LPENS
LABORATOIRE DE PHYSIQUE
DE L'ÉCOLE NORMALE SUPÉRIEURE

Room temperature polaritonics in all-inorganic cesium lead halide perovskite

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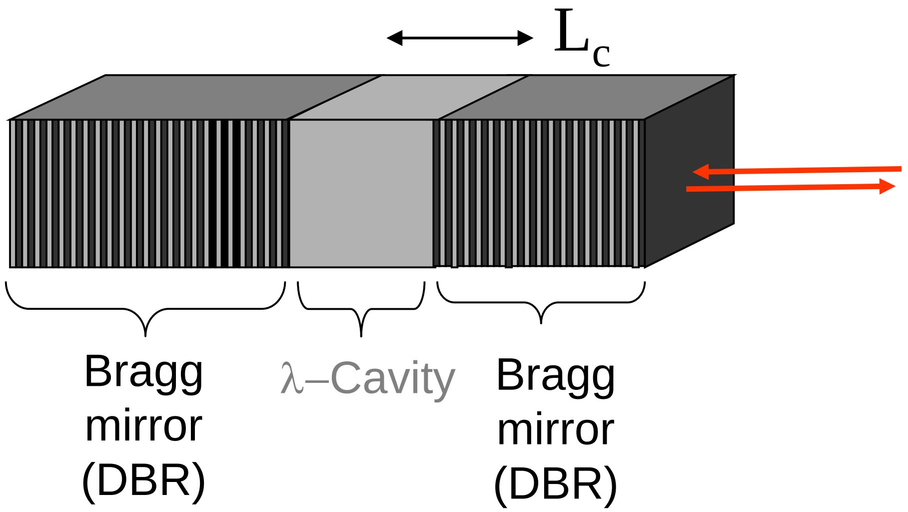
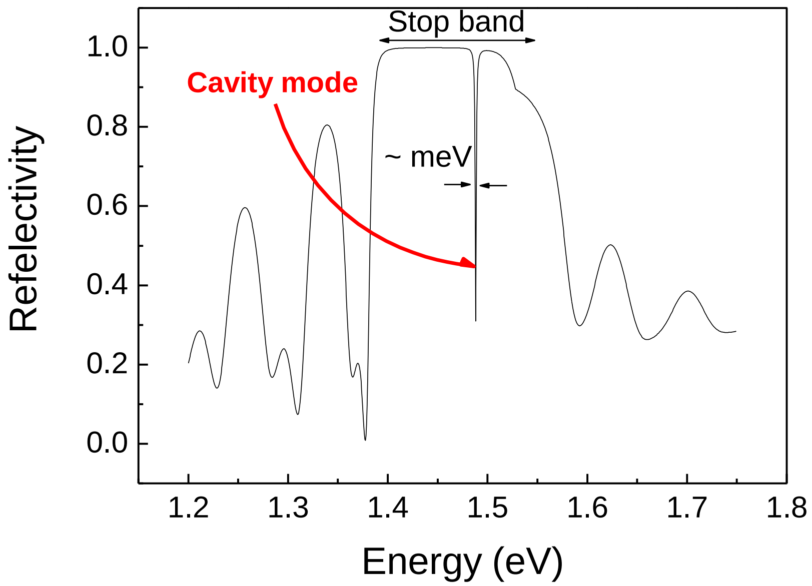
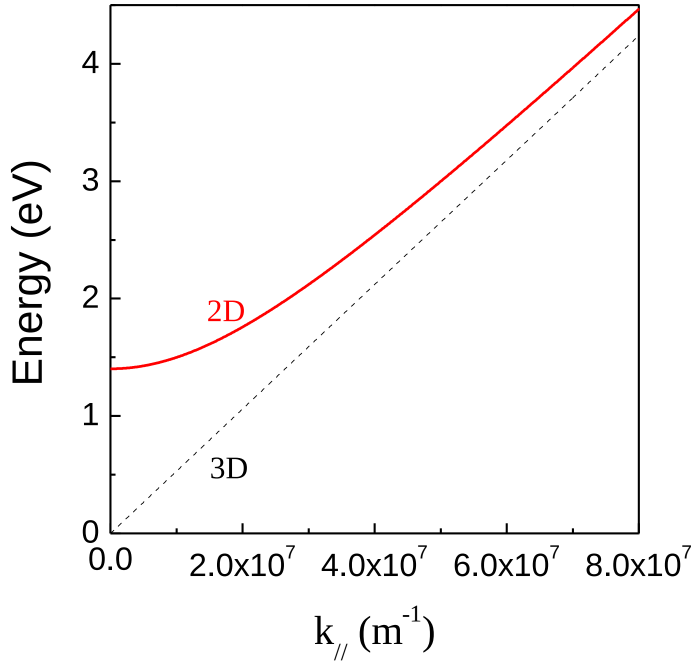
Light-Matter strong coupling regime in semiconductor microcavities

Mixed light-matter quasi-particles : exciton-polariton

A Fabry-Pérot microcavity → confined photons







Photons confined in an optical cavity:

- **Very light**
- Very fast
- No interaction

$$E_C(k) = \frac{\hbar c}{n_c}\sqrt{k_\parallel^2 + \left(\frac{p\pi}{L_c}\right)^2} \longrightarrow E_C(k) = E_C(0) + \frac{\hbar^2 k^2}{2M_{phot}}$$

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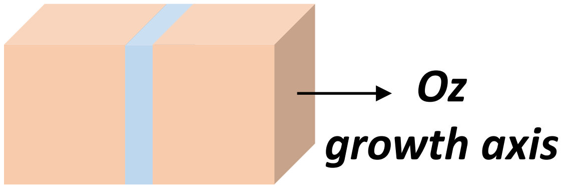
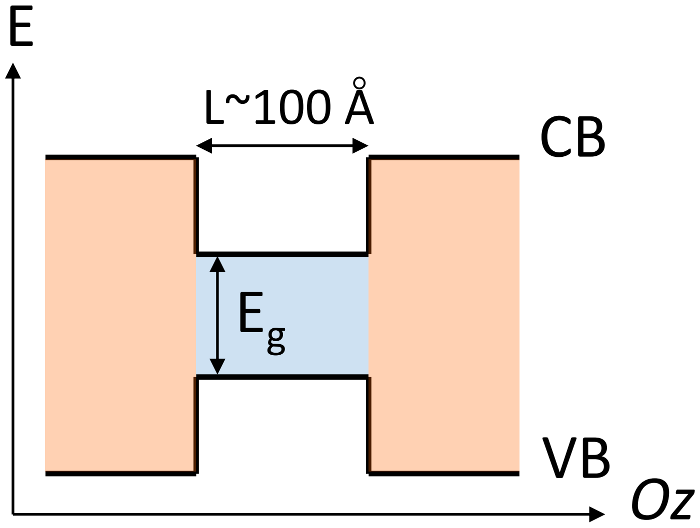
Mixed light-matter quasi-particles : exciton-polariton

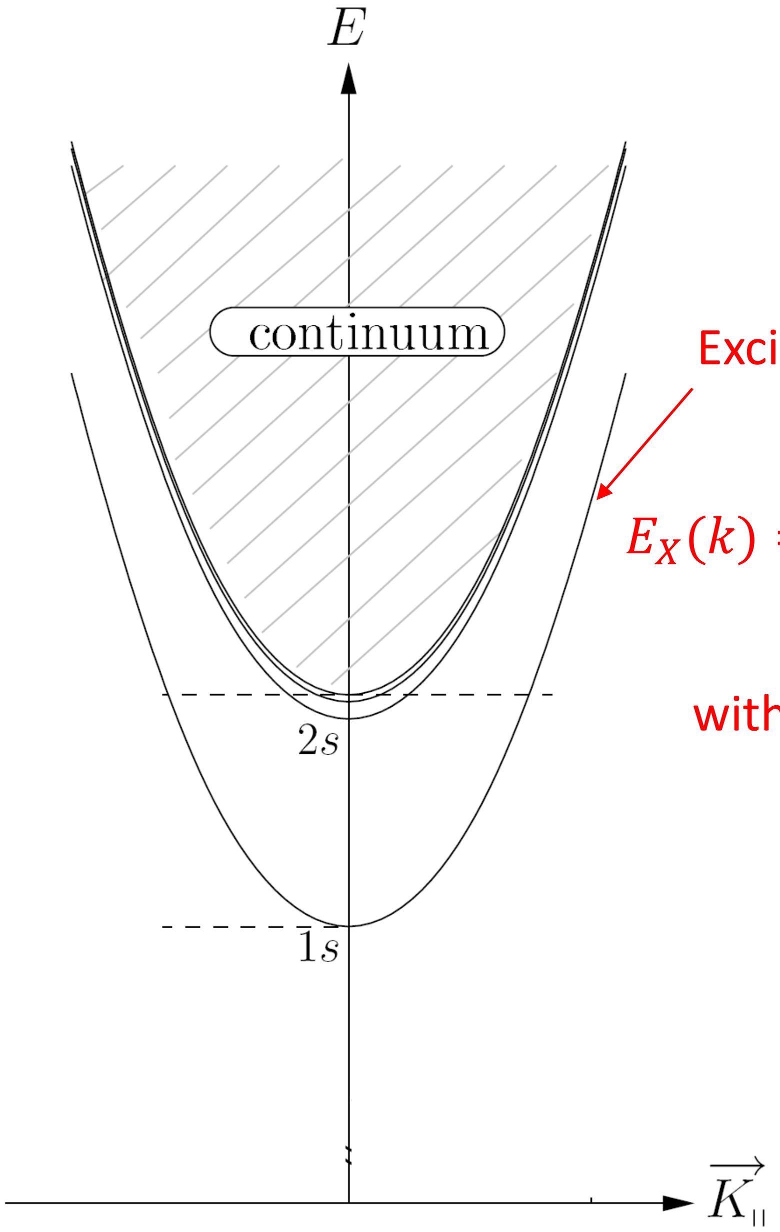
An active medium - quantum well → confined excitons





Excitons confined in a quantum well:

- Very heavy
- Very slow
- **Interaction**



Exciton 1s

$$E_X(k) = E_X(0) + \frac{\hbar^2 k^2}{2M_X}$$

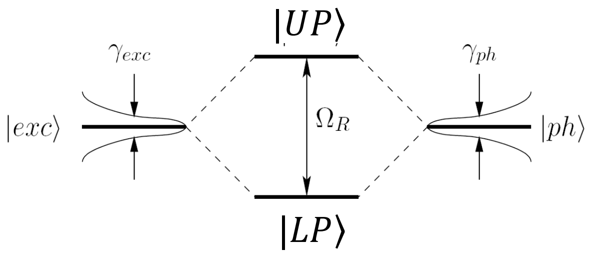
with $\frac{M_X}{M_{phot}} \sim 10^4$

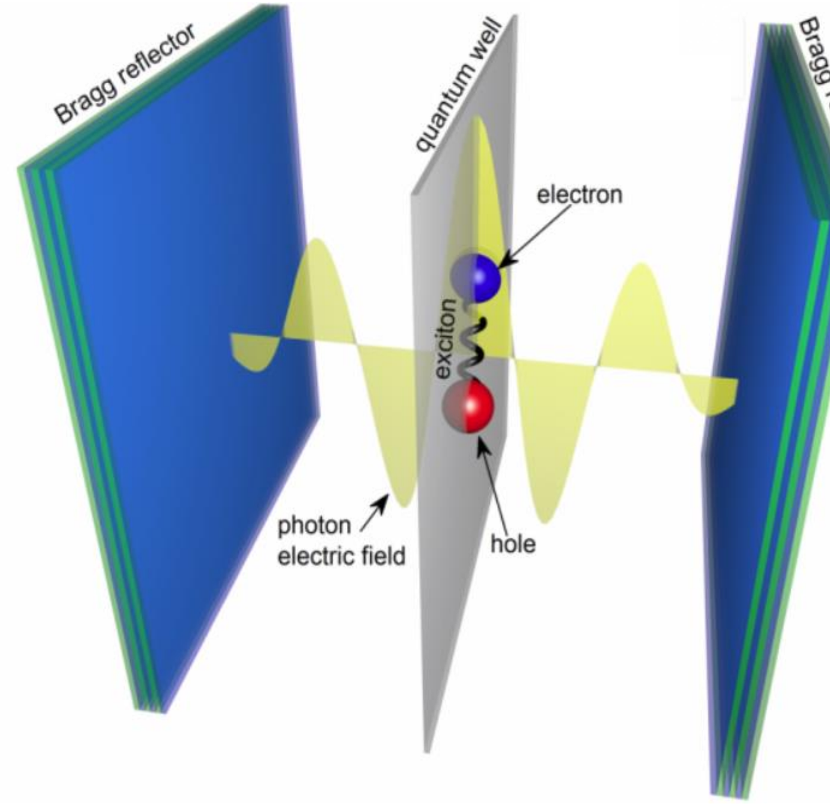
Light-Matter strong coupling regime in semiconductor microcavities

Mixed light-matter quasi-particles : exciton-polariton

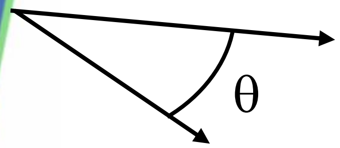
Confined excitons coupled to confined photons → **polaritons**

First demo:
C. Weisbuch *et al.*
PRL **69**, 3314 (1992)



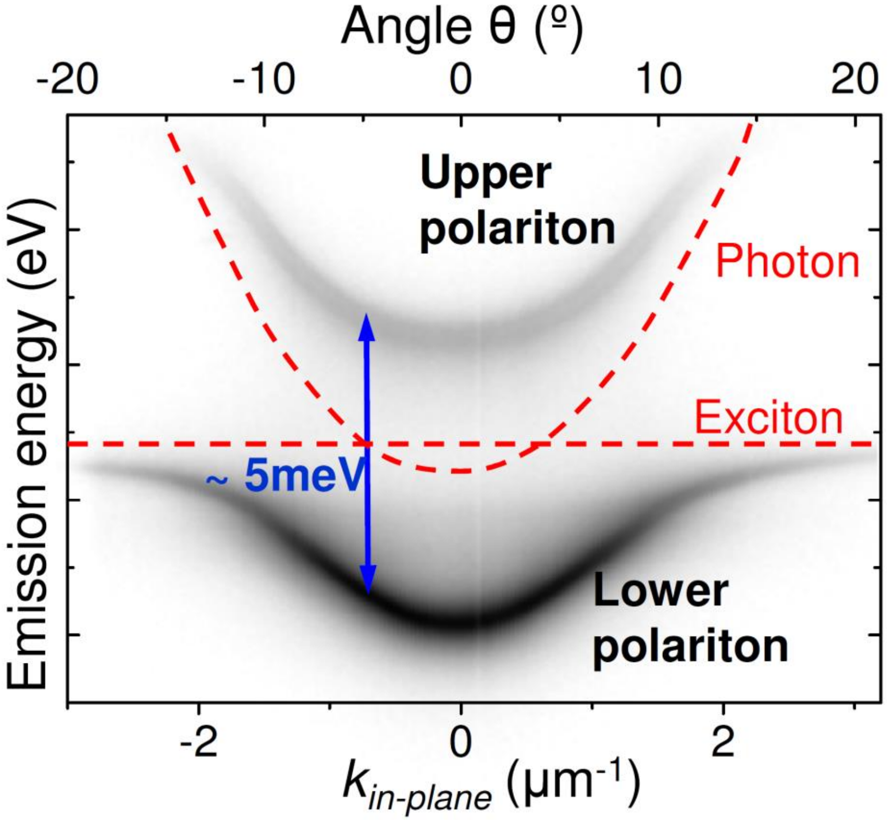


M. Sich *et al.*, C. R. Phys **17**, 908 (2016)



Imaging the Fourier space

$$k_{\parallel} = \frac{\omega}{c}\sin\theta$$

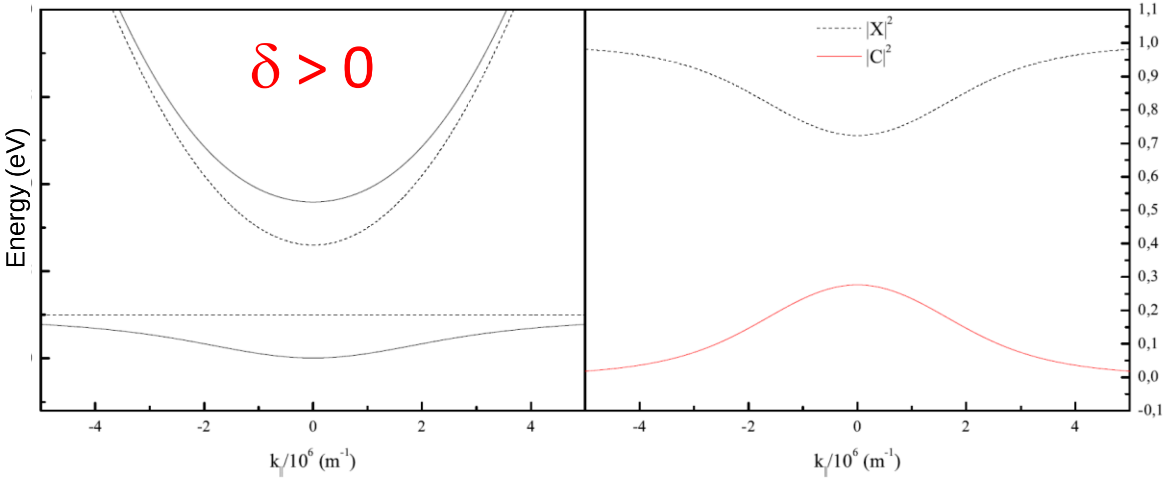


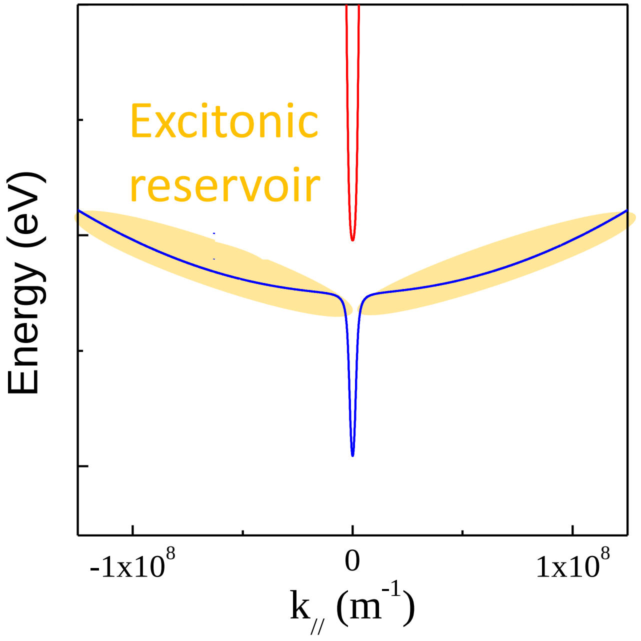
A. Amo *et al.*, Nature **457**, 291 (2009)

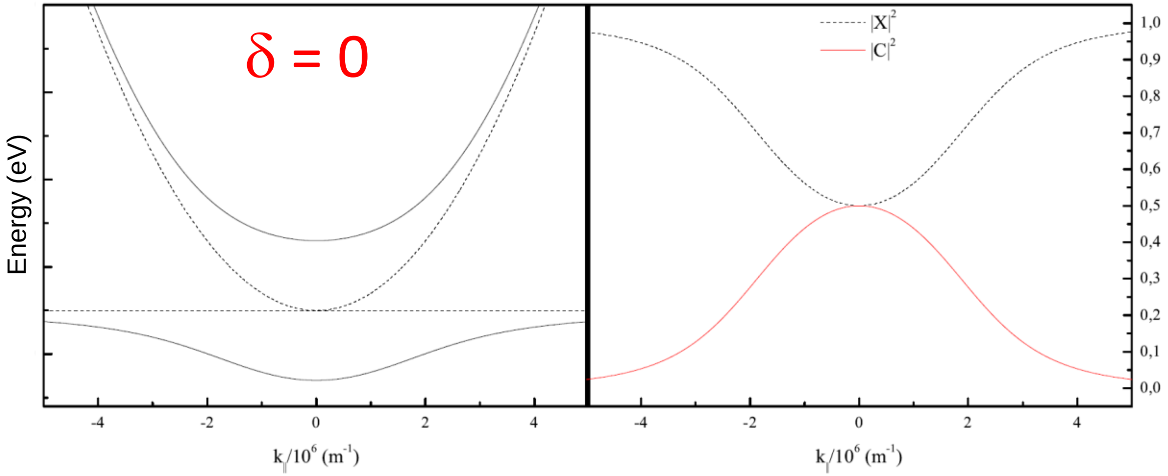
Light-Matter strong coupling regime in semiconductor microcavities

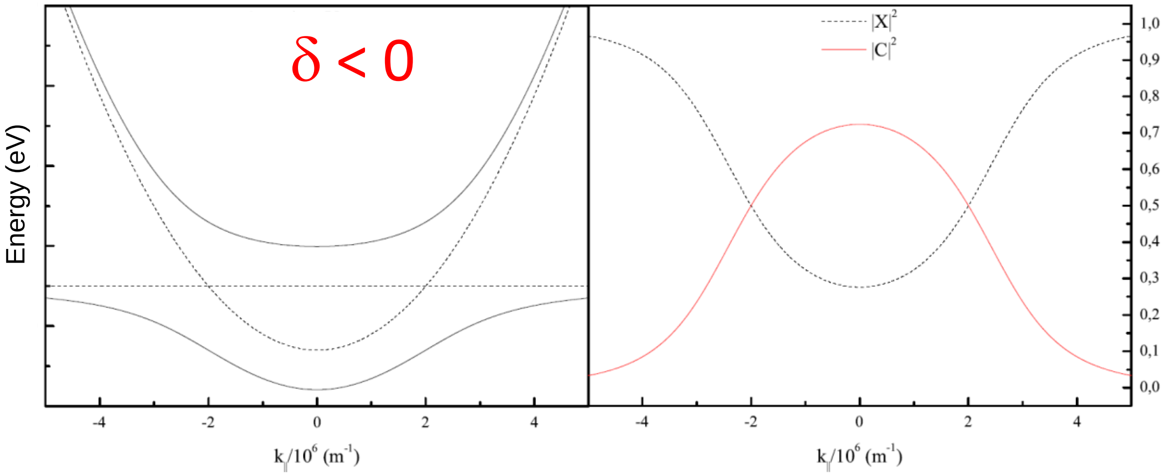
Mixed light-matter quasi-particles : exciton-polariton

S-shaped polariton dispersion





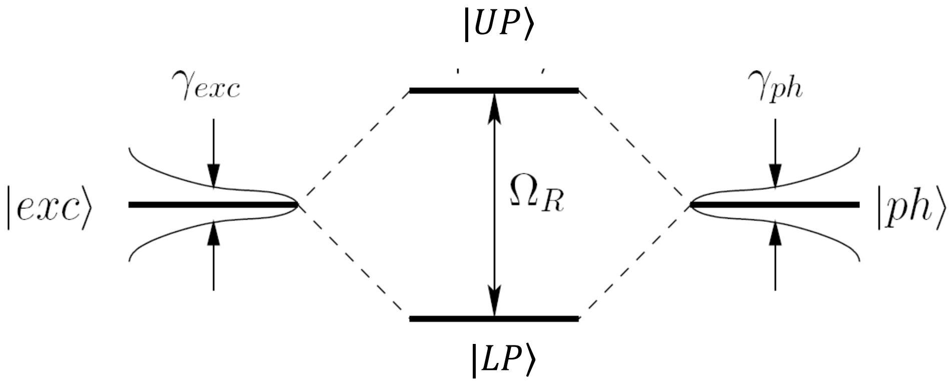




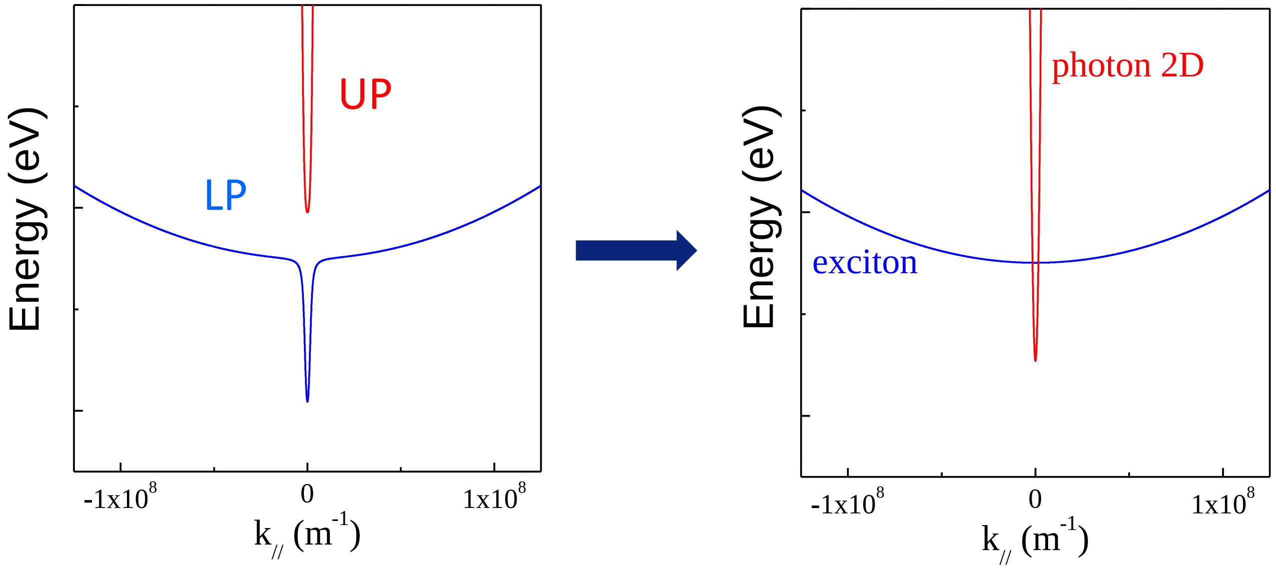
Light-Matter strong coupling regime in semiconductor microcavities

Mixed light-matter quasi-particles : exciton-polariton

- Composite bosons
- Excitonic components → **Strong interactions**
- Photonic component → **Low mass**
- Short lifetime (few ps) → Coupling to free space



Strong to weak coupling regime



- Low-cavity finesse
- Phonons interactions → **Low temperature**
- Coulomb interactions, many body effects (collisional broadening) → **Low optical densities**

Exciton-polariton condensation

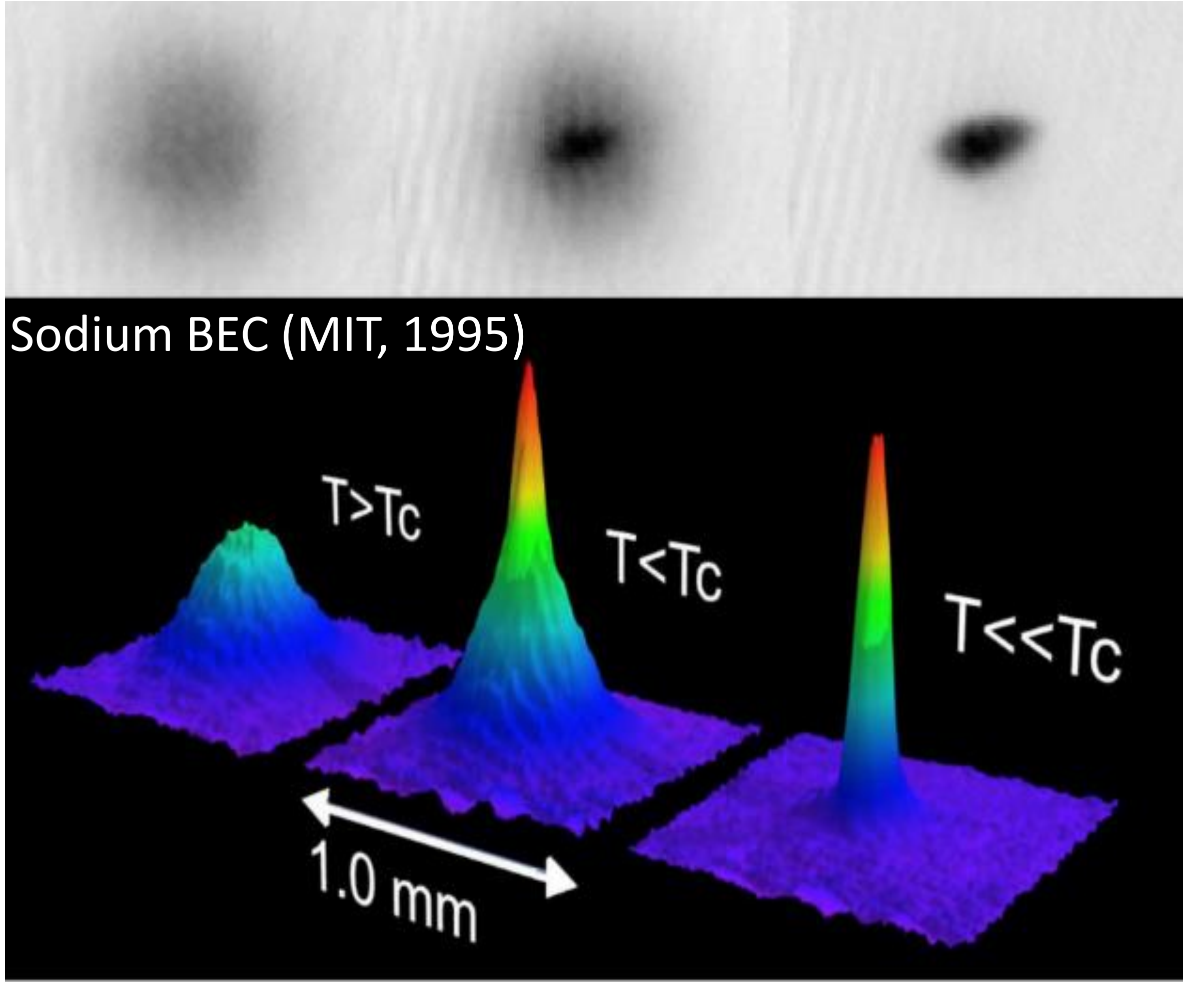
At the heart of polaritonics applications

Bose-Einstein Condensation in atomic physics (Nobel Prize 2001) :

→ A group of atoms cooled to temperatures close to absolute zero (~ 100 nK)

→ A large fraction of bosons occupy a single quantum state

→ Coherence properties (temporal and spatial)



Sodium BEC (MIT, 1995)

$$T_c = \left(\frac{n}{\zeta(3/2)}\right)^{2/3} \frac{2\pi\hbar^2}{mk_B} \approx 3.3125\,\frac{\hbar^2 n^{2/3}}{mk_B}$$

What about exciton-polariton ?

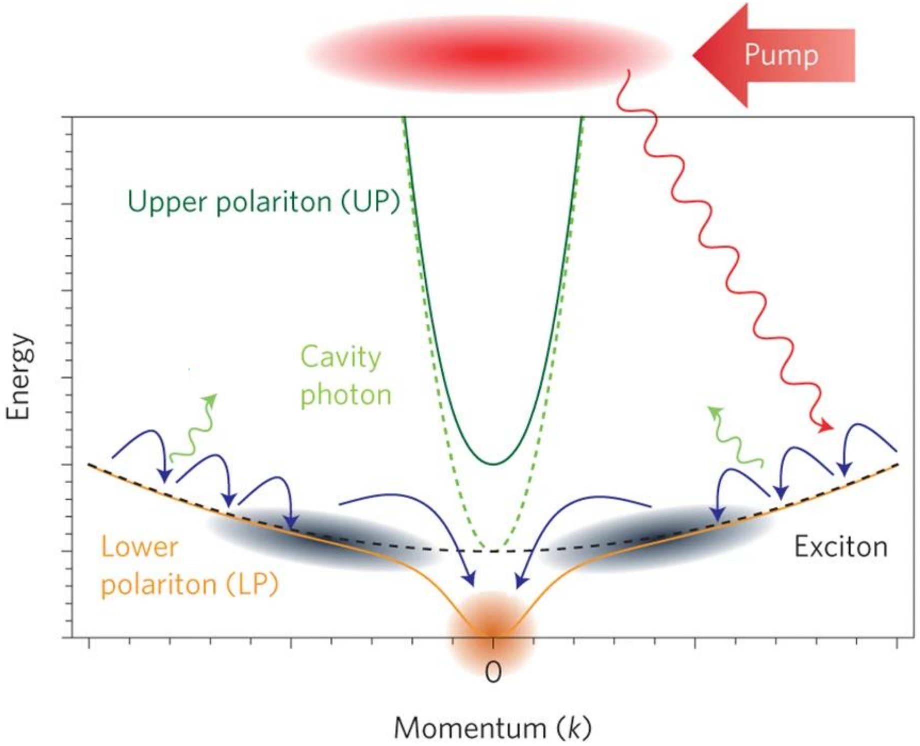
→ Key parameter: low effective mass polariton (10^{-8} m_{at}) → $T_c^{pol} \propto \frac{\hbar^2 n^{2/3}}{m_{pol}k_B} \propto 10^8 T_c^{at} \propto 10K$

→ Polariton-Polariton interactions

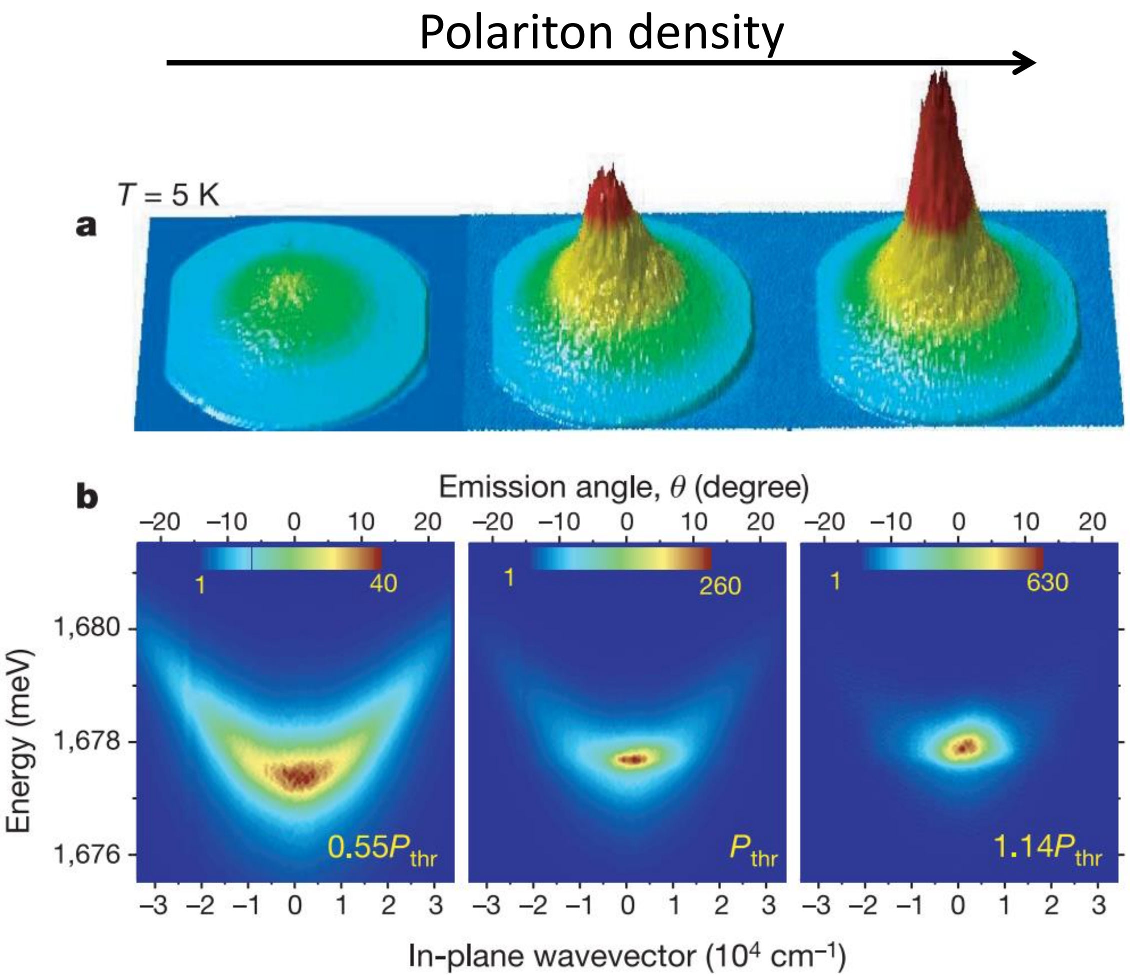
Exciton-polariton condensation

At the heart of polaritonics applications

First demo of polariton condensate *at non-thermal equilibrium*:
J. Kasprzak *et al.*, Nature **443**, 409 (2006)



T. Byrnes *et al.*, Nature Physics **10**, 803 (2014)



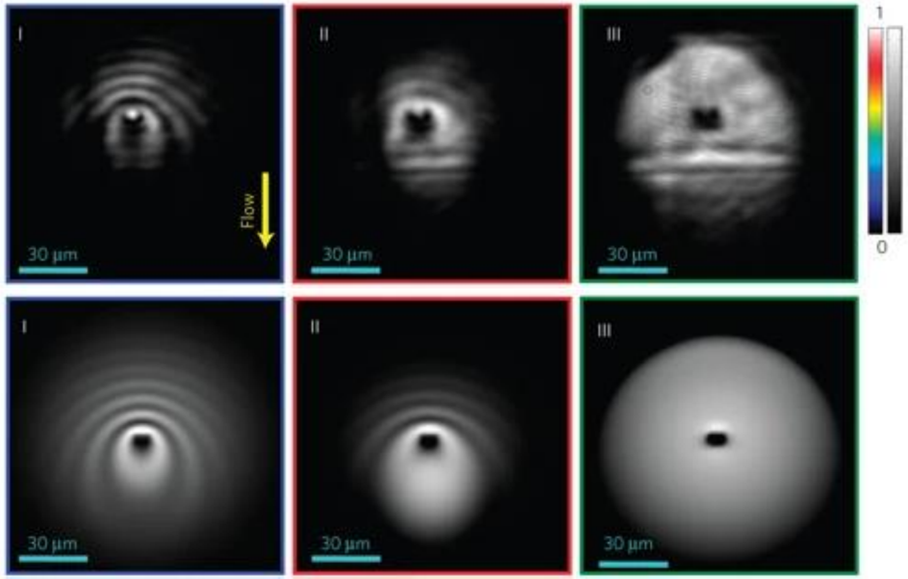
Non-resonant pumping (optical or electrical)
→ Polariton scattering to the excitonic reservoir
→ Polariton – Phonon interactions
→ Polariton – Polariton interactions ("magic angle")
→ Macroscopic occupation of the LP branch at k=0

Exciton-polariton condensation

At the heart of polaritonics applications

Solid state platform to study the physics of BEC

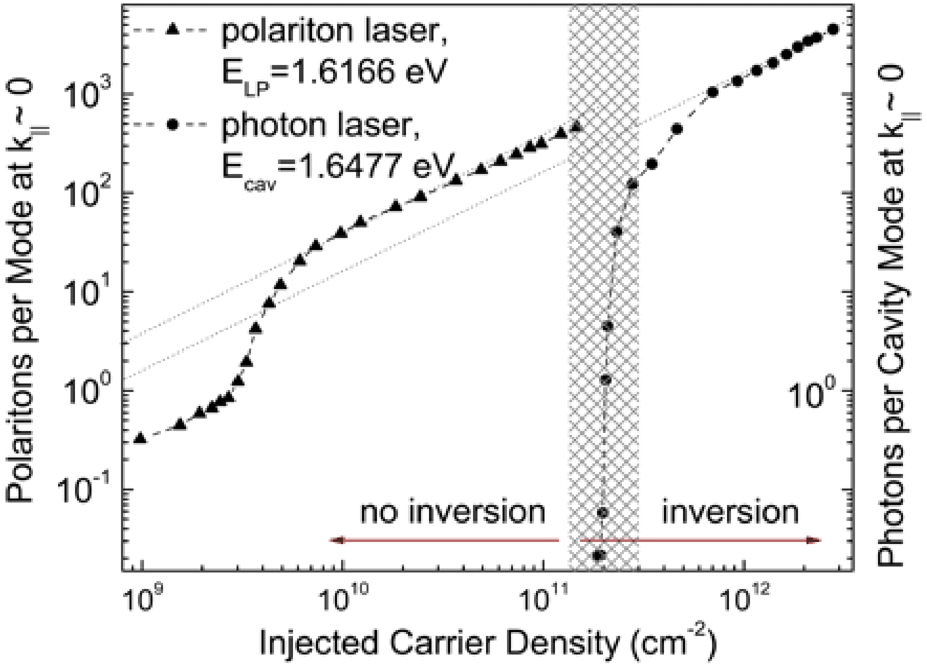
- Superfluidity
- Vortices
- Quantum fluid of light



A. Amo *et al.*, Nature Physics **5**, 805 (2009)

Low-threshold polariton laser

- Analogy with VCSELs (QW in a µcavity)
- Short polariton lifetime (~ps)
- Out of equilibrium BEC
- Coherent emission in strong coupling regime, without population inversion



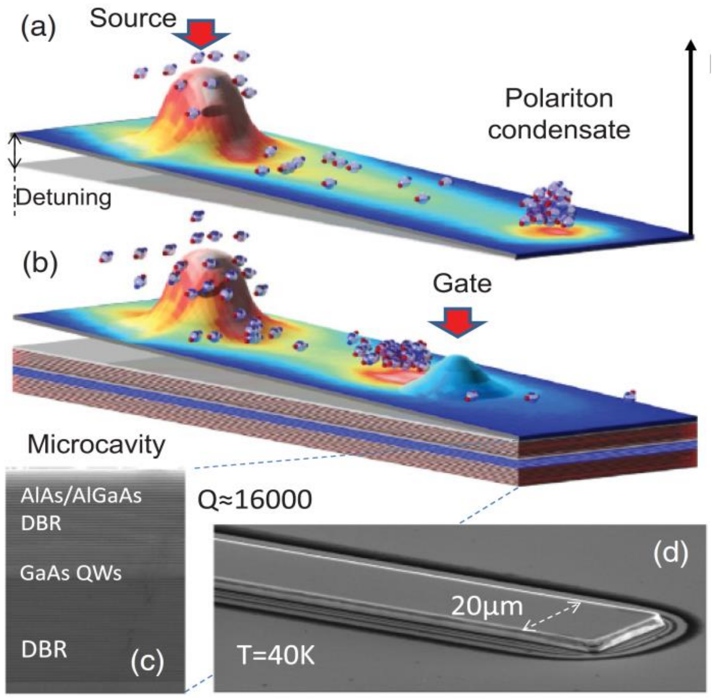
H. Deng *et al.*, PNAS **100**, 15318 (2003)

Exciton-polariton condensation

At the heart of polaritonics applications

Exciton-polariton circuits

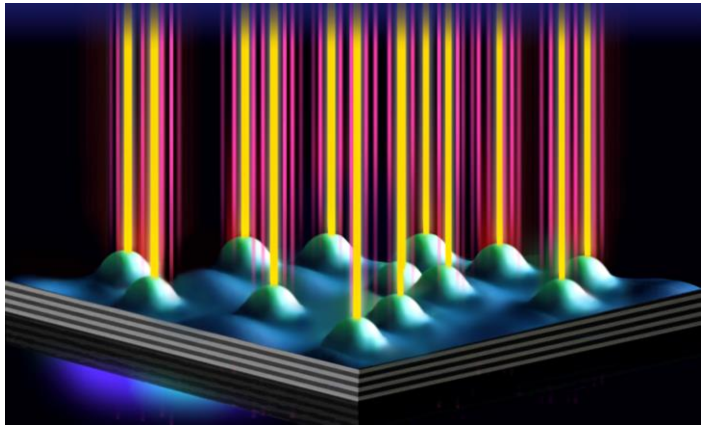
- Propagation of polariton condensates
- All-optical information processing elements



T. Gao *et al.*, PRB **85**, 235102 (2012)

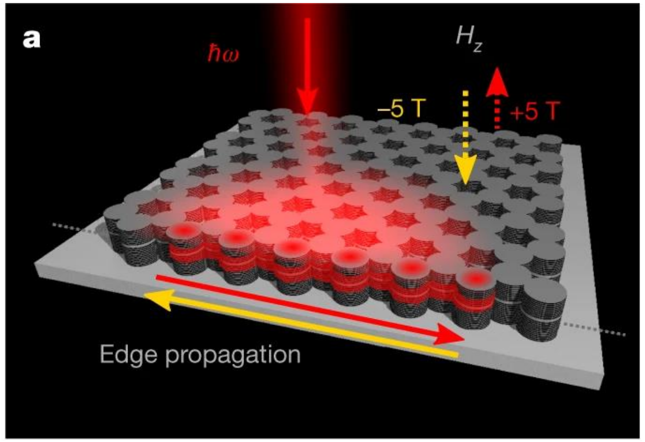
Exciton-polariton condensates in lattices

- Quantum simulators



Credits to N. Berloff (Univ. of Cambridge)

- Topological insulators

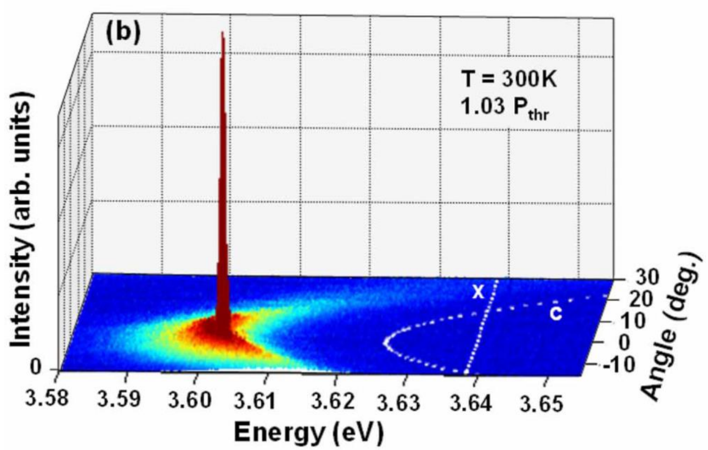


S. Klembt *et al.*, Nature **562**, 552 (2018)

Polariton condensation and polariton lasing at room temperature

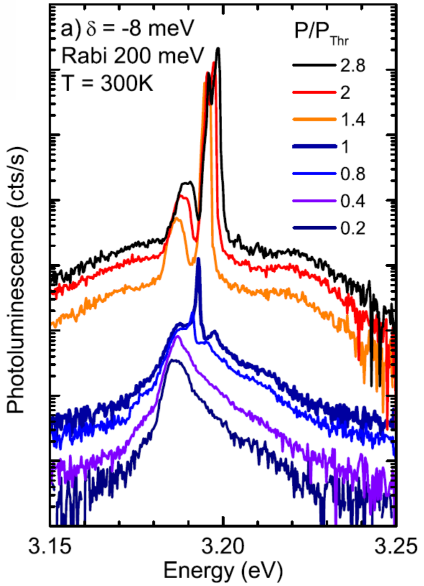
Inorganic wide bandgap semiconductors

- Wannier-Mott excitons with large binding energies (25 - 100meV)
- Sophisticated epitaxial fabrication techniques



GaN QWs

G. Christmann *et al.*, APL **93**, 051102 (2008)

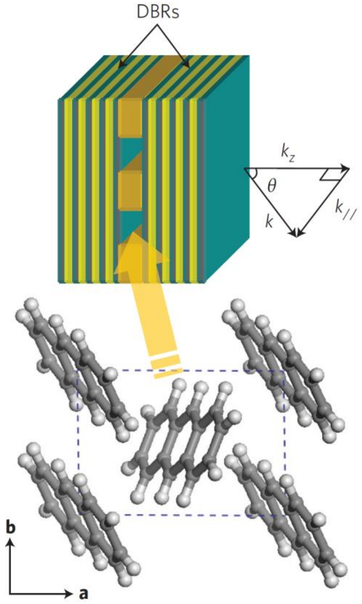


Bulk ZnO microcavity

Feng Li *et al.*, APL **102**, 191118 (2013)

Organic materials

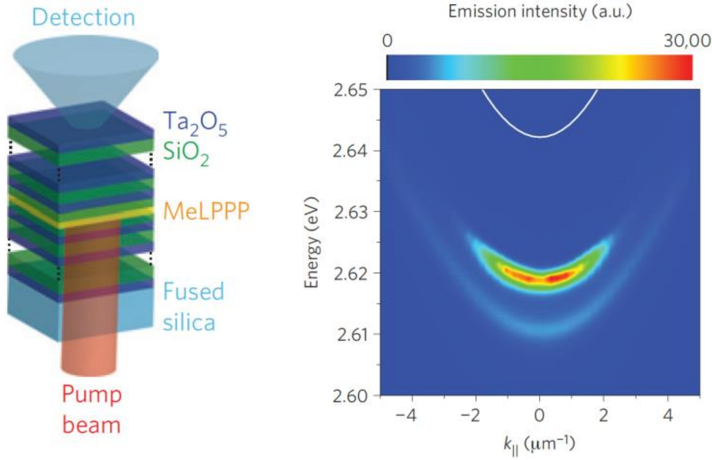
- Frenkel excitons: large exciton oscillator strengths and binding energies (0.2 – 1 eV)
- Higher thresholds due to weaker exciton interactions



Anthracene

S. Kéna-Cohen *et al.*, Nat. Photon. **4**, 371 (2010)

Polymer

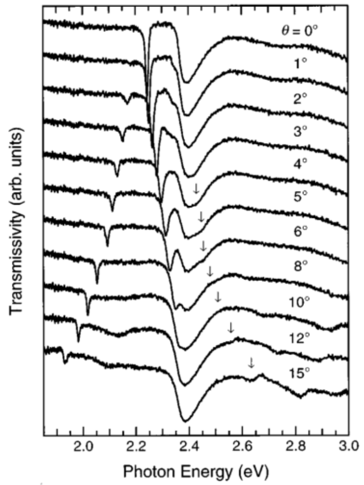


J. Plumhof *et al.*, Nat. Mat. **13**, 247 (2014)

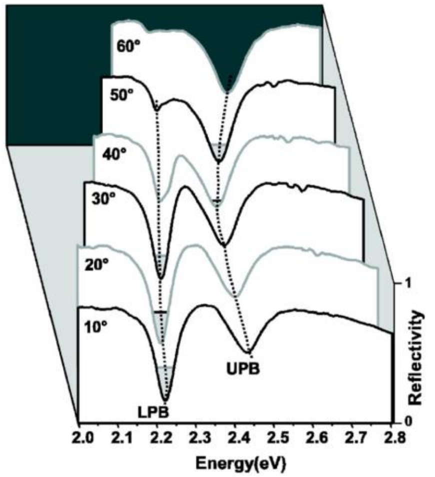
Hybrid organic-inorganic perovskite at room temperature

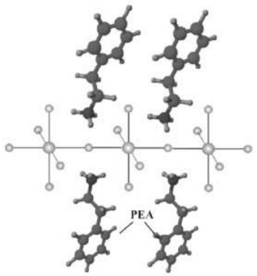
Strong coupling in layered 2D perovskite

T. Fujita *et al.*, PRB **57**, 7456 (1998)

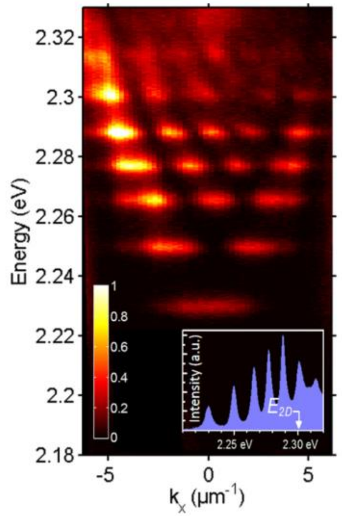


A. Brehier *et al.*, APL **89**, 171110 (2006)

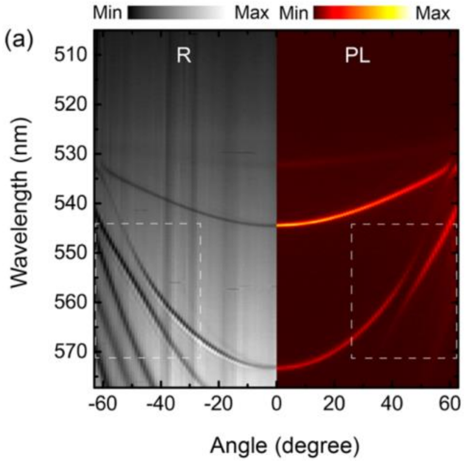




$(C_6H_5C_2H_4–NH_3)_2PbI_4$



H.S. Nguyen *et al.*, APL **104,** 081103 (2014)

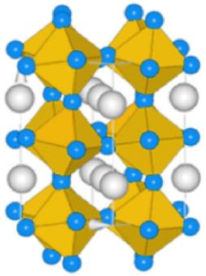


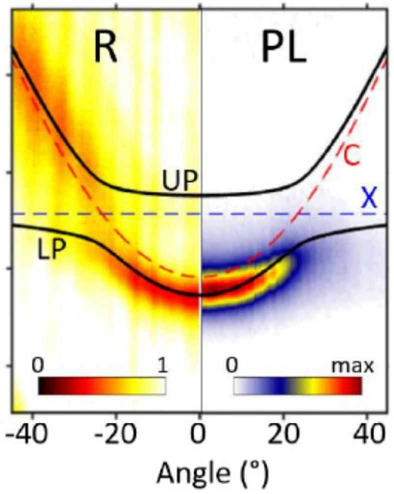
J. Wang *et al.*, ACS Nano **12**, 8382 (2018)

Strong coupling in 3D perovskite thin films

○ $CH_3NH_3^+$
● Pb
● Br

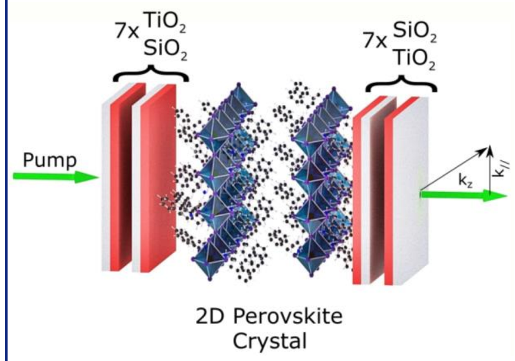
$(CH_3NH_3)_2PbBr_3$

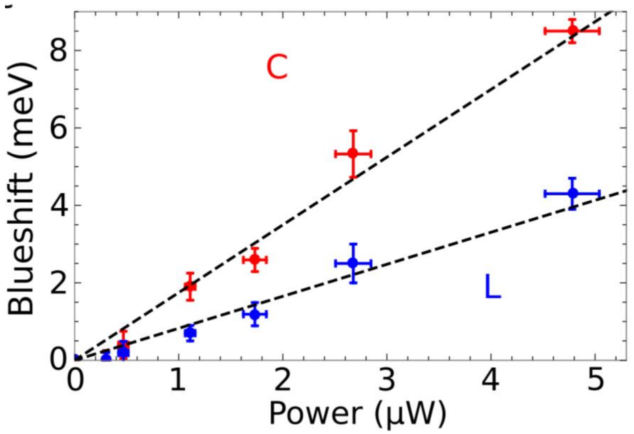




P. Bouteyre *et al.*, ACS Photonics **6**, 1804 (2019)

Polariton interactions in layered 2D perovskite



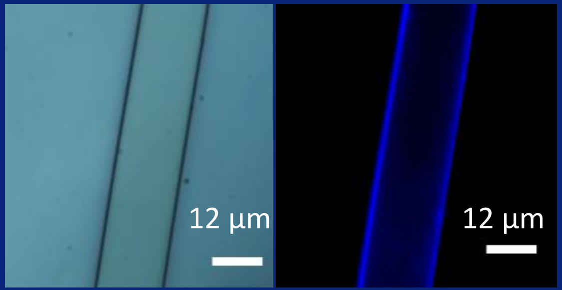


A. Fieramosca *et al.*, Sci. Adv. **5**, eaav 9967 (2019)

Experimental results in all-inorganic perovskite-based microcavities at room temperature

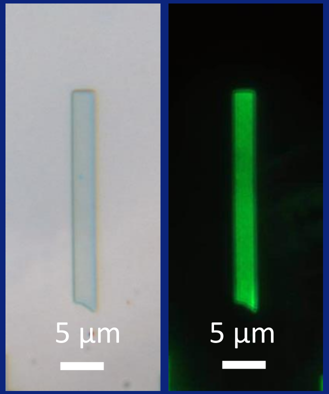
- Polariton condensation in $CsPbCl_3$ microplatelets

  R. Su *et al.*, Nano Letters **17**, 3982 (2017)



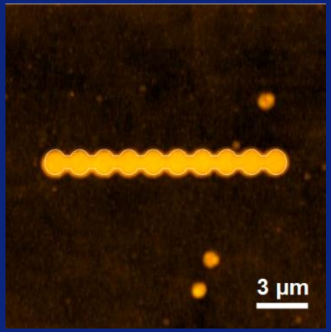
- Polariton condensate flow in $CsPbBr_3$ microwires

  R. Su *et al.*, Science Advances **4**, eaau0244 (2018)



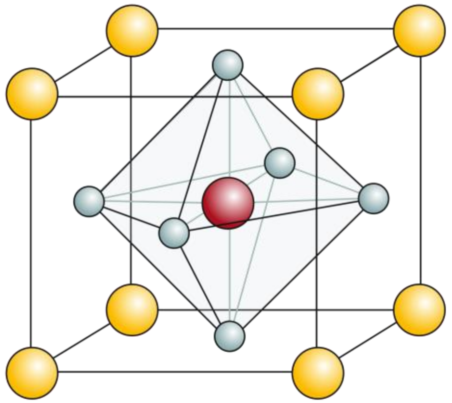
- Polariton condensation in a $CsPbBr_3$ lattice

  R. Su *et al.*, Nature Physics **16**, 301 (2020)



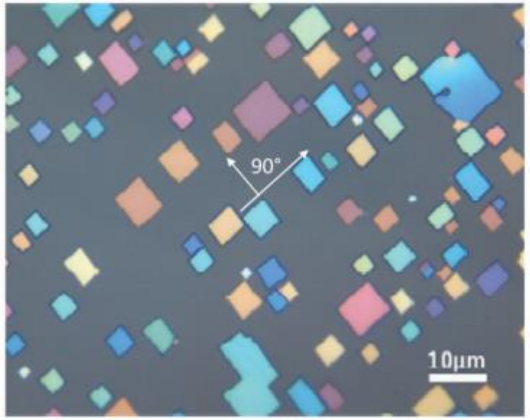
All-inorganic Cesium Lead Halide perovskite

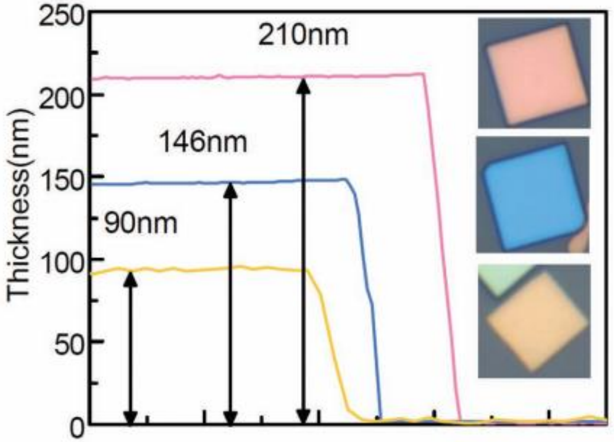
A new class of materials for photonics and polaritonics

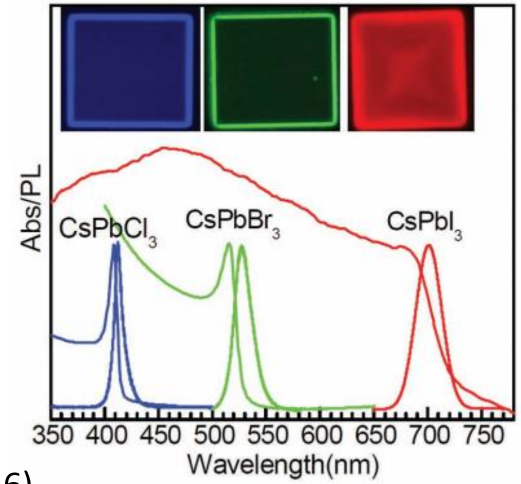


Cs Pb X_3

(X = Cl, Br, I or mixture)

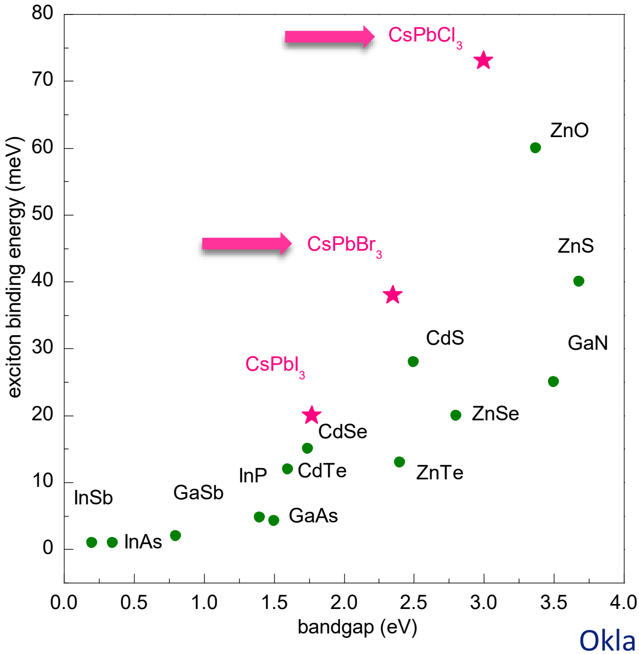


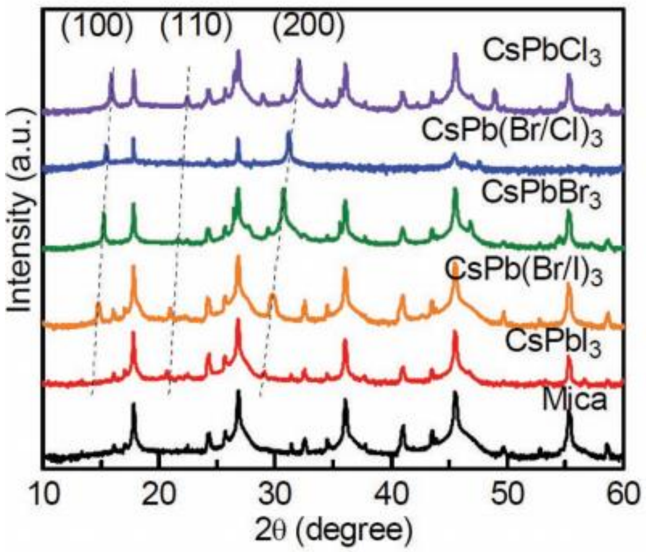




Q. Zhang *et al*., Adv. Funct. Mater. **26**, 6238 (2016)

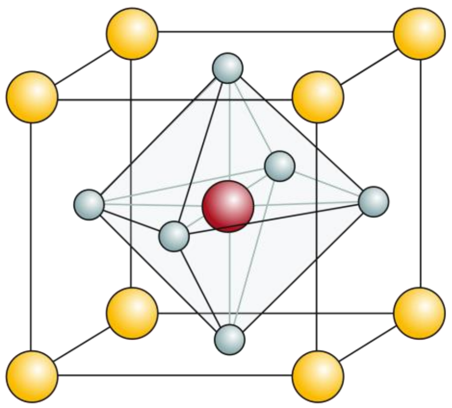
- Ease of platelets synthesis by CVD
- Direct bandgap semiconductors
- Wavelength tunability in the visible range
- Large exciton binding energies > k_BT
- High crystalline quality by CVD growth
- High PL quantum efficiencies (~70% @ RT)
- Better stability than hybrid perovskite





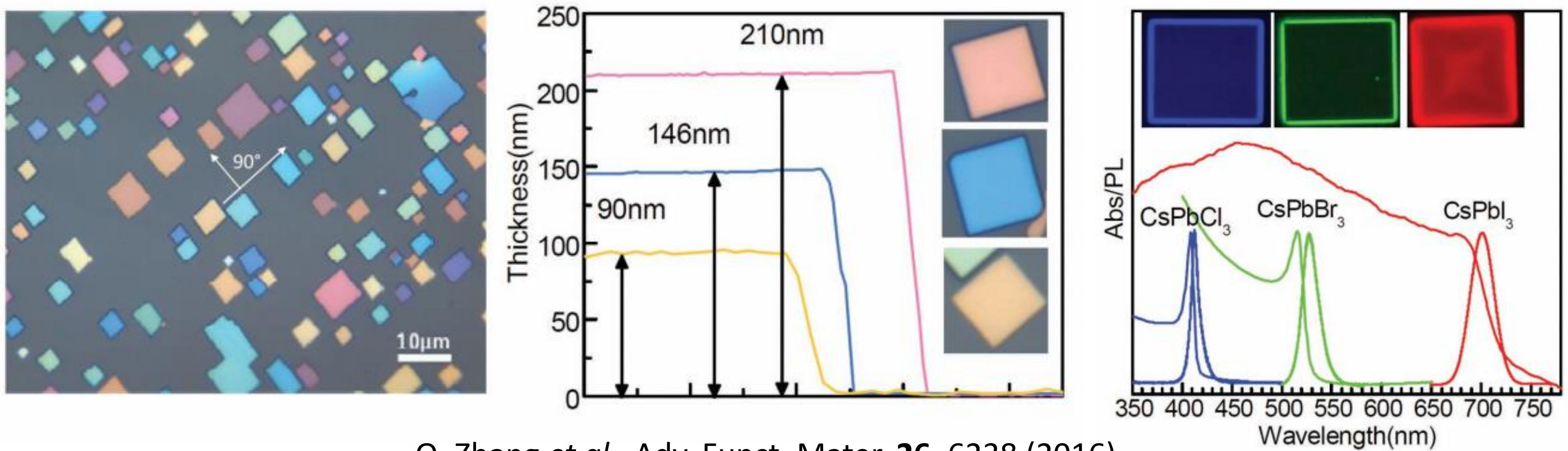
All-inorganic Cesium Lead Halide perovskite

Whispering Gallery Mode photonic lasing

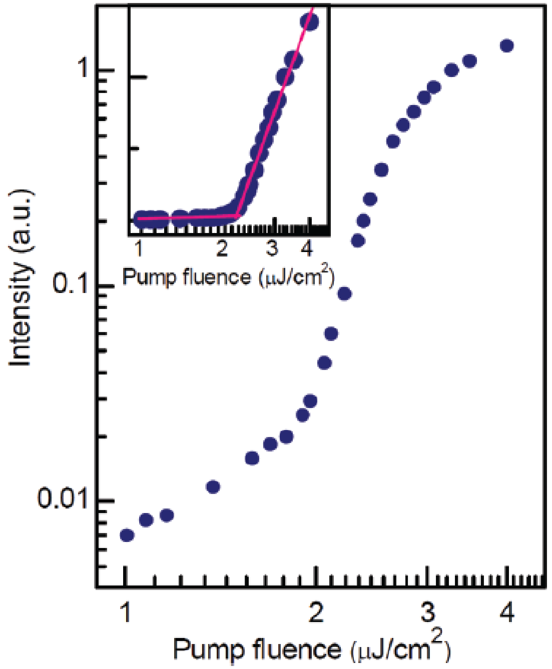


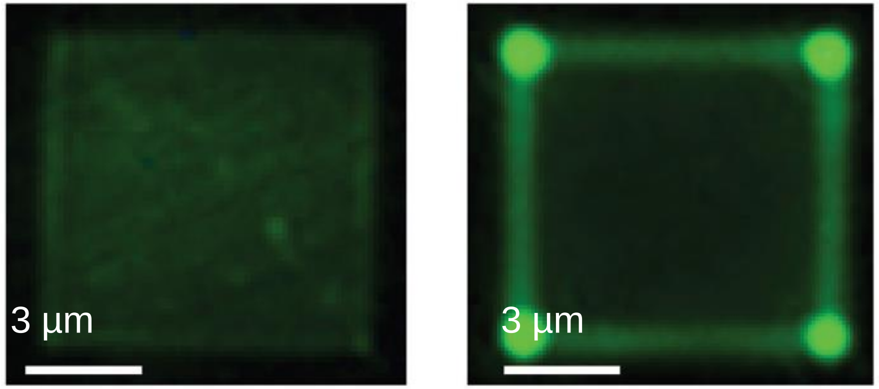
Cs Pb X_3

(X = Cl, Br, I or mixture)

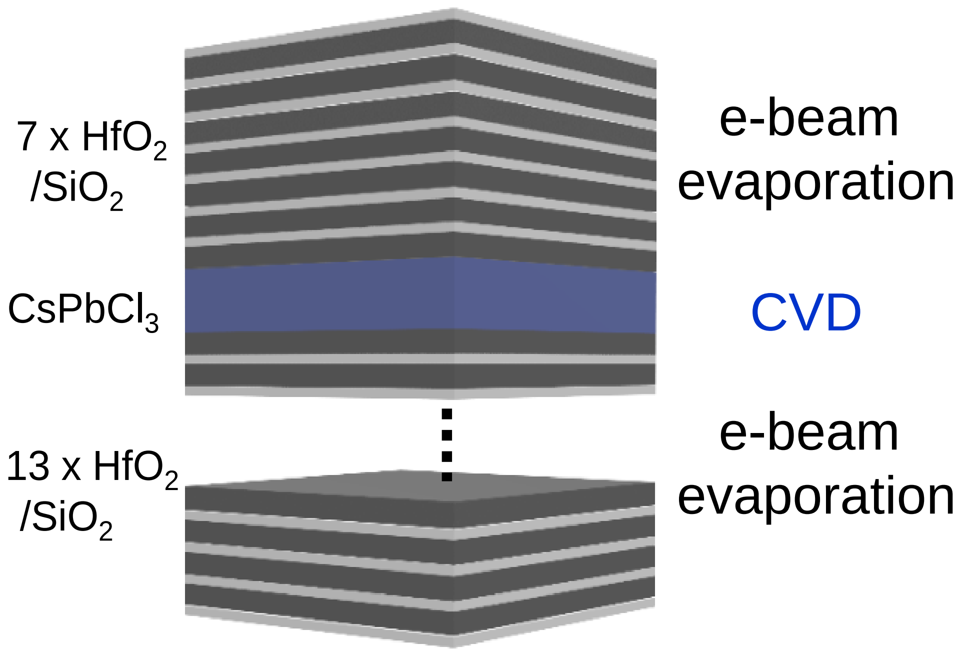


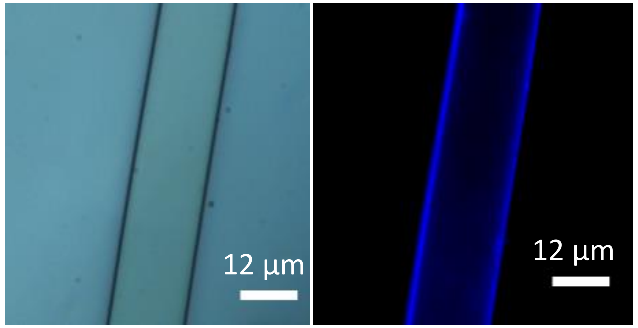
Q. Zhang *et al*., Adv. Funct. Mater. **26**, 6238 (2016)

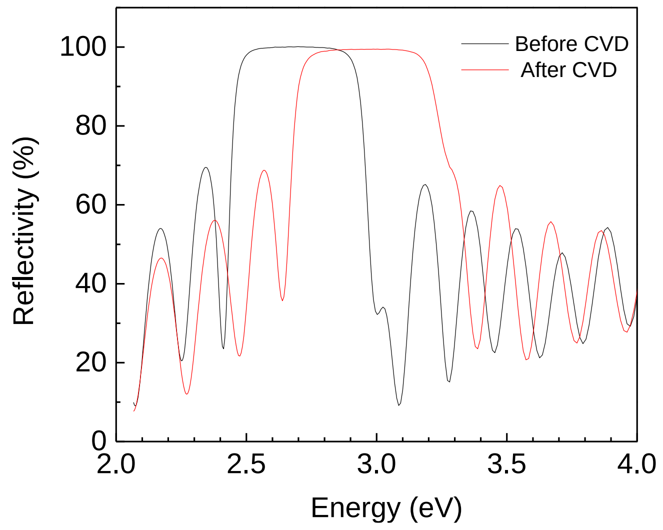


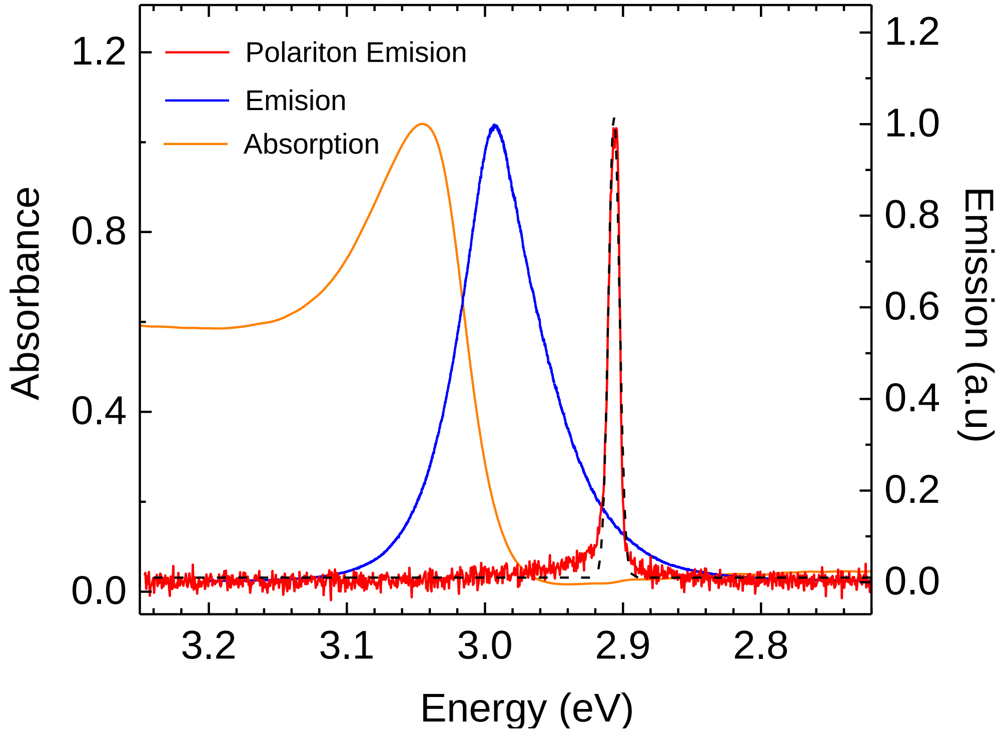


Perovskite-based microcavity





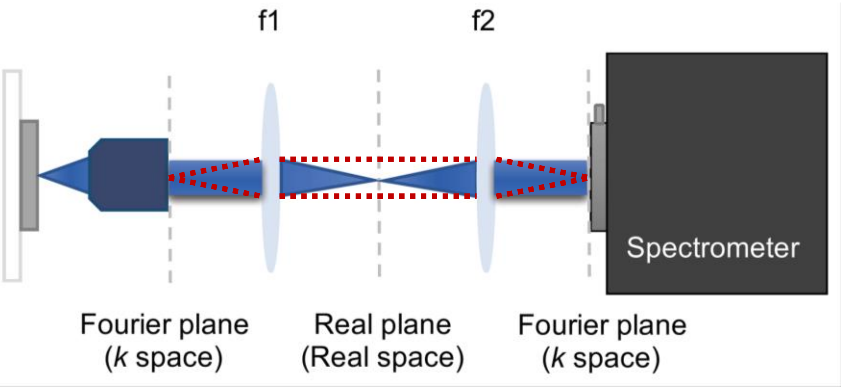


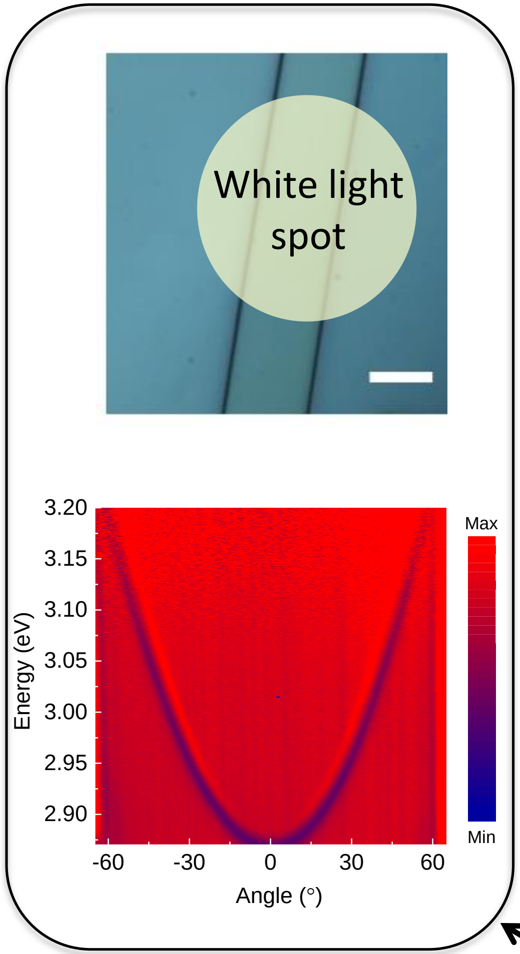


- Epitaxy-free fabrication techniques
- In-situ growth or dry transfer of perovskite on the bottom DBR
- Stop band (2.75 eV to 3.15 eV) with maximum reflectivity of 99.3% after CVD
- Various platelet thicknesses (~ 370 nm)
  → different detunings
- Quality factor Q ~ 300

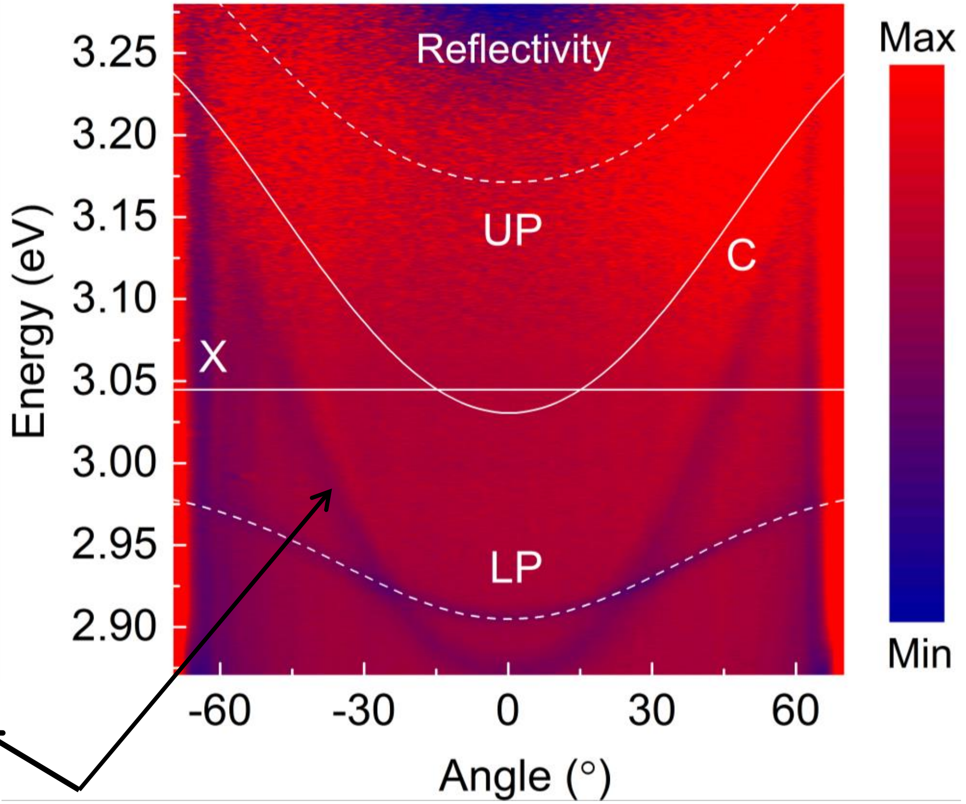
Room temperature exciton-photon strong coupling

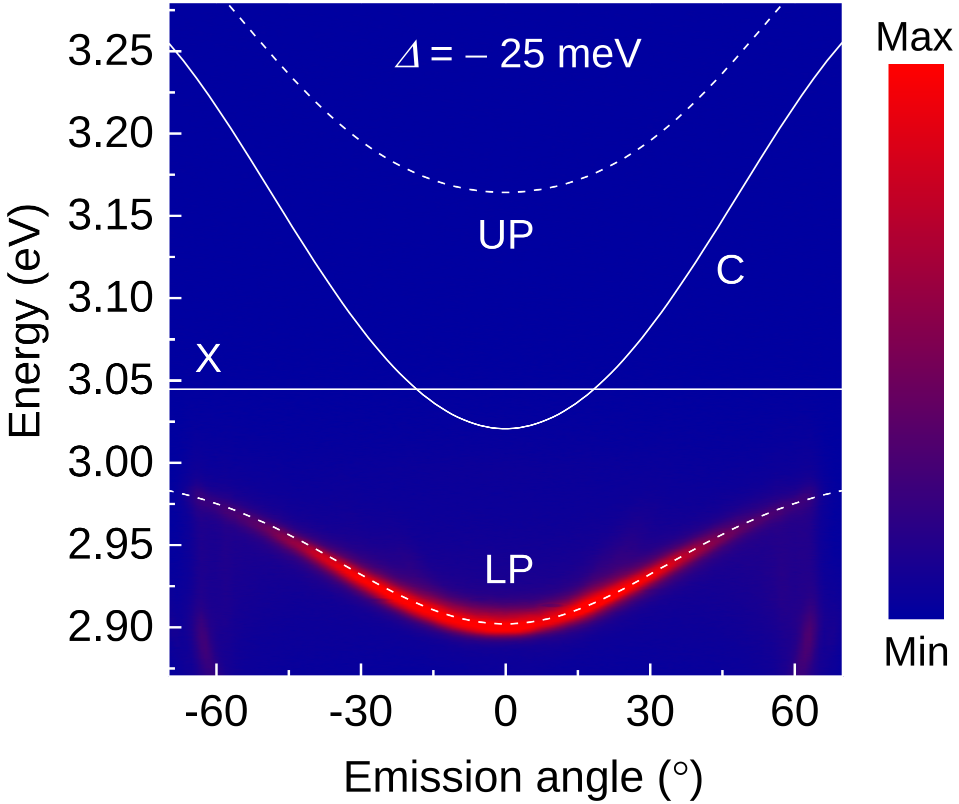
Angle-resolved spectroscopy
(image of the Fourier plane)





Rabi splitting ~ 265 meV

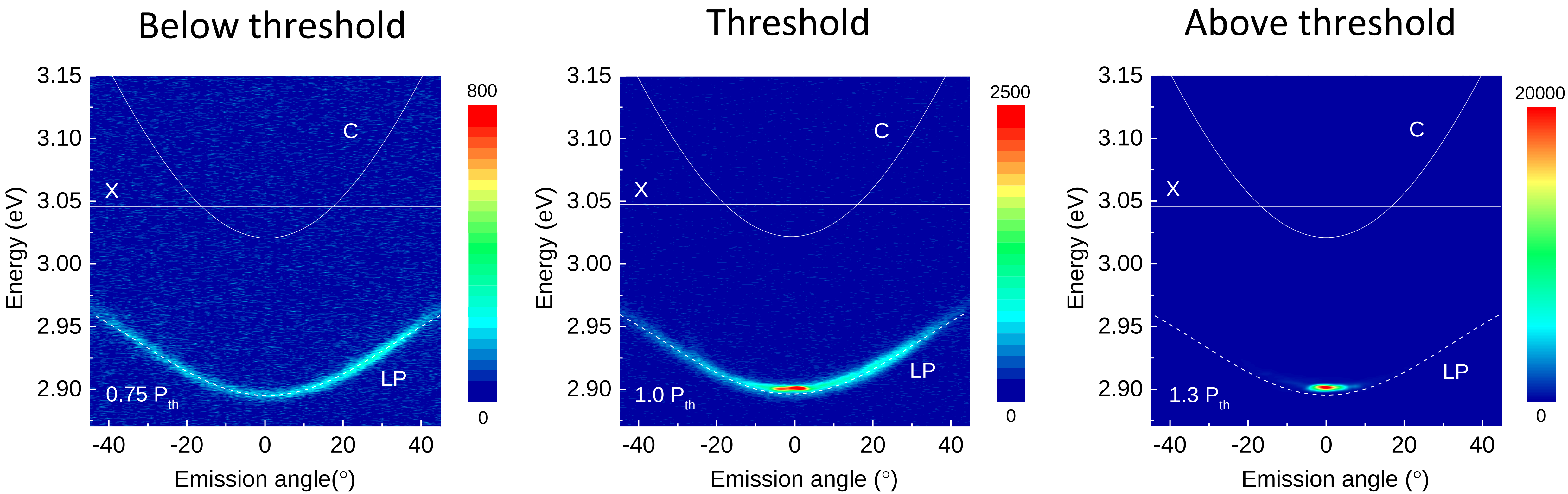




Bare cavity mode

Room temperature exciton-polariton condensation

- Negative detuning Δ = - 25 meV
- Pulsed excitation (100 fs @ 1 kHz)



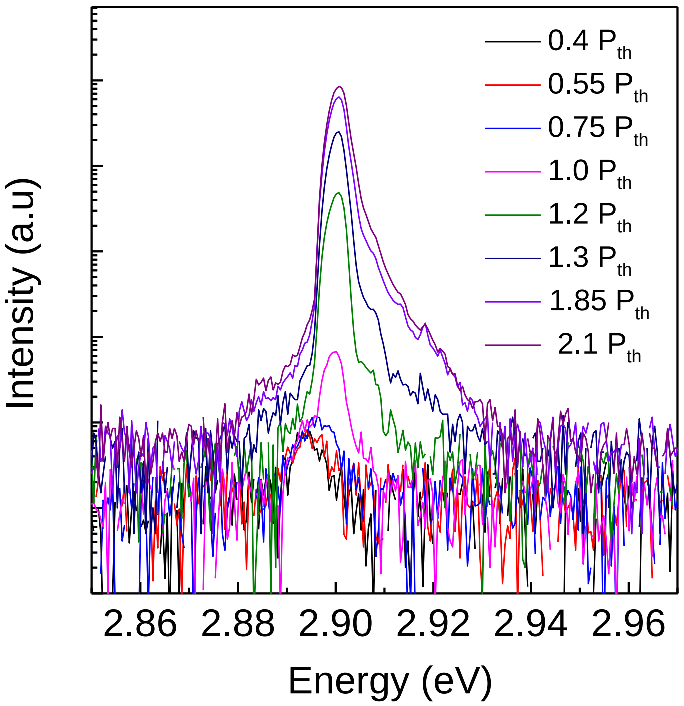
Below threshold

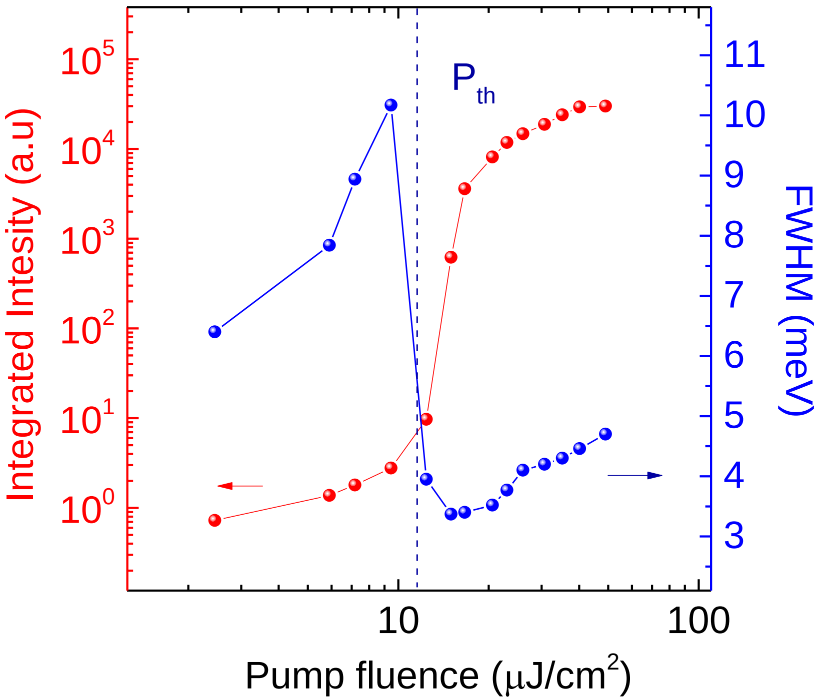
Threshold

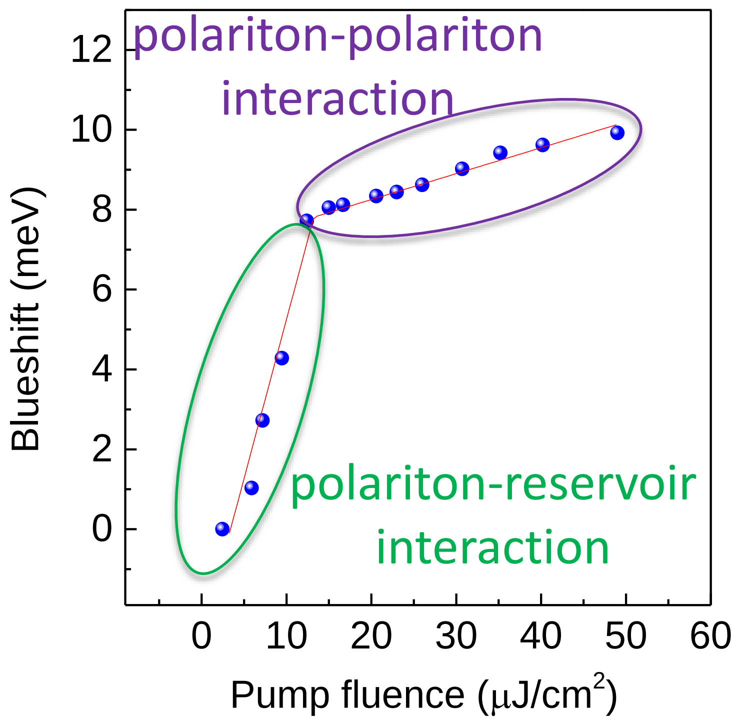
Above threshold

Macroscopic occupation of the LP ground state above a threshold

Polariton condensate & polariton lasing properties





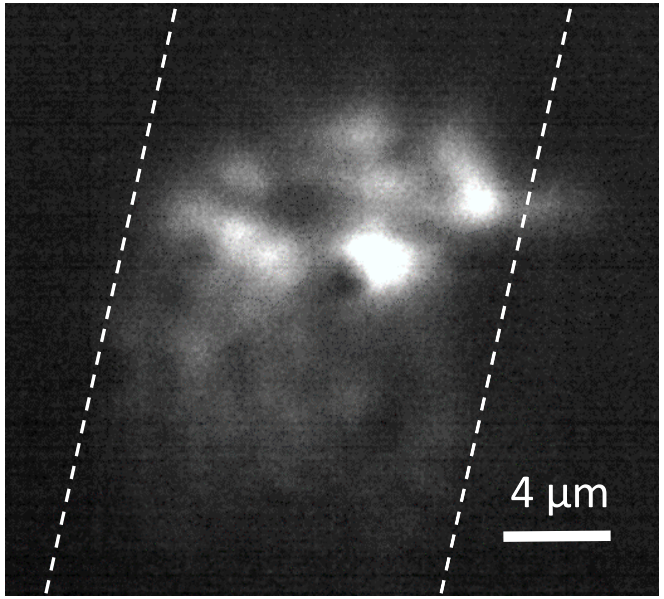


- Linewidth narrowing → Temporal coherence
- Blueshift of 10 meV << $\Delta E = E_C - E_{LP}$ = 120 meV → **still in strong coupling**
- Modeled by the driven dissipative GP equation coupled to an excitonic reservoir

Polariton condensate & polariton lasing properties

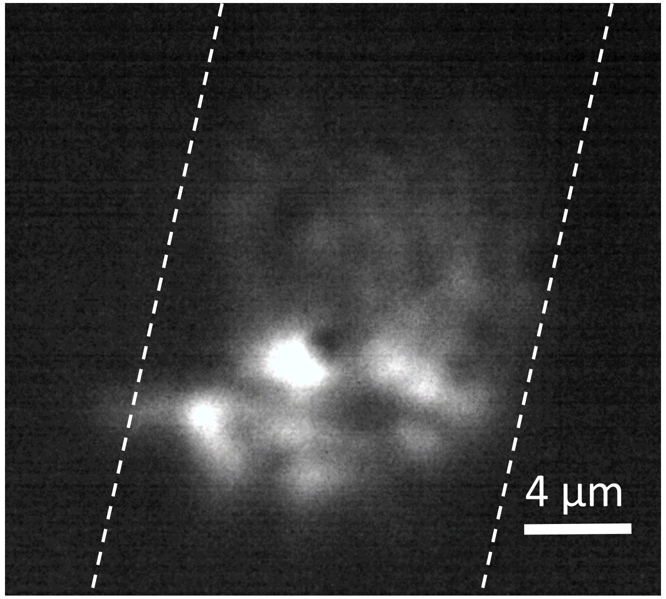
- Michelson interferometer in the retroreflector configuration
- First-order spatial coherence $g^{(1)}(\mathbf{r}, -\mathbf{r})$

Real space image



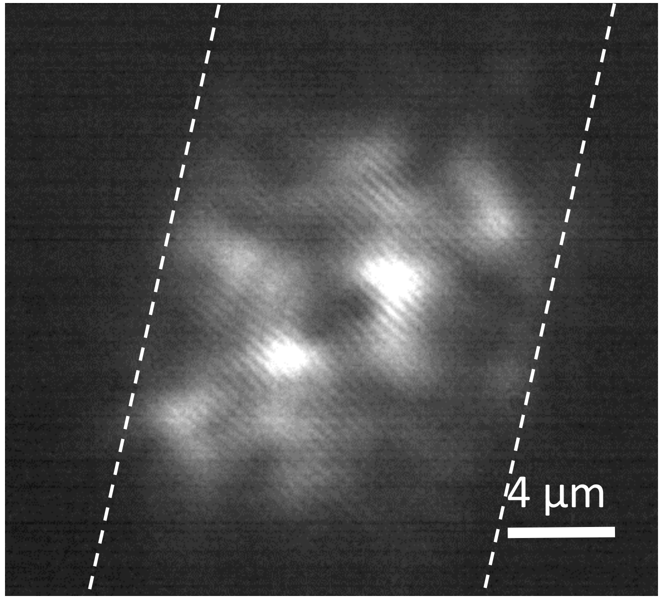
+

Centro-symmetric real space image



=

Interference fringes

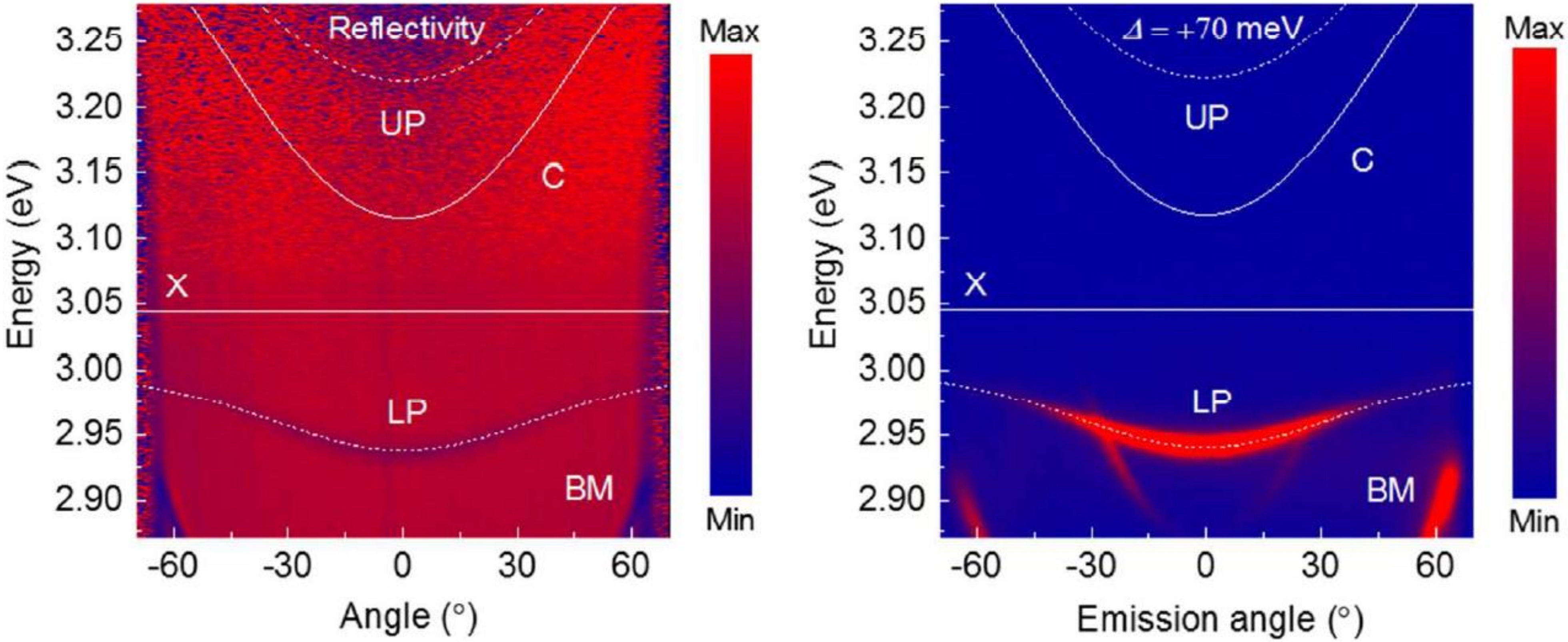


Build-up of a long range spatial coherence in the condensate

From strong coupling to weak coupling regime

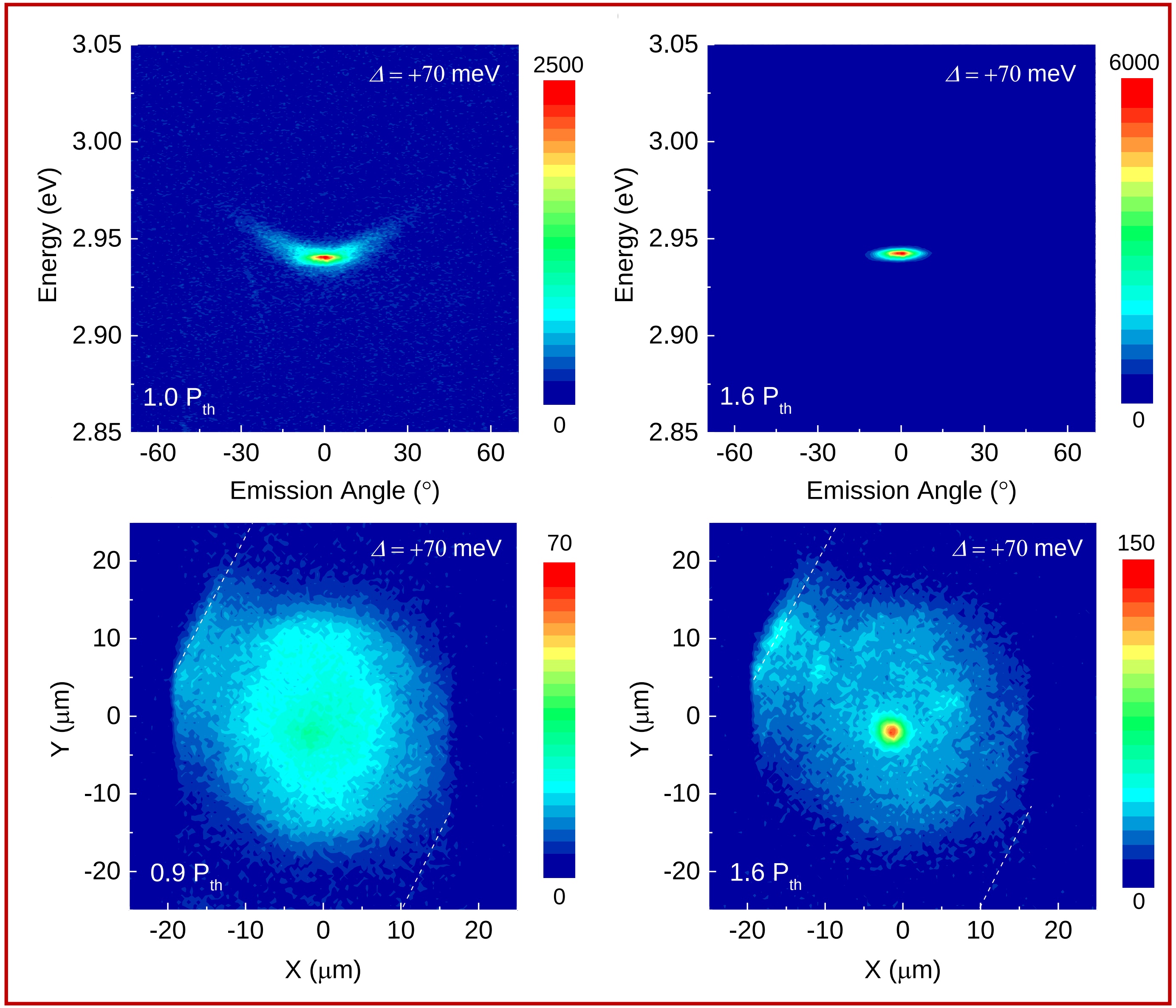
- Positive detuning Δ = + 70 meV
- Room temperature
- CW excitation

Rabi splitting ~ 273 meV

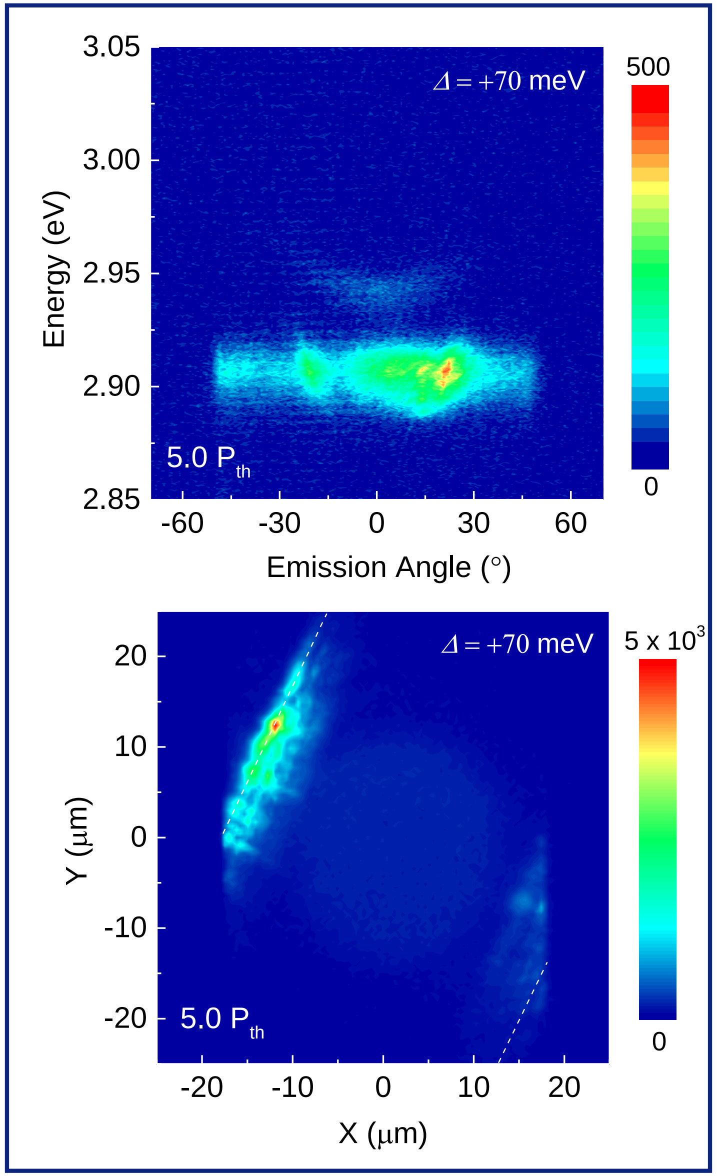


From strong coupling to weak coupling regime

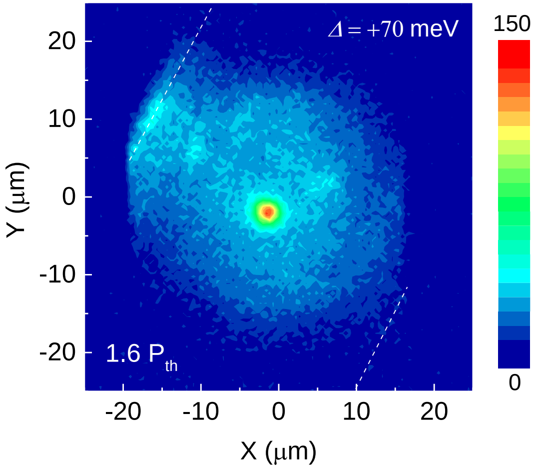
Strong coupling

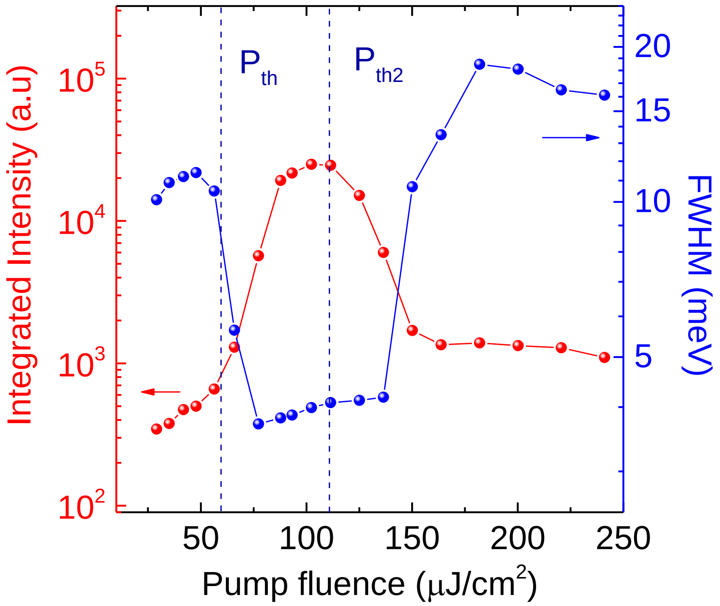


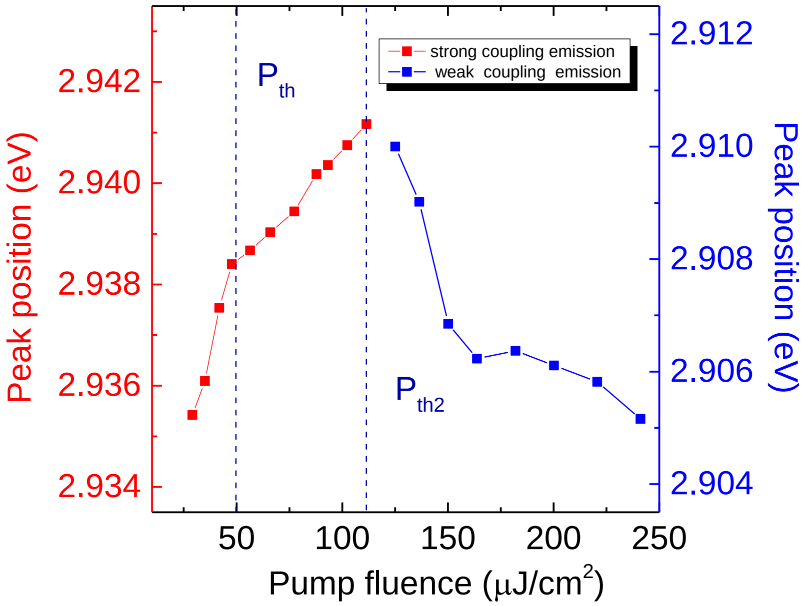
Weak coupling

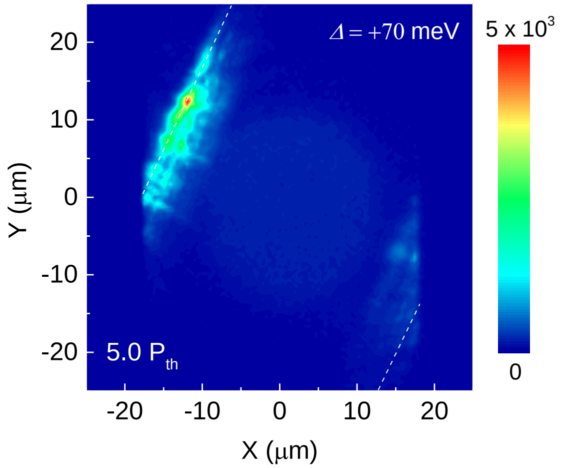


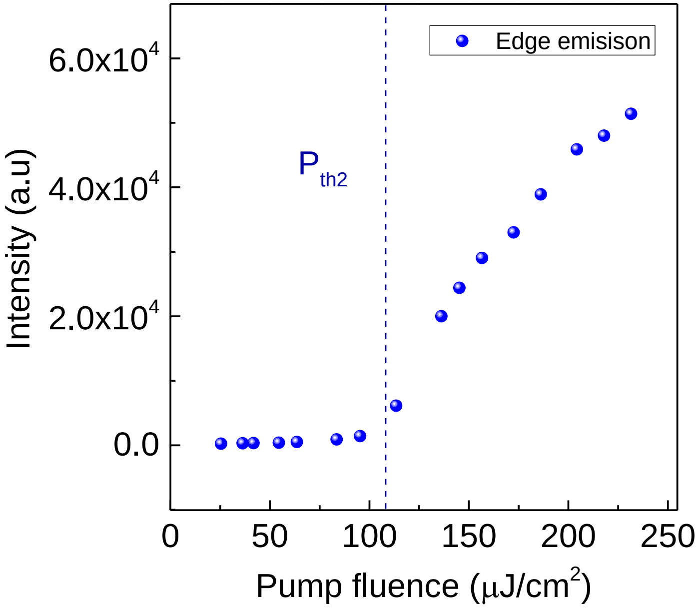
From strong coupling to weak coupling regime

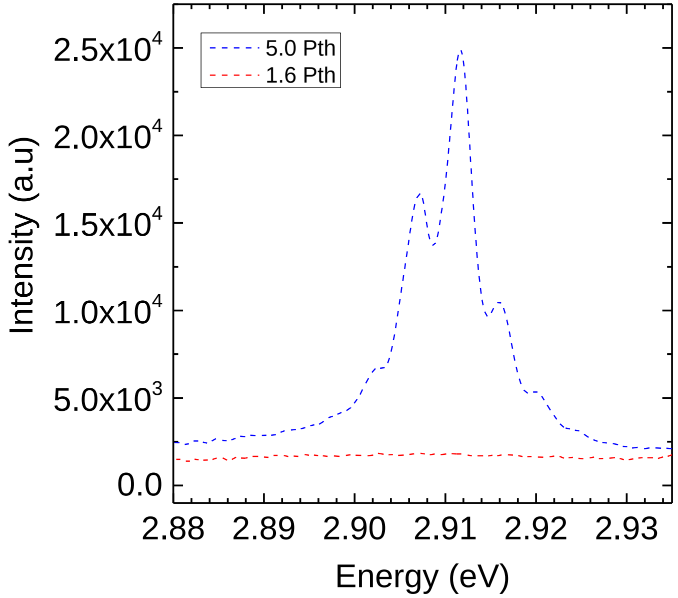








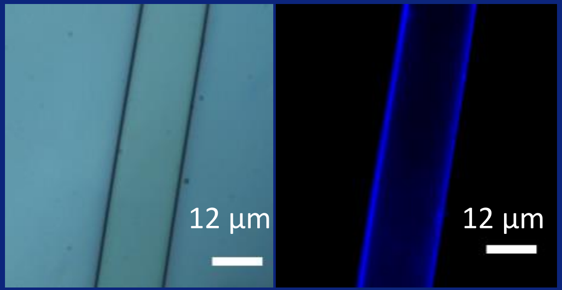




Experimental results in all-inorganic perovskite-based microcavities at room temperature

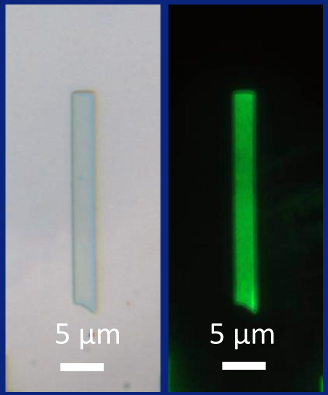
- Polariton condensation in $CsPbCl_3$ microplatelets

  R. Su *et al.*, Nano Letters **17**, 3982 (2017)



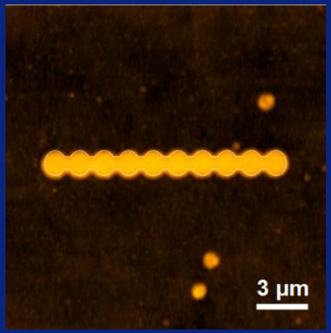
- Polariton condensate flow in $CsPbBr_3$ microwires

  R. Su *et al.*, Science Advances **4**, eaau0244 (2018)



- Polariton condensation in a $CsPbBr_3$ lattice

  R. Su *et al.*, Nature Physics **16**, 301 (2020)

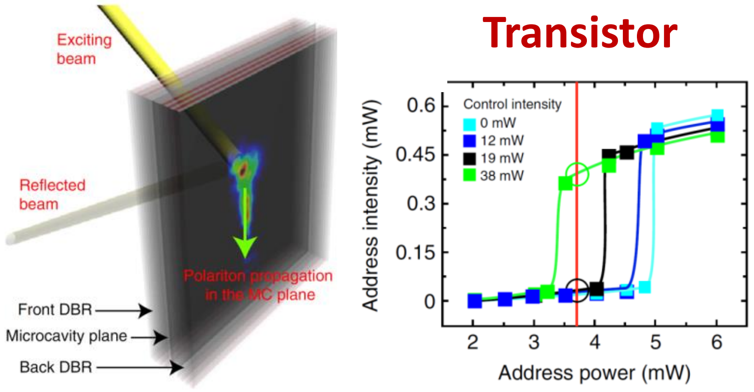


1D microwire microcavities

Motivation

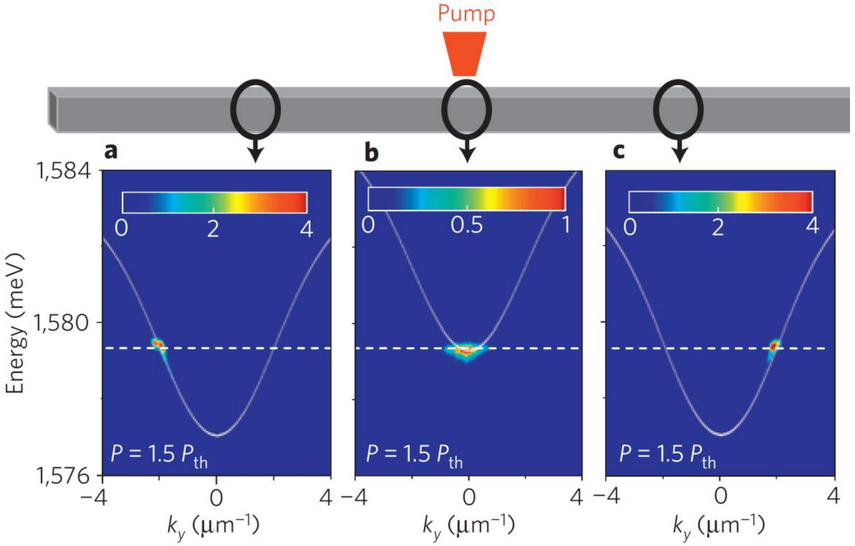
Ideal platform for polariton propagation
→ Flow / momentum controlled by the incident angle of a resonant laser or the spot size of a non resonant laser
→ Toward all-optical information processing elements and polaritonic circuits

Transistor



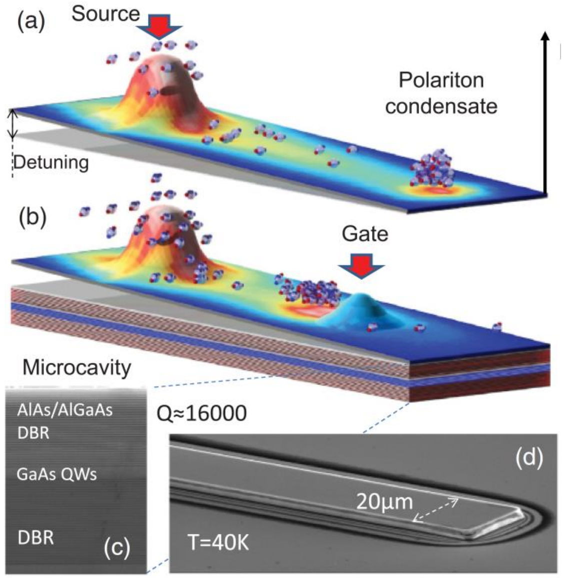
D. Ballarini *et al.*, Nat. Commun. **4**, 1778 (2013)

Propagation (condensate)



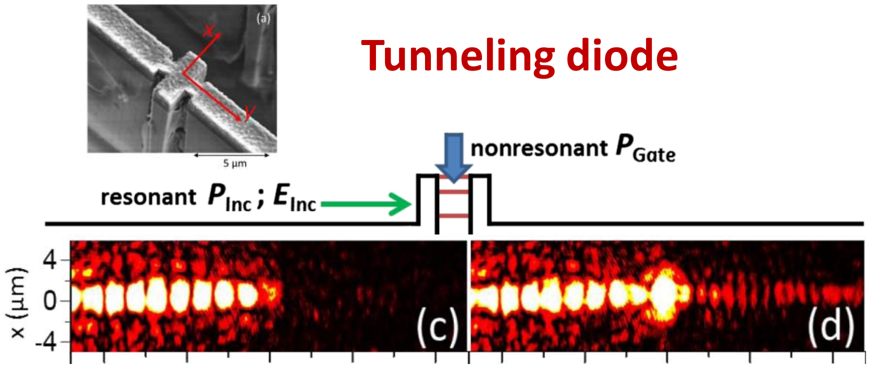
E. Wertz *et al.*, Nat. Phys. **6**, 860 (2010)

Gate / Switch (condensate)



T. Gao *et al.*, PRB **85**, 235102 (2012)

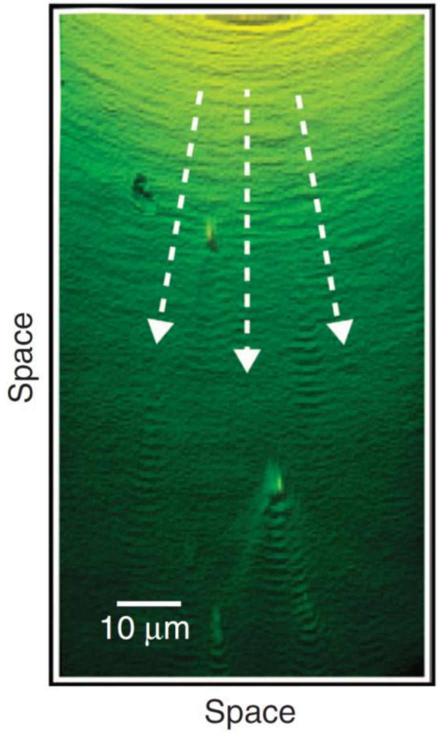
Tunneling diode



H. S. Nguyen *et al.*, PRL **110**, 236601 (2013)

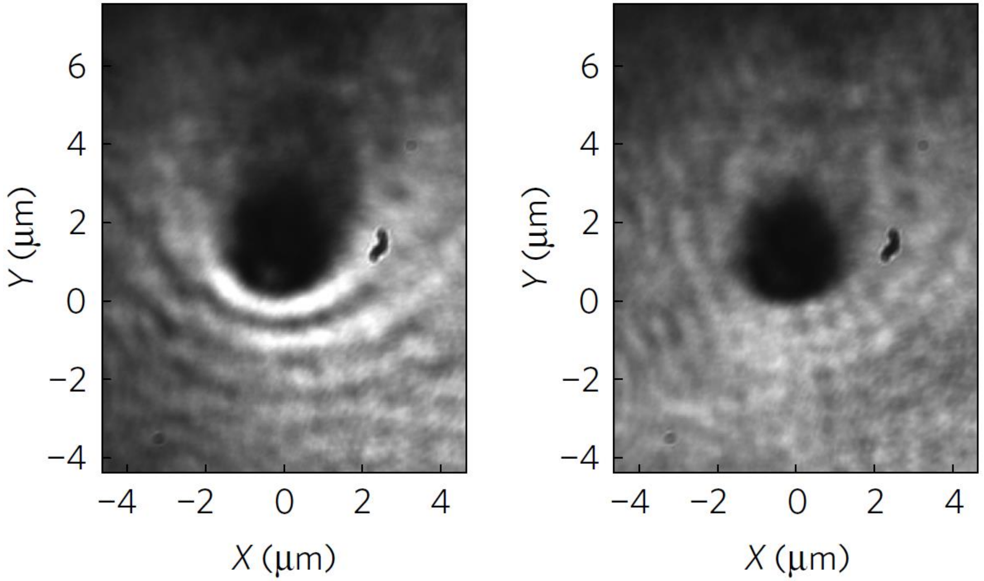
Polariton propagation at room temperature

Bloch surface wave polaritons
(no condensation)



G. Lerario *et al.*, Light Sci. Appl. **6**, e16212 (2017)

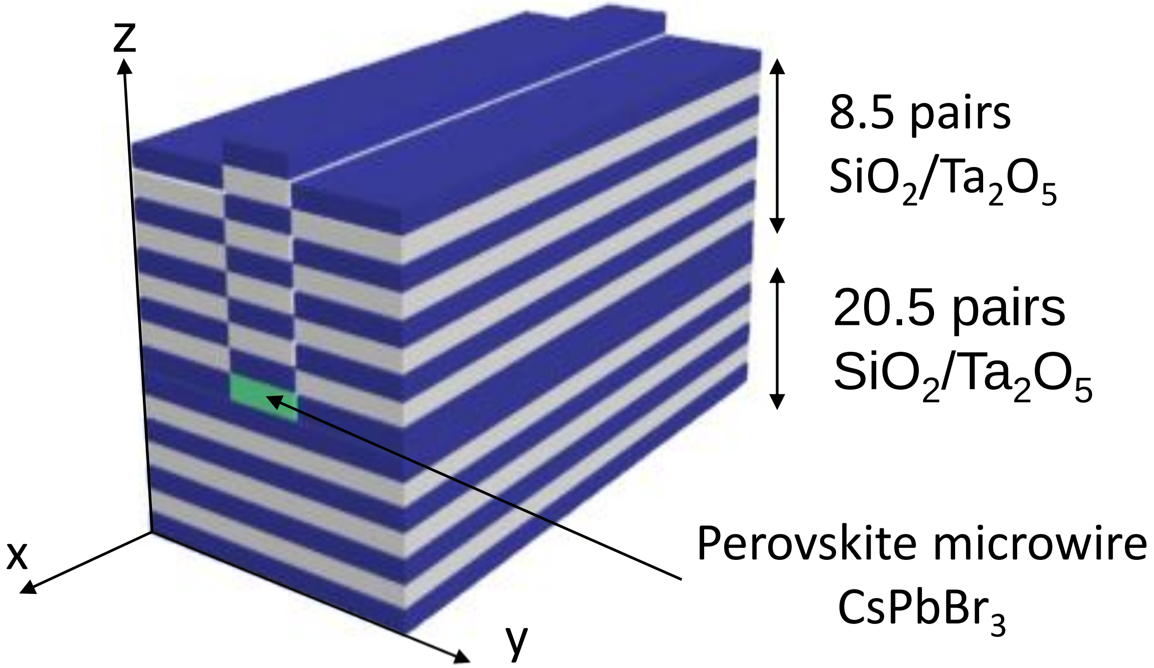
Polariton superfluid
(resonant excitation)

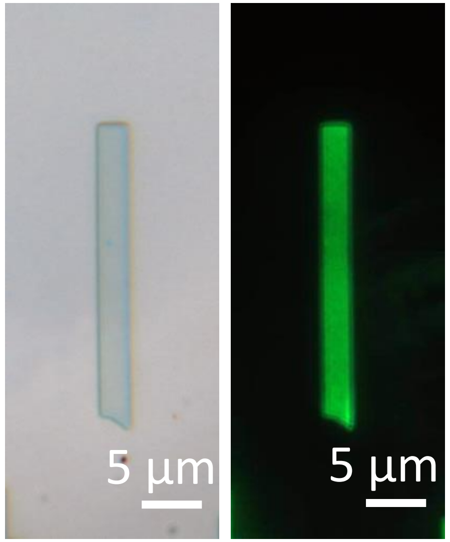


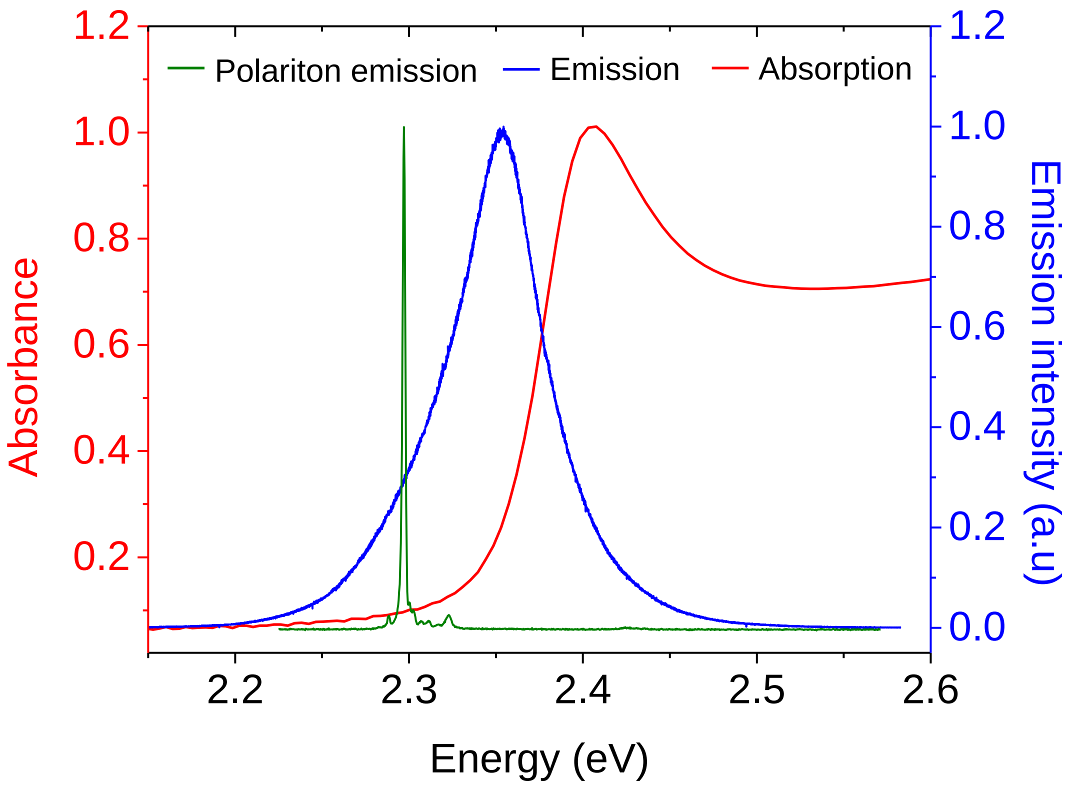
G. Lerario *et al.*, Nat Phys. **13**, 837 (2017)

Room temperature long-range propagation of a coherent polariton condensate under non resonant excitation in perovskite ?

Perovskite microwire microcavity



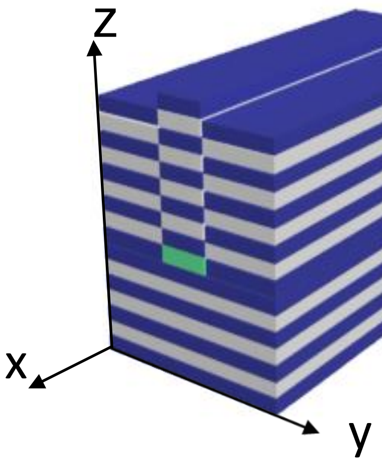




- Etching-free 1D microcavity
- Microwire of length ~ 30 µm and width ~ 2 µm
- PMMA protection layer
- Quality factor Q ~ 1200

Perovskite microwire microcavity

Room temperature strong coupling regime – 1D polaritons

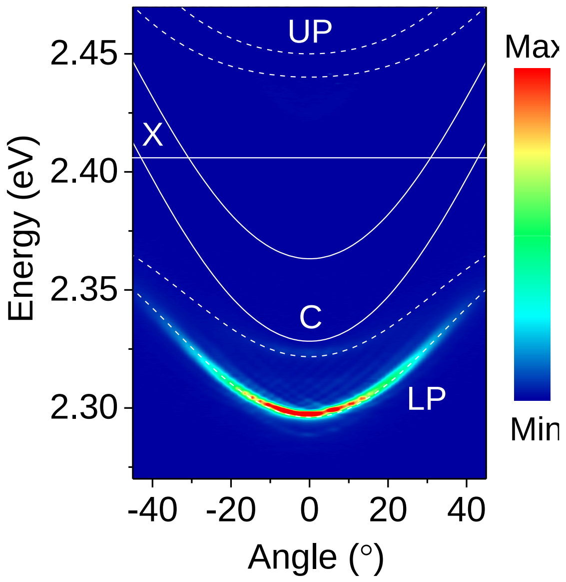


Lateral confinement along y
→ additional quantization

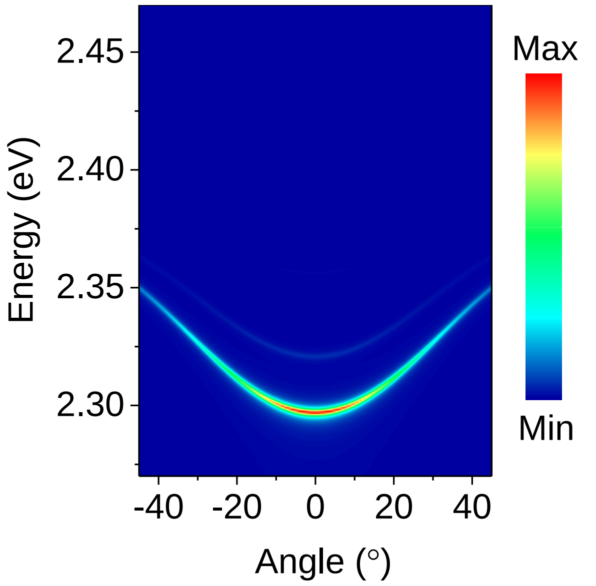
$$E^{c}{}_{1\mathrm{D}}(j,k_x) = E_0\sqrt{1+\underbrace{\left[\frac{(j+1)\pi}{L_y}\right]^2}_{k_y\,(\,j\,=\,0,1,2\ldots\,)}\frac{1}{k^2{}_z}+\left(\frac{k_x}{k_z}\right)^2}$$

- $2\Omega \sim 120$ meV
- $\Delta_1 = -80$ meV ; $\Delta_2 = -40$ meV

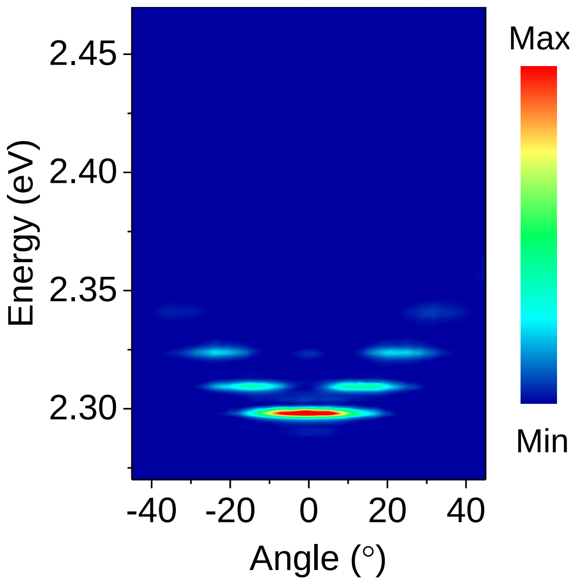
Along x



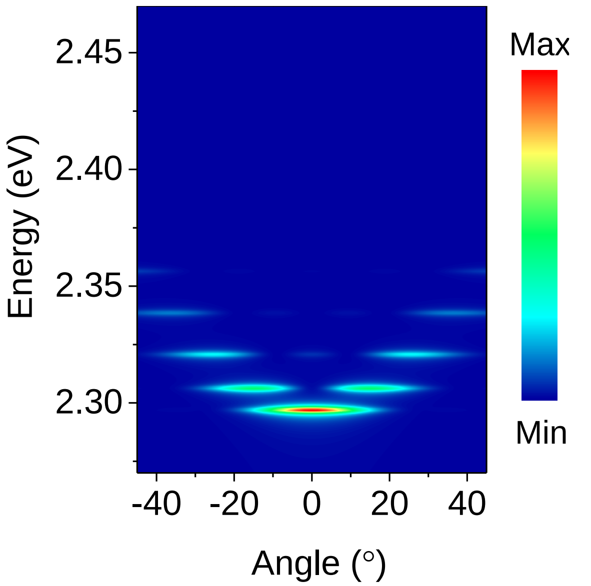
Simulation



Along y

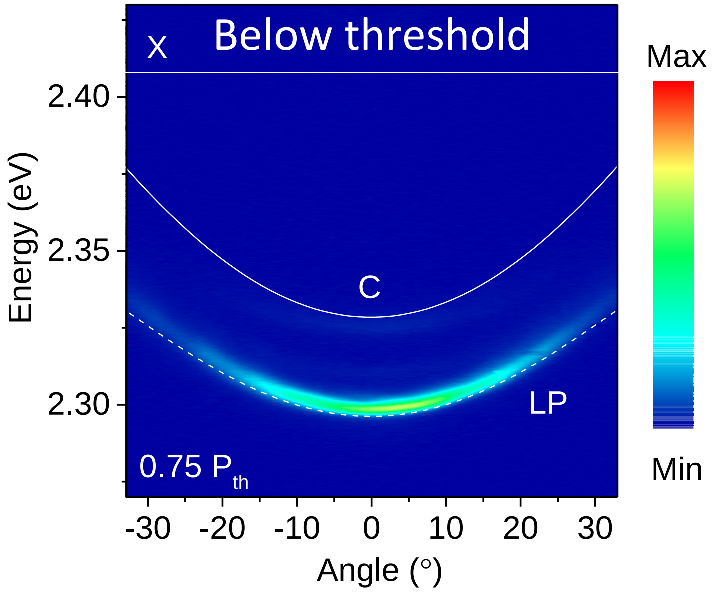


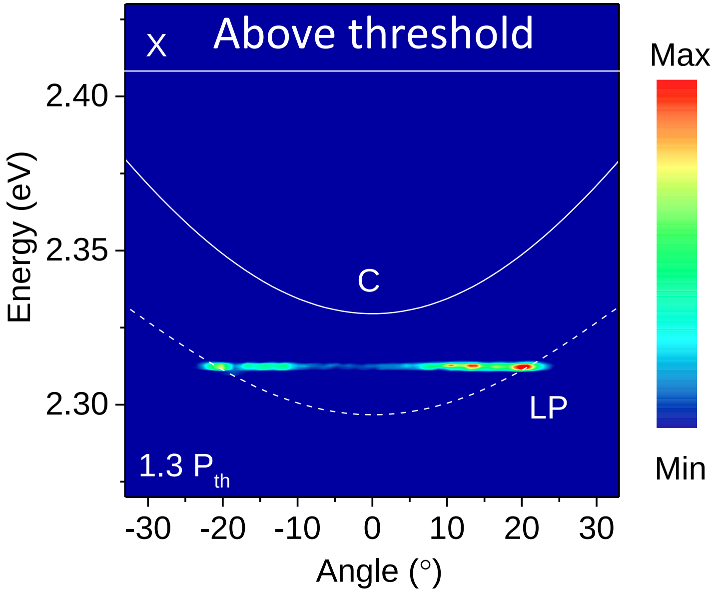
Simulation

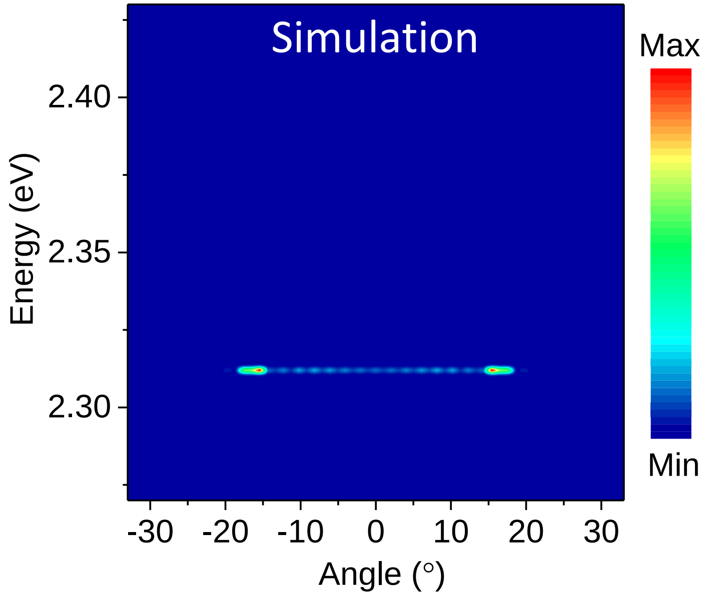


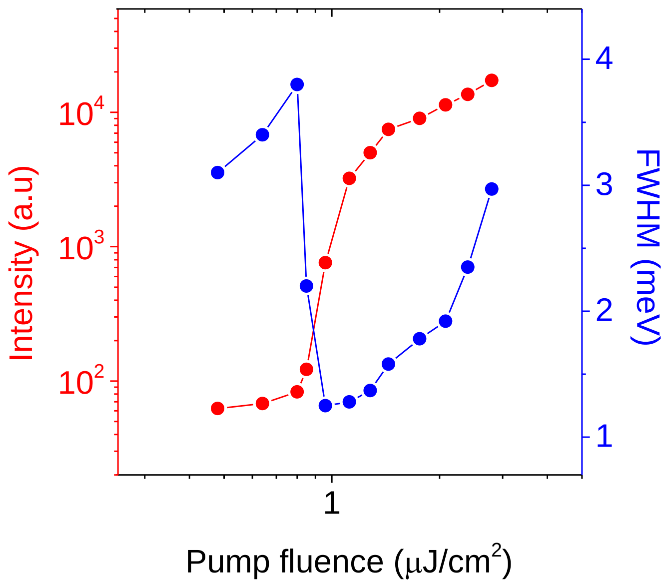
Perovskite microwire microcavity

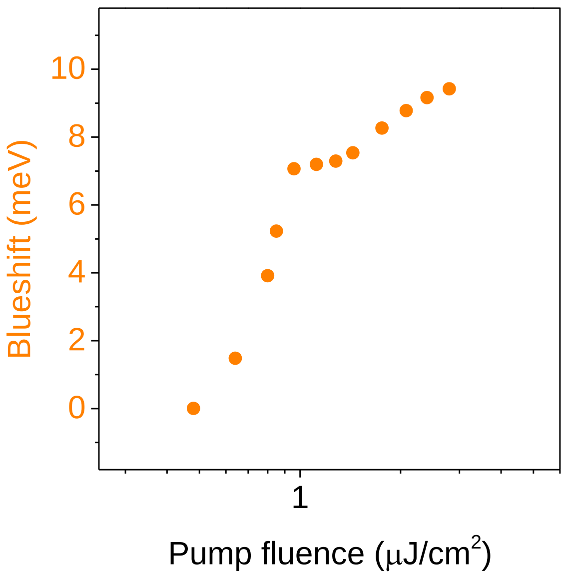
Room temperature exciton-polariton condensation

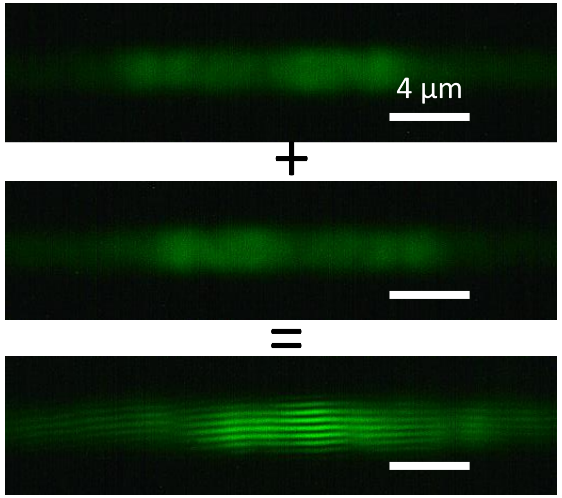






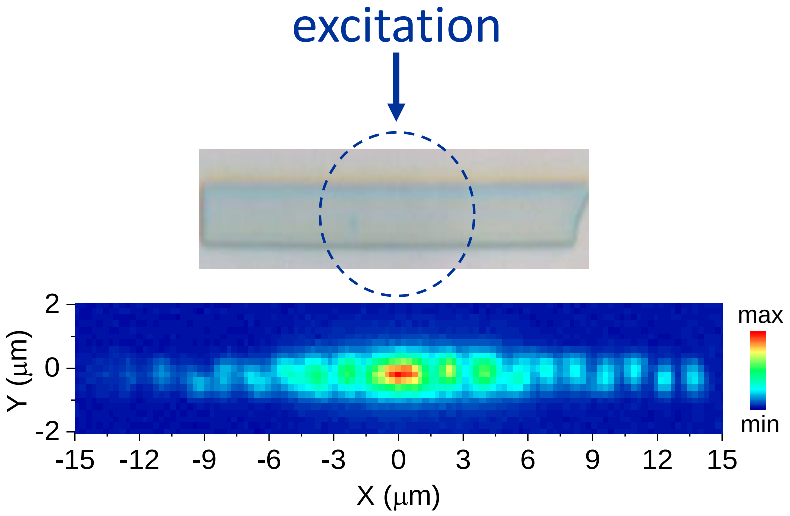


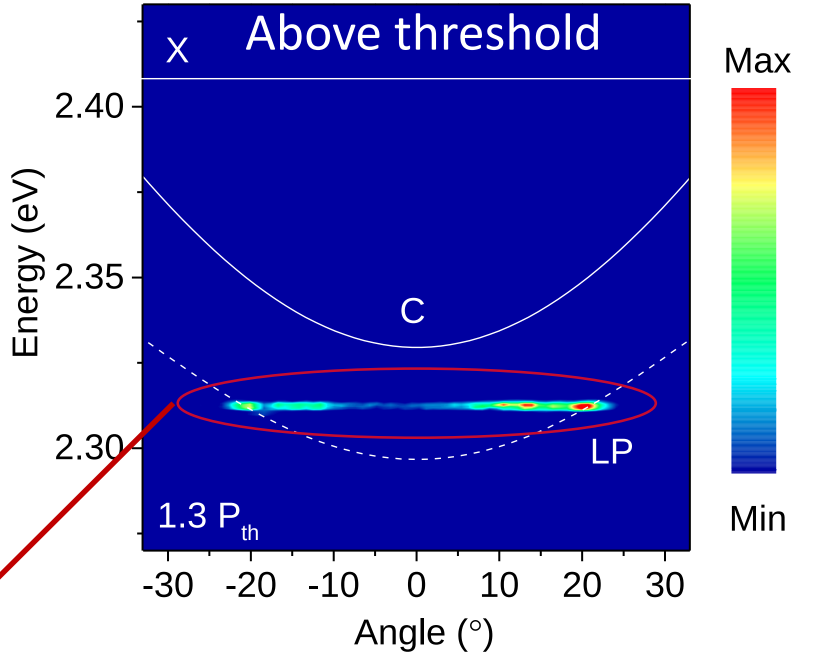


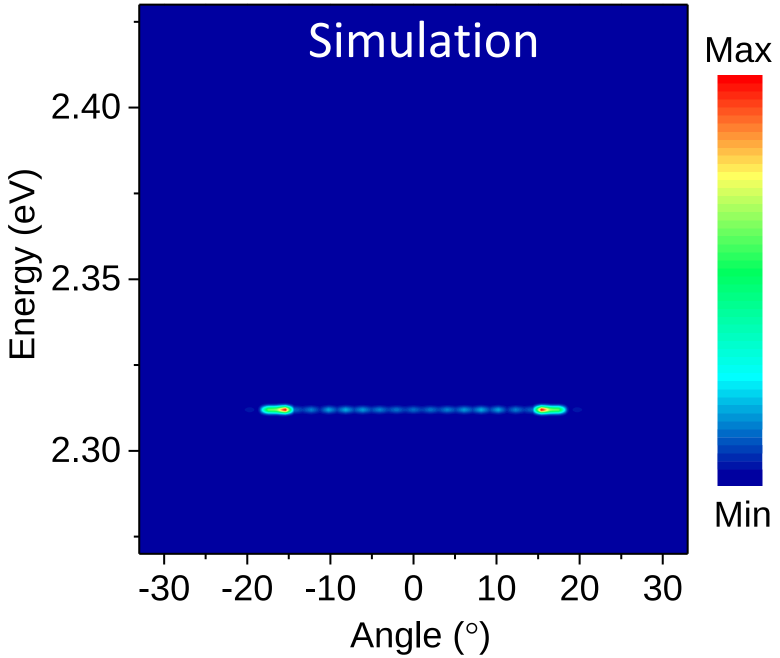


Perovskite microwire microcavity

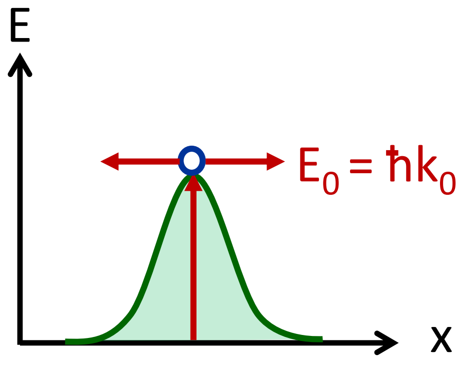
Polariton condensate flow

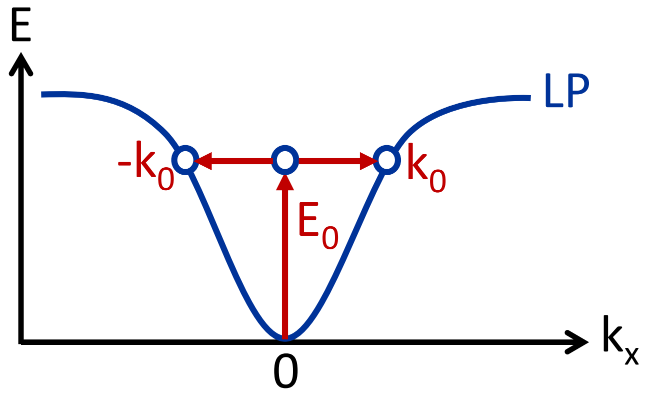






Non-equilibrium condensate





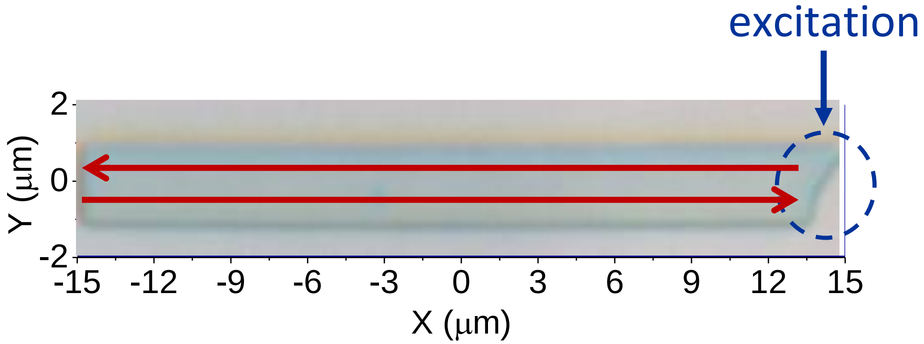
Initial blueshift converted into kinetic energy

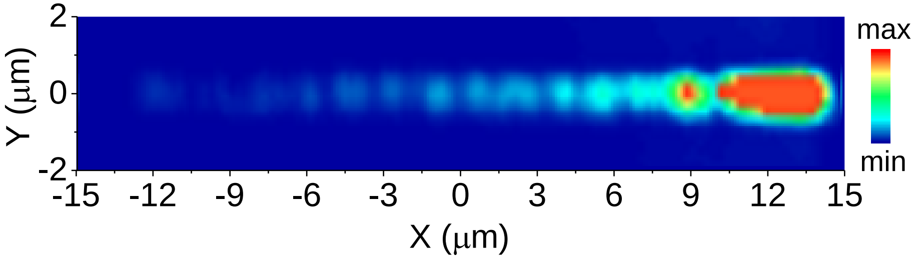
Ballistic propagation of the condensate outwards the excitation spot

M. Wouters et al., PRB **77**, 115340 (2008)
E. Wertz et al., Nat. Phys. **6**, 860 (2010)

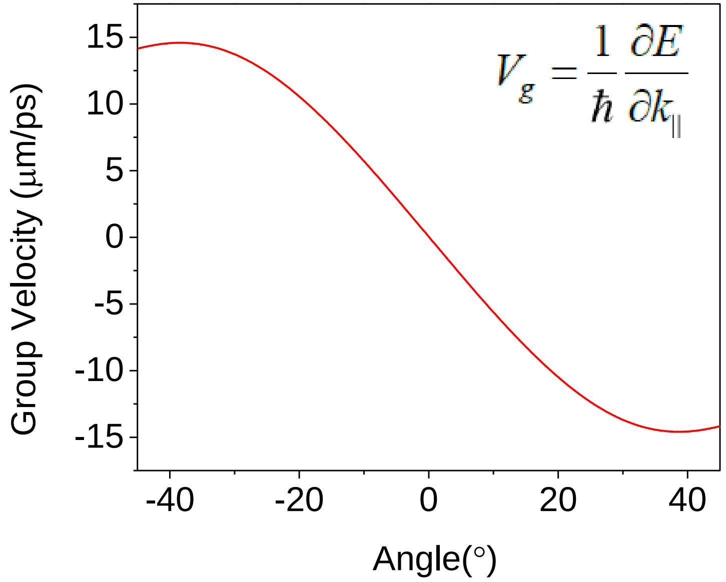
Perovskite microwire microcavity

Polariton condensate flow

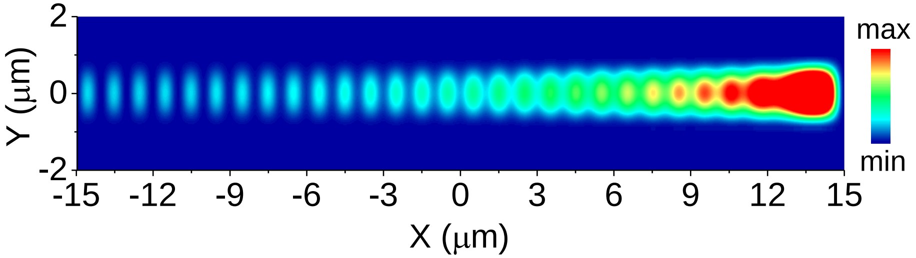




Interference pattern throughout the whole microwire
→ **some polaritons have propagated over 60 µm**



Simulation

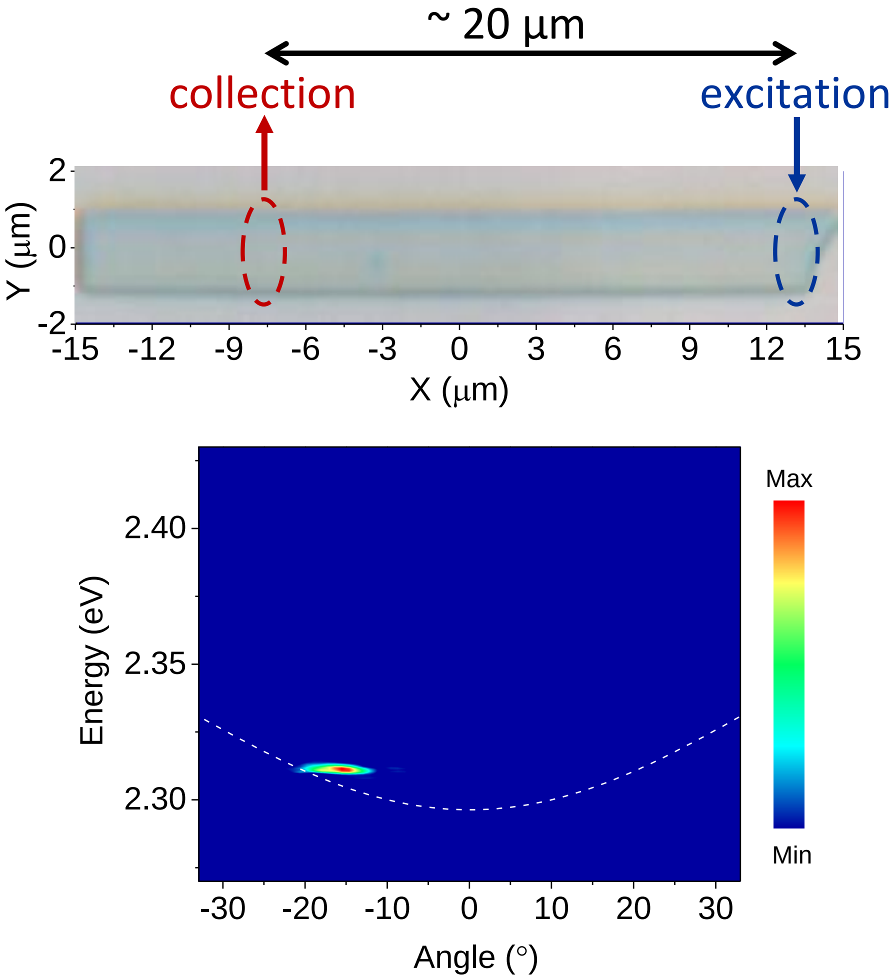


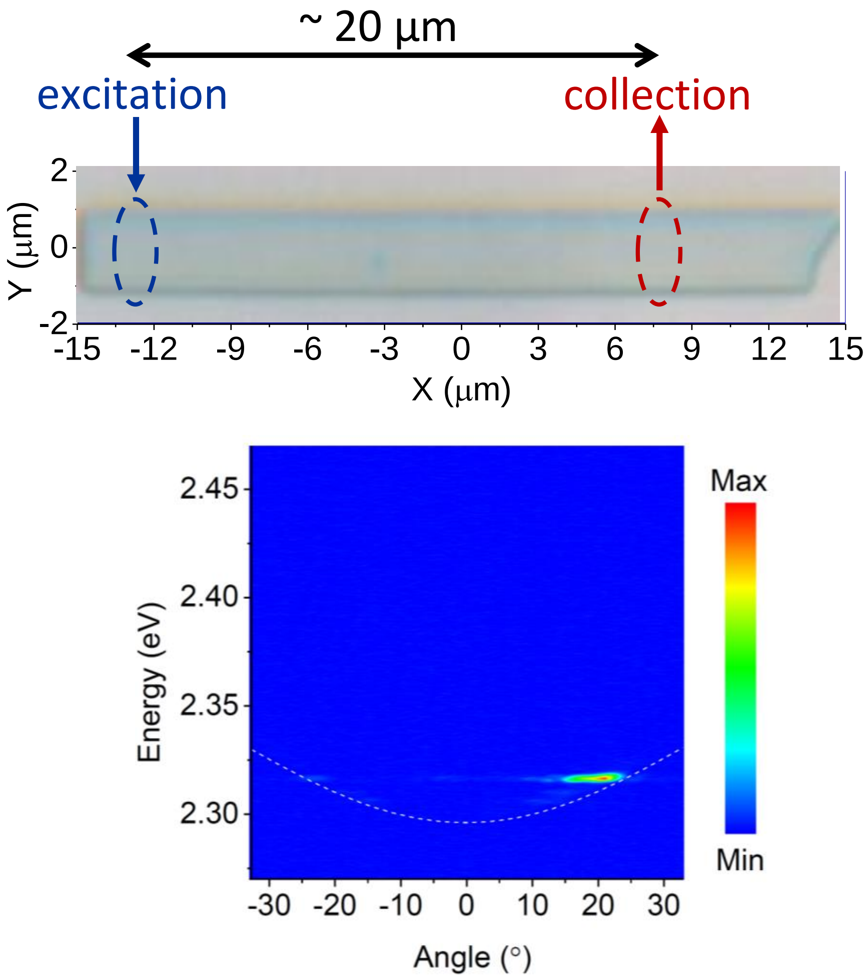
- Solving the driven-dissipative mean field dynamics
- Polariton group velocity < 10 µm/ps
- Observation of the interference fringes depends on the polariton decay rate in the calculation (0.2 meV)

→ **Polariton lifetime of 3 ps**

Perovskite microwire microcavity

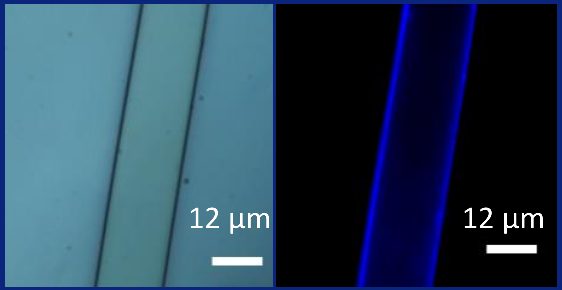
Control of the polariton condensate flow





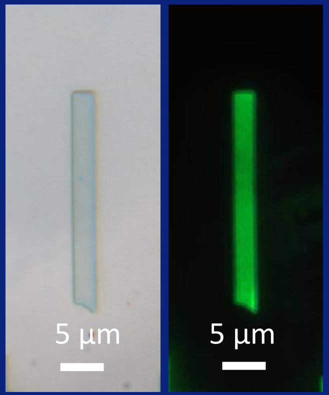
- Non-symmetric far-field emission due to dominant propagation in one direction
- Propagation controlled by changing the position of the pumping spot

Experimental results in all-inorganic perovskite-based microcavities at room temperature



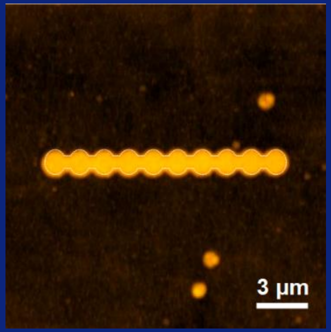
- Polariton condensation in $CsPbCl_3$ microplatelets

  R. Su *et al.*, Nano Letters **17**, 3982 (2017)



- Polariton condensate flow in $CsPbBr_3$ microwires

  R. Su *et al.*, Science Advances **4**, eaau0244 (2018)



- Polariton condensation in a $CsPbBr_3$ lattice

  R. Su *et al.*, Nature Physics **16**, 301 (2020)

Polariton condensation in lattices

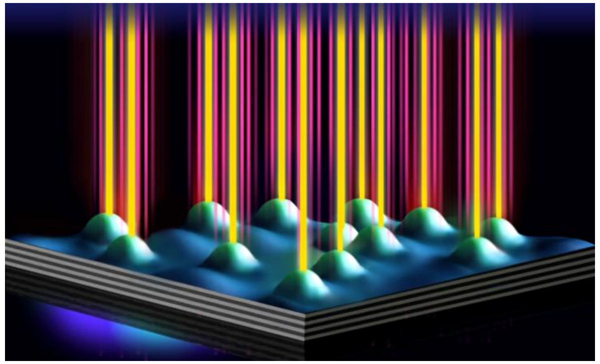
Motivation

Strong lattice

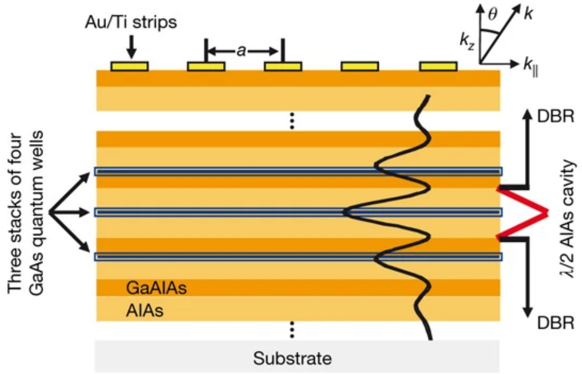
→ Robust trapping of polariton condensates in periodic potentials (large forbidden bandgap opening)

→ Strong inter-site coupling for coherent motion of polariton within the lattice (large lattice bandwidth)

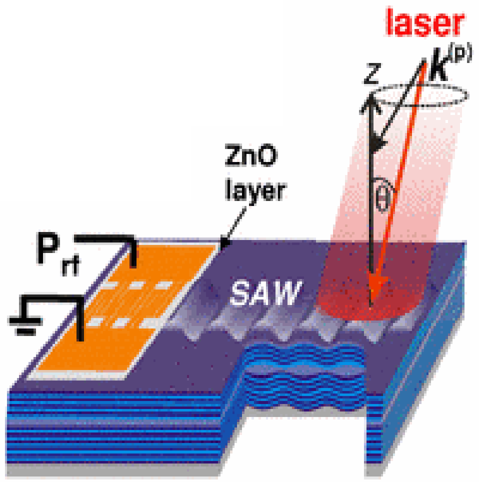
Quantum simulators



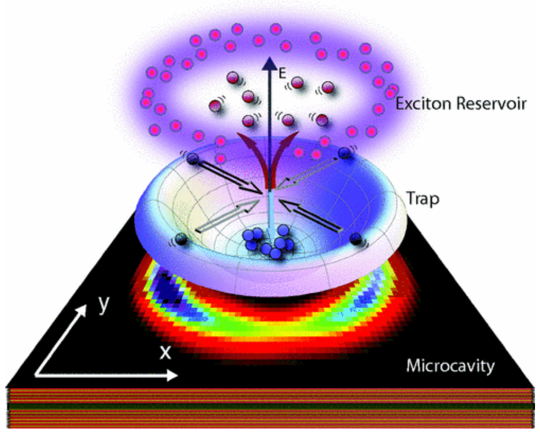
Credits to N. Berloff (Univ. of Cambridge)



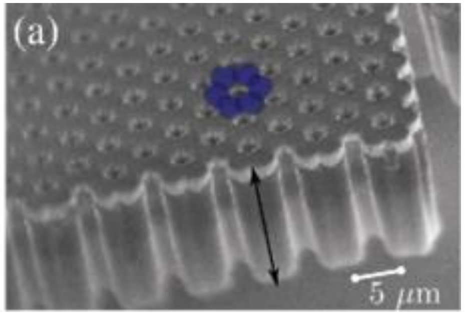
C. Lai *et al.*, Nature **450**, 529 (2007)



E. Cerda-Mendez *et al.*, PRL **105**, 116402 (2010)

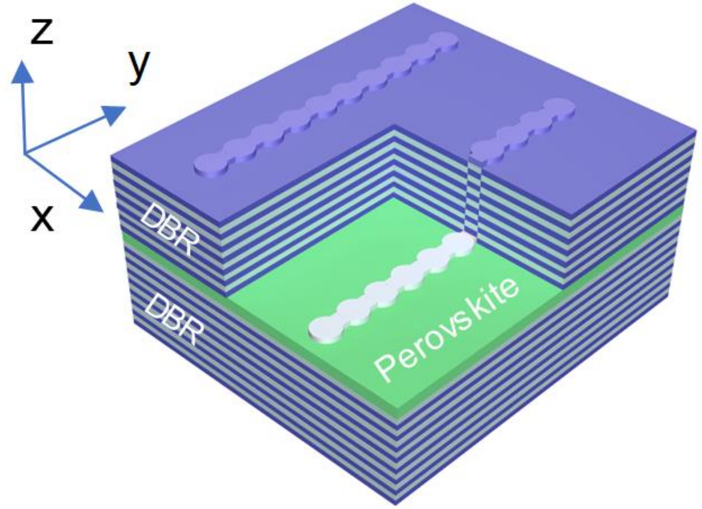


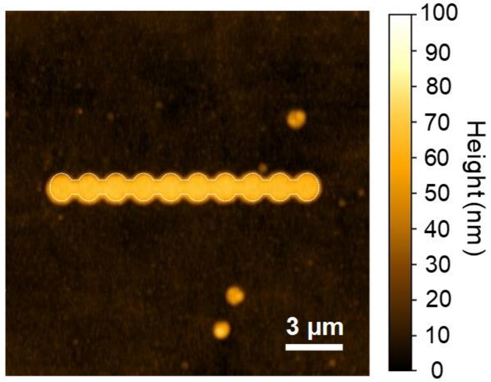
A. Askitopoulos *et al.*, PRB **88**, 041308(R) (2013)



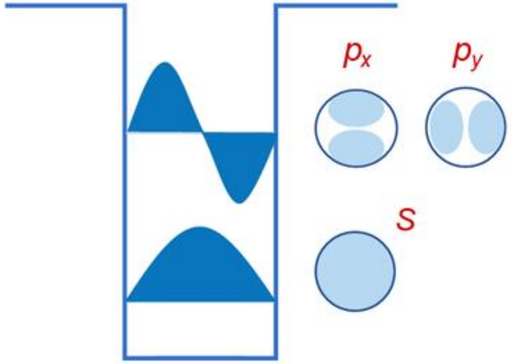
T. Jacqmin *et al.*, PRL **112**, 116402 (2014)

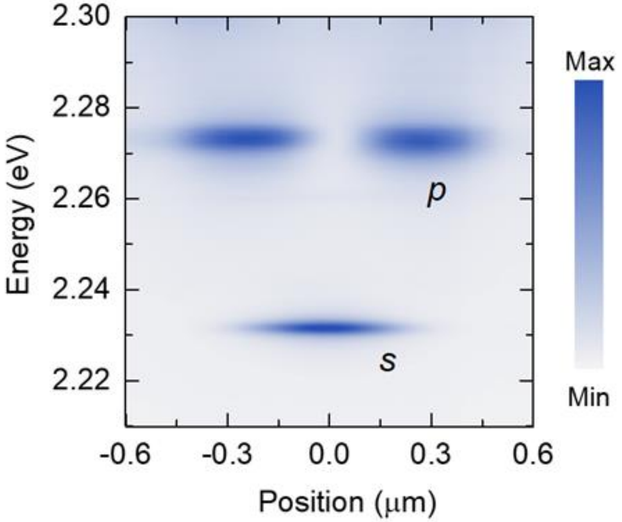
1D perovskite lattice





- 150 nm-thick $CsPbBr_3$ perovskite platelet
- Patterning of the 60 nm-thick PMMA spacer layer on top of the perovskite
- Array of 10 pillars of 1 µm diameter connected with channels of 0.5 µm width
- Deep periodic potential of 400 meV (to compare to the 6 meV linewidth)

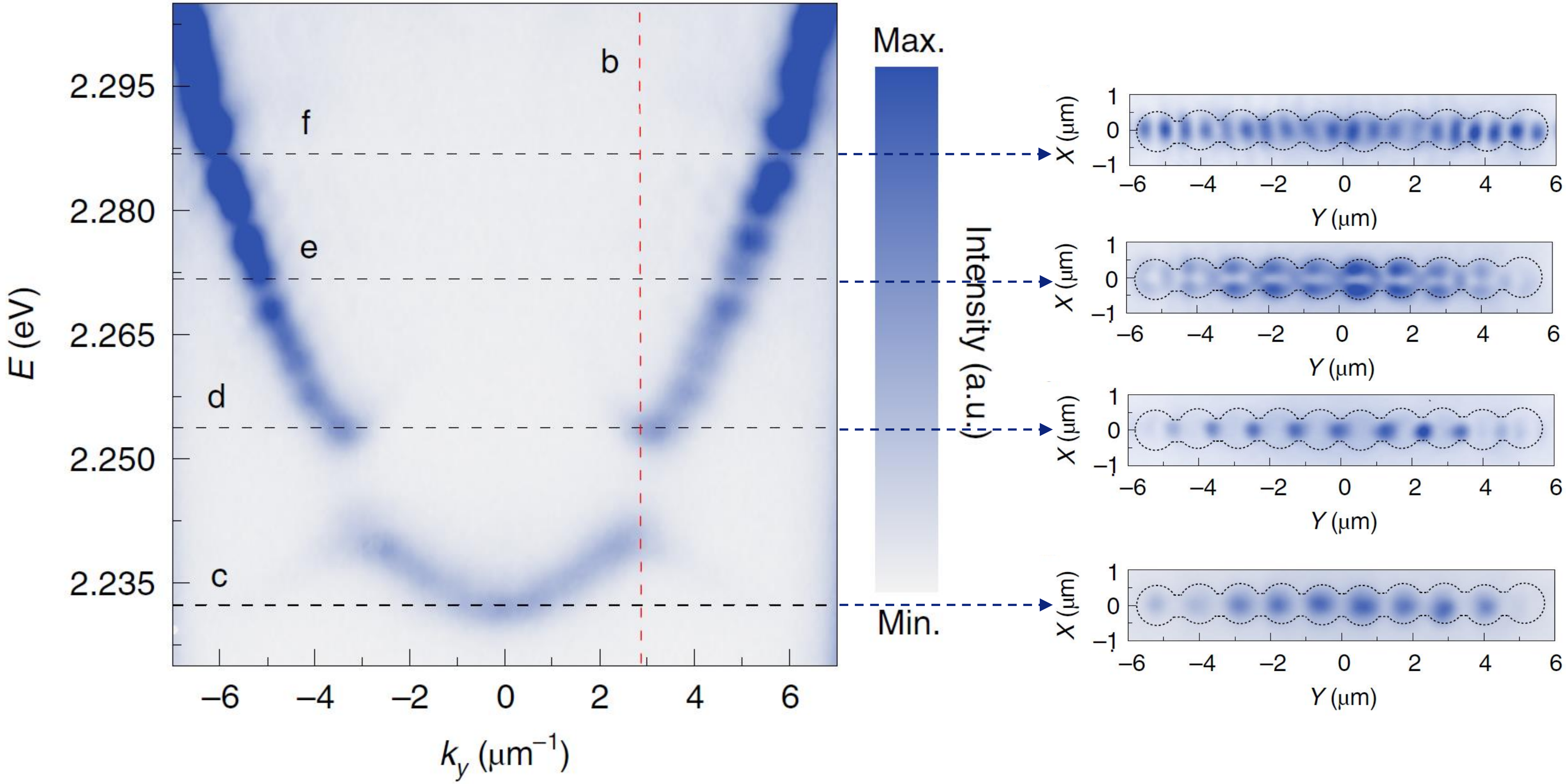




- 3D confinement in a pillar
- Orbital states in a single pillar: a non-degenerate symmetric s state and a twofold-degenerate antisymmetric p state

1D perovskite lattice

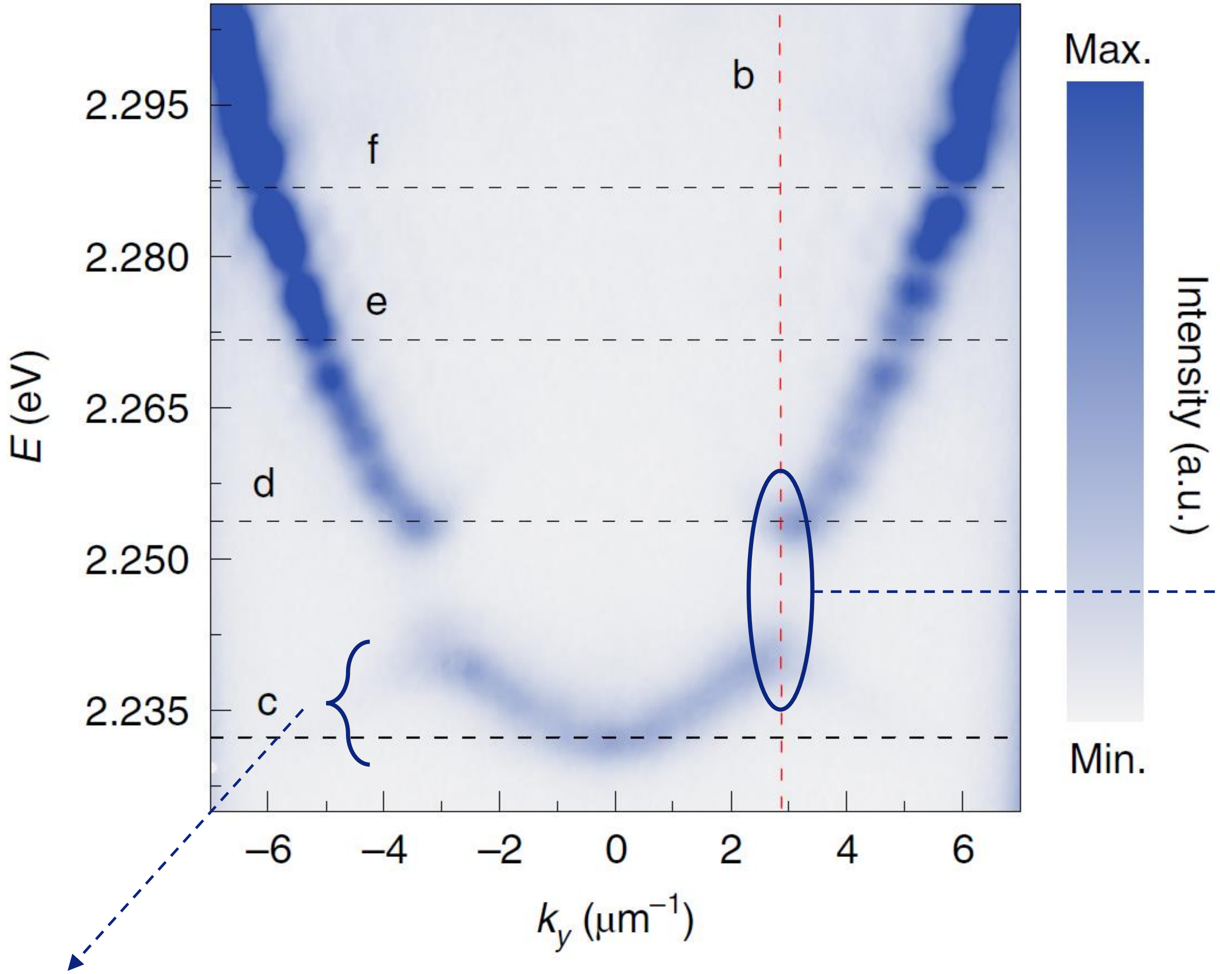
Room temperature strong coupling regime



- Lower band = s-orbital state of the pillars + channel states
- Upper band = p-orbital states of the pillars + channel states

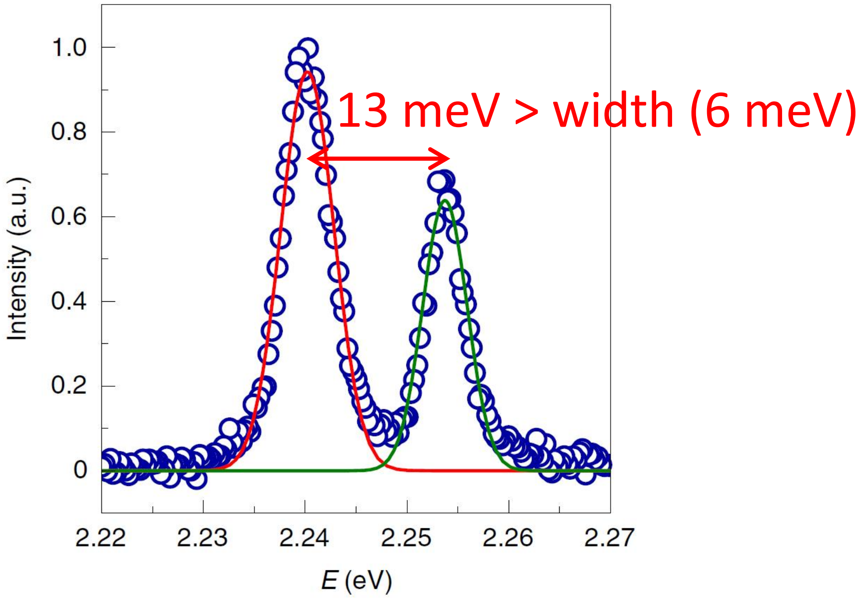
1D perovskite lattice

Room temperature strong coupling regime



Bandgap opening

Robust confinement of polaritons within the band

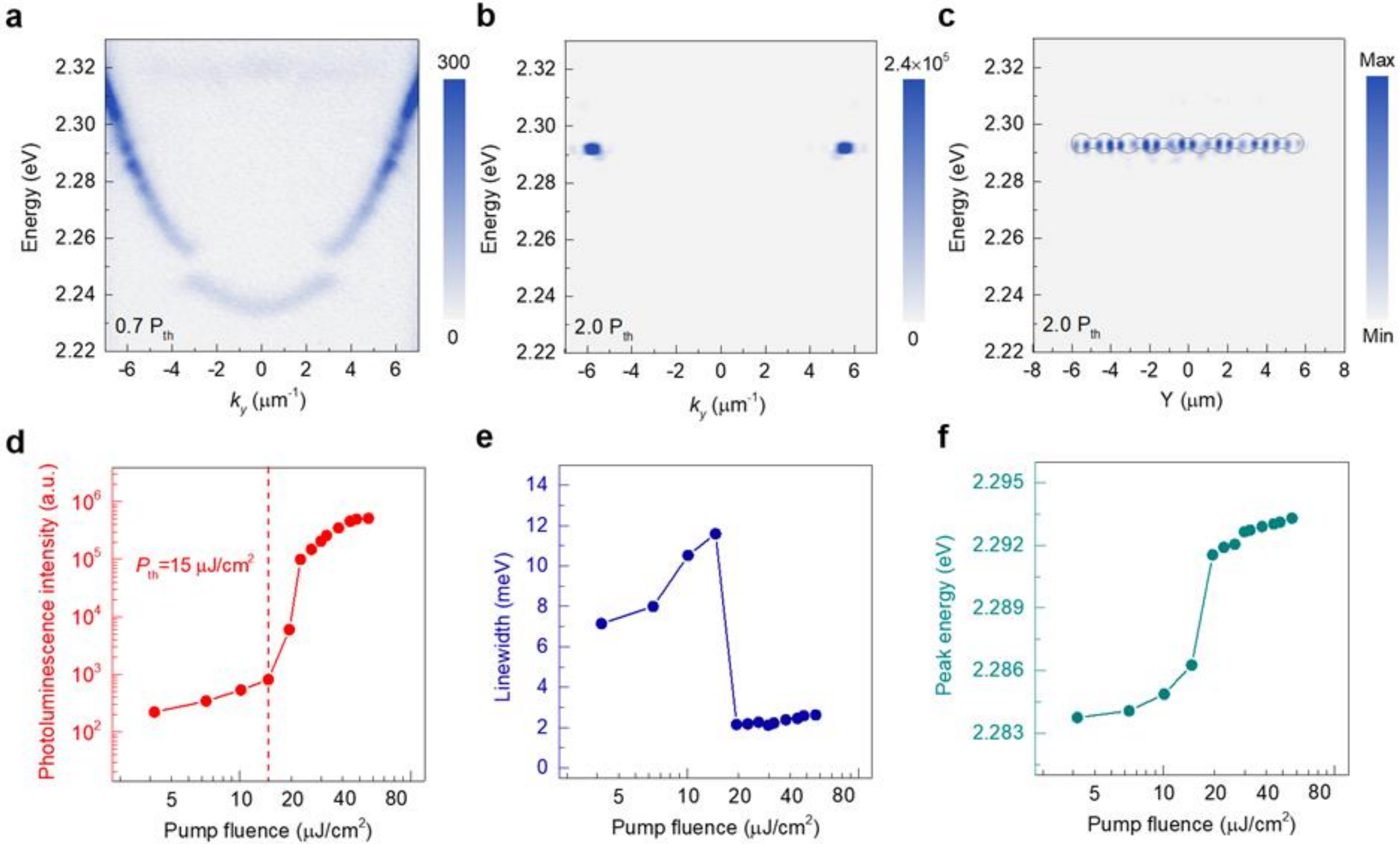


Large lattice bandwidth (8.5 meV)

Inter-site coupling (2 meV) allowing motion of the polaritons within the lattice sites

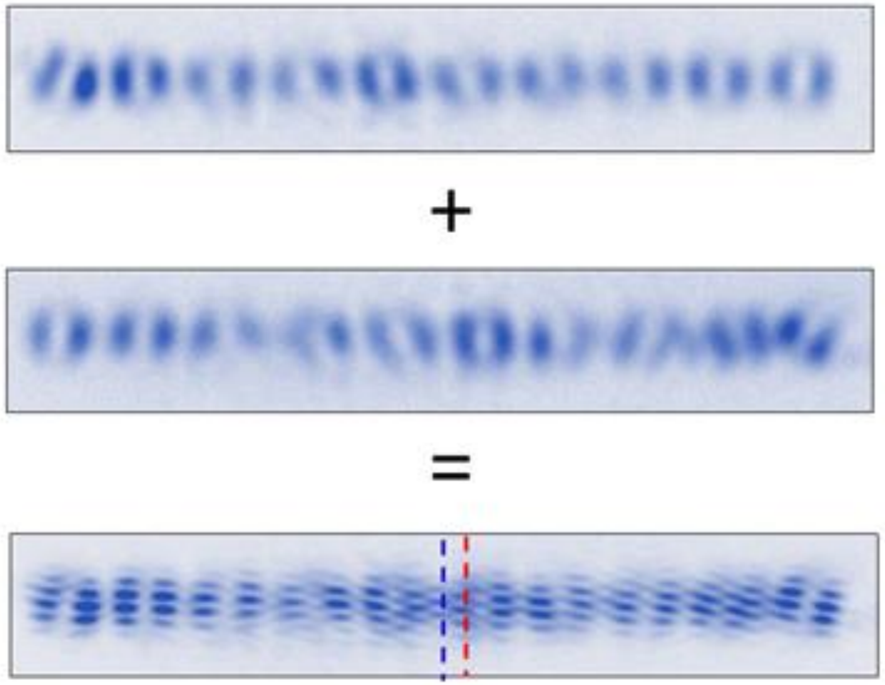
1D perovskite lattice

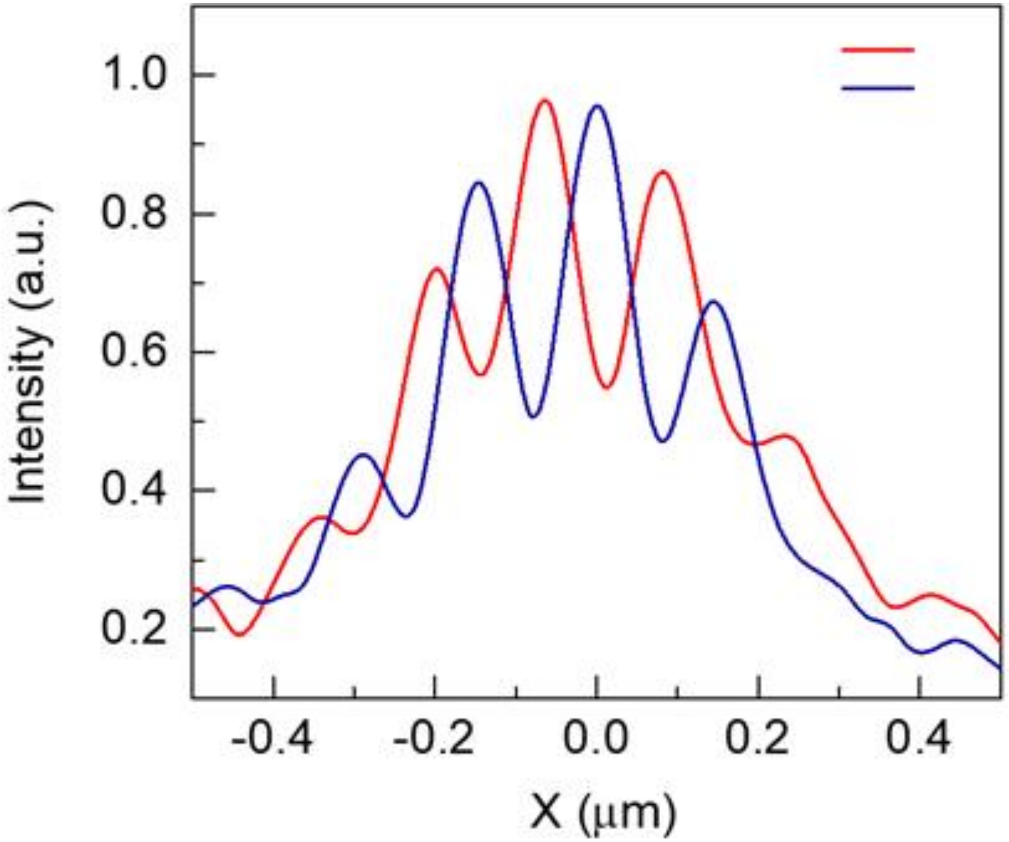
Room temperature polariton condensation



1D perovskite lattice

Room temperature polariton condensation





Superposition of the real-space image and its inverted image
→ interference fringes within a distance as large as 12 μm
→ build-up of the long-range spatial coherence

Conclusion

- Room temperature polariton condensation in perovskite of different compositions and different geometries
  - → Low cost room-temperature polariton devices based on wavelength tunable epitaxy-free materials

- Room temperature long range polariton condensate flow in perovskite microwires
  - → Polaritonic circuits

- Room temperature polariton condensation in a perovskite lattice with sizable tunability in terms of potential landscape engineering and lattice design
  - → Realization of arbitrary lattice geometries for polaritonic devices