- Diego Abujetas
- Francisco Garcia-Vidal
- Antonio Fernandez-Dominguez
- Johannes Feist

Conclusion

Nanoparticle arrays (metallic & dielectric) supporting surface lattice resonances offer a versatile platform for several applications:
SSL, optical communication, exciton-polariton condensation