

Control of light-matter interaction in 2D Van der Waals materials

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EFRI 2DARE
ECCS 1509551
DMR 1120923
MRSEC: PAS³

ICTS – OQS, 2017

Team



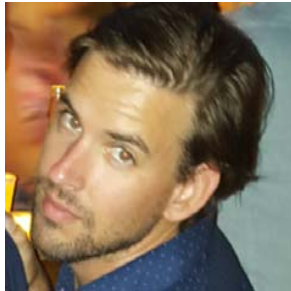
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Jie Gu
Grad Student – CUNY



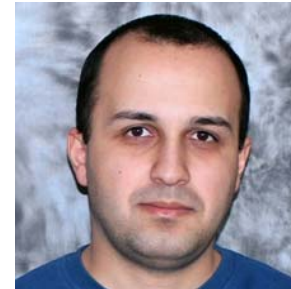
Dr. Biswanath Chakraborty
Postdoc– CCNY



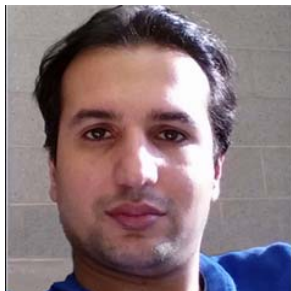
Ryan Considine
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Michael Dollar
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CUNY



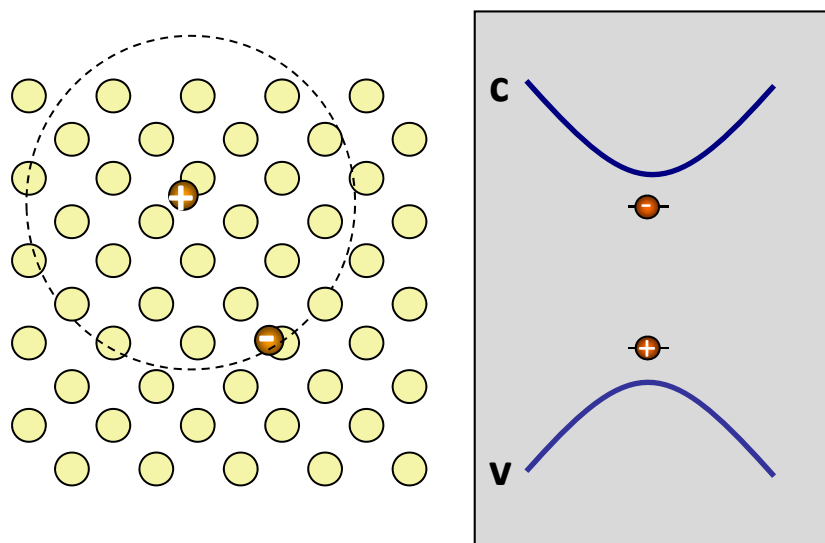
Prof. Stéphane Kéna-Cohen
École Polytechnique de Montréal



Prof. Yi-Hsien Lee
NTHU

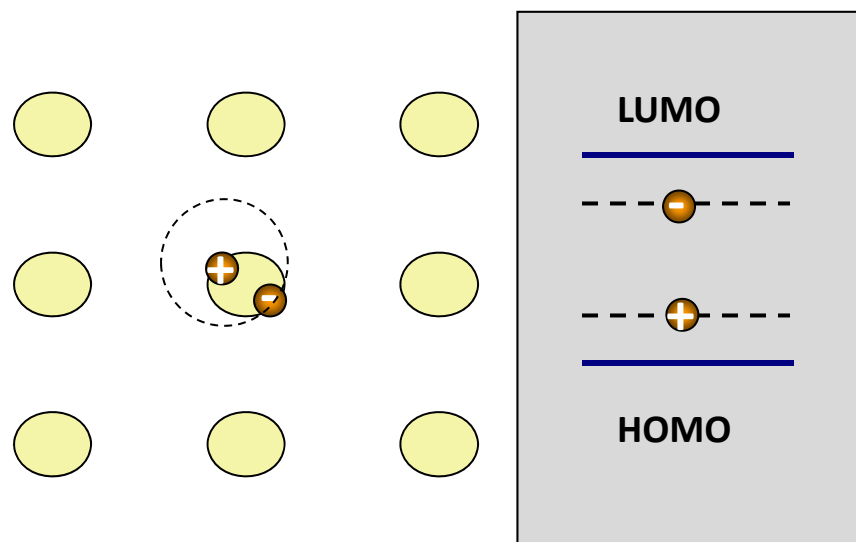
Excitons

Wannier-Mott excitons



Inorganic Materials

Frenkel excitons



Organic Materials

Excitons

Wannier-Mott excitons Inorganic Materials

- Large exciton radius $\sim 100\text{\AA}$
- Small binding energy (10-60meV)
- Long range ballistic transport
- Condensation (Low T)
- Polariton Lasing ($\sim$ Room T)
- Small saturation density

Frenkel excitons Organic Materials

- Localized at molecular site
- Large binding energy (0.5 – 1 eV)
- Hopping transport
- Forster / Dexter energy transfer
- Polariton lasing (Room T)
- Large saturation density

Transition Metal Di-Chalcogenides

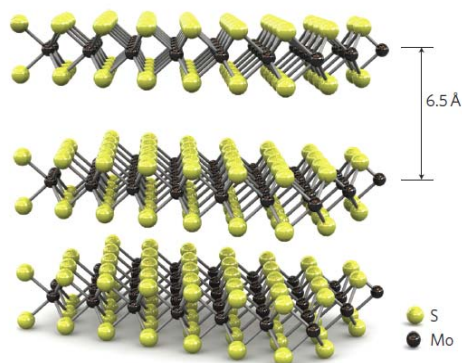
2D materials with optical bandgap

H	MX₂ M = Transition metal X = Chalcogen M = Mo and W, X = S, Se and Te.																He
Li	Be											B	C	N	O	F	Ne
Na	Mg	3	4	5	6	7	8	9	10	11	12	Al	Si	P	S	Cl	Ar
K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr
Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe
Cs	Ba	La-Lu	Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po	At	Rn
Fr	Ra	Ac-Lr	Rf	Db	Sg	Bh	Hs	Mt	Ds	Rg	Cn	Uut	Fl	Uup	Lv	Uus	Uuo

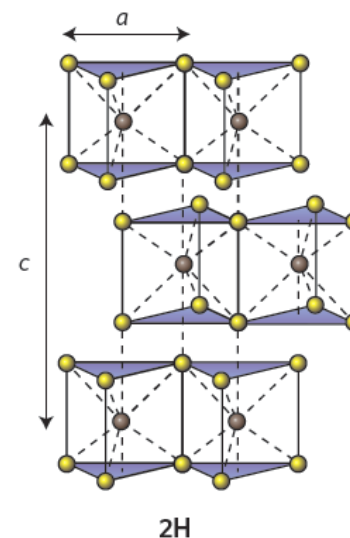
MoS_2
1L : 1.8 eV, Bulk: 1.2 eV

WS_2
1L: 2.1 eV, Bulk: 1.4 eV

Chhowalla, M. et al (2013), *Nat. Chem.*, 5, 263–275.



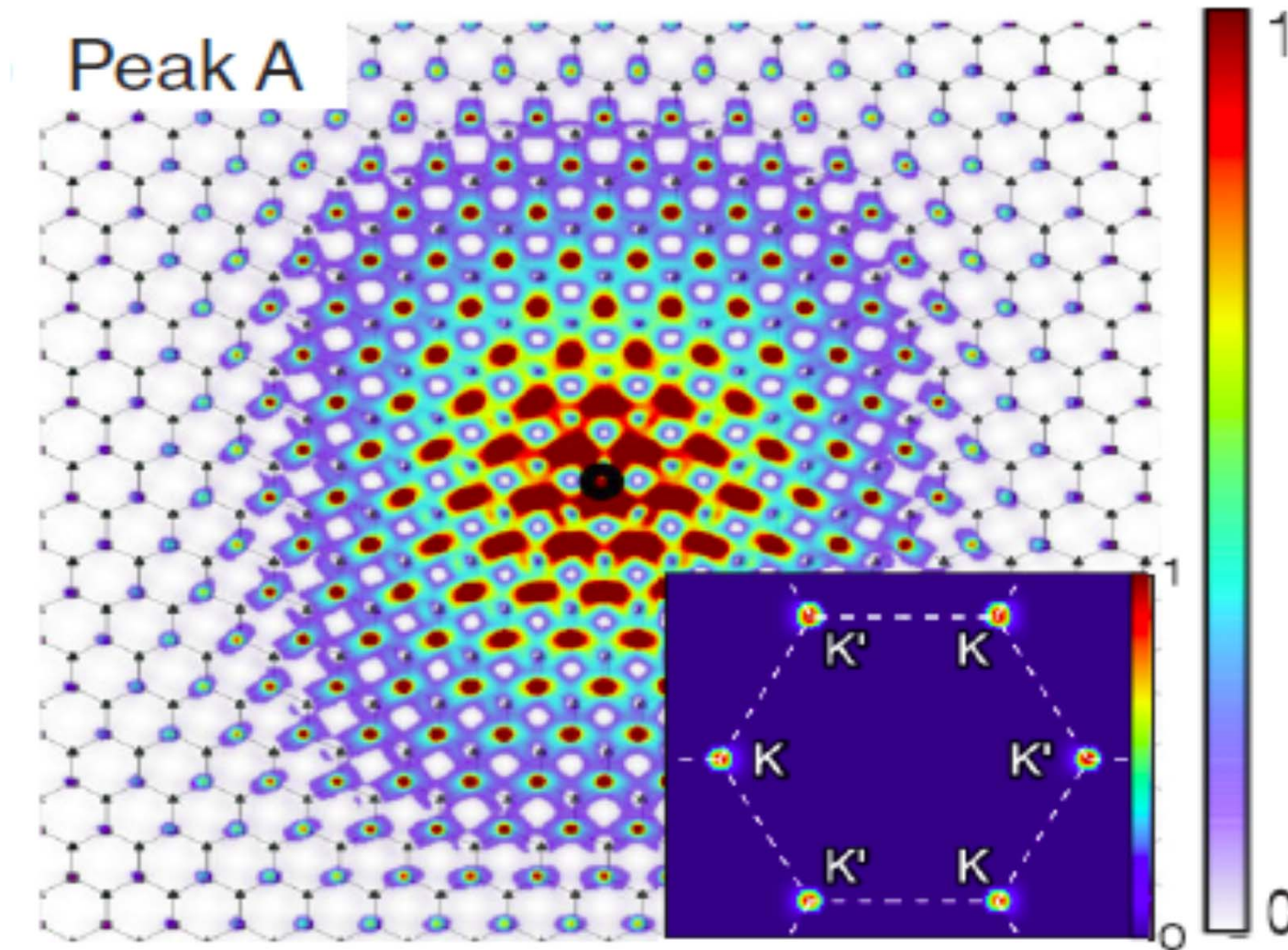
Radisavljevic, B. et al (2011), *Nat. Nanotechnol.*, 6, 147–150.



Wang, Q. H. et al (2012), *Nat. Nanotechnol.*, 7, 699–712.

Excitons in 2D TMDs: Best of both worlds?

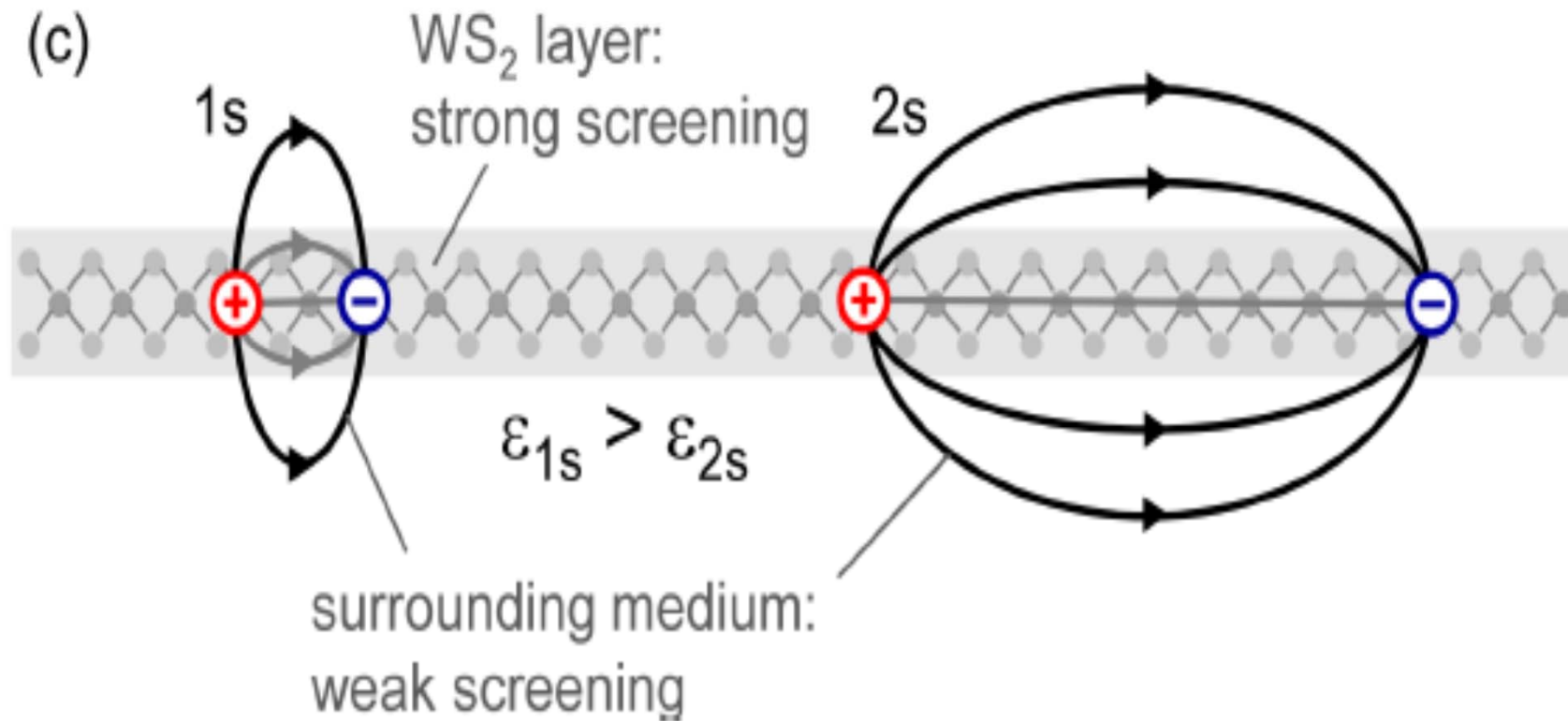
Bohr Radius ~ 1 nm (MoS_2)



D. Y. Qiu et al. PRL 111, 216805 (2013)

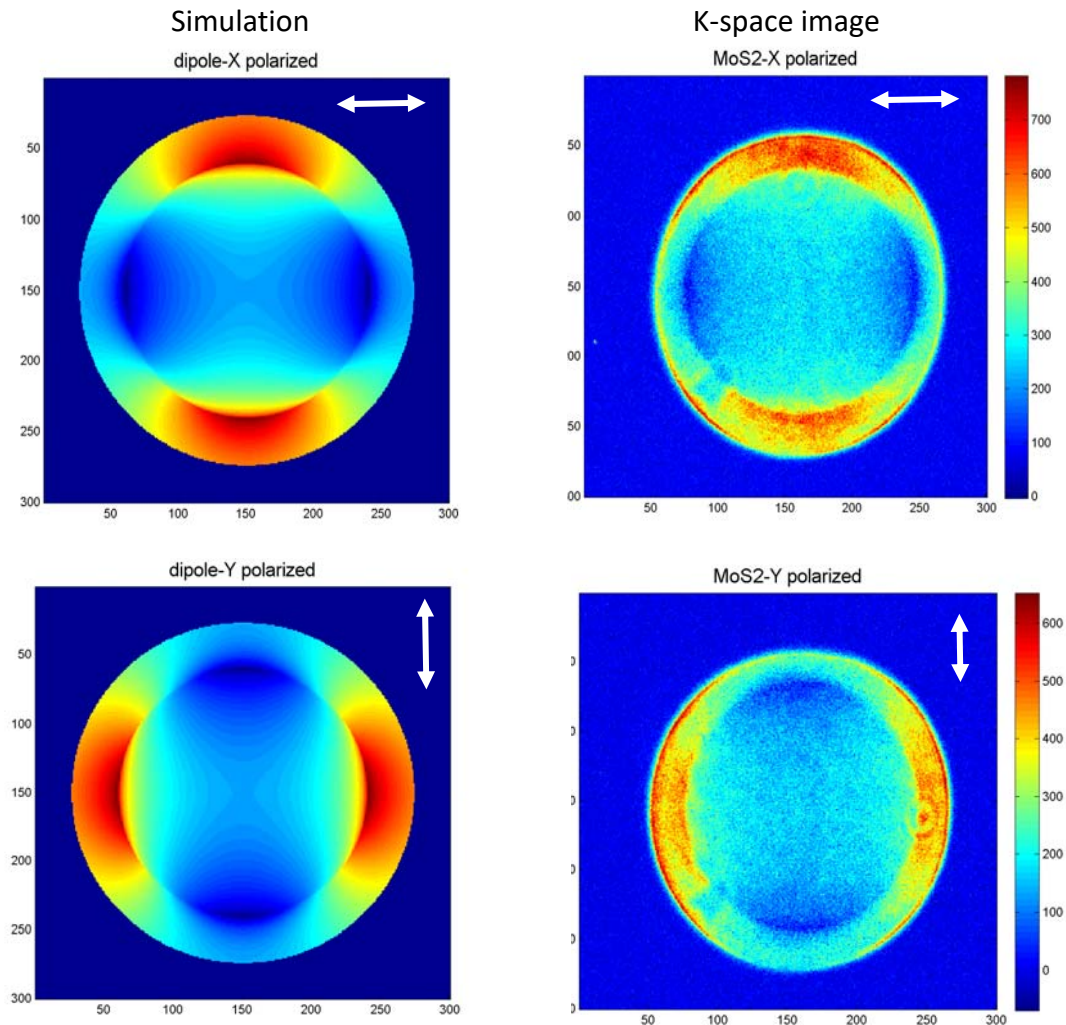
Excitons in 2D TMDs: Best of both worlds?

Binding Energy (0.3-0.5 eV)



In-plane Dipoles

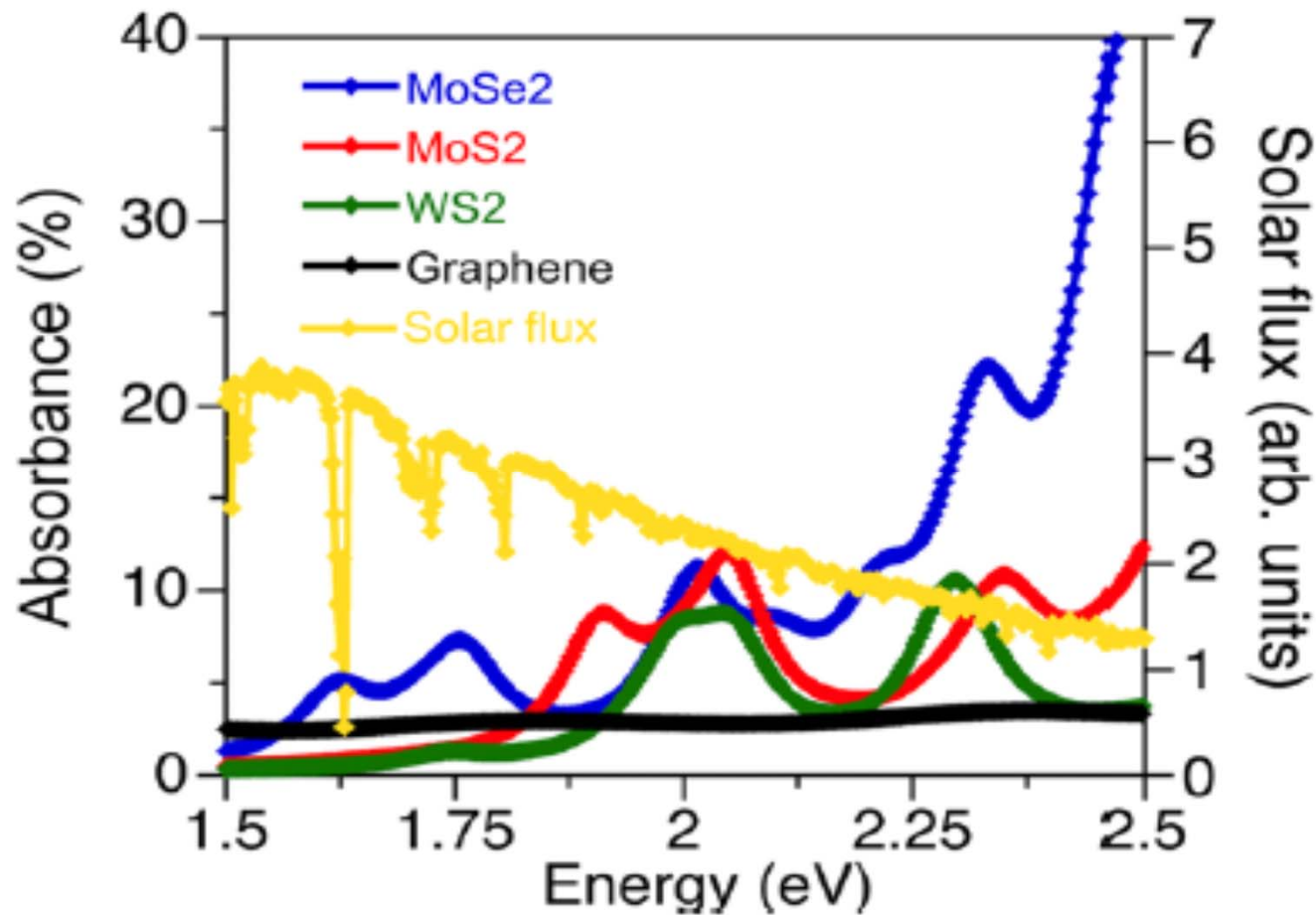
2D dipole orientation.



J. Gu...VMM (unpublished)

Also: J. Schuller et al., Nat. Nanotech **8** 271 (2013)

Large oscillator strength

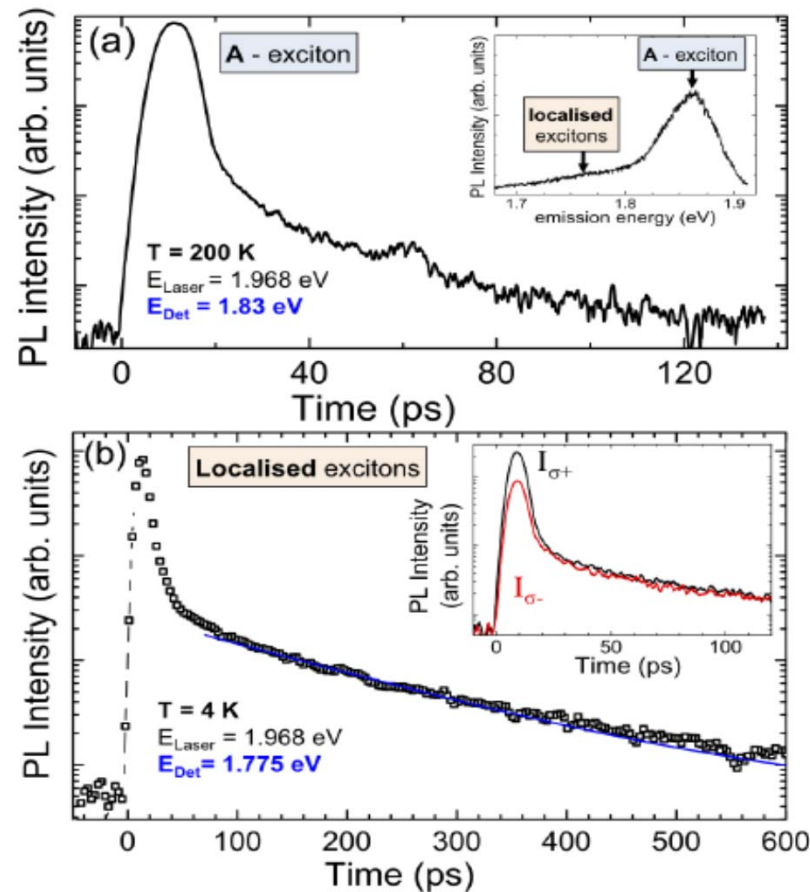


1 ML absorbs 5-10% light !

Exciton Lifetime

CVD grown 2D MoS₂ : < 4 ps [Sie et al. arxiv 1312.2918]

Exfoliated 2D MoS₂: 5-6 ps, 125 ps (localized excitons) [Lagarde et al. 1308.0696 (2013)]



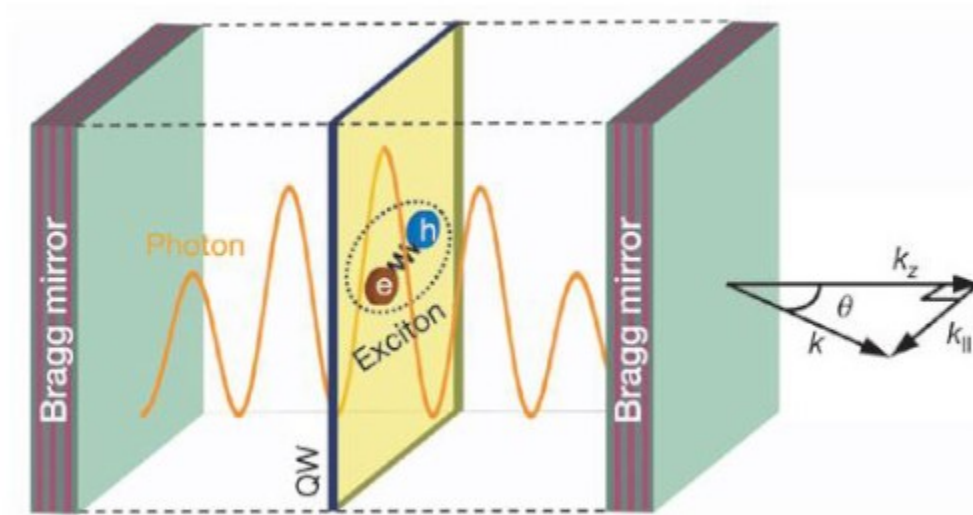
Lagarde et al. *Physical Review Letters* **112**, 047401 (2014)

Mai et al. *Nano Lett.* **14**, 202 (2013)

Wang, G. et al. *arXiv:1402.6009* (2014)

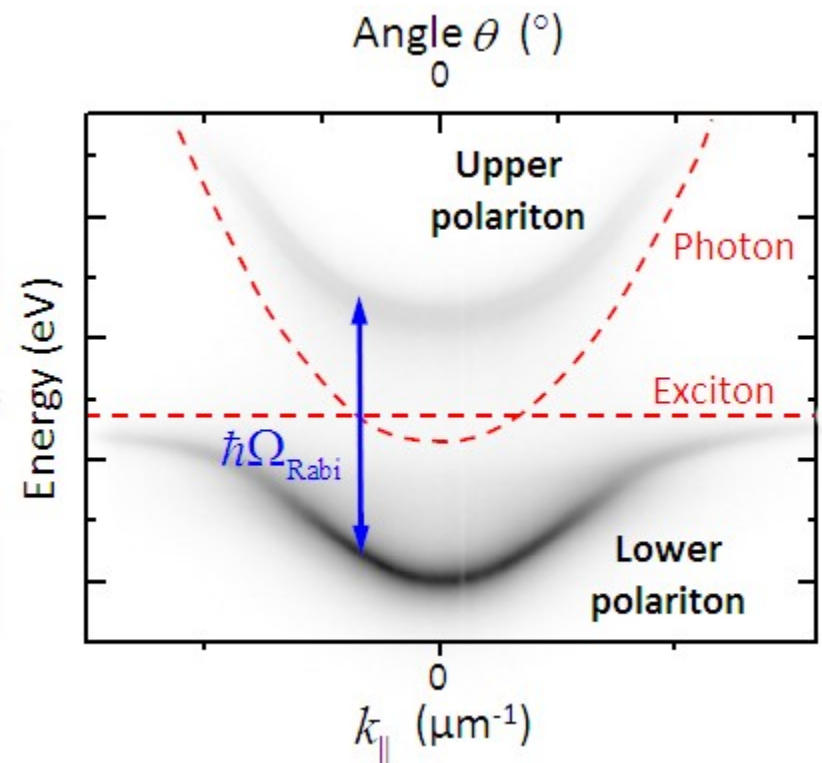
Microcavity Polaritons

(a)



J. Kasprzak. et al(2006), *Nature*, 443(7110), 409–414.

(b)



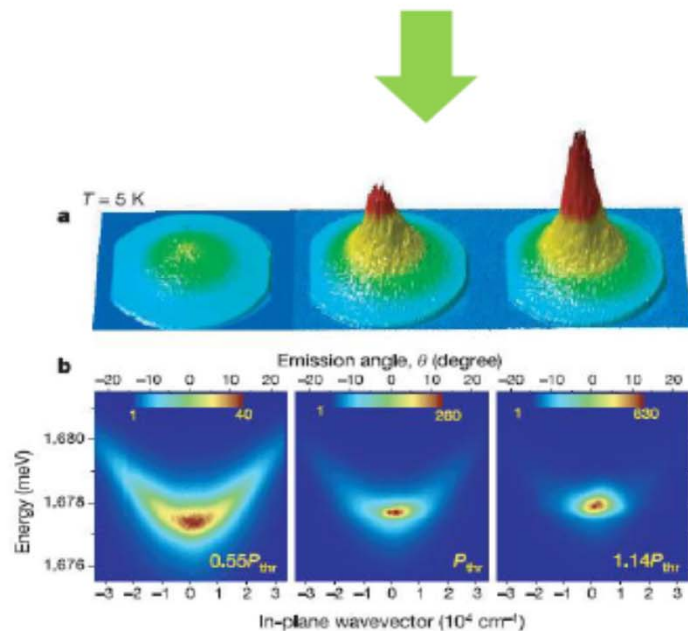
Microcavity Polaritons

Composite bosons: half-exciton half-photon

Excitonic component – Electronic response, scattering and nonlinearity

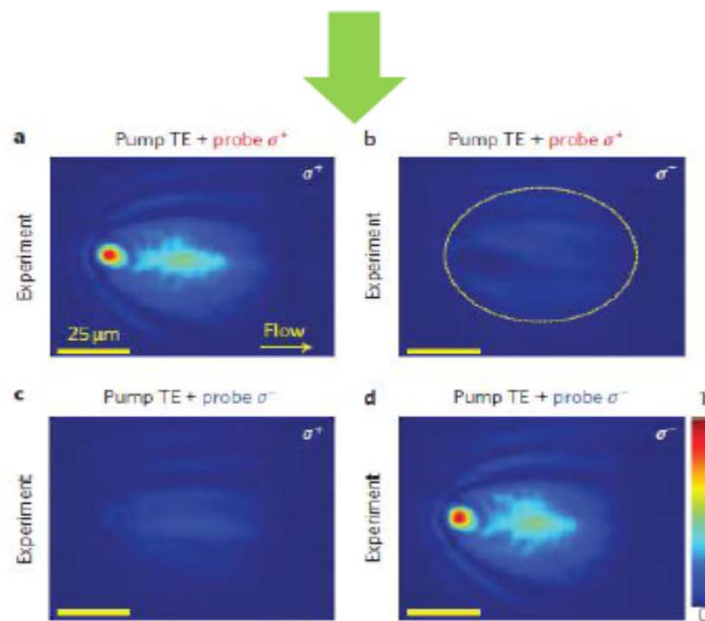
Photonic component – Small mass, spatial coherence and long propagation

Polariton condensation



Kasprzak, J. et al. *Nature* **443**, 409 (2006)
Balili, R., et al. *Science* **316**, 1007 (2007).

Polariton spin switch

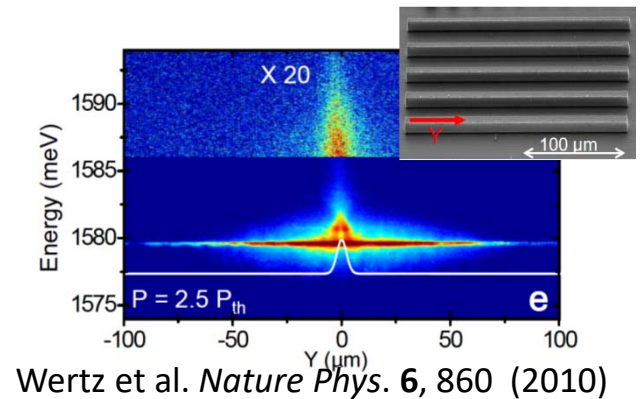


Amo et al. *Nature Photonics* **4**, 361 (2010)

Microcavity Polaritons

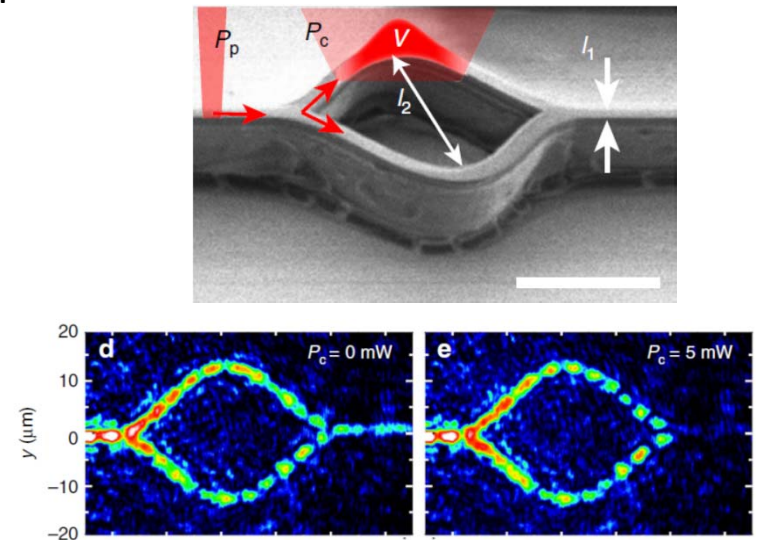
Cryogenic Temperatures

- Condensation – Kasperzak, Balili
- Superfluidity – Amo, Lagoudakis,
- Thermalization of polaritons – Snoke, Pfeiffer, West
- 2DEG polaritons – Imamoglu, Faist
- Polaritons in Intersubband systems – Sirtori, Tredicucci,
- Polariton lattices – Amo, Bloch, Tureci
- Confined polaritons – Wertz, Bloch, Deveaud
- Mach Zehnder/Sagnac interferometer – Amo, Bloch



Room Temperature

- Organic (Holmes, Kena-Cohen, Forrest, Lidzey...)
- Hybrid (Holmes, VMM, Forrest, Lidzey...)
- ZnO (Deng, van Vugt, Trichet, Malpuech, VMM...)
- GaN (Baumberg, Bhattacharya...)
- 2D materials (VMM, Imamoglu, Tartakovskii...)

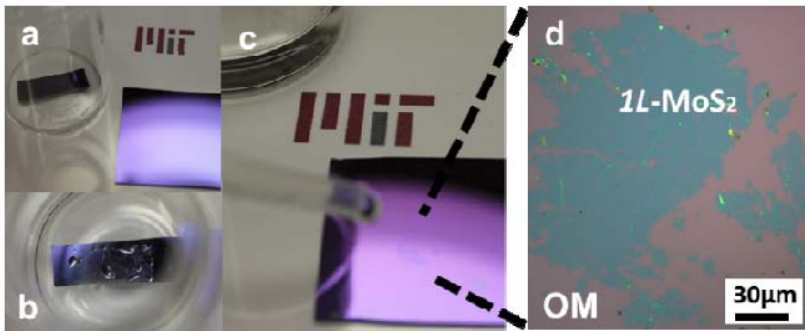
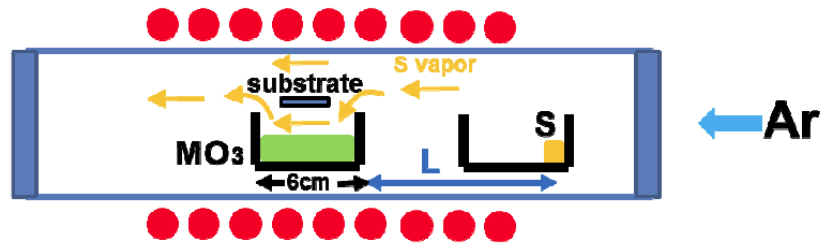


Sturm et al. *Nature Comm.* **5** 3278 (2014)

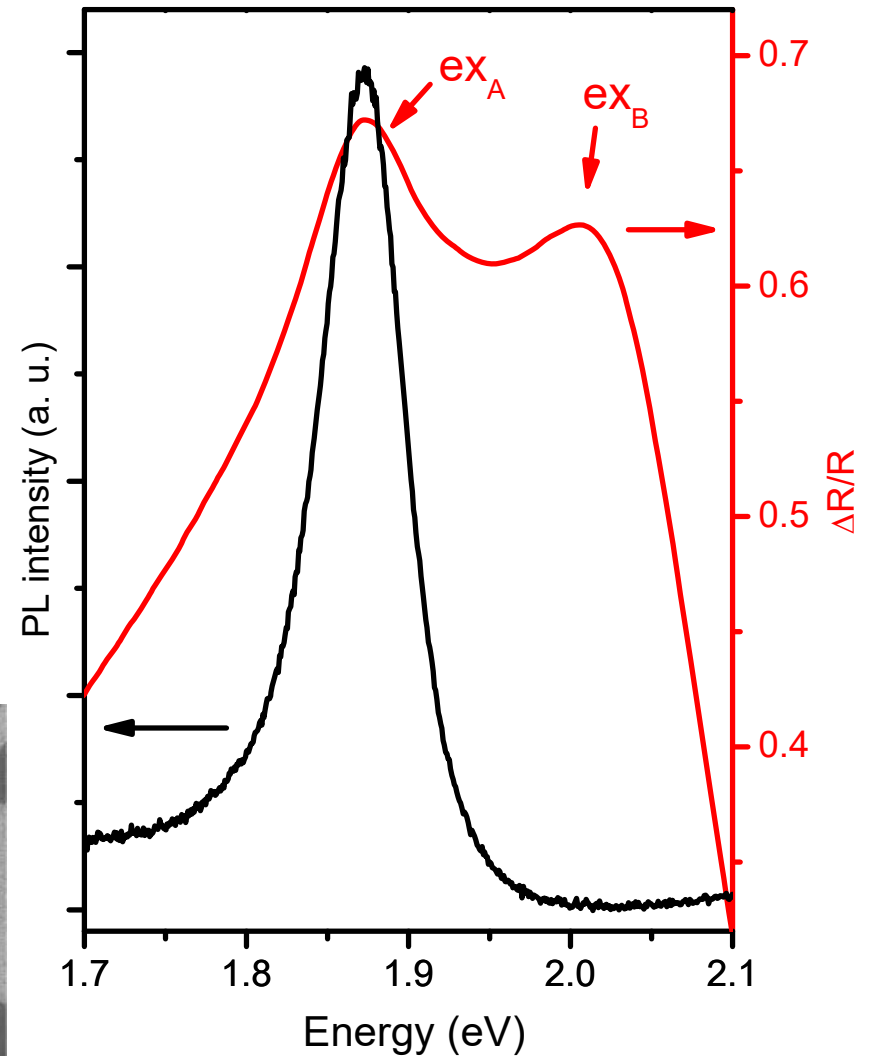
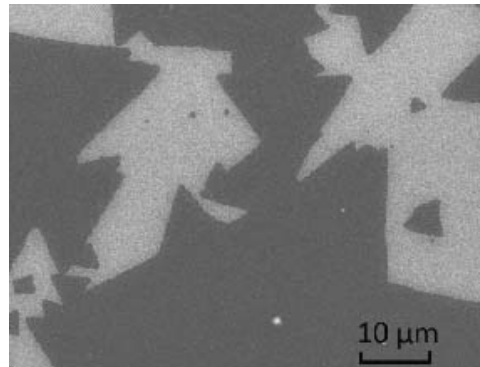
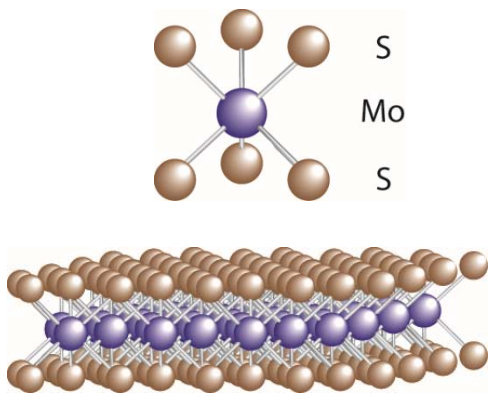
Strong coupling in 2D materials

	Monolayer TMDs	GaAs Quantum wells
Thickness	< 1 nm	> 3 nm
Excitons	Wannier-Mott-like	Wannier-Mott
Binding energy	$\sim 0.3 - 0.5$ eV	~ 10 meV
Bohr radius	~ 2 nm	~ 10 nm
Oscillator strength	High	Low
Mobility	~ 400 cm ² /V·s	$\sim 10^3$ cm ² /V·s

CVD grown MoS₂ Monolayer

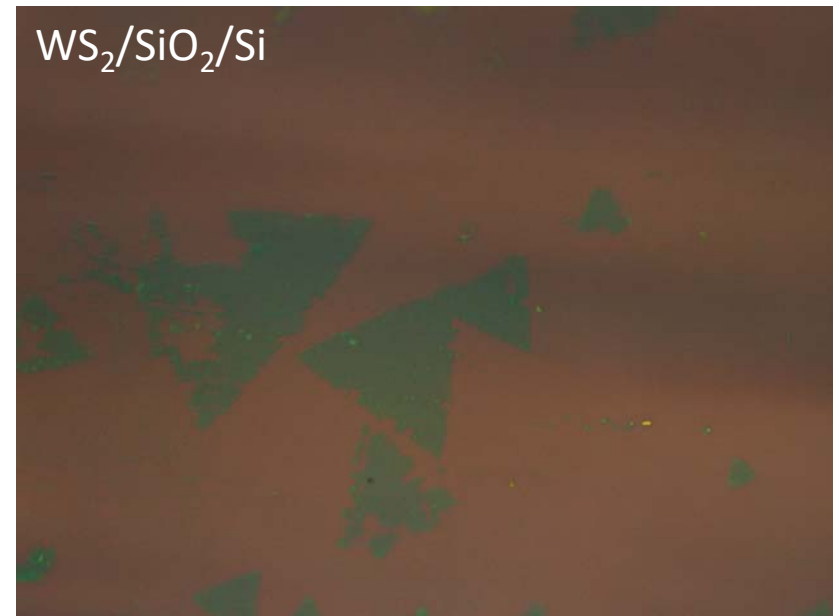
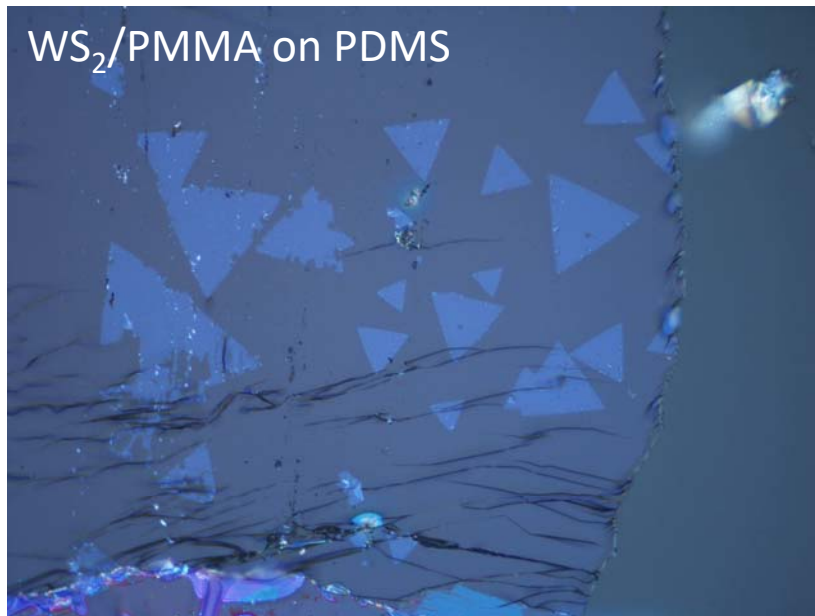


Lee, Y.-H. *et al* (2013), *Nano Lett.* **13**, 1852–1857.



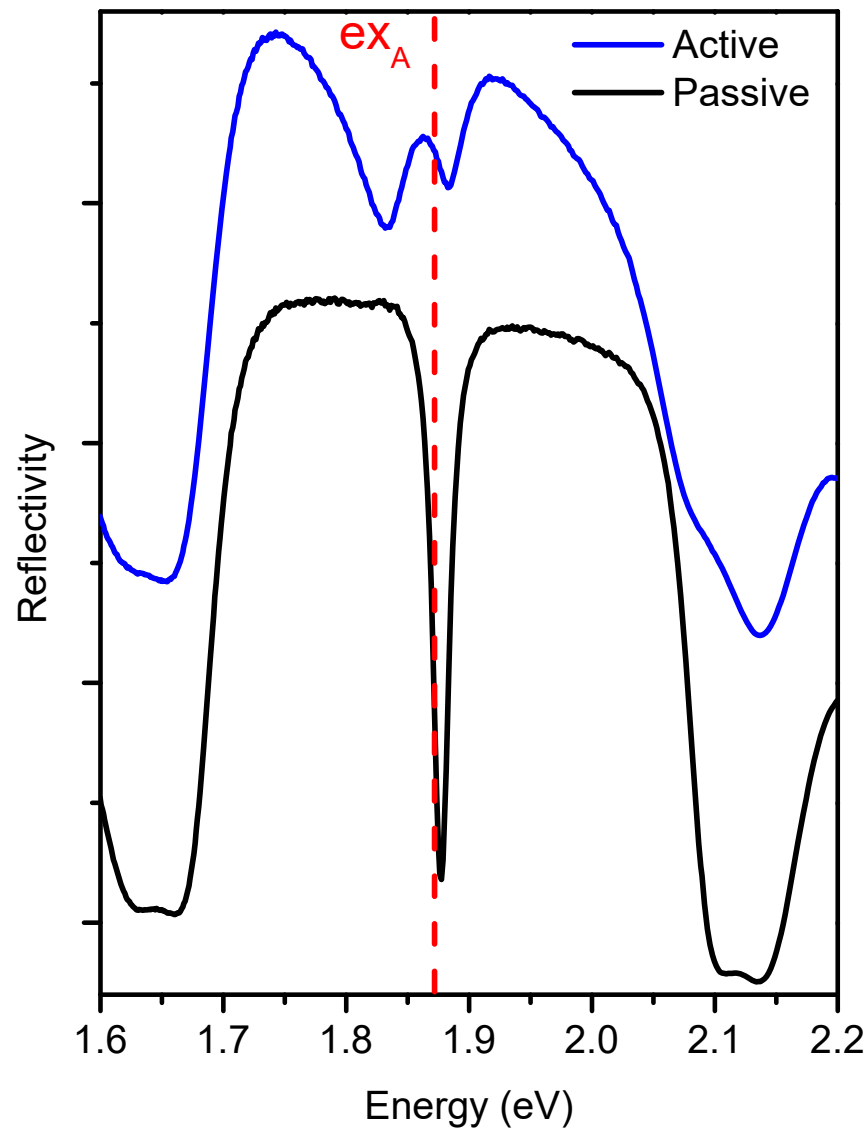
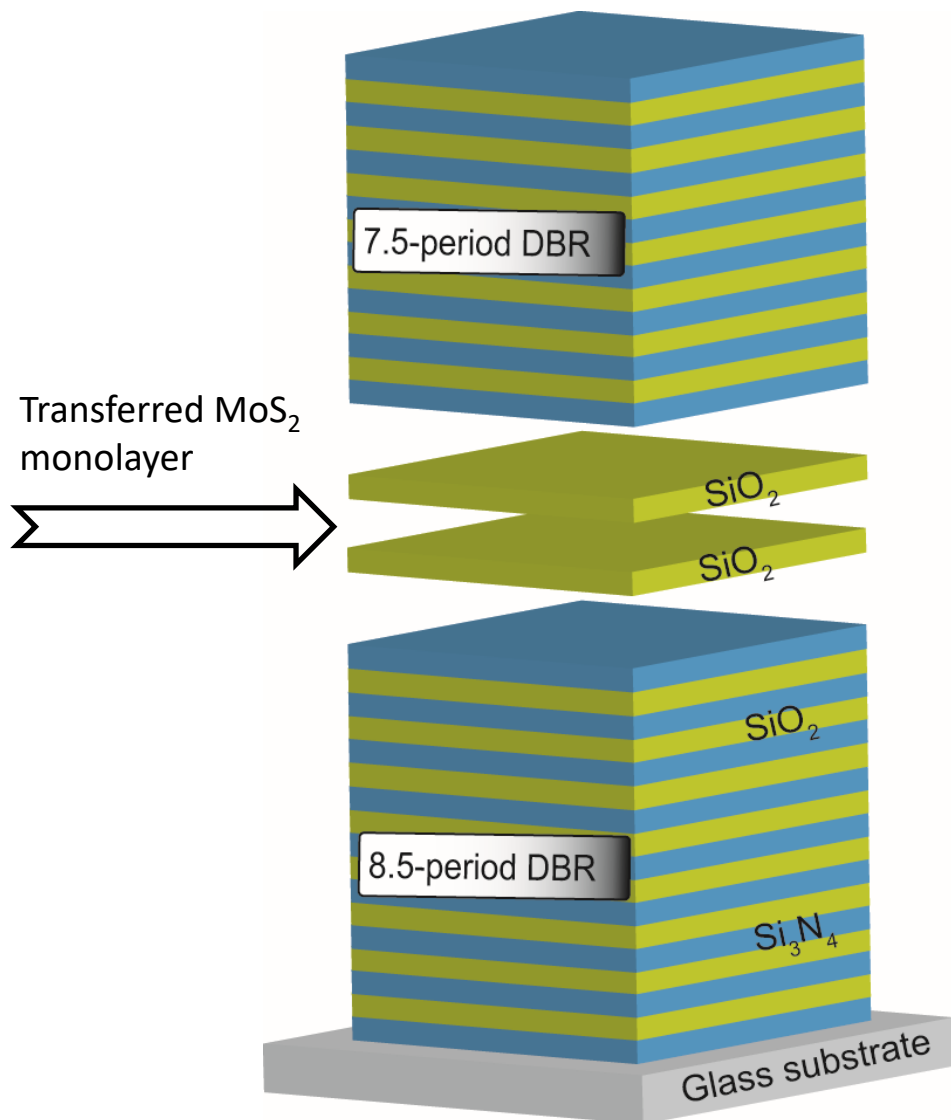
Growth of 2D Materials by Y. H. Lee's group – NTHU, Taiwan

Transfer of CVD grown 2D layers

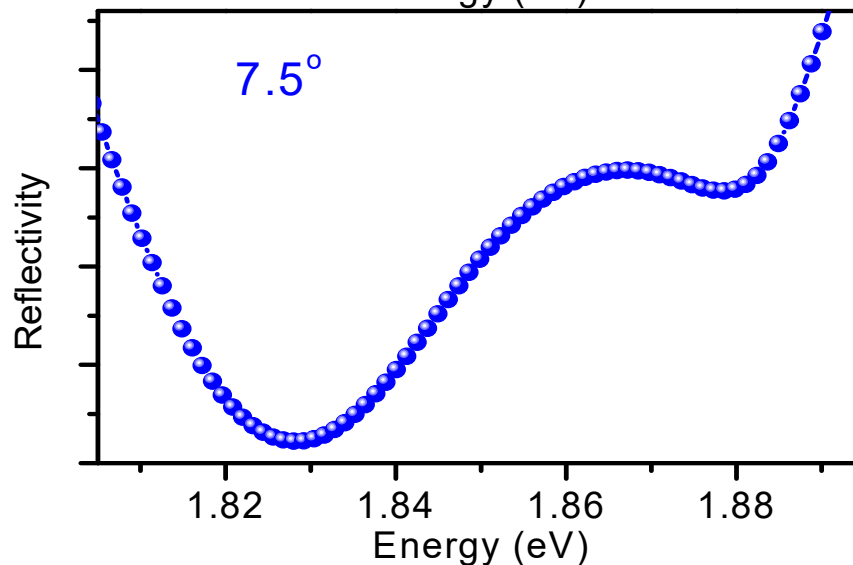
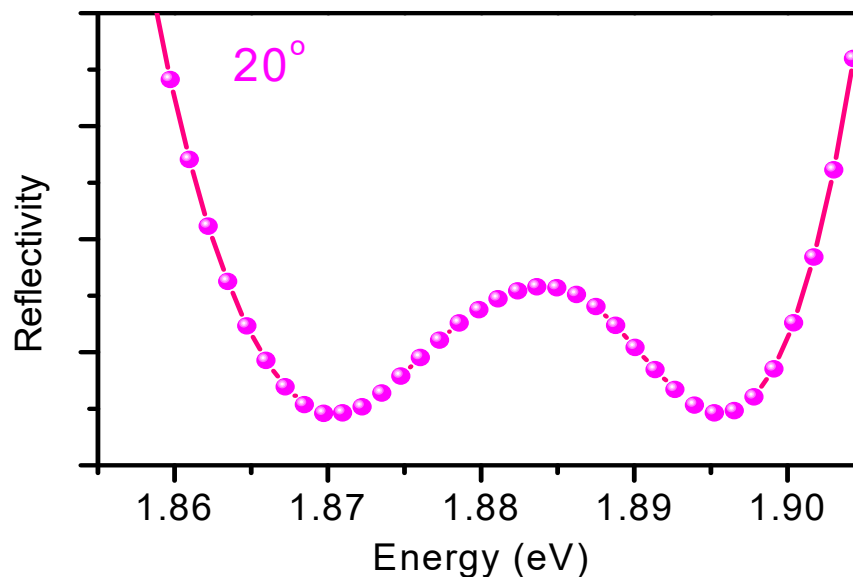
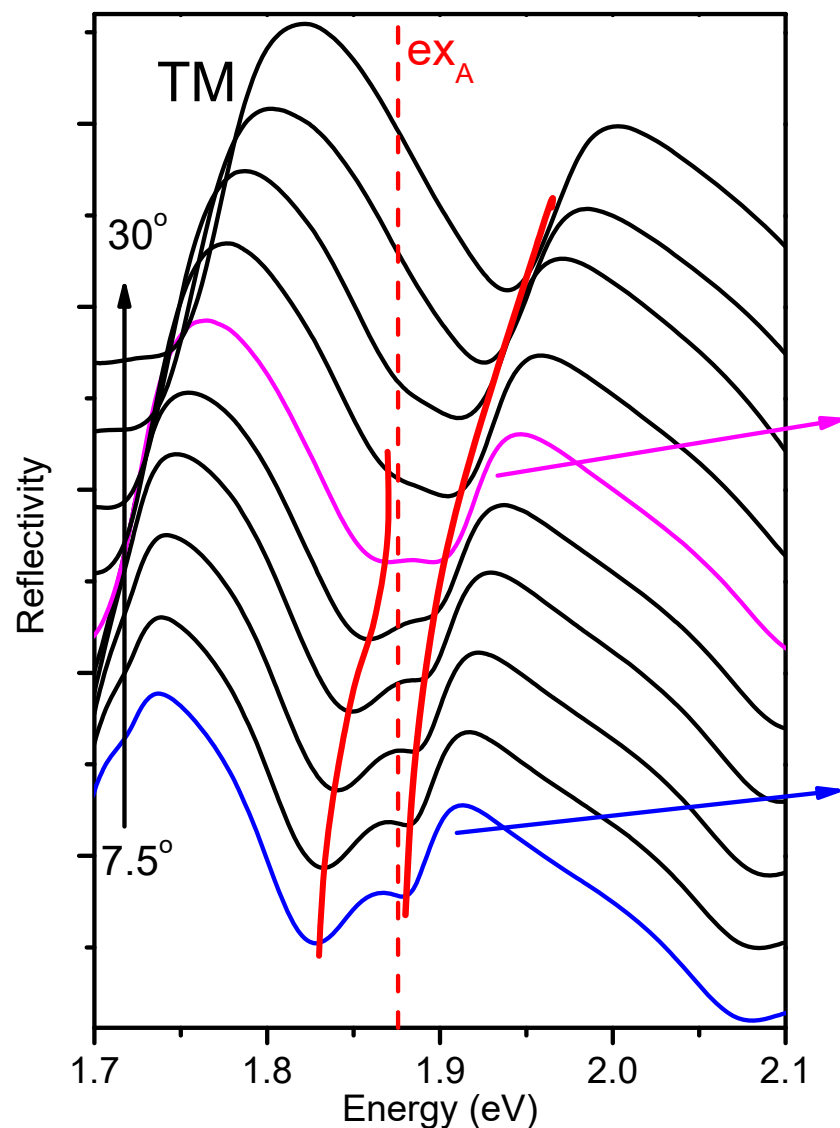


PDMS based transfer
KOH etch

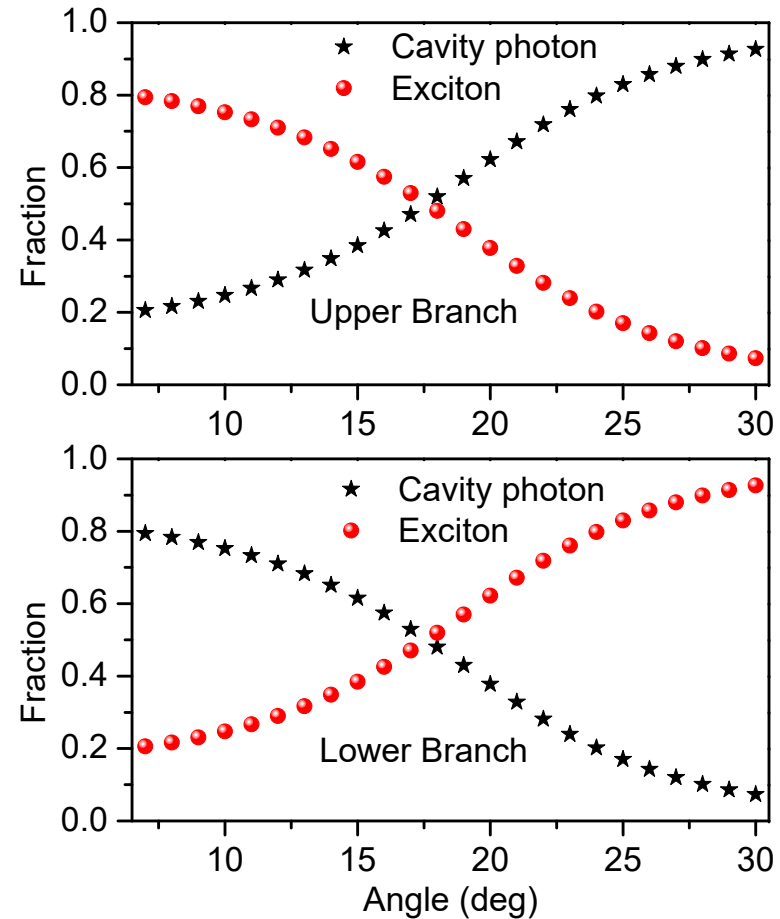
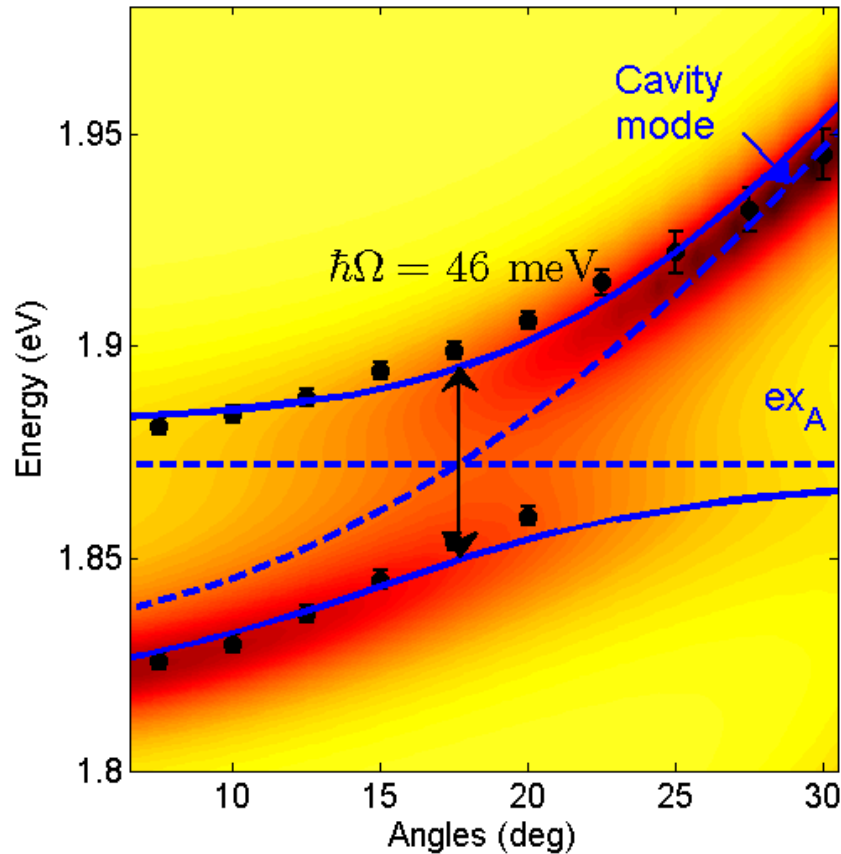
MoS₂ Microcavity



Angle-resolved reflectivity



Reflectivity Dispersions

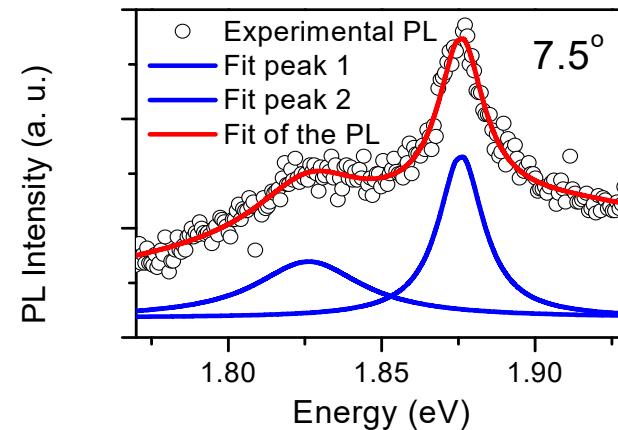
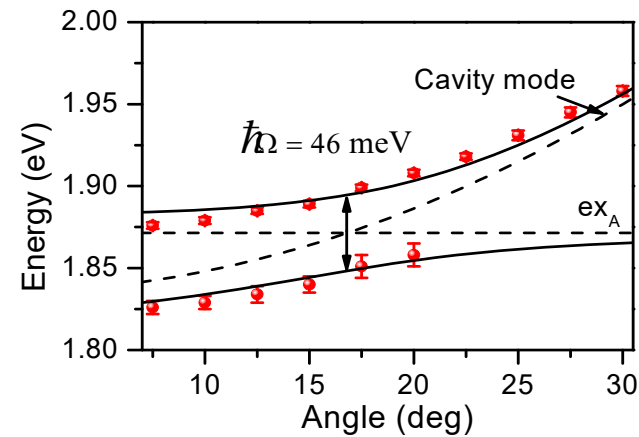
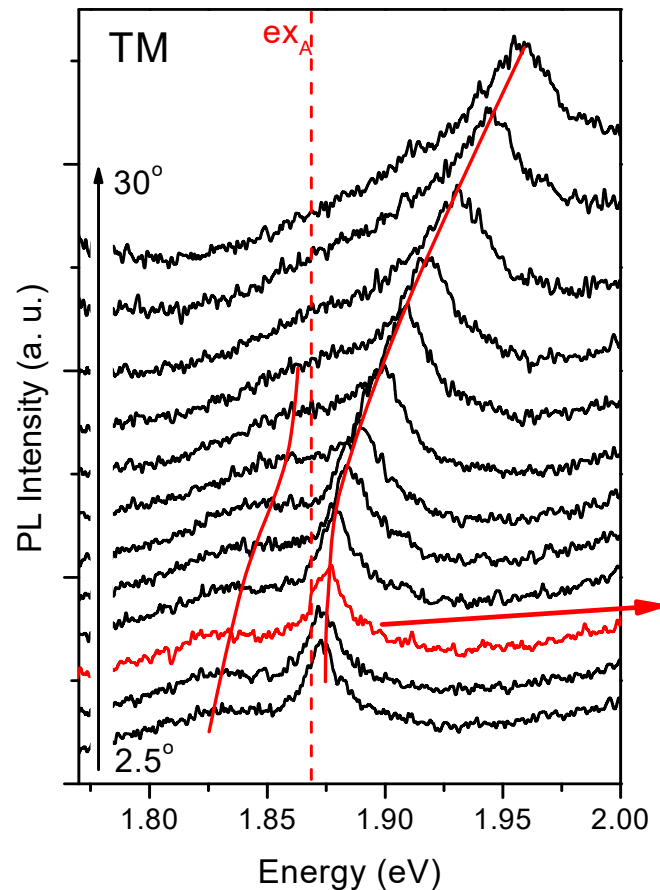


Determining the strong coupling regime:

$$\hbar\Gamma_{ex} = 60 \text{ meV}, \quad \hbar\Gamma_{cavity} = 17 \text{ meV}, \quad \hbar\Omega_{Rabi} = 46 \text{ meV}$$

$$\Rightarrow V = 31.5 \text{ meV} \Rightarrow V^2 > \left(\frac{\hbar\Gamma_{ex} - \hbar\Gamma_{cav}}{2} \right)^2$$

Angle-resolved PL



X. Liu,...VMM, *Nature Photonics* **9**, 30 (2015)

Other reports:

Dufferwiel, S. *et al. Nat. Commun.* **6**, 8579 (2015).

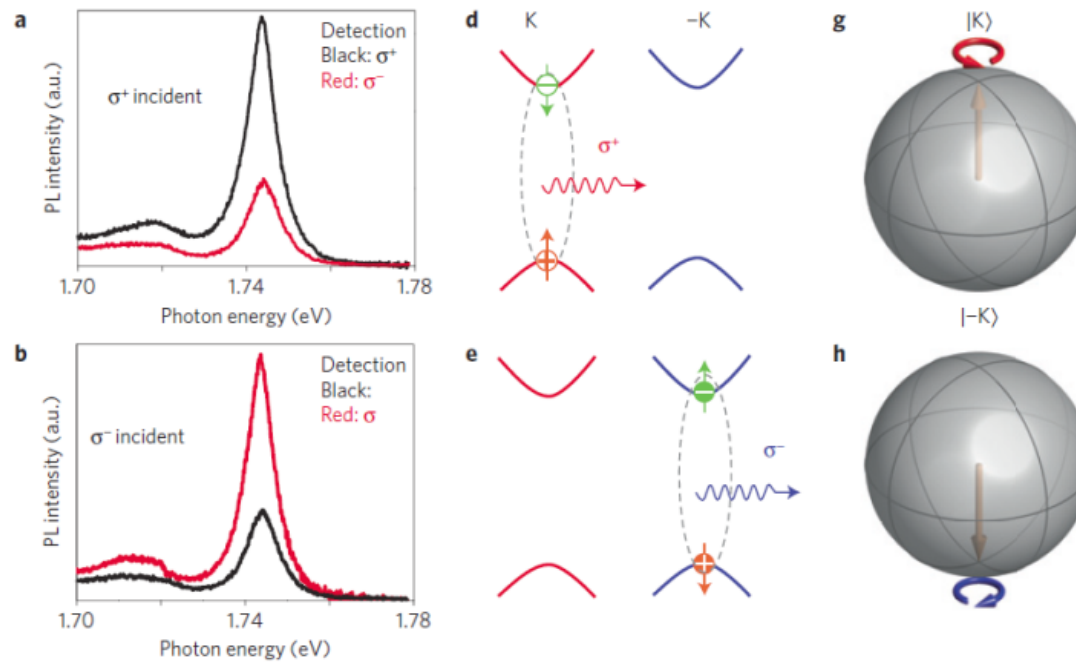
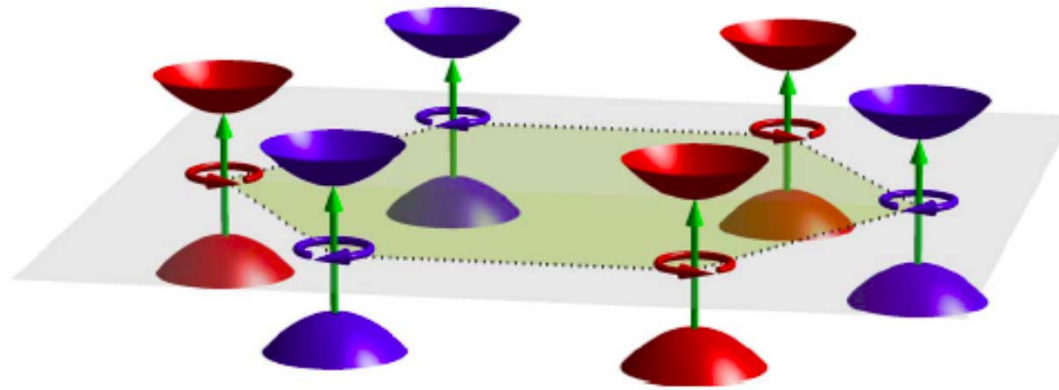
Sidler, M. *et al. Nat. Phys.* (2016). doi:10.1038/nphys3949

Liu, W. *et al. Nano Lett.* **16**, 1262–1269 (2016).

Wang, S. *et al. Nano Lett.* **16**, 4368–4374 (2016).

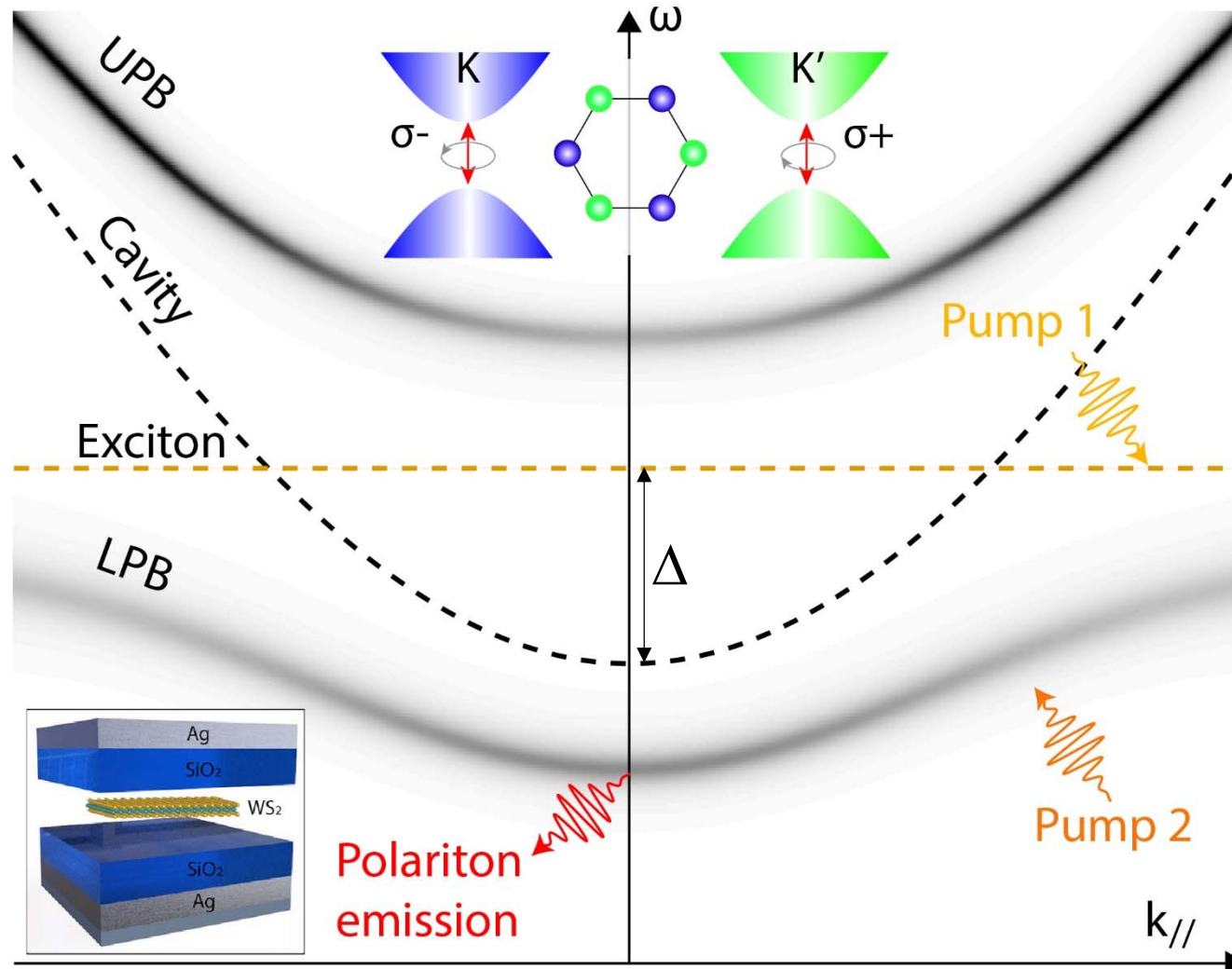
Lundt, N. *et al. Nat. Commun.* **7**, 13328 (2016).

Valley Polarization



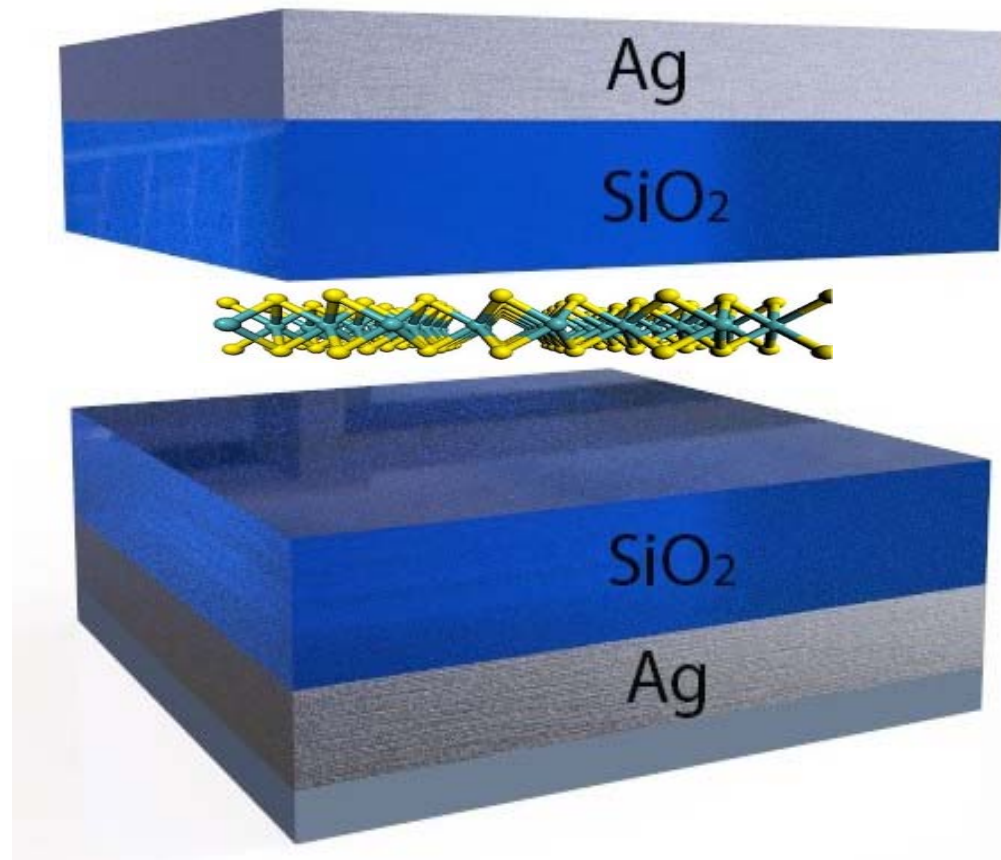
Xu, X. et al. *Nat. Phys.*, **10**, 343–350 (2014).

Valley Polaritons

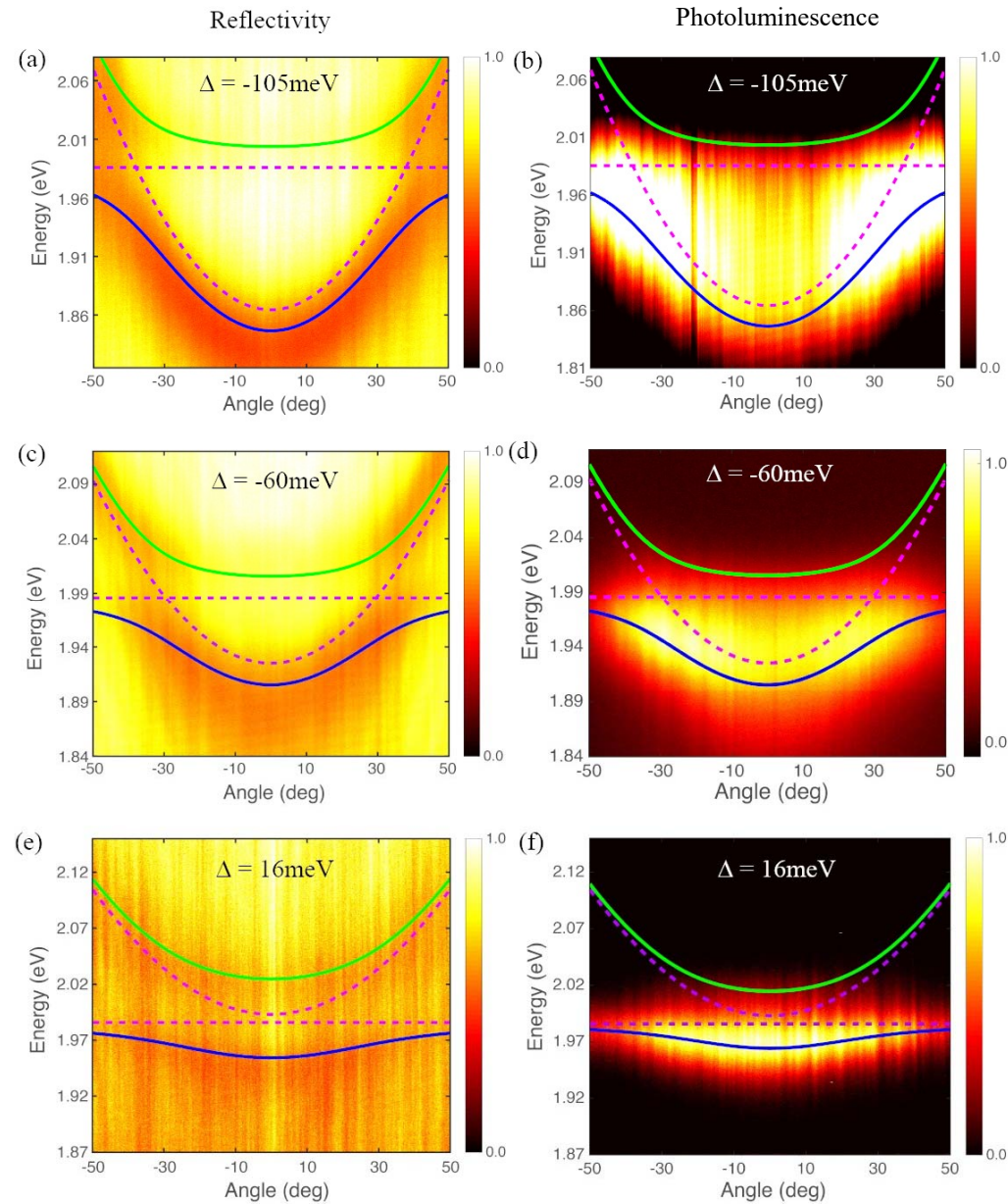


$$\text{Helicity: } \rho = \frac{|I(\sigma+) - I(\sigma-)|}{|I(\sigma+) + I(\sigma-)|}$$

Metal mirror cavity

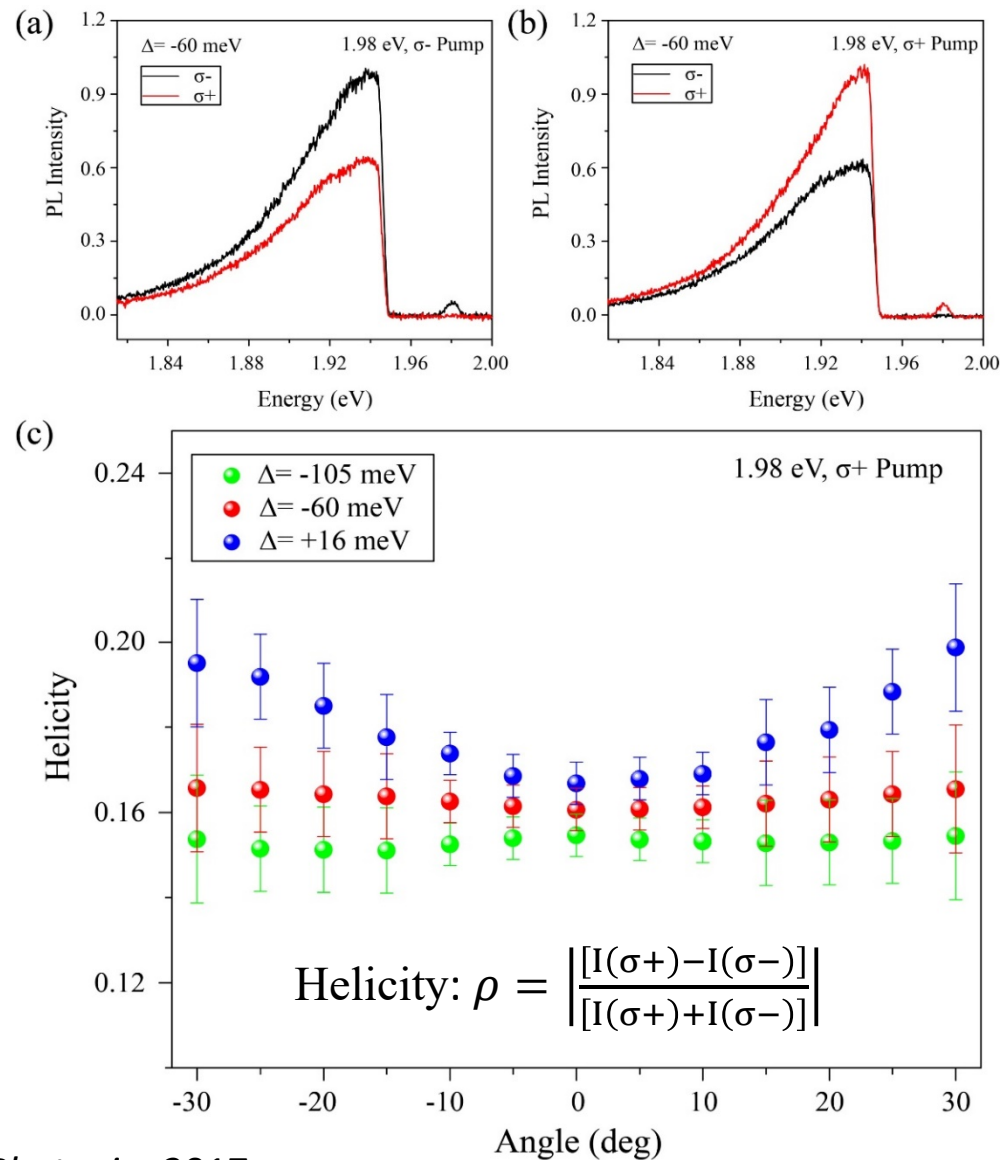


Dispersion



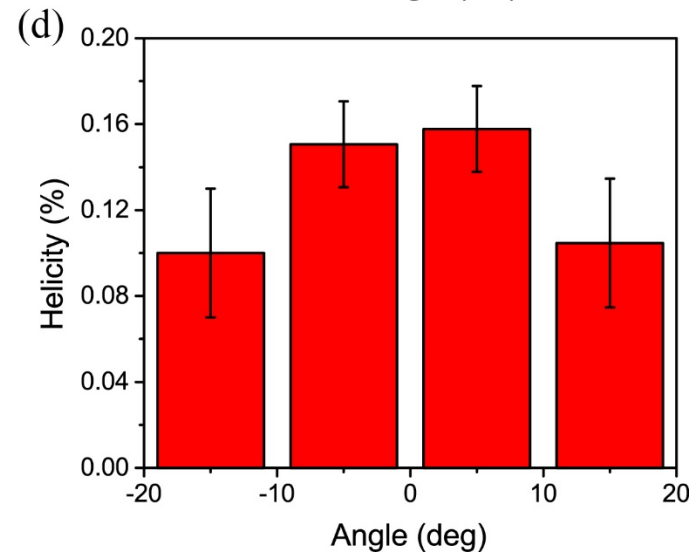
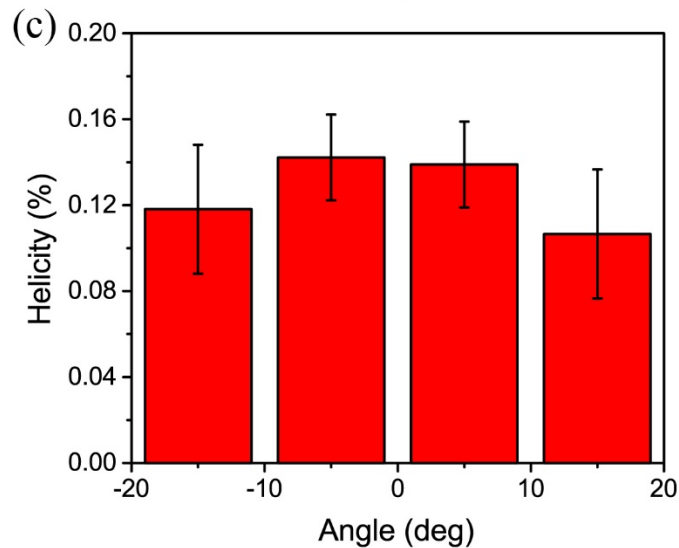
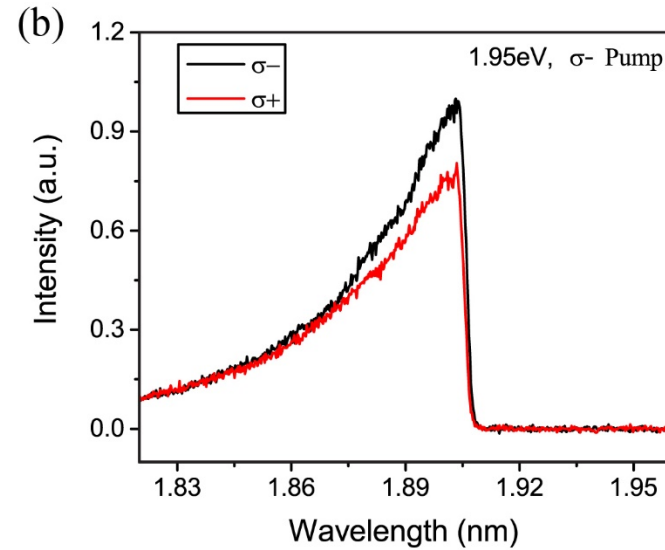
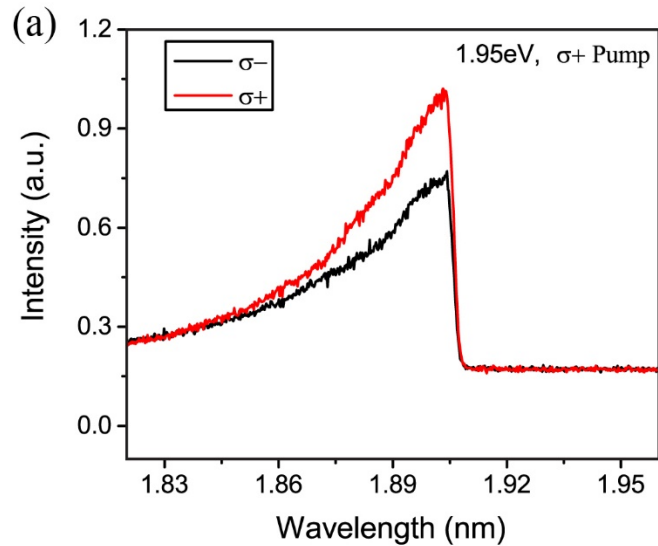
Helicity of polariton emission – Pump 1

Pump 1: Resonant with exciton energy

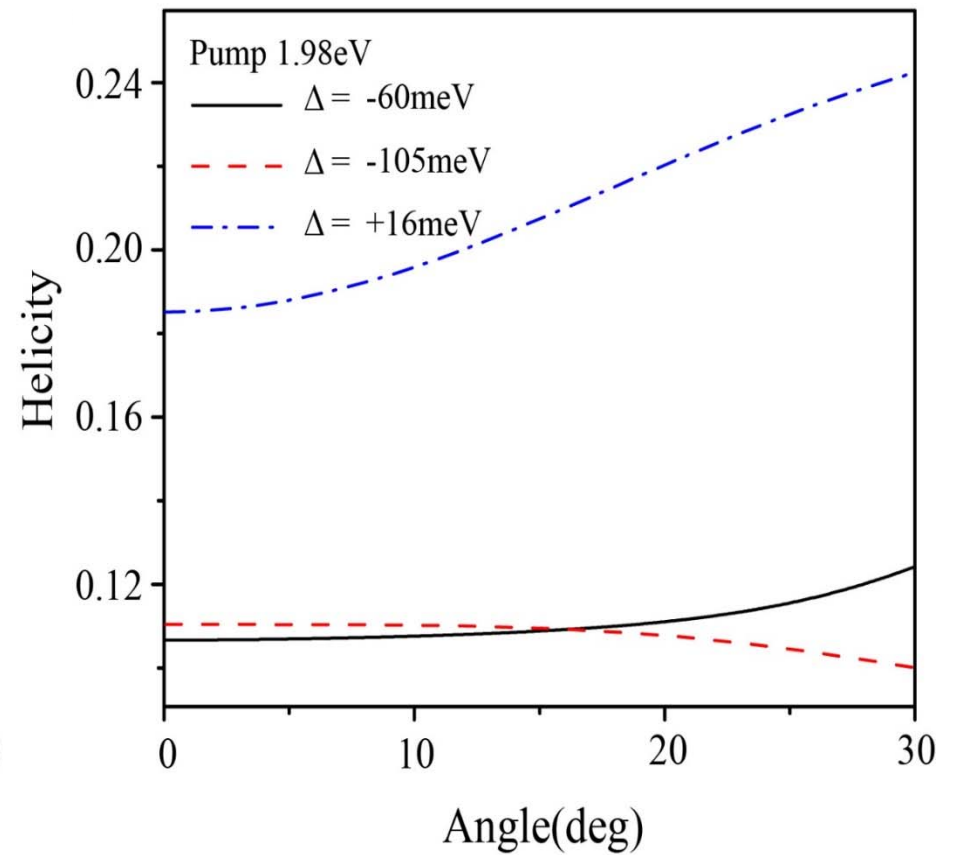
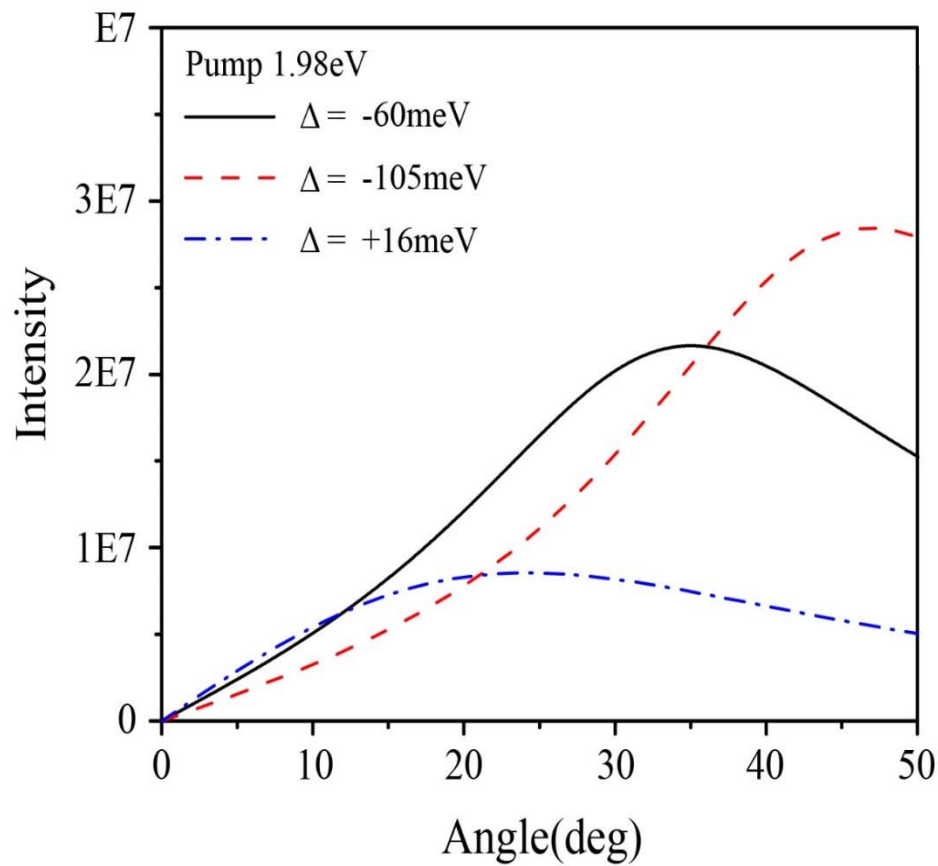


Helicity of polariton emission – Pump 2

Pump 2: Resonant with lower polariton branch



Theory simulations



Valley-polarized exciton-polaritons in a monolayer semiconductor

Yen-Jung Chen¹, Jeffrey D. Cain^{2,3}, Teodor K. Stanev¹, Vinayak P. Dravid^{2,3} and Nathaniel P. Stern^{1*}

Valley-addressable polaritons in atomically thin semiconductors

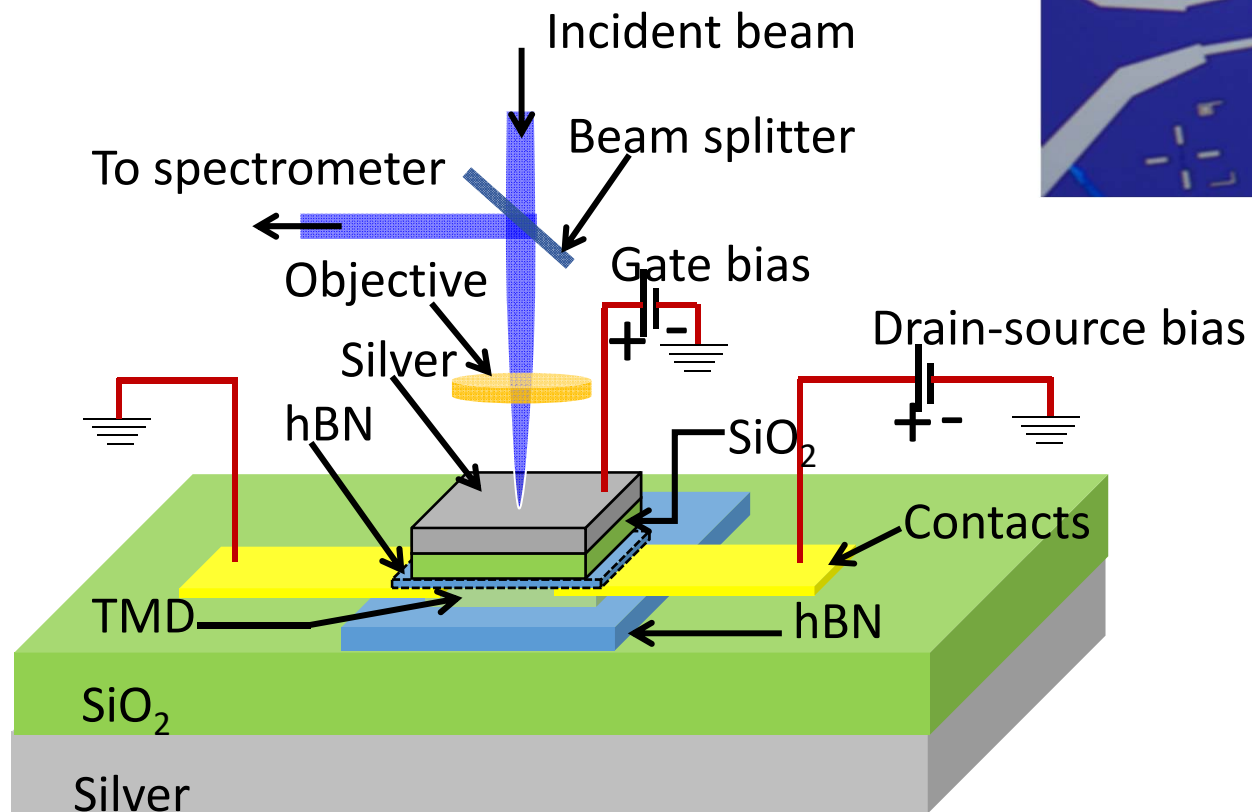
S. Dufferwiell^{1*}, T. P. Lyons¹, D. D. Solnyshkov², A. A. P. Trichet³, F. Withers^{4,5}, S. Schwarz¹, G. Malpuech², J. M. Smith³, K. S. Novoselov⁴, M. S. Skolnick¹, D. N. Krizhanovskii¹ and A. I. Tartakovskii^{1*}

Optical control of room-temperature valley polaritons

Zheng Sun^{1,2}, Jie Gu^{1,2}, Areg Ghazaryan¹, Zav Shotan¹, Christopher R. Consideine¹, Michael Dollar¹, Biswanath Chakraborty¹, Xiaoze Liu^{1,2†}, Pouyan Ghaemi^{1,2}, Stéphane Kéna-Cohen³ and Vinod M. Menon^{1,2*}

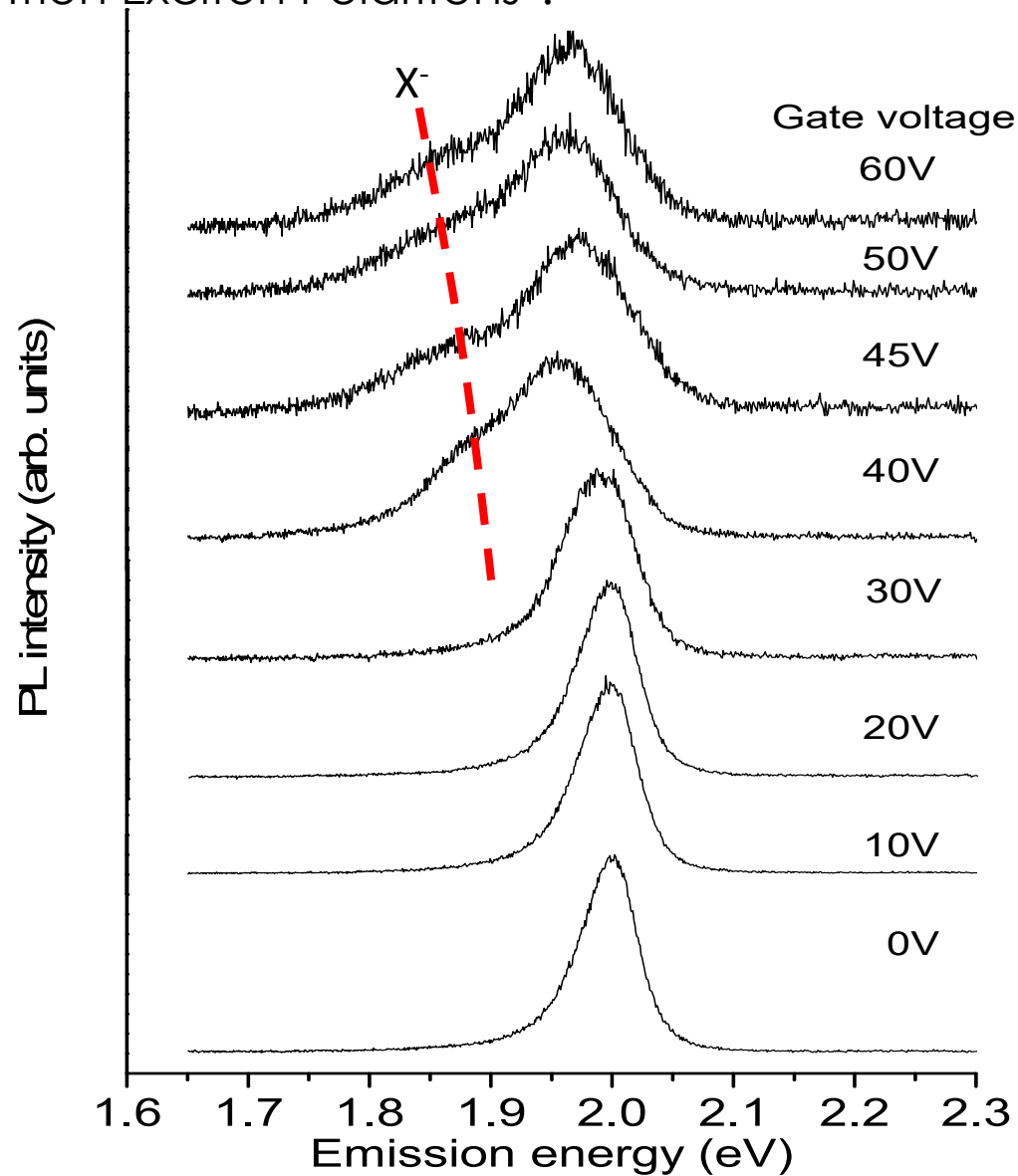
Outlook

Electrical control – Trion Exciton Polaritons ?



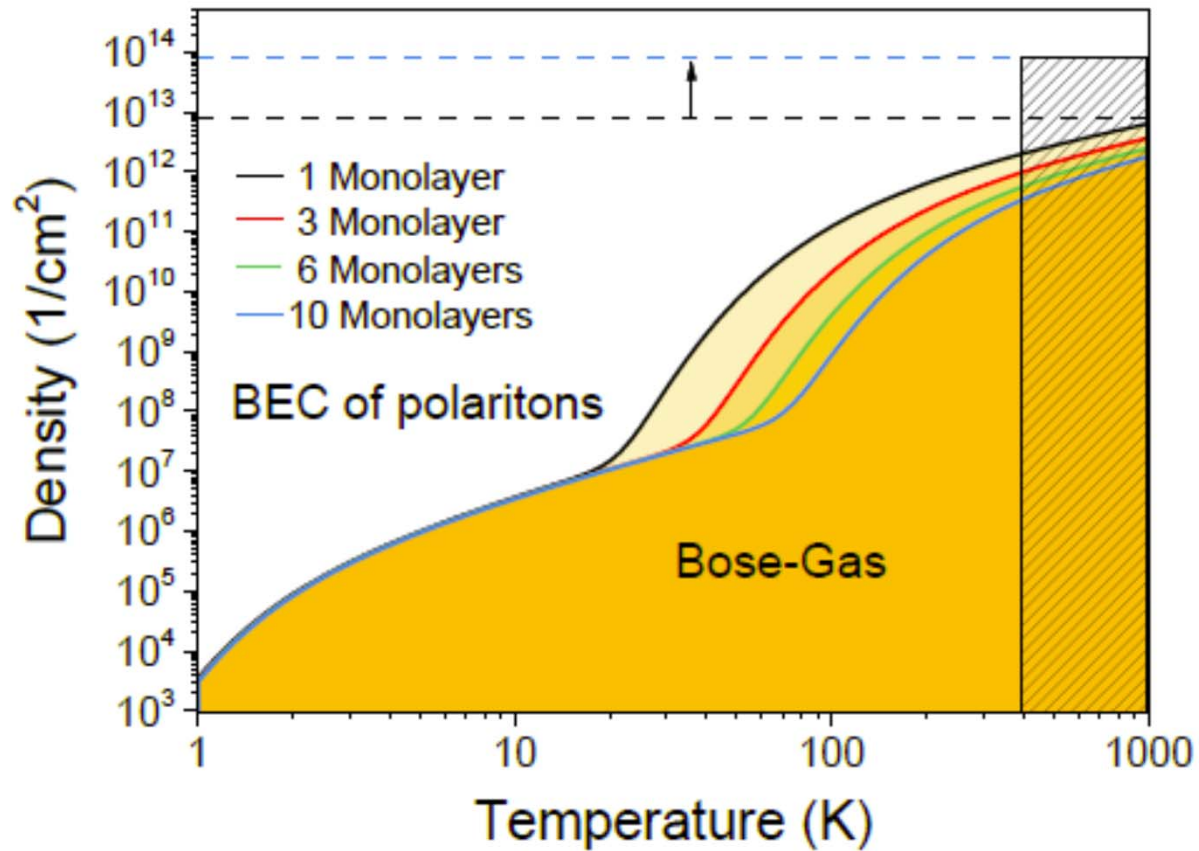
Outlook

Electrical control – Trion Exciton Polaritons ?



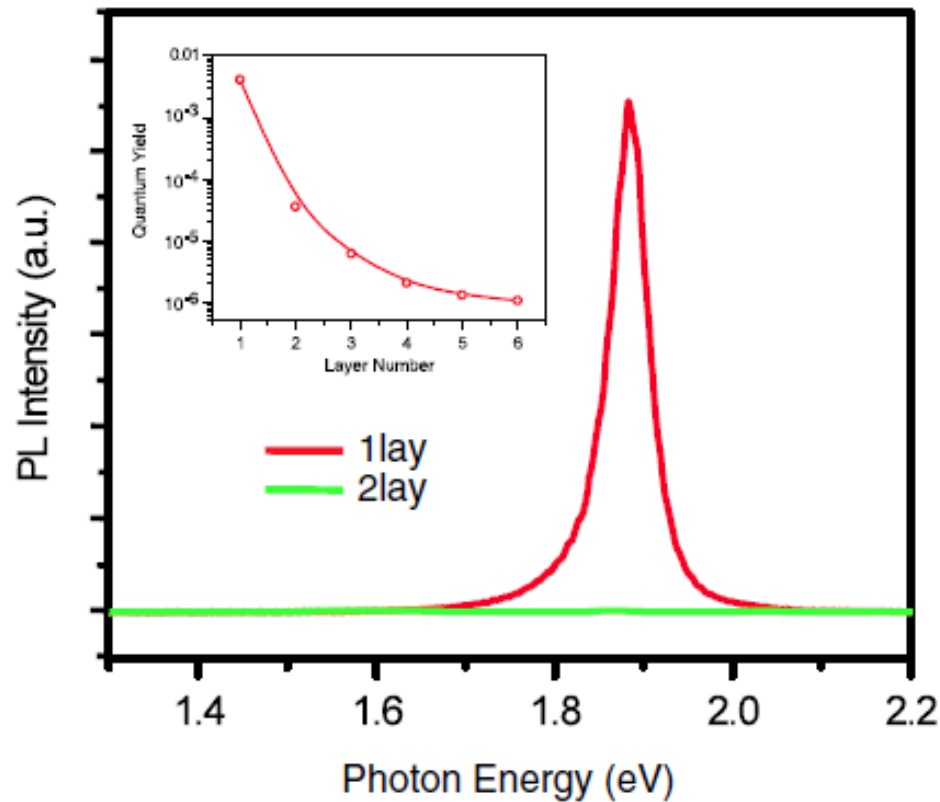
Outlook

Condensation?



Lundt et al. 2D Materials, **4**, 015006 (2016)

Quantum Yield?



$\sim 10^{-3}$

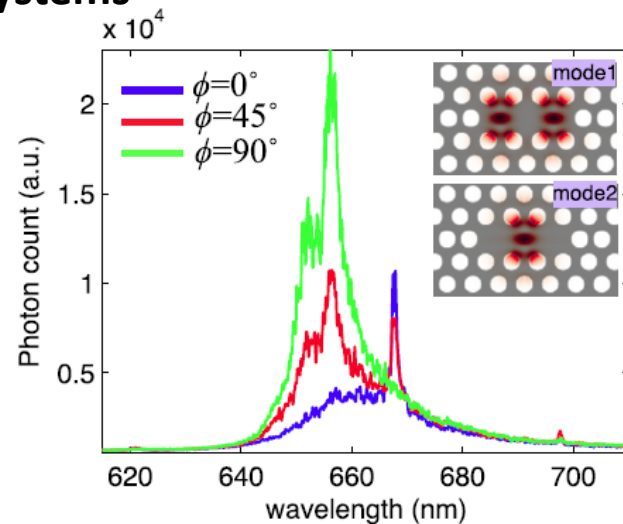
Mak, K. F. et al *Phys. Rev. Lett.*, **105**, 136805 (2010)

Chemical approaches to increasing quantum yield

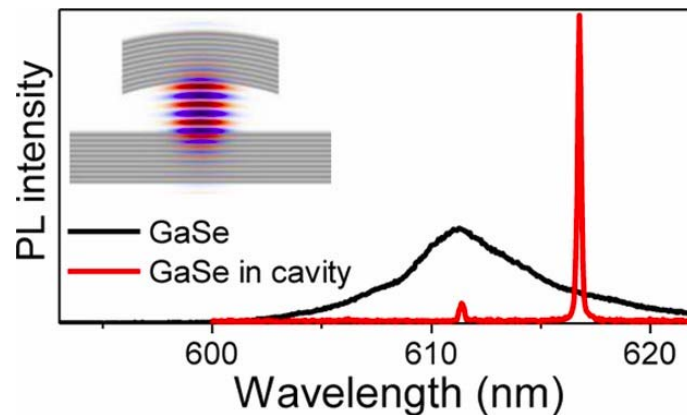
Amani et al. *Science* 350, 1065 (2015)

Control of Spontaneous Emission in 2D Materials

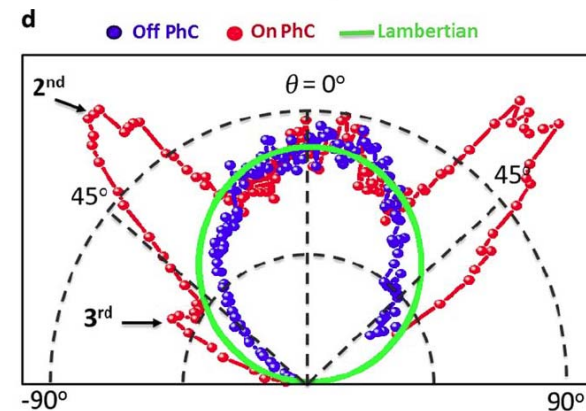
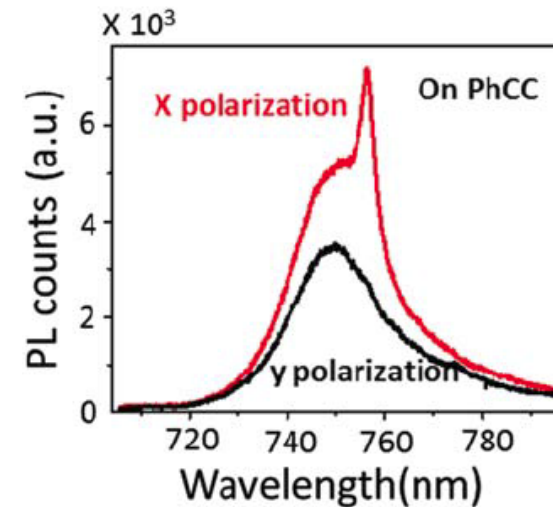
Cavity Systems



Gan, X. et al (2013). *Appl. Phys. Lett.* 103, 181119.



Schwarz, S. et al (2014). *Nano Lett.* 14, 7003

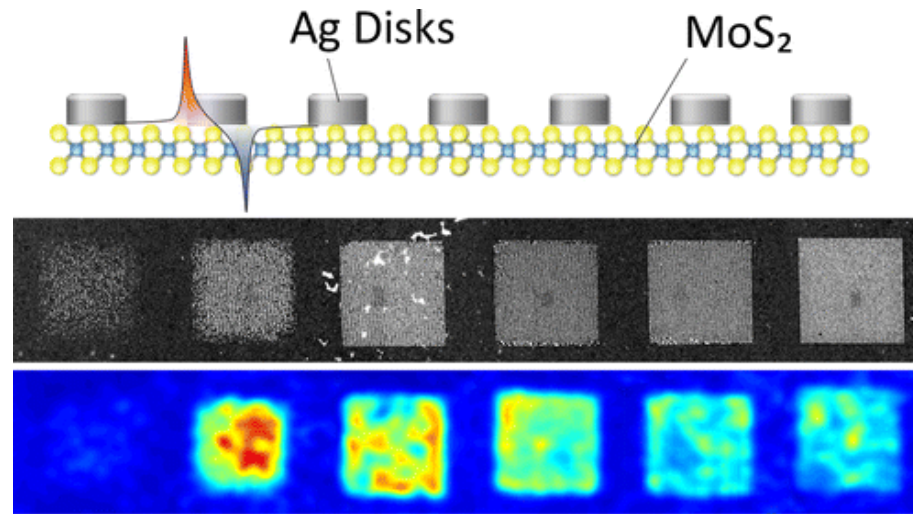


Wu, S. et al (2014). *2D Materials*, 1, 011001.

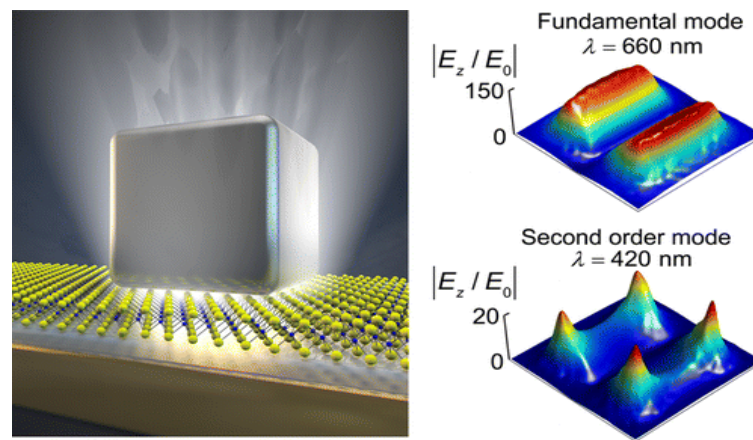
Demonstration of enhanced spontaneous emission and directionality

Control of Spontaneous Emission in 2D Materials

Plasmonic Systems



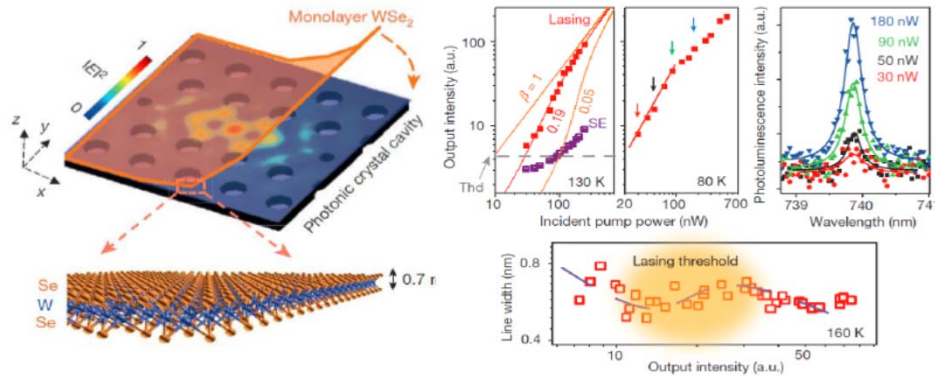
S. Butun. et al (2015). *Nano Lett.* 15, 2700



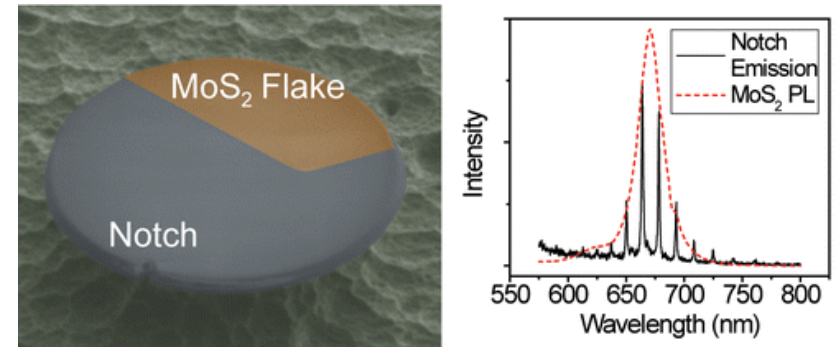
Akselrod et al. (2015). *Nano Lett.* 15, 3578

and several other groups

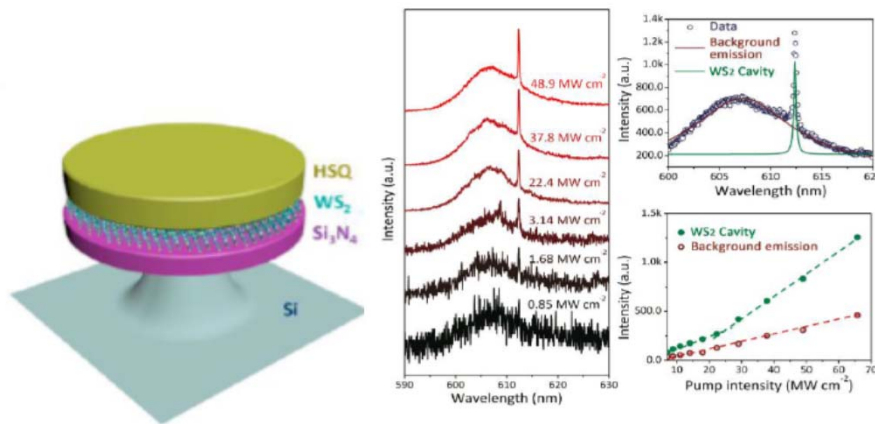
2D Materials Integrated with Microcavities



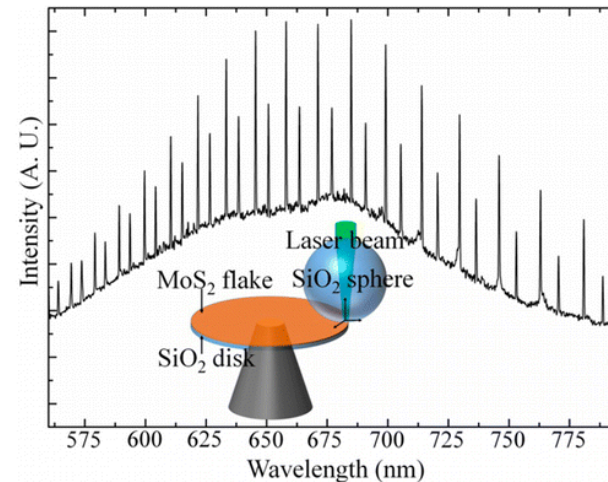
Wu, S., et al. *Nature*, 520, 69-72 (2015)



Reed, J., et al. *Nano Lett.* 15, 1967 (2015)



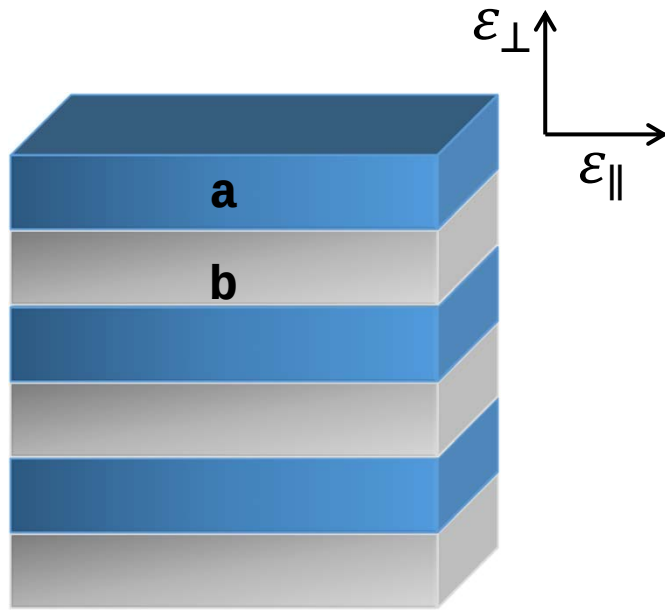
Ye, Y., et al. *Nature Photonics* 2015.



Salehzadeh, O., et al. *Nano Lett.* 15, 5302 (2015)

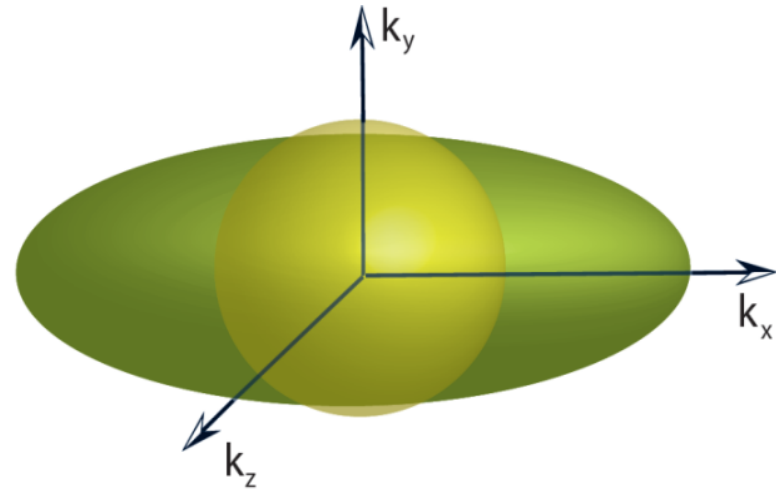
Broadband Enhancement of Spontaneous Emission?

Anisotropic Materials and Dispersion



$$\frac{1}{\epsilon_{eff}^{\perp}} = \frac{f_a}{\epsilon_a} + \frac{f_b}{\epsilon_b}$$

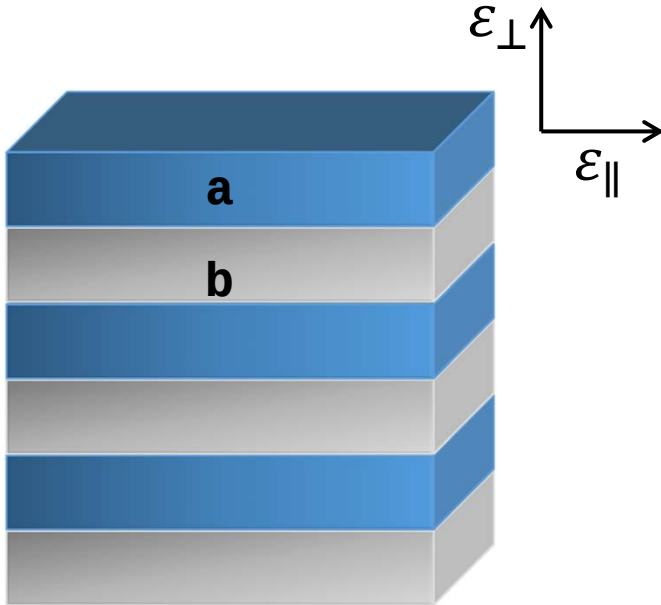
$$\epsilon_{eff}^{\parallel} = f_a \epsilon_a + f_b \epsilon_b$$



$$\epsilon_{\parallel}, \epsilon_{\perp} > 0; \epsilon_{\parallel} \ll \epsilon_{\perp}$$

$$\frac{k_z^2}{\epsilon_{\parallel}} + \frac{k_x^2 + k_y^2}{\epsilon_{\perp}} = \frac{\omega^2}{c^2}$$

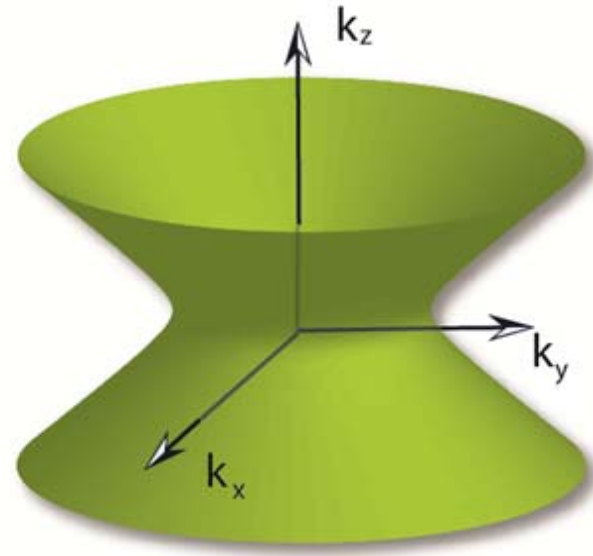
Anisotropic Materials and Dispersion



$$\frac{1}{\epsilon_{eff}^{\perp}} = \frac{f_a}{\epsilon_a} + \frac{f_b}{\epsilon_b}$$

$$\epsilon_{eff}^{\parallel} = f_a \epsilon_a \boxed{-} f_b \epsilon_b$$

Metals: $\epsilon < 0$ below plasma frequency
and $f_b > f_a$



$$\epsilon_{\parallel} < 0; \epsilon_{\perp} > 0;$$

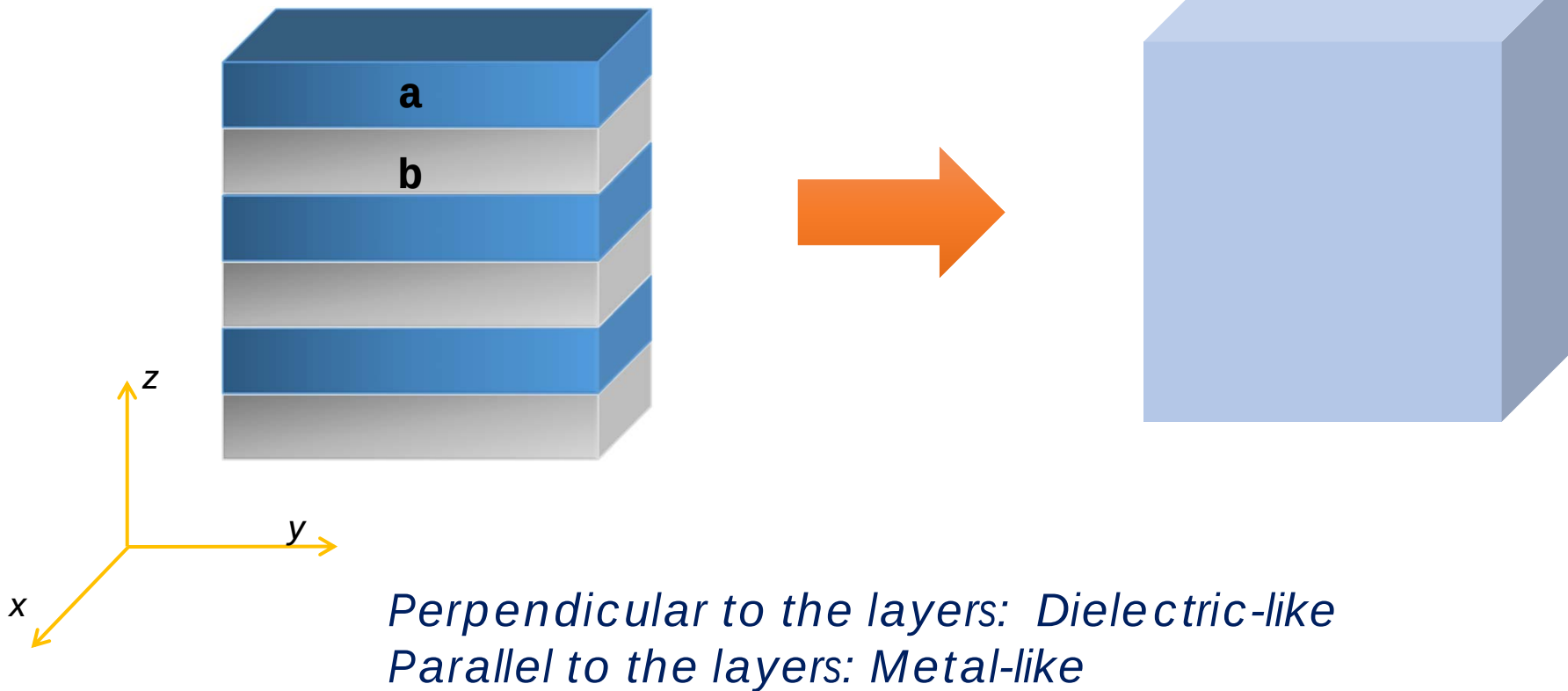
$$\frac{k_x^2 + k_y^2}{\epsilon_{\perp}} - \frac{k_z^2}{\epsilon_{\parallel}} = \frac{\omega^2}{c^2}$$

Indefinite Media (Hyperbolic Media)

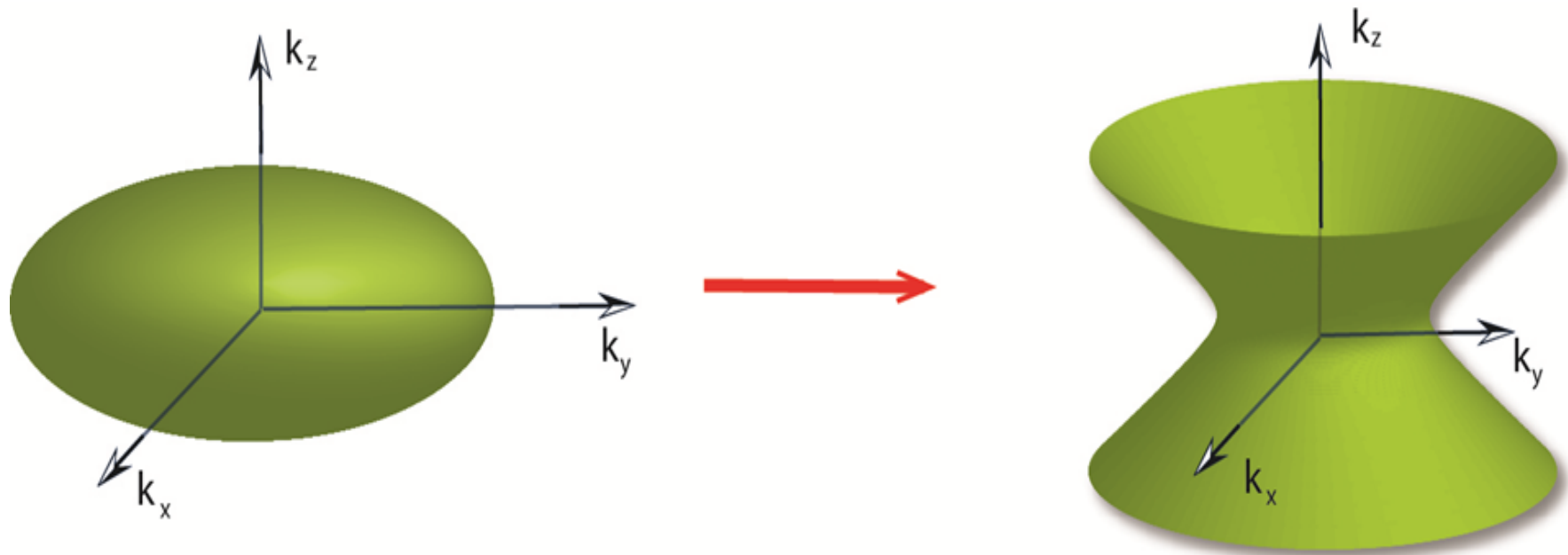
$$\epsilon_{xy(\parallel)} < 0, \epsilon_{z(\perp)} > 0$$

Subwavelength metal-dielectric structure

Effective Anisotropic Medium



Topology of Iso-frequency Surface

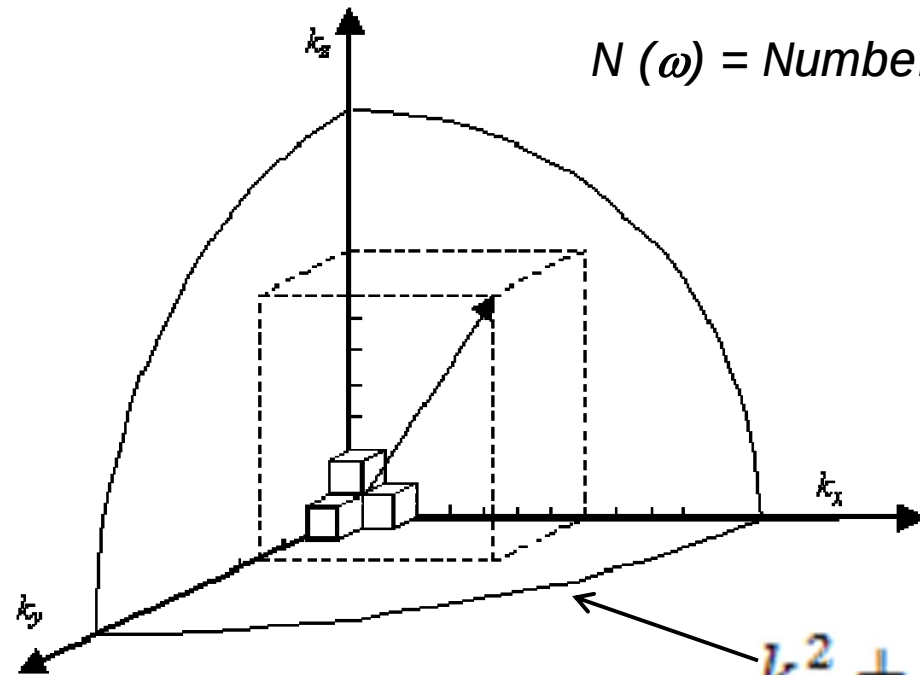


What is the physical manifestation of this transition in Optics?

M. Lifshitz, Sov. Phys. JETP. **11**, 1130 (1960).

Krishnamoorthy, Jacob, Narimanov, Kretzschmar, VMM, *Science* **336**, 205 (2012)

Photon Density of States



$N(\omega)$ = Number of states with frequency below ω

$$\rho(\omega) = \frac{dN}{d\omega}$$

$$k_x^2 + k_y^2 + k_z^2 = c \frac{\omega^2}{c^2}$$

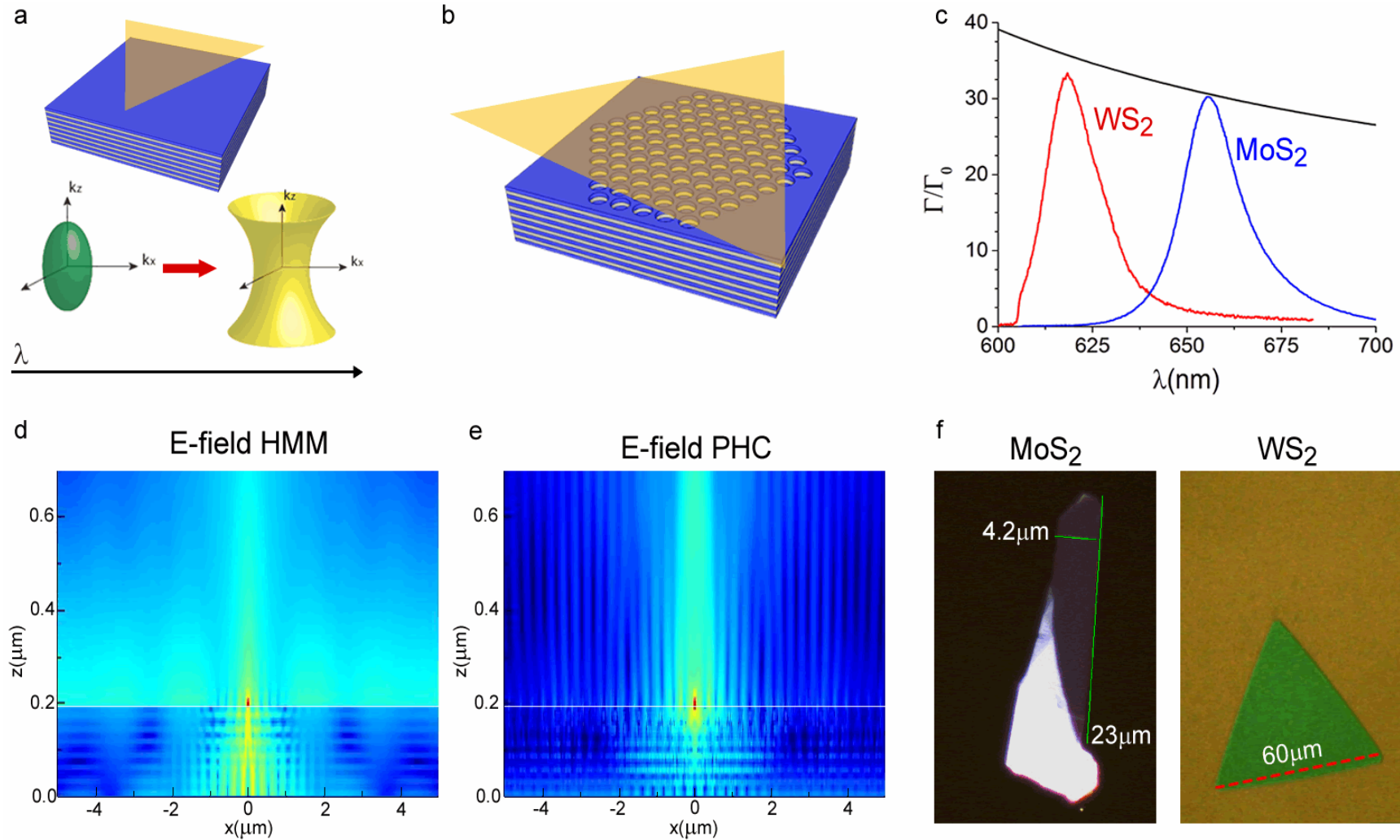
Purcell Effect

Fermi Golden Rule

$$\Gamma = \frac{2\pi}{\hbar} \rho(\omega) \times (\text{Dipole Matrix Element})^2$$

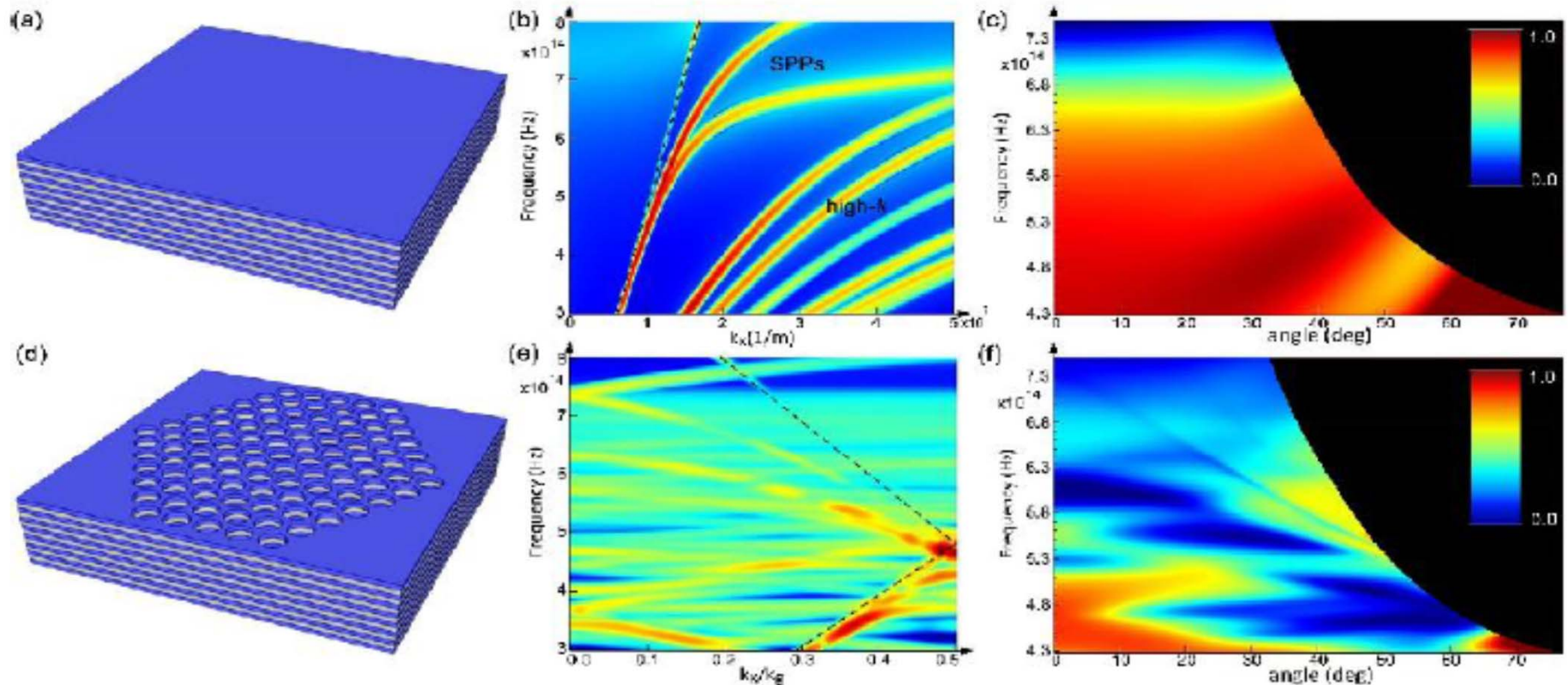
Purcell Factor = Rate in cavity / Rate in free space

Hyperbolic Metamaterials for Enhanced Spontaneous Emission from 2D TMD



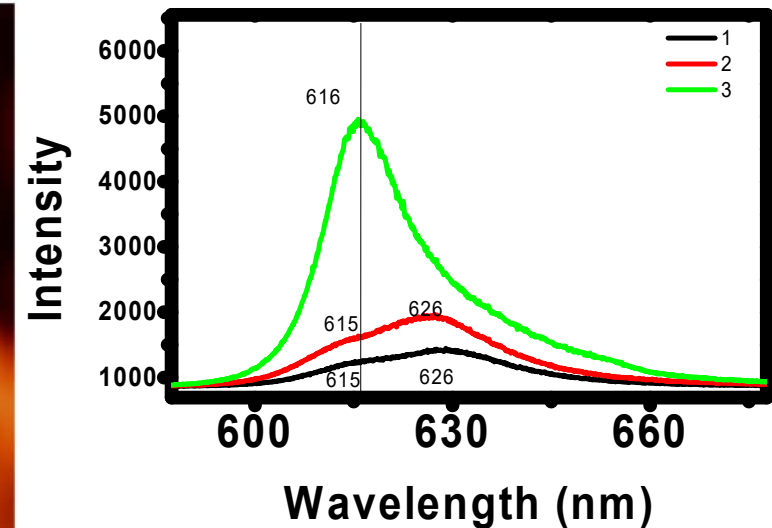
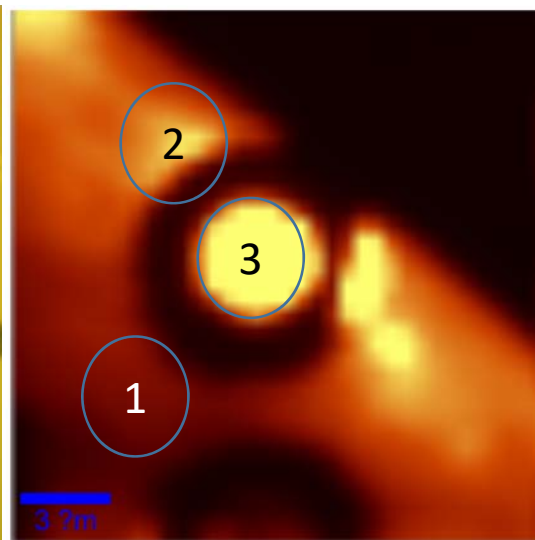
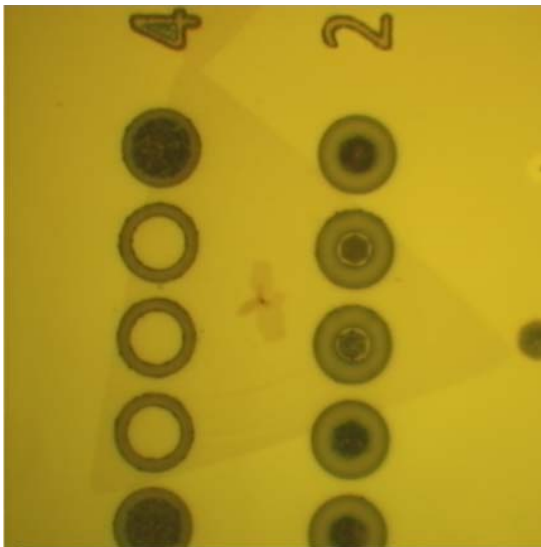
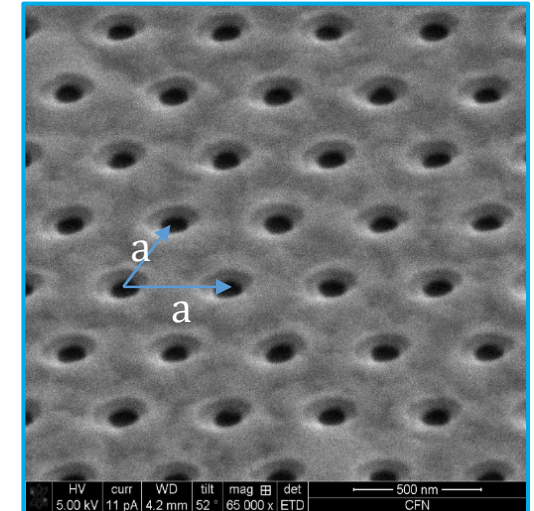
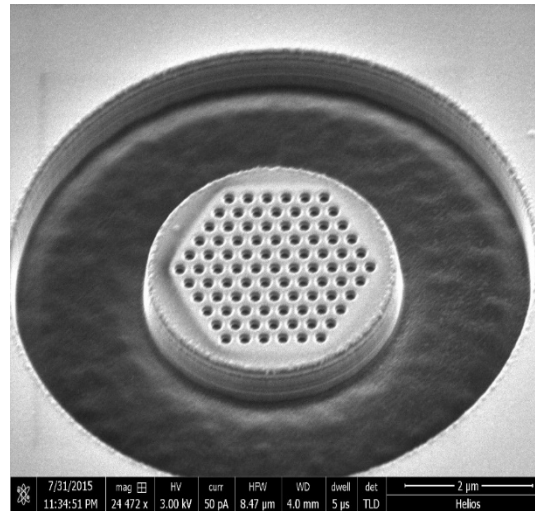
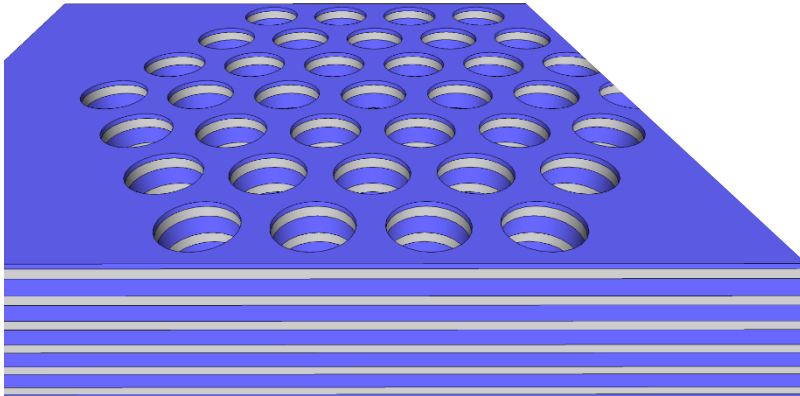
T. Galfsky...VMM, *Nano Lett.* DOI: 10.1021/acs.nanolett.6b01558, (2016)

Photonic Hypercrystal



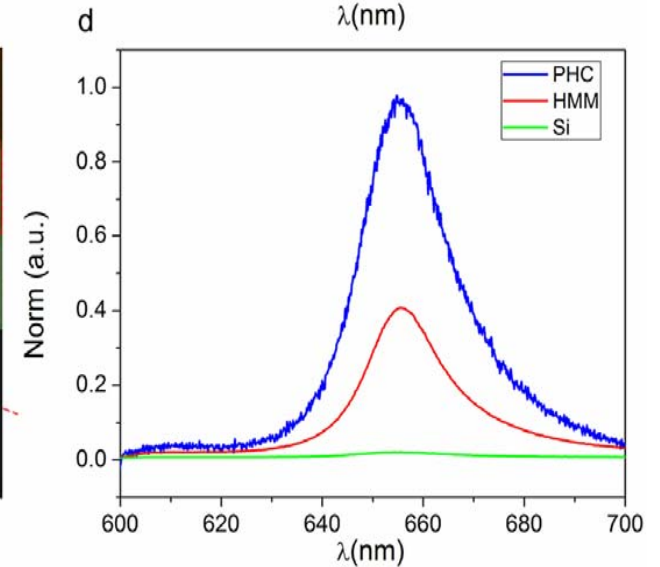
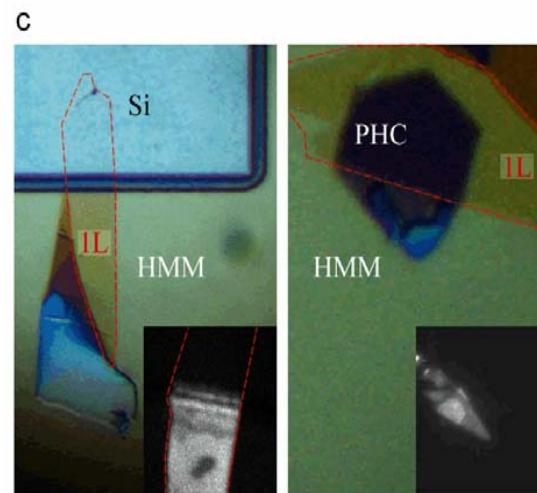
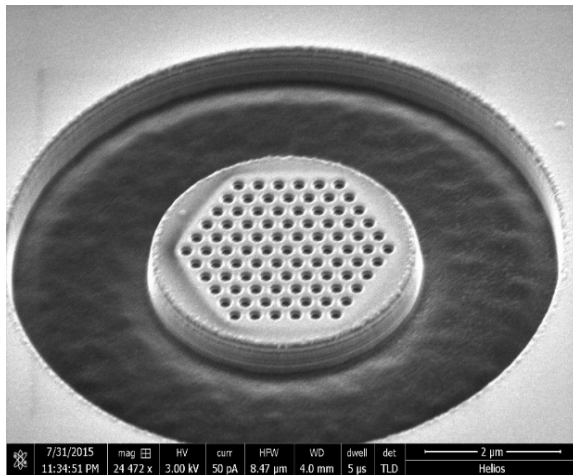
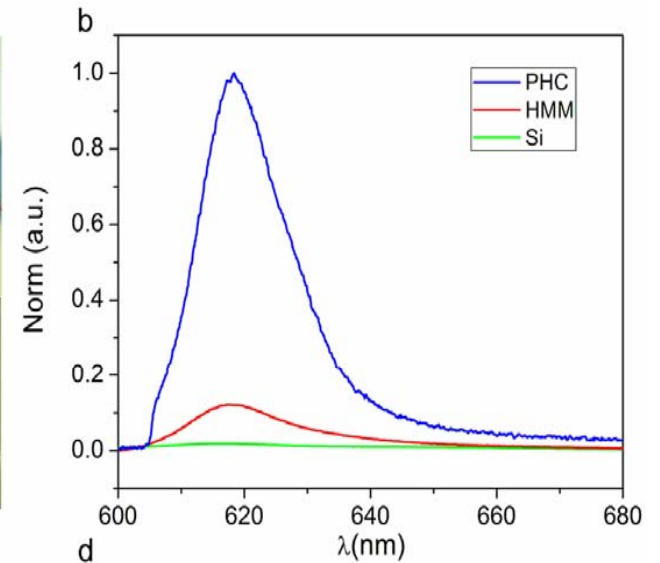
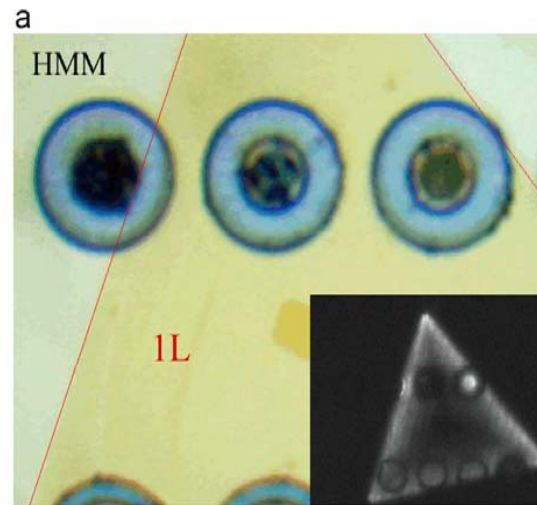
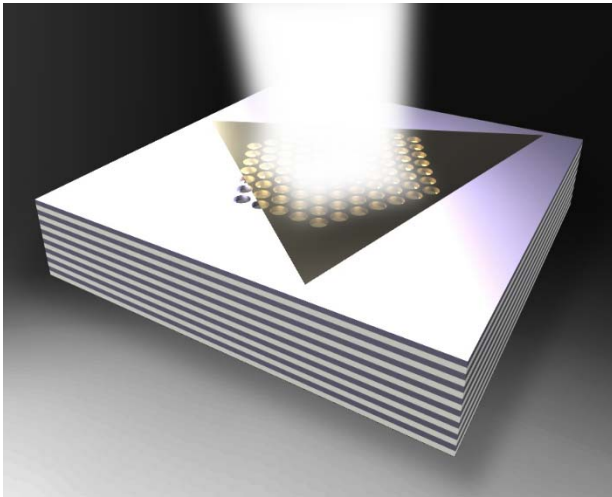
E. Narimanov, "Photonic Hypercrystals," *Phys Rev X* **4** (2014)
 T.Galfsky, J. Gu, E. Narimanov and VMM, *PNAS* **114**, 5125 (2017)

Photonic Hypercrystal

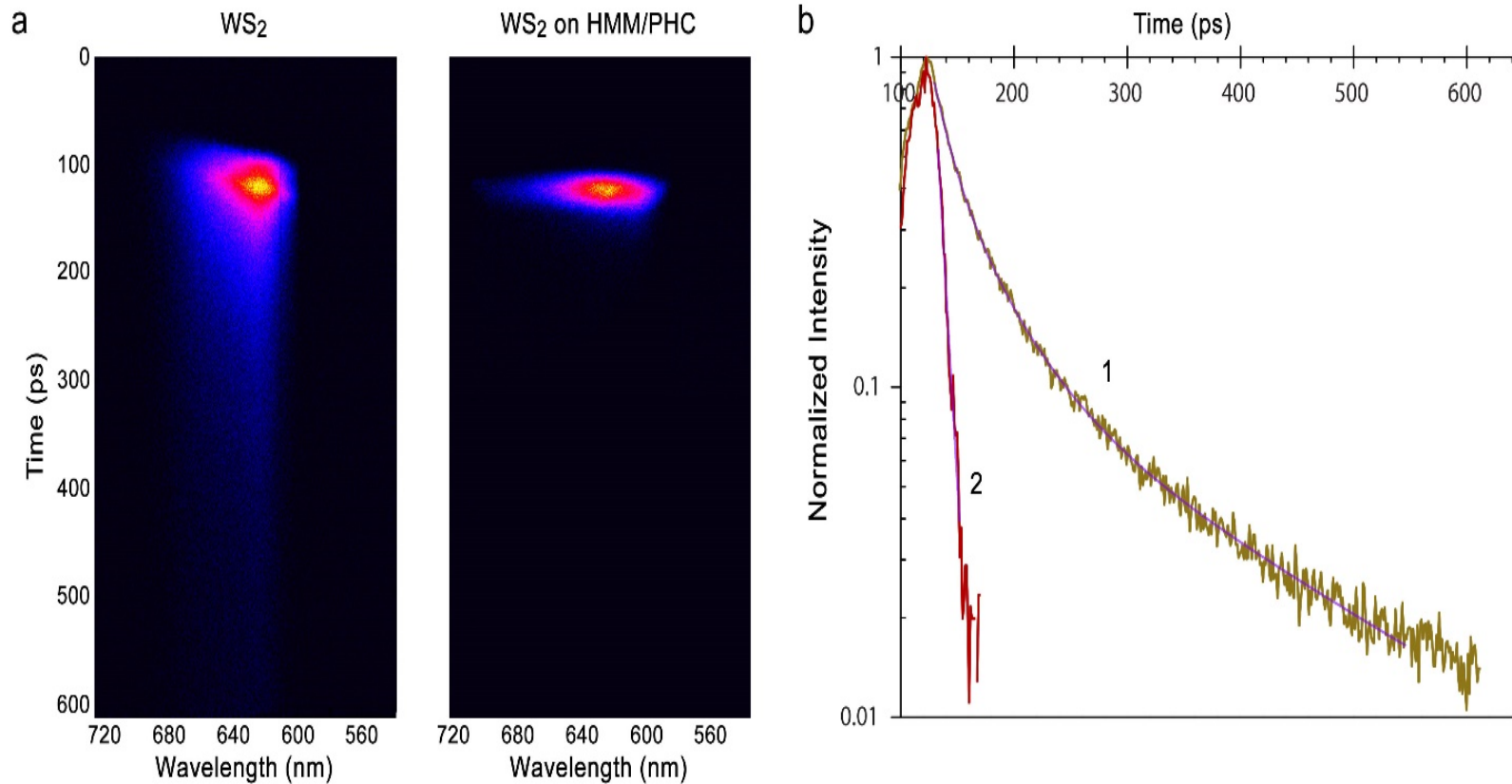


E. Narimanov, *Phys Rev X*. **4** (2014)
 T. Galfsky...VMM, *PNAS* **114**, 5125 (2017)

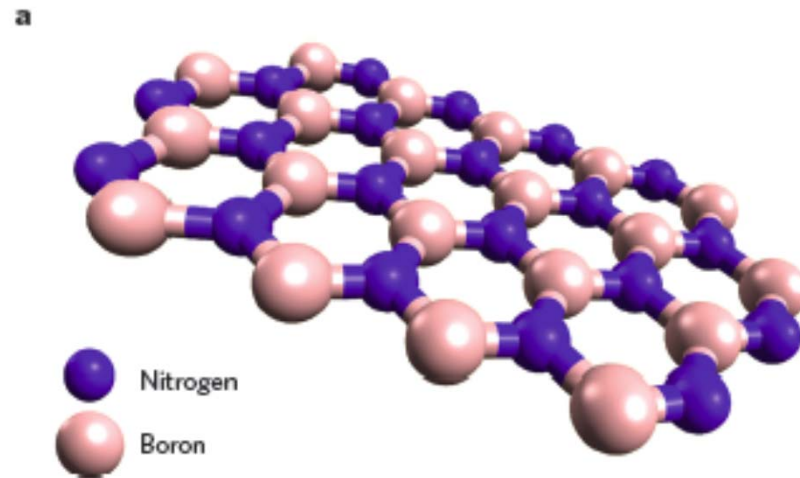
Spontaneous Emission



Spontaneous Emission Rate

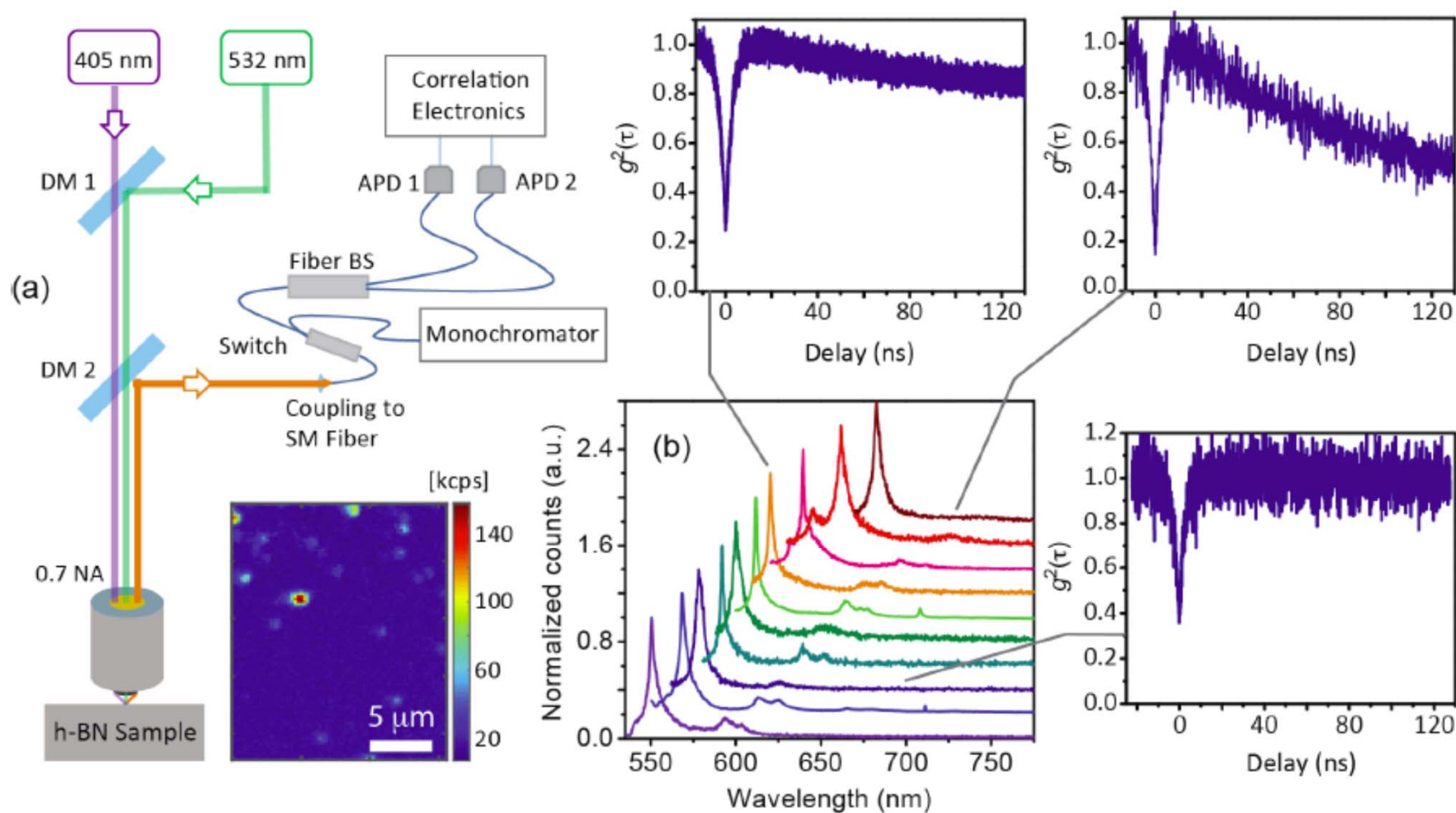


Quantum Emitters in hBN



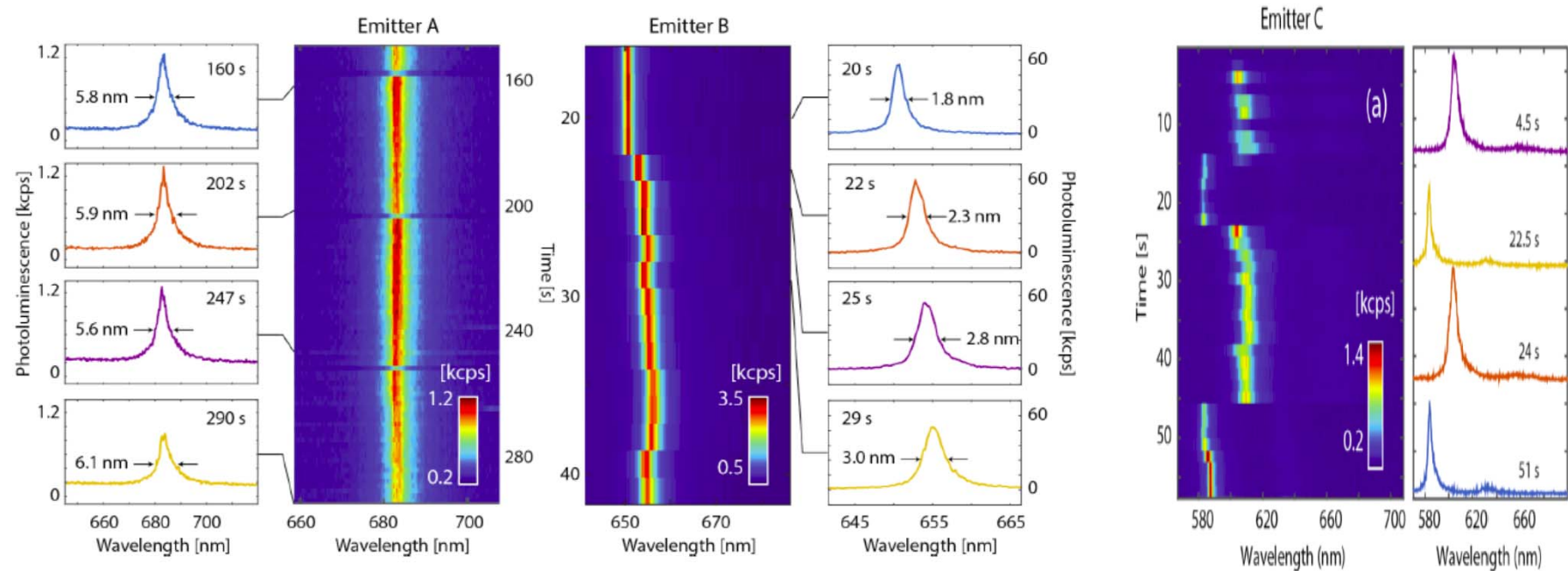
Tran et al. *Nat. Nanotechnol.* **11**, 37–41 (2015).

Single Photon Emission from hBN



Single Photon Emission from hBN

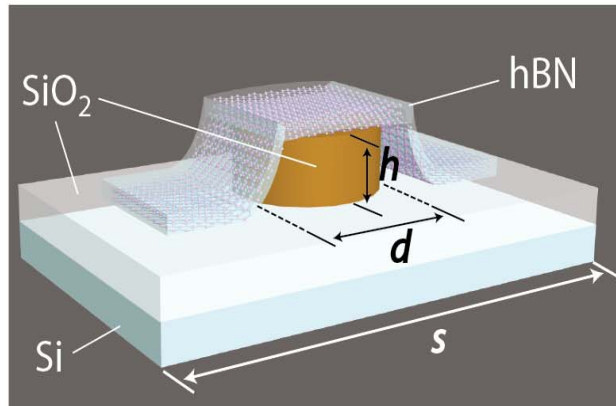
Stability Issues: Spectral diffusion, jumps



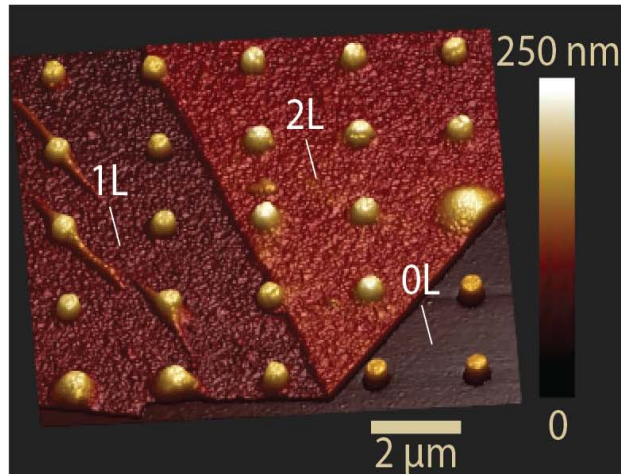
Emission wavelength and spatial location are not controlled

Towards Deterministic Single Photon Emission from hBN

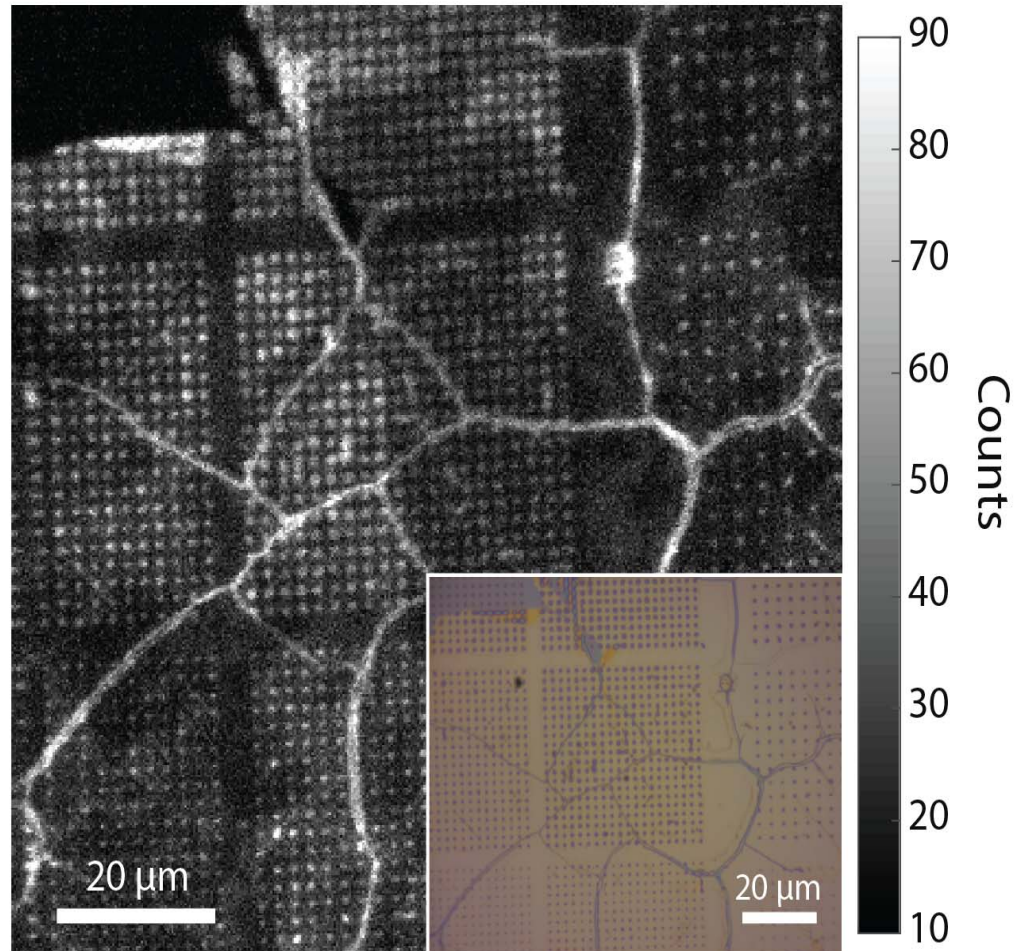
(a)



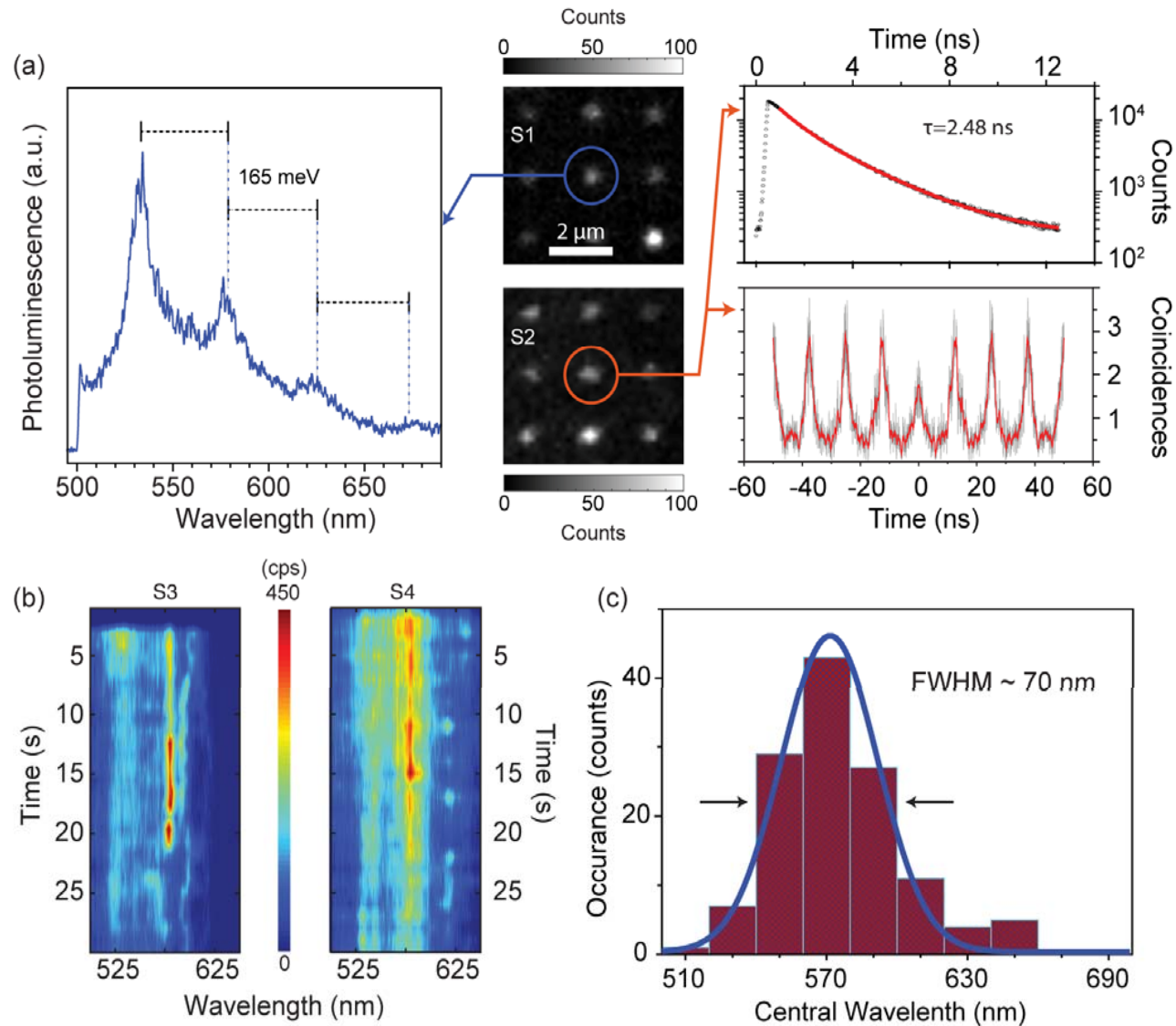
(b)



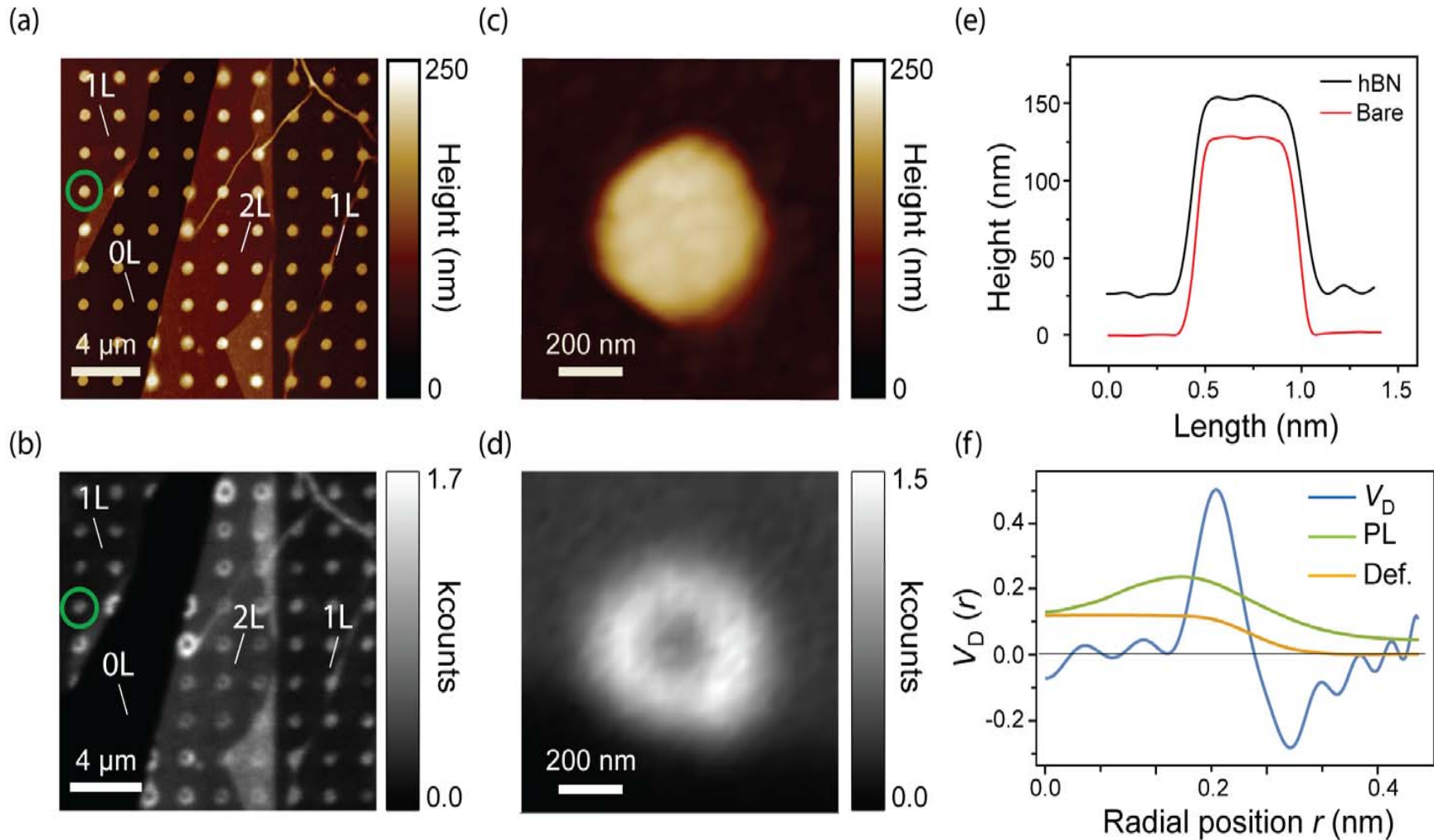
(c)



Towards Deterministic Single Photon Emission from hBN



Towards Deterministic Single Photon Emission from hBN



Summary

- Formation of microcavity polaritons in a metal cavity with monolayer WS_2 .
- Demonstration of the angle dependent valley polarized polaritons at room temperature with peak helicity around 27% .
- Observation of valley polarization of polariton emission when excited in resonance with the lower polariton branch.
- Enhanced spontaneous emission from 2D TMDs using photonic hypercrystal
- Room temperature single photon emission from defects in hBN

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Relevant Publications

“Room Temperature valley polaritons”, Z. Sun, J. Gu, A. Ghazaryan, Z. Shotan, M. Dollar, B. Chatterjee, P Ghaemi, S-K. Cohen and V. M. Menon, *Nature Photonics*, DOI: 10.1038/NPHOTON.2017.121

“Photonic hypercrystals for control of light-matter interaction,” T. Galfsky, J. Gu, E. Narimanov and V. M. Menon, *PNAS* **114**, 5119 (2017).

“Broadband enhancement of spontaneous emission in 2D semiconductors using photonic hypercrystals,” T. Galfsky, Z. Sun, C. R. Considine, C-T Chou, W-C Ko, Y-H Lee, E. Narimanov and V. M. Menon, *Nano Lett.* **16**, 4940 (2016).

“Microcavity enhanced second harmonic generation in 2D MoS₂,” J. K. Day, M. Chung, Y-H. Lee, and V. M. Menon, *Opt. Materials Express* **6**, 2360 (2016).

“Strong light-matter coupling in two-dimensional atomic crystals,” X. Liu, T. Galfsky, Z. Sun, F. Xia, E-C. Lin, Y-H. Lee, S. Kena-Cohen, and V. M. Menon, *Nature Photonics* **9**, 30 (2015).

“Topological transitions in metamaterials,” H. N. S. Krishnamoorthy, Z. Jacob, E. Narimanov, I. Kretzschmar, and V. M. Menon, *Science* **336**, 205 (2012).

Thank you!