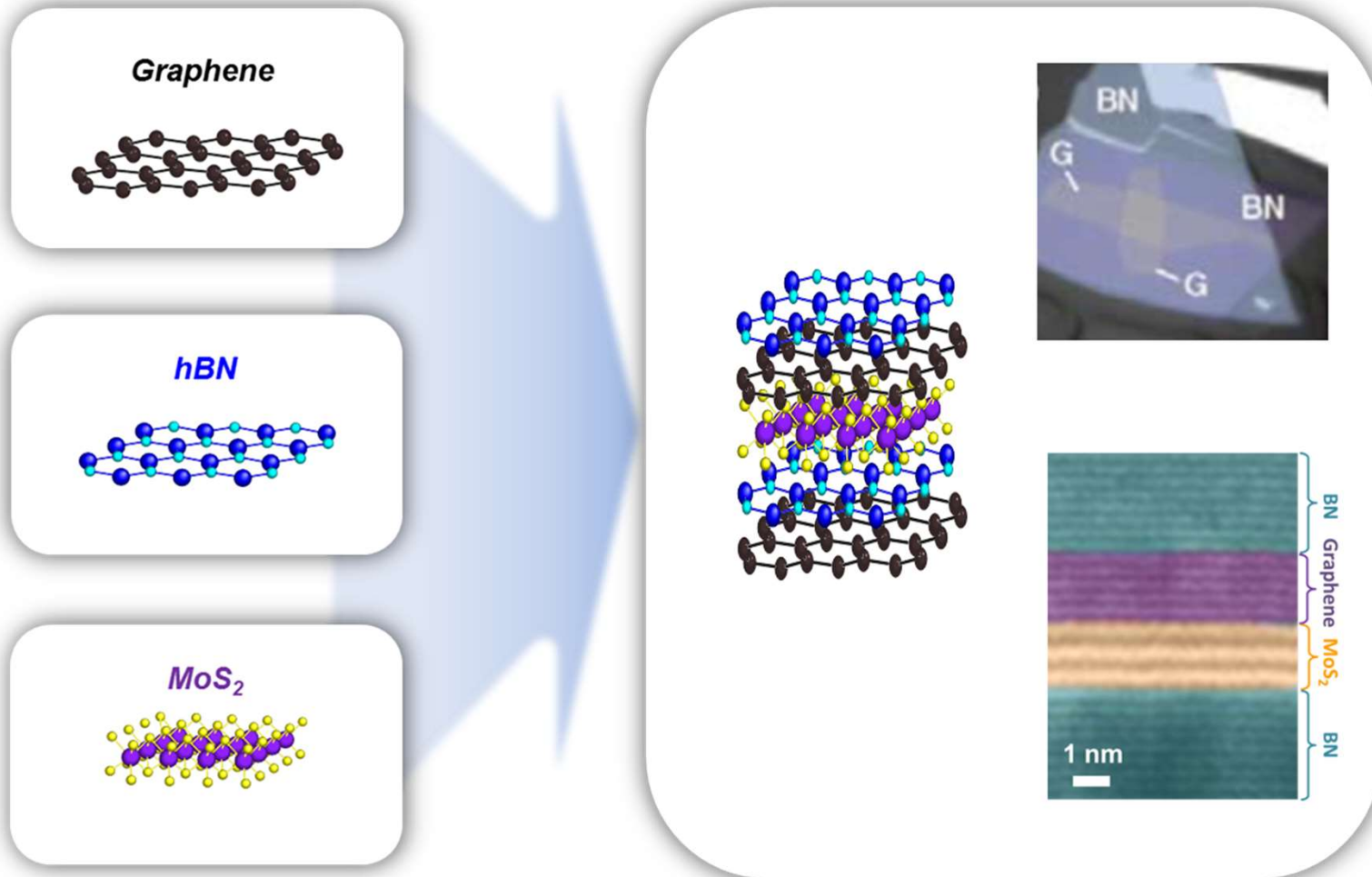


# Two-dimensional materials and applications

## 8. van der Waals Heterostructures

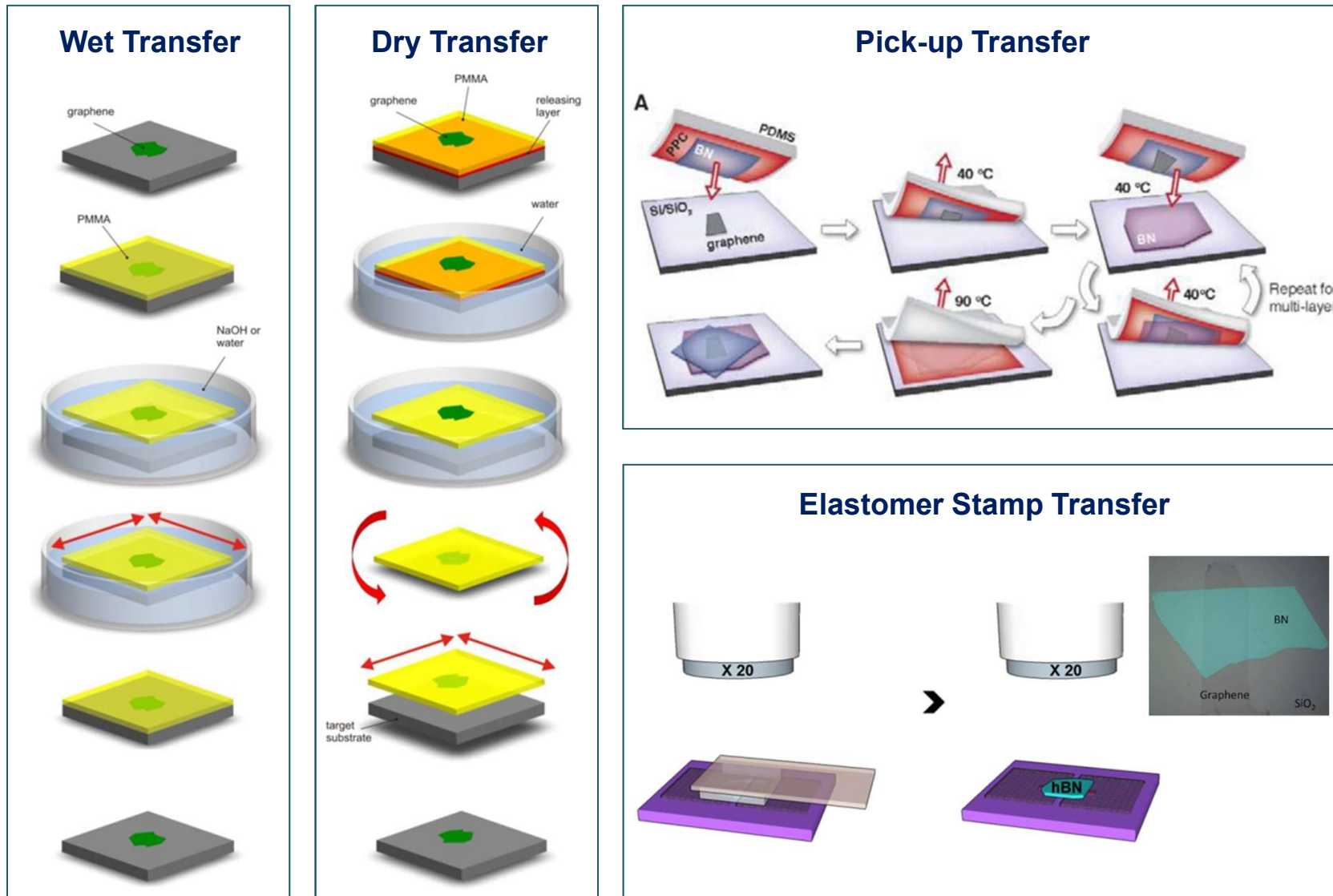


# van der Waals Heterostructures



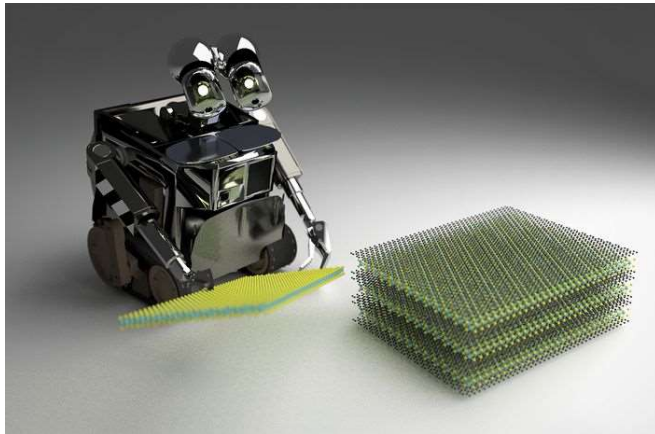
Ultrathin devices for flexible & transparent  
Ultrasharp heterointerface formed through vdW force

# Stacking Technique for 2D Heterostructures

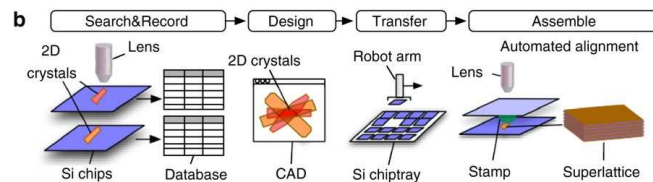
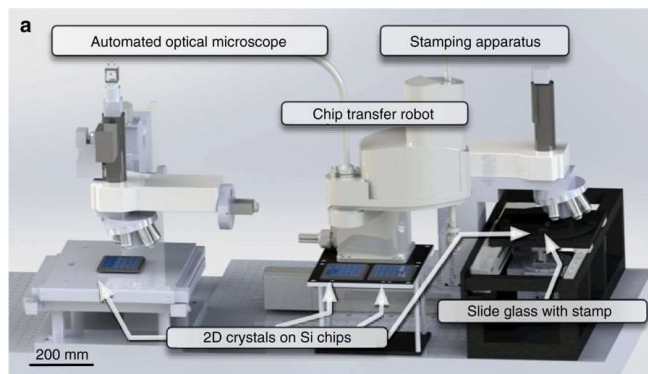


# Stacking Technique for 2D Heterostructures

## Robotic assembly of artificial nanomaterials



A. Castellanos-Gomez et al. *Nat. Nanotechnol.* (2018)

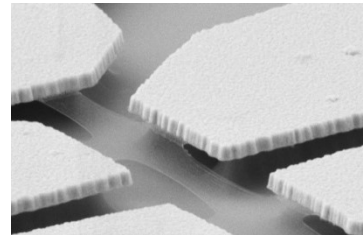
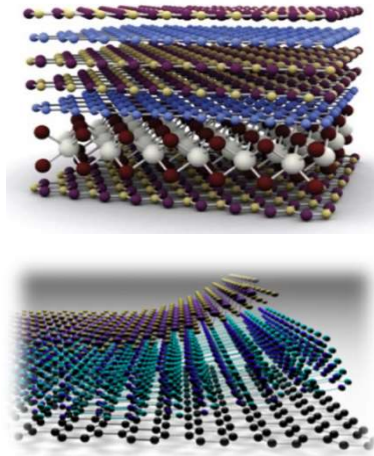


T. Machida et al. *Nat. Commun.* (2018)



# van der Waals Heterostructures

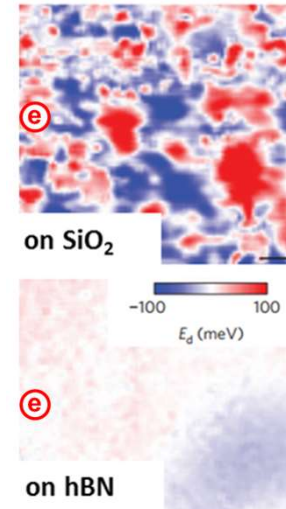
## Effect of hBN on Electrical Transport of Graphene



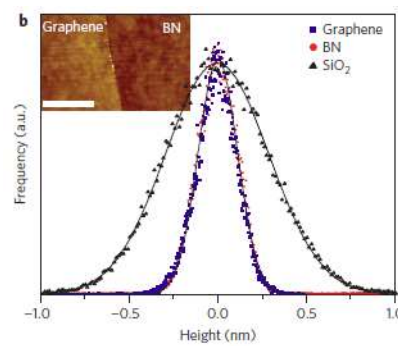
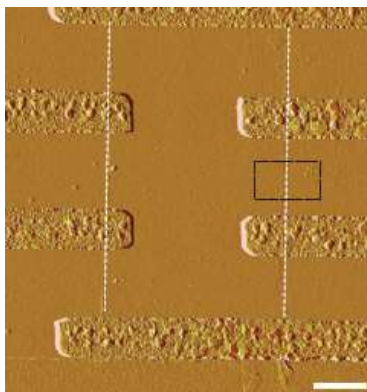
Graphene on SiO<sub>2</sub>



Graphene on hBN

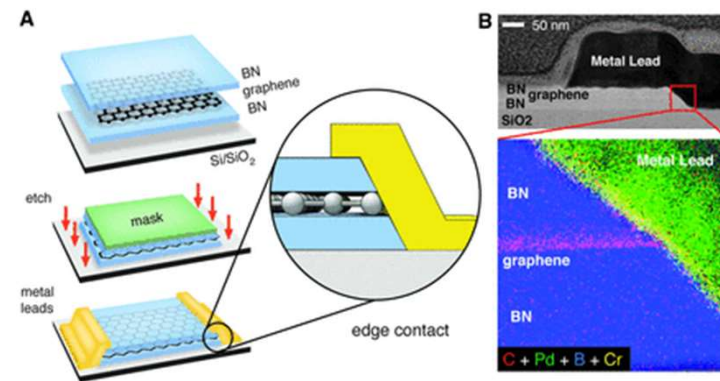


*J. Xue et al. Nat. Mater. (2011)*



**40,000 cm<sup>2</sup>/V·s at 300K**

*C. Dean et al. Nat. Nanotech. (2010)*

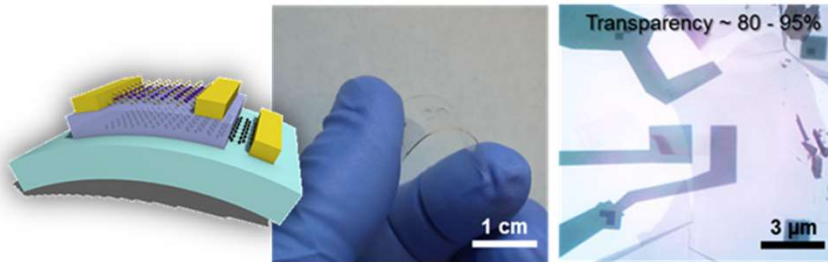


**Room-temperature mobility > 140,000 cm<sup>2</sup>/Vs**

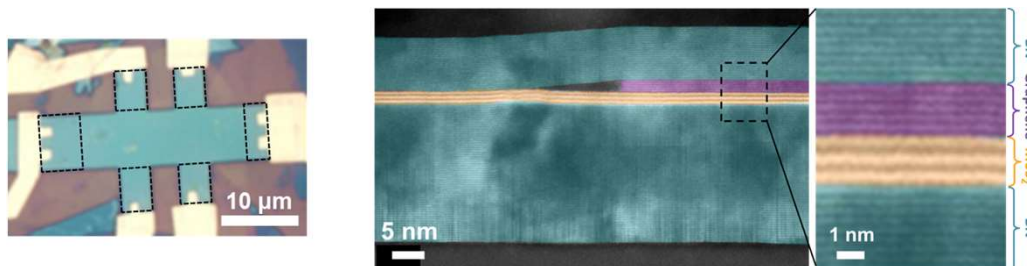
*L. Wang et al. Science (2014)*

# Advantages of van der Waals Heterostructures

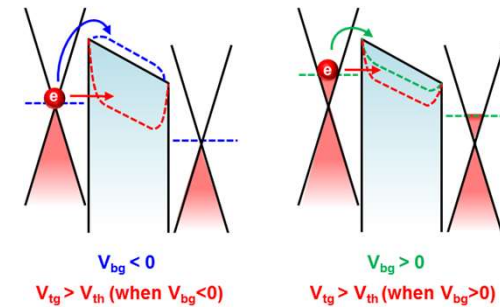
## 1. Ultrathin devices for flexible & transparent electronics



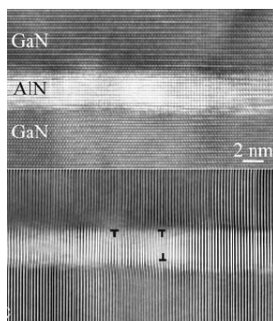
## 2. Ultrasharp heterointerface



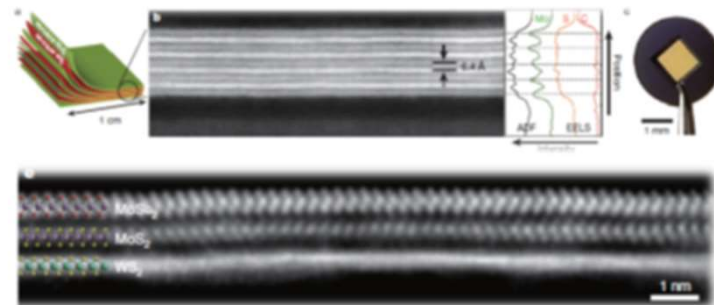
*G.H. Lee et al. ACS Nano (2013)*



## 3. Arbitrary stacking



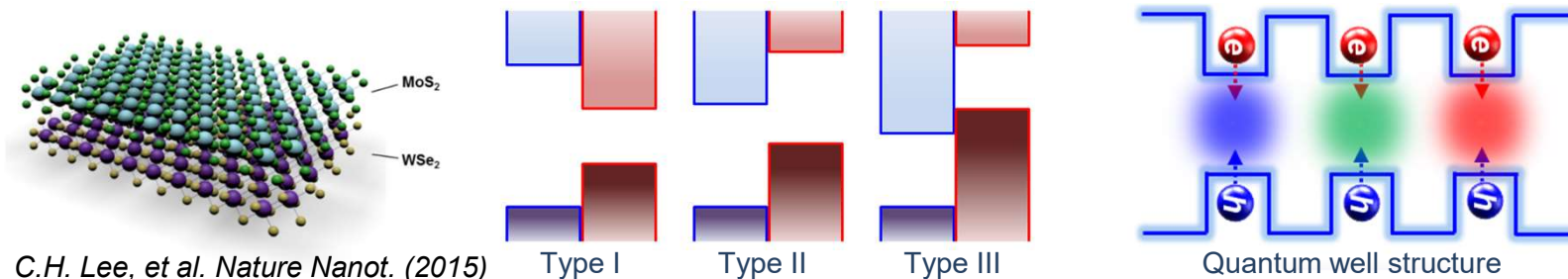
vs.



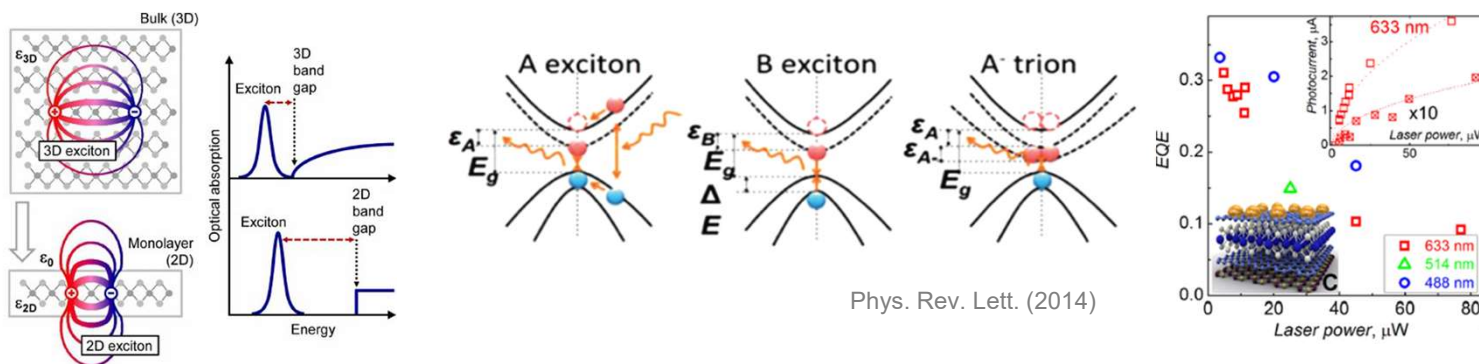
*J. P et al. Nature (2017)*

# Advantages of van der Waals Heterostructures

## 4. Band engineering

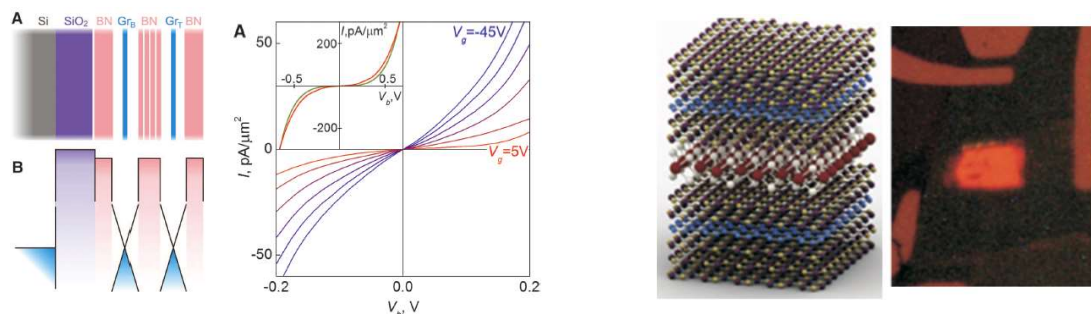


## 5. Strong light-matter interaction



L. Britnell et al. *Science* (2013)

## 6. Vertical confinement in 2D space (quantum well structure)



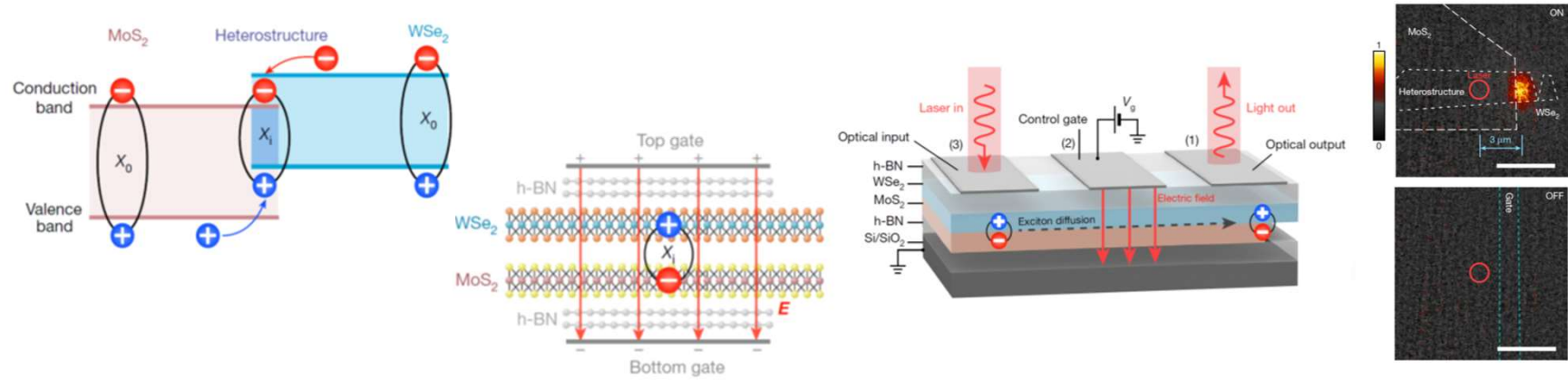
R. V. Gorbachev et al. *Science* (2012)

F. Withers et al., *Nat. Mater.* (2015)



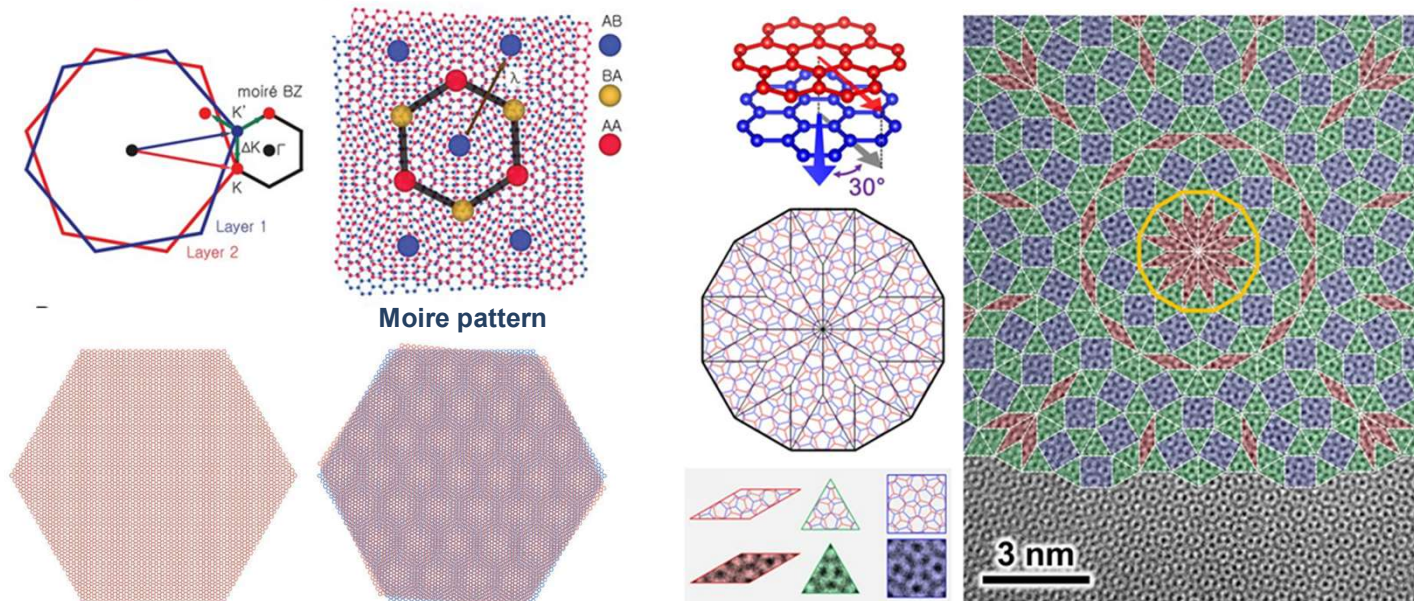
# Advantages of van der Waals Heterostructures

## 7. Interlayer interaction (interlayer exciton)



A. Kis et al. Nature (2018)

## 8. Periodic potential modulation / Novel symmetry material systems (Quasicrystal)

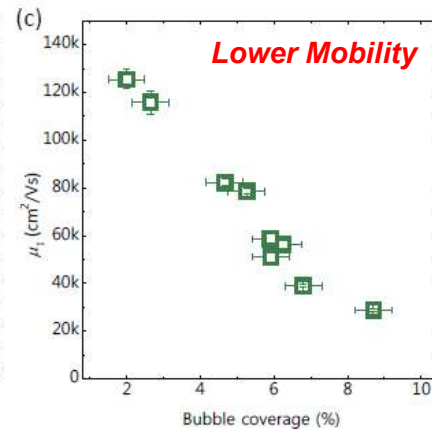
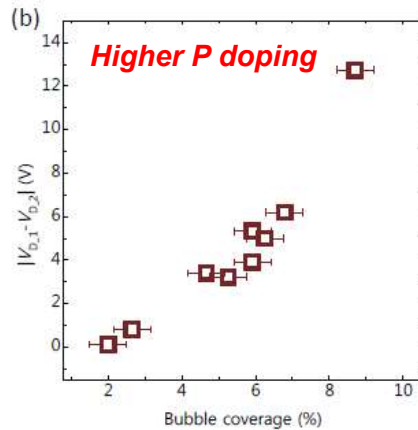
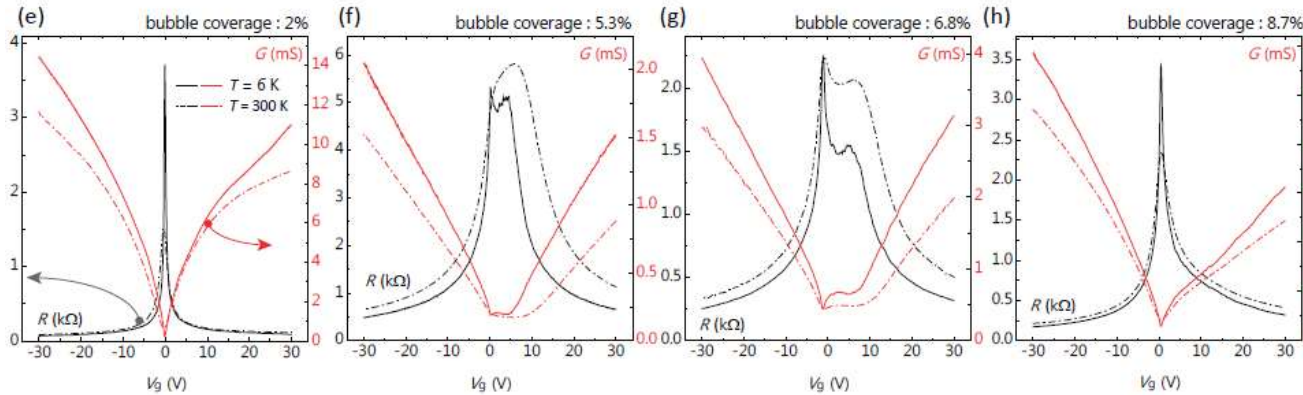
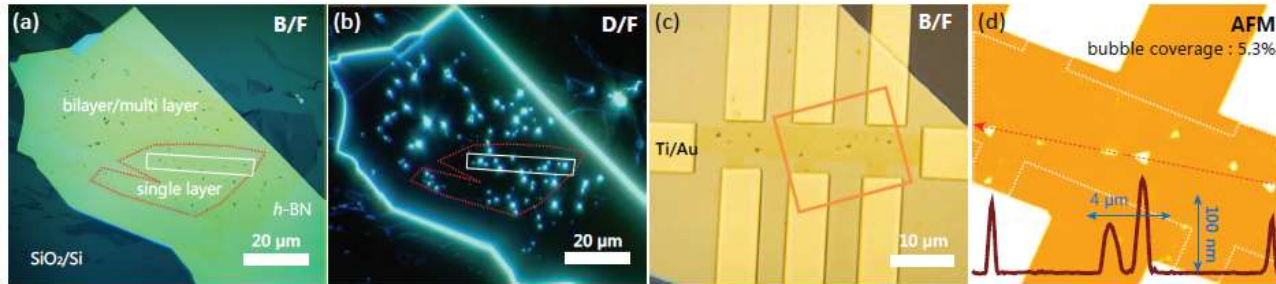


L. A. Ponomarenko et al. Science (2019)

Y.W. Son et al. Science (2018)

# Issue in Stacking

## Formation of bubbles

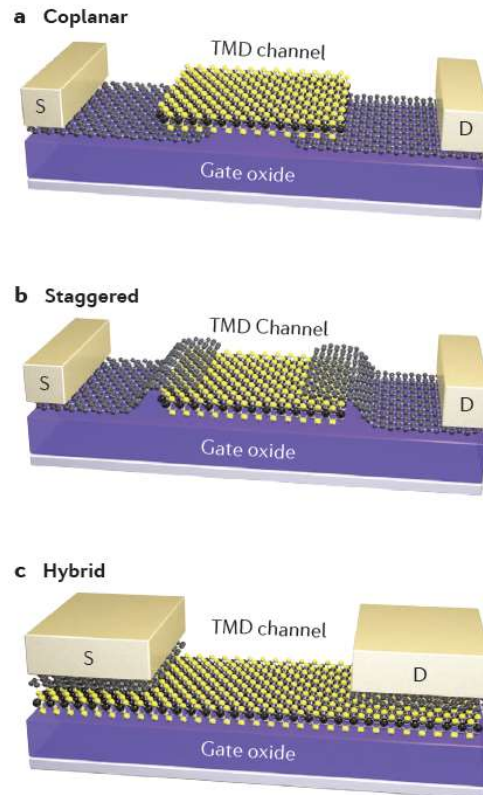


- Contaminant inside graphene bubbles are mostly hydrocarbon. And hydrocarbons behave like **p-type dopants**
- **Low mobility** attribute to scattering effects from long and short range disorders induced by bubble graphene



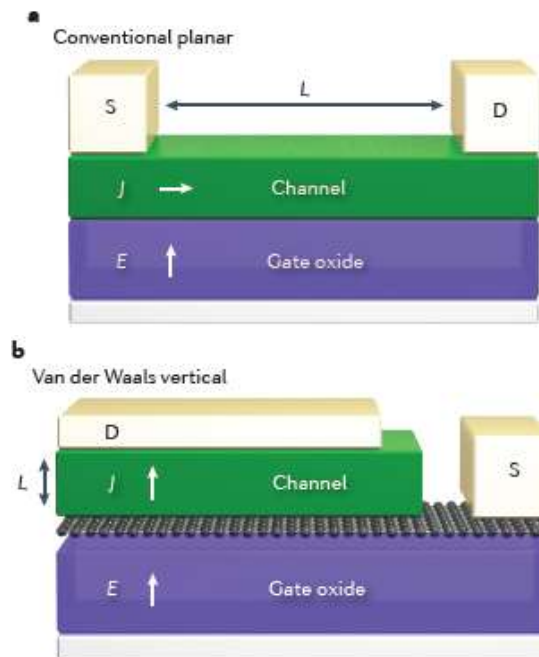
# Various Types of vdW Heterostructure Devices

## 1. Vertical contact



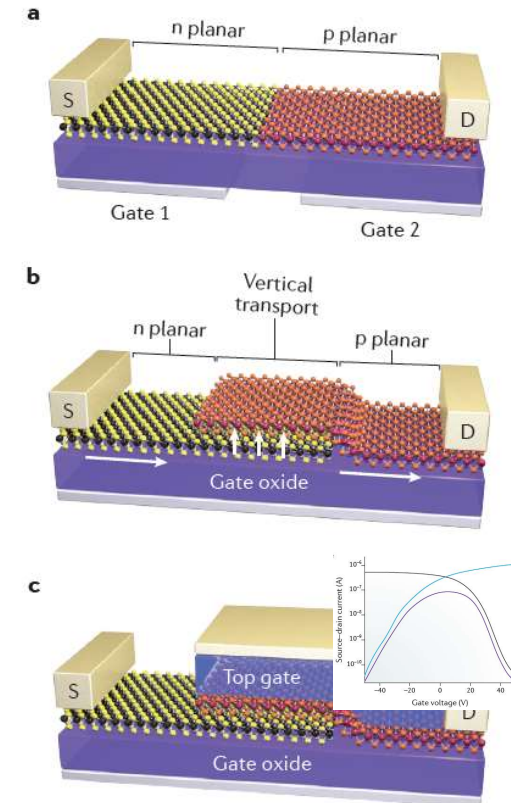
- Versatile work-function tunable contact
- Less contamination
- Non-damaging interface between metal and 2D channel

## 2. Vertical transistors



- High-speed, low-power and flexible
- Tunneling vertical transistors

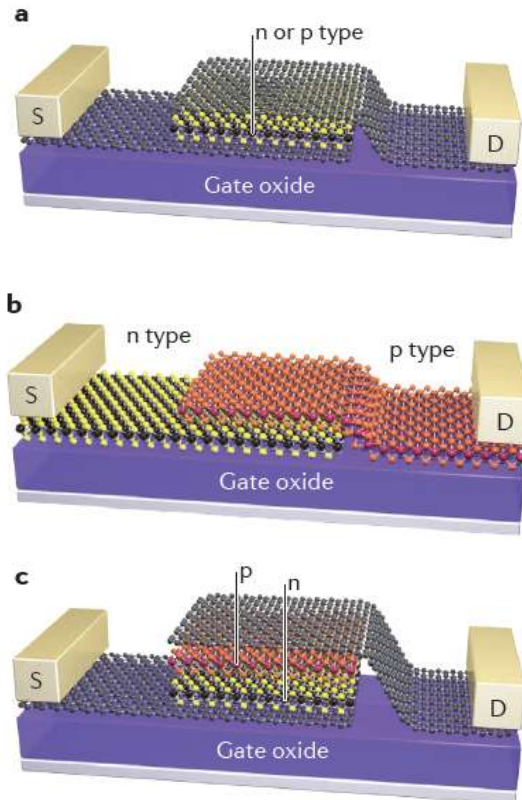
## 3. Vertical diodes



- Unique E-field controlled p-n diodes
- Anti-ambipolar behavior

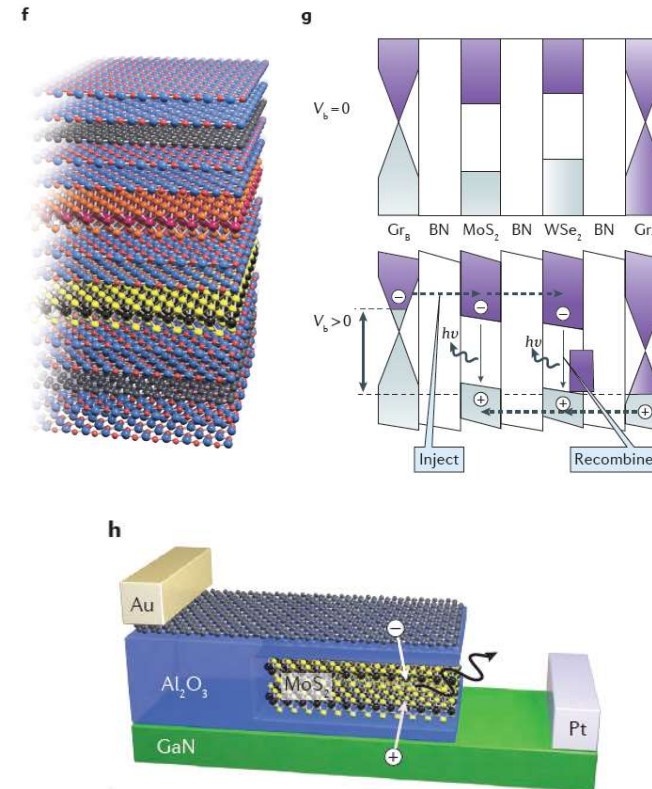
# Various Types of vdW Heterostructure Devices

## 4. Light harvesters and detectors



- Gate-dependent photoresponse (tunable carrier density)
- Dynamic modulation of diode characteristics
- Rapid extraction of exciton

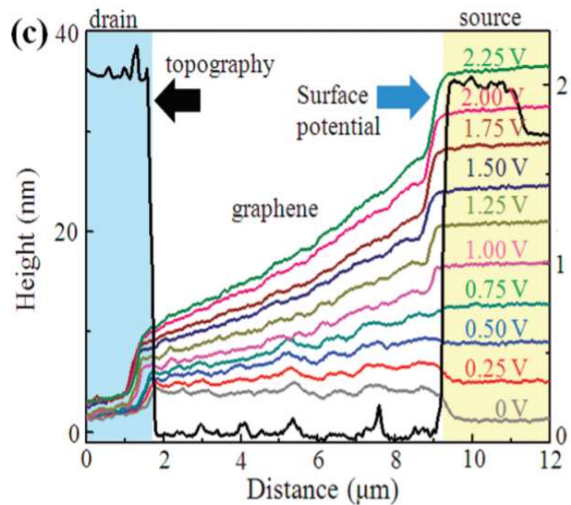
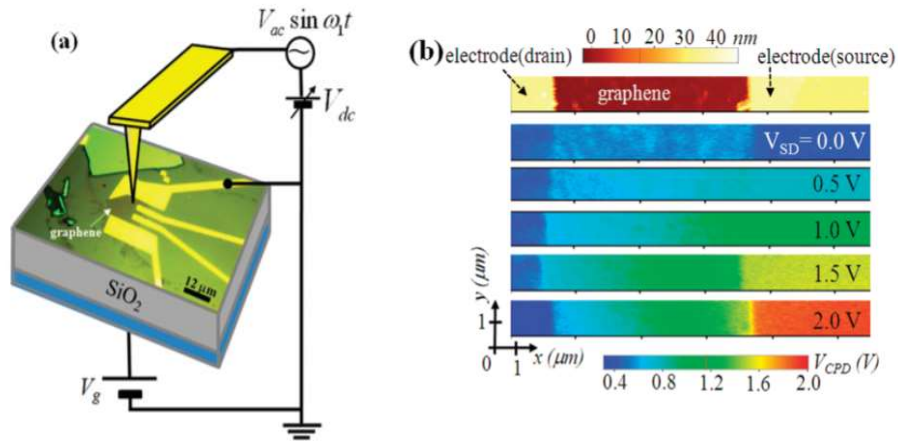
## 5. Light emitting devices



- Vertical device structure
- Vertical current injection
- Quantum well structure

# Heterostructure of Graphene/hBN

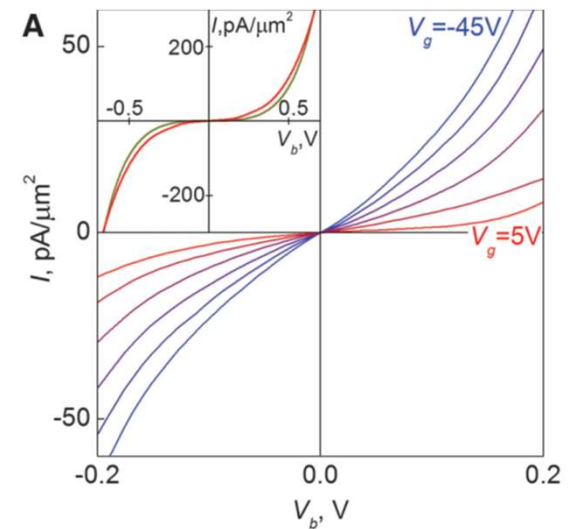
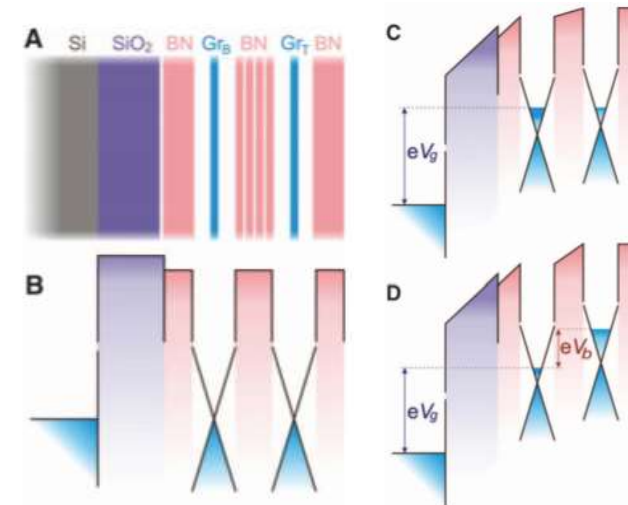
## Tunable Schottky barrier in graphene



## Tuning the graphene work function by electric field

*P. Kim et al. Nano. Lett. (2009)*

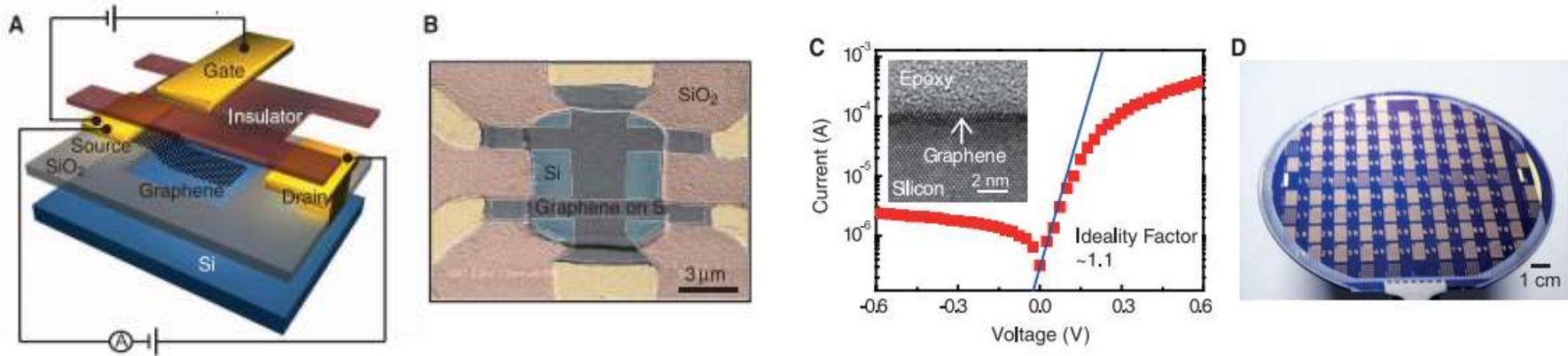
## Field-effect tunneling transistor



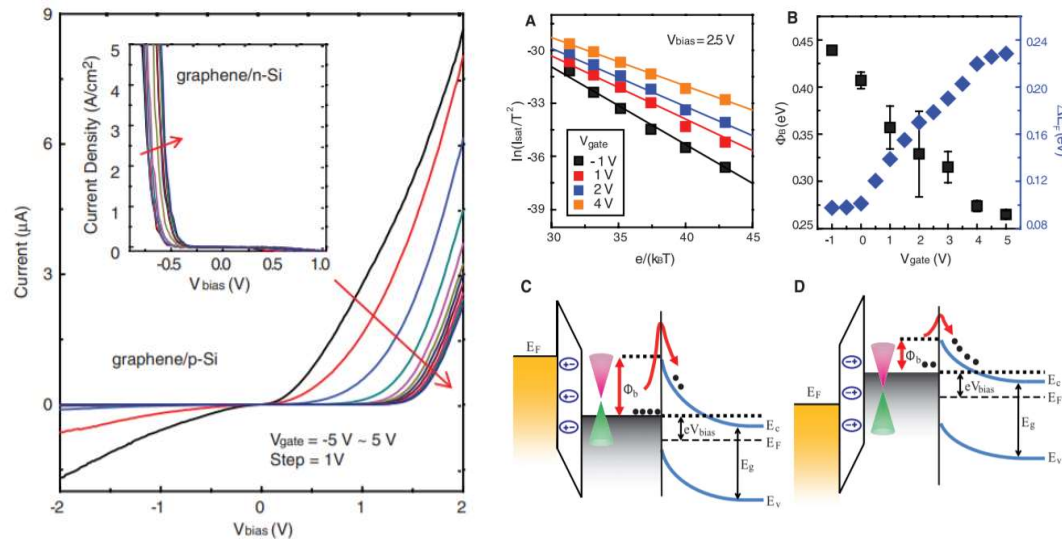
*L. A. Ponomarenko et al. Science (2012)*

# Barristor

## Graphene barristor



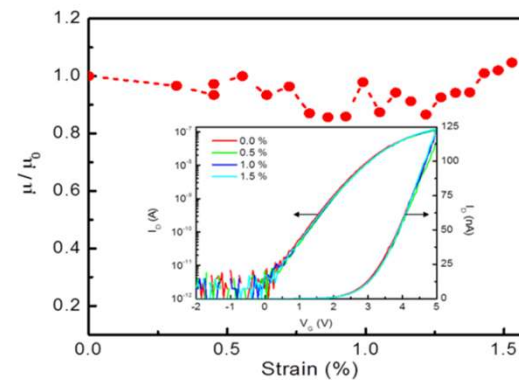
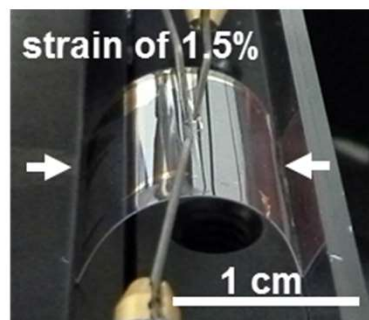
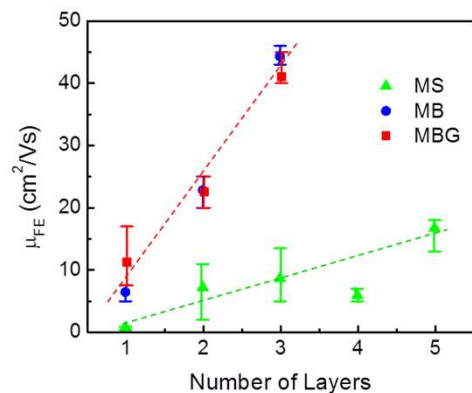
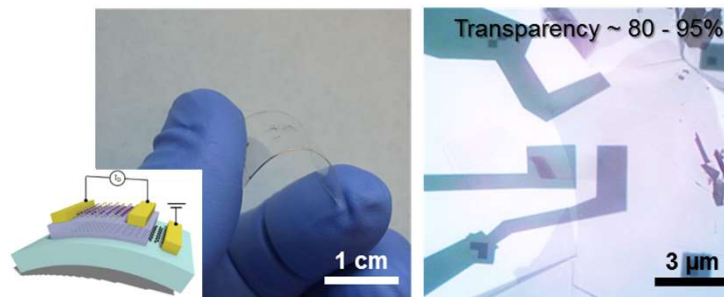
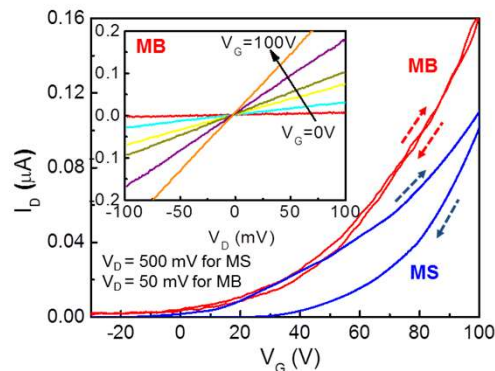
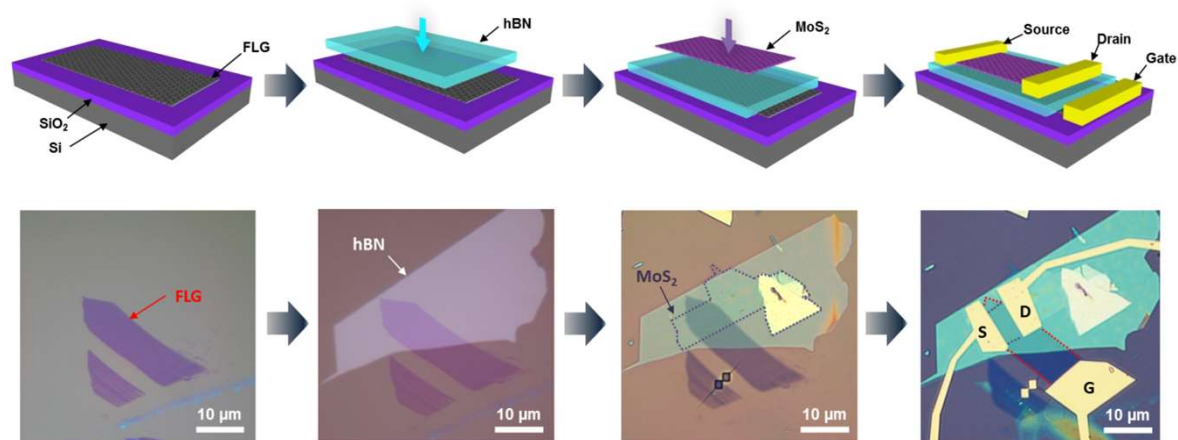
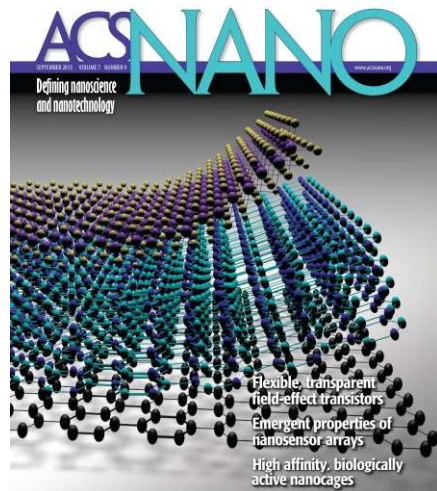
## Tunable Schottky barrier



- Barristor = Barrier + transistor
- Barristor is a device that controls the on-off ratio by changing the Schottky barrier of graphene and silicon via the applied gate voltage.

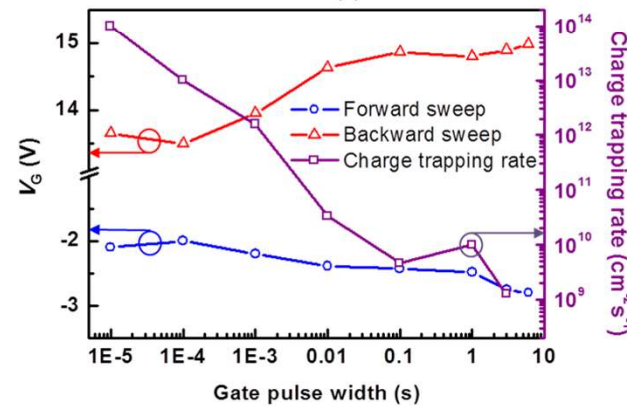
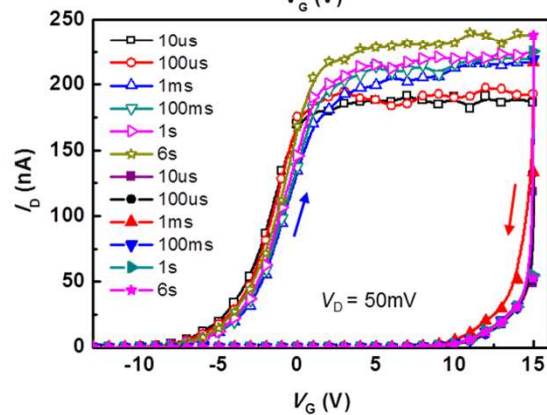
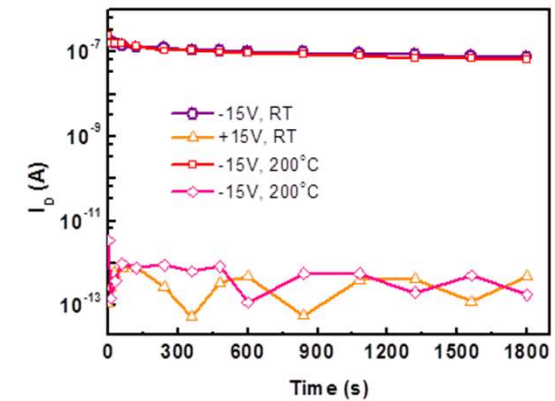
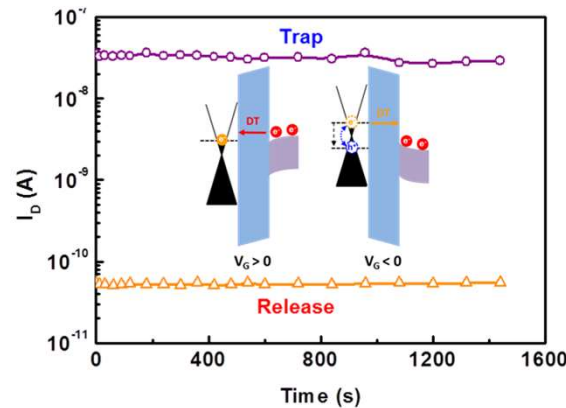
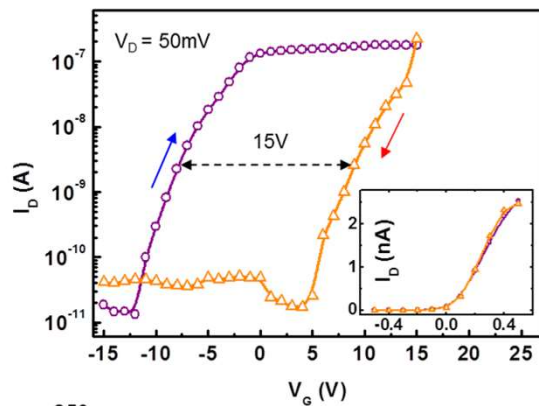
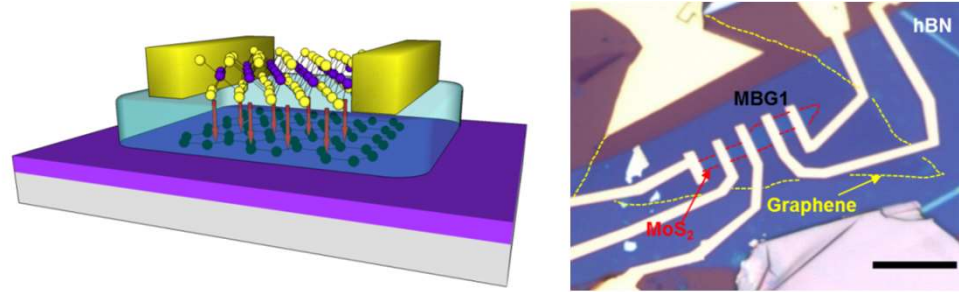


# Flexible and Transparent Heterostructure Devices





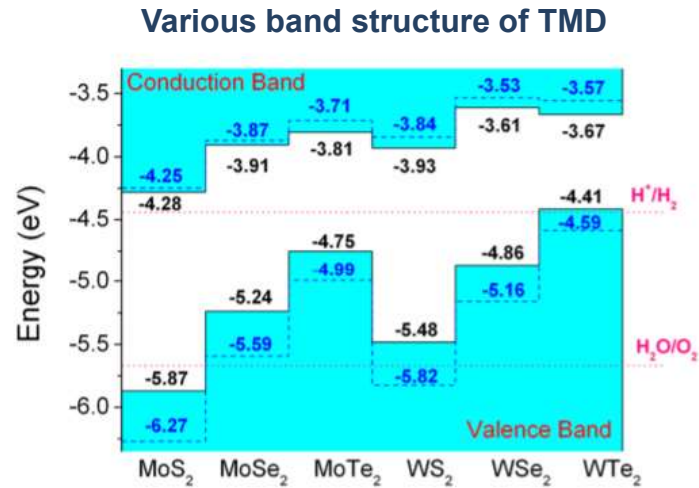
# Heterostructure Charge Trap Memory



- High stability
- High carrier mobility
- high on/off ratio
- Large memory window
- Low operating voltage
- Small dimension
- Transparency and flexibility

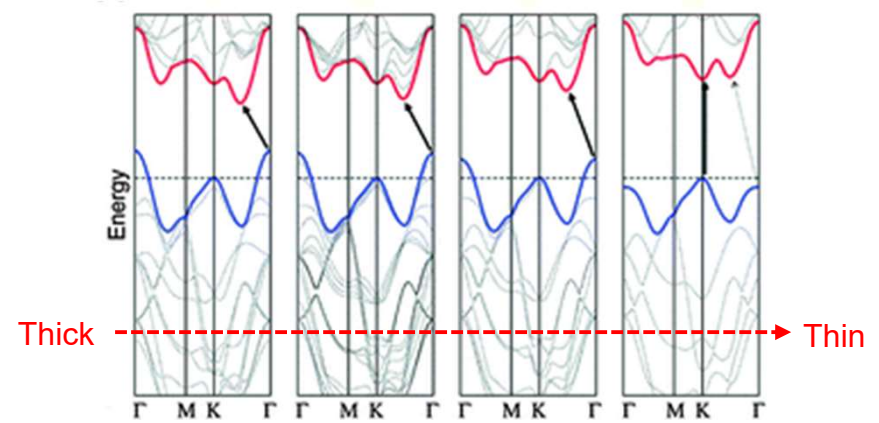
# Heterostructure of TMD/TMD

## Band engineering



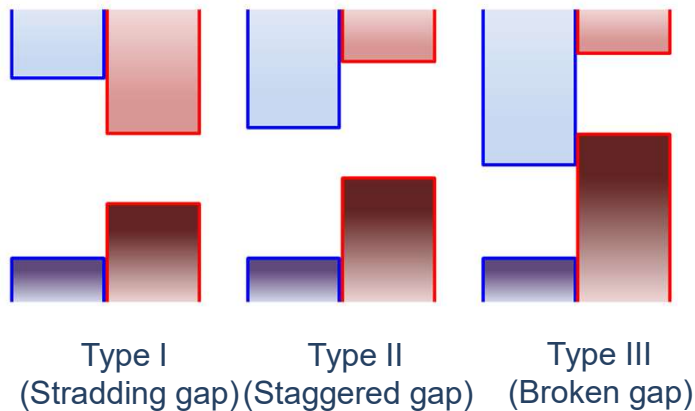
*J. Wu et al. Appl. Phys. Lett. (2013)*

## Band structure dependent on TMD thickness

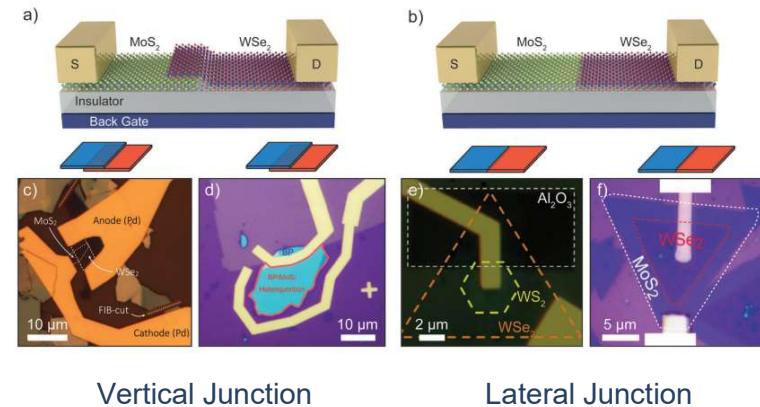


*F. Wang et al. Nano Lett. (2010)*

## Type of band alignment



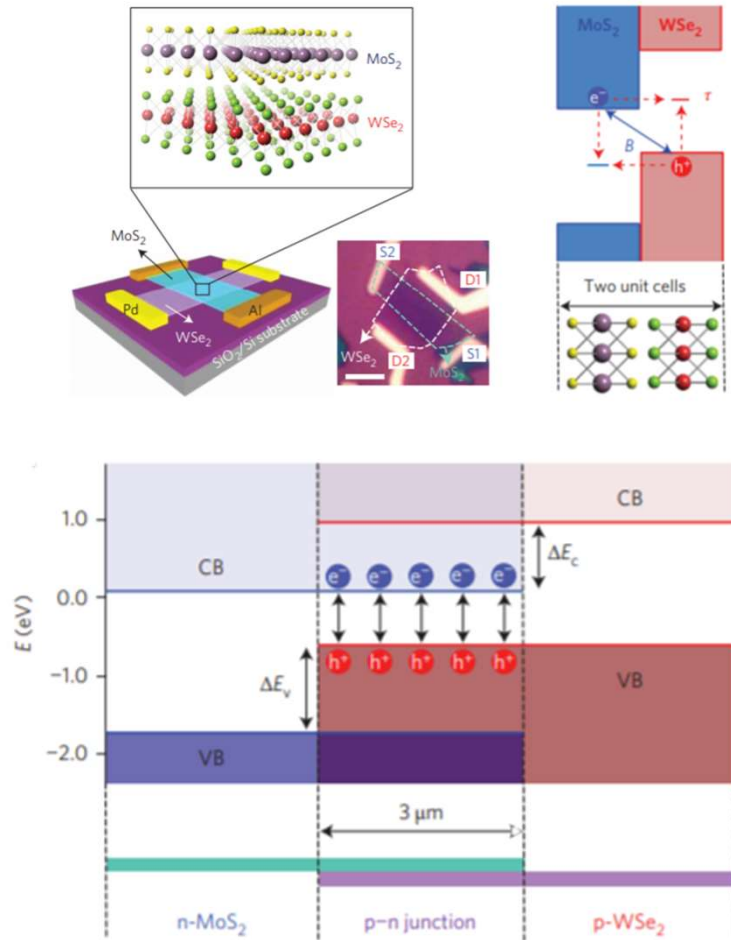
## Type of heterojunction



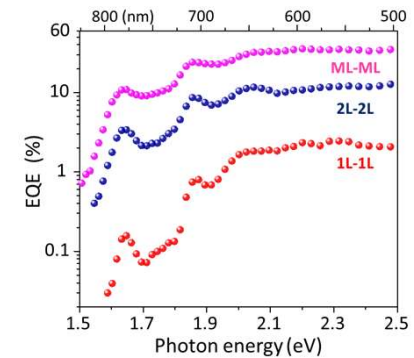
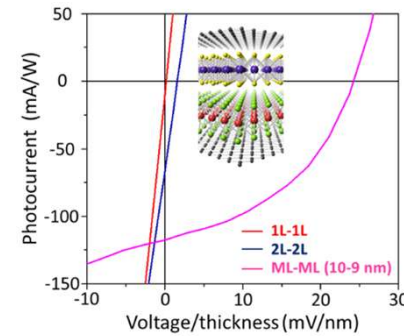
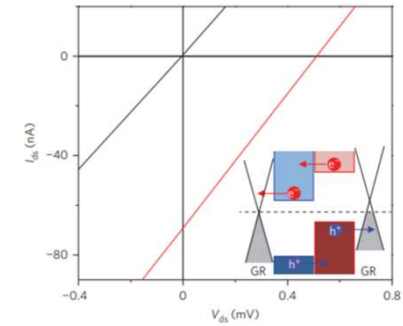
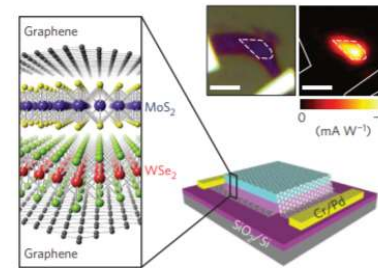
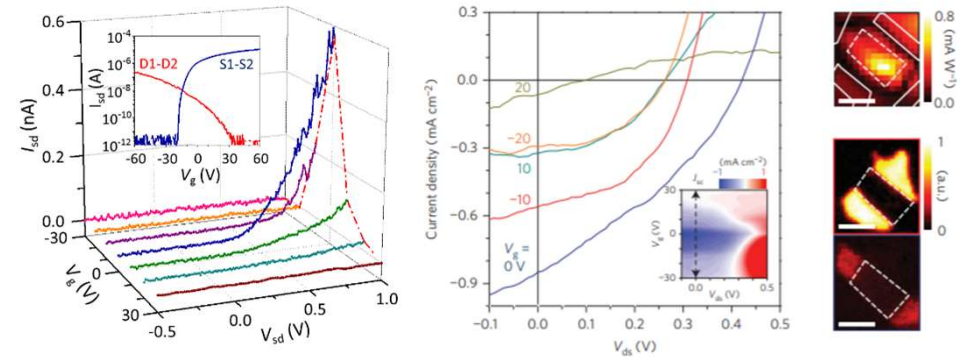
*H. S. J. van der Zant et al Chem. Soc. Rev. (2018)*

# Heterostructure of TMD/TMD

## Atomically PN junction



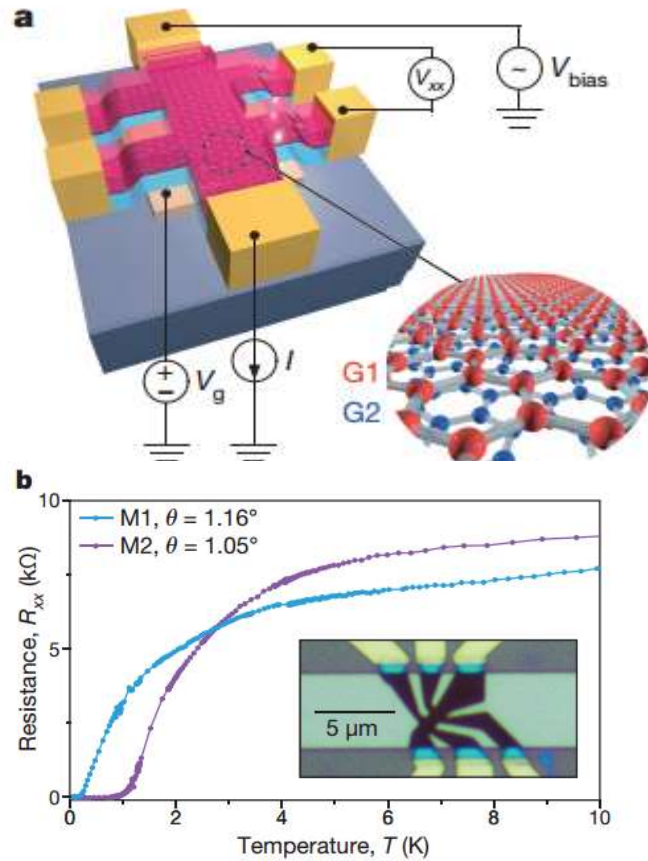
## PN Junction device





# Heterostructure of Graphene/Graphene

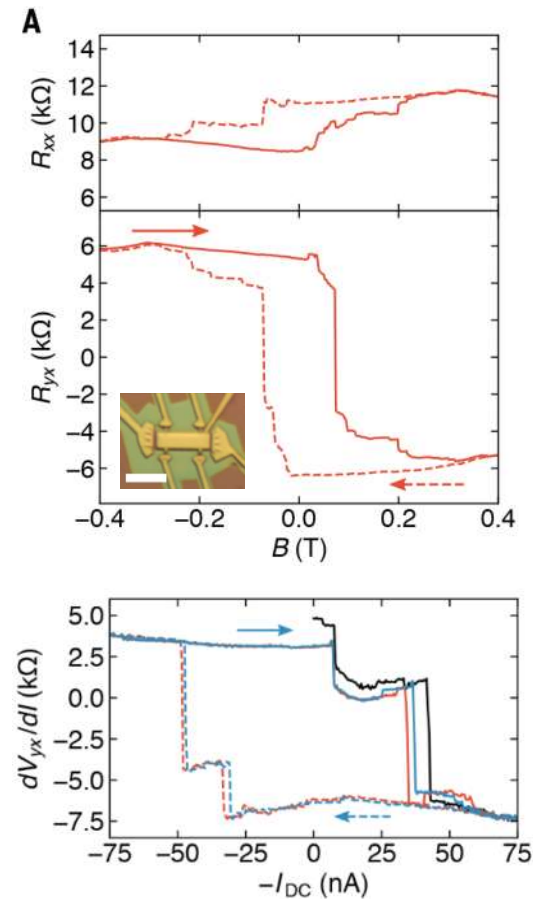
## Twisted bilayer graphene



**Superconductivity**

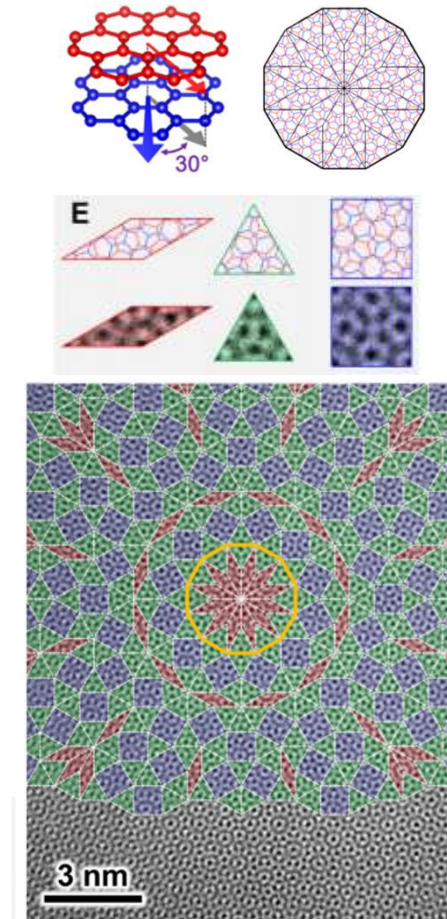
*P Jarillo-Herrero et al. Nature. (2018)*

## Graphene quasi-crystal



**Ferromagnetism**

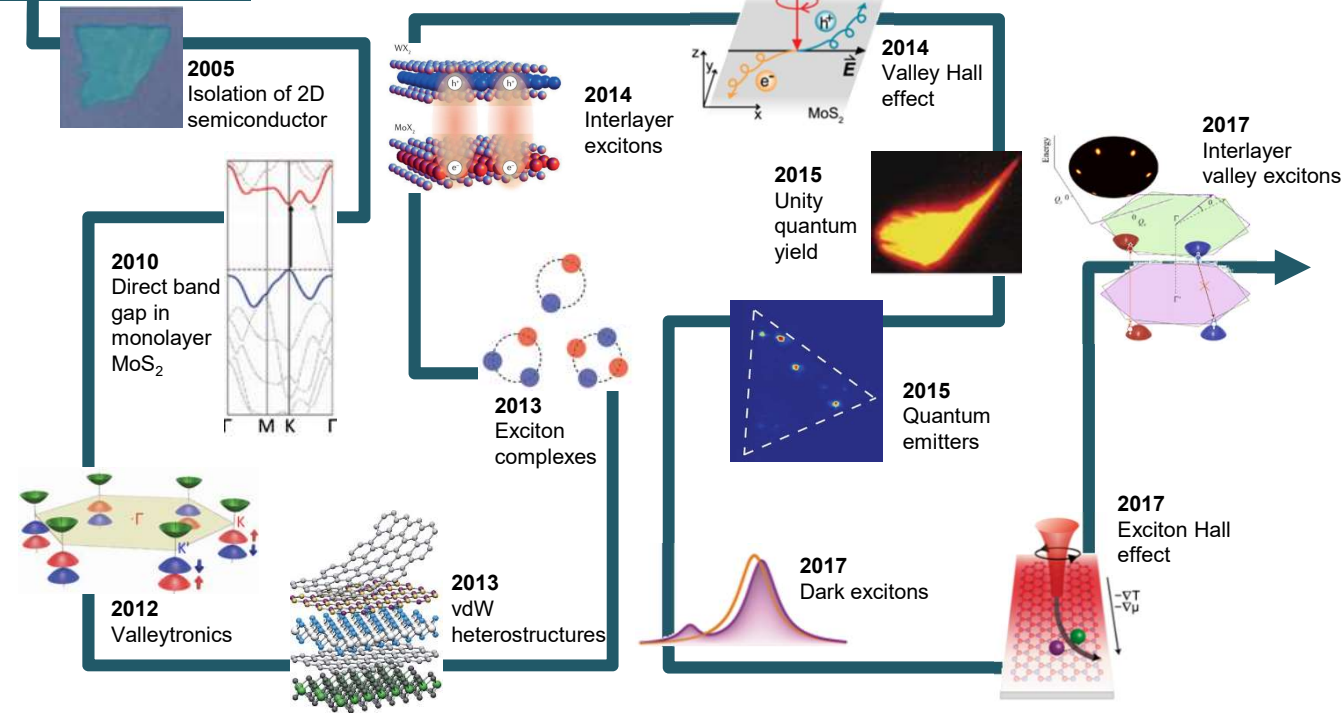
*L. A. Ponomarenko et al. Science (2019)*



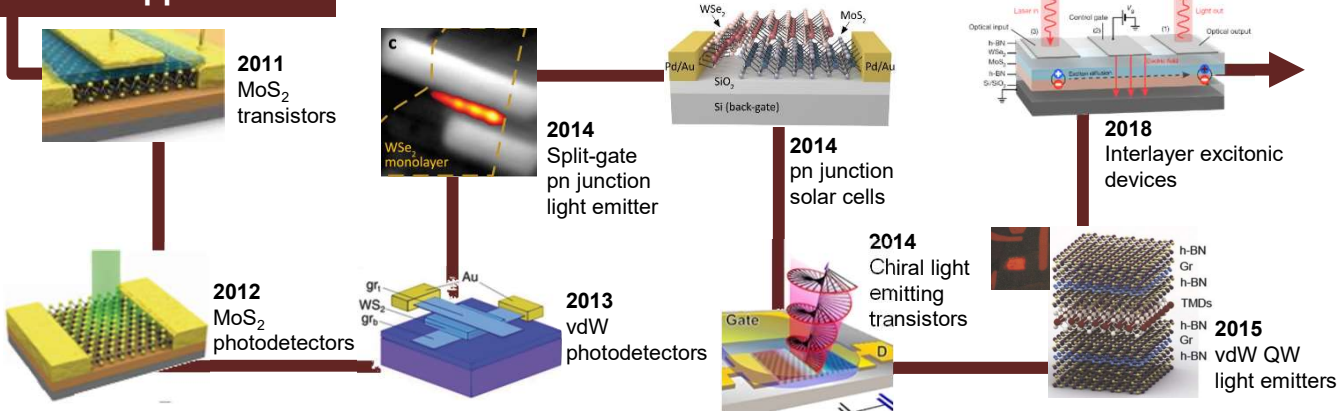
*Y.W. Son et al. Science (2018)*

# Excitons and Excitonic Devices

## Fundamentals



## Device Applications



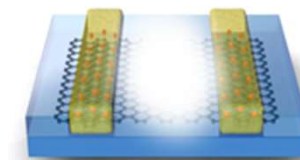
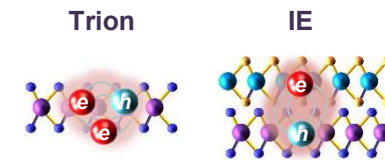
## Excitonic EL Devices

### 1. Issues in 2D LEDs

- Low efficiency
- Low temperature operation only
- Absence of gate tunability

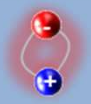
### 1. Control of exciton complex

- Trion and interlayer exciton(IE)

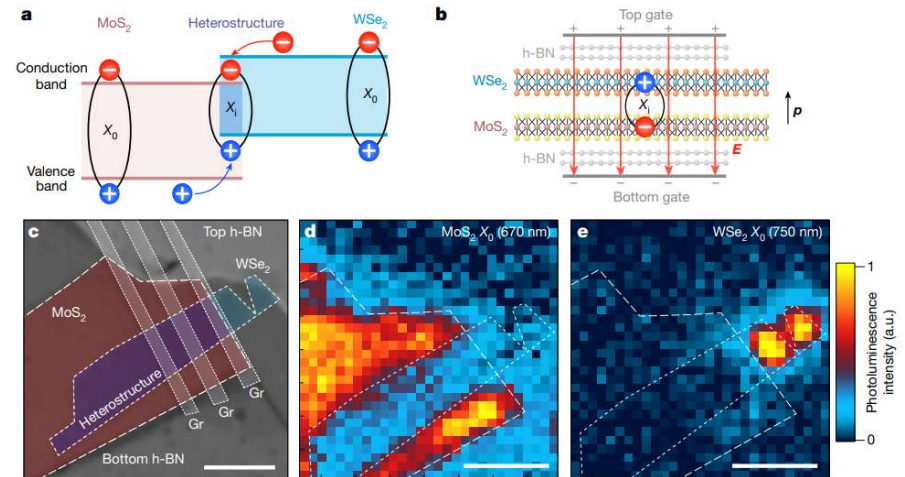




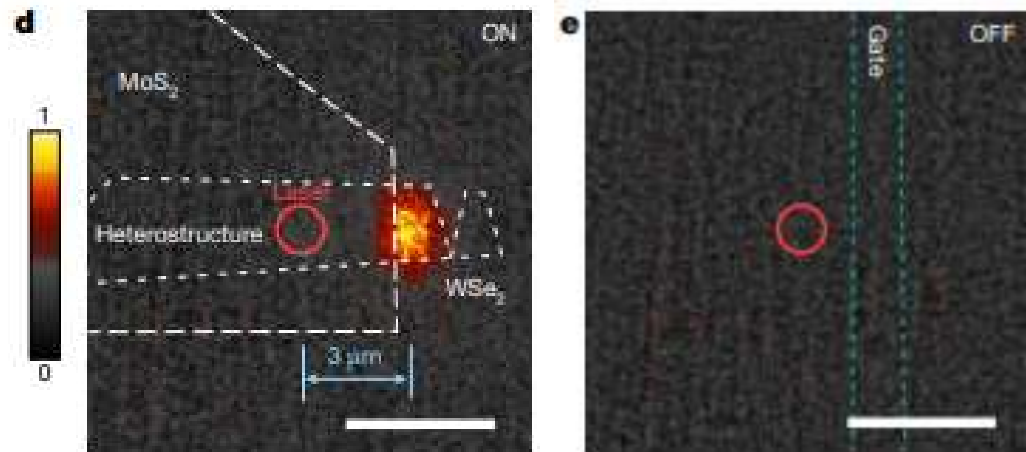
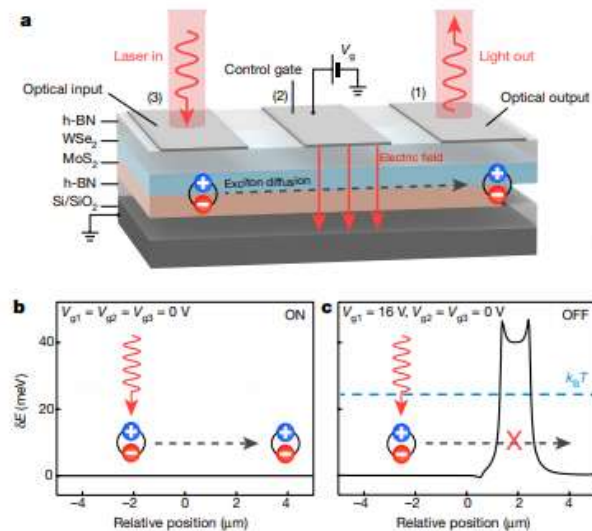
# Interlayer exciton

 <b>Interlayer exciton</b>	
<b>Charge</b>	<b>Neutral Charge</b>
<b>Transport mechanism</b>	<b>Diffusion</b>
<b>Control parameter</b>	<b>Exciton density</b>

## Interlayer excitons in the vdW heterostructure



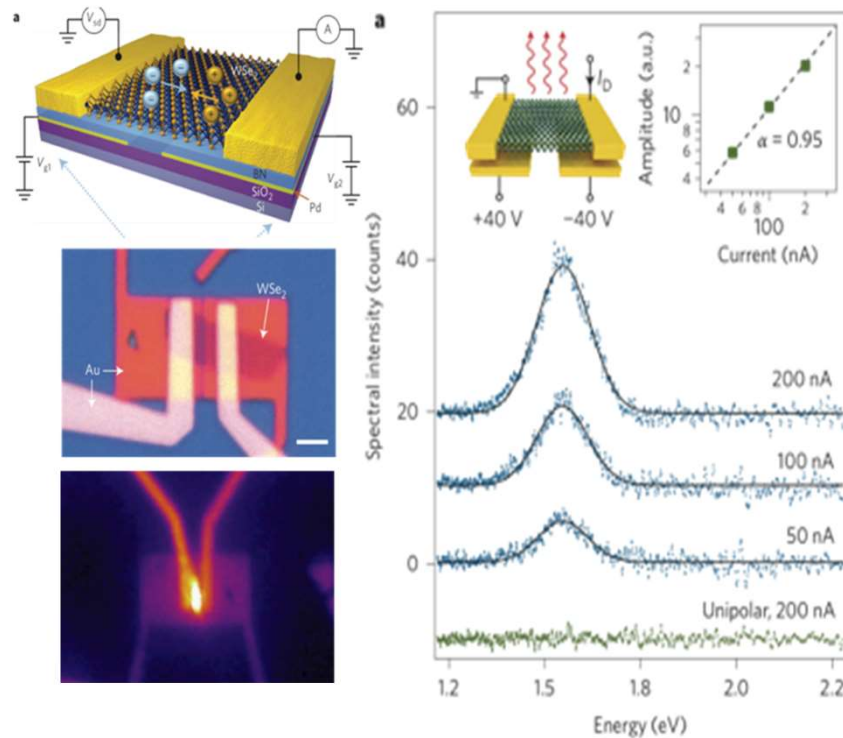
## Excitonic transistor operation by electric field



*A. Kis et al. Nature (2018)*

# Light Emitting Devices

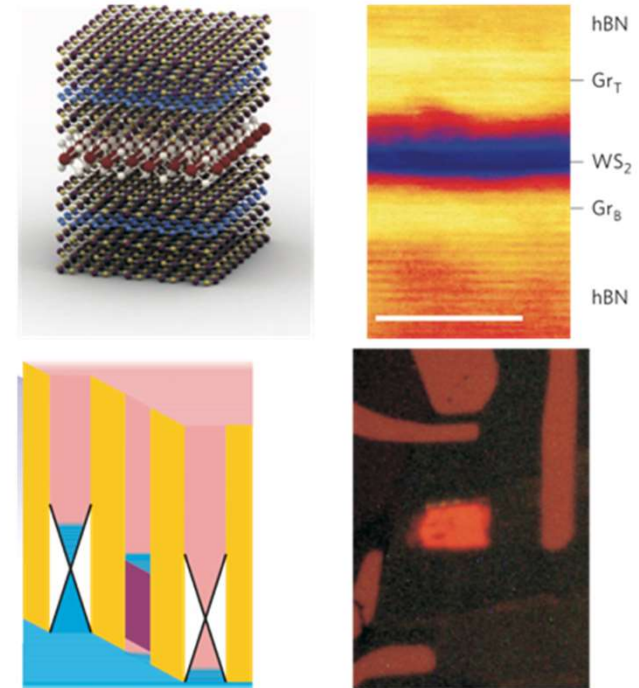
## TMDC Light Emitter



- Atomically thin LEDs (pn junction by split gate) from WSe<sub>2</sub>, MoS<sub>2</sub>, WS<sub>2</sub>
- EQE: ~ 0.2 % (limited by contact and thickness)

A. Pospischil et al. *Nature Nanotech.* 9, 257 (2015)  
 B. W. H. Baugher et al. *Nature Nanotech.* 9, 262 (2015)  
 J. S. Ross et al. *Nature Nanotech.* 9, 268 (2015)

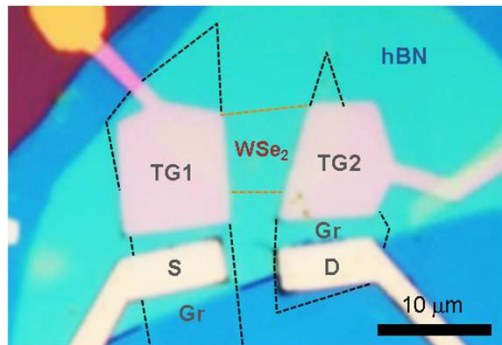
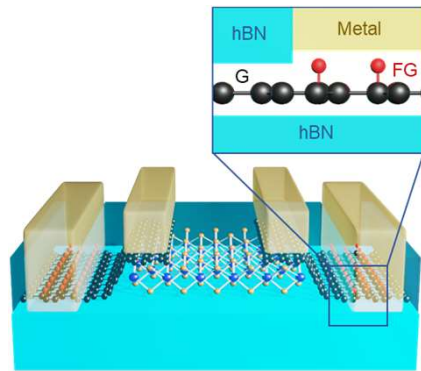
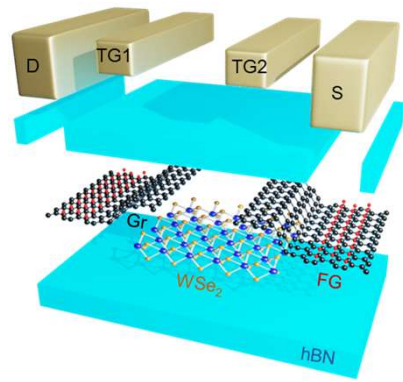
## van der Waals Light Emitter



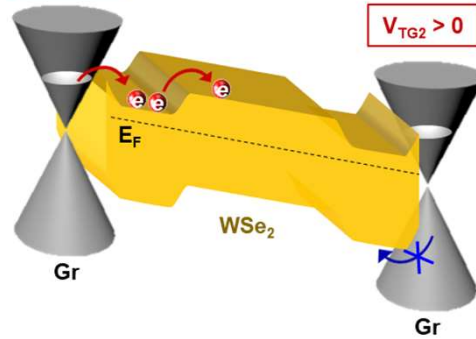
- Atomically thin quantum well with van der Waals heterostructure
- Efficient electron and hole injection by tunneling through hBN
- Flexible and transparent light emission devices
- EQE: 2.0 ~ 8.4 % (at low T)

F. Withers et al. *Nature Materials* 14, 301 (2015)

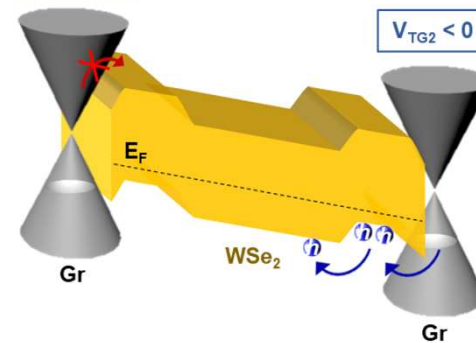
# Monolayer WSe<sub>2</sub> LETs with Tunable Schottky Barrier



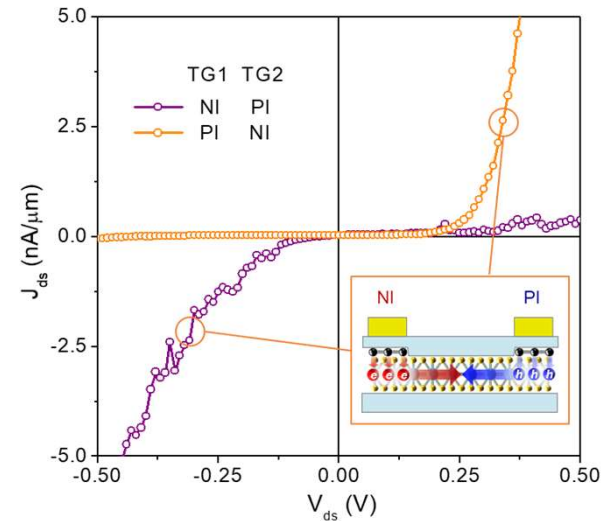
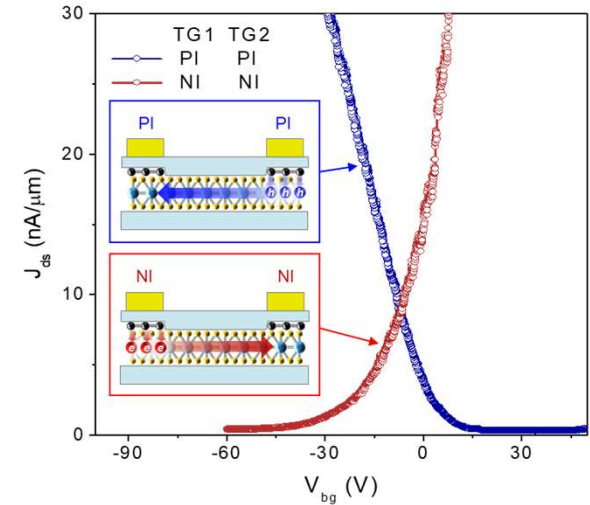
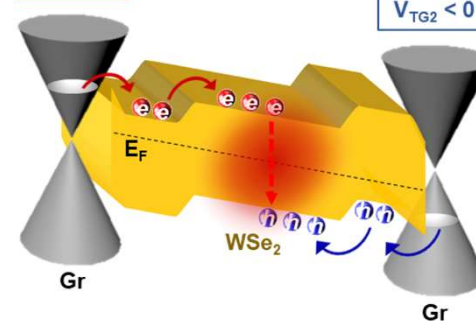
$V_{TG1} > 0$  NI (negative charge injection)



$V_{TG1} < 0$  PI (positive charge injection)



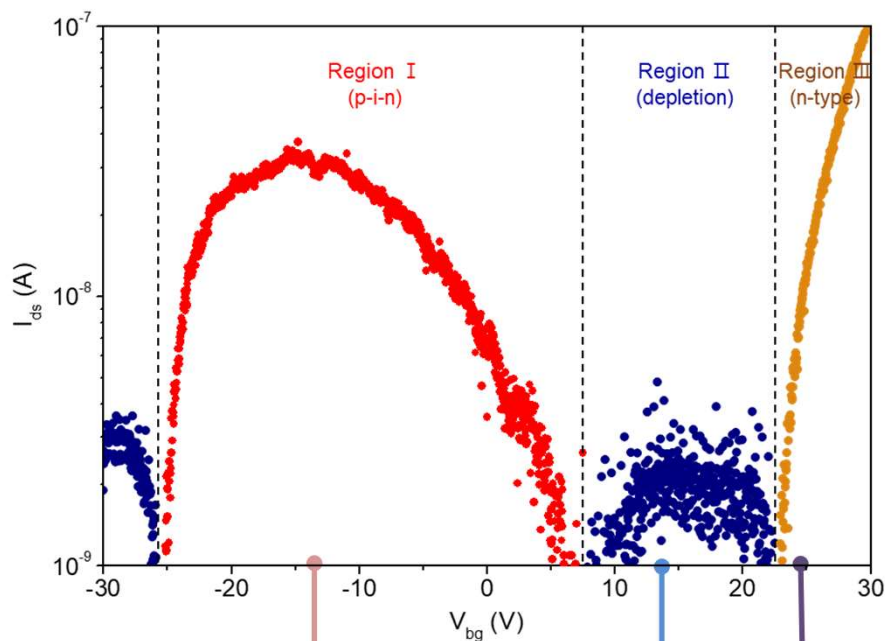
$V_{TG1} > 0$  NI-PI



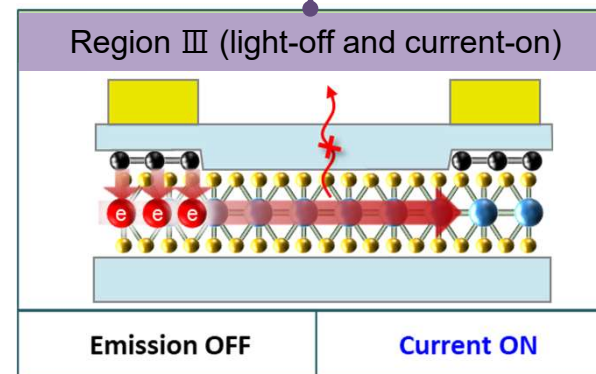
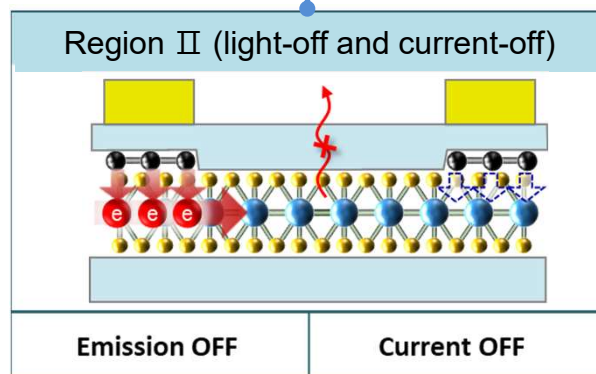
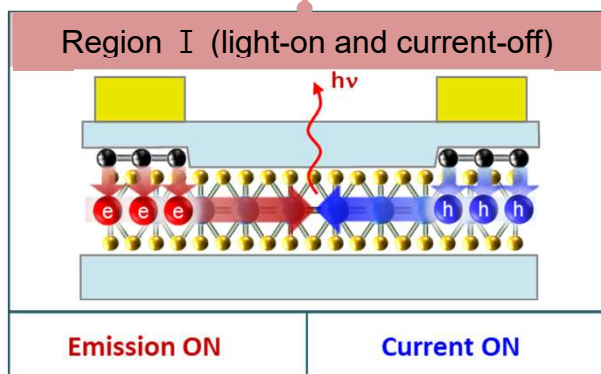
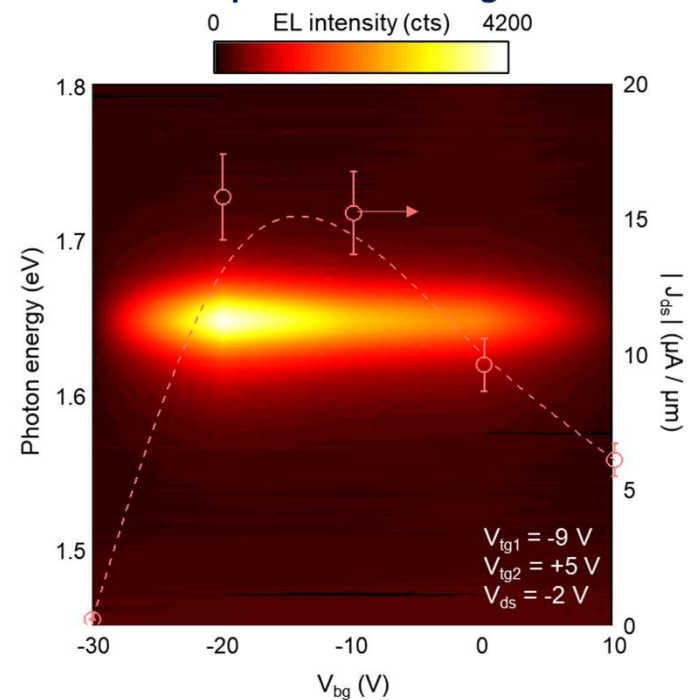


# Multi-operation Modes of WSe<sub>2</sub> LETs

Gate-dependence in transfer curve

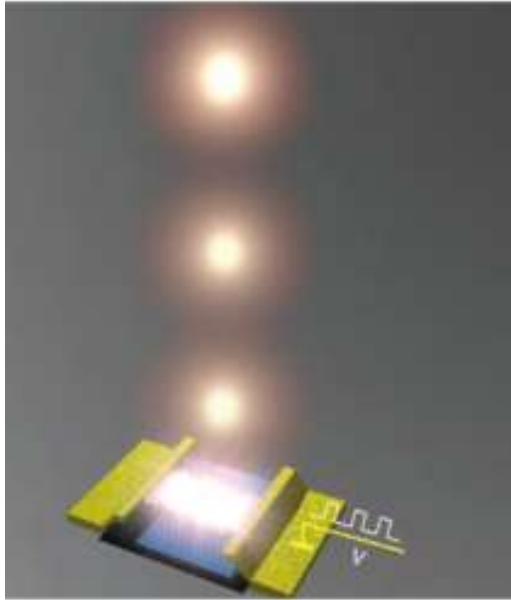


Gate-dependent EL in region I

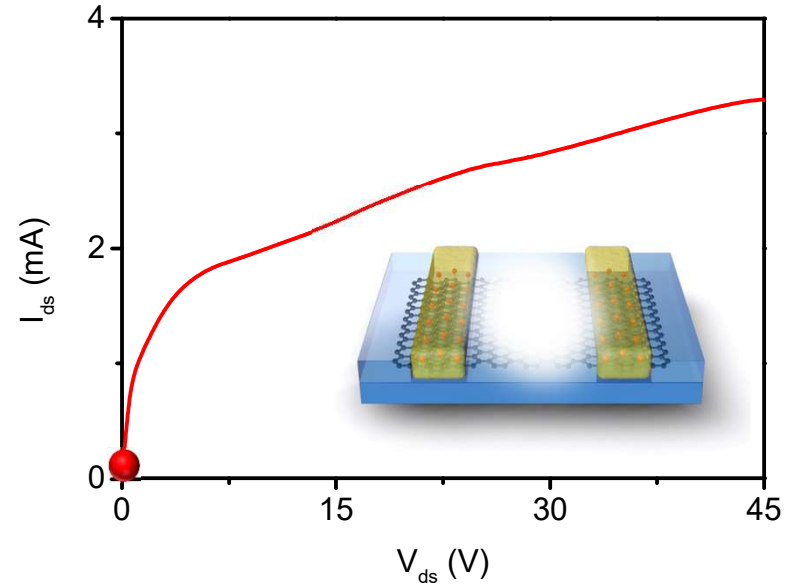


# Light Emitting Devices

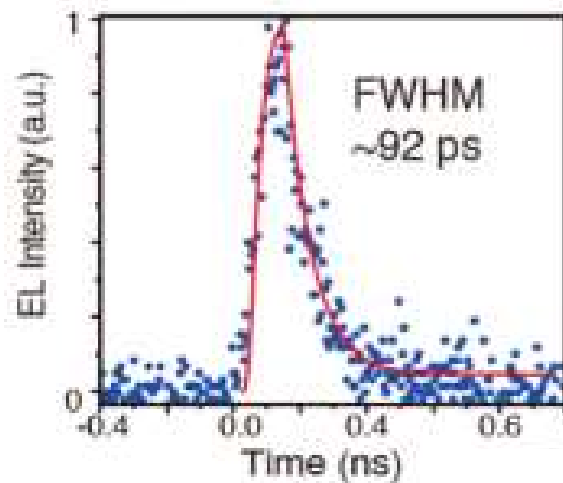
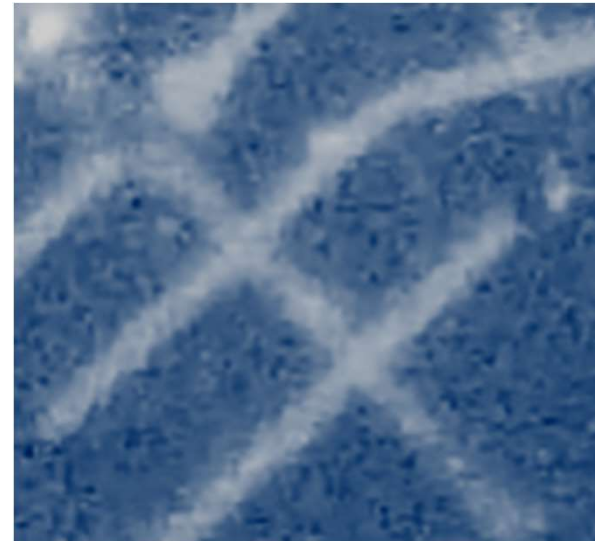
## Graphene light emitting device



## High breakdown current



## Light emission from graphene

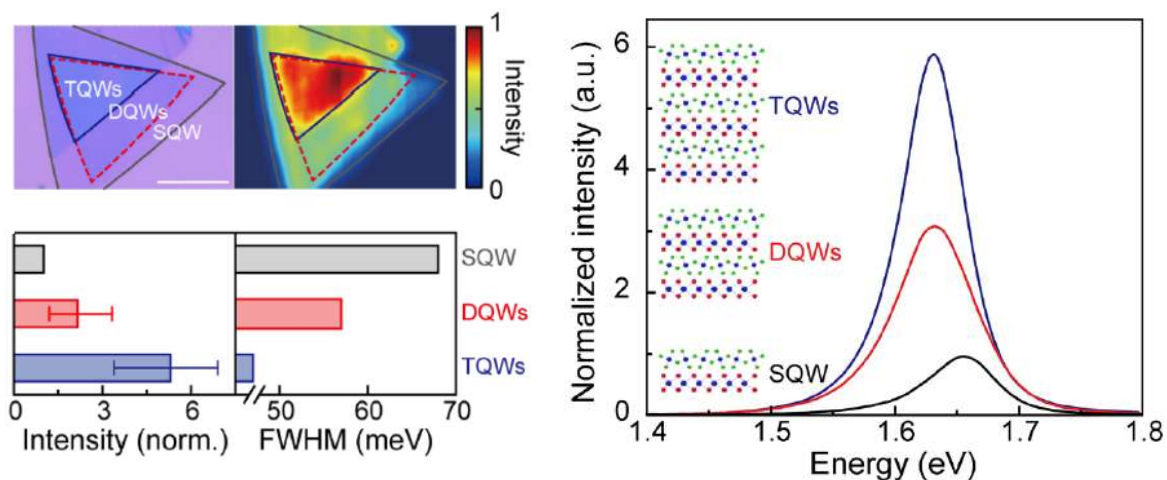
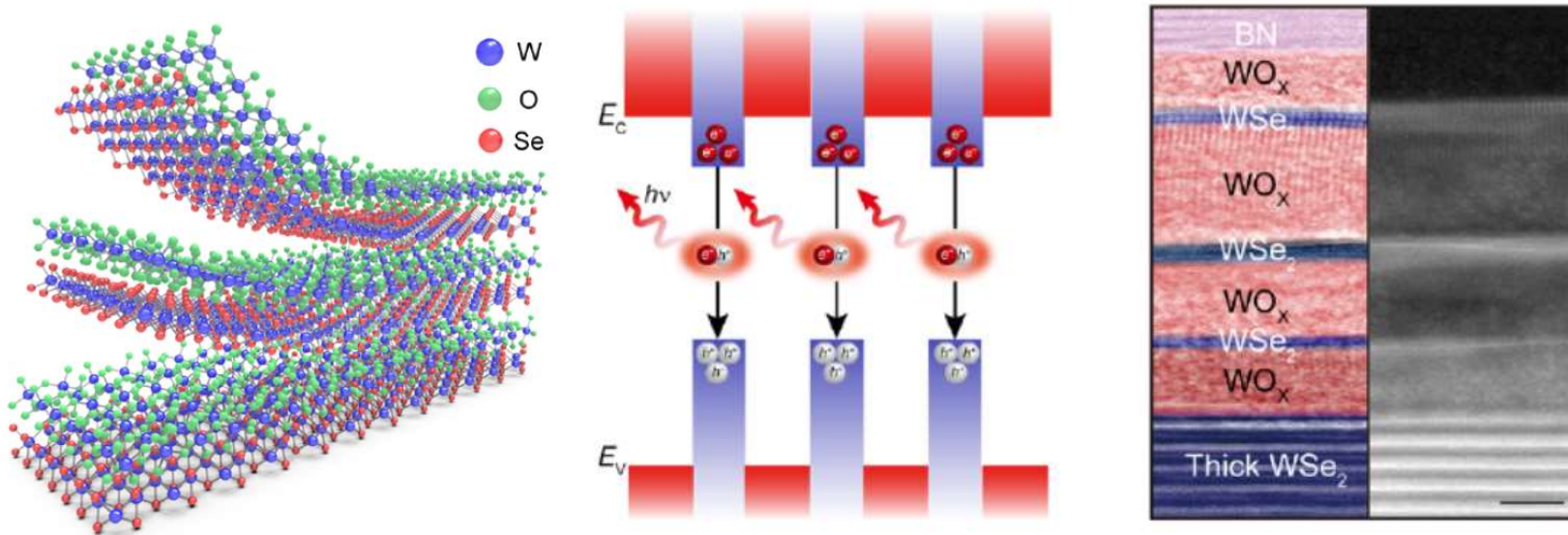


*J. Hone. et al. Nano Lett. (2018)*



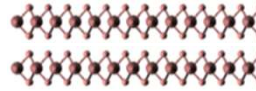
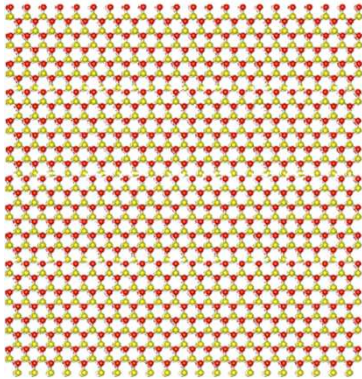
# Multiple Quantum Well of TMDs

Fabrication process of  $\text{WO}_x/\text{WSe}_2/\text{WO}_x/\text{WSe}_2$  by Layer-by-layer oxidation

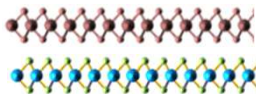
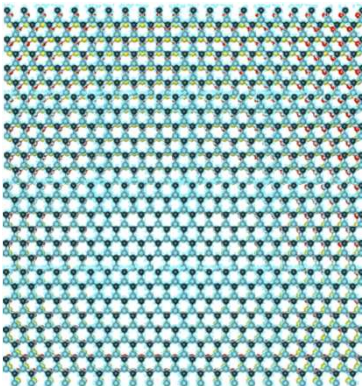


# Twist Angle of Stacked 2D Layers

**MoS<sub>2</sub>/MoS<sub>2</sub>**



**MoS<sub>2</sub>/WSe<sub>2</sub>**



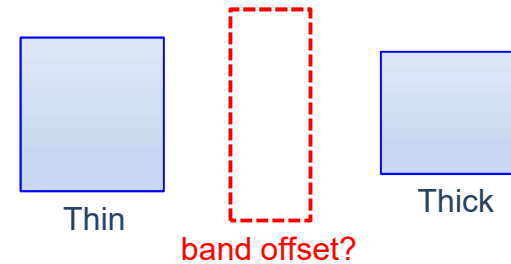
**Lateral junction**



**Vertical junction**



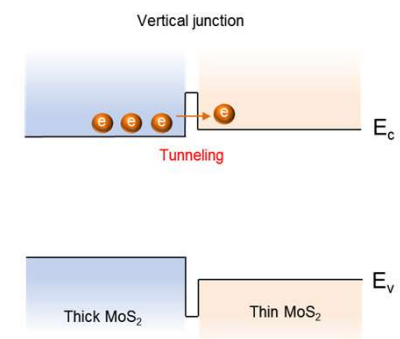
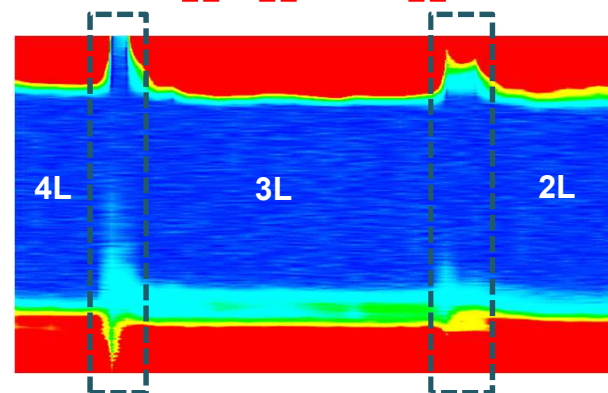
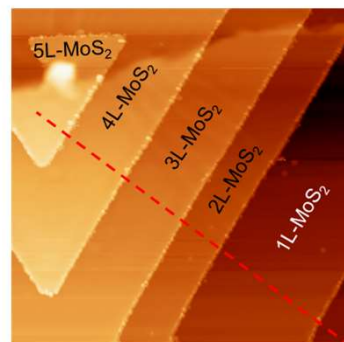
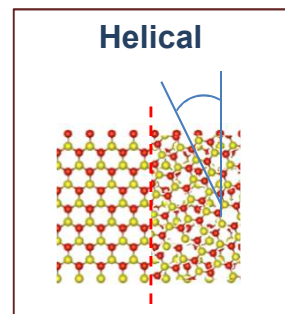
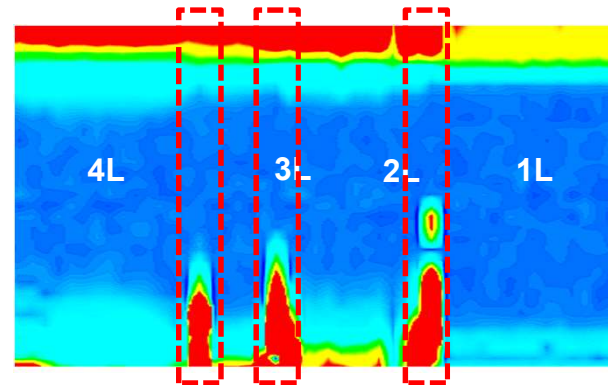
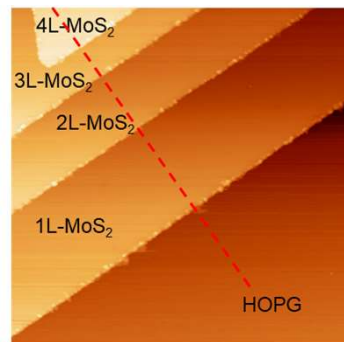
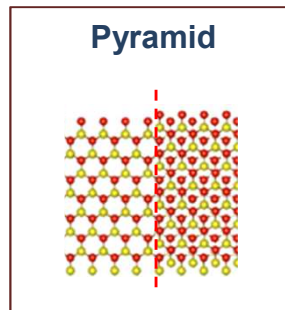
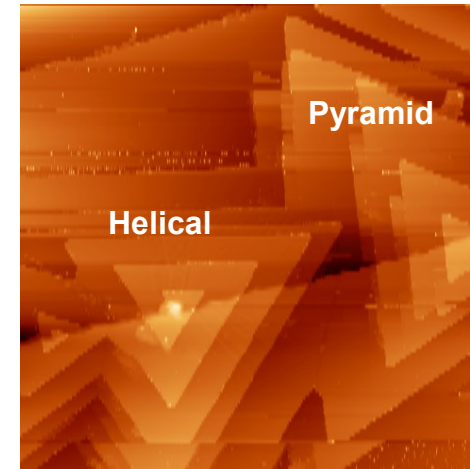
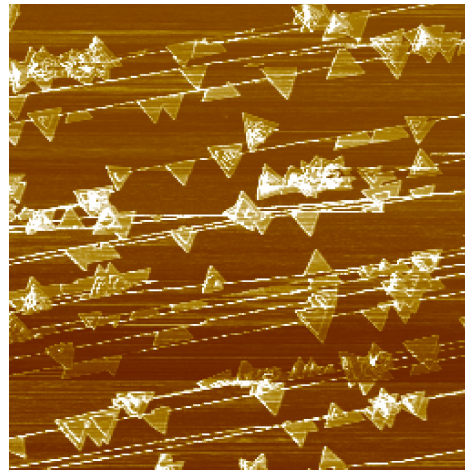
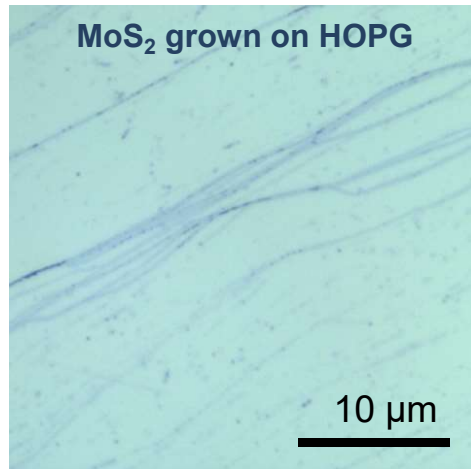
**Junction band structure**



## Charge transfer at interface of stacked 2D layers

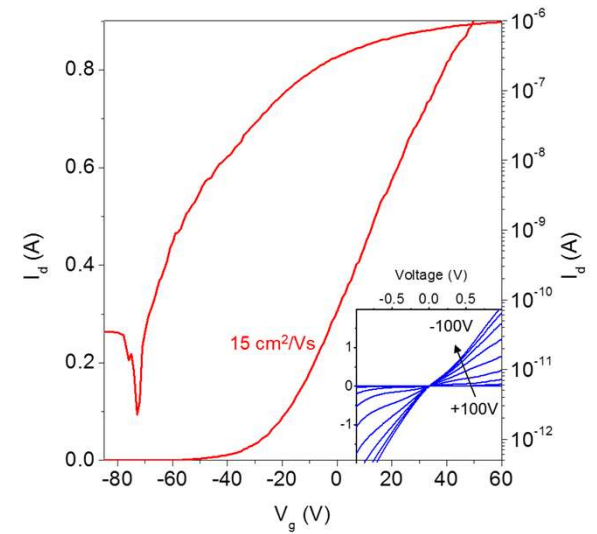
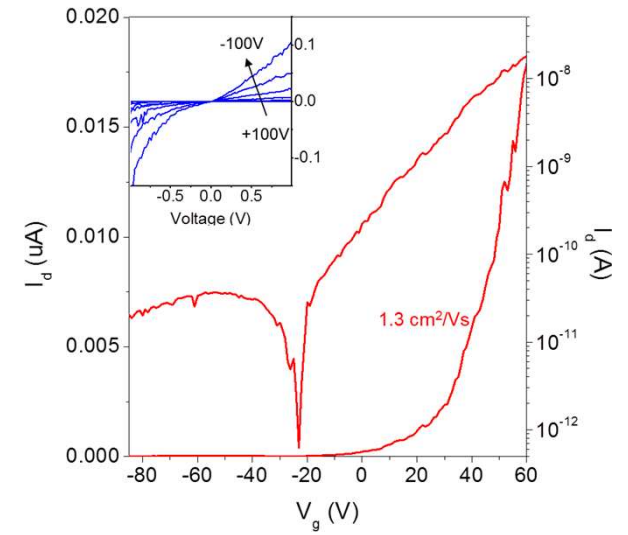
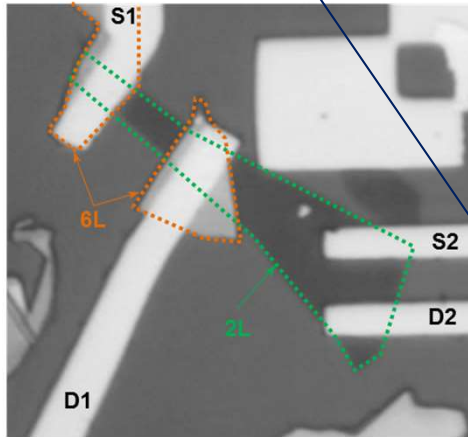
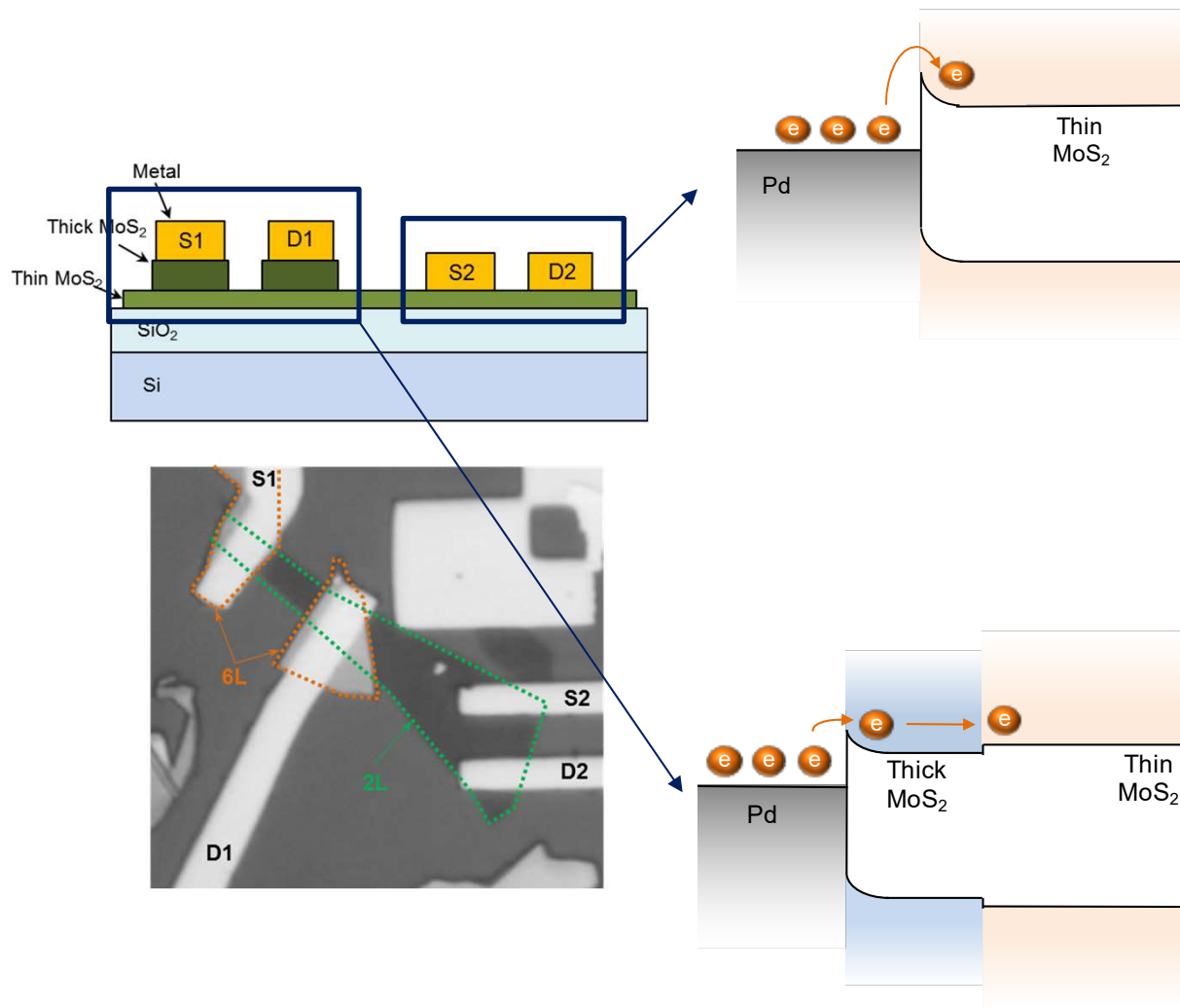
- Electrical properties are influenced by stacking angle?
- The stacked layers are coupled or decoupled?
- Can we modify interfacial properties and band structure with twist angle and stacking structure?

# Band Offset at Heterointerface



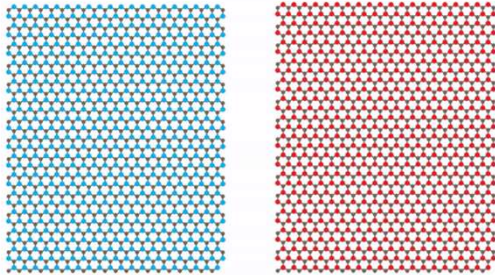


# Hetero-structured Contacts with Low Resistance



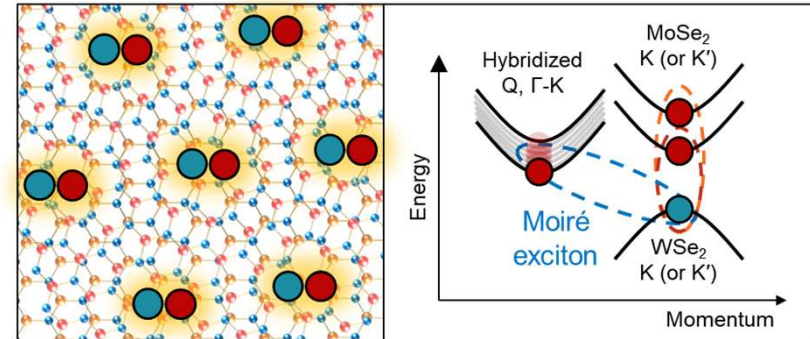
# Moiré Crystals

## Moiré superlattice

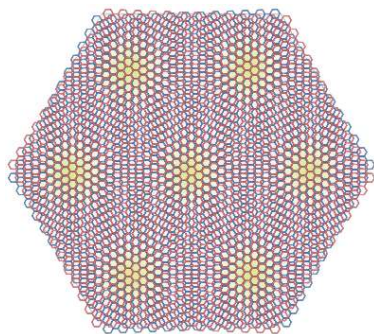


Stack together two monolayer semiconductors.

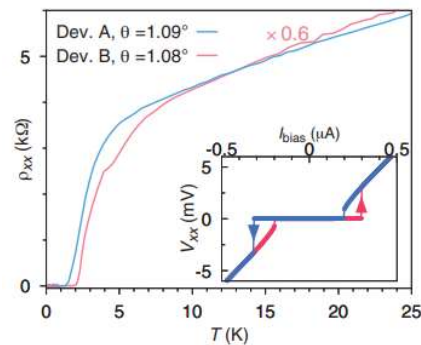
## Moiré potential trapped interlayer exciton



## Superconductivity in magic angle graphene

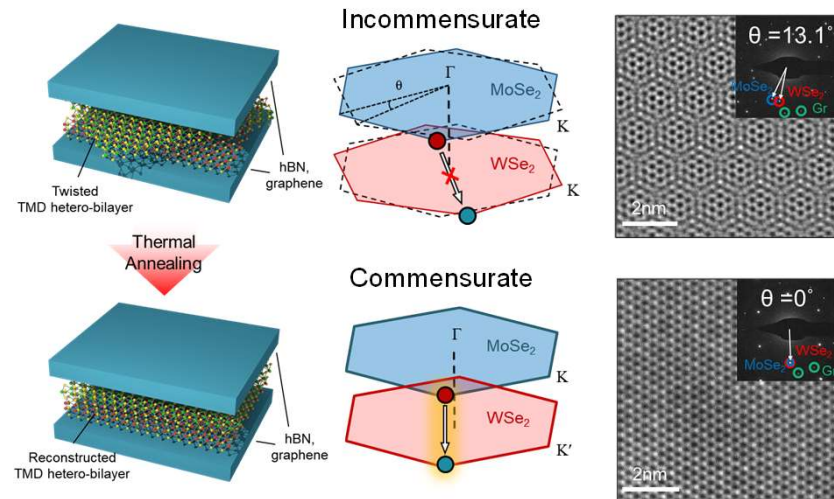


$\theta = 1.09^\circ$



Science (2018)

## Twist angle control in TMDs heterobilayers



J. Baek, G.H. Lee *In preparation*

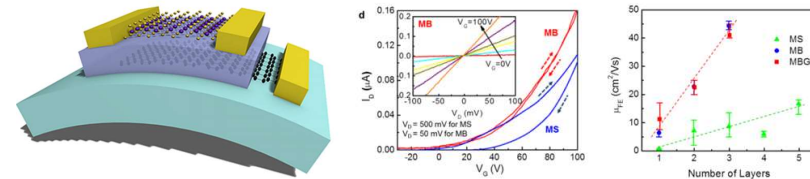
- Moiré potential trapped charge carriers contribute to novel quantum physics in 2D materials.
- Twisted bilayer graphene shows superconducting behavior at specific angle ( $\theta \approx 1.1^\circ$ ).

- Interlayer excitons are laterally confined from the moiré potential
- Atomic reconstruction occurred in twisted TMDs heterobilayer by thermal annealing.

# Type of van der Waals heterostructure

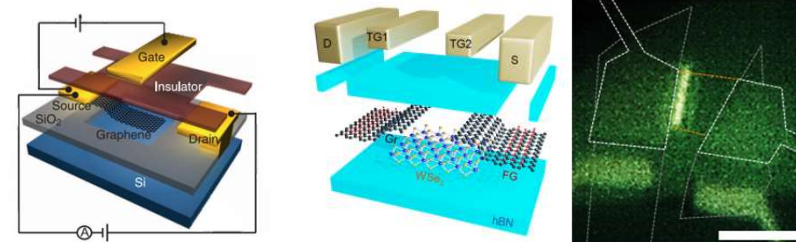
## 1. Heterostructure of Graphene-hBN / TMD-hBN

- High mobility
- No hysteresis



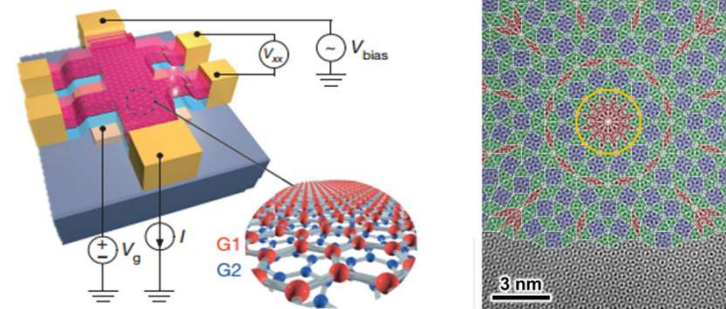
## 2. Heterostructure of Graphene-TMD-hBN

- Tunable Schottky barrier
- Fast charge transfer
- Barristor / LET



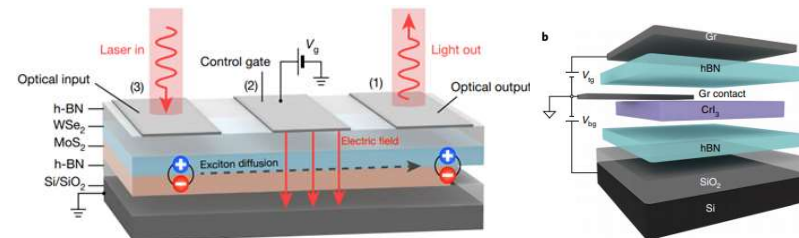
## 3. Heterostructure of Graphene – Graphene

- Twisted bilayer graphene
- Quasi crystal



## 4. Heterostructure of TMD-TMD

- Band engineering
- PN junction
- Interlayer exciton



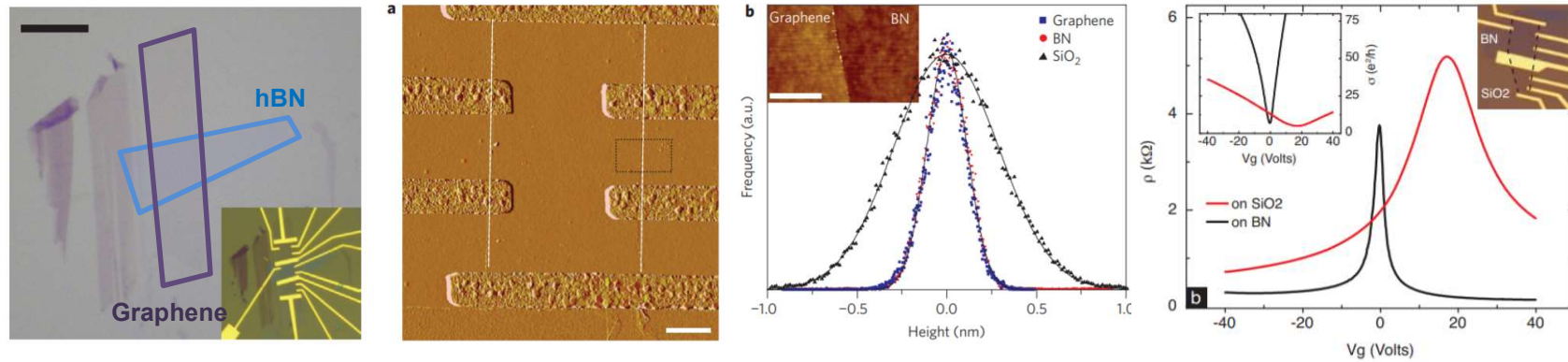
## 5. Heterostructure of various 2D material

- 2D magnetic : spin
- 2D ferroelectric : NC transistor



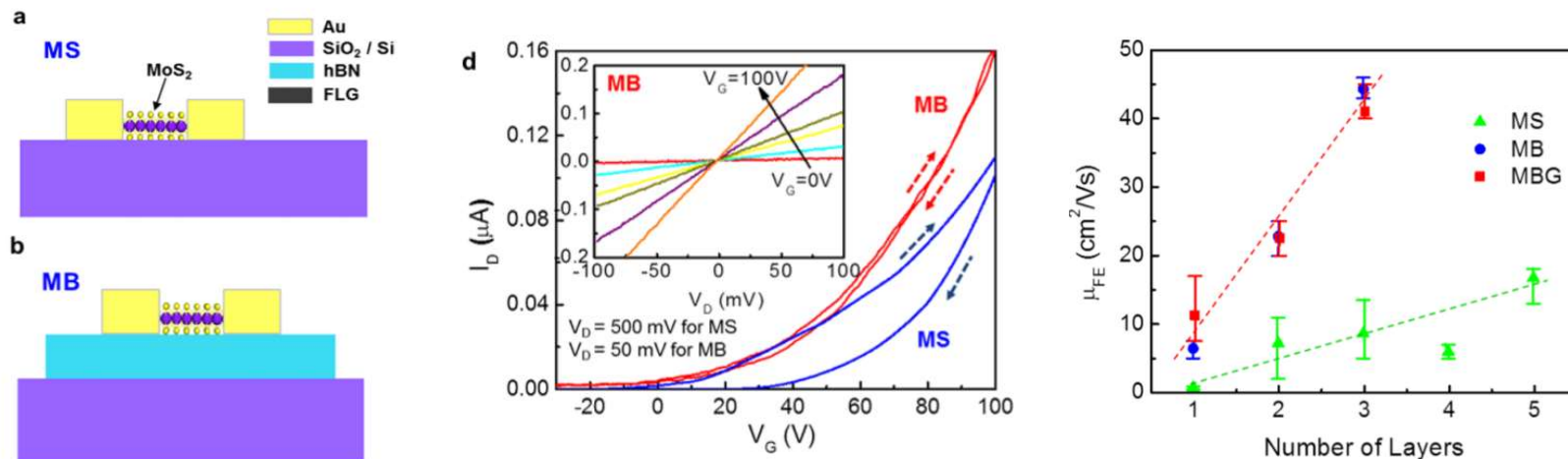
# Heterostructure of Graphene-hBN / TMD-hBN

## 1-1. Heterostructure of Graphene-hBN



*J. Hone et al. Nat. Nanotechnol (2010)*

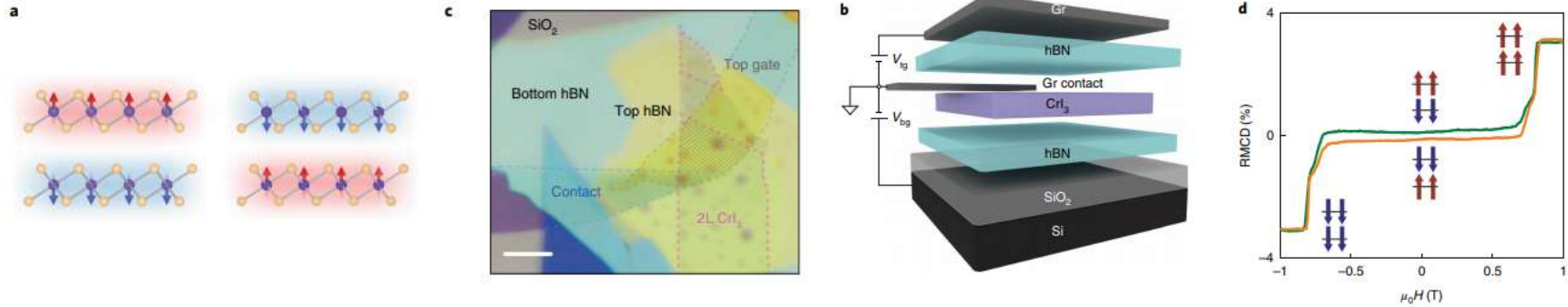
## 1-2. Heterostructure of TMD-hBN



*G.H. Lee, J. Hone et al Nat. Nanotech (2013)*

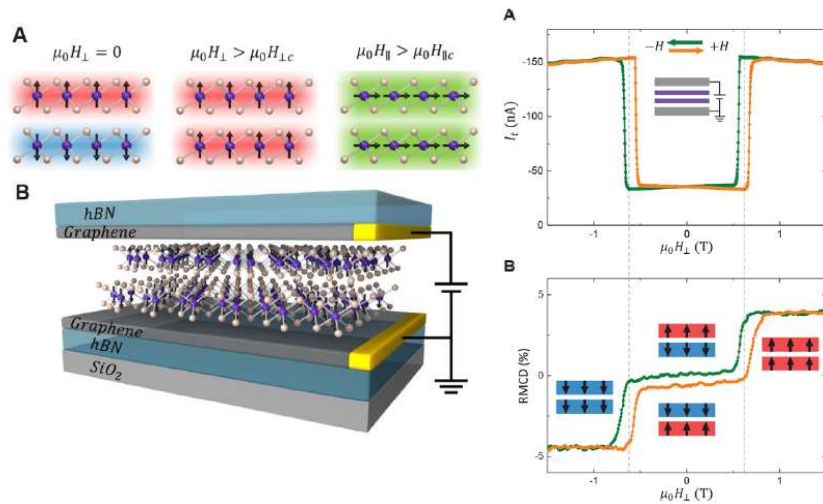
# Heterostructure of magnetic material

## 2D magnetic material CrI3



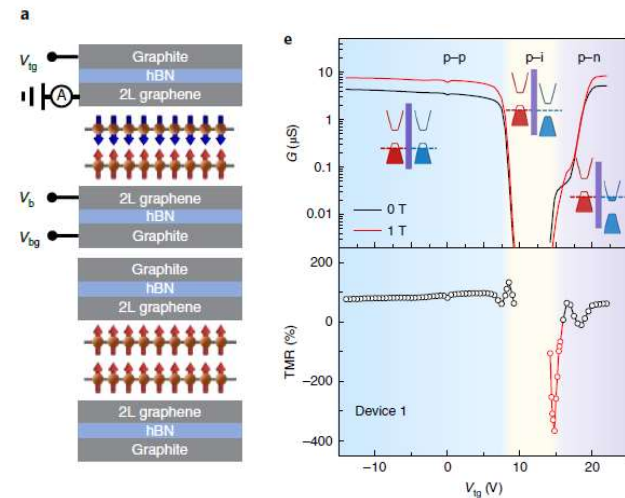
X. Xu et al. Nat. Nanotechnol. (2018)

## Giant tunneling magnetoresistance in spin filter



X. Xu et al. Science (2018)

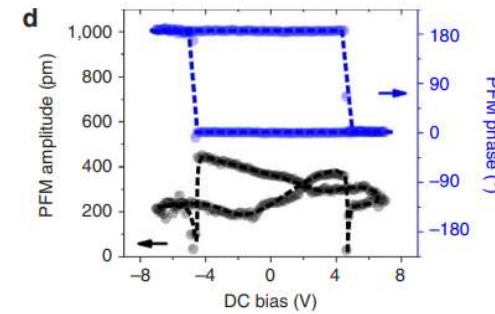
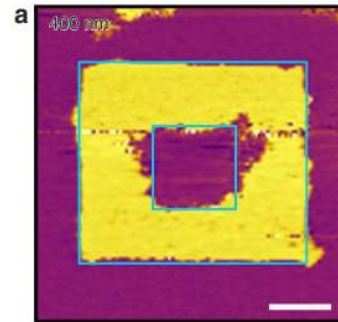
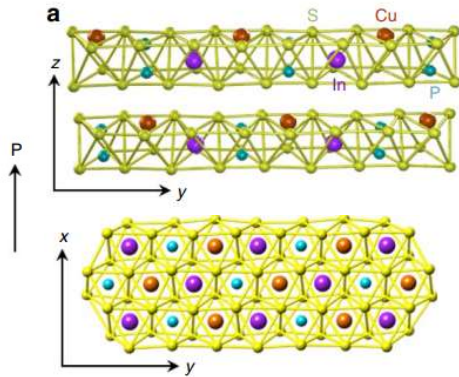
## Spin tunnel field effect transistor



K.F. Mak et al. Nat. Electron. (2019)

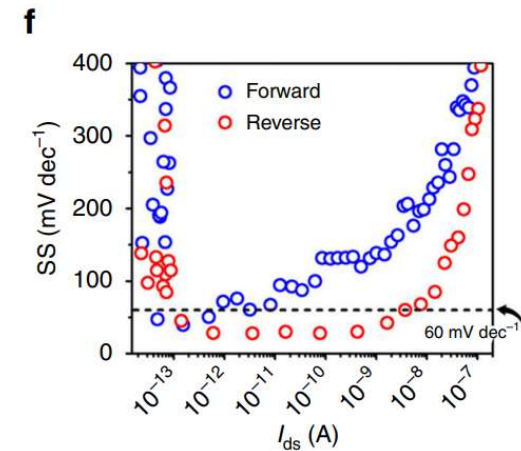
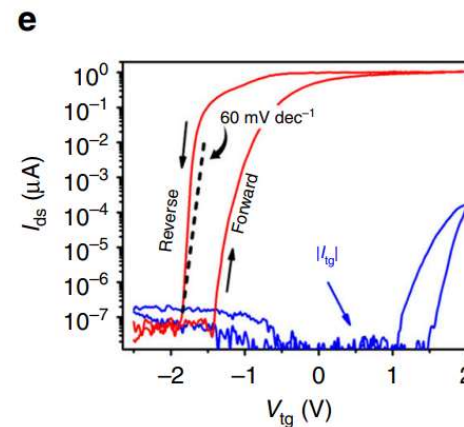
# Heterostructure of ferroelectricity material

## 2D ferroelectric material $\text{CuInP}_2\text{S}_6$



Z. Liu et al. Nat. Commun. (2016)

## Negative capacitance transistor



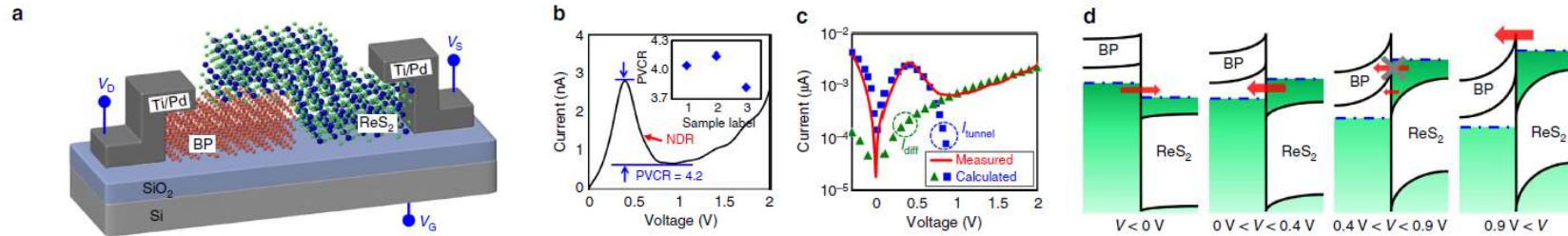
- vdW NC transistor has  $28 \text{ mV dec}^{-1}$  subthreshold swing(SS) and can overcome theoretically thermionic limit of  $60 \text{ mV dec}^{-1}$

Z. Liu et al. Nat. Commun. (2019)

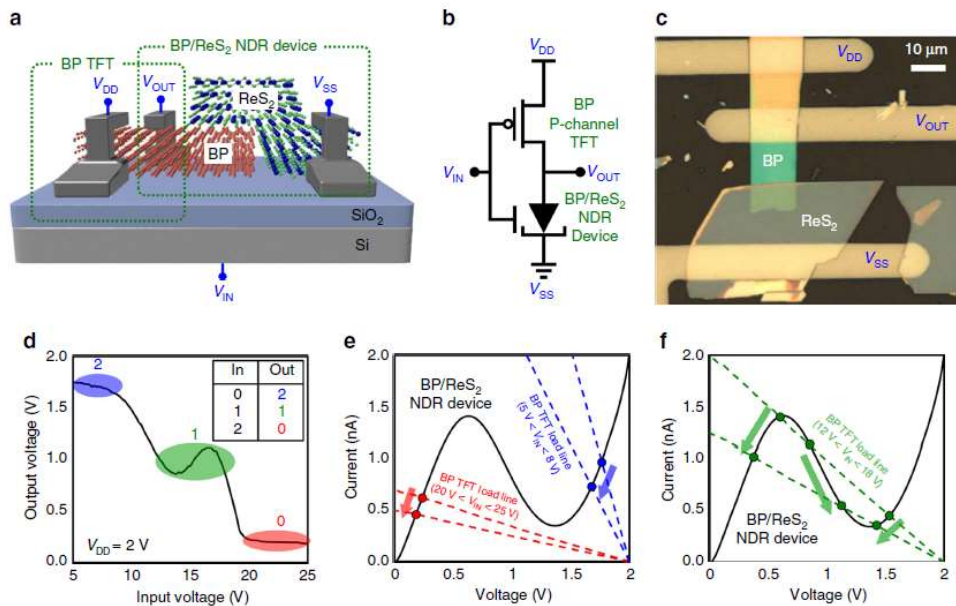


# Negative differential resistance device

## NDR device



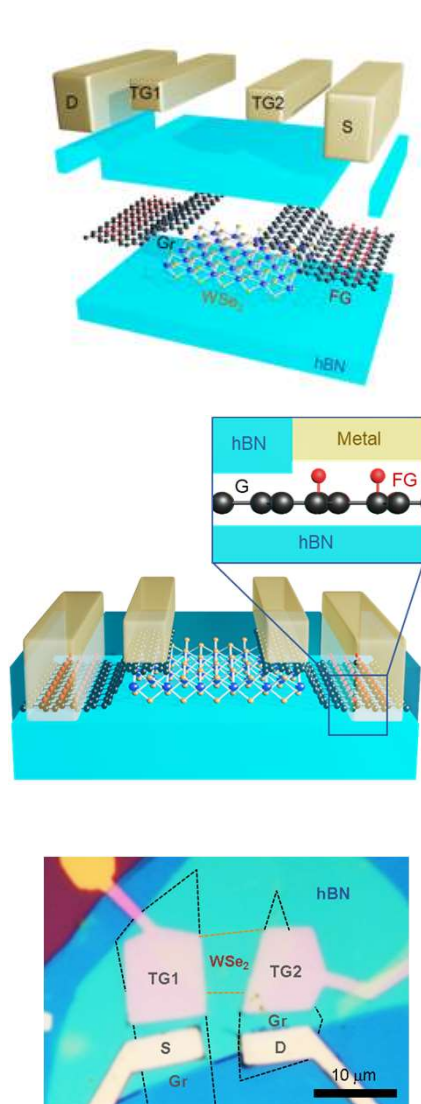
## Ternary inverter with three logical states



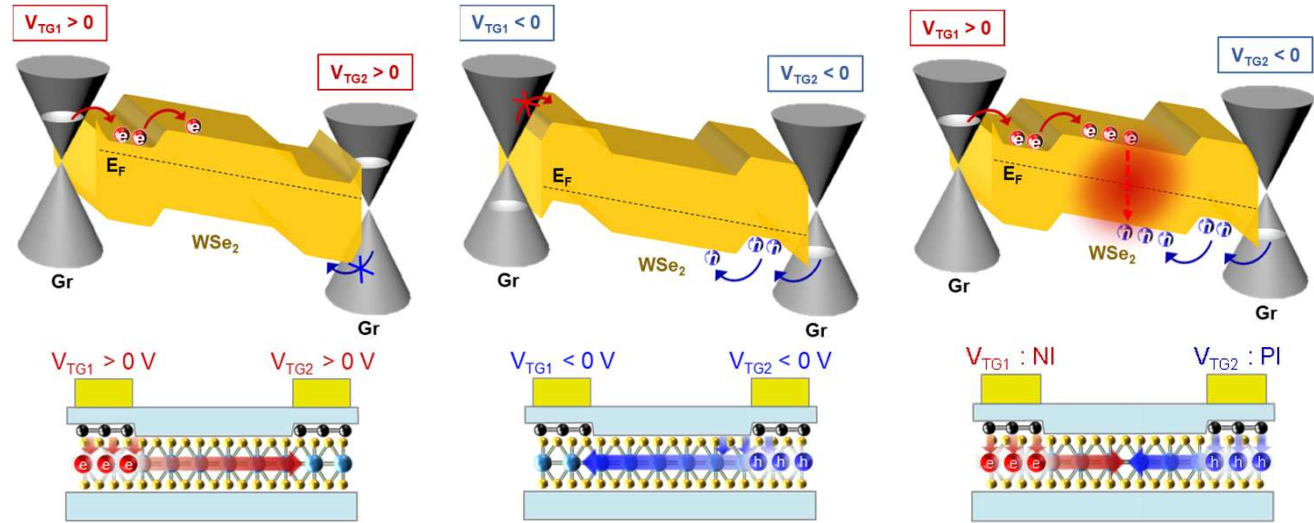
- NDR device using band engineering of vdW heterostructure
- NDR characteristic can be attributed to change types of band alignment from type III to type II.
- Multi-value level can be demonstrated, as ternary inverter fabricated using NDR characteristics

# Light emitting transistor with tunable Schottky barrier

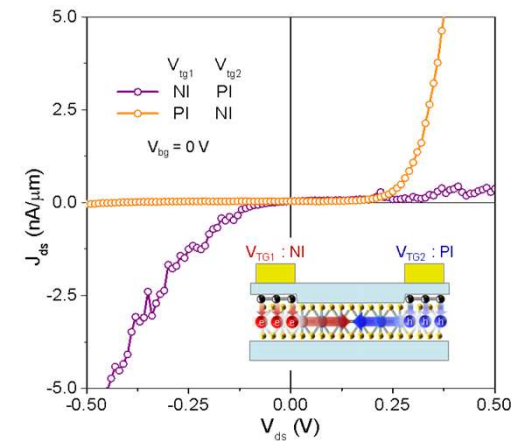
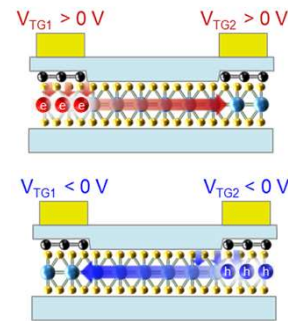
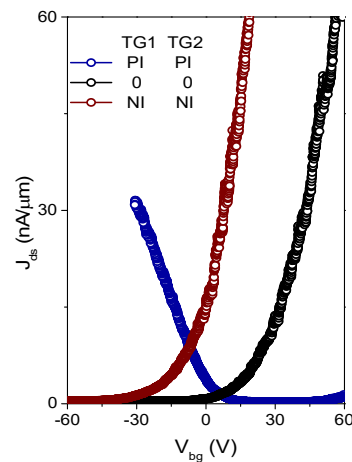
## Device geometry



## Working principle

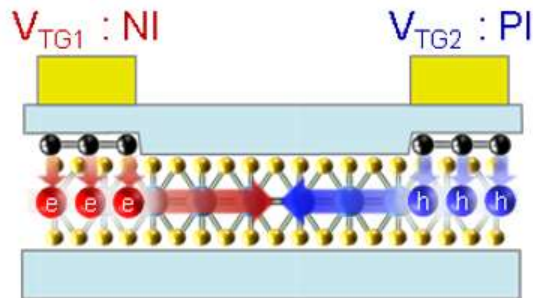
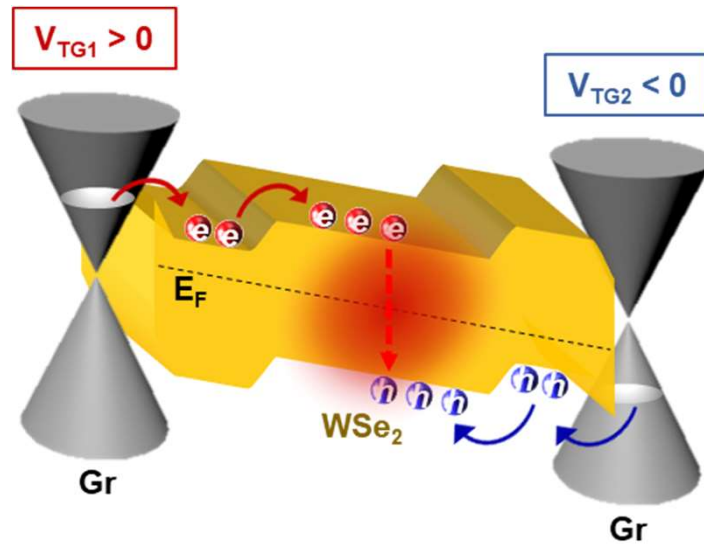


## Polarity Control

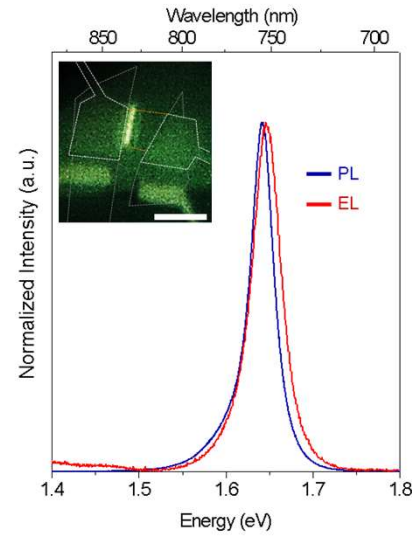


# Light emitting transistor with tunable Schottky barrier

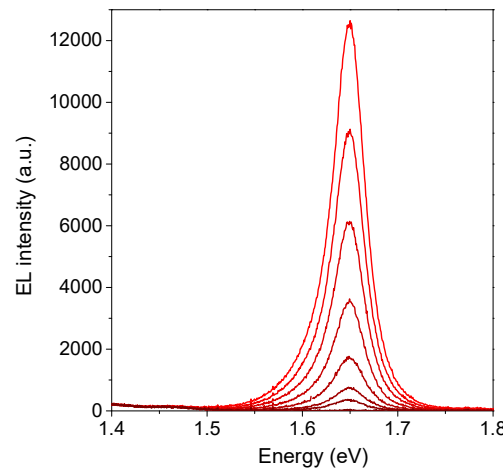
## Light emission



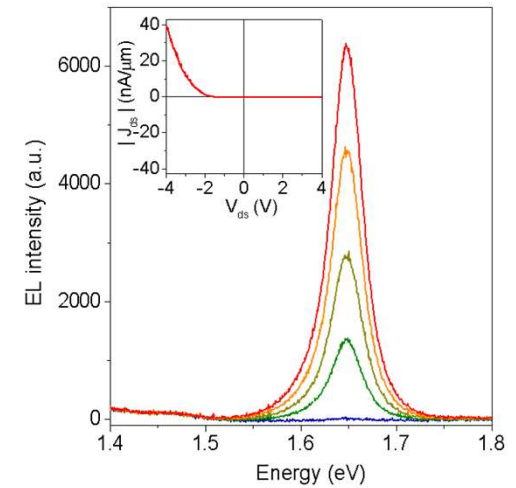
## EL vs PL



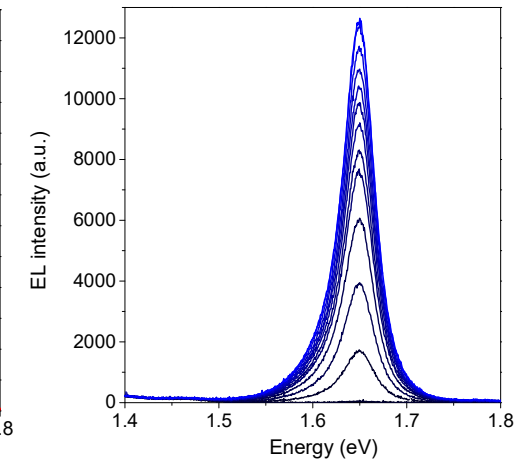
## Control of hole injection ( $V_{tg1}$ modulation)



## $V_{ds}$ dependence of EL



## Control of electron injection ( $V_{tg2}$ modulation)

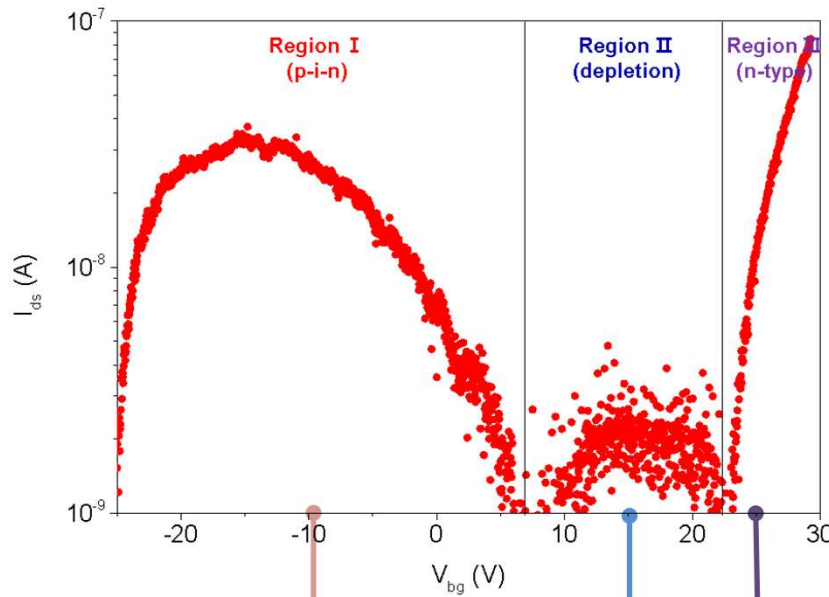




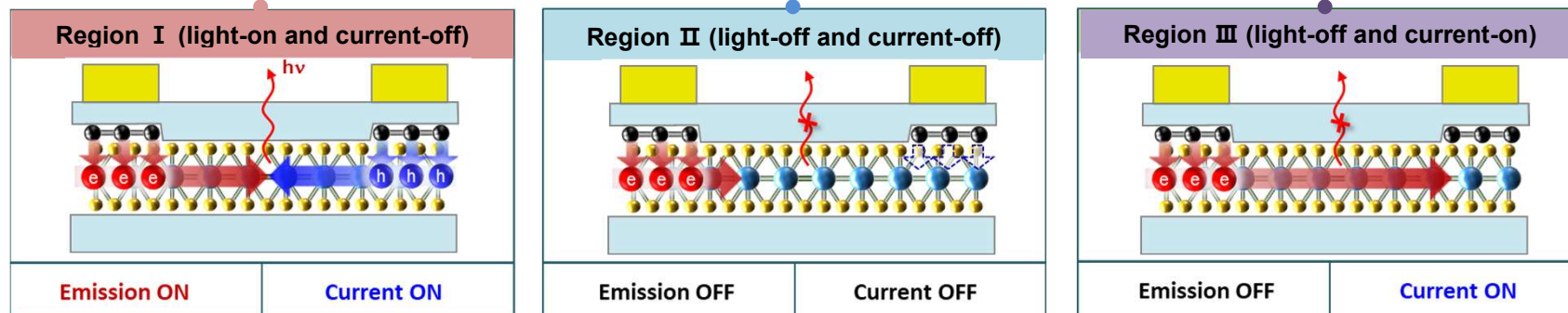
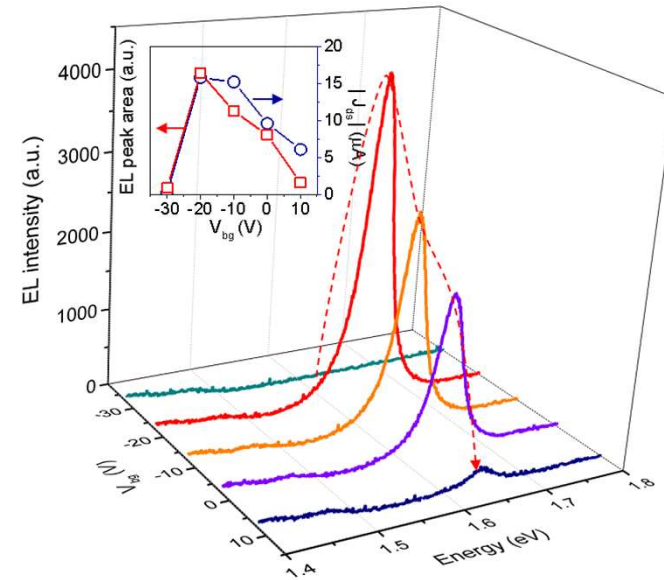
# Light emitting transistor with tunable Schottky barrier

## Multi-mode Operation

Gate-dependence in transfer curve

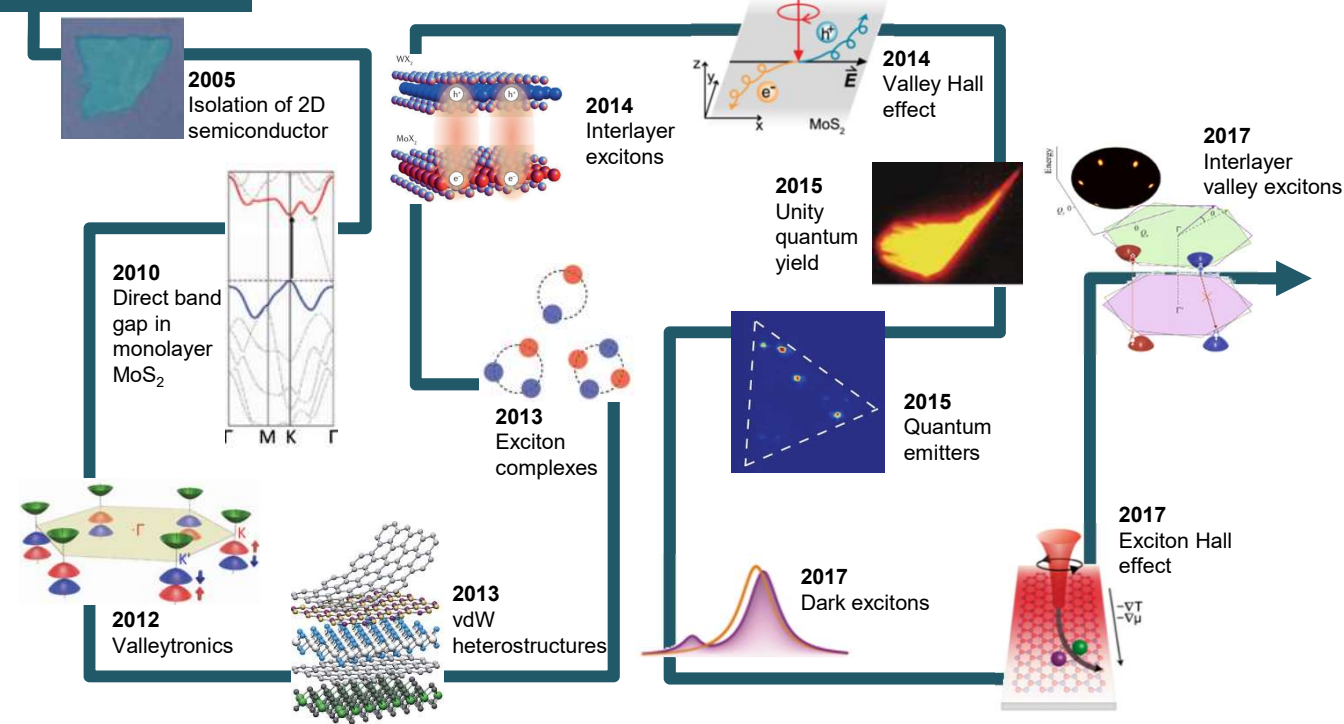


Gate-dependent EL in region I

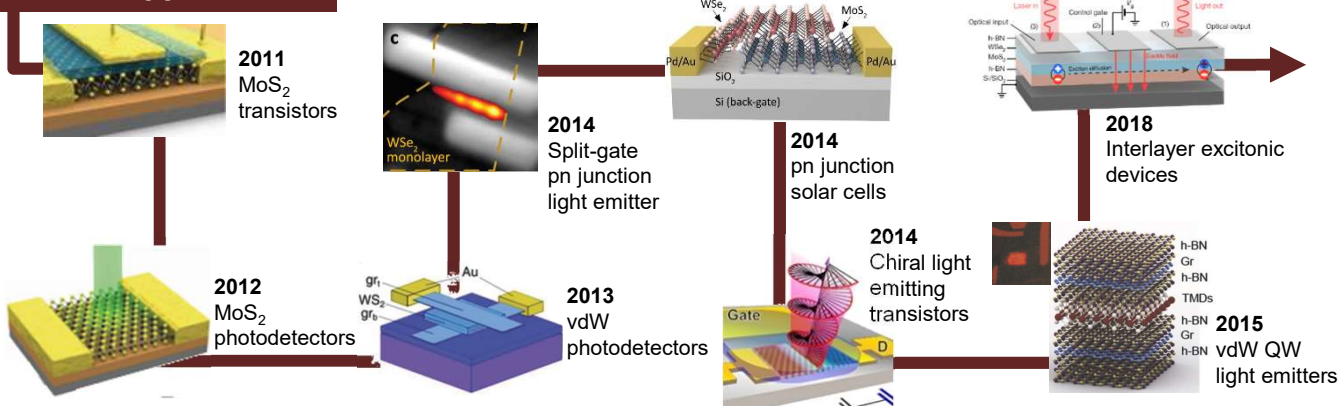


# Excitonic Light Emitting Devices

## Fundamentals

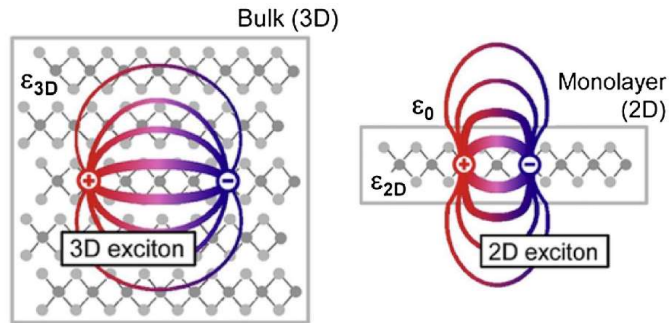


## Device Applications



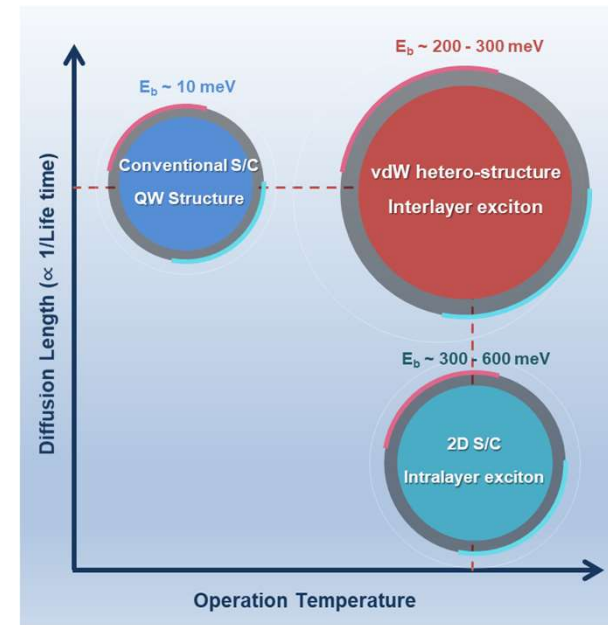
# Interlayer exciton

## Strong light-matter interaction of TMD

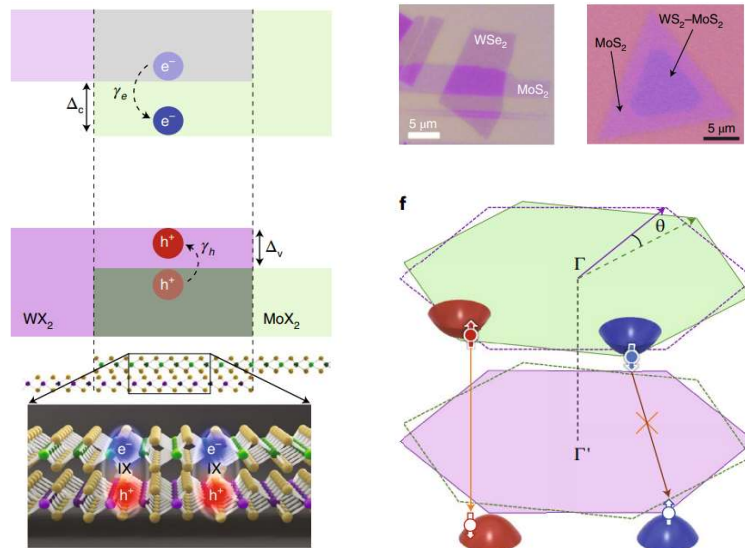


*P. S. Toth et al. Appl. Mater. Today (2017)*

## Possibility of interlayer excitation



## Interlayer excitation generation



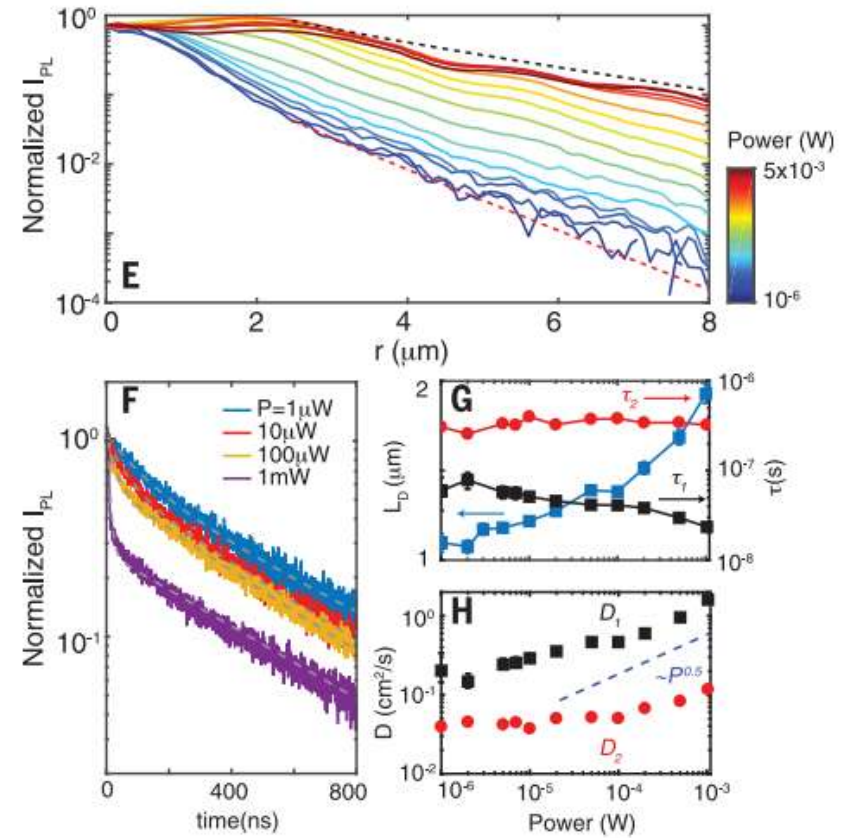
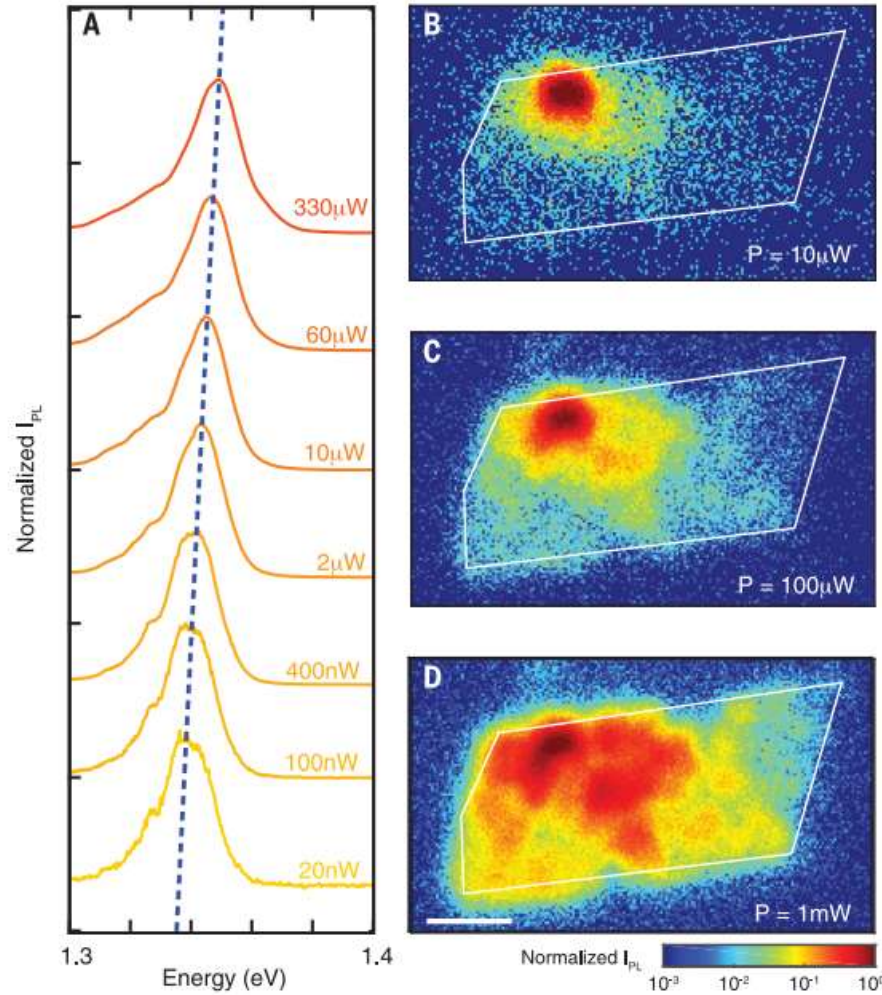
*X. Xu et al. Nat. Nanotechno. (2018)*

TMD	Binding energy	Life time
Conventional S/C	1~20meV	300ns
2D S/C	300~500meV	0.001~20ns
Vdw heterostructure	200~300meV	100ns



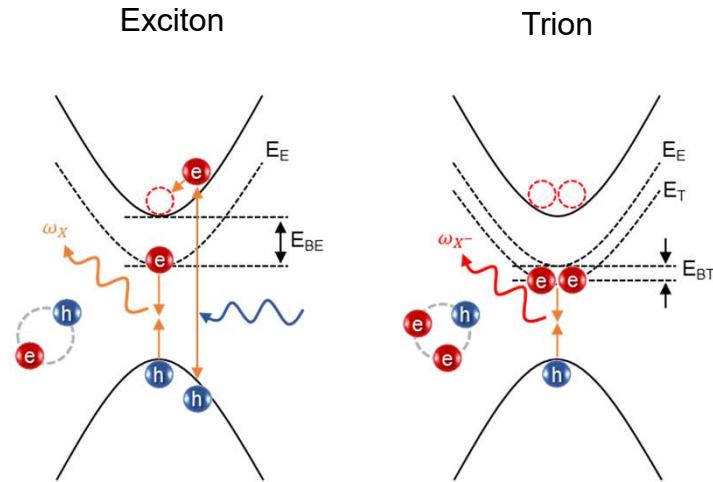
# Interlayer exciton

## Diffusion of Interlayer exciton



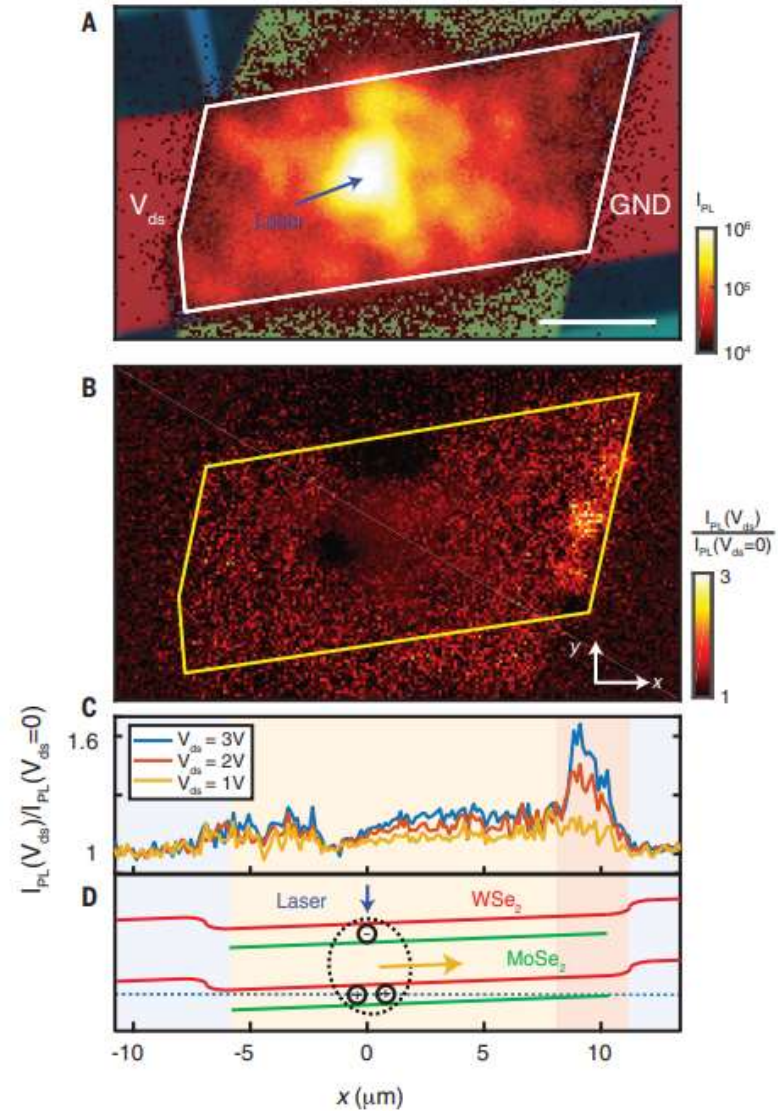
# Interlayer trion

## Exciton vs Trion



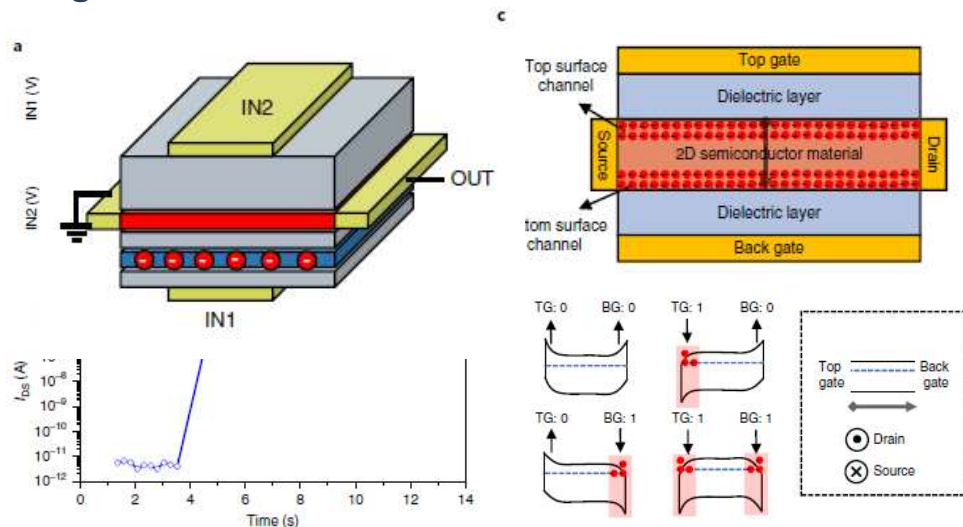
Interlayer trion	
Charge	Positive Charge Negative Charge
Transport mechanism	Drift (+Diffusion)
Control parameter	Electric field

## Diffusion of Interlayer trion

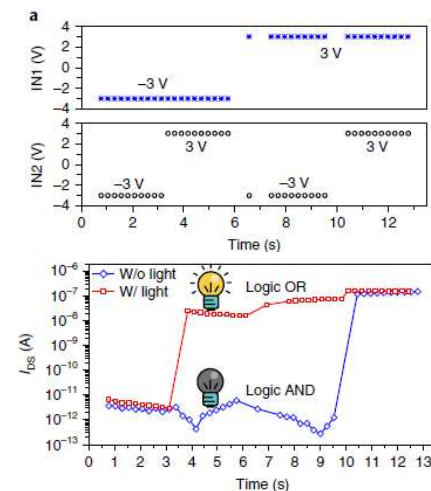


# Photoswitching logic and memory

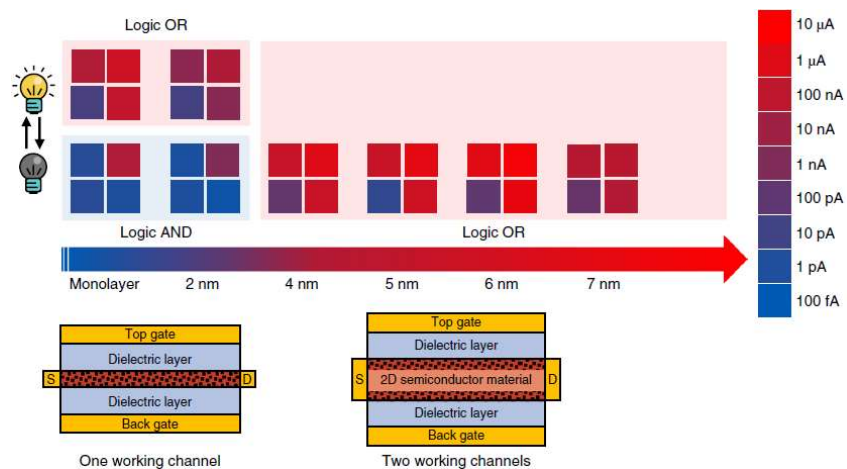
## OR gate



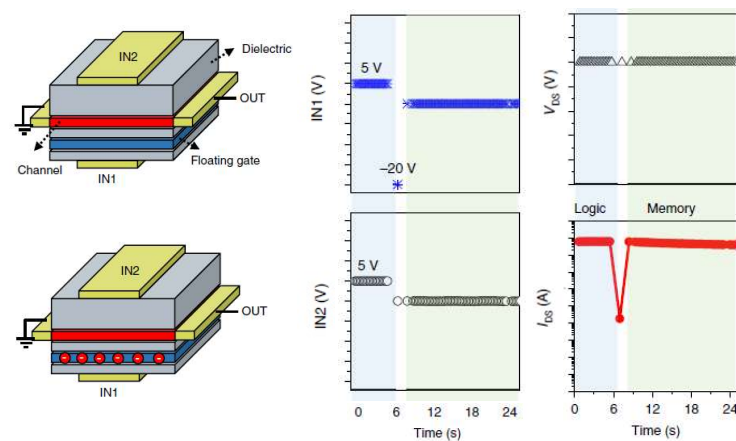
## AND gate



## Different operation depend on channel thickness

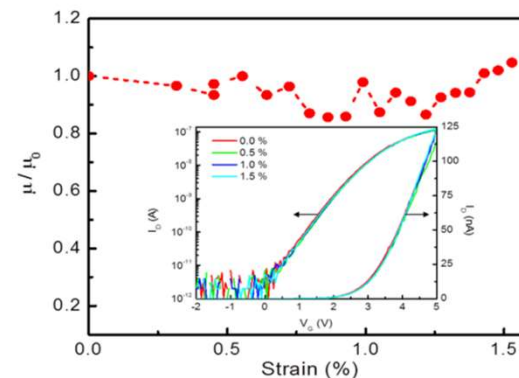
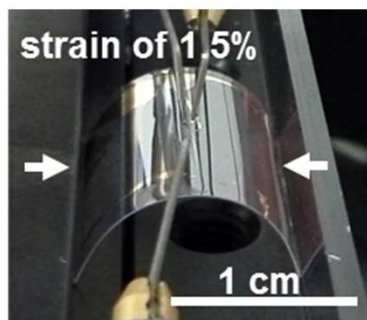
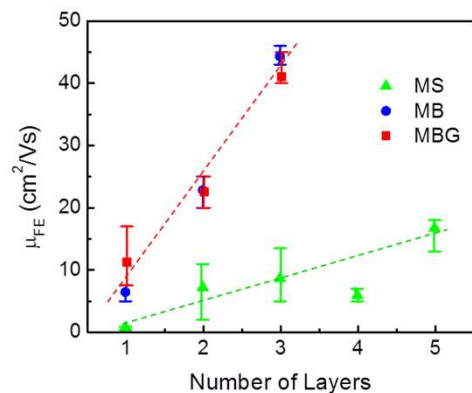
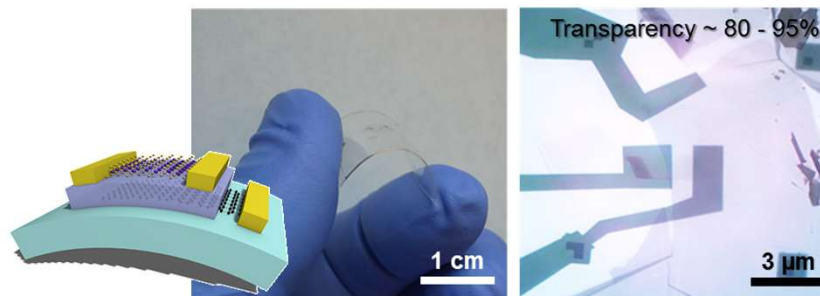
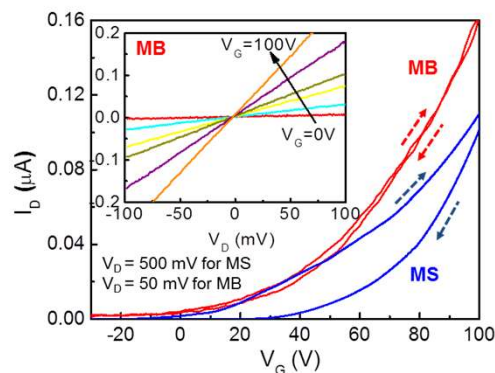
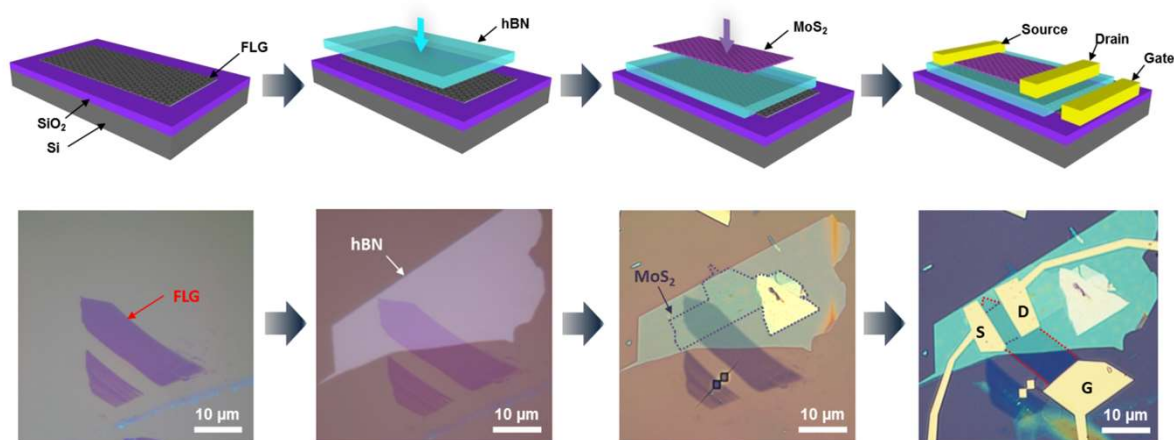
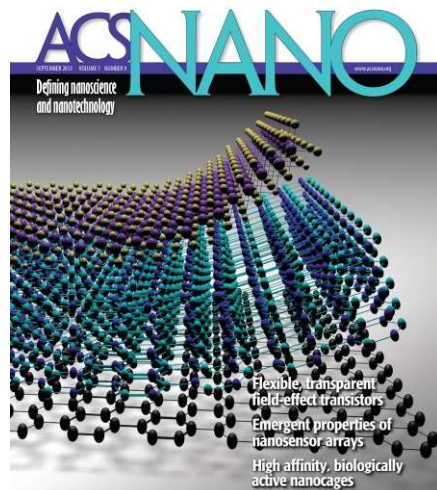


## Memory using floating gate



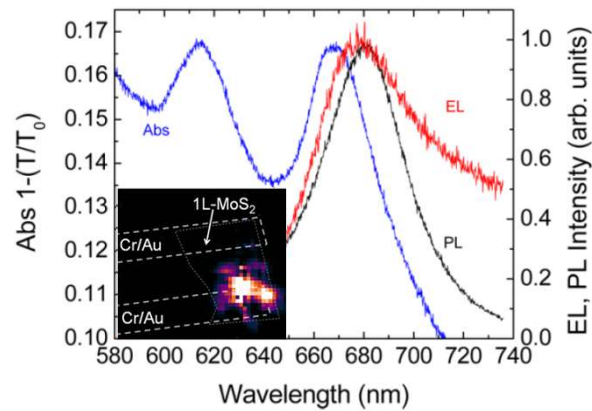


# Flexible and Transparent Heterostructure Devices



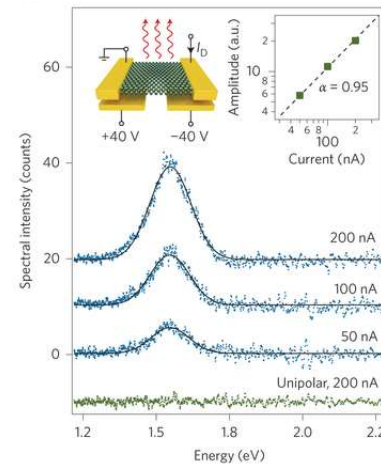
# Light Emitting Devices based on 2D Materials

## Thermionic emission



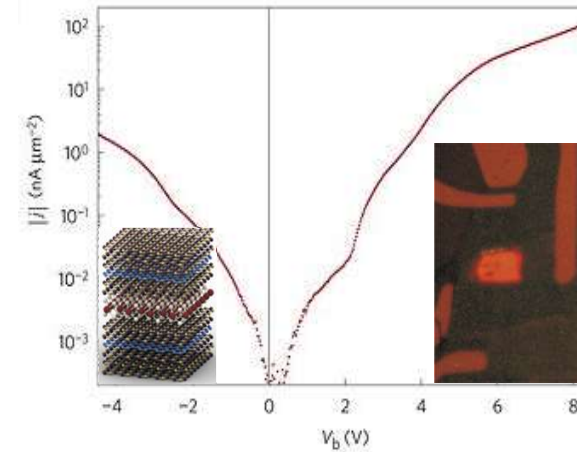
M. Steiner et al. *Nano Lett.* (2013)

## Lateral p-n diode

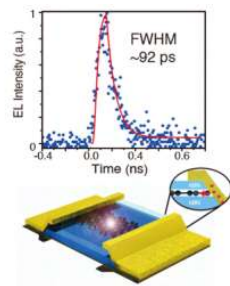
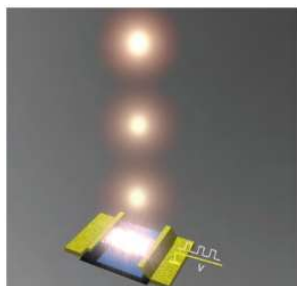


A. Pospischil et al. *Nat. Nanotech.* (2015)

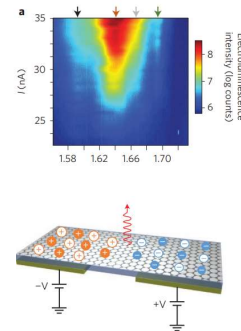
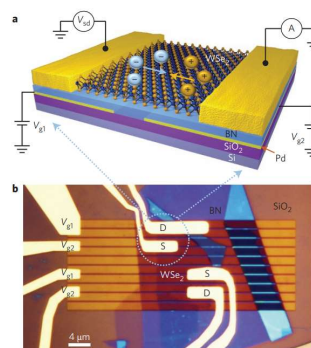
## Tunnel device



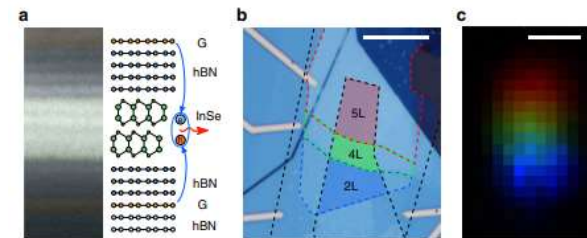
F. Withers et al. *Nat. Mater.* (2015)



J. Hone. et al. *Nano Lett.* (2018)

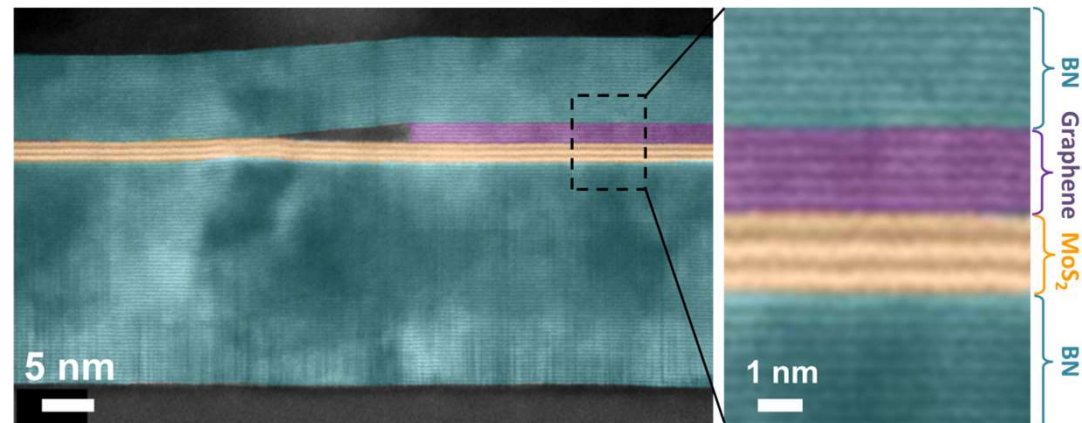
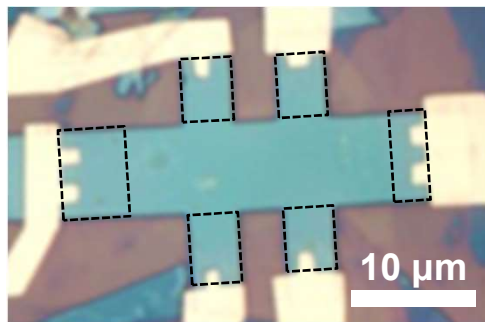
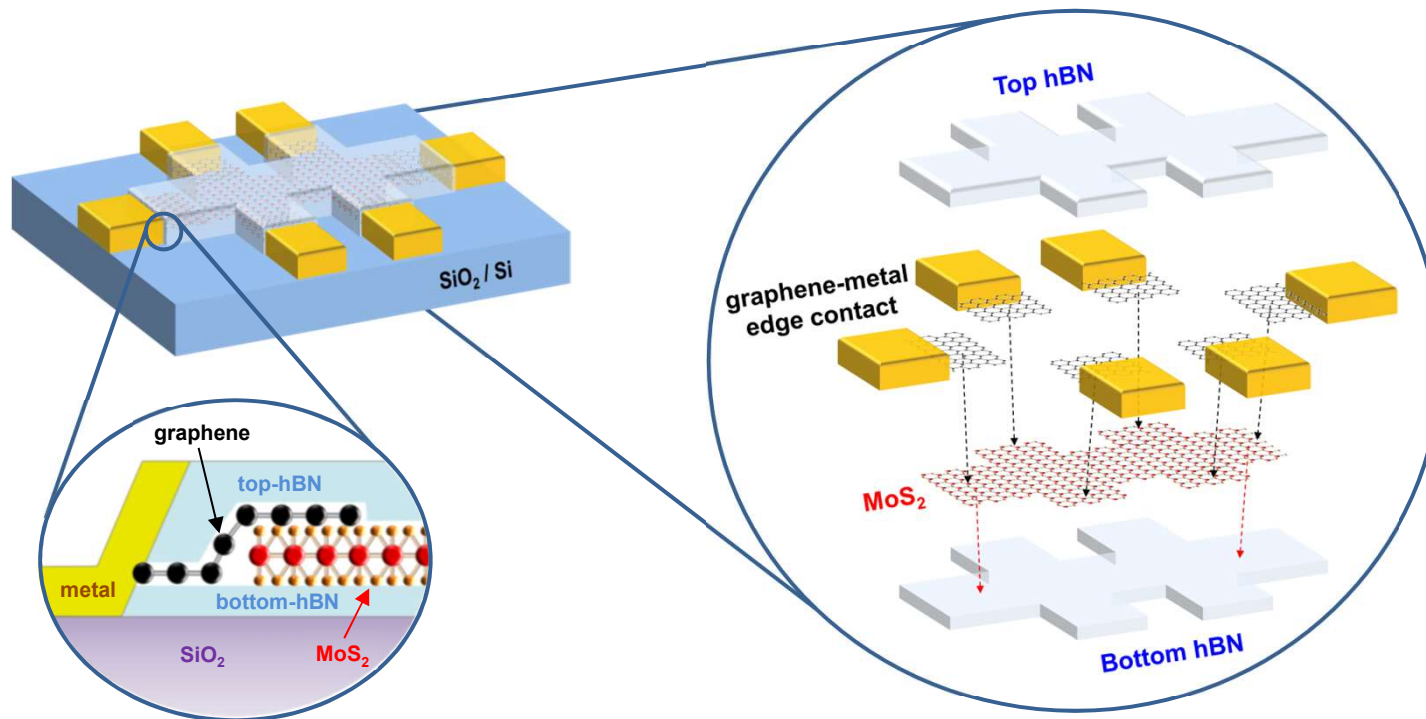


X. Xu. et al. *Nat. Nanotechno.* (2014)



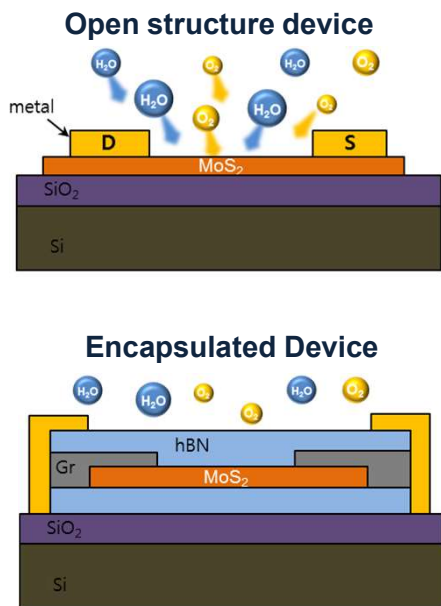
R. Gorbachev et al. *Nat. Commun.* (2020)

# vdW Heterostructure Device Platform

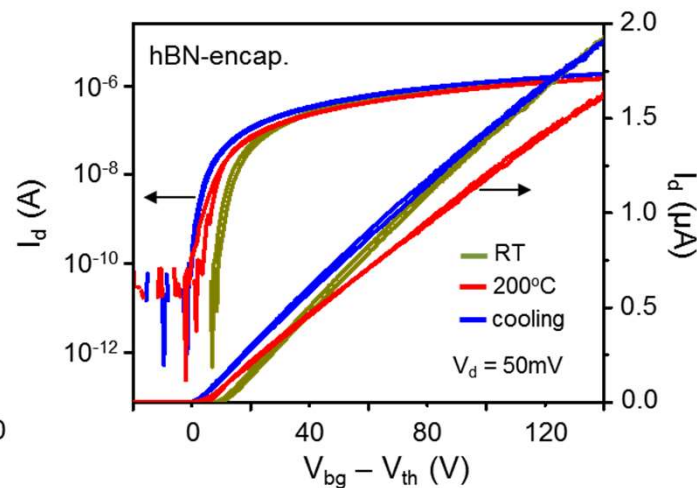
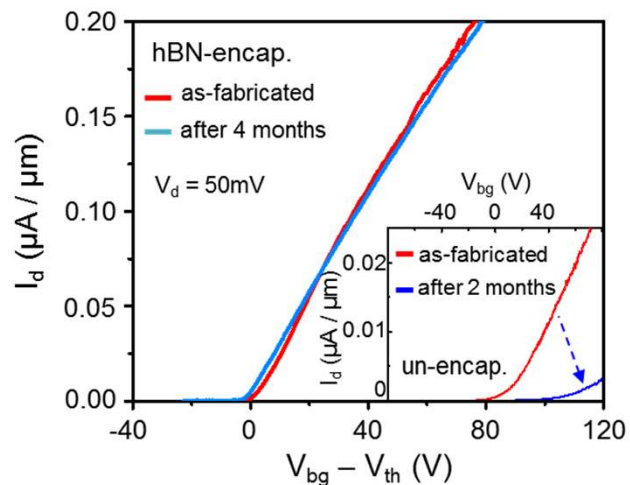




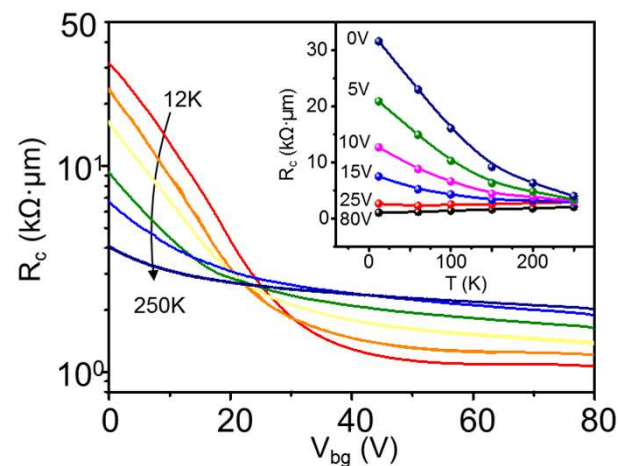
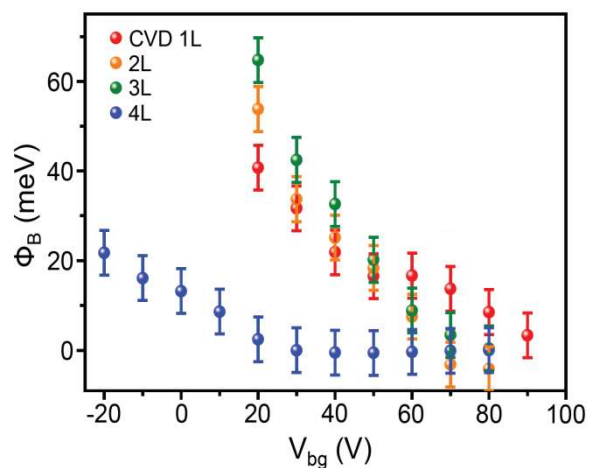
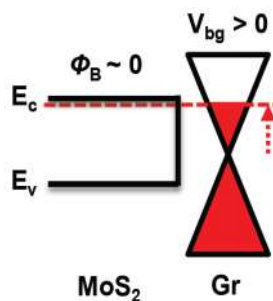
# High Stability and Low Contact Resistance



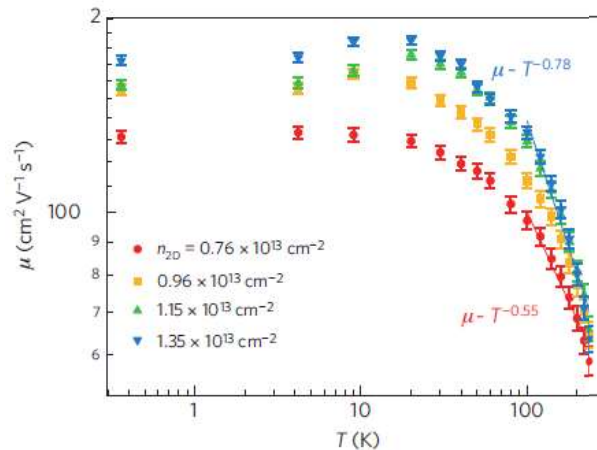
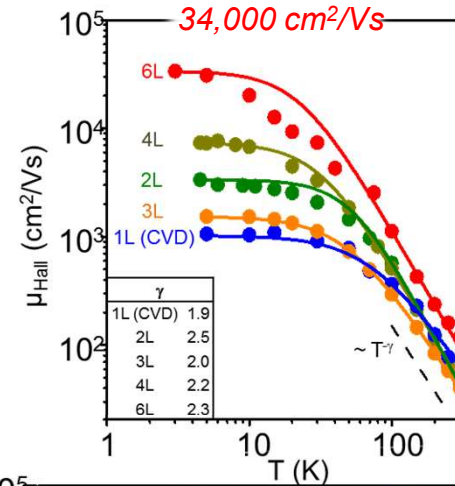
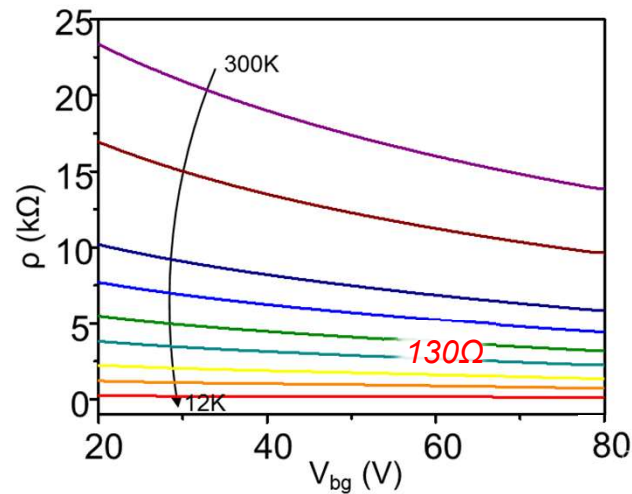
Perfect protection from environment



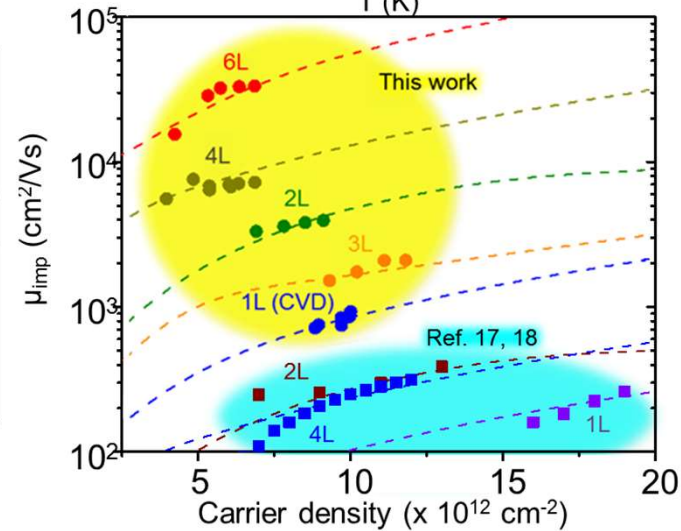
Gate-tunable graphene electrodes



# Ultrahigh Mobility of MoS<sub>2</sub>

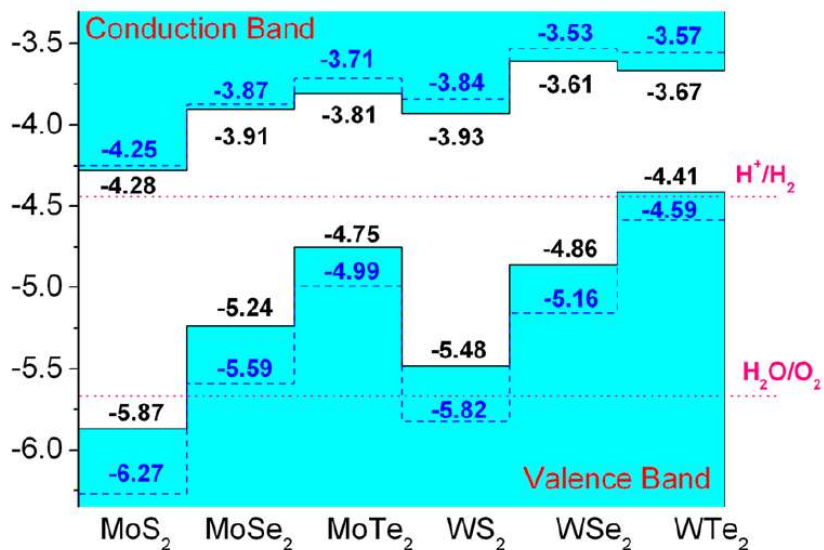


B. Radisavjevic et al. Nature Mater. (2014)

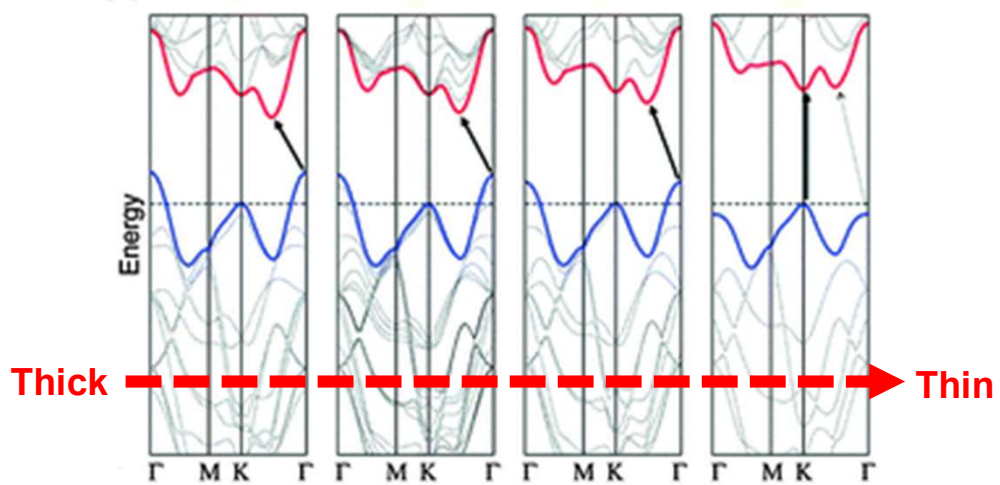
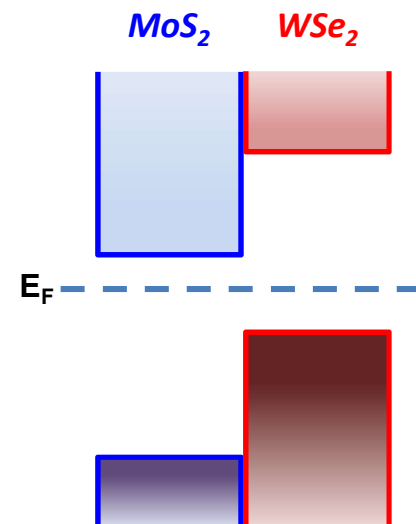
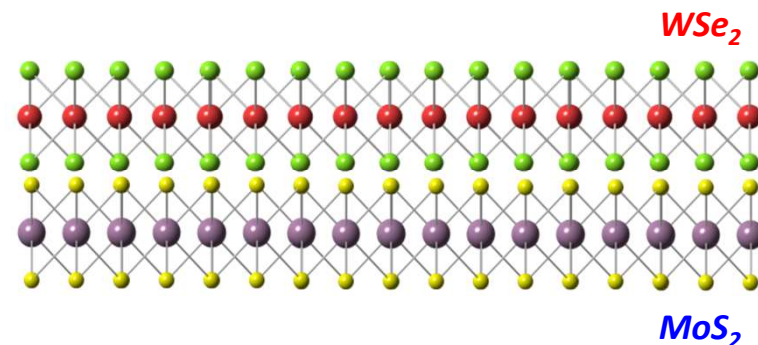


- Promising carrier mobility of 2D semiconductors
- Device platform for measurement of intrinsic electrical properties of 2D materials

# Heterostructures of p-type and n-type



*Appl. Phys. Lett.* **102**, 012111 (2013)



**Tunable photovoltaic effect?**  
**Light-emitting?**  
**New band structure at the interface?**



# Ultra-thin p-n Junction Devices

