

# TEMPLATED SELF-ASSEMBLY OF PEROVSKITE NANOCRYSTALS INTO 2D-CHIRAL PHOTONIC STRUCTURES FOR CIRCULARLY POLARIZED PHOTOLUMINESCENCE

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# PRESENCE OF CIRCULARLY POLARIZED LIGHT

Spectroscopy & sensing

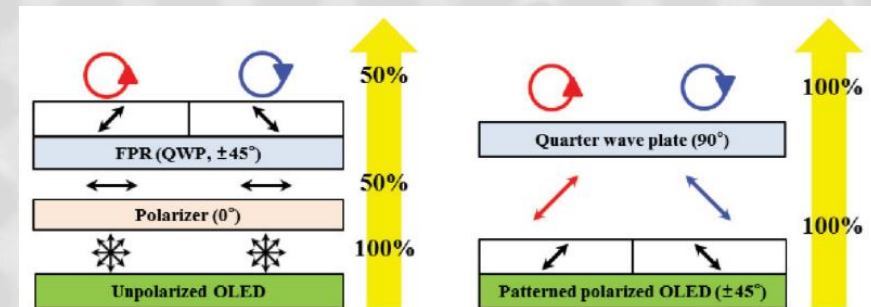
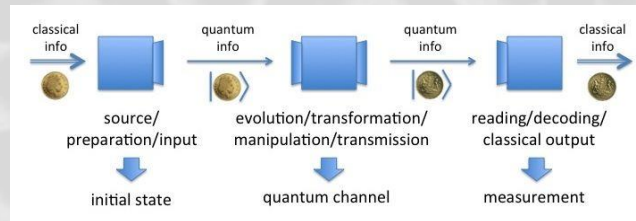
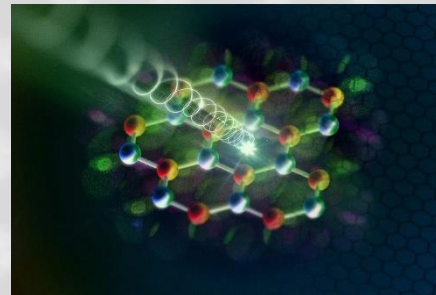
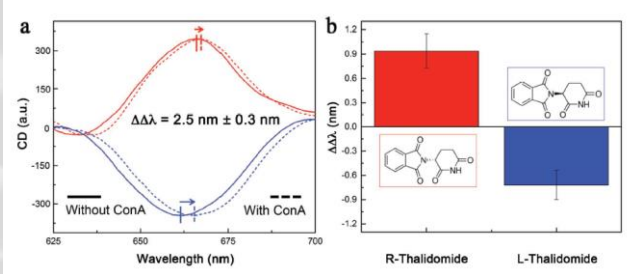
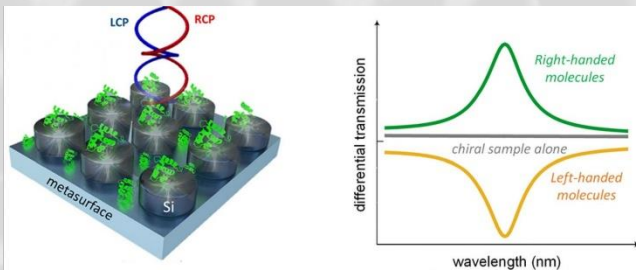
Quantum information processing

Display devices

$$\text{Circular Dichroism (CD)} = \frac{A_{LCP} - A_{RCP}}{\text{Extinction}}$$

$$\psi = a_1|L\rangle + a_2|R\rangle$$

Increased efficiency

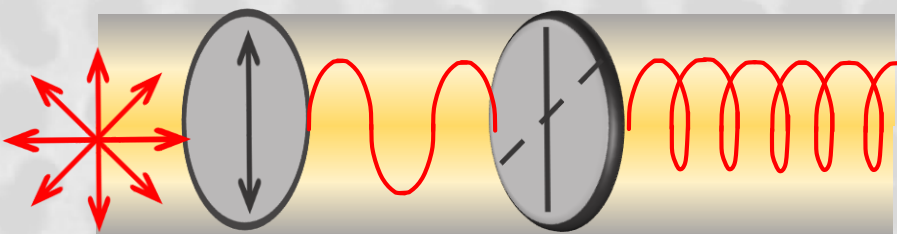


# CIRCULARLY POLARIZED LIGHT EMISSION

## Common way: filtering out

## Chirality transfer from photonic structure

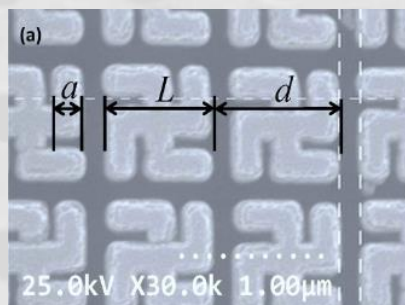
## Chiral emitters



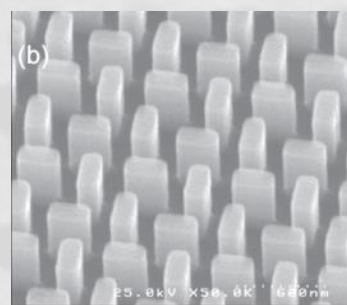
Linear polarizer

$\lambda/4$  waveplate

- Full dissymmetry
- Difficult to miniaturize

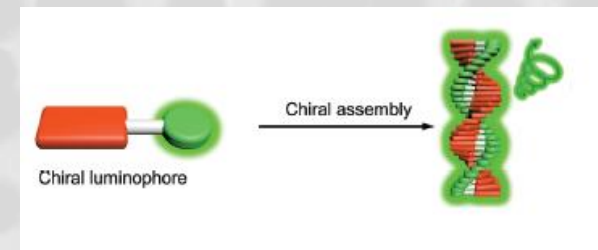


PRB **89**, 045316 (2014)



PRB **92**, 205309 (2015)

- High dissymmetry factor
- Expensive and time consuming in fabrication
- Not in visible spectra



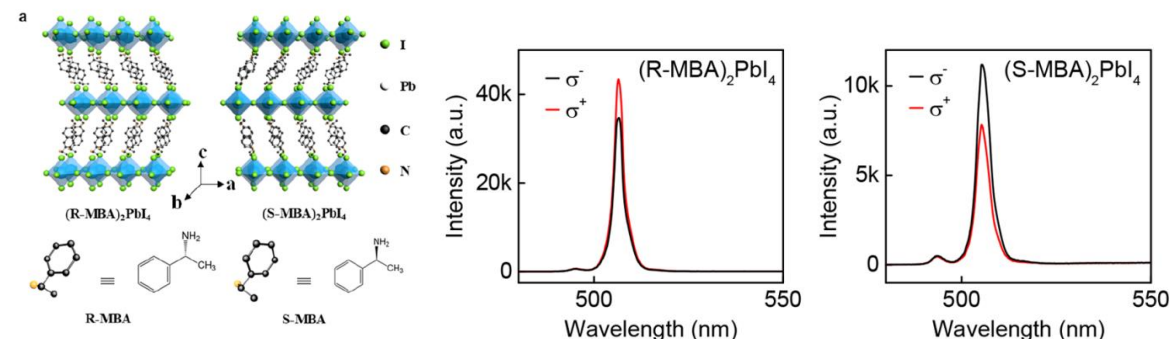
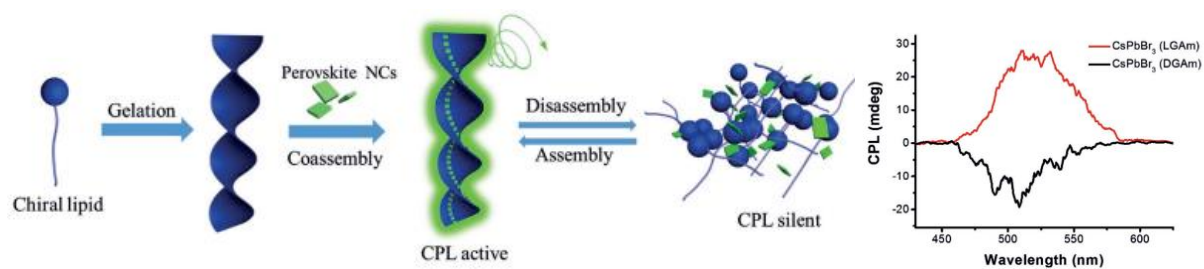
Adv. Mater. **2020**, 32, 1900110

- Low dissymmetry factor
- Emission in visible range

# CHEMICALLY MODIFIED PEROVSKITES

- Assembled in chiral structures
- Need of chiral reagents to induce chiral assembly

$$g_{lum} = 2 \cdot \frac{I_{LCP} - I_{RCP}}{I_{LCP} + I_{RCP}}$$



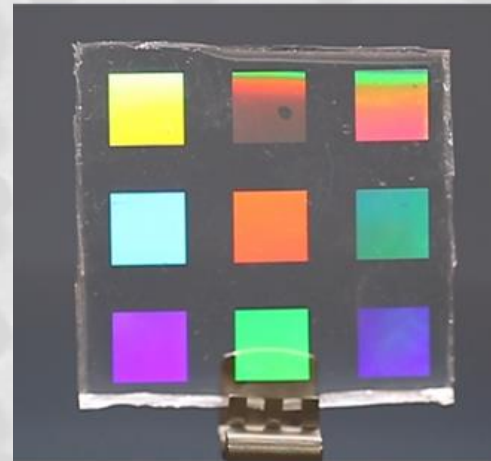
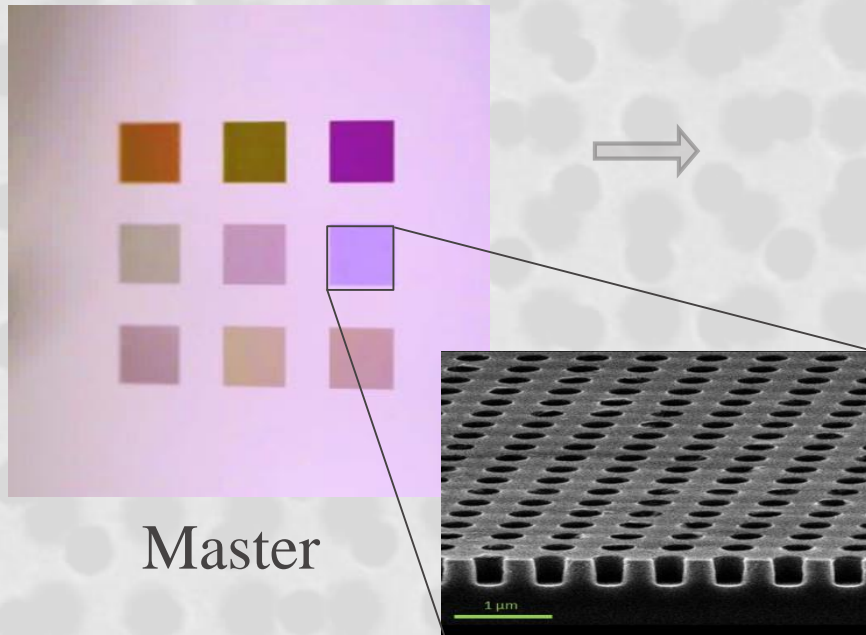
Adv. Mater. 2018, 30, 1705011

•  $g_{lum} \sim 1e-3$

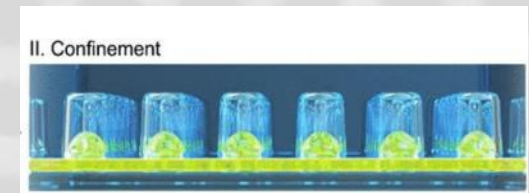
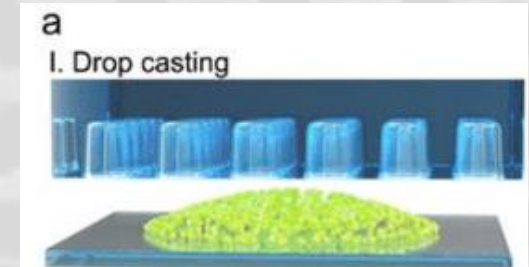
ACS Nano 2019, 13, 3659–3665

•  $g_{lum} \sim 0.2$

# SOFT NANOIMPRINTING LITHOGRAPHY (NIL)



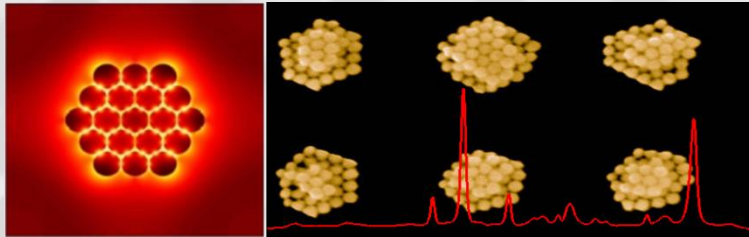
## Self assembly



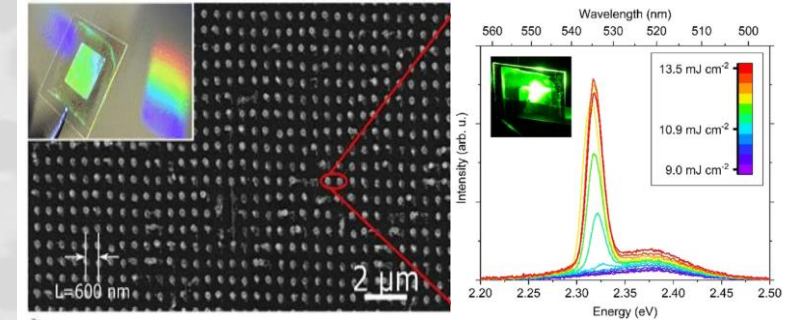
1. Nanometric resolution
2. Large area/highly scalable
3. Cleanroom free
4. Reusable components

# VERSATILE IN BOTH MATERIALS AND APPLICATIONS

ACS Nano  
2018, 12,  
8531–8539



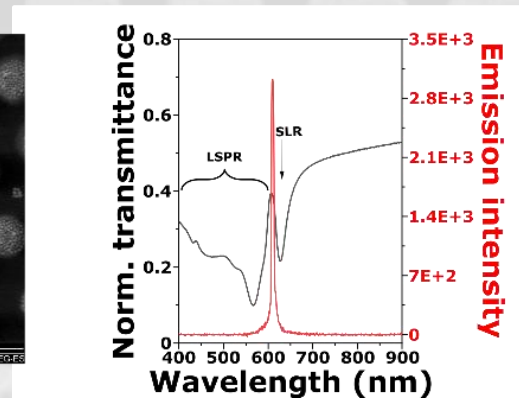
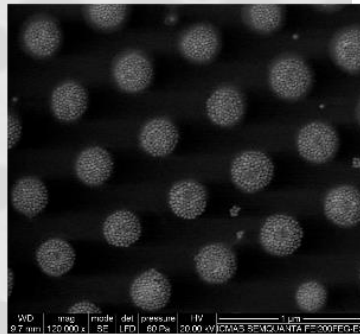
- 50nm Gold nanoparticles: Use of the near fields for SERS



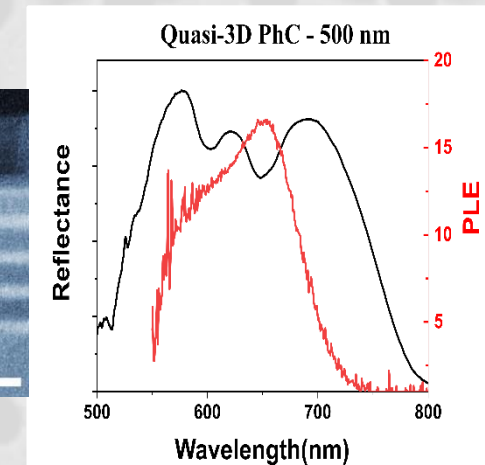
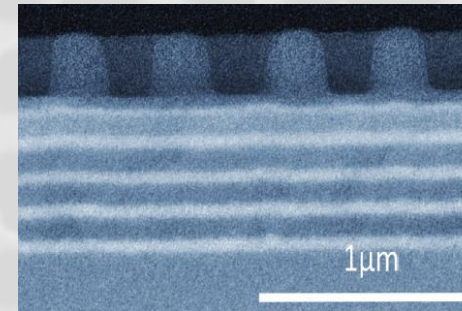
Angew. Chem.  
Int. Ed. 2020, 59,  
17750 – 17756

- CsPbBr<sub>3</sub> NCs: Lattice mode for amplified spontaneous emission

Conti et al.  
to be  
submitted



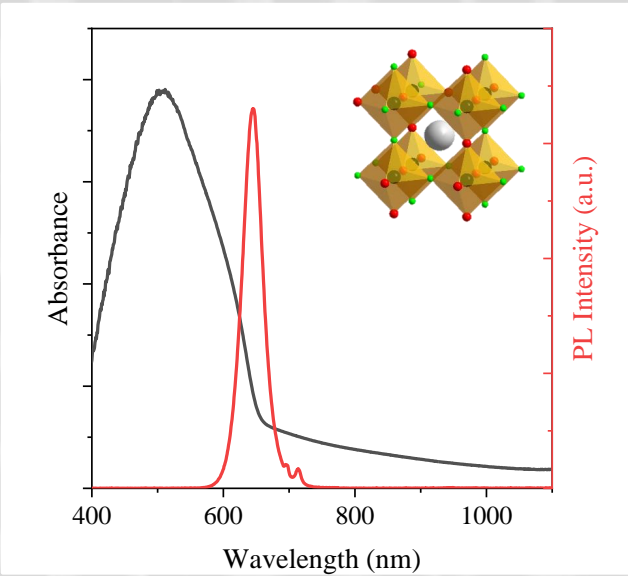
- Ag 25nm: use of lattice mode for stimulated emission



Adv. Optical  
Mater. 2022,  
10, 2101324

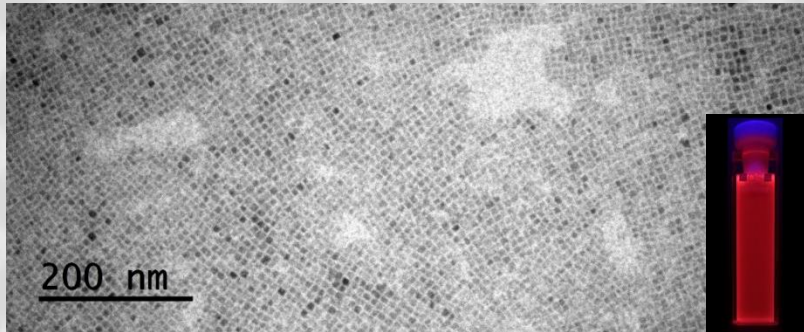
- Enhanced photoluminescence by quasi-3D PhC

# OUR APPROACH



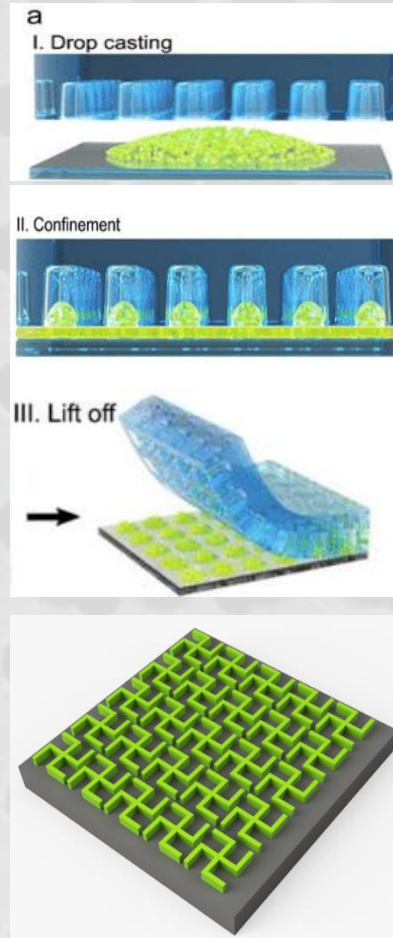
- A = Cs    ● X = Br
- B = Pb    ● X = I

- PL peak at 640nm
- FWHM ~ 40nm



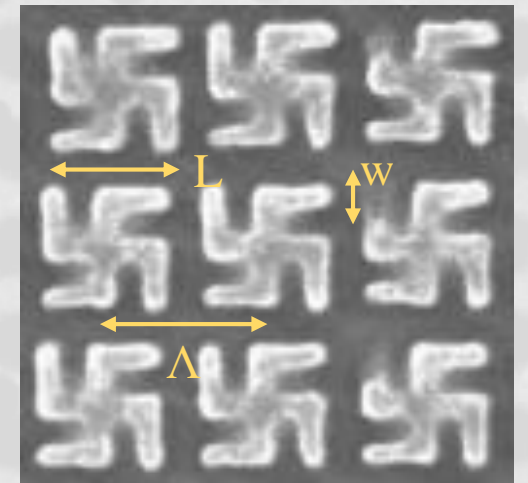
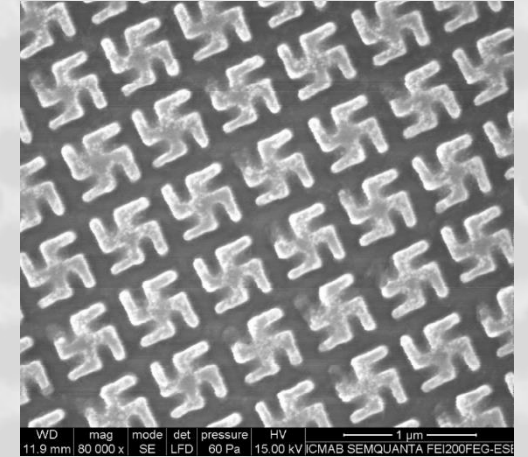
- CsPbBr<sub>3</sub>I<sub>2</sub> cubic nanocrystals
- 8-10 nm size

**Nanopt**

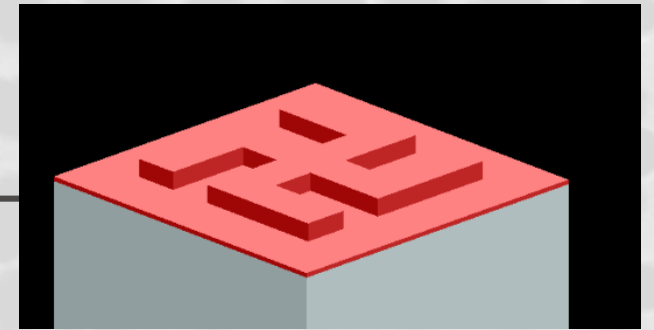


2D-Chiral self-assembly

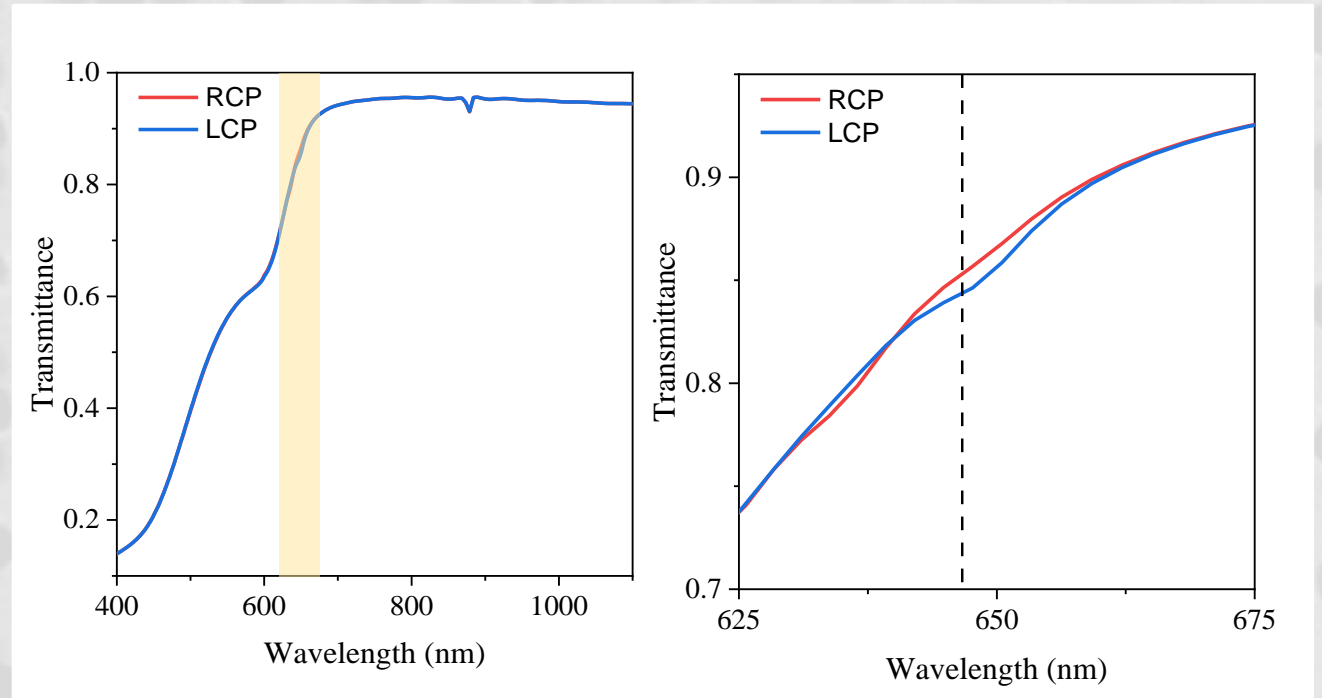
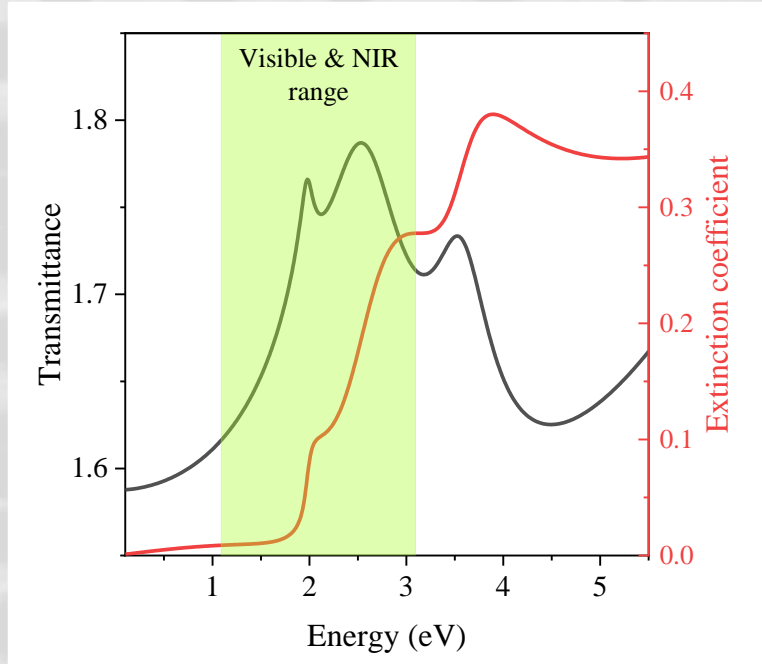
$L = 500\text{nm}$   
 $w = 100\text{nm}$   
 $\Lambda = 600\text{nm}$   
 $h = 125\text{nm}$



# OPTICAL DESIGN



- Refractive index 1.6-1.8
- $E_{\text{gap}} = 2\text{eV}$



- Uniform monolayer
- Mixture between NCs and air

- Small CD in transmittance
- Low gammadion height

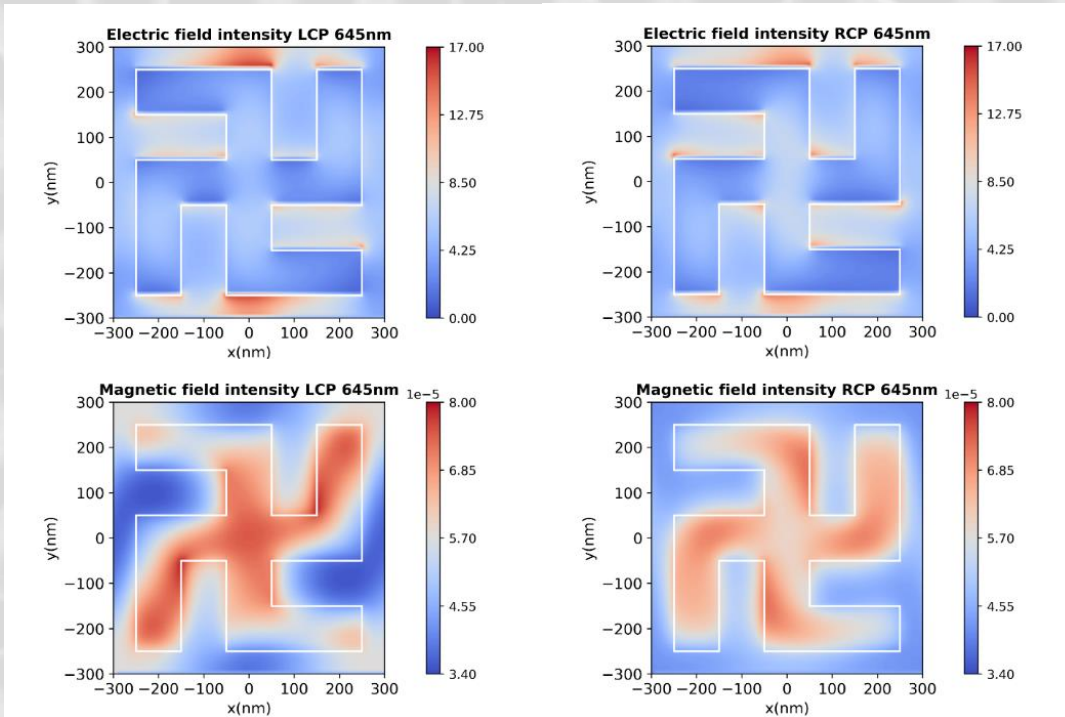


# OPTICAL DESIGN

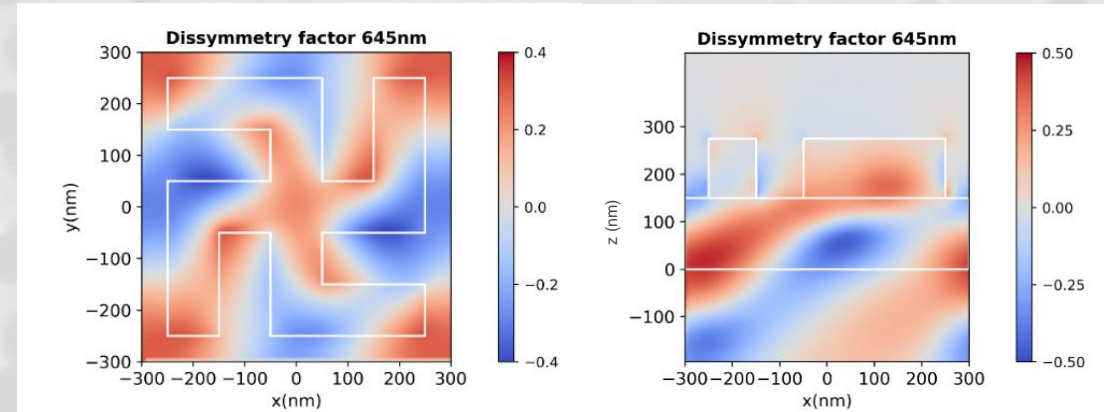
LCP ↻

RCP ↻

Electric field  
Magnetic field



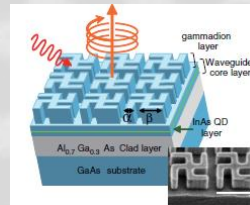
$$I = \frac{1}{2} \mu H^2 = \frac{1}{2} \epsilon_0 E^2 \quad g_{lum} = 2 \cdot \frac{I_{LCP} - I_{RCP}}{I_{LCP} + I_{RCP}}$$



## Waveguide and gammadion coupling

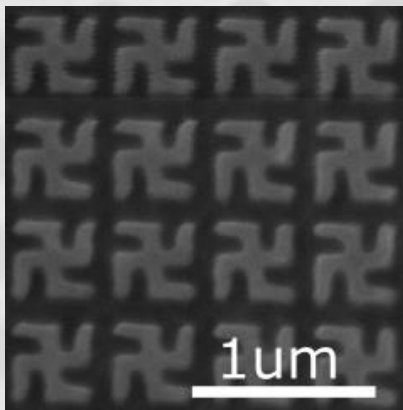
- QDs in waveguide layer and gammadion structure show strong CD
- Related works assign CDPL to Fabry-Pérot interference to waveguiding layer

Nanopt 



PRL 106, 057402 (2011)

# OPTICAL CHARACTERIZATION SETUP



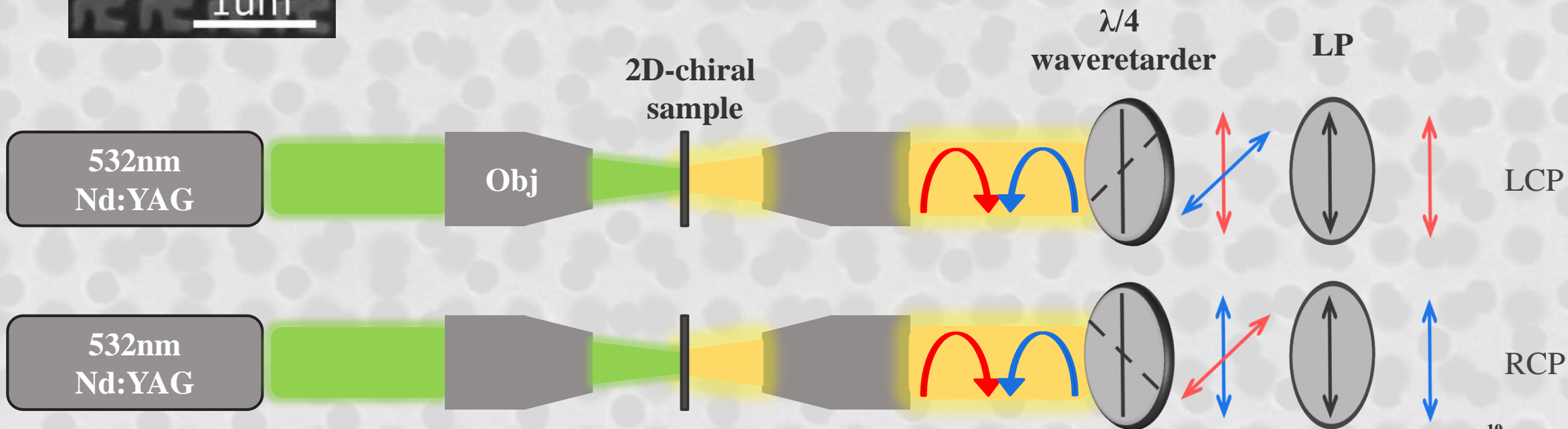
- 4-fold symmetry  $\rightarrow C_4$
- Glass substrate breaks symmetry in z

$$I^j = \iint dx dy (|E_x^j|^2 + |E_y^j|^2),$$

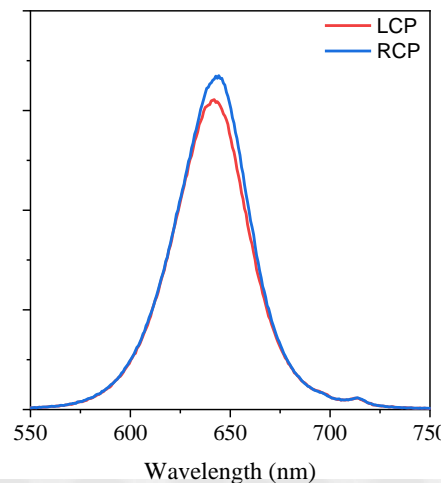
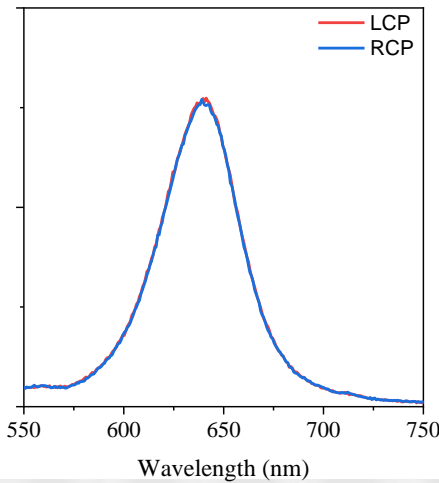
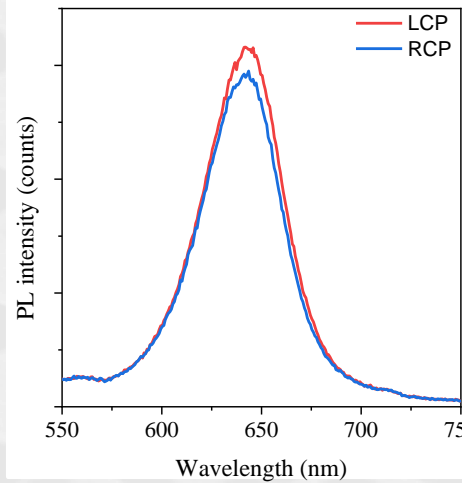
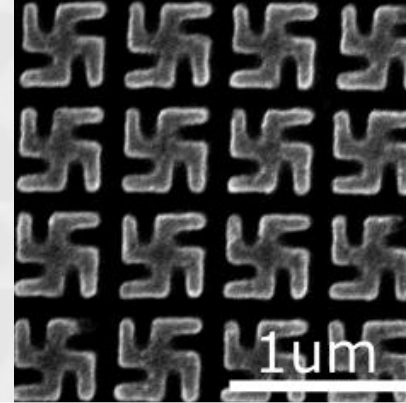
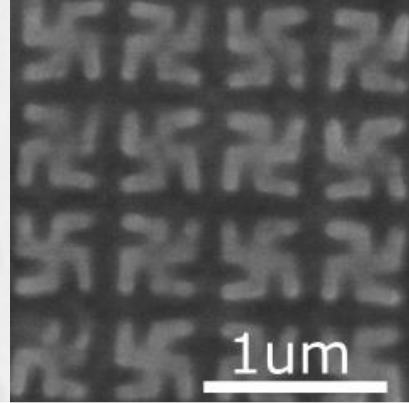
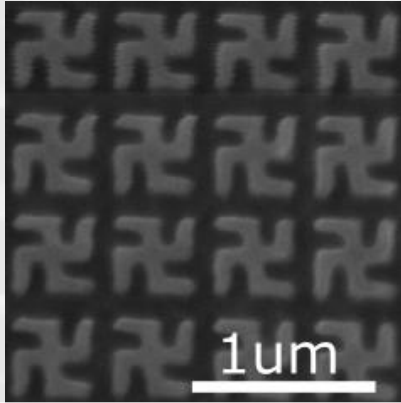
$$E = E_x \quad \frac{\pi}{2} \longrightarrow \quad E = E_y \quad I_x = I_y$$

$$\rho_{xy} = (I^x - I^y)/(I^x + I^y) = 0,$$

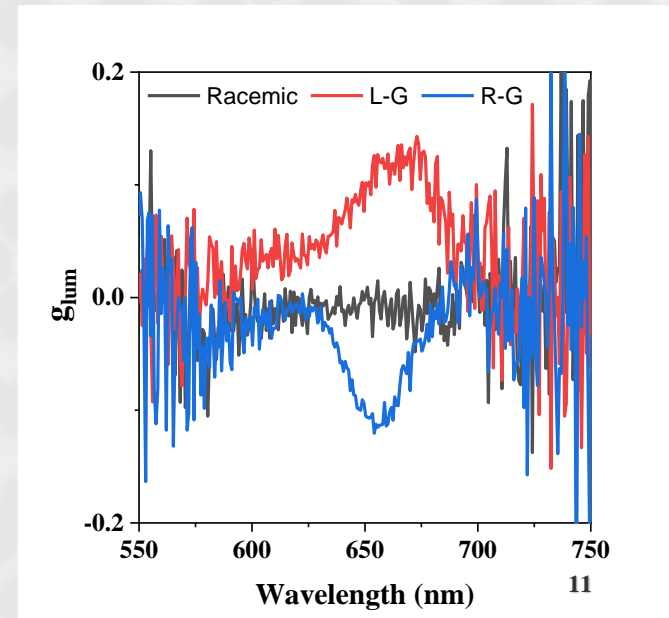
No LP emission  
dissymmetry



# 2D-CHIRAL PHOTONIC STRUCTURES

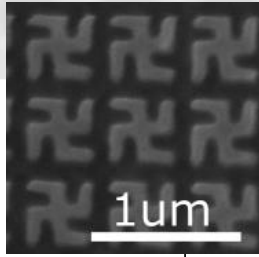
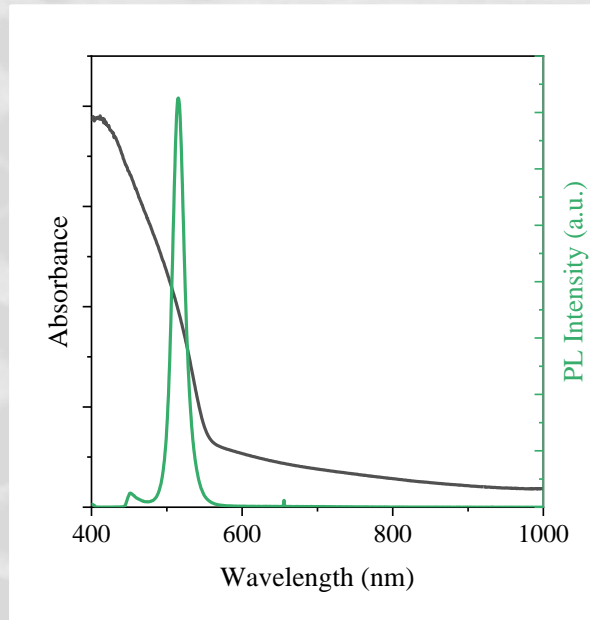


- Coupled emission redshifted
- $g_{lum} \sim 0.1$

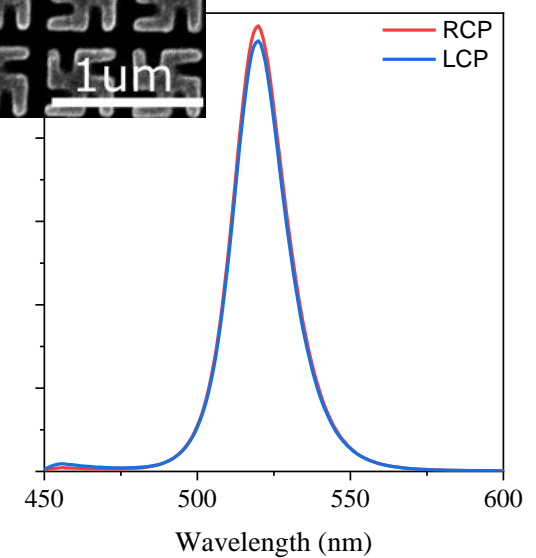
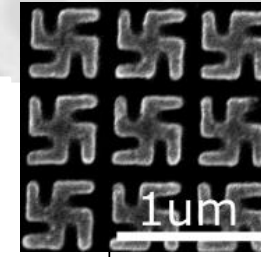
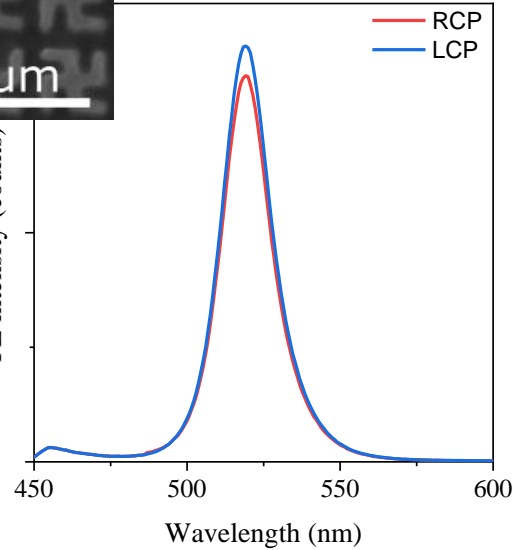


# VERSATILE FABRICATION FOR DIFFERENT EMITTERS

$\text{CsPbBr}_3$

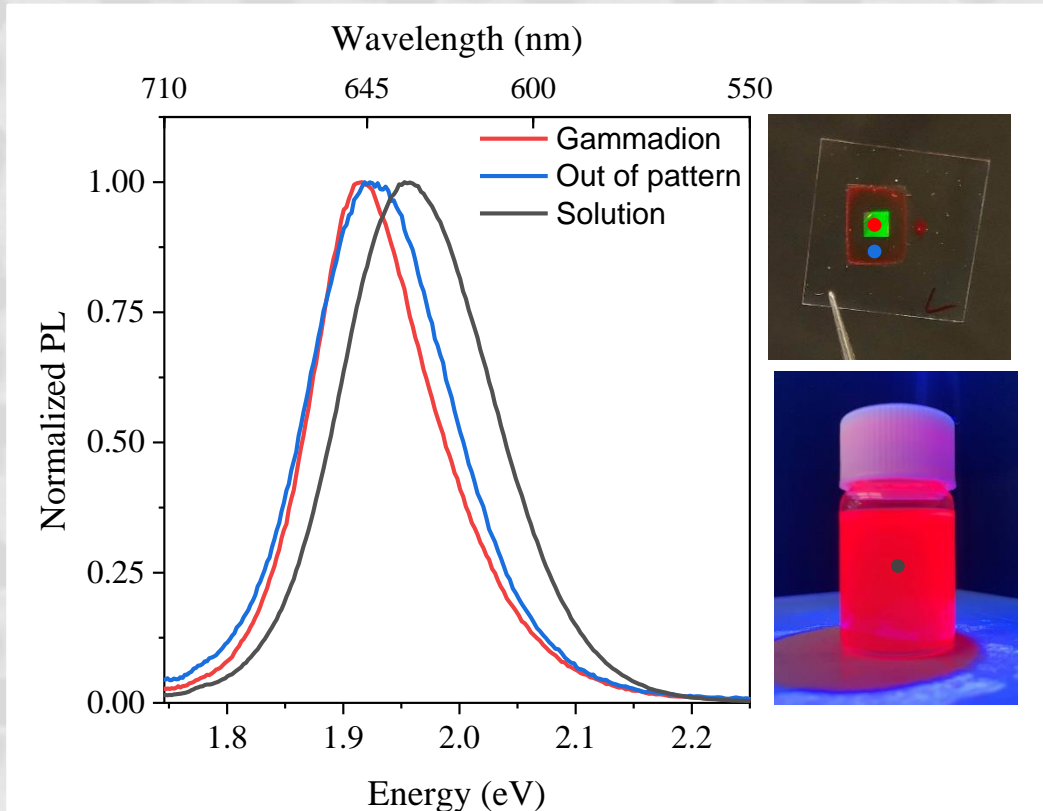


PL intensity (counts)



- Similar dissymmetry PL factor
- Optical activity intrinsic to the chiral photonic structure

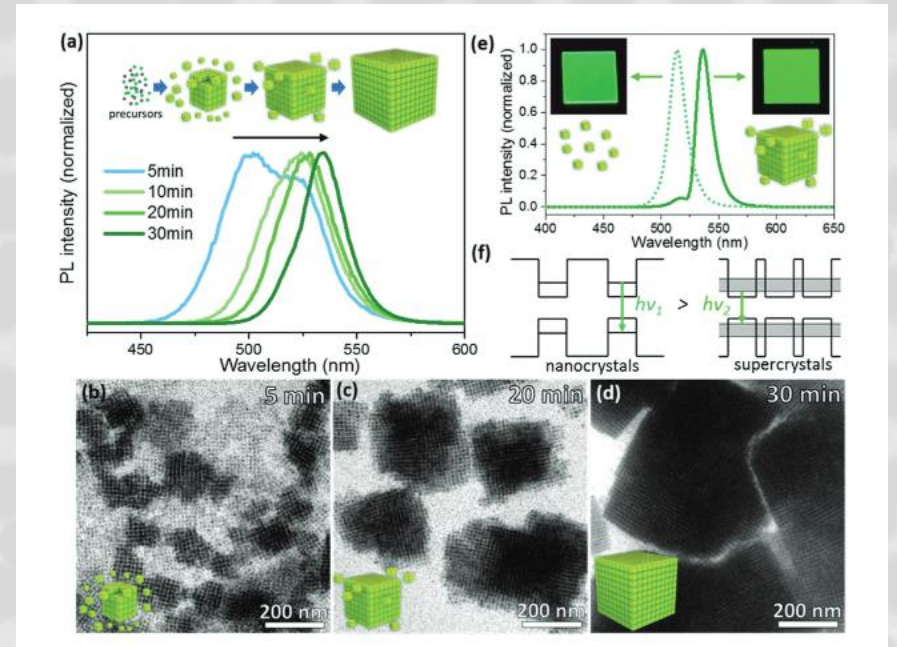
# PHOTOLUMINESCENCE AND EXCITON COUPLING



Transition from  
single nanocrystal  
to supercrystal

Hypothesis

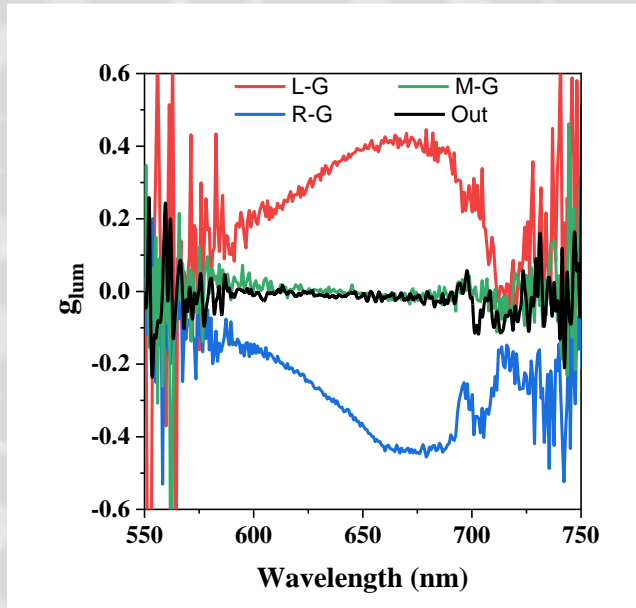
Wavefunction  
and exciton  
coupling



Adv.Mater.2018, 30, 1801117

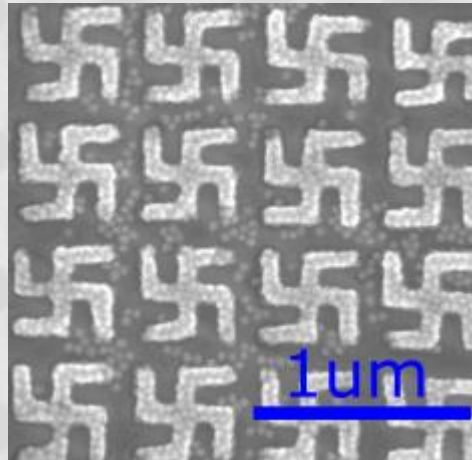
# FUTURE PROSPECTS

## Thickness gradients



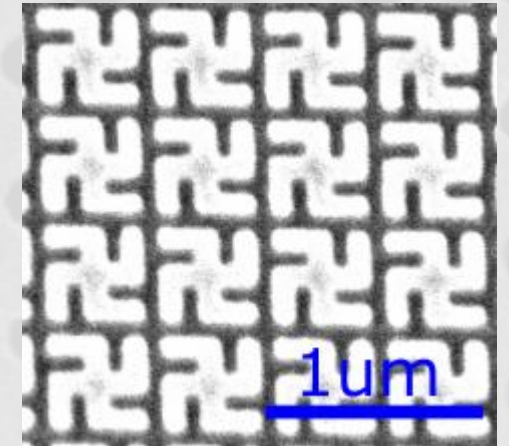
$g_{lum} \sim 0.4$

## Ag NPs on doped waveguide



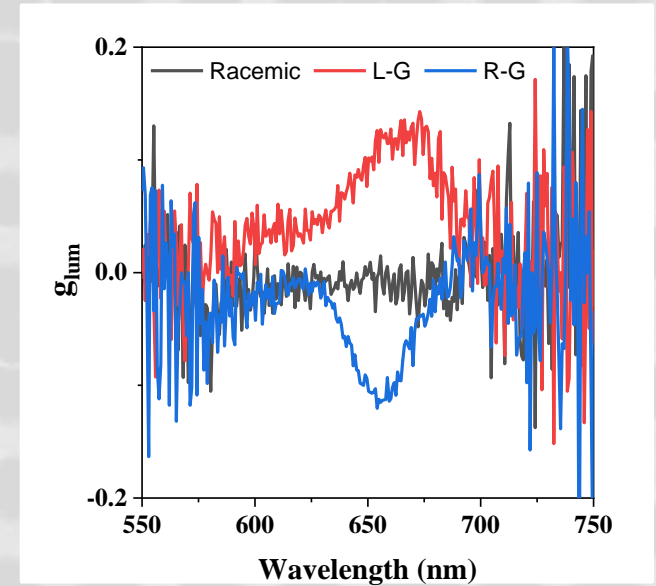
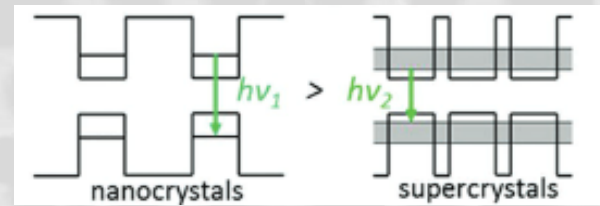
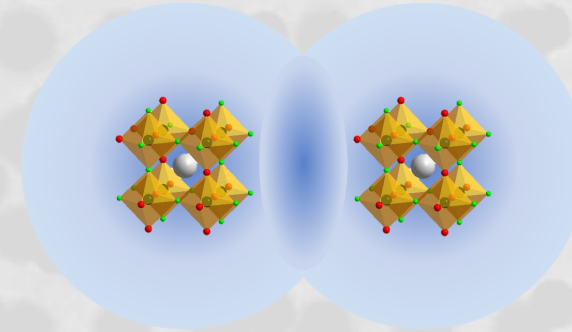
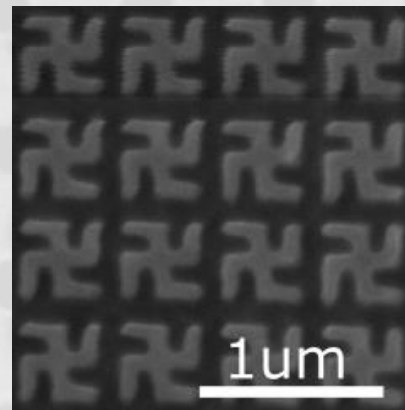
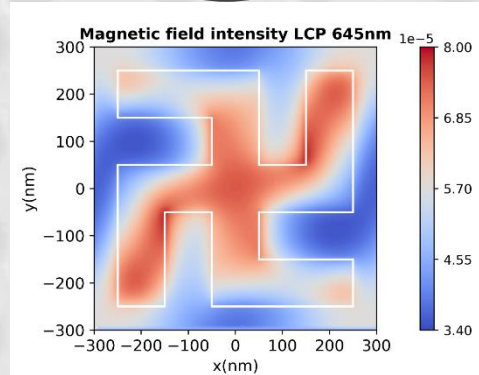
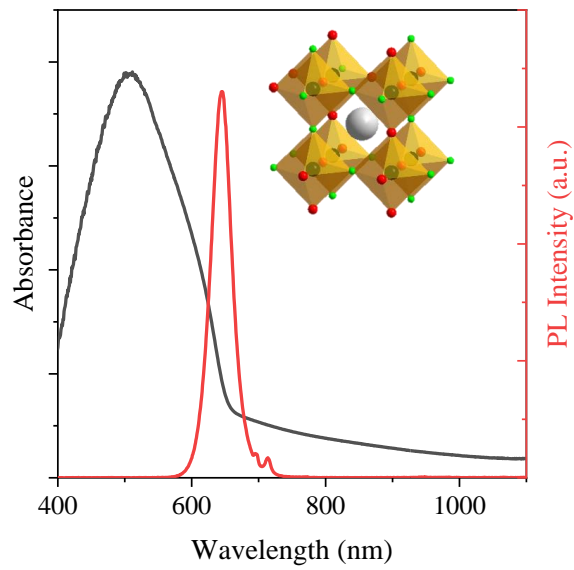
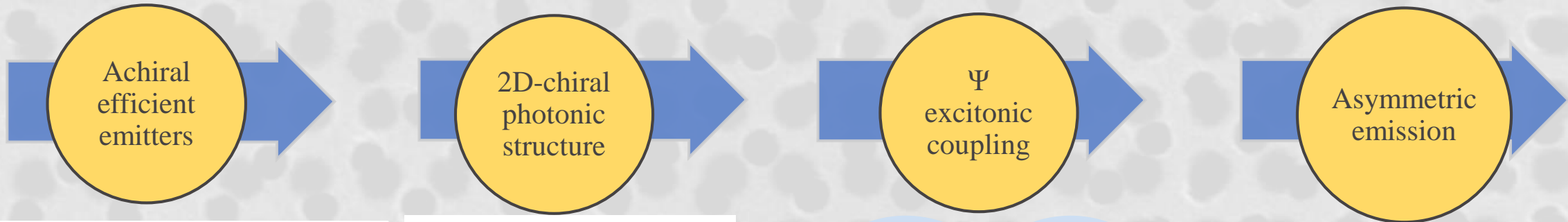
Plasmonic nanoparticles

## Structured doped polymer



Plasmonic (Au) and high refractive index (TiO<sub>2</sub>) coatings

# SUMMARY



# ACKNOWLEDGEMENTS & FUNDING



<https://www.icmab.es/>



<https://nanopto.icmab.es/>



<https://enlightment.icmab.es/>  @Enlightment\_BCN



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