

*Domes made of two
dimensional crystals:
magneto- and
quantum-optical properties*

Antonio Polimeni

Dipartimento di Fisica



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UNIVERSITÀ DI ROMA



Acknowledgements I



Salvatore
Cianci

Marzia
Cuccu

Atanu
Patra

Marco
Felici

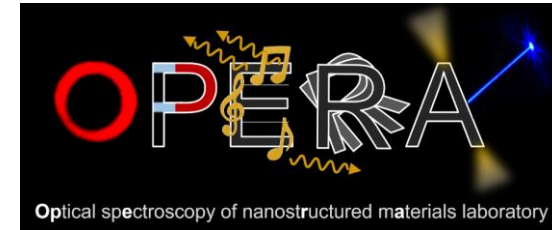
Elena
Blundo

Federico
Tuzi



CNR IFN
Istituto di Fotonica e Nanotecnologie

Dr. Giorgio Pettinari



Prof. Antonio Polimeni
Prof. Marco Felici
Elena Blundo
Antonio Miriametro
Marzia Cuccu
Federico Tuzi
Eirini Parmenopoulou
Djeero Peters

Acknowledgements II

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Marco Felici, Marzia Cuccu
Salvatore Cianci, Federico Tuzi
Antonio Miriametro



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Acknowledgements II

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University of Salerno, Fisciano, Italy

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Katarzyna Olkowska-Pucko Tomasz Kazimierczuk Adam Babiński Maciej Molas

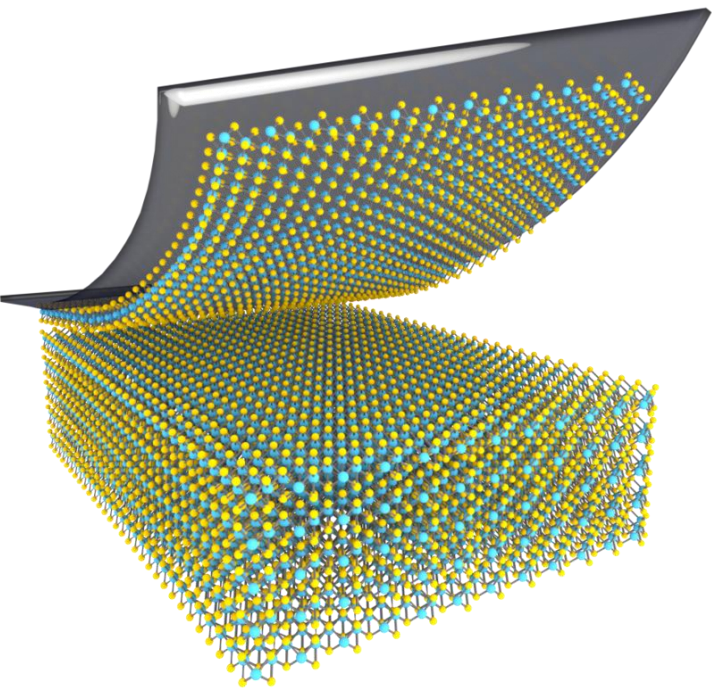


Institute of Experimental Physics, Faculty of Physics,
University of Warsaw, Poland

Outline

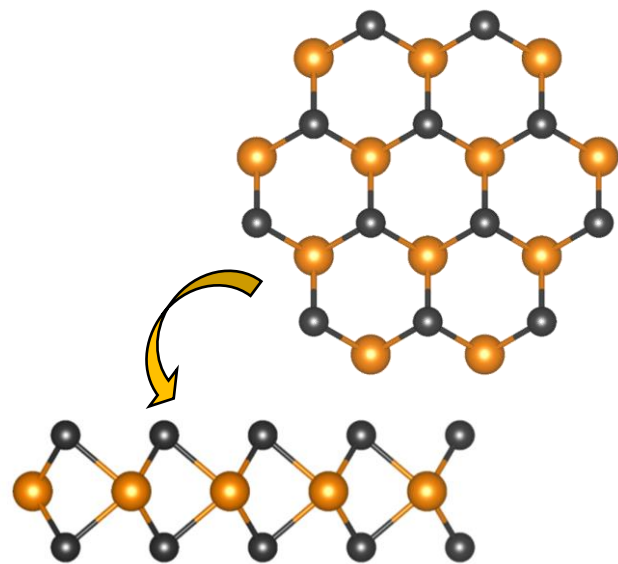
- Strain in 2D crystals
- Formation, characteristics and control of artificial domes in exfoliable materials
- Strain fields in curved membranes: optical, vibrational and magneto-optical properties
- Applications for site-controlled quantum light sources

Two-dimensional crystals

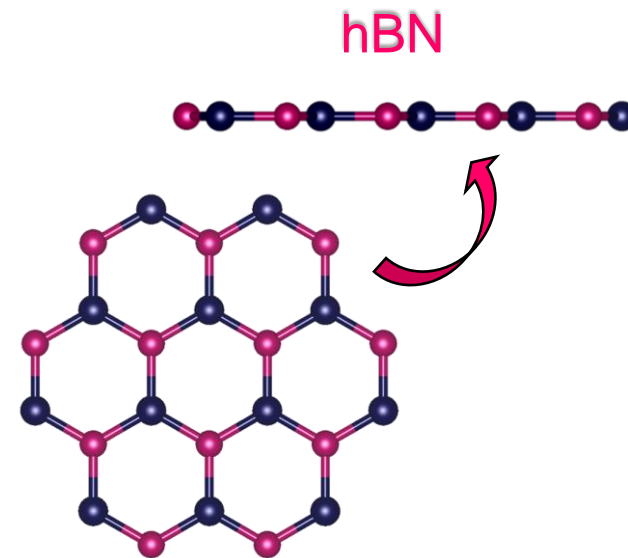


M: Mo, W

X: S, Se, Te

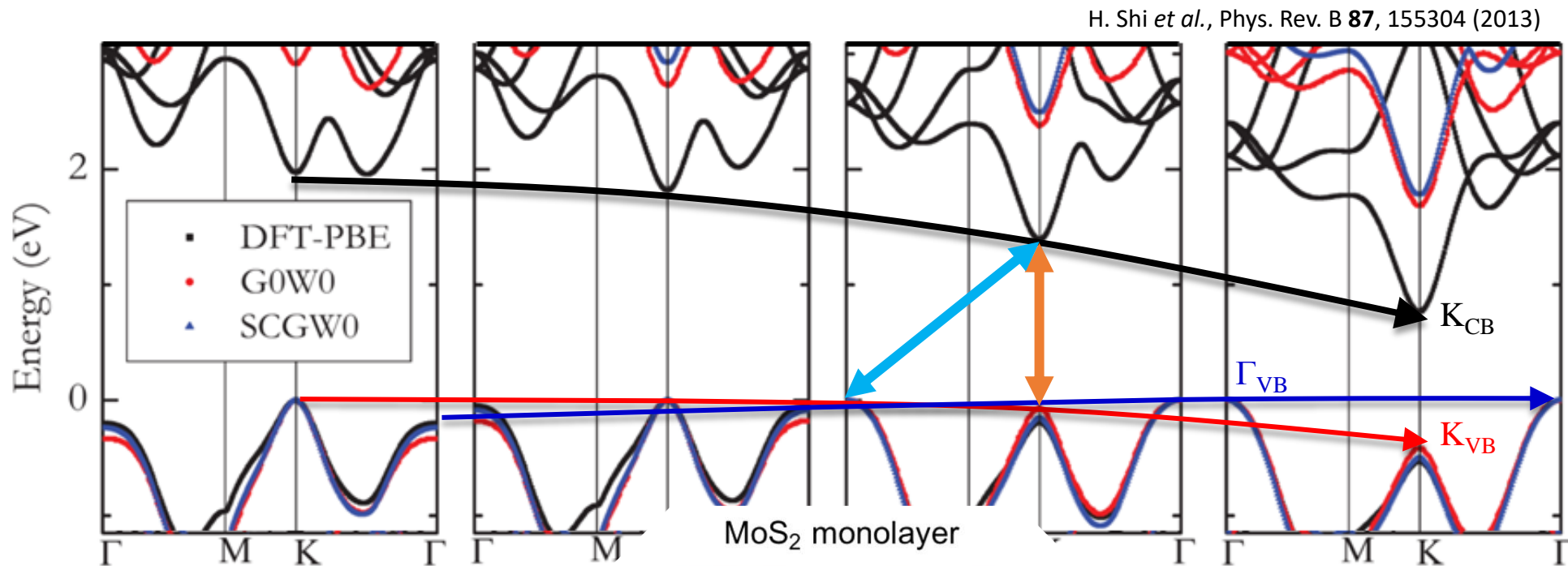
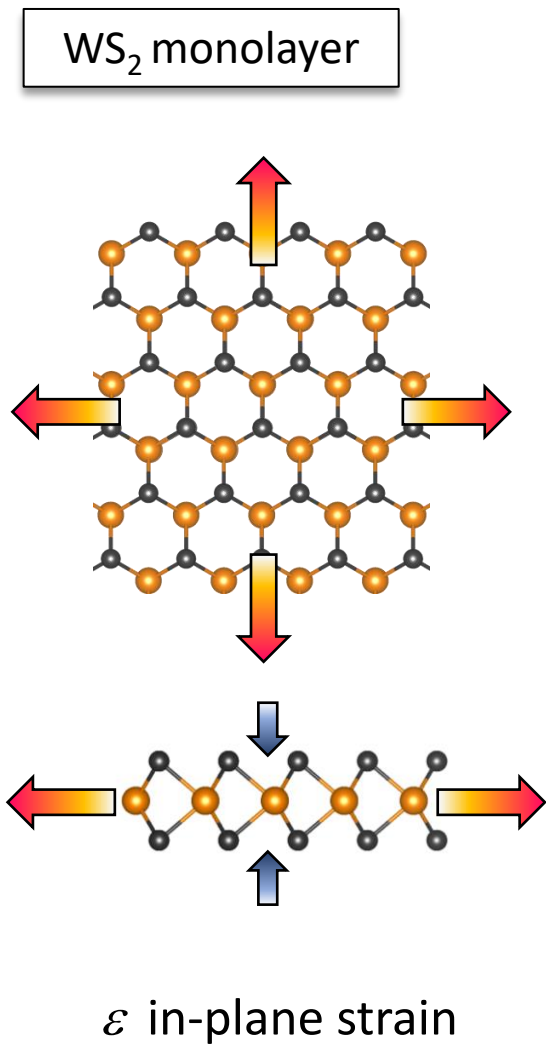


TMDs



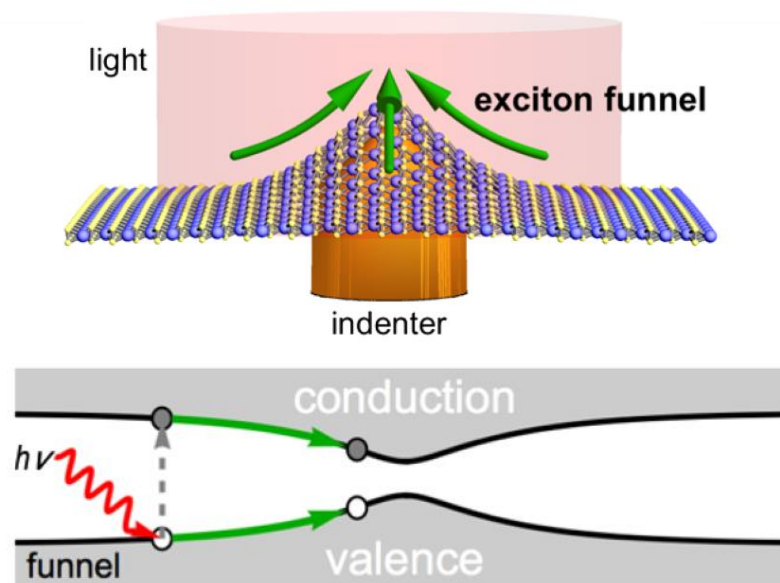
hBN

Why strain?

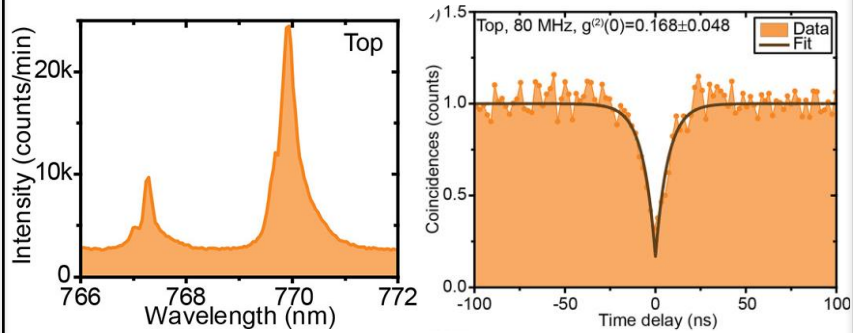
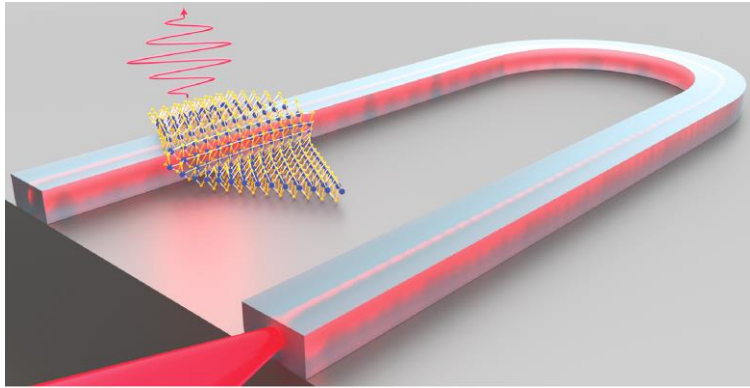


$\epsilon = 0\%$

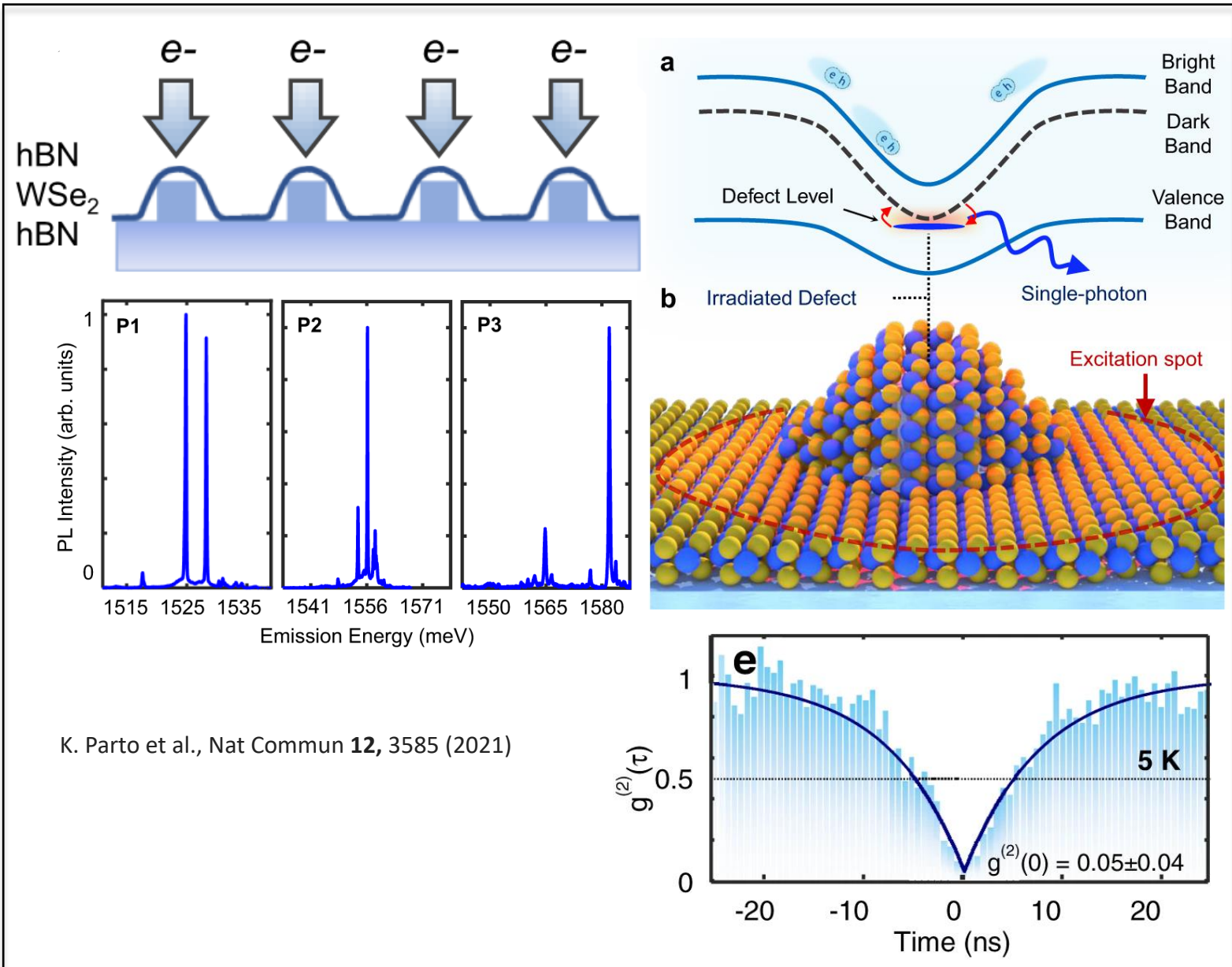
$\epsilon = 12\%$



Why strain?



Carlos Errando-Herranz et al., ACS Photonics **8**, 1069 (2021)

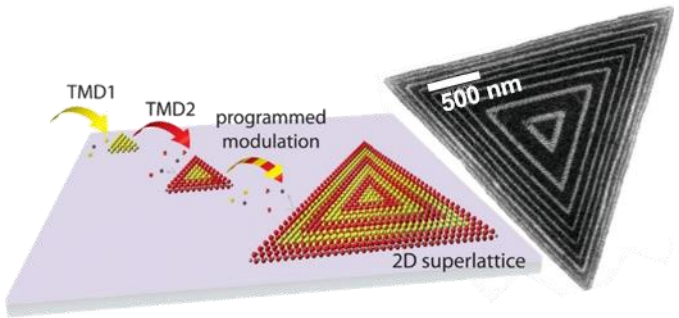


K. Parto et al., Nat Commun **12**, 3585 (2021)

Straining methods

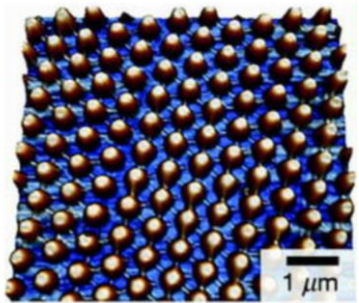
E. Blundo et al.
Appl. Phys. Rev. **8**, 021318 (2021)

Growth of superlattices

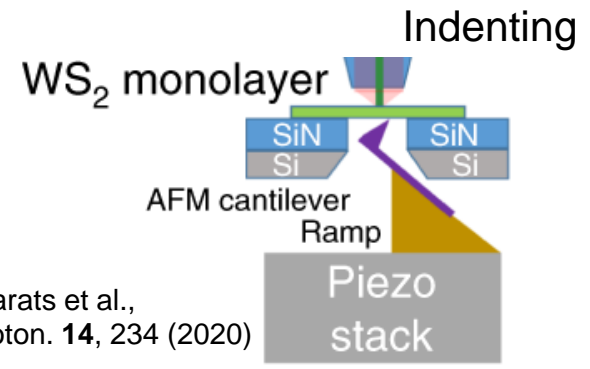


Xie et al., Science **359**, 1131 (2020)

Deposition on nanocones/pillars

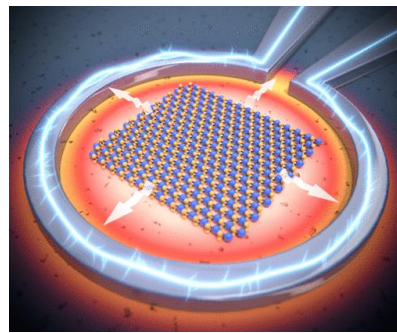


Li et al., J. Am. Chem. Soc. **138**, 5123 (2016)



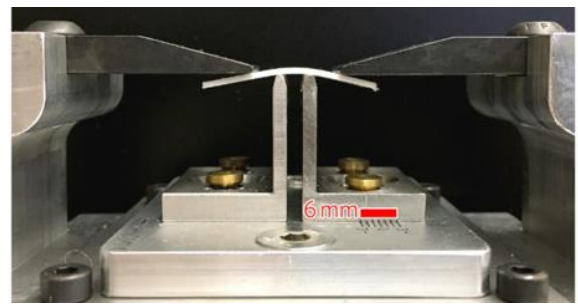
M. G. Harats et al.,
Nat. Photon. **14**, 234 (2020)

Stretching



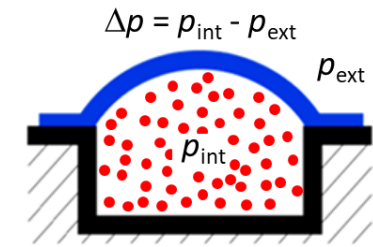
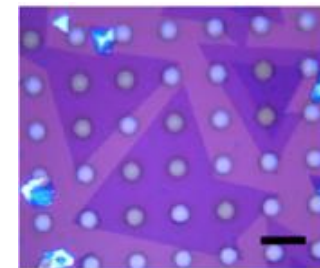
Ryu et al., Nano Lett. **20**, 5339 (2020)

Bending



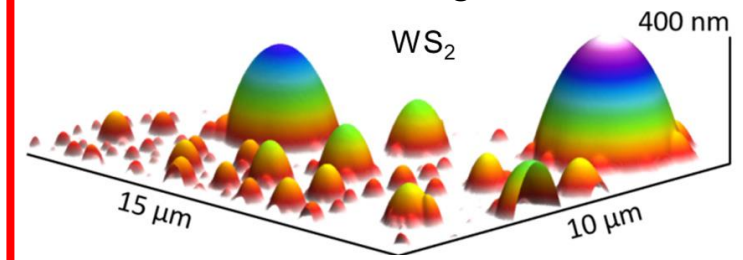
Wu et al., Nano Lett. **18**, 2351 (2018)

Bulging



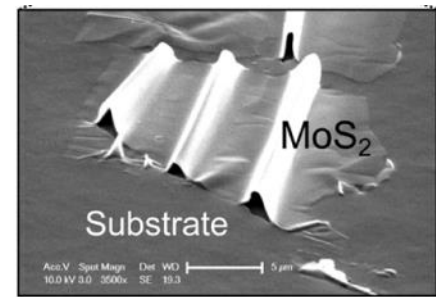
Lloyd et al., Nano Lett. **16**, 5836 (2016)

Bubbling



Blundo et al, Phys. Rev. Research **2**, 012024 (2020)

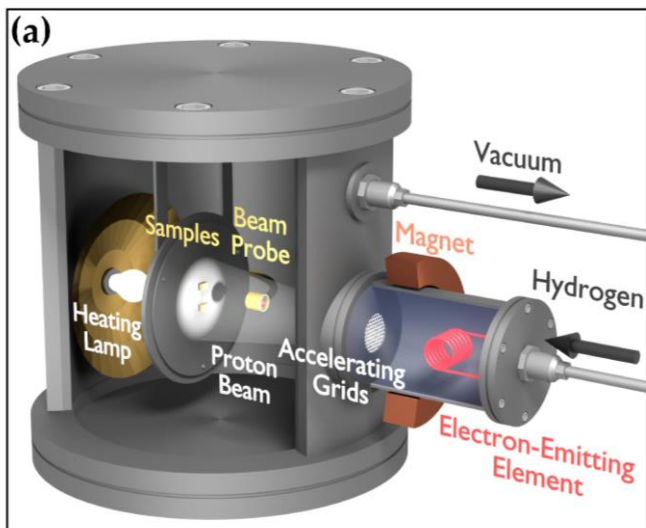
Wrinkling



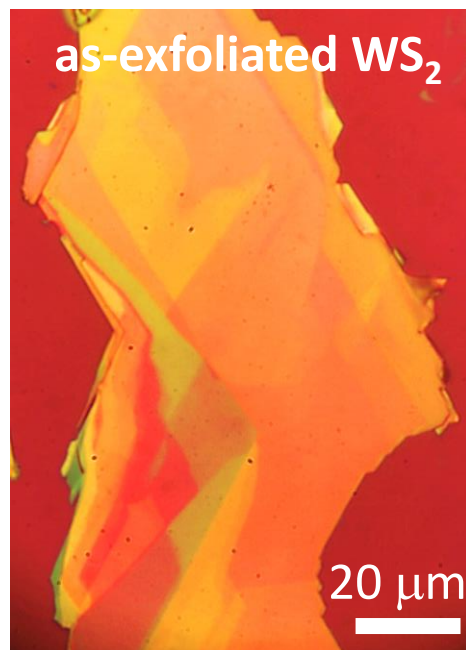
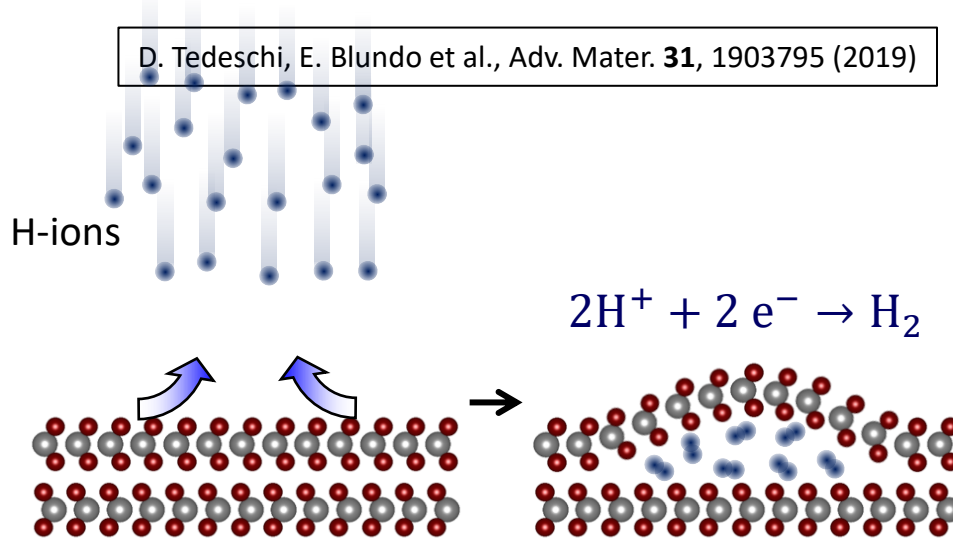
Castellanos-Gomez et al., Nano Lett. **13**, 5361 (2013)

Hydrogen irradiation of bulk TMDs

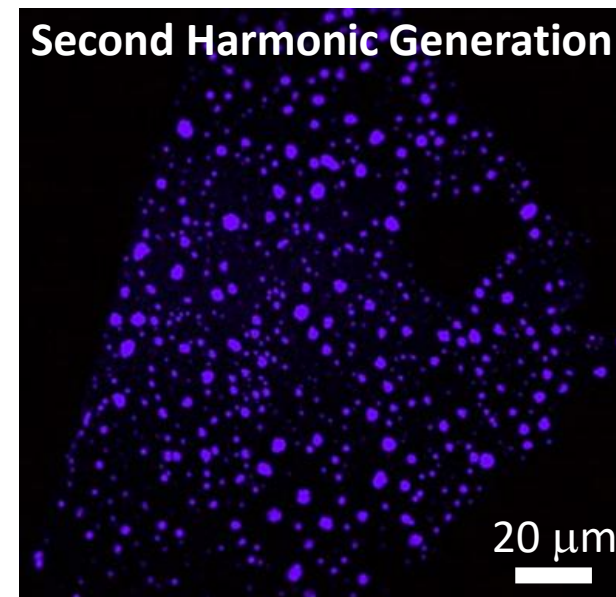
D. Tedeschi, E. Blundo et al., Adv. Mater. **31**, 1903795 (2019)



J. Felton, E. Blundo et al., Molecules **25**, 2526 (2020)

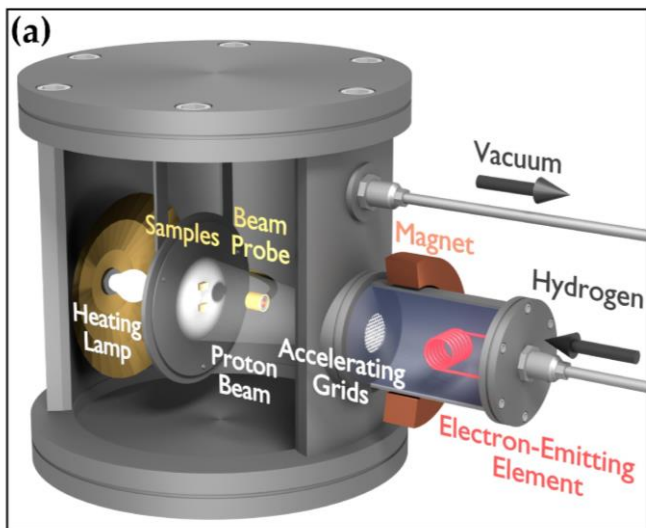


1 monolayer thick !

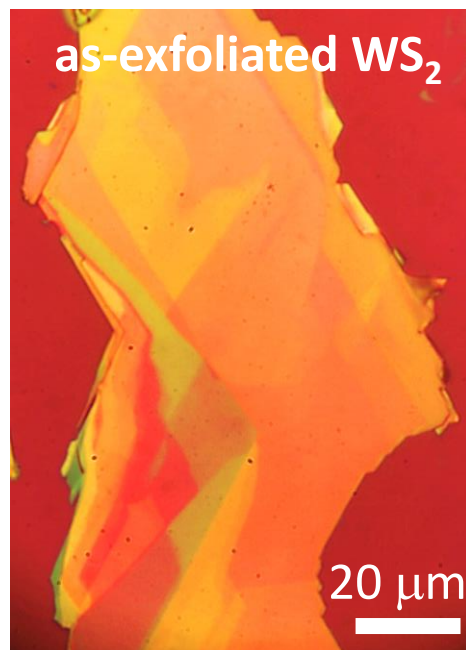
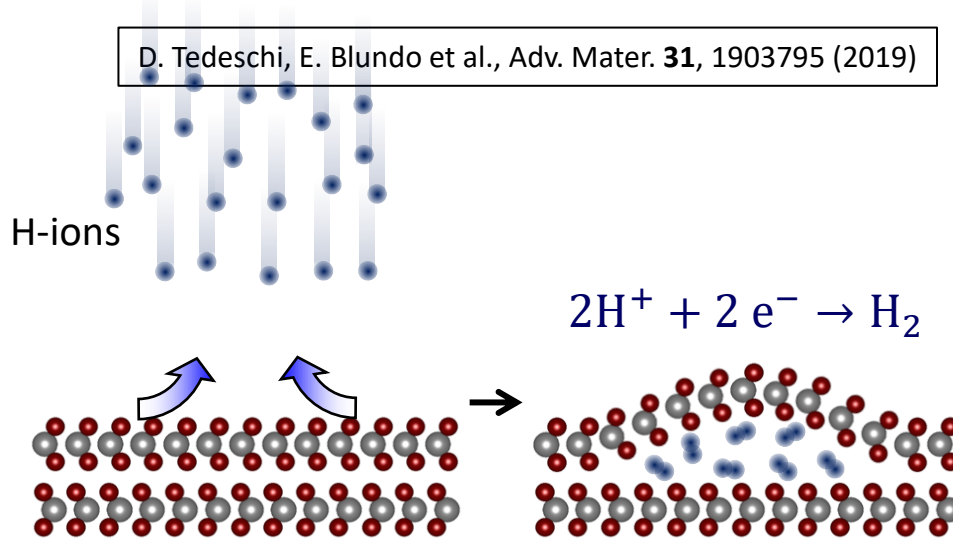


Hydrogen irradiation of bulk TMDs

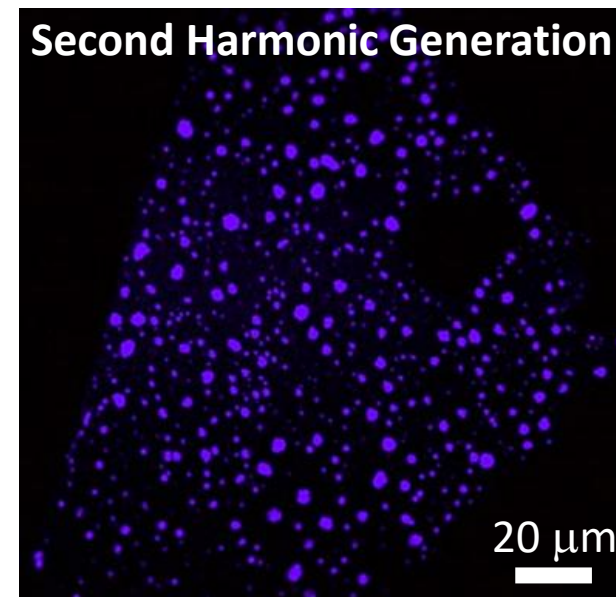
D. Tedeschi, E. Blundo et al., Adv. Mater. **31**, 1903795 (2019)



J. Felton, E. Blundo et al., Molecules **25**, 2526 (2020)



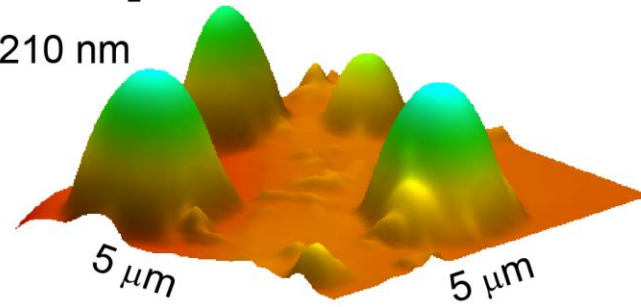
1 monolayer thick !



Domes in TMDs

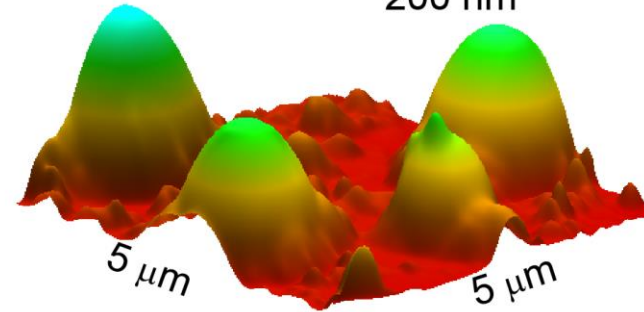
A WS_2

210 nm



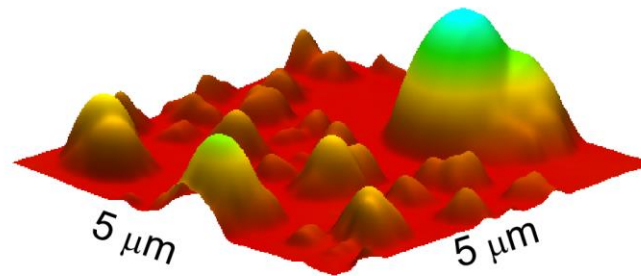
B MoS_2

200 nm



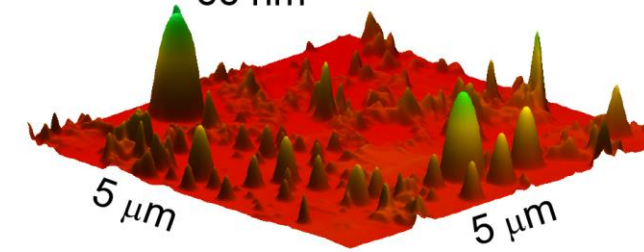
C WSe_2

150 nm



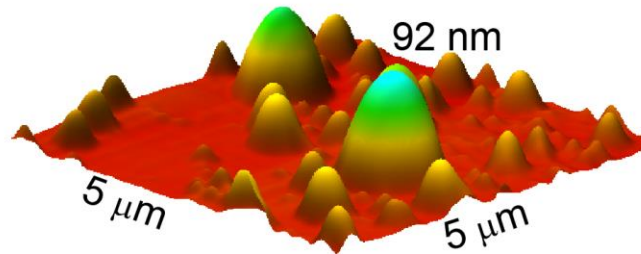
D MoSe_2

66 nm



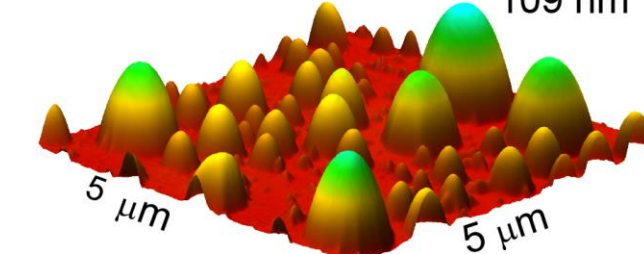
E WTe_2

92 nm

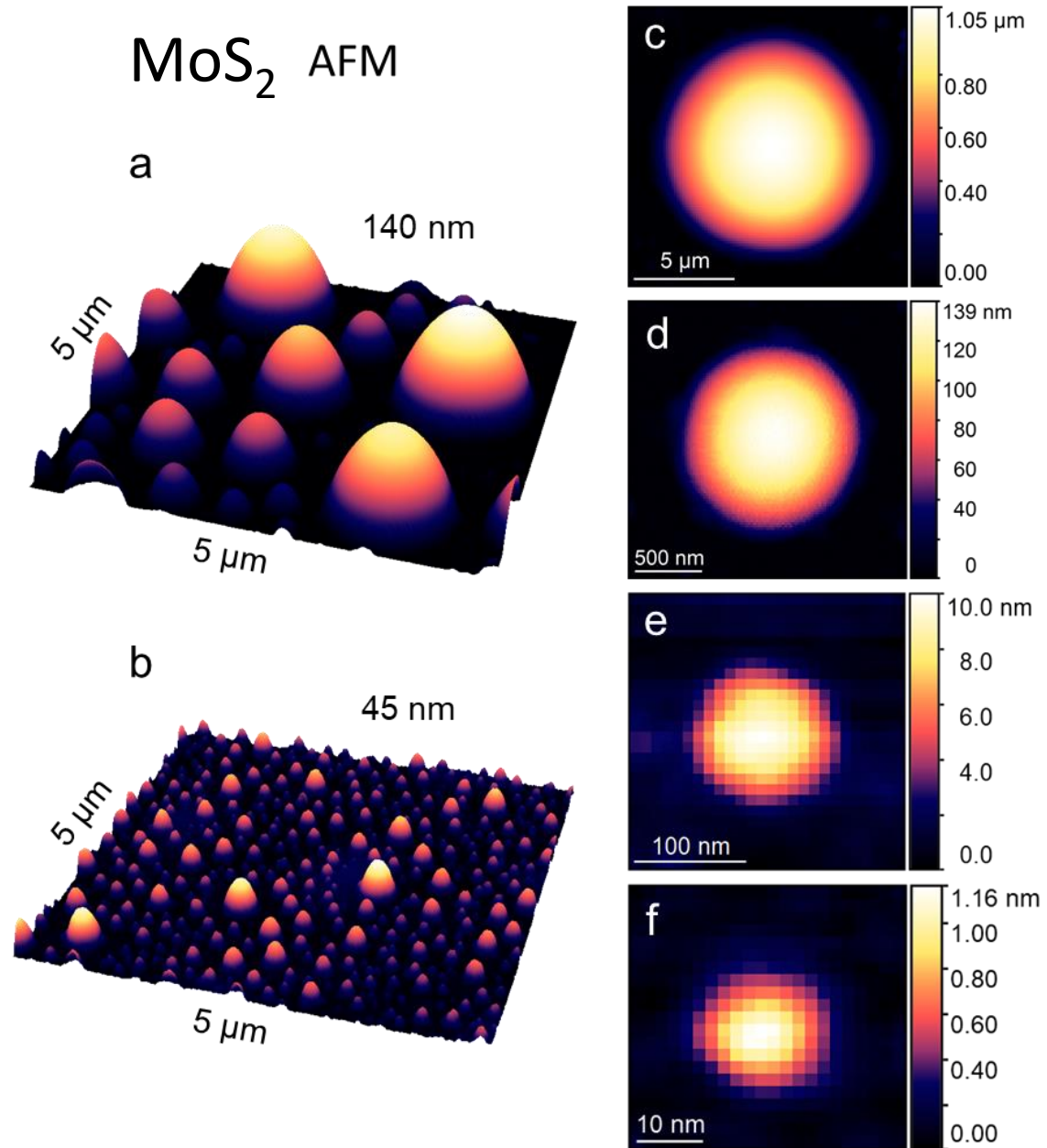


F MoTe_2

109 nm



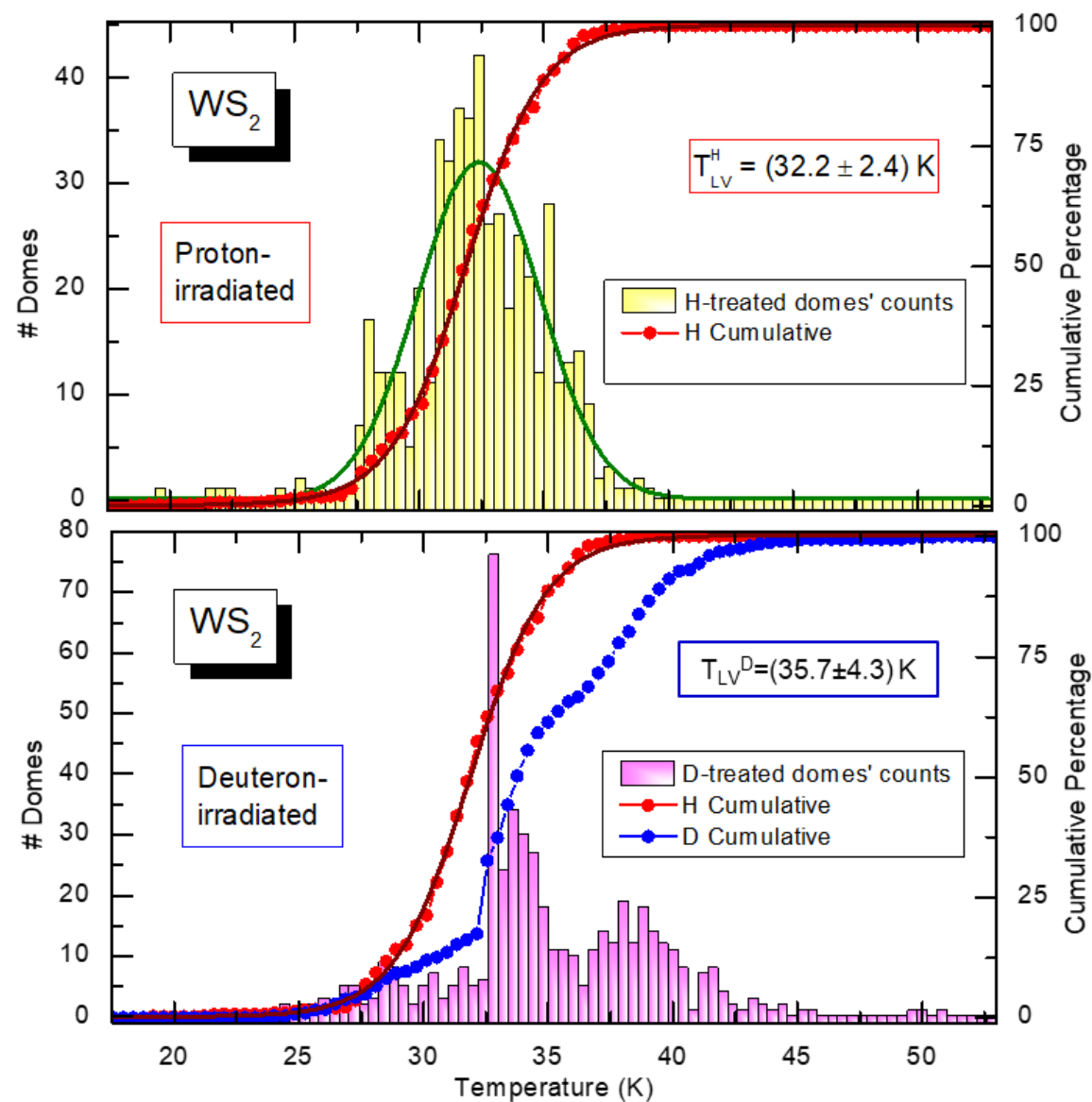
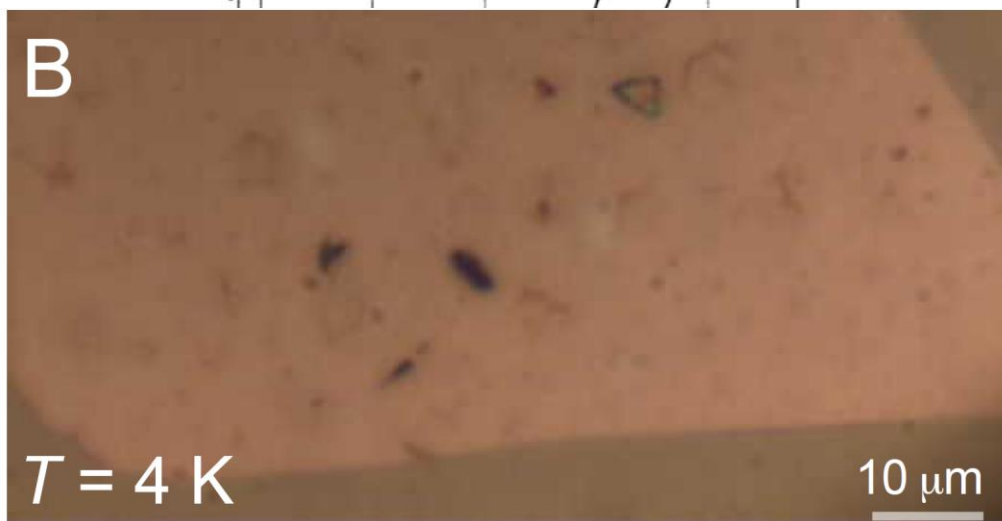
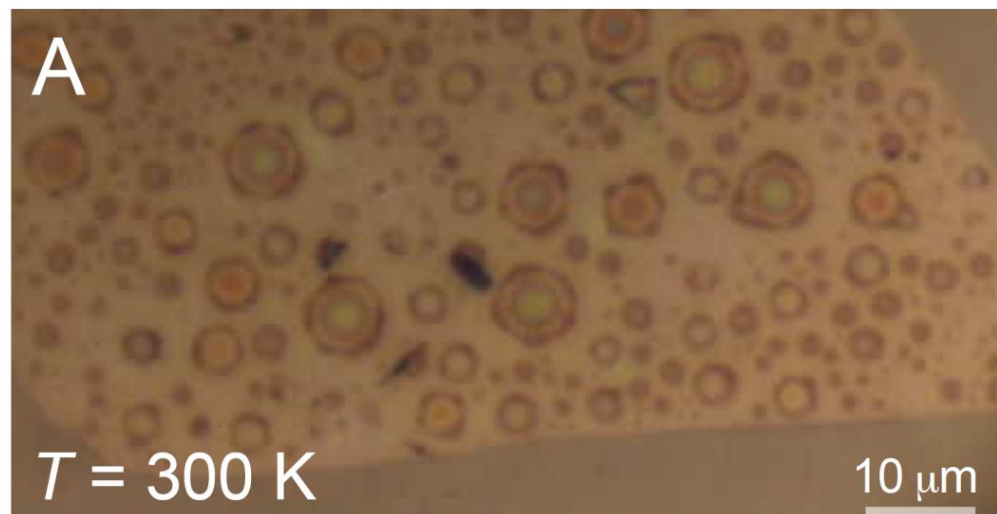
Domes in TMDs: Size



Three orders of magnitude variation in size is possible

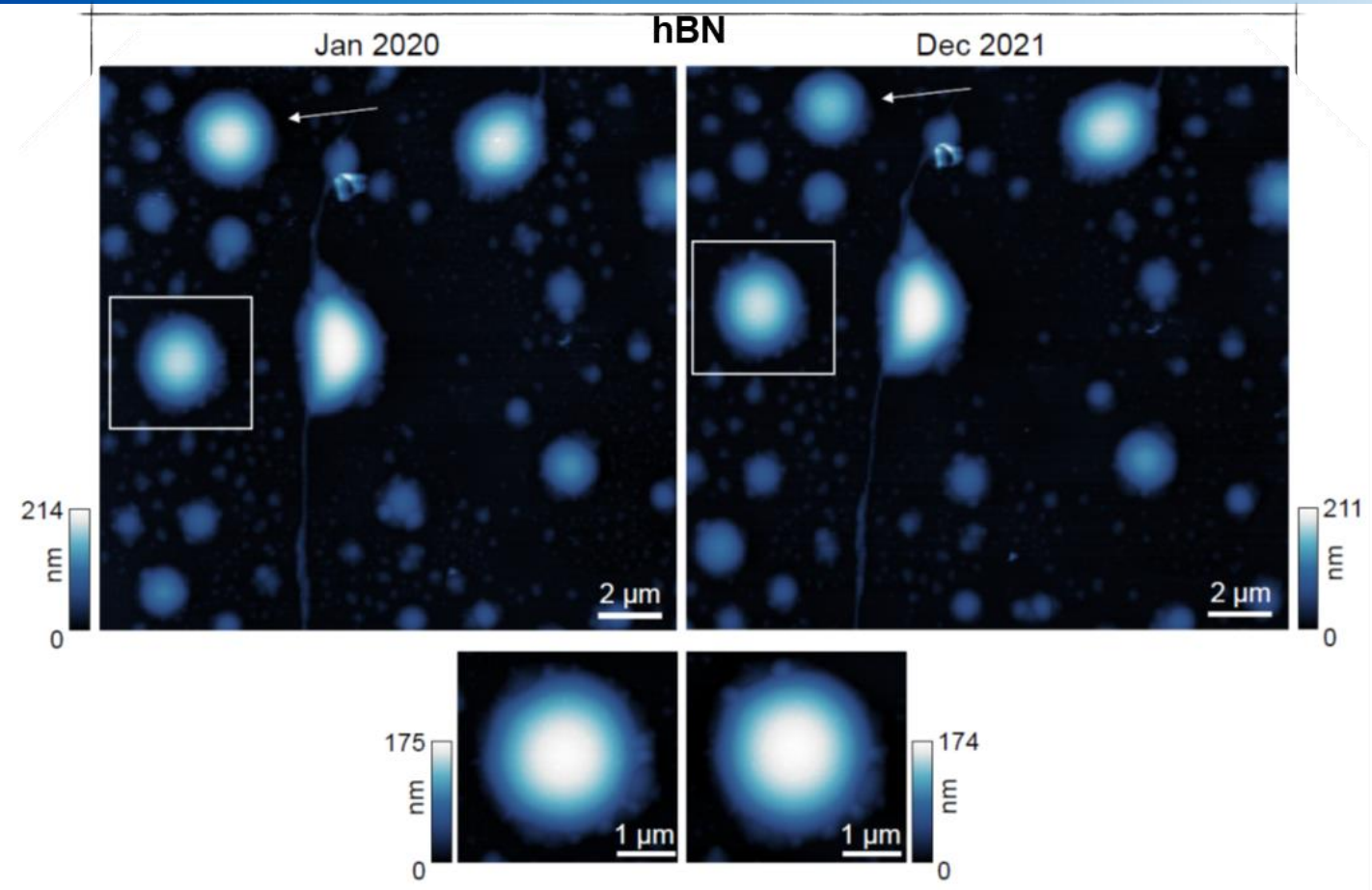
Evidence of H_2 molecules within the domes

$$d_H = 8 \times 10^{16} \text{ protons/cm}^2$$



Domes in TMDs: Main features

Durability > 3 yrs

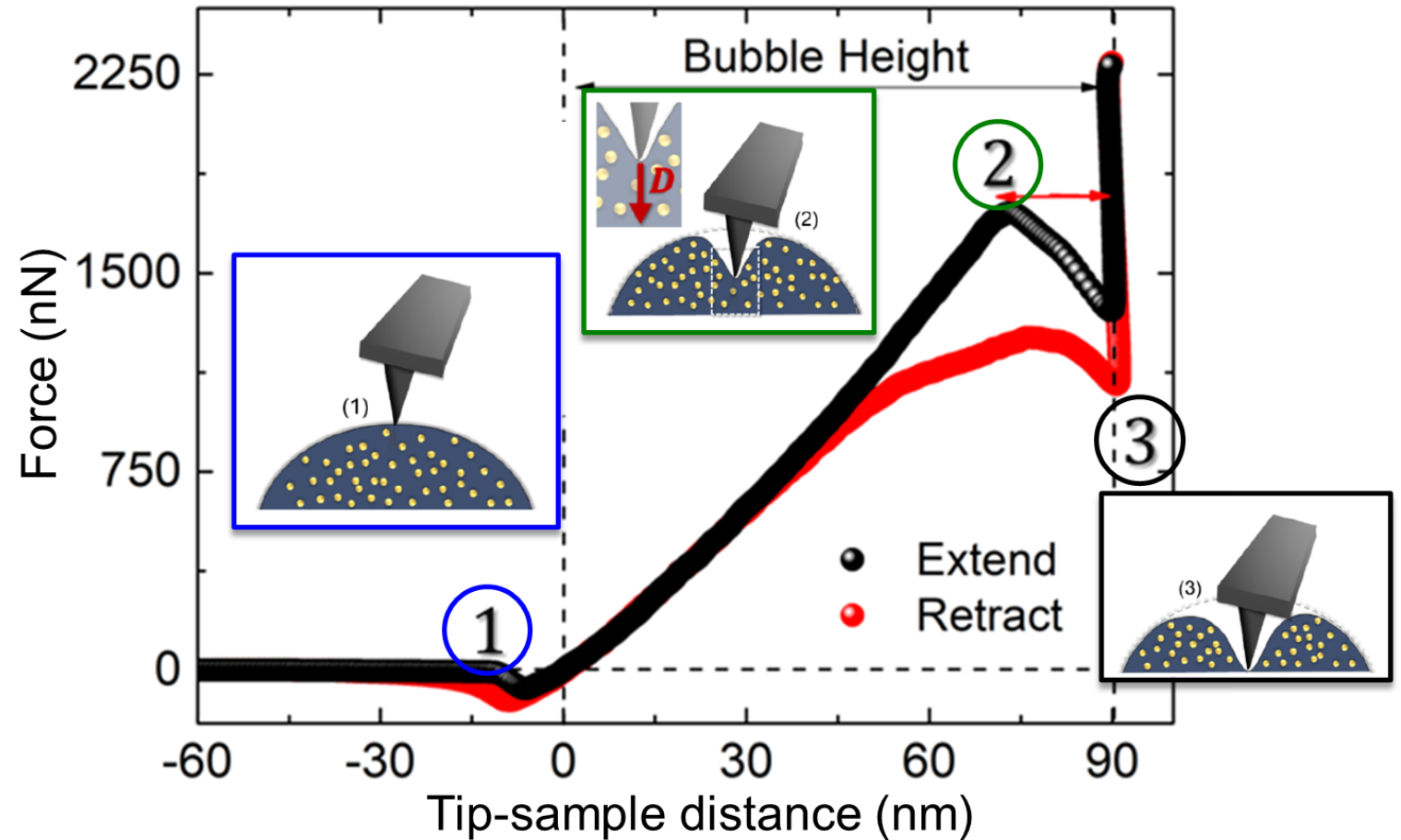
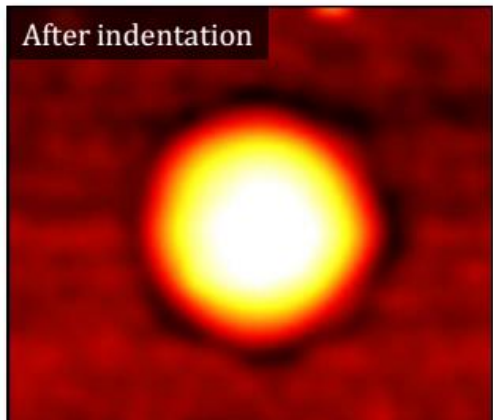
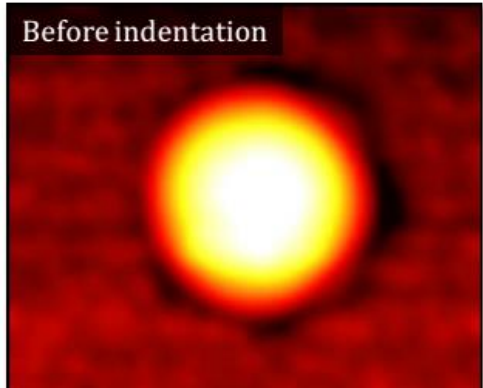


E. Blundo et al., Nano Lett. **22**, 1525 (2022)

Domes in TMDs: Main features

Durability > 3 yrs

Robustness



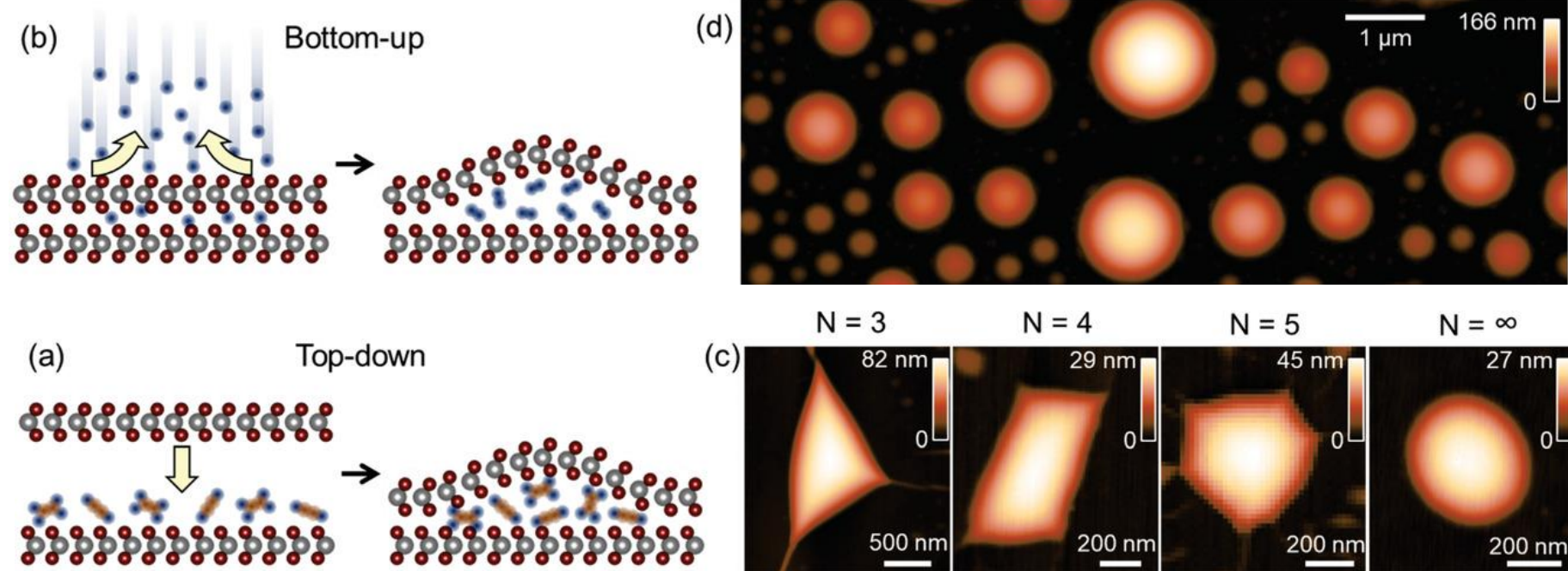
C. Di Giorgio et al., ACS Appl. Mater. Interfaces **13**, 48228 (2021)

Domes in TMDs: Main features

Durability > 3 yrs

Robustness

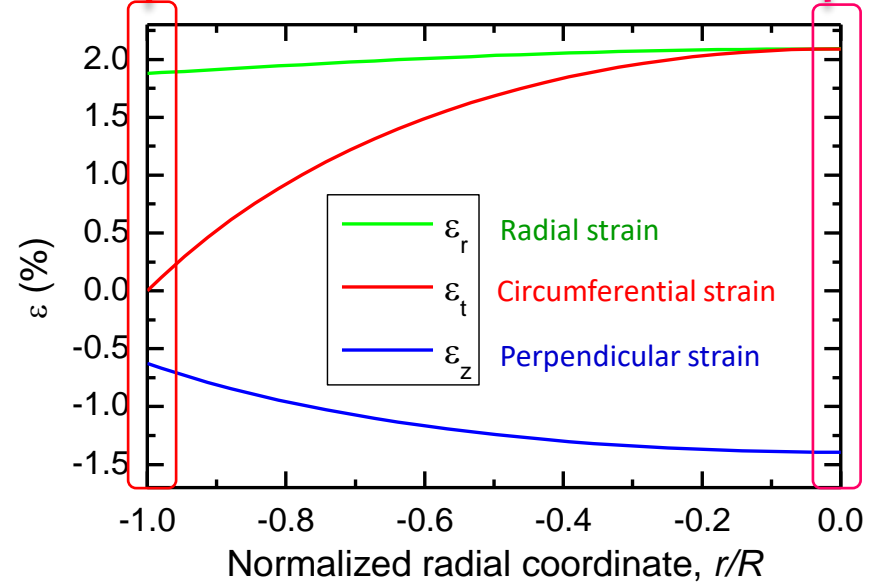
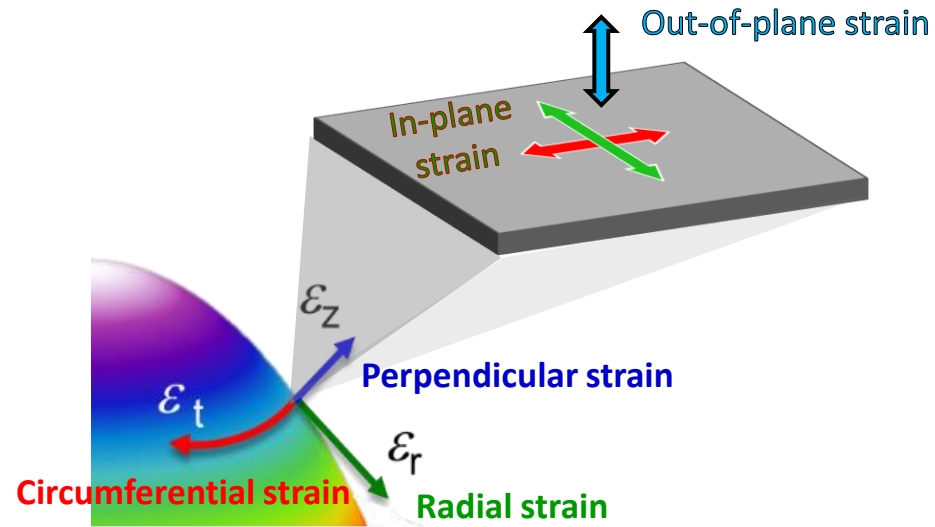
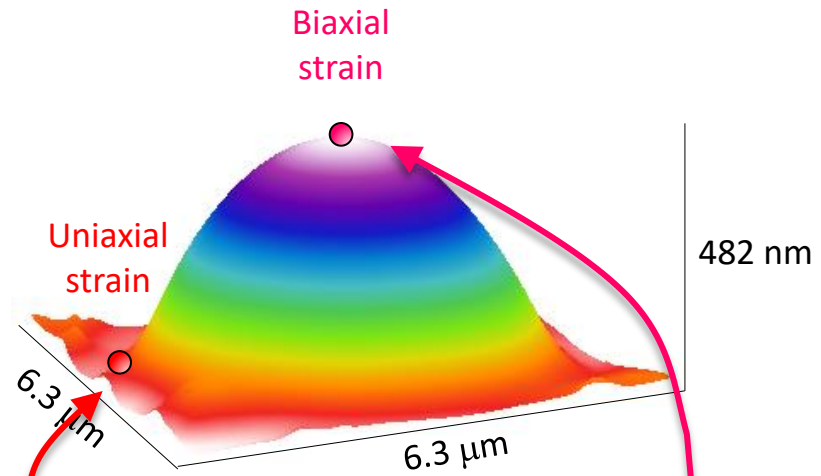
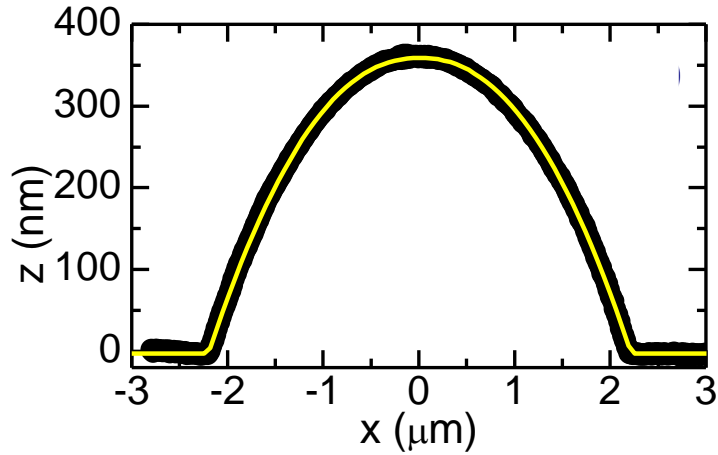
Regular/
reproducible shape



Strain field

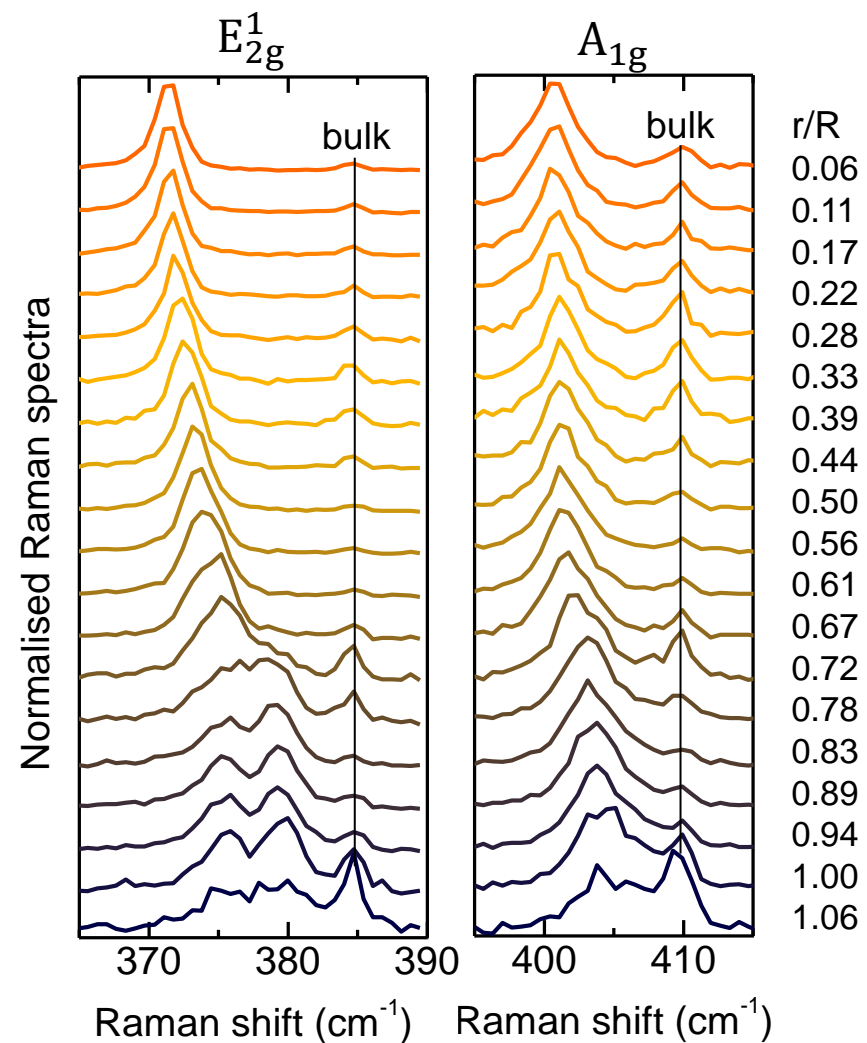
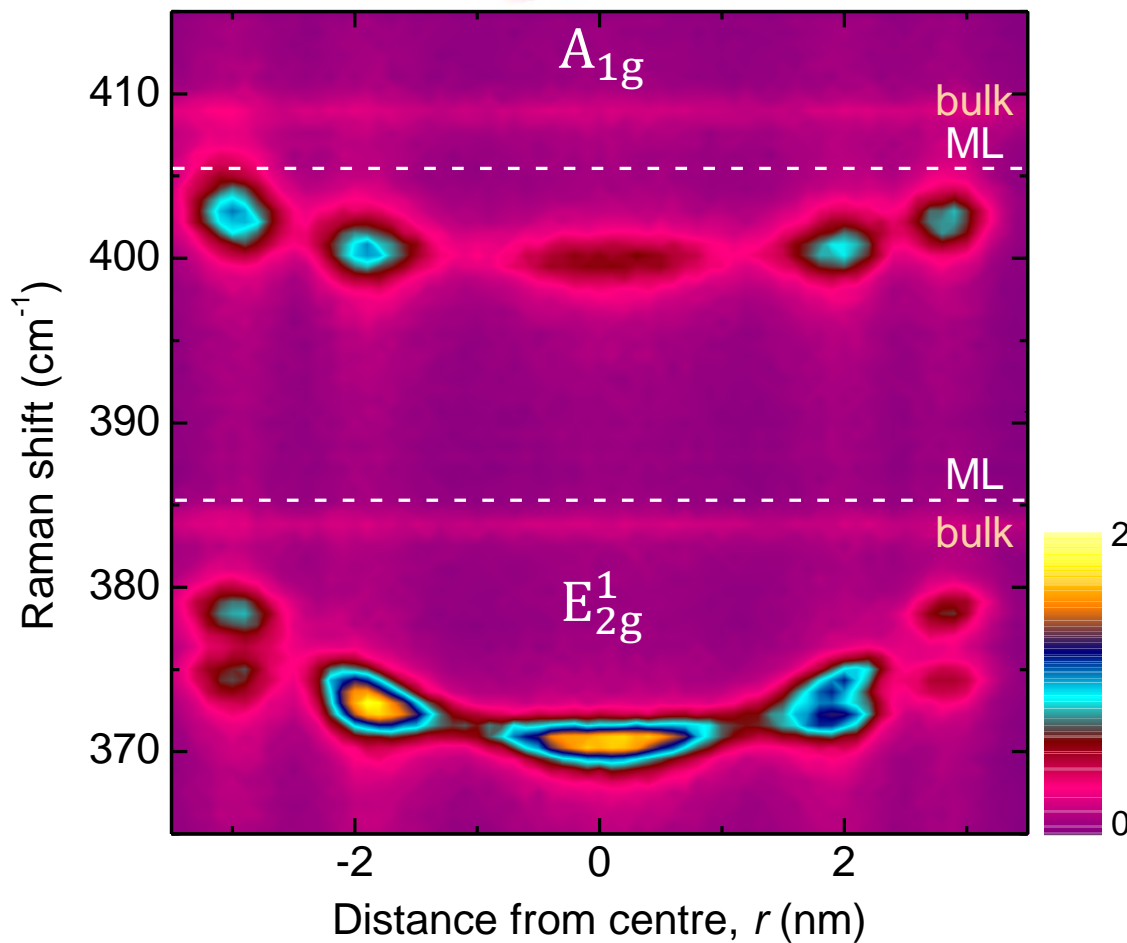
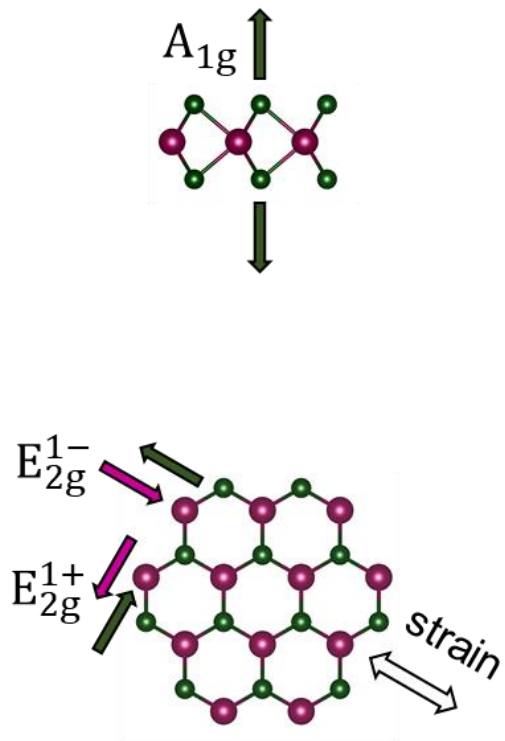
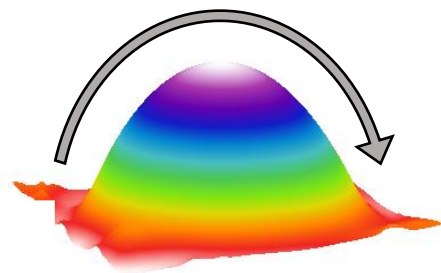
FEM (Finite-Element Method) calculations, within the framework of nonlinear membrane theory

E. Blundo et al, Phys. Rev. Lett. **127**, 46101 (2021)



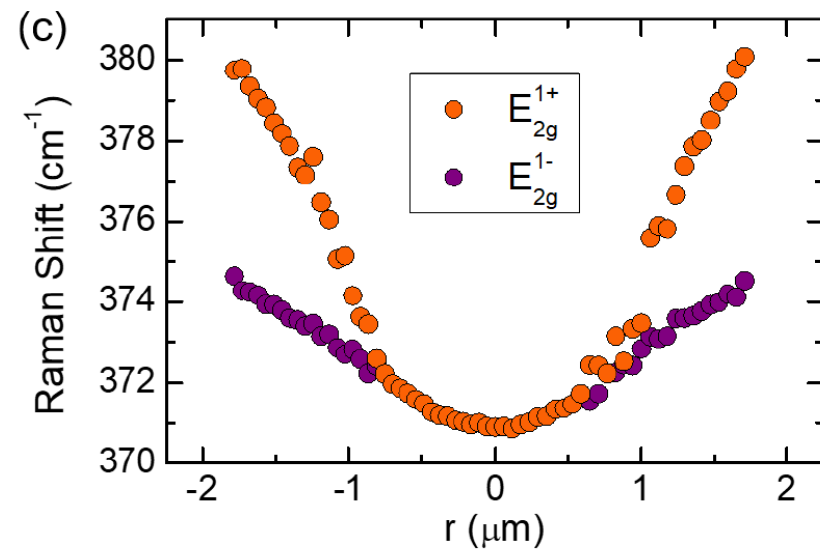
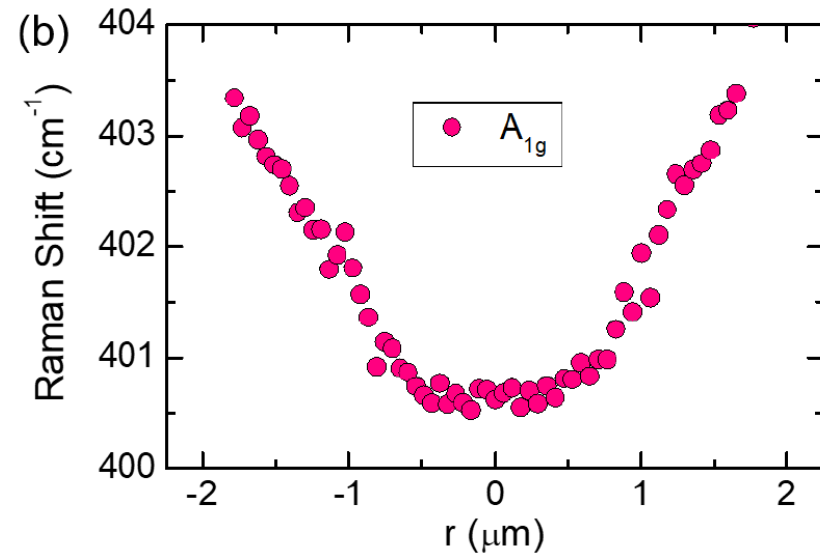
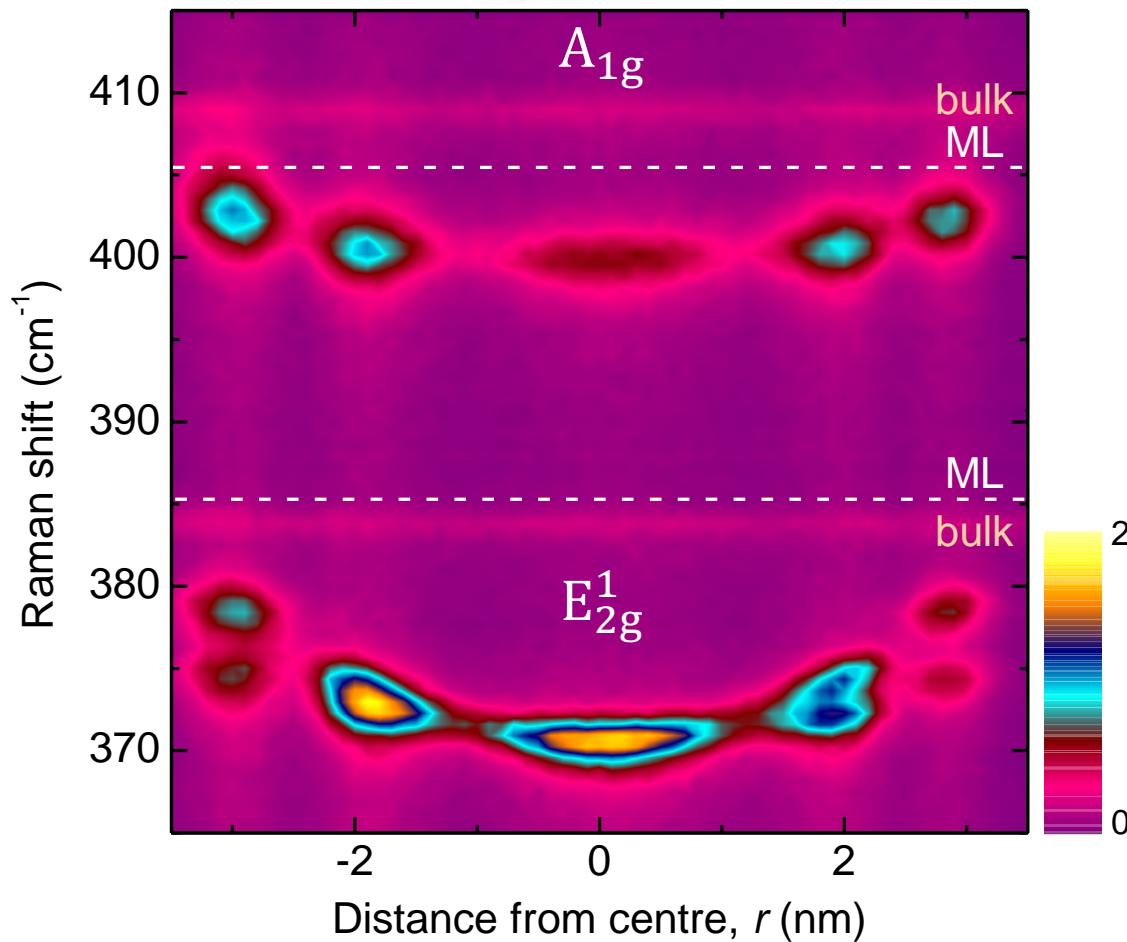
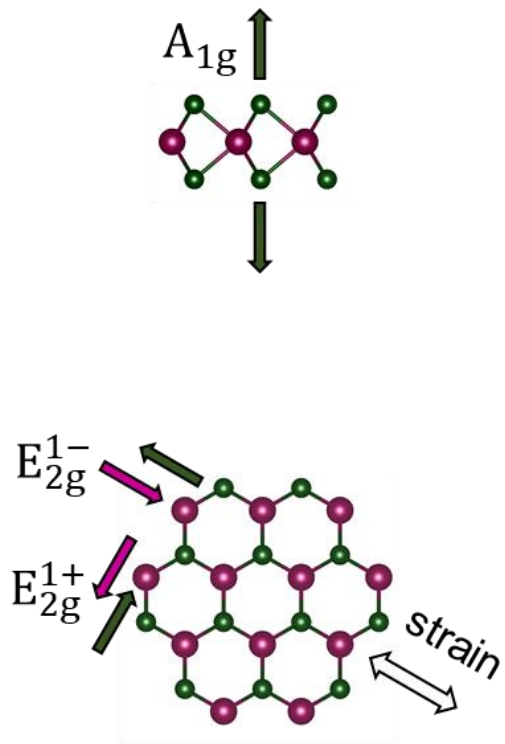
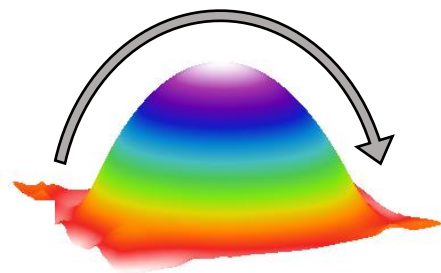
Micro-Raman measurements – MoS₂

E. Blundo et al, Phys. Rev. Lett. **127**, 46101 (2021)

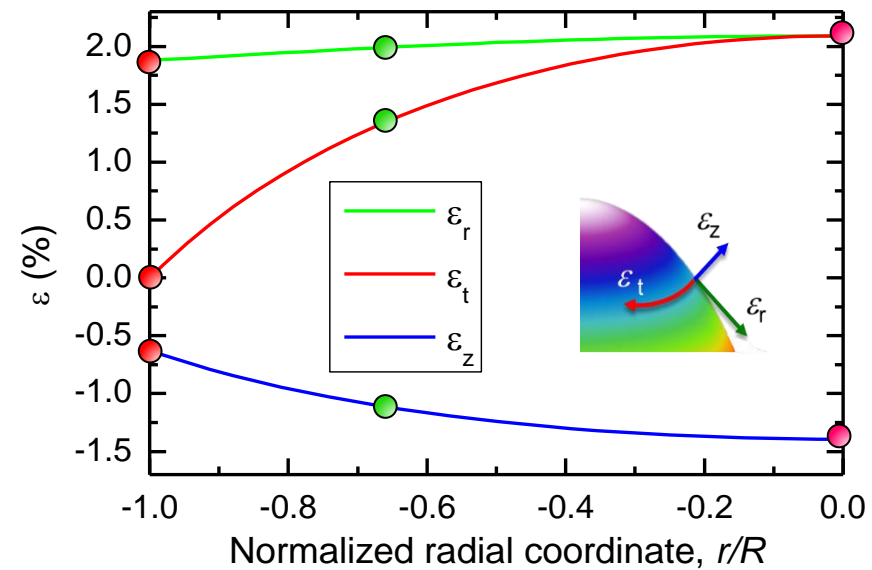
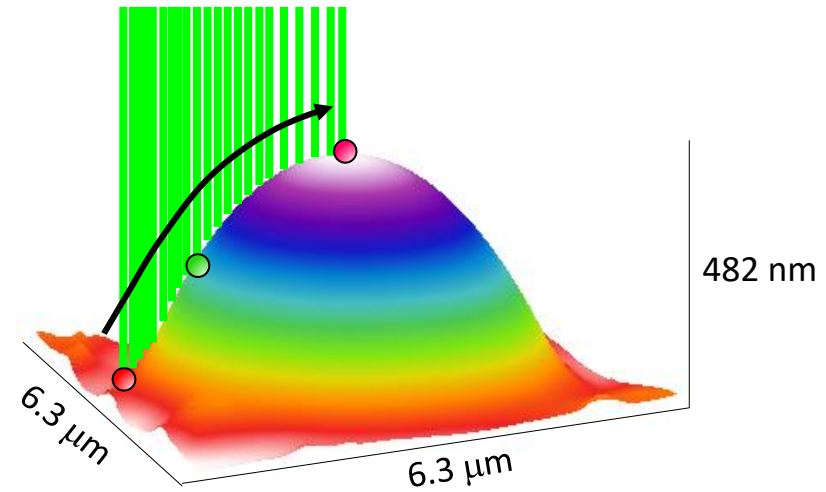
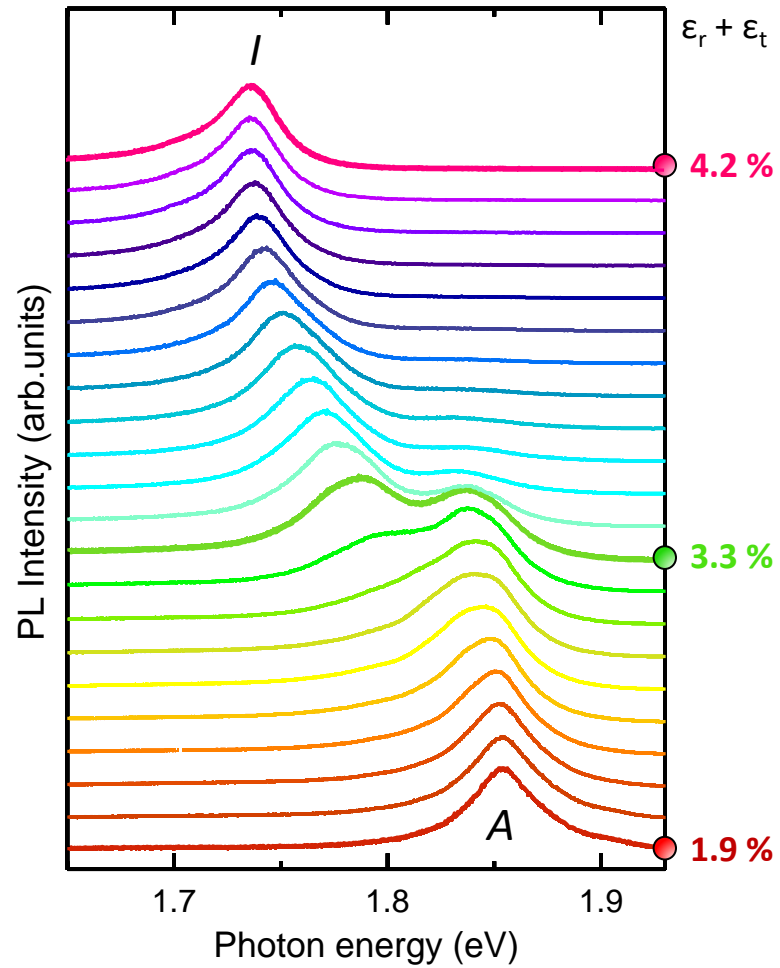


Micro-Raman measurements – MoS₂

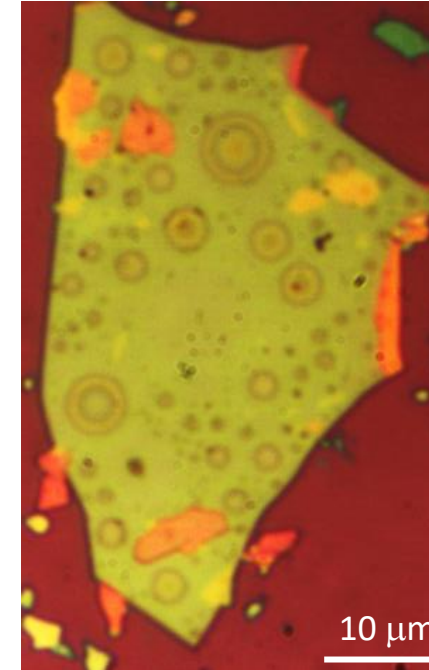
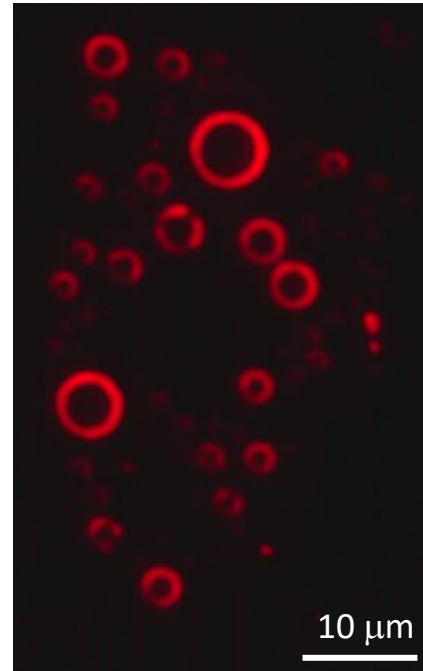
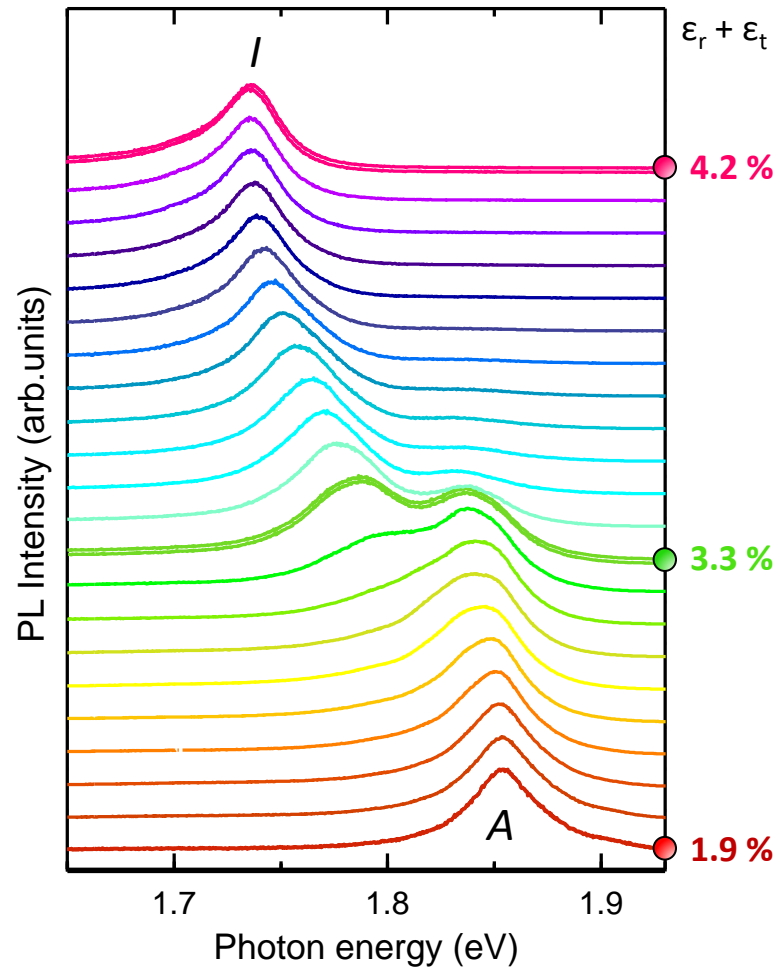
E. Blundo et al, Phys. Rev. Lett. **127**, 46101 (2021)



Micro-PL mapping of WS_2 domes

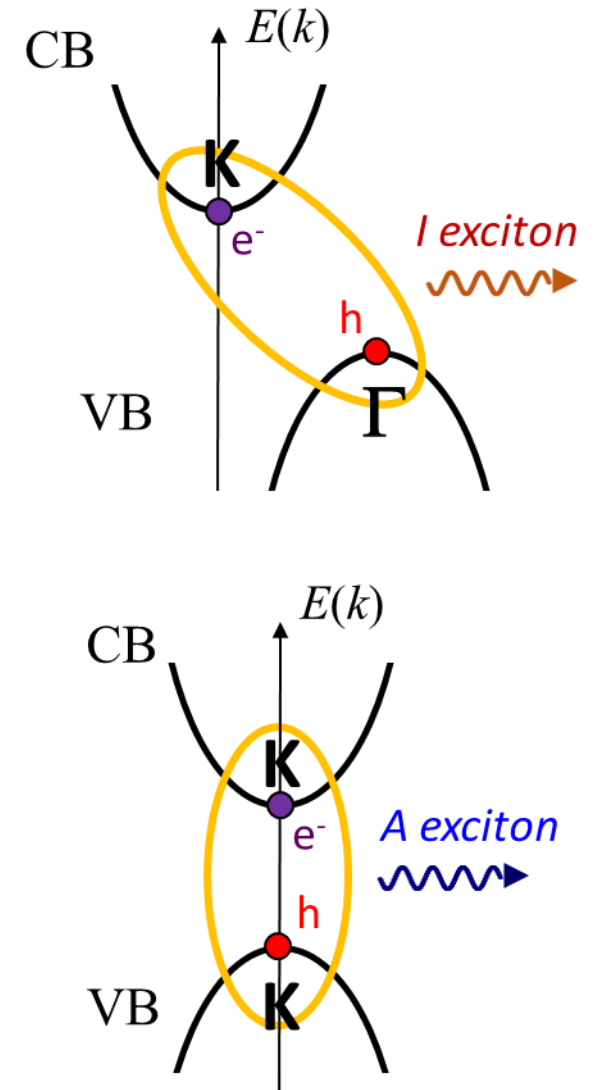
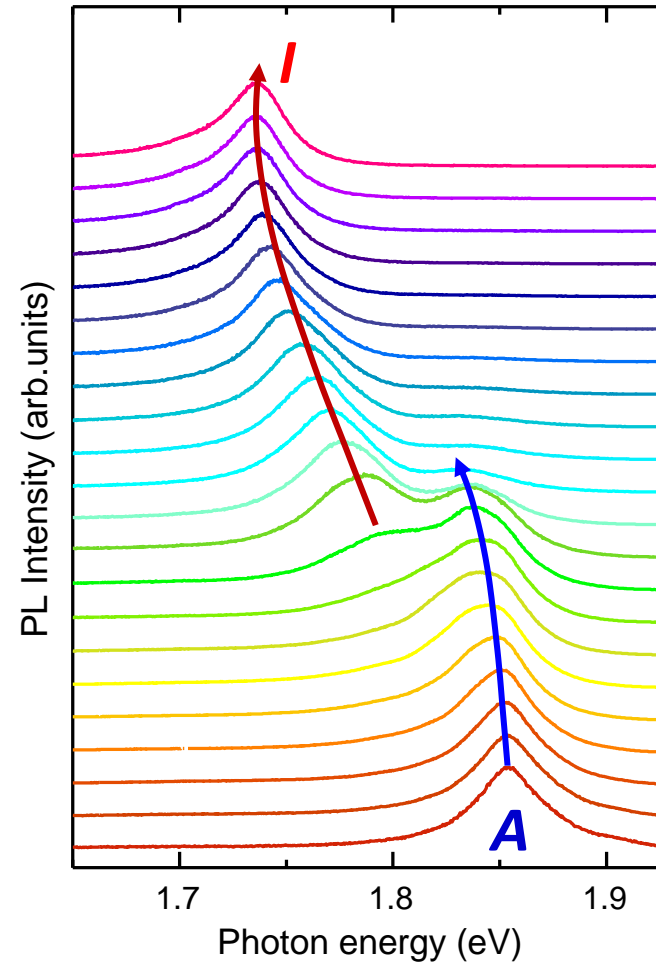
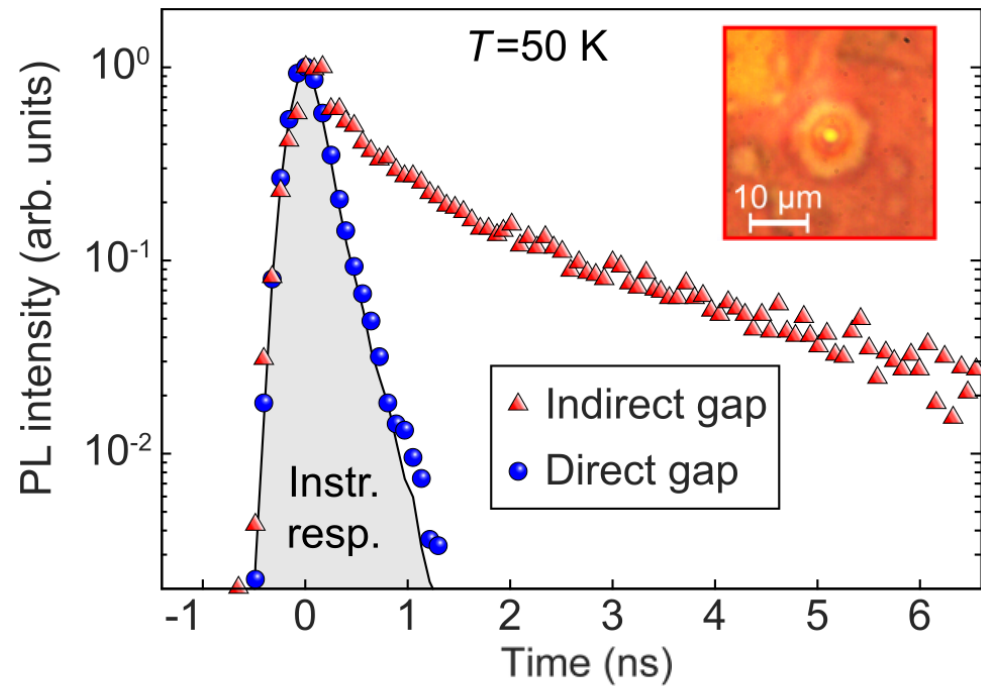


Micro-PL mapping of WS_2 domes



Creation of highly efficient anular regions

Micro-PL mapping of WS_2 domes



Magneto-PL measurements of WS_2 domes

Proposal for magnet time at the
High Field Magnet Laboratory
(up to 30 T). PI: A. Polimeni



Proposal for magnet time (**ISABEL**) at
the **Regional Partner 'University of
Warsaw'** (up to 12 T). PI: E. Blundo

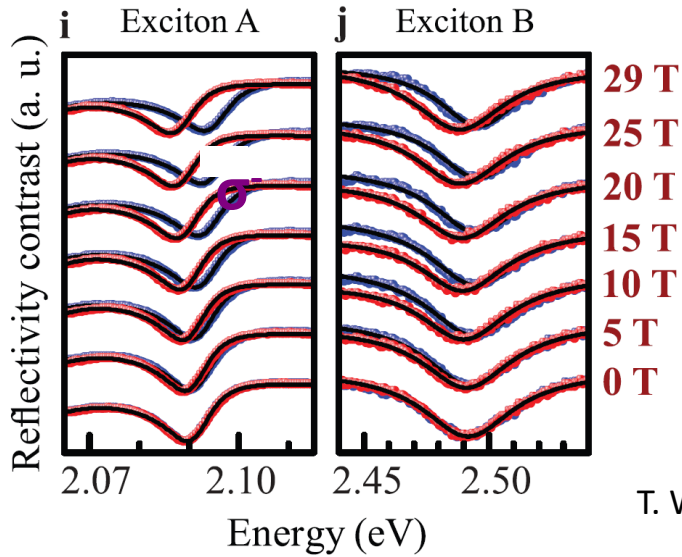


Magneto-PL measurements of WS_2 monolayers

WS_2 monolayer

M. Koperski et al., 2D Mater. **6**, 015001 (2019)

E. Blundo et al., Phys. Rev. Lett. **129**, 067402 (2022)

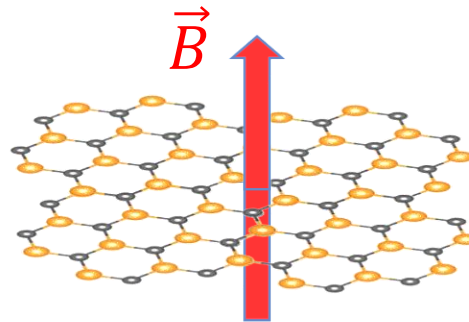
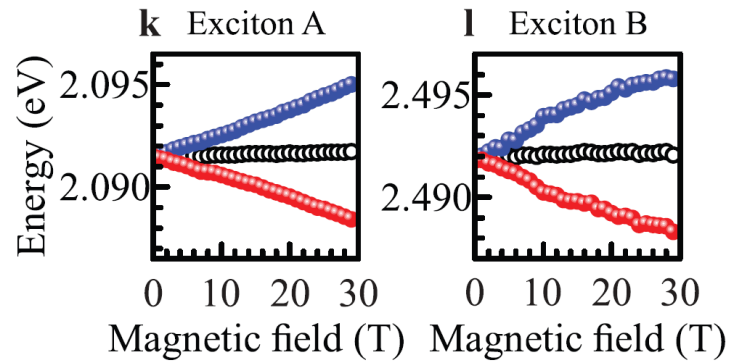
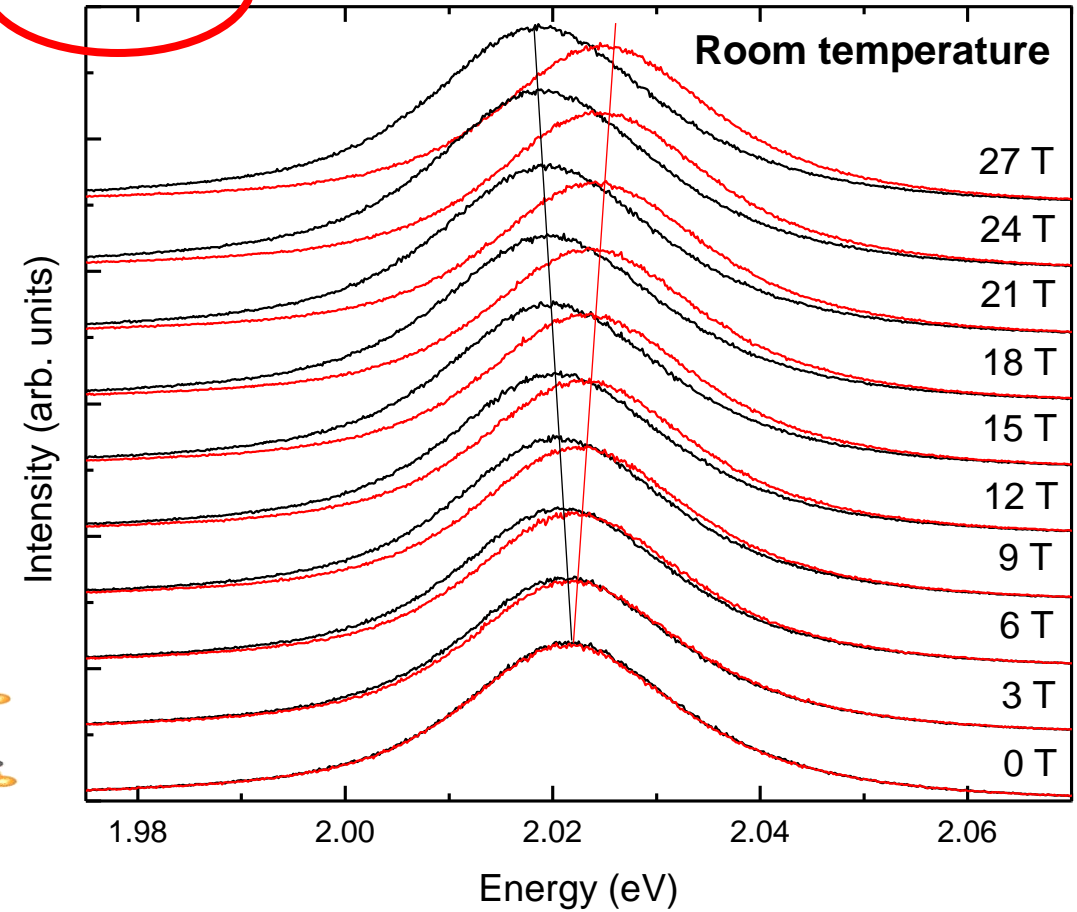


$$\Delta E_Z^A = g_A \mu_B B$$

$$g_A = 2[L_Z(CB_+) - L_Z(VB_+)]$$

T. Woźniak et al., Phys. Rev. B **101**, 235408 (2020)

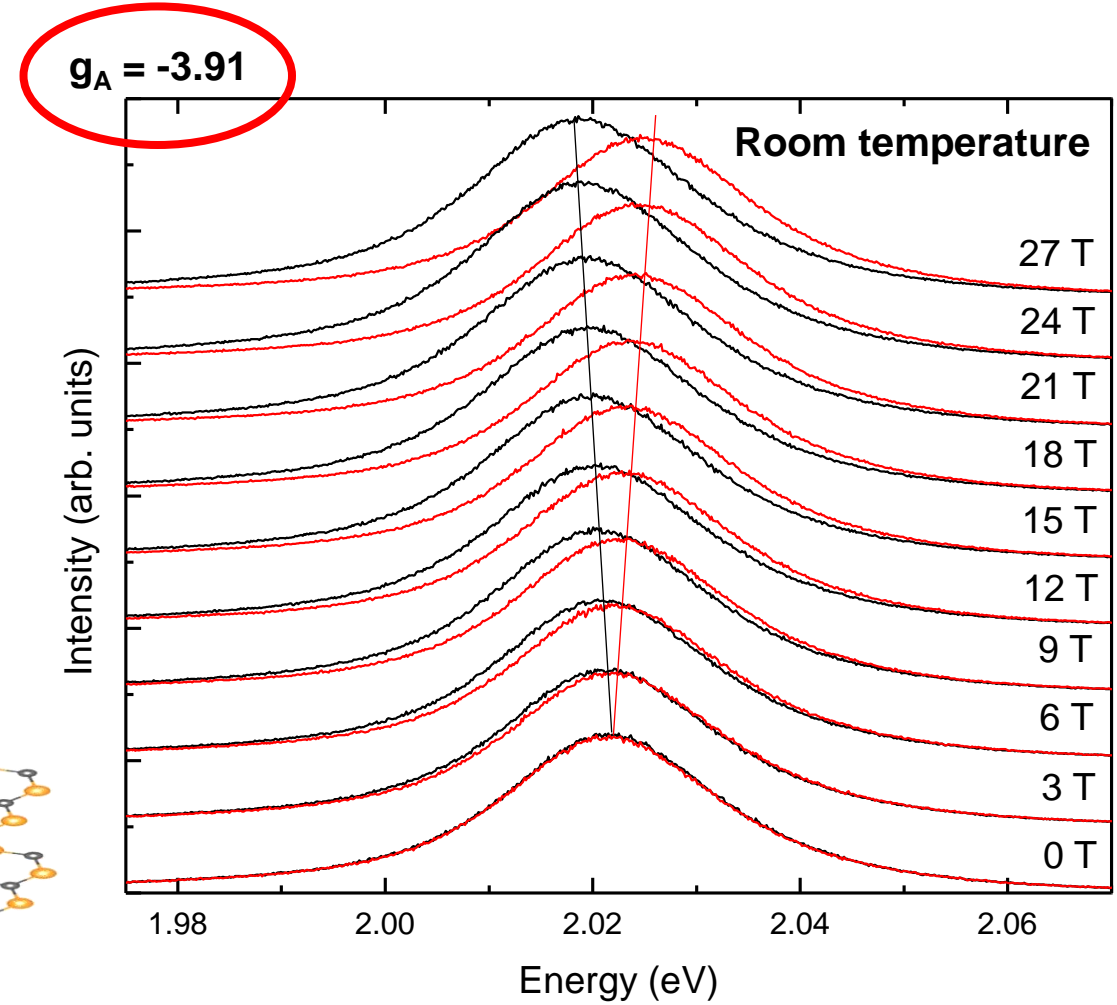
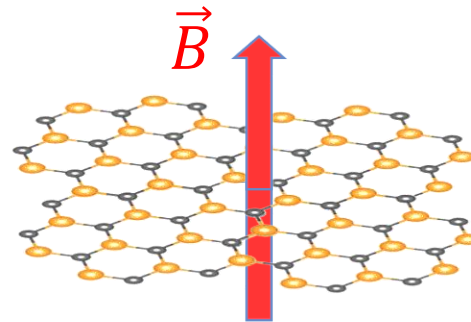
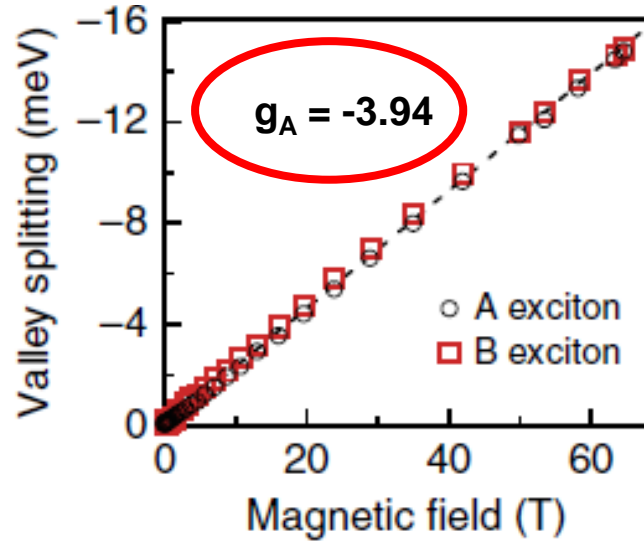
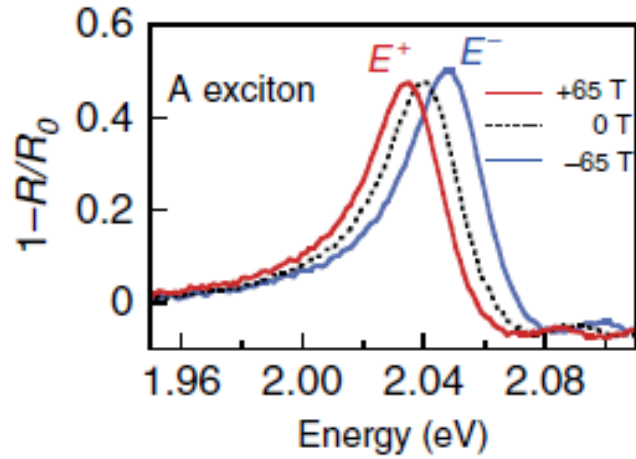
$$g_A = -3.91$$



Line	g-factor
Exciton A	-3.7 ± 0.2
Exciton B	-4.9 ± 1.2

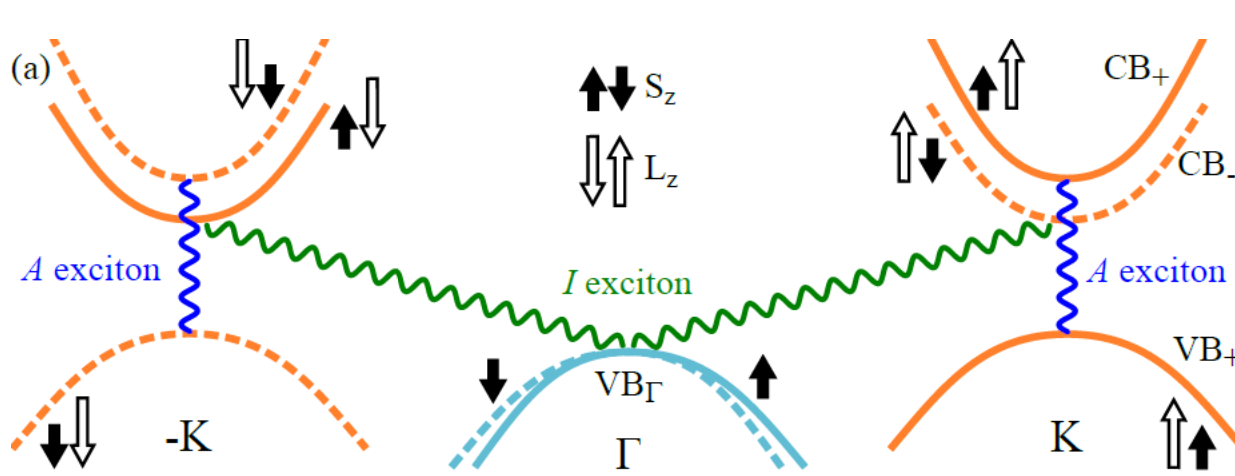
Magneto-PL measurements of WS_2 monolayers

E. Blundo et al., Phys. Rev. Lett. **129**, 067402 (2022)



Stier et al., Nat. Commun. **7**, 10643 (2016)

Magneto-PL measurements of WS_2 domes

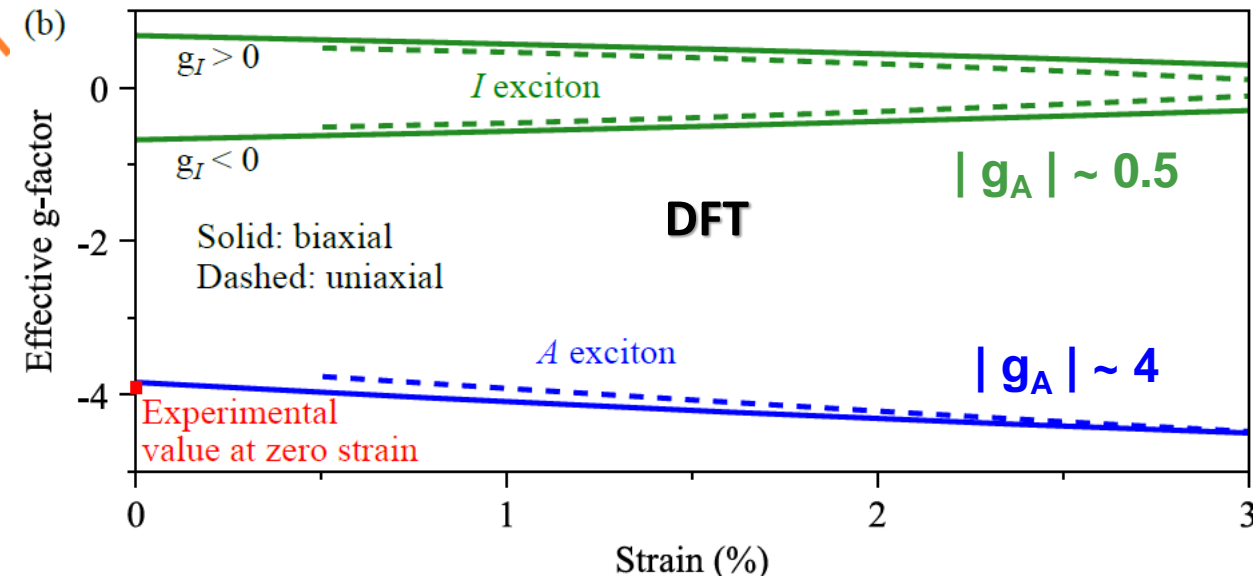
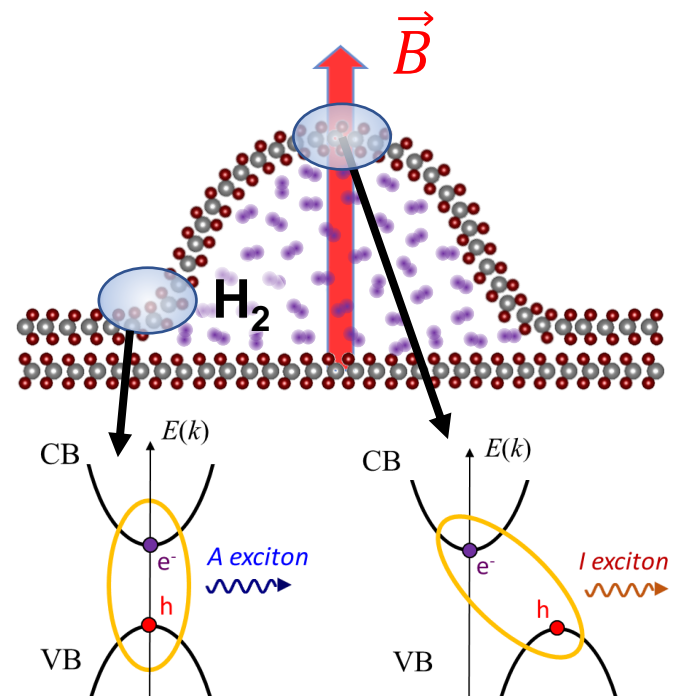


$$\Delta E_Z^{A(I)} = g_{A(I)} \mu_B B$$

$$g_A = 2[L_z(CB_+) - L_z(VB_+)]$$

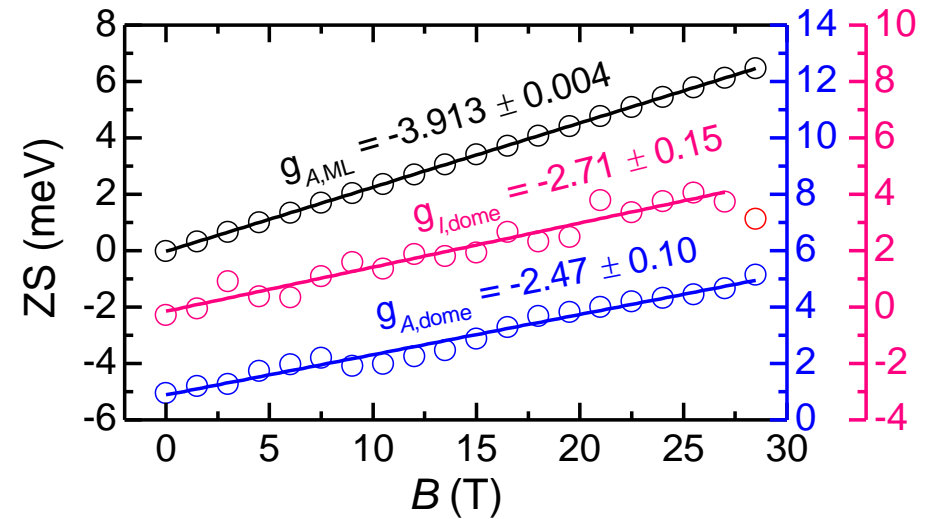
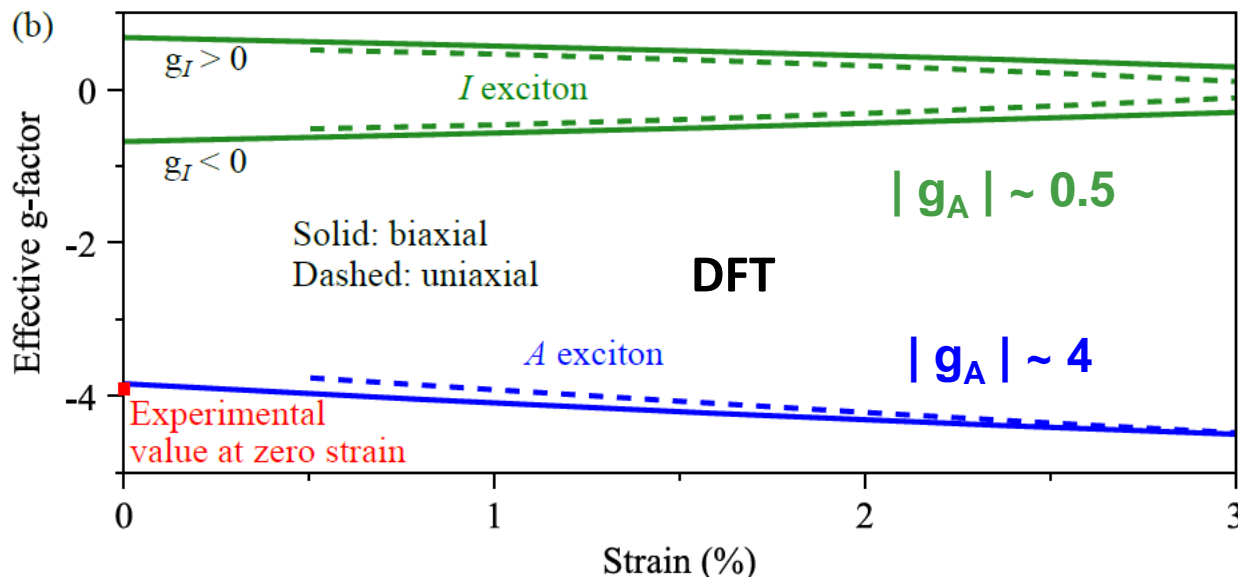
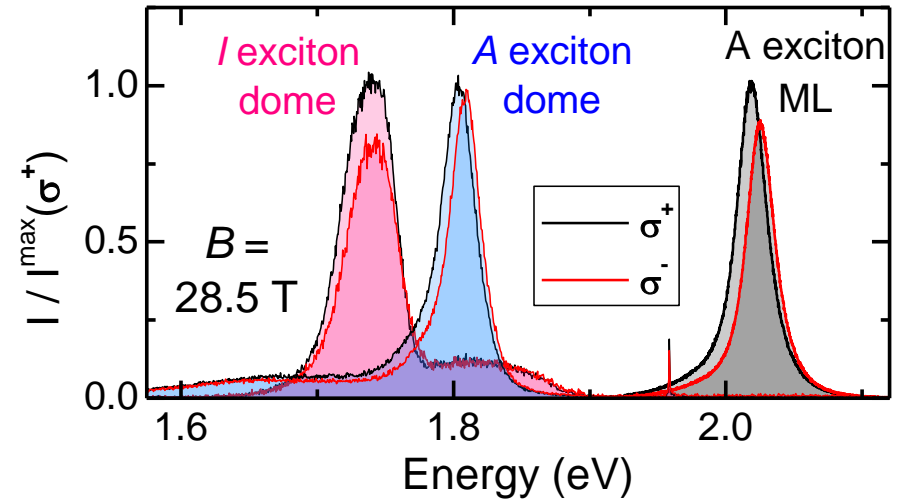
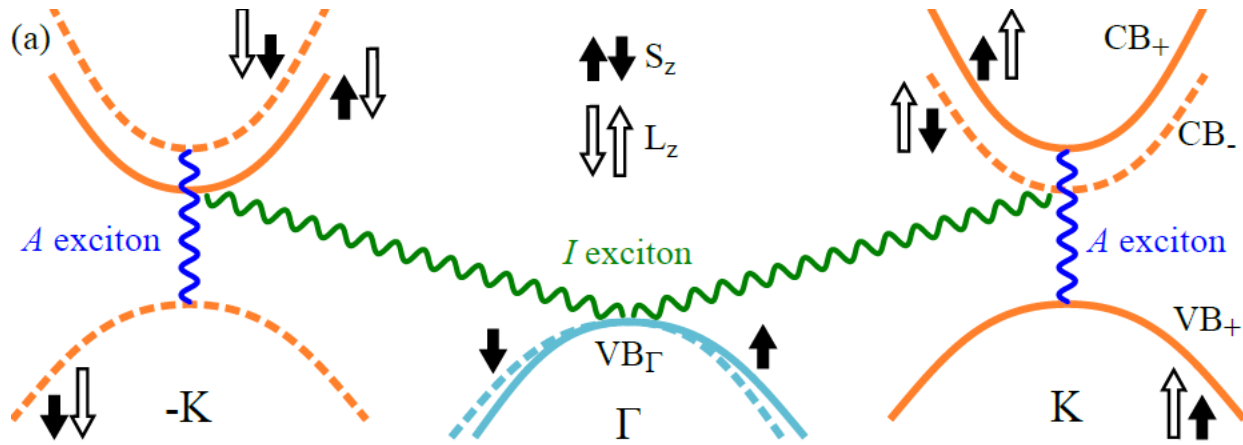
$$g_I = 2[L_z(CB_-) - 2]$$

How does $g_{A(I)}$ change with strain?



$g_{A(I)}$ should not change with strain
 A and I excitons should show very different splittings

Magneto-PL measurements of WS_2 domes

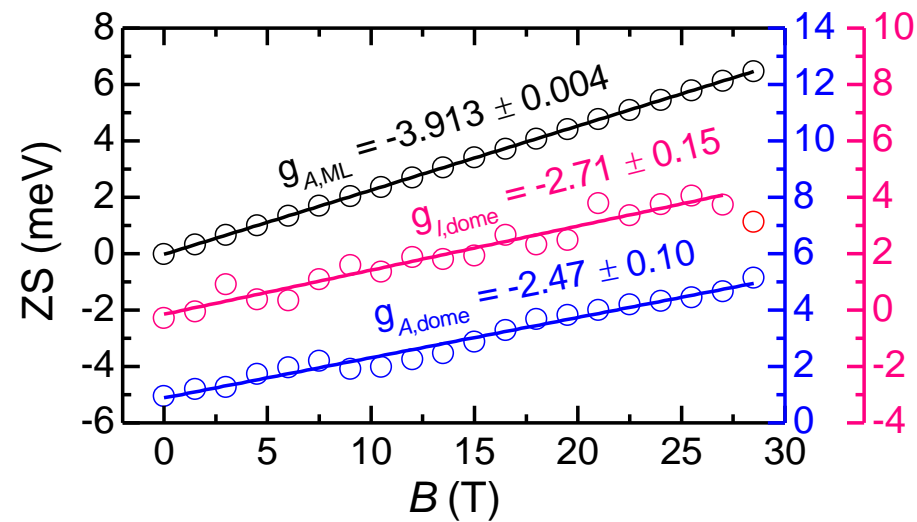
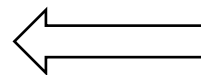
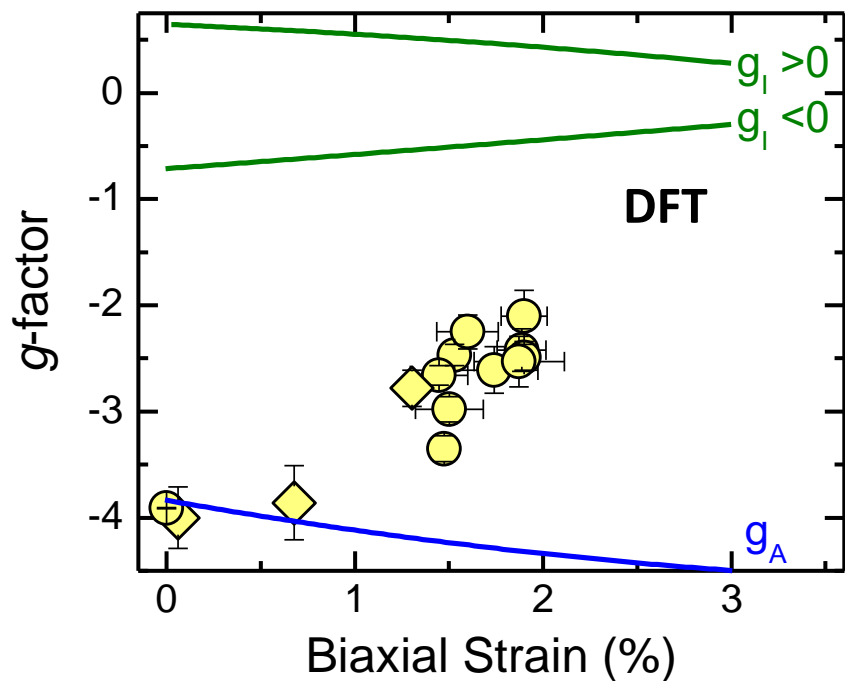
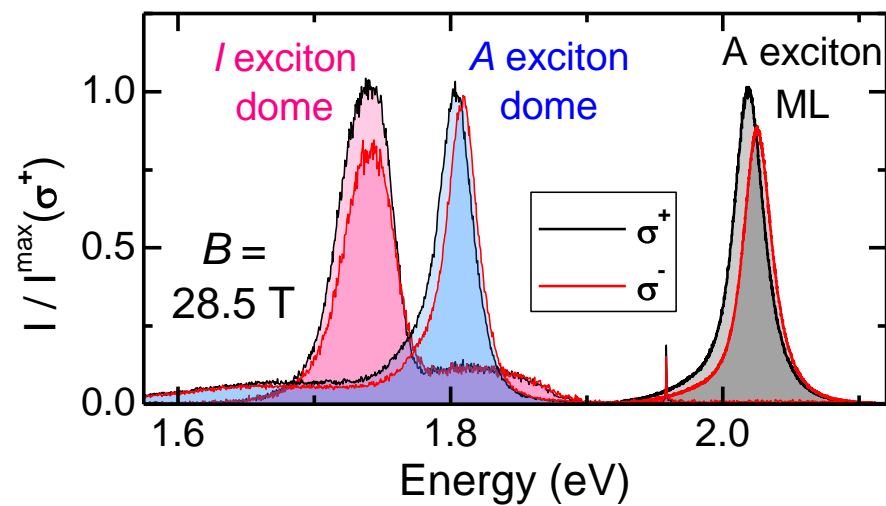
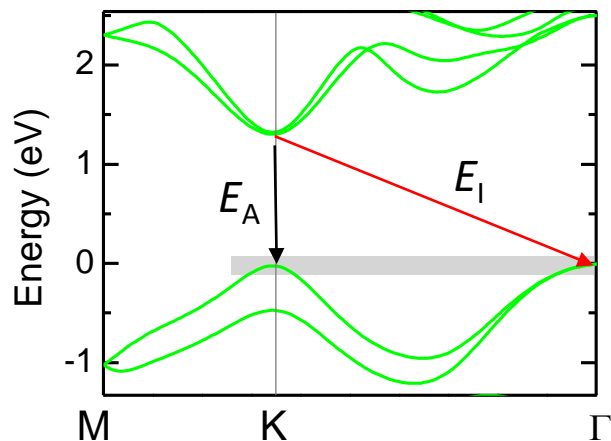


A and I excitons show very similar splittings

$g_{A(I)}$ should not change with strain

Theory-wise A and I excitons should show very different splittings

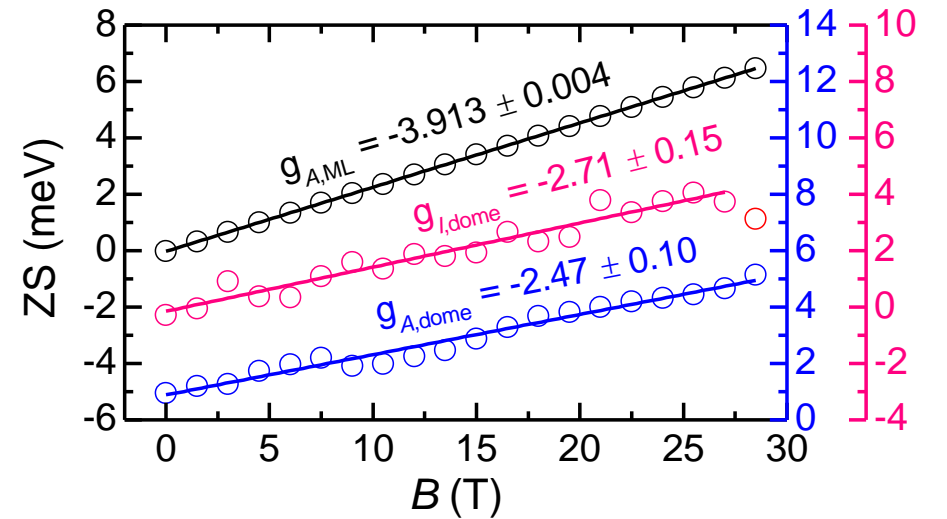
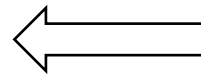
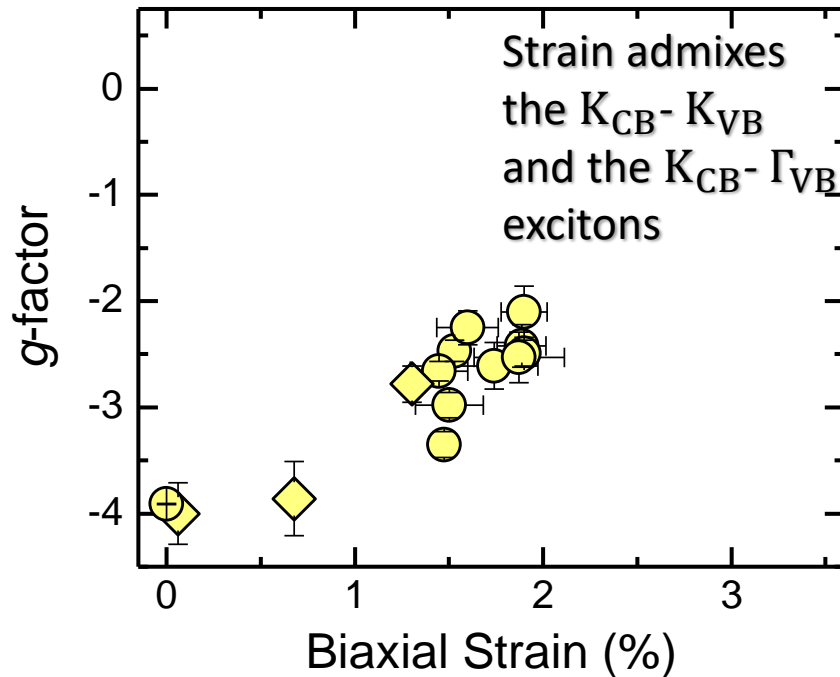
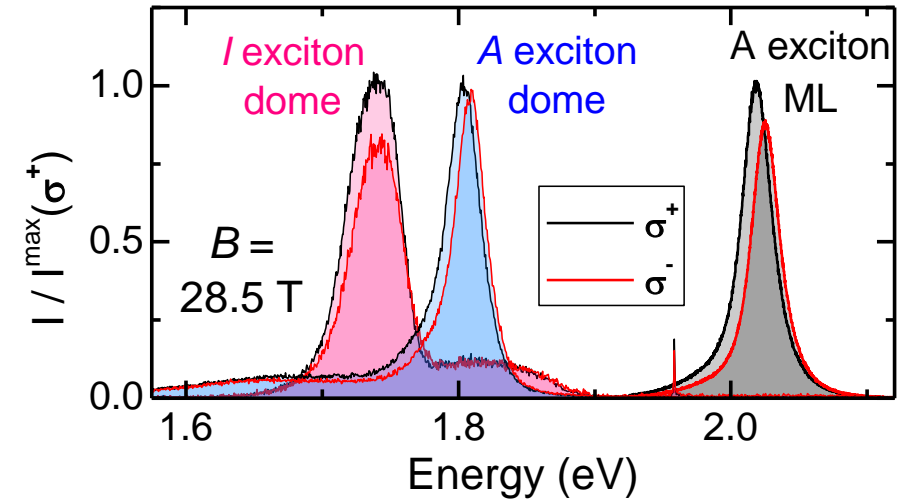
Magneto-PL measurements of WS_2 domes



Magneto-PL measurements of WS_2 domes

Strain-induced A and I exciton hybridisation

$$\begin{bmatrix} E_A + \frac{1}{2}g_A\mu_B B & 0 & \Delta_1 & \Delta_2 \\ 0 & E_A - \frac{1}{2}g_A\mu_B B & -\kappa\Delta_2^* & \kappa\Delta_1^* \\ \Delta_1^* & -\kappa\Delta_2 & E_I + \frac{1}{2}g_I\mu_B B & 0 \\ \Delta_2^* & \kappa\Delta_1 & 0 & E_I - \frac{1}{2}g_I\mu_B B \end{bmatrix}$$

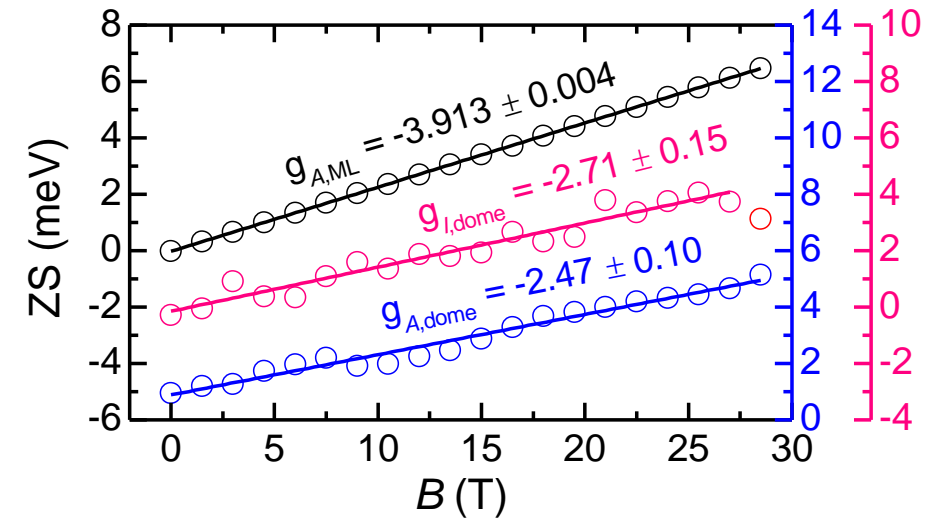
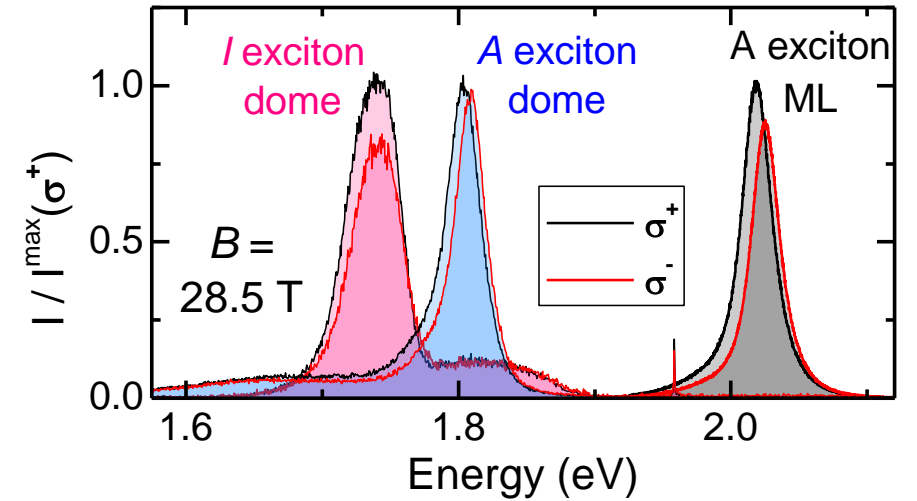
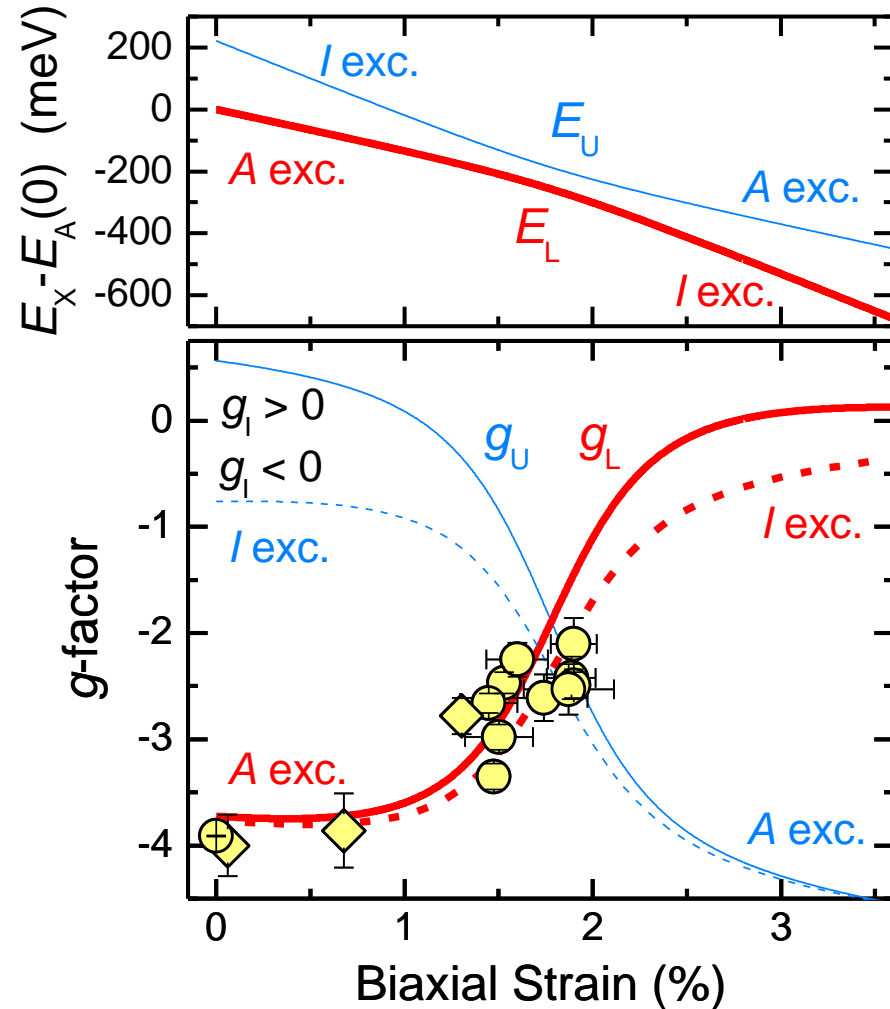


Magneto-PL measurements of WS_2 domes

$$\begin{bmatrix} E_A + \frac{1}{2}g_A\mu_B B & 0 & \Delta_1 & \Delta_2 \\ 0 & E_A - \frac{1}{2}g_A\mu_B B & -\kappa\Delta_2^* & \kappa\Delta_1^* \\ \Delta_1^* & -\kappa\Delta_2 & E_I + \frac{1}{2}g_I\mu_B B & 0 \\ \Delta_2^* & \kappa\Delta_1 & 0 & E_I - \frac{1}{2}g_I\mu_B B \end{bmatrix}$$

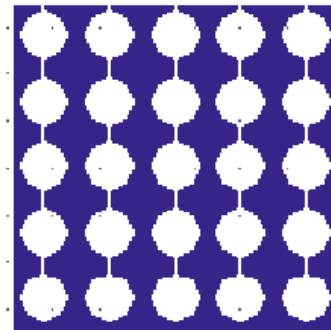
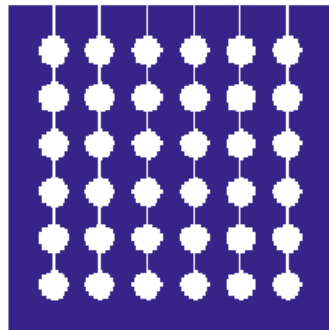
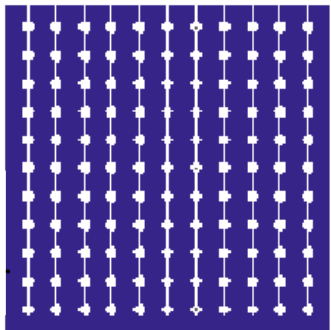
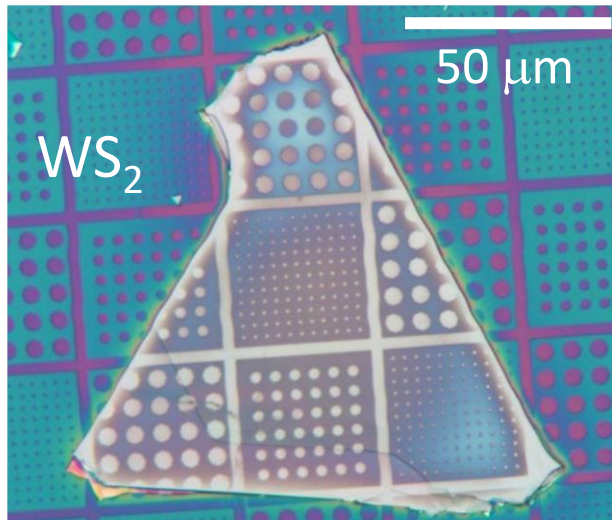
$$\Delta_1 = 35 \text{ meV}$$

$$\Delta_2 = 0$$



Zeeman-splitting measurements unveil exciton hybridization
 Phonons? Disorder?

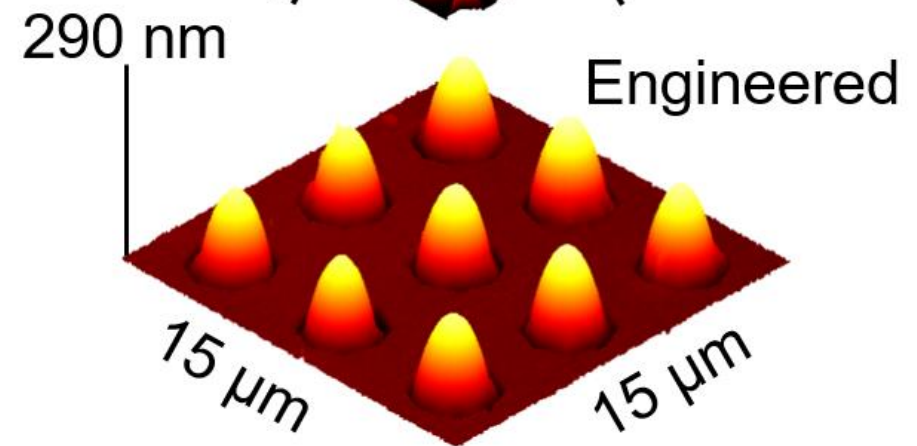
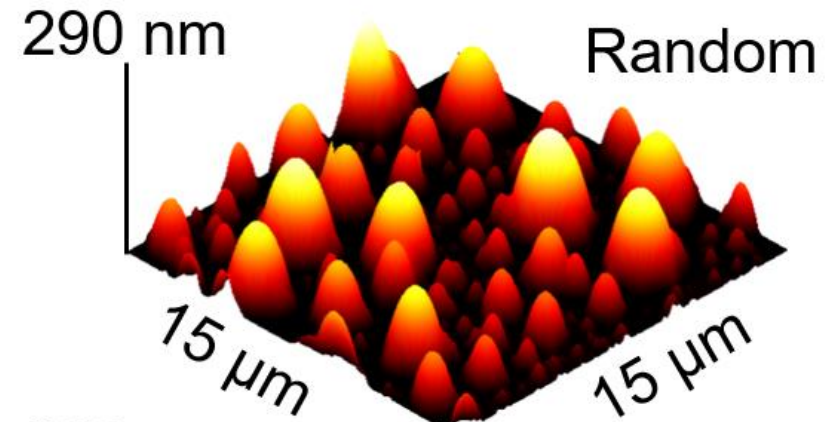
Controlled dome formation



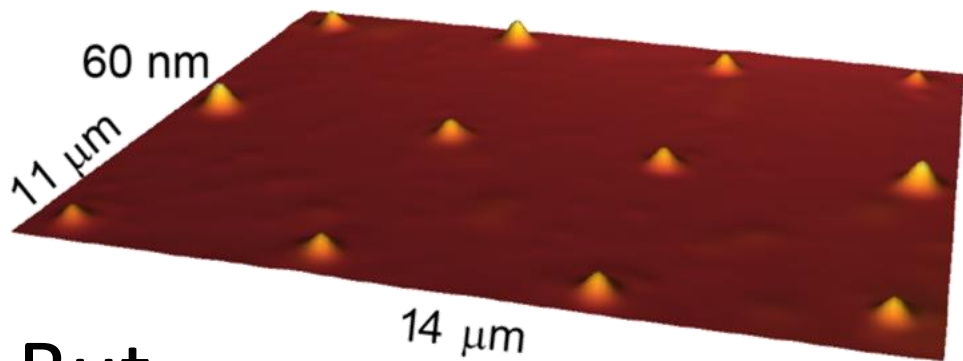
$S = 1 \mu\text{m}$

$3 \mu\text{m}$

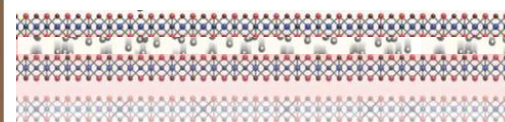
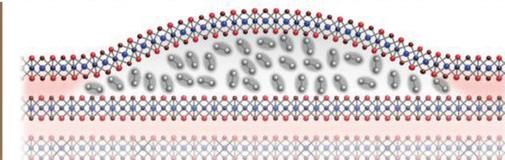
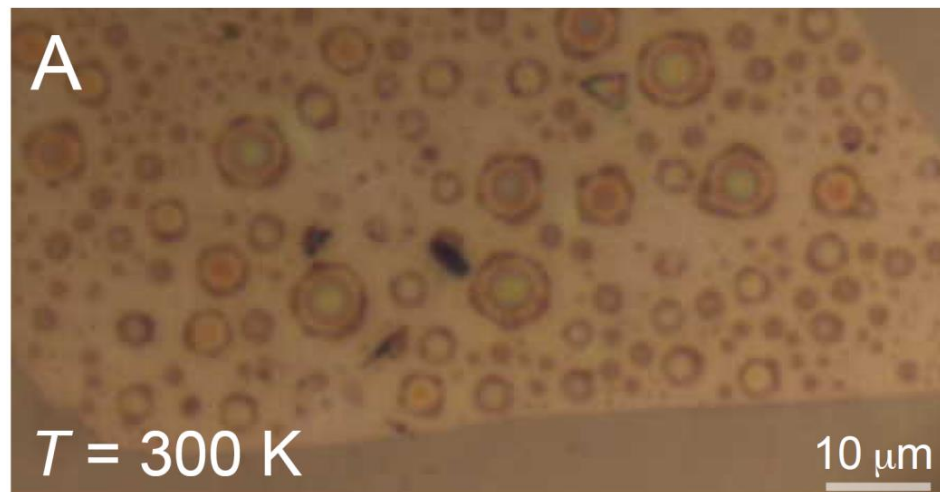
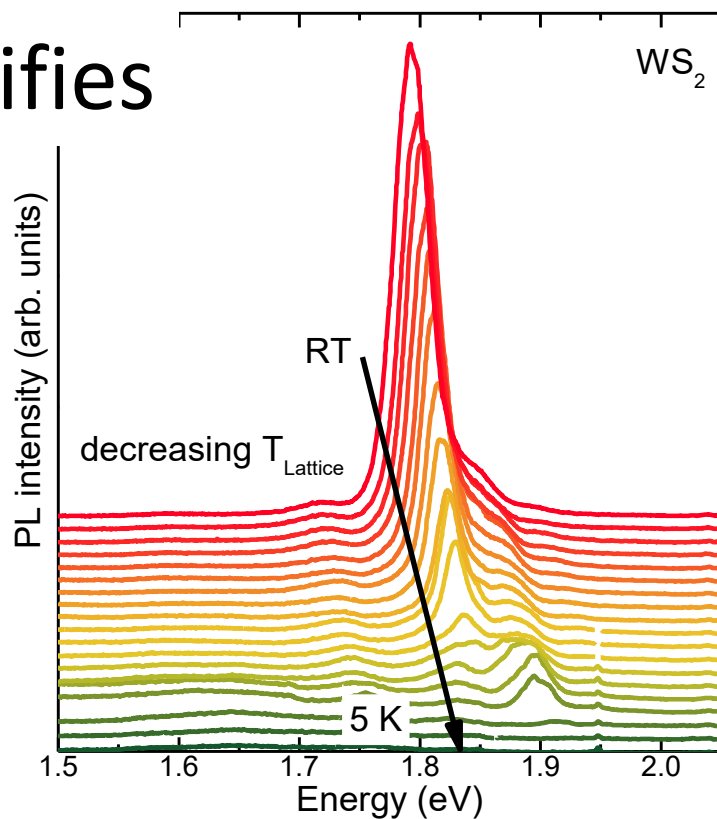
$5 \mu\text{m}$



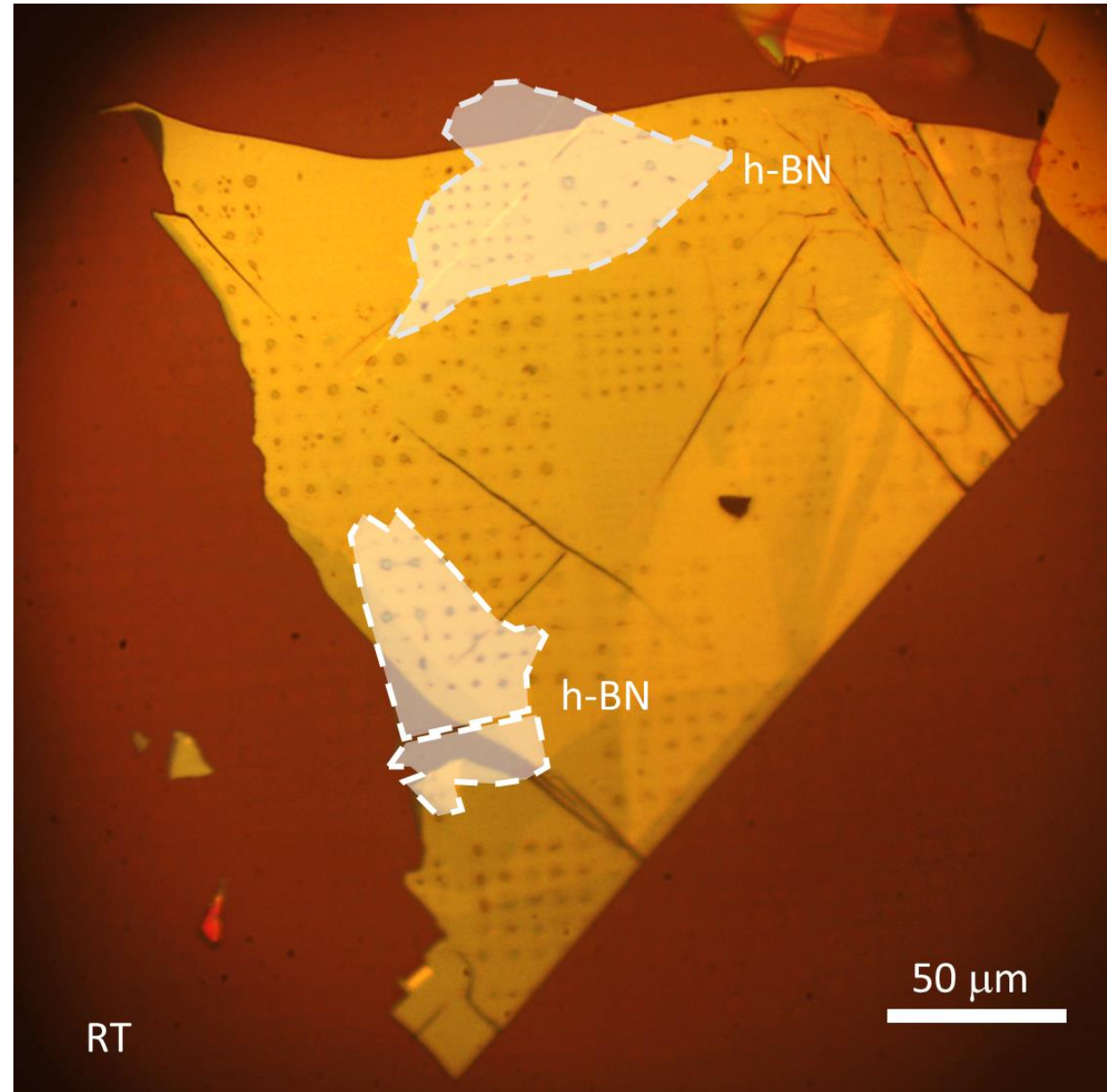
Space-controlled emitters?



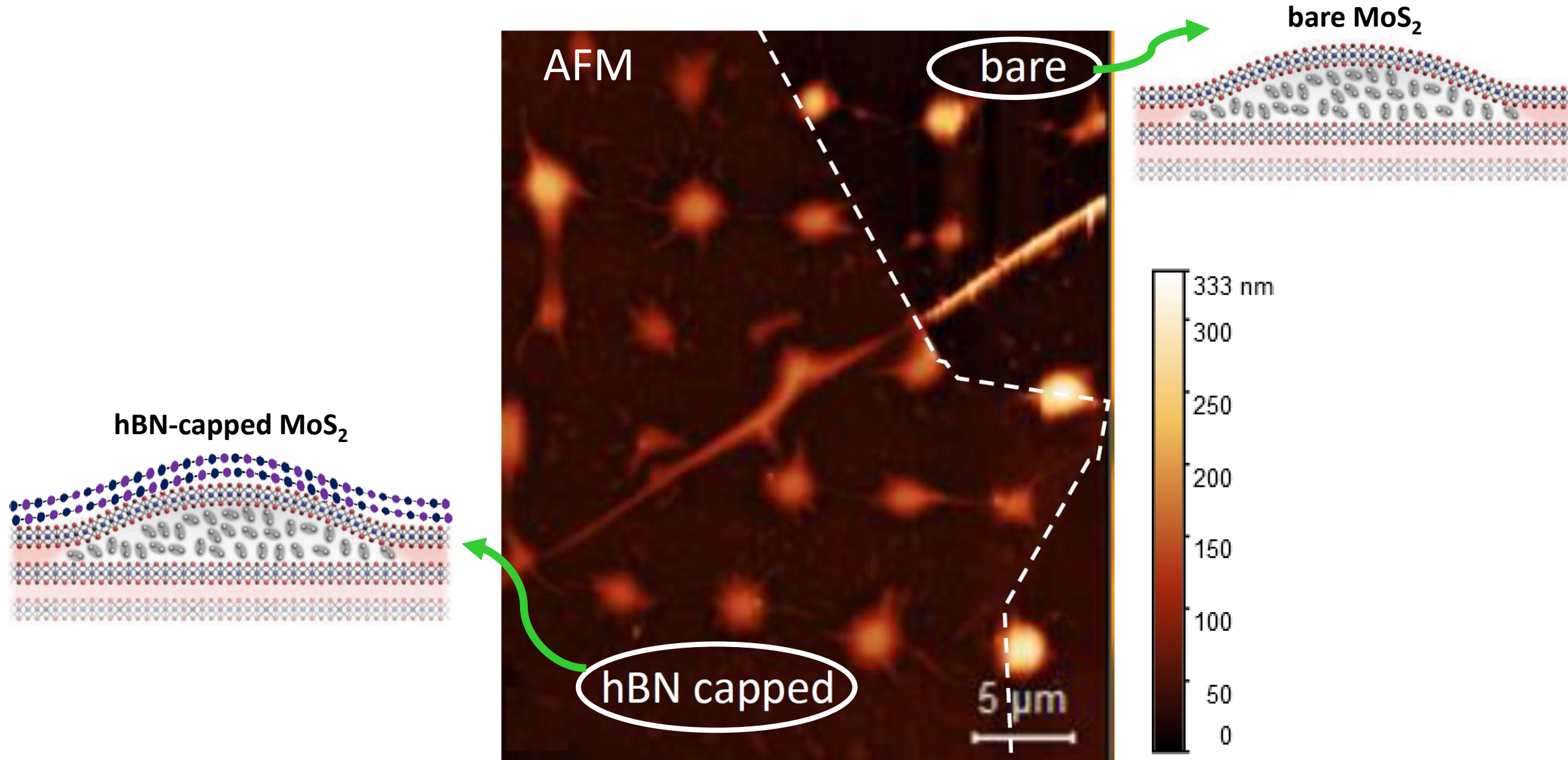
But...
 H_2 liquifies



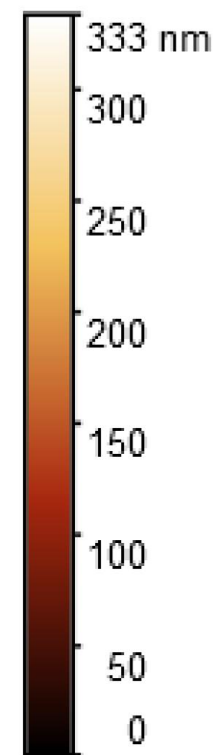
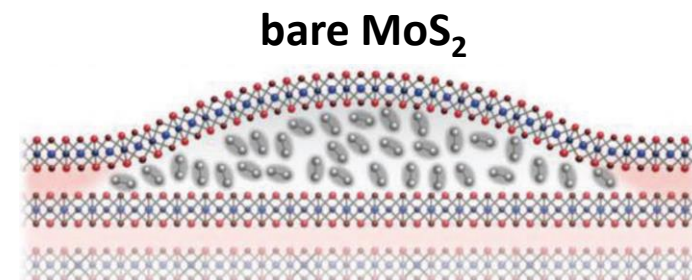
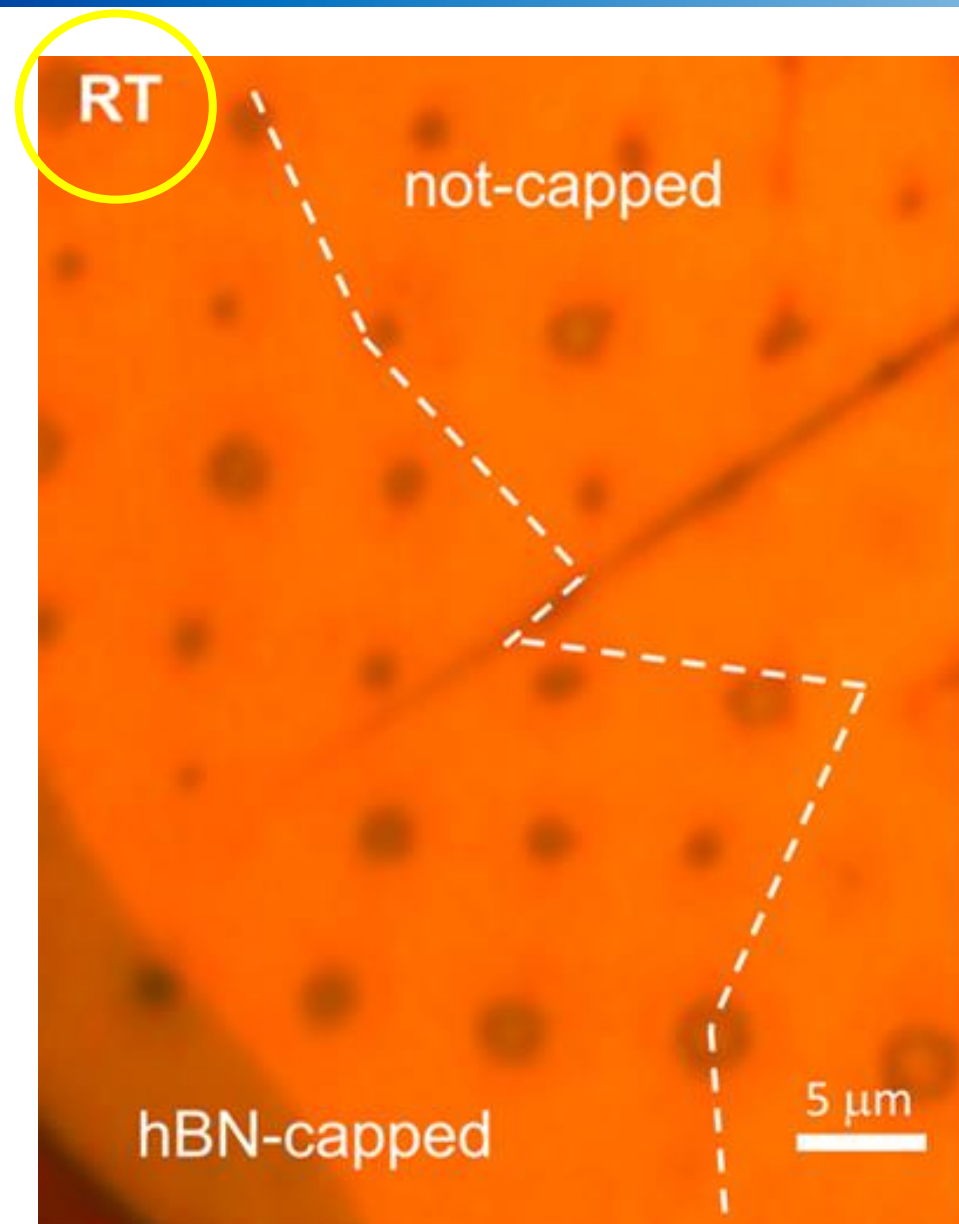
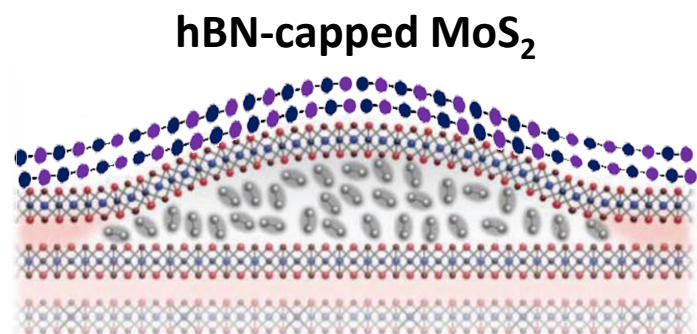
How to circumvent H₂
liquefaction?



hBN heterostructuring

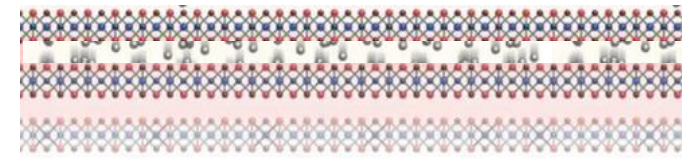
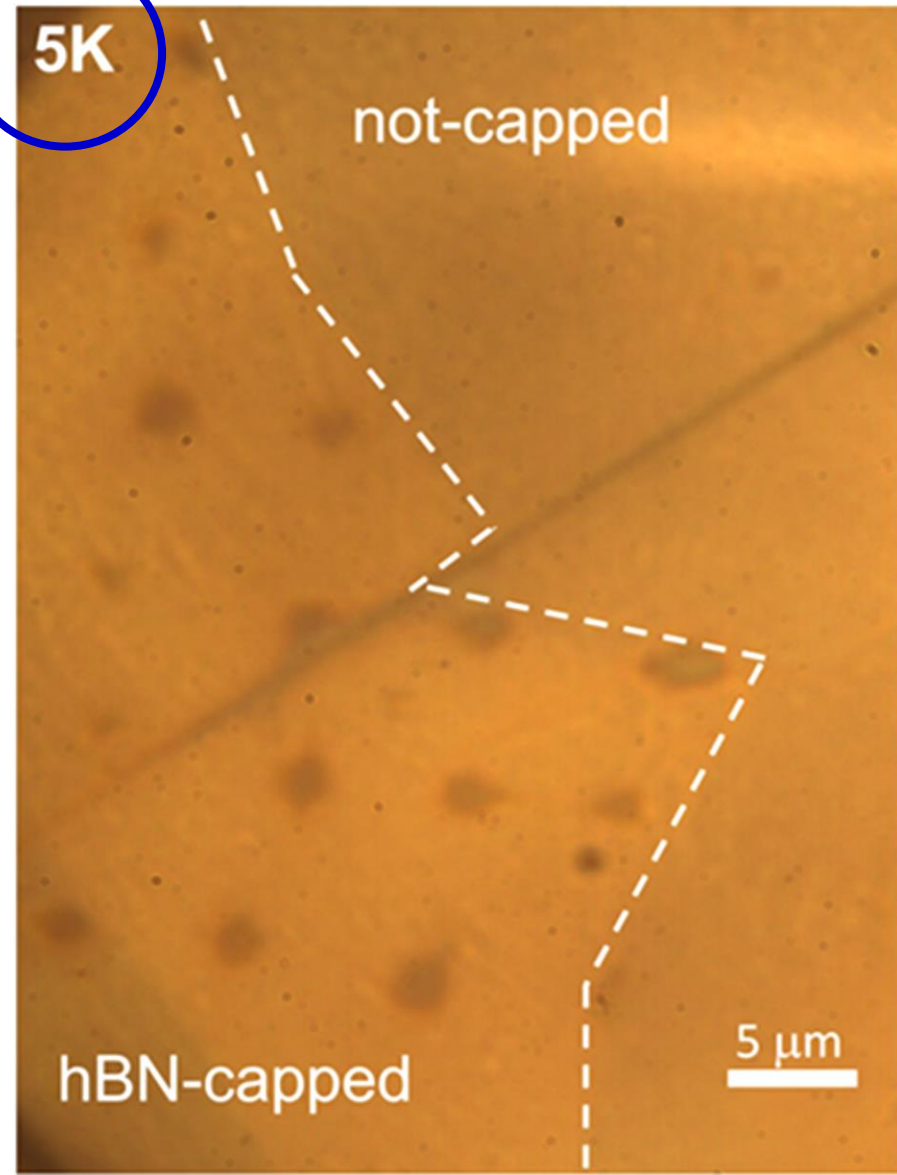


hBN heterostructuring

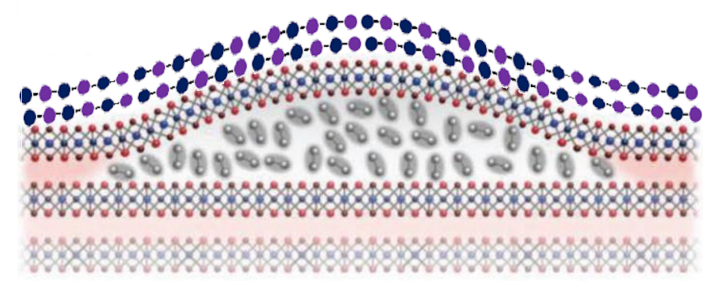


hBN heterostructuring

5K



hBN-capped MoS₂

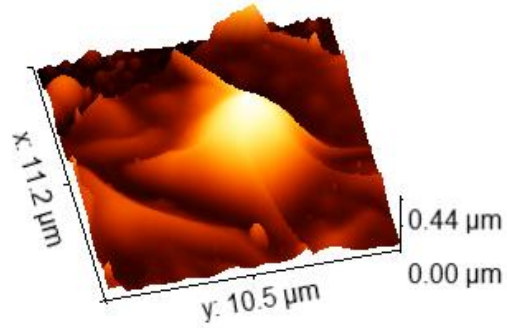
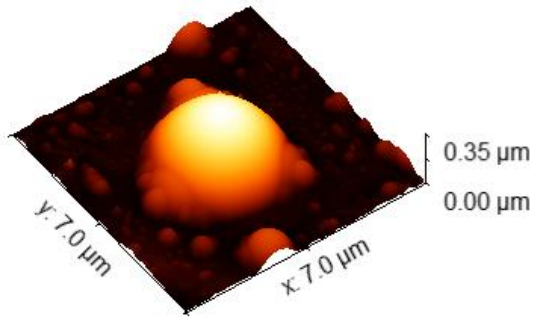


capped domes do not deflate

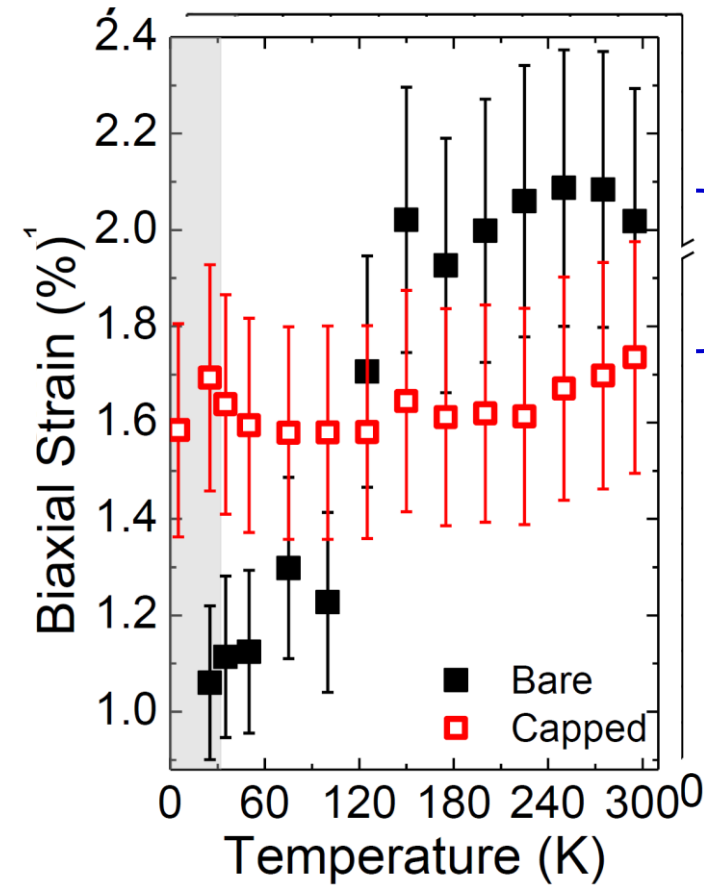
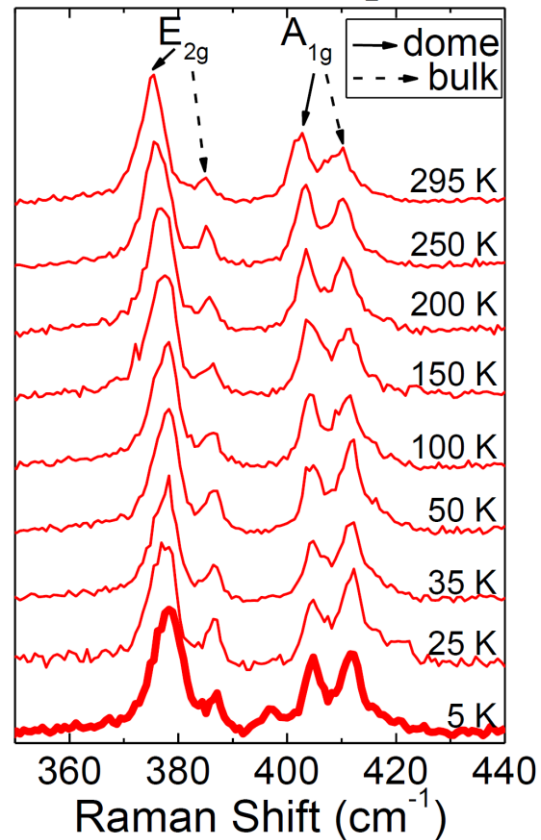
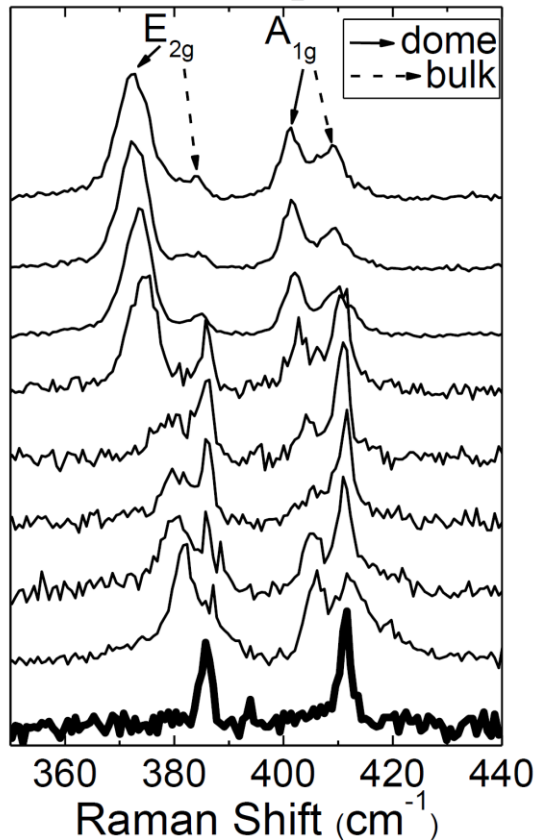
hBN heterostructuring

bare MoS₂ dome

hBN-capped MoS₂ dome



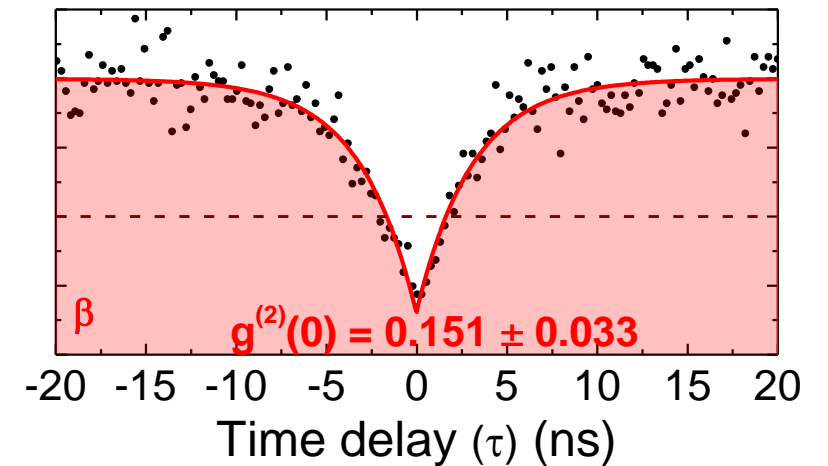
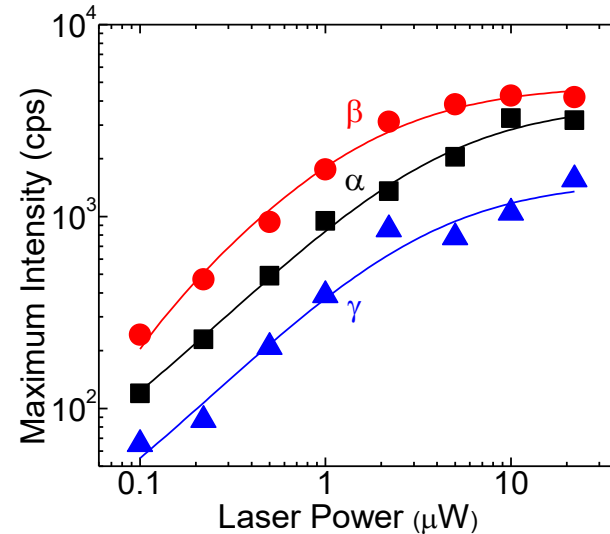
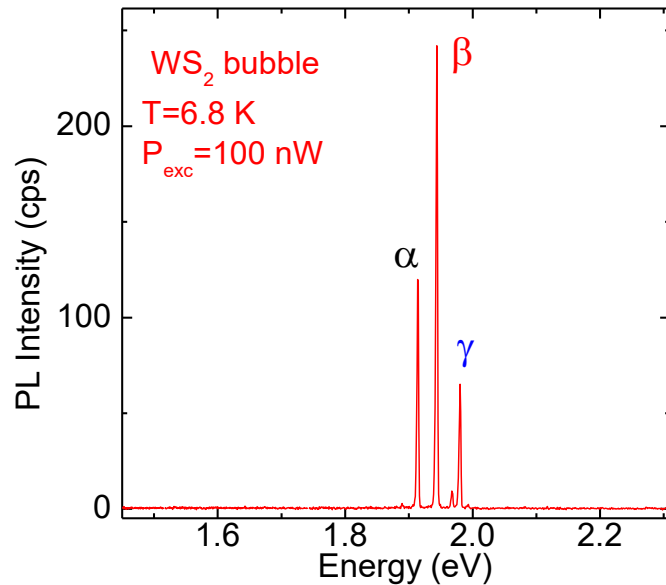
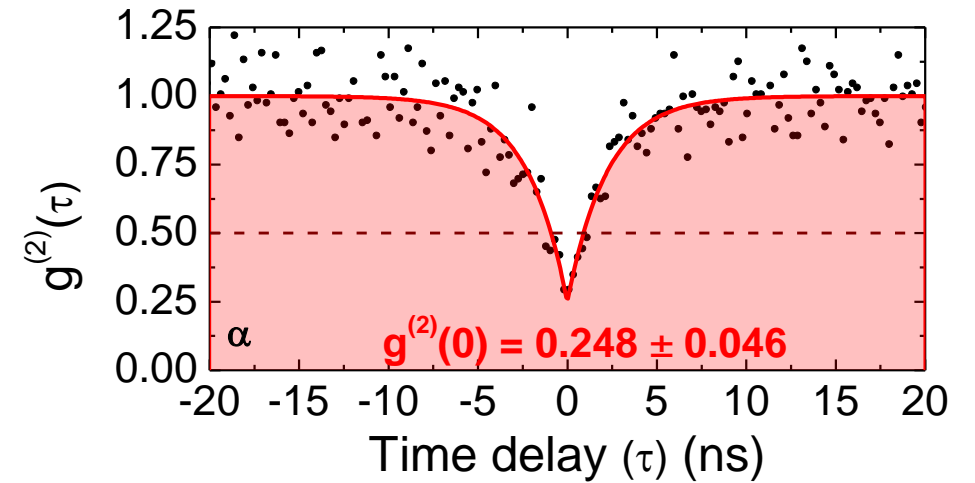
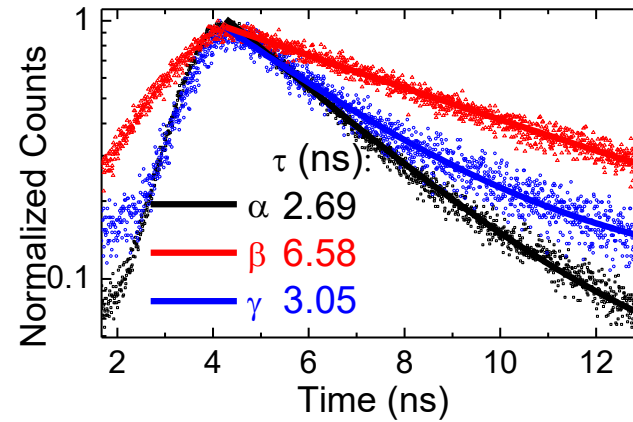
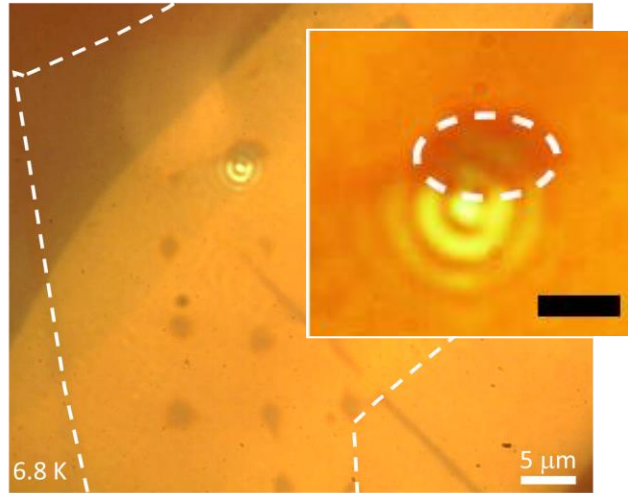
Normalized Raman Intensity (arb. units)



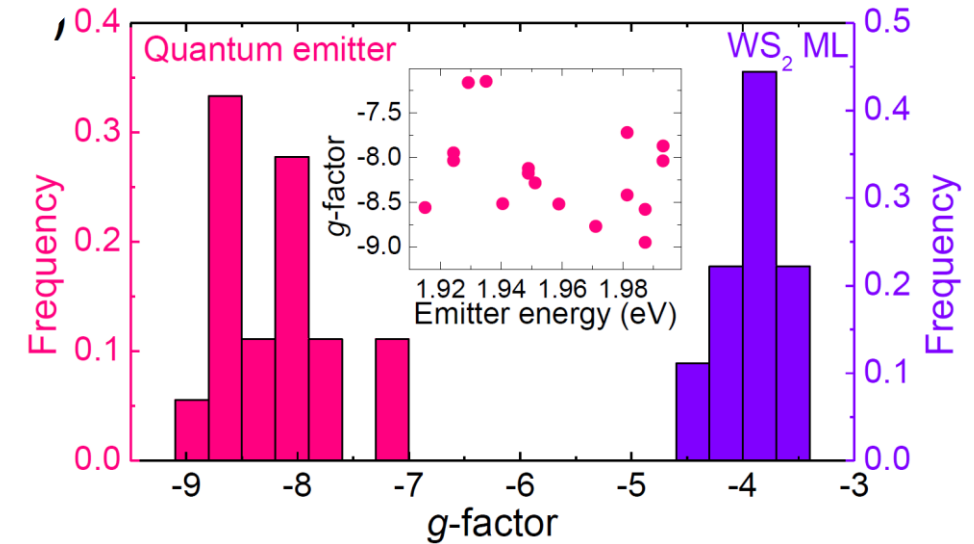
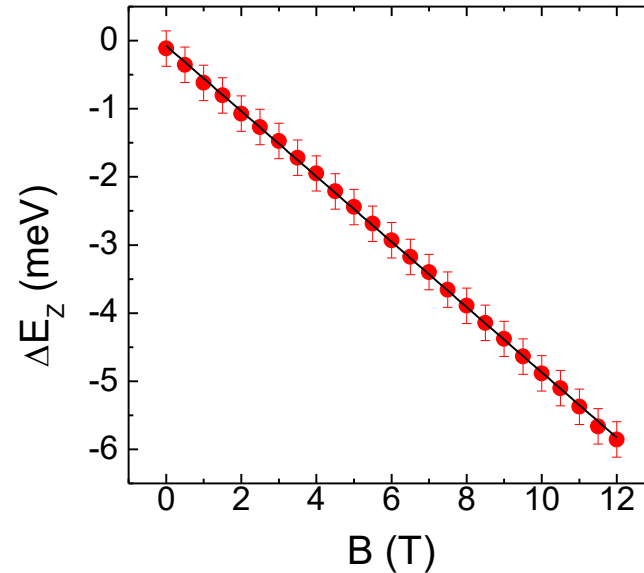
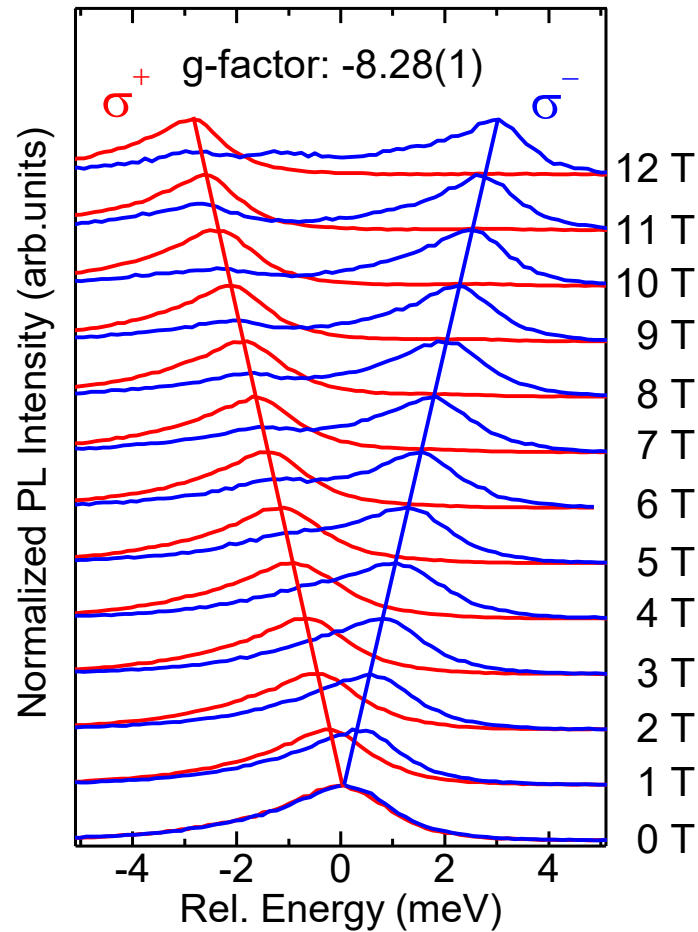
hBN capping causes strain decrease: Elastic energy transfer

$$\varepsilon(T) = \frac{1}{2} \cdot \left(\frac{\omega_{E_{2g}}^{ML}(T) - \omega_{E_{2g}}^{dome}(T)}{\omega_{E_{2g}}^{ML}(T)} \right) \frac{1}{\gamma_{E_{2g}}}$$

Space-controlled quantum emitters



Space-controlled quantum emitters



Like for WSe₂ quantum emitters, the large g value indicates the involvements of an electron in a defect state and a hole in the valence band

$$\Delta E_Z = g_{\text{exc}} \mu_B B$$

Conclusions

- Durable, spatially controlled domes can be created in TMDs and hBN

- The domes host complex strain field and act as efficient light emitters

- Complex strain fields give access to exciton hybridization phenomena

- Applications for site-controlled quantum light sources

