



# Single photon emitters in sc-TMD ( $\text{WSe}_2$ ) structures (and in hexagonal boron nitride)

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J.-Y. Veuillen,<sup>3</sup> J. Marcus,<sup>3</sup> P. Kossacki,<sup>1,2</sup> and M. Potemski<sup>1</sup>

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# LNCMI

<http://lncmi.cnrs.fr/>

- Development of high magnetic field installations & instrumentation
- In house research in high magnetic fields
- Give access to all qualified French and European high field users



Grenoble; DC fields

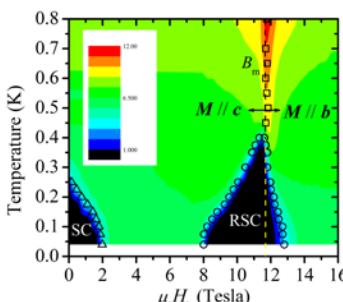
37 T / 43 T in project



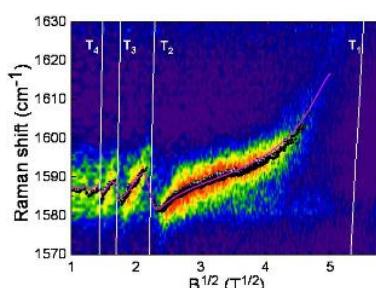
Toulouse; Pulsed fields

91 T 10 ms





**Correlated fermions,  
superconductivity**



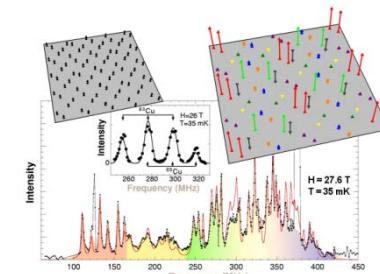
### Semiconductors and nanophysics

atomically thin 2D systems (graphene, TMDs)  
Dirac matter, topological insulators  
quantum dots  
optics and transport

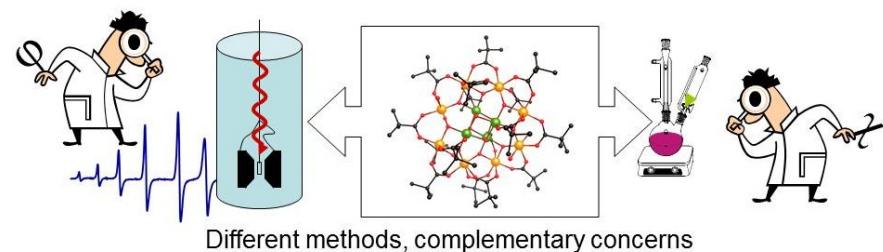


**Applied superconductivity**

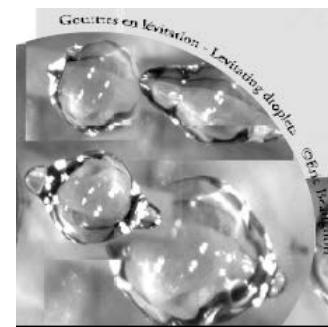
# Research topics at LNCMI-Grenoble



**Quantum magnetism**



**Chemistry**



**Magnetoscience** levitation,  
elaboration under magnetic field



# Acknowledgements



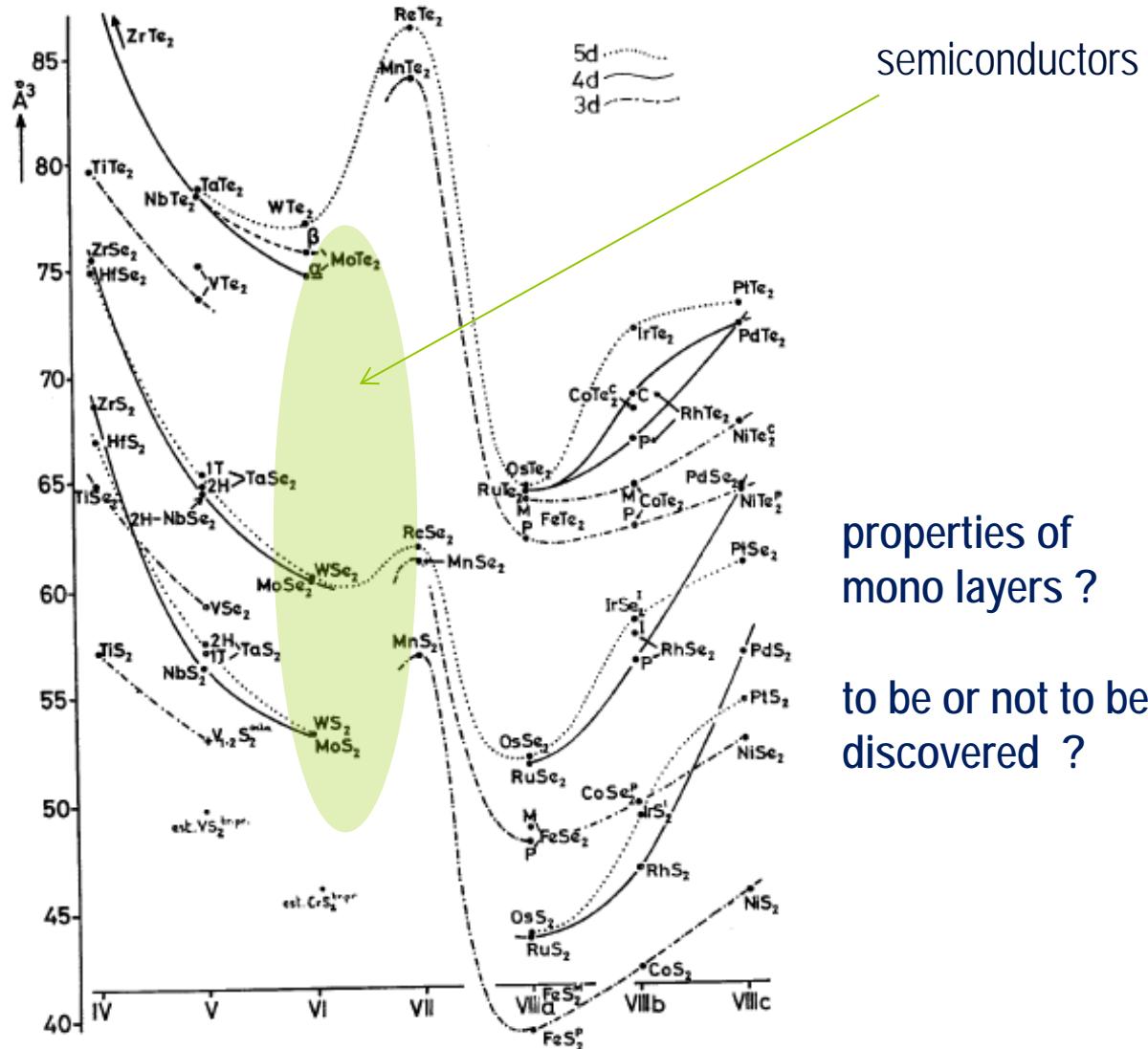
+ research groups  
from

Warsaw  
NEEL Grenoble

Manchester  
Columbia  
Geneva  
Münster



## Beyond graphene; other layered compounds, e.g., TM Dichalcogenides



### Advances in Physics

Publication details, including instructions for authors and subscription information:

<http://www.tandfonline.com/loi/tadp20>

**The transition metal dichalcogenides discussion and interpretation of the observed optical, electrical and structural properties**

J.A. Wilson <sup>a</sup> & A.D. Yoffe <sup>a</sup>

<sup>a</sup> Cavendish Laboratory, Cambridge

Available online: 02 Jun 2006

semiconductors

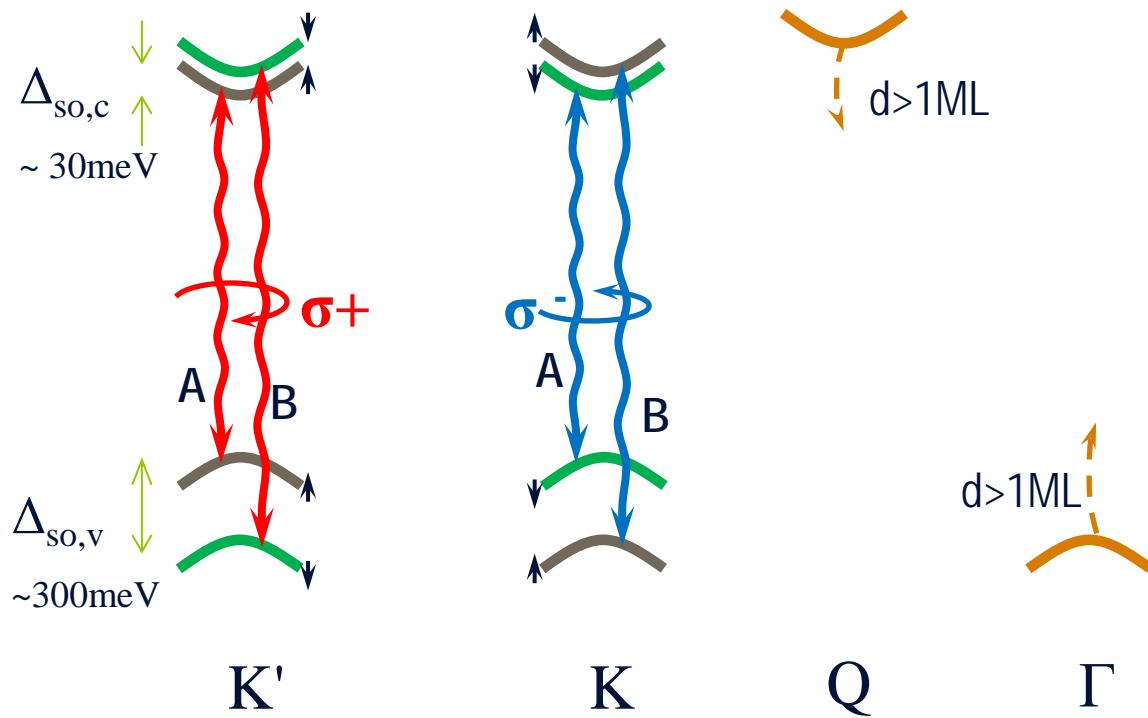
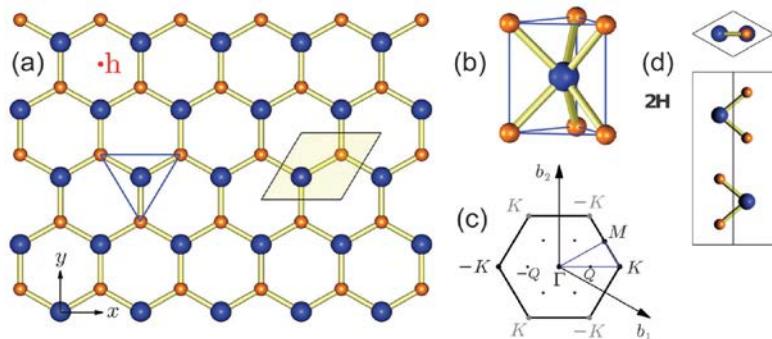
properties of  
mono layers ?

to be or not to be  
discovered ?



# Atomically thin semiconducting transition metal dichalcogenides

$\text{WSe}_2, \text{MoSe}_2, \text{WS}_2, \text{MoS}_2, \text{MoTe}_2, \dots$  (S-TMDs)



light emitting devices  
(optically bright monolayers)

+

opto-valley-tronics  
(robust valley pseudospin ?)

+

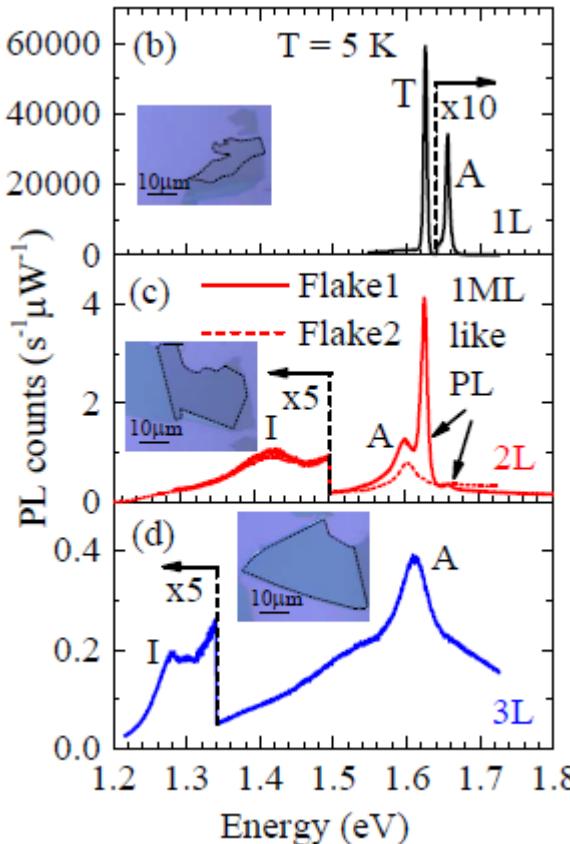
...., interesting physics

# Direct gap monolayers and indirect gap N-layers

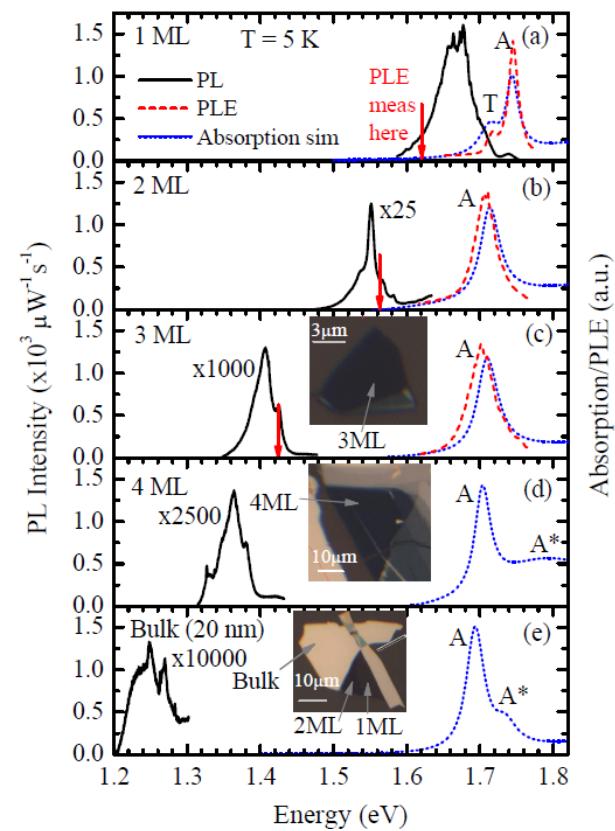
A. Splendiani *et al.*, Nano Lett. 10, 1271 (2010).  
 F.K. Mak *et al.*, Phys. Rev. Lett. 105, 136805, (2010).

LNCMI:

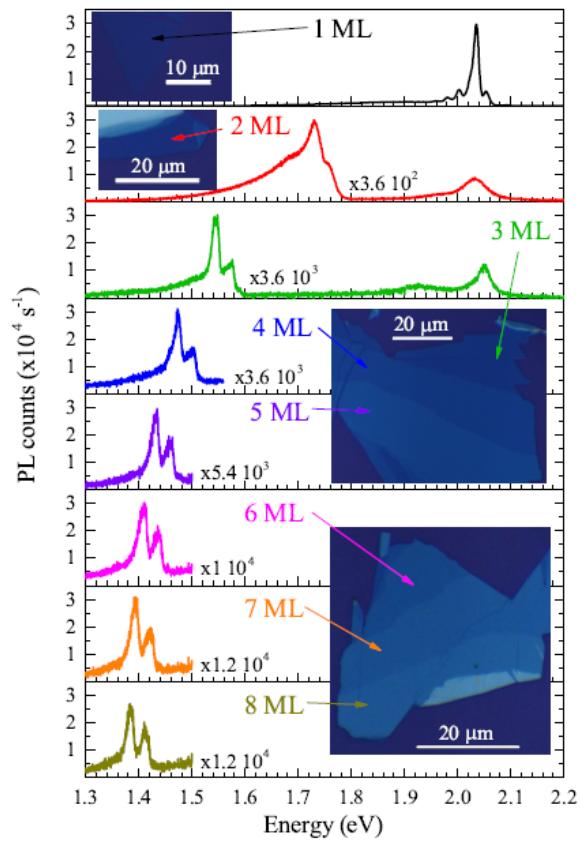
$\text{MoSe}_2$



$\text{WSe}_2$



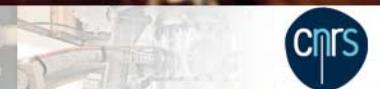
$\text{WS}_2$



A. Arora *et al.*  
 Nanoscale (2015)

A. Arora *et al.*  
 Nanoscale (2015)

M. Molas *et al.*  
 to be published



# 2D materials → (1D), 0D materials ?

quantum (single electron) electronics (**graphene**) ✓  
nano – lithography (~100 nm) + electrostatic gating

quantum (single photon) photonics  
single objects (~10nm) ?

making profit of imperfections !



# 2D → 0D (past experience)

## semiconductor heterojunctions/quantum wells : interface imperfections (lattice mismatch)

VOLUME 66, NUMBER 23

PHYSICAL REVIEW LETTERS

10 JUNE 1991

### Effect of Strain on Surface Morphology in Highly Strained InGaAs Films

C. W. Snyder, B. G. Orr, D. Kessler, and L. M. Sander

*H. M. Randall Laboratory, University of Michigan, Ann Arbor, Michigan 48109-1120*

(Received 28 February 1991)

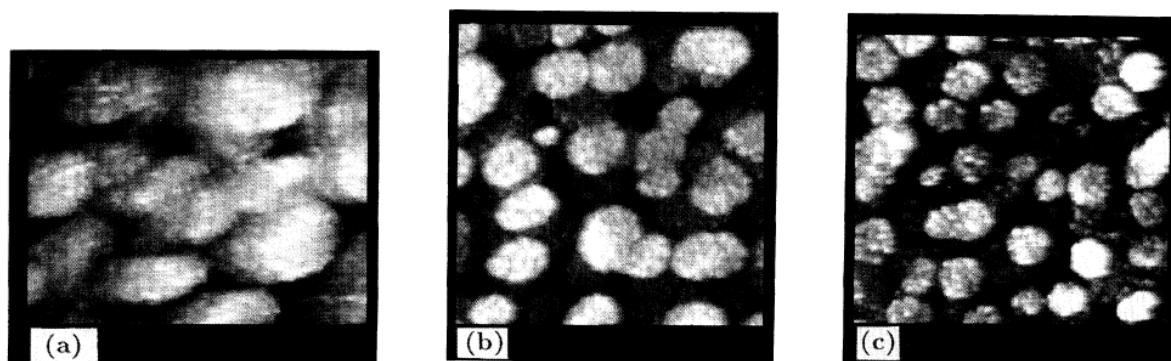


FIG. 3. STM images of  $\text{In}_x\text{Ga}_{1-x}\text{As}$  on  $\text{GaAs}(100)$ . (a) 14 ML's,  $x=0.3$  (2.2% mismatch), scan range =  $154 \text{ nm} \times 130 \text{ nm}$ , full-scale height range (from black to white) is  $8.0 \text{ nm}$ ,  $V_t=2.6 \text{ V}$ ,  $I_t=0.4 \text{ pA}$ . (b) 11 ML's,  $x=0.4$  (2.9% mismatch), scan range =  $150 \text{ nm} \times 154 \text{ nm}$ , full-scale height range is  $8.5 \text{ nm}$ ,  $V_t=2.6 \text{ V}$ ,  $I_t=60 \text{ pA}$ . (c) 12 ML's,  $x=0.5$  (3.6% mismatch),  $150 \text{ nm} \times 147 \text{ nm}$ , full-scale height range is  $9.0 \text{ nm}$ ,  $V_t=2.7 \text{ V}$ ,  $I_t=20 \text{ pA}$ . For the films represented in (b) and (c), 4–5 ML's of additional material were deposited after the RHEED pattern became spotty. During this extra growth the pattern sharpened. For the film represented in (a) there was no extra growth.

# 2D → 0D (past experience)

## semiconductor heterojunctions/quantum wells : self assembled InAs/GaAs quantum dots

### Photoluminescence of Single InAs Quantum Dots Obtained by Self-Organized Growth on GaAs

J.-Y. Marzin, J.-M. Gérard, A. Izraël, and D. Barrier

France Telecom, Centre National d'Etudes des Télécommunications-PAB, Laboratoire de Bagneux, BP107,  
F92225 Bagneux, France

G. Bastard

Laboratoire de Physique de la Matière Condensée, Ecole Normale Supérieure, 24 rue Lhomond, F75005 Paris, France  
(Received 11 March 1994)

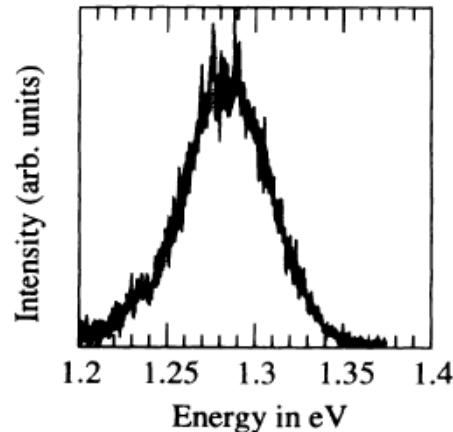


FIG. 1. 10 K PL spectrum of a 5000 nm mesa in sample A.

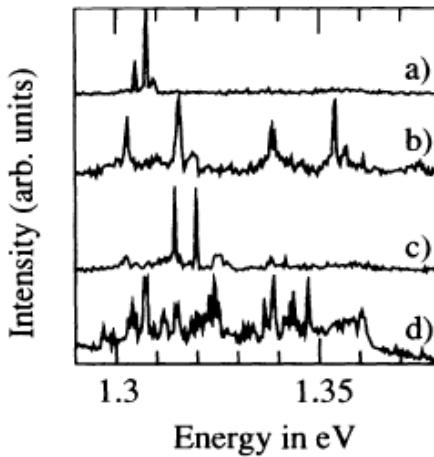


FIG. 5. (a), (b), and (c): 10 K PL spectra of three different 200 nm mesas of sample B. (d) sum of 20 spectra recorded on different such mesas.

# 2D → 0D (past experience)

## semiconductor heterojunctions/quantum wells : well width fluctuations (monolayer steps)



### Quantum Dots Formed by Interface Fluctuations in AlAs/GaAs Coupled Quantum Well Structures

A. Zrenner, L. V. Butov,\* M. Hagn, G. Abstreiter, G. Böhm, and G. Weimann  
*Walter Schottky Institut, Technische Universität München, D-85748 Garching, Germany*  
(Received 21 December 1993)

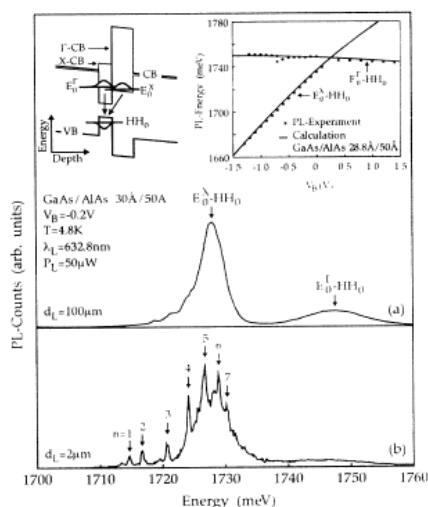


FIG. 1. (a) PL response of a GaAs/AlAs 30 Å/50 Å coupled QW structure in the indirect regime for  $V_B = -0.2 \text{ V}$ . The diameter of the optically probed area  $d_L$  is 100  $\mu\text{m}$ . A schematic band diagram of the structure and the observed PL energies as a function of  $V_B$  are shown in the inset. (b) Same as (a) only with higher spatial resolution ( $d_L = 2 \mu\text{m}$ ). New narrow emission lines (labeled from  $n=1$  to 7) appear in the region of the indirect PL.

### Fine Structure Splitting in the Optical Spectra of Single GaAs Quantum Dots

D. Gammon, E. S. Snow, B. V. Shanabrook, D. S. Katzer, and D. Park  
*Naval Research Laboratory, Washington, D.C. 20375-5347*  
(Received 17 August 1995)

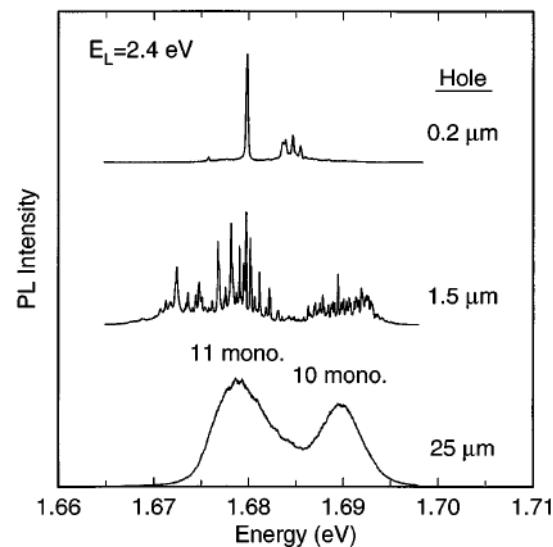


FIG. 2. Nonresonant PL spectra excited and detected through apertures with diameters listed.

# 2D → 0D (past experience)

## semiconductor heterojunctions/quantum wells : well width fluctuations (monolayer steps)



### Quantum Dots Formed by Interface Fluctuations in AlAs/GaAs Coupled Quantum Well Structures

A. Zrenner, L. V. Butov,\* M. Hagn, G. Abstreiter, G. Böhm, and G. Weimann  
*Walter Schottky Institut, Technische Universität München, D-85748 Garching, Germany*  
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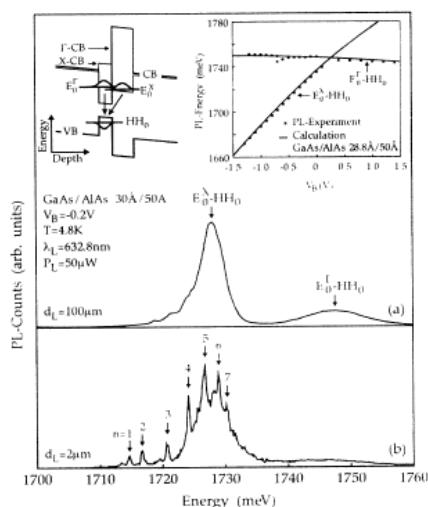


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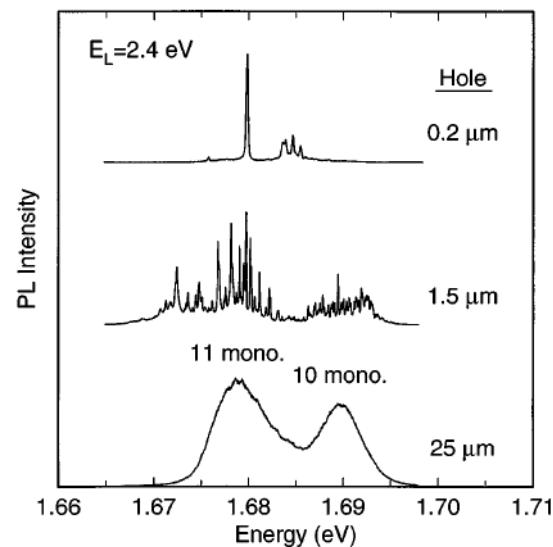
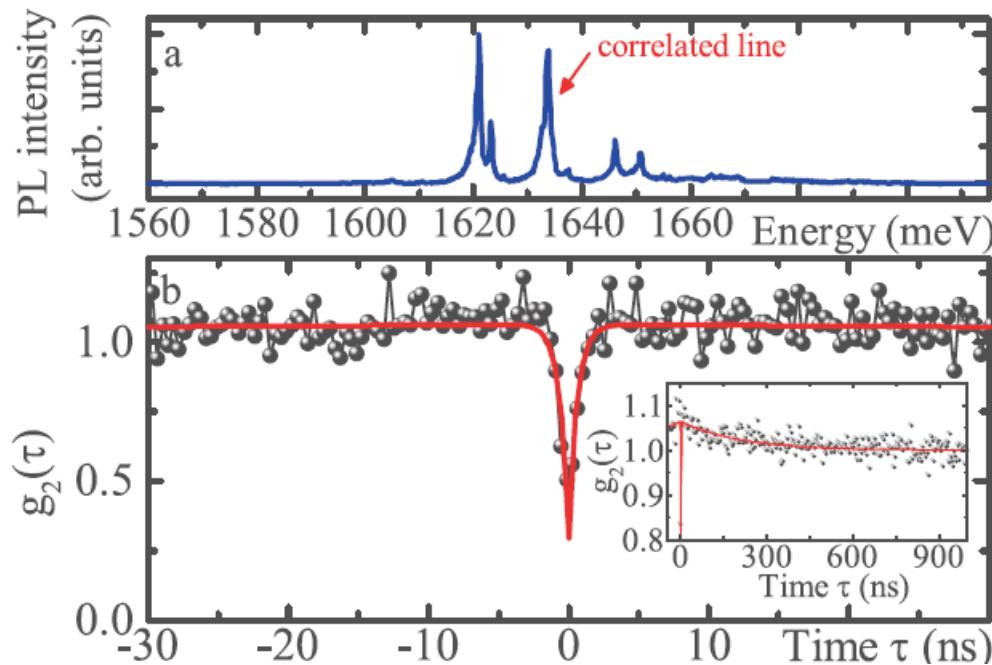


FIG. 2. Nonresonant PL spectra excited and detected through apertures with diameters listed.



# Single photon sources (narrow emission lines, ~0.1meV)) fingerprint: photon antibunching photon correlation measurements



semiconductor (single) quantum dots, nanocrystals (colloidal dots)  
NV centers in diamond (SiC), organic molecules  
interesting applications; issues: stability, room temperature operation, electrical pumping



# Single photon emitters in WSe<sub>2</sub> (MX<sub>2</sub>) flakes ?

( Nature Nanotechnology, 2015)

A. Srivastava et al., Optically active quantum dots in monolayer WSe<sub>2</sub> (Switzerland)

M. Koperski et al., Single photon emitters in exfoliated WSe<sub>2</sub> structures (France, Poland)

Y-M, He et al., Single quantum emitters in monolayer semiconductors (China, USA)

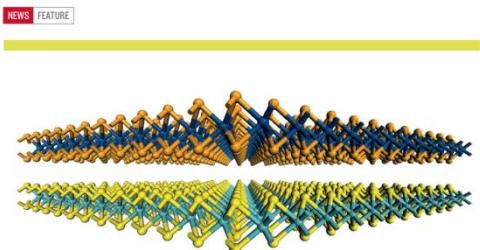
C. Chakraborty et al., Voltage controlled of quantum light from an atomically thin semiconductor (USA)

news & views

METAL DICHALCOGENIDES

## Two dimensions and one photon

Single-photon sources have been demonstrated in two-dimensional semiconductors.



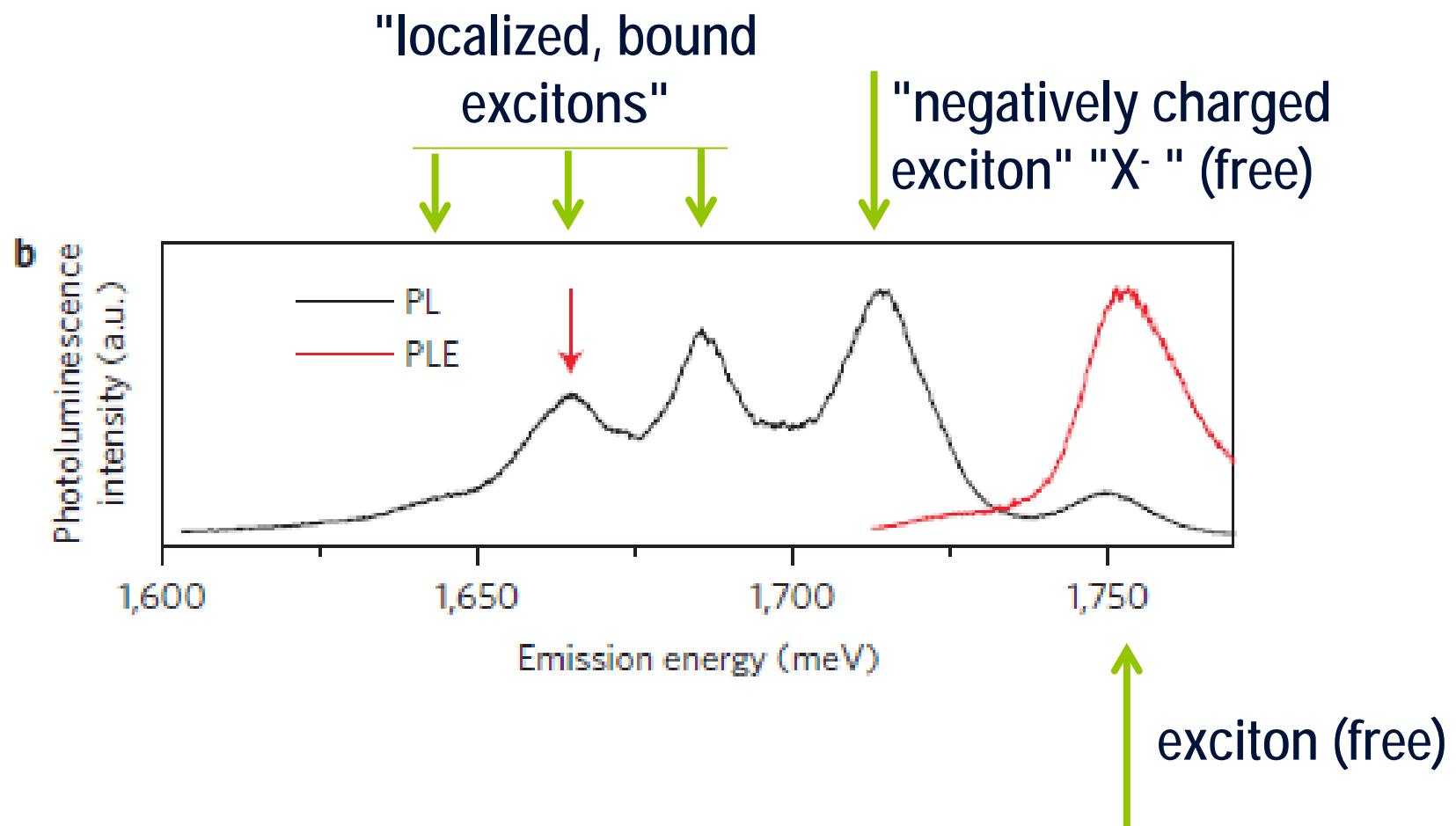
2D OR NOT 2D



# 2D WSe<sub>2</sub> monolayer

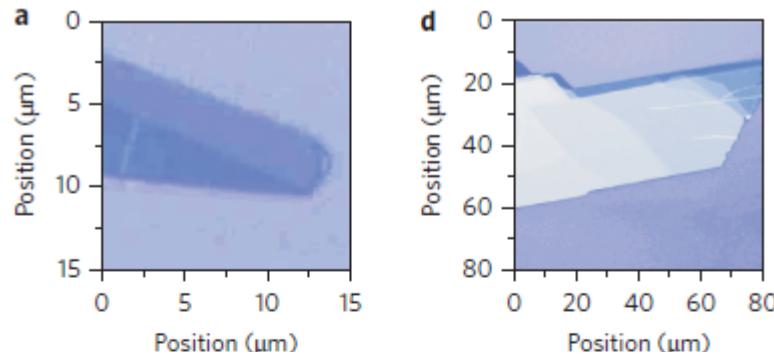
typical photoluminescence/absorption response

broad (10-20meV) emission/absorption peaks

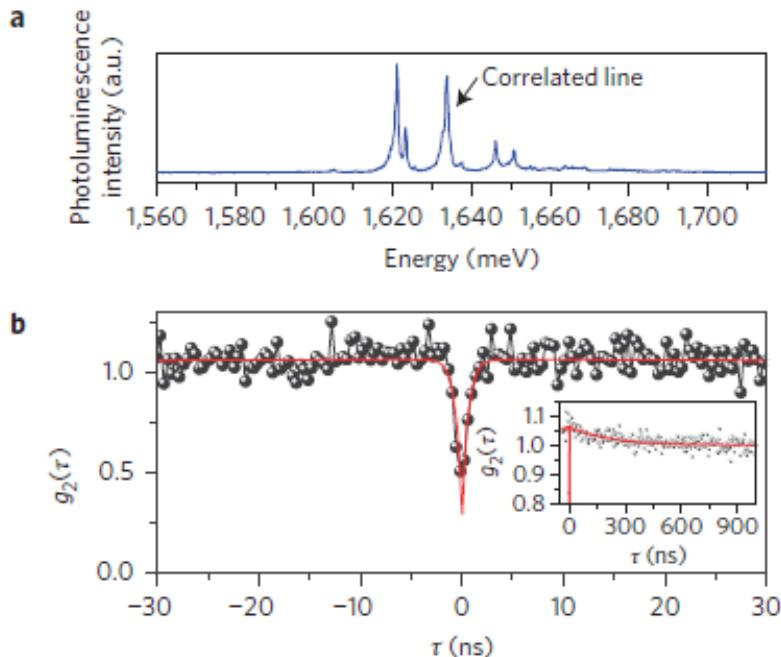




# M. Koperski et al., Single photon emitters in exfoliated WSe<sub>2</sub> structures



( Imperfections at edges ? )



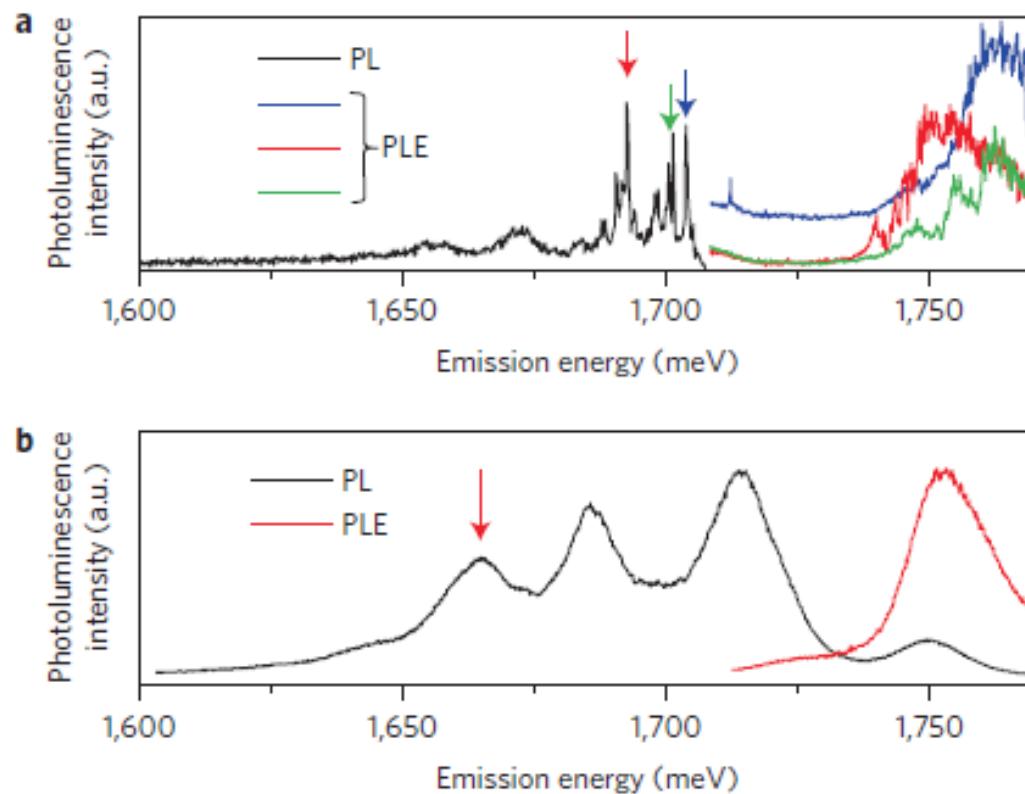
## Photon antibunching

=

an unambiguous attribute  
of single photon emitters

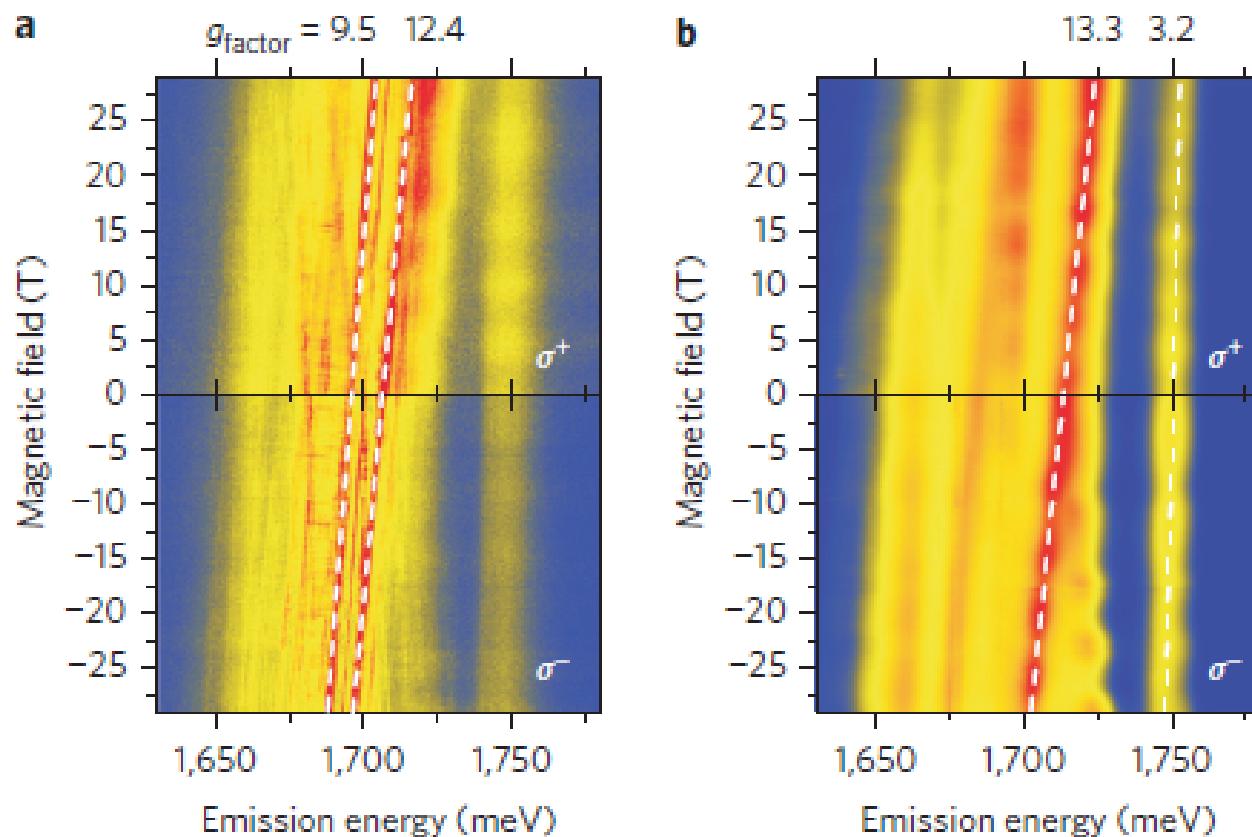


## Optical response of narrow lines , linked to the 2D properties of a WSe<sub>2</sub> monolayer

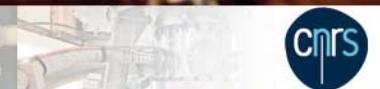


Emission in the same energy range, similar excitation spectra

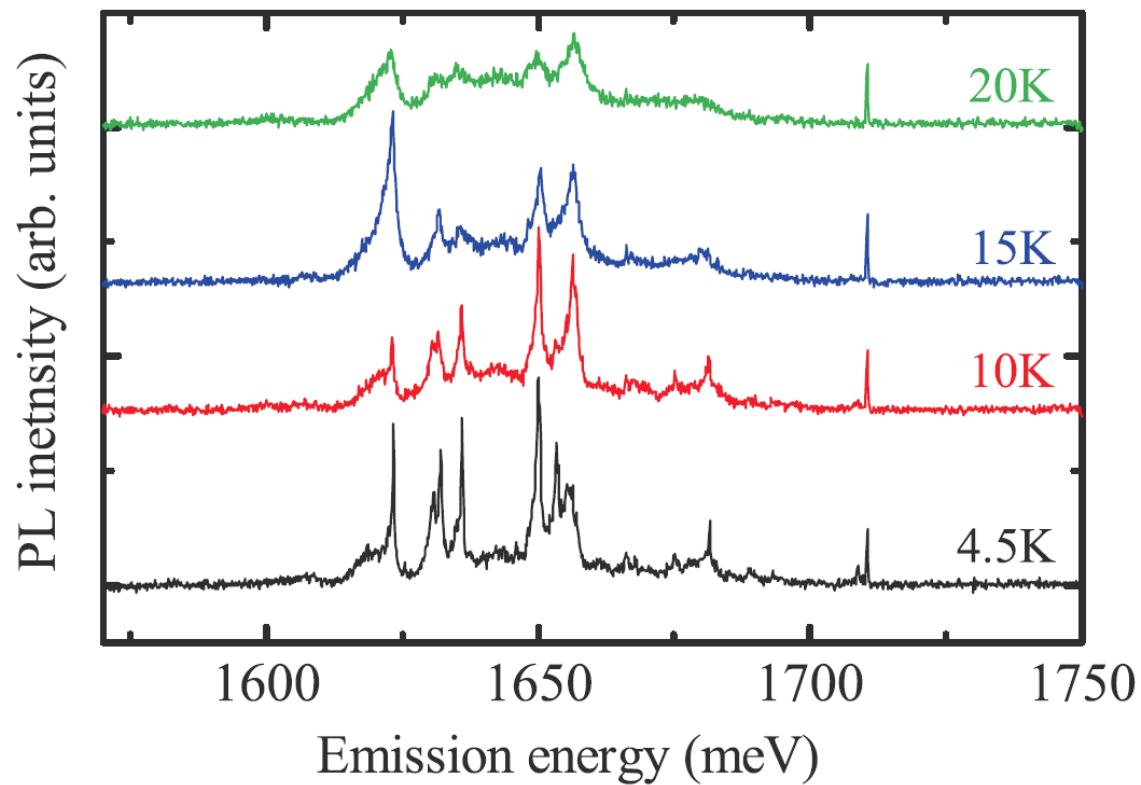
# Optical response of narrow lines , linked to the 2D properties of a WSe<sub>2</sub> monolayer



Similar, anomalously large "Zeeman" splitting



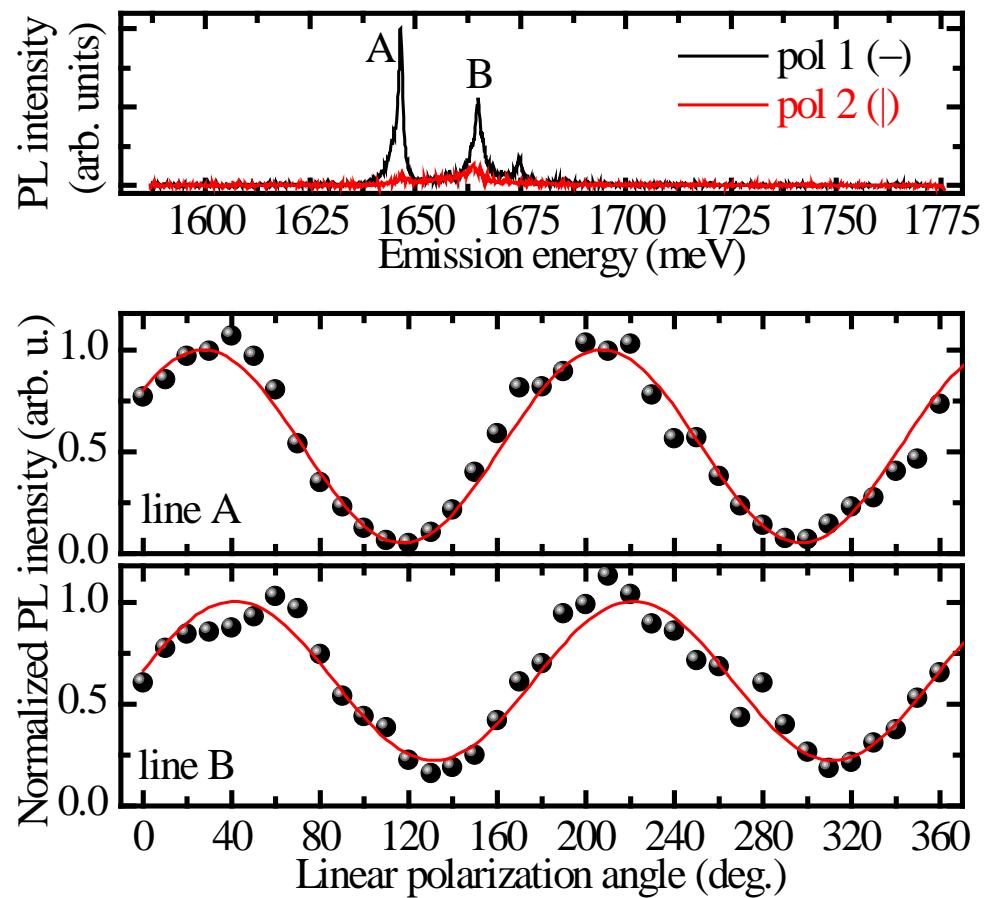
Temperature effect:



Quench of intensity (weakly confined electron hole pairs ?)  
Broadening (similar to acoustic phonon effect in semiconductor QDs)



Pronounced (sometimes) linear anisotropy:



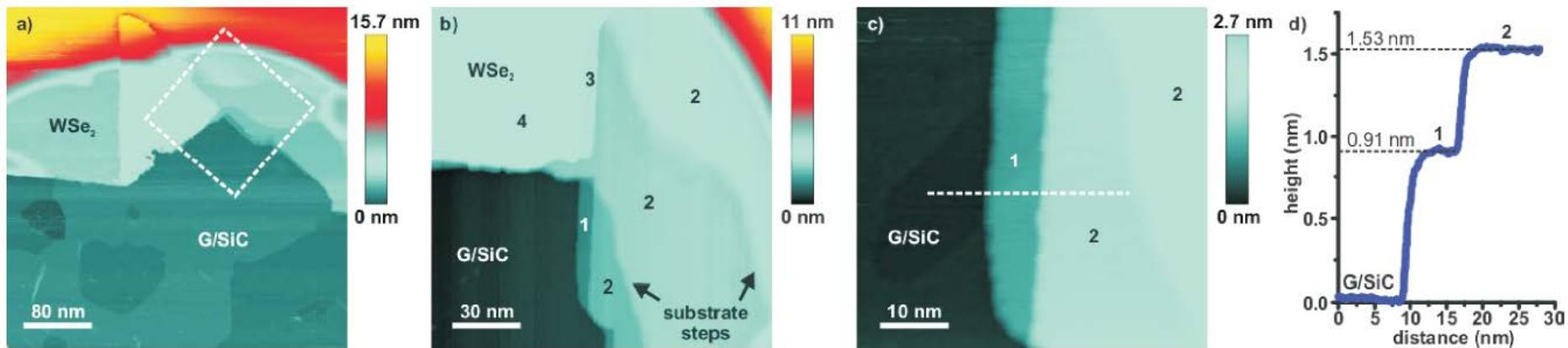
Similar to fine structure splitting in QDots with anisotropic shape



## What are they ?

Monolayer "nanoflakes" at the edges of monolayers and thicker flakes ???

STM: Monolayer nanoflake at the edge of a thicker flake

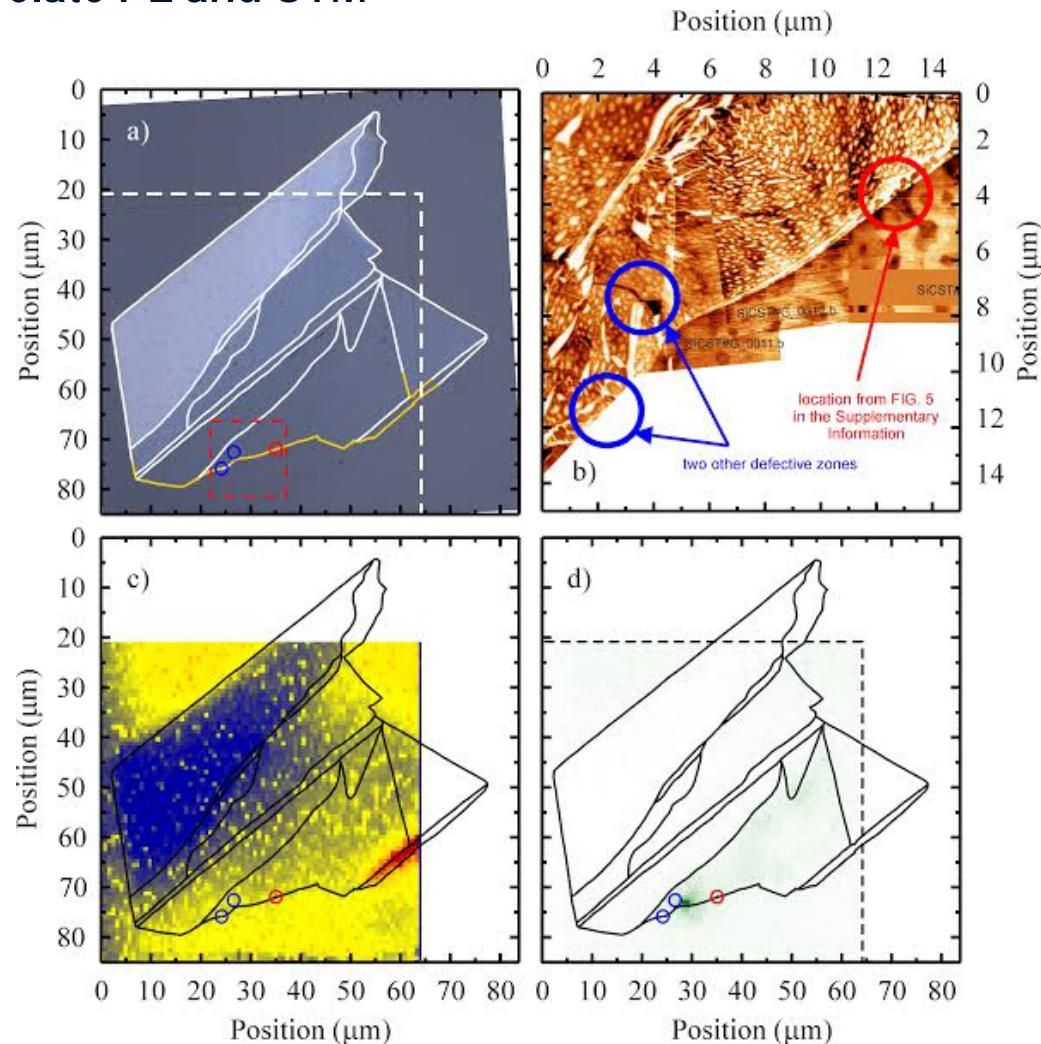




# What are they ?

Monolayer "nanoflakes" at the edges of monolayers and thicker flakes ?

An attempt to correlate PL and STM



# Flow of works :

- [117] Kumar S, Kaczmarczyk A, Gerardot BD. Strain-Induced Spatial and Spectral Isolation of Quantum Emitters in Mono- and Bilayer WSe<sub>2</sub>. *Nano Lett* 2015;15:7567–7573.
- [118] Clark G, Schaibley JR, Ross J, Taniguchi T, Watanabe K, Hendrickson JR, Mou S, Yao W, Xu X. Single Defect Light-Emitting Diode in a van der Waals Heterostructure. *Nano Lett* 2016;16:3944–3948.
- [119] Schwarz S, Kozikov A, Withers F, Maguire JK, Foster AP, Dufferwiel S, Hague L, Makhonin MN, Wilson LR, Geim AK, Novoselov KS, Tartakovskii AI. Electrically pumped single-defect light emitters in WSe<sub>2</sub>. *2D Mater* 2016;3:25038.
- [120] He Y-M, Iff O, Lundt N, Baumann V, Davanco M, Srinivasan K, Hofling S, Schneider C. Cascaded emission of single photons from the biexciton in monolayered WSe<sub>2</sub>. *Nat Commun* 2016;7:13409.
- [121] Palacios-Berraquero C, Barbone M, Kara DM, Chen X, Goykhman I, Yoon D, Ott AK, Beitner J, Watanabe K, Taniguchi T, Ferrari AC, Atature M. Atomically thin quantum light-emitting diodes. *Nat Commun* 2016;7:12978.
- [122] Palacios-Berraquero C, Kara DM, Montblanch AR-P, Barbone M, Latawiec P, Yoon D, Ott AK, Loncar M, Ferrari AC, Atature M. Large-scale quantum-emitter arrays in atomically thin semiconductors. *ArXiv E-Prints* 2016;Preprint at arXiv:1609.04244.
- [123] Kern J, Niehues I, Tonndorf P, Schmidt R, Wigger D, Schneider R, Stiehm T, Michaelis de Vasconcellos S, Reiter DE, Kuhn T, Bratschitsch R. Nanoscale Positioning of Single-Photon Emitters in Atomically Thin WSe<sub>2</sub>. *Adv Mater* 2016;28:7101–7105.
- [124] Branny A, Wang G, Kumar S, Robert C, Lassagne B, Marie X, Gerardot BD, Urbaszek B. Discrete quantum dot like emitters in monolayer MoSe<sub>2</sub>: Spatial mapping, magneto-optics, and charge tuning. *Appl Phys Lett* 2016;108:142101.



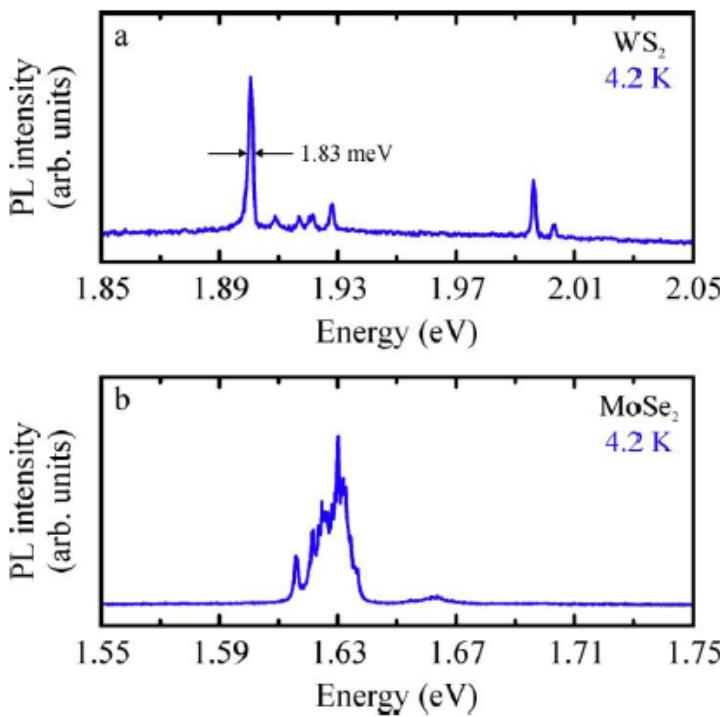
# Sharp emission lines in other S-TMDs ? Electric excitation ?

C. Palacios-Berraquero *et al.*, Nat. Commun. (2016)

A. Branny *et al.*, APL (2016)

S. Schwarrz *et al.*, 2D Materials (2016)

.....



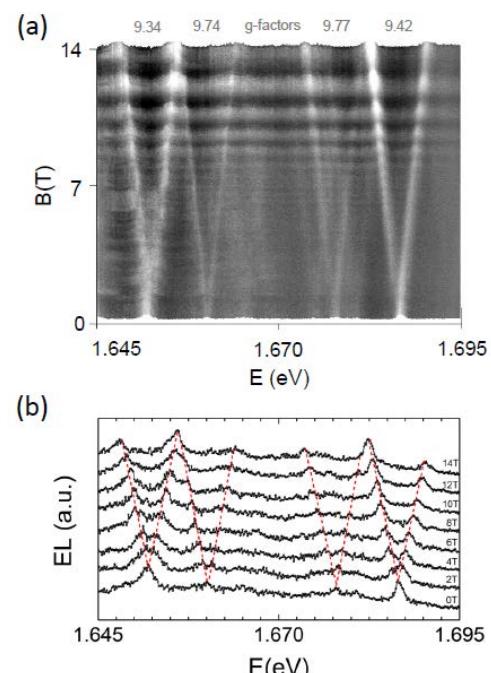
M. Molas, M. Koperski *et al.*,  
unpublished data



J. Binder *et al.*,  
unpublished data

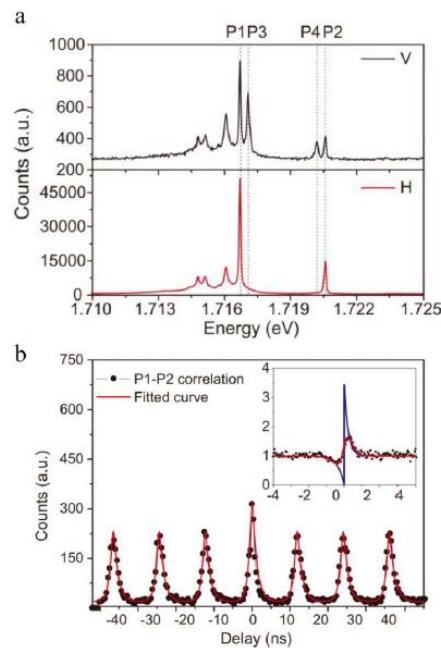


electroluminescence  
hBN/WSe<sub>2</sub>/hBN LED





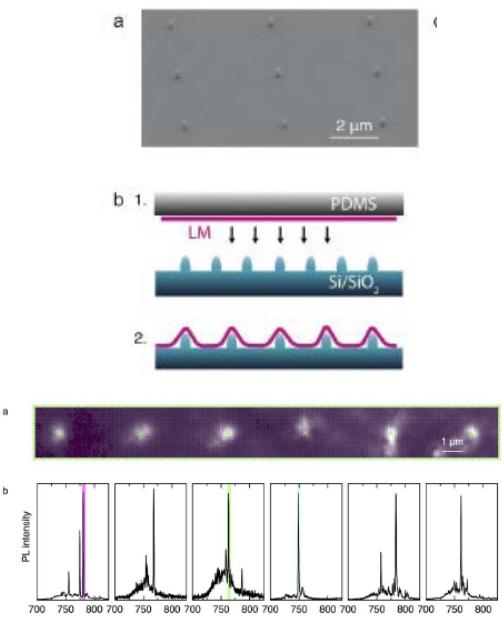
# 3D confinement (or defects ?)



biexciton-exciton cascade ?

Y-M. He *et al.*,  
Nat. Commun. (2016)

Control,  
on demand  
- strain engineering



C. Palacios-Berraquero *et al.*,  
arXiv (2016)

J. Kern *et al.*, Adv. Mater. (2016)



# hexagonal boron nitride (hBN): yet another class of single photon emitters

T.T. Train *et al.*, Nature Nanotech. (2015)

...

N.R. Jungwirth *et al.*, Nano Lett. (2016)

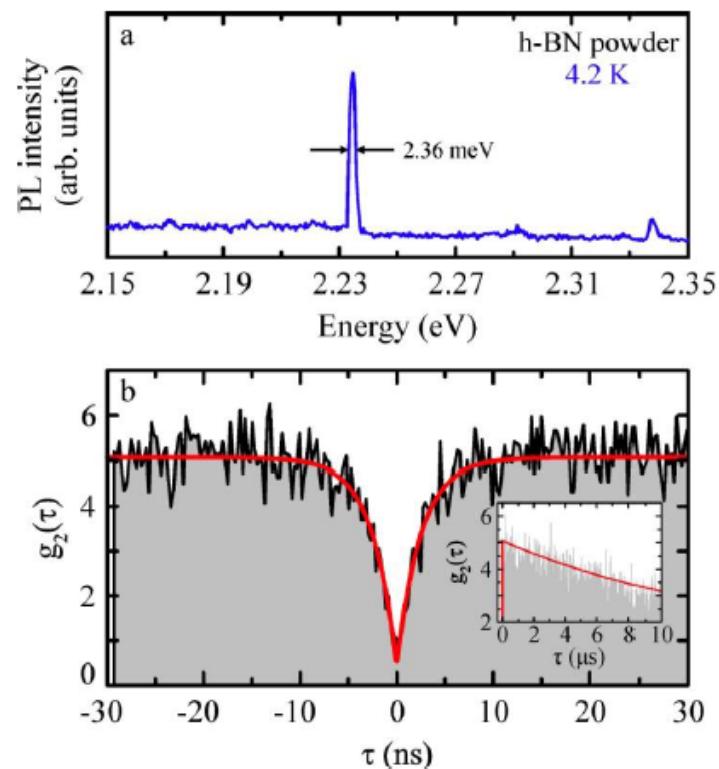
A.W. Shell, et al., APL Photonics (2016)

L.J. Martinez, *et al.*, PRB(2016)

N. Chejanovsky *et al.*, arXiv (2016)

A.L. Exarhos *et al.*, arXiv (2016)

...



M. Koperski et al  
to be published



- [125] Tran TT, Bray K, Ford MJ, Toth M, Aharonovich I. Quantum emission from hexagonal boron nitride monolayers. *Nat Nanotechnol* 2015;11:37–41.
- [126] Tran TT, Zachreson C, Berhane AM, Bray K, Sandstrom RG, Li LH, Taniguchi T, Watanabe K, Aharonovich I, Toth M. Quantum Emission from Defects in Single-Crystalline Hexagonal Boron Nitride. *Phys Rev Appl* 2016;5:34005.
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- [128] Tran TT, Elbadawi C, Totonjian D, Lobo CJ, Grosso G, Moon H, Englund DR, Ford MJ, Aharonovich I, Toth M. Robust Multicolor Single Photon Emission from Point Defects in Hexagonal Boron Nitride. *ACS Nano* 2016;10:7331–7338.
- [129] Schell AW, Tran TT, Takashima H, Takeuchi S, Aharonovich I. Non-linear excitation of quantum emitters in hexagonal boron nitride multiplayers. *APL Photonics* 2016;1:91302.
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- [131] Martínez LJ, Pelini T, Waselowski V, Maze JR, Gil B, Cassabois G, Jacques V. Efficient single photon emission from a high-purity hexagonal boron nitride crystal. *Phys Rev B* 2016;94:121405.
- [132] Chejanovsky N, Rezai M, Paolucci F, Kim Y, Rendler T, Rouabeh W, de Oliveira FF, Herlinger P, Denisenko A, Yang S, Gerhardt I, Finkler A, et al. Topological attributes and photo-dynamics of visible spectrum quantum emitters in hexagonal boron nitride. *Nano Lett* 2016;16:7037-7045.
- [133] Exarhos AL, Hopper DA, Grote RR, Alkauskas A, Bassett LC. Optical Signatures of Quantum Emitters in Suspended Hexagonal Boron Nitride. *ArXiv E-Prints* 2016:Preprint at arXiv:1609.02641.



# Déjà vu again

## Single photon emitters in sc-TMDs

### quantum dot like objects

## Single photon emitters in hBN

### similar to NV centers in diamond/SiC



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