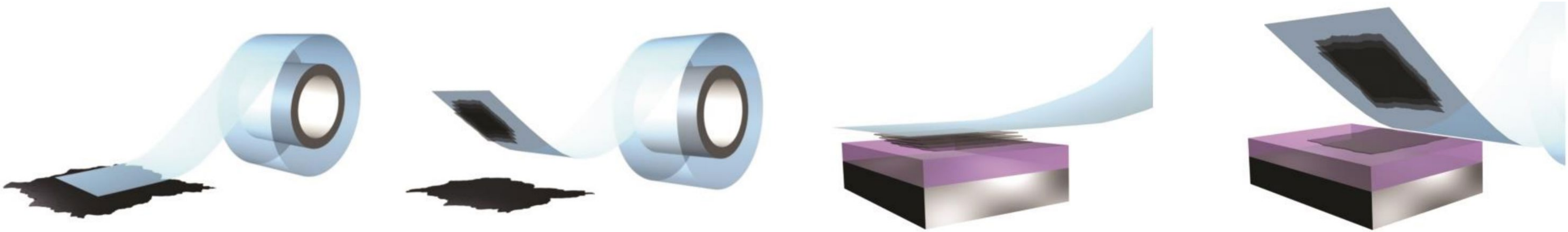
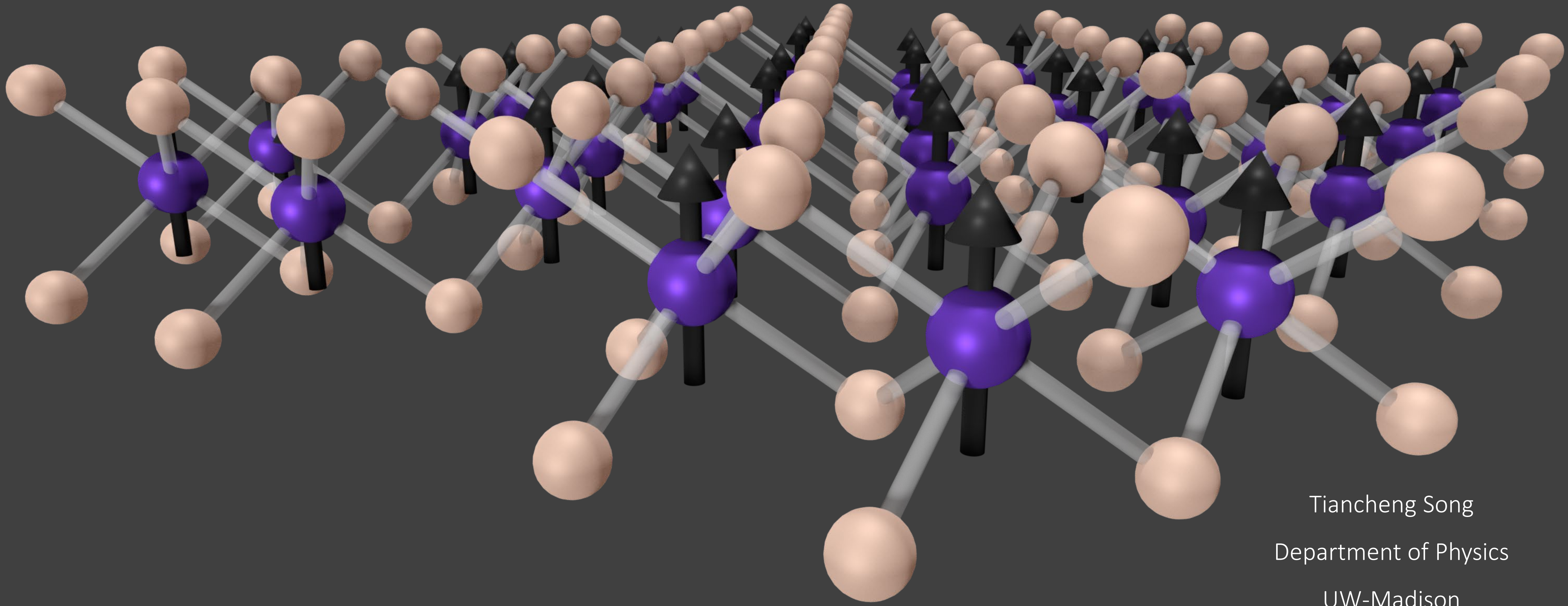
# University of Wisconsin-Madison

# Lab tour this afternoon!



# Quantum "LEGO" in 2D flatland

# 2D materials and van der Waals heterostructures

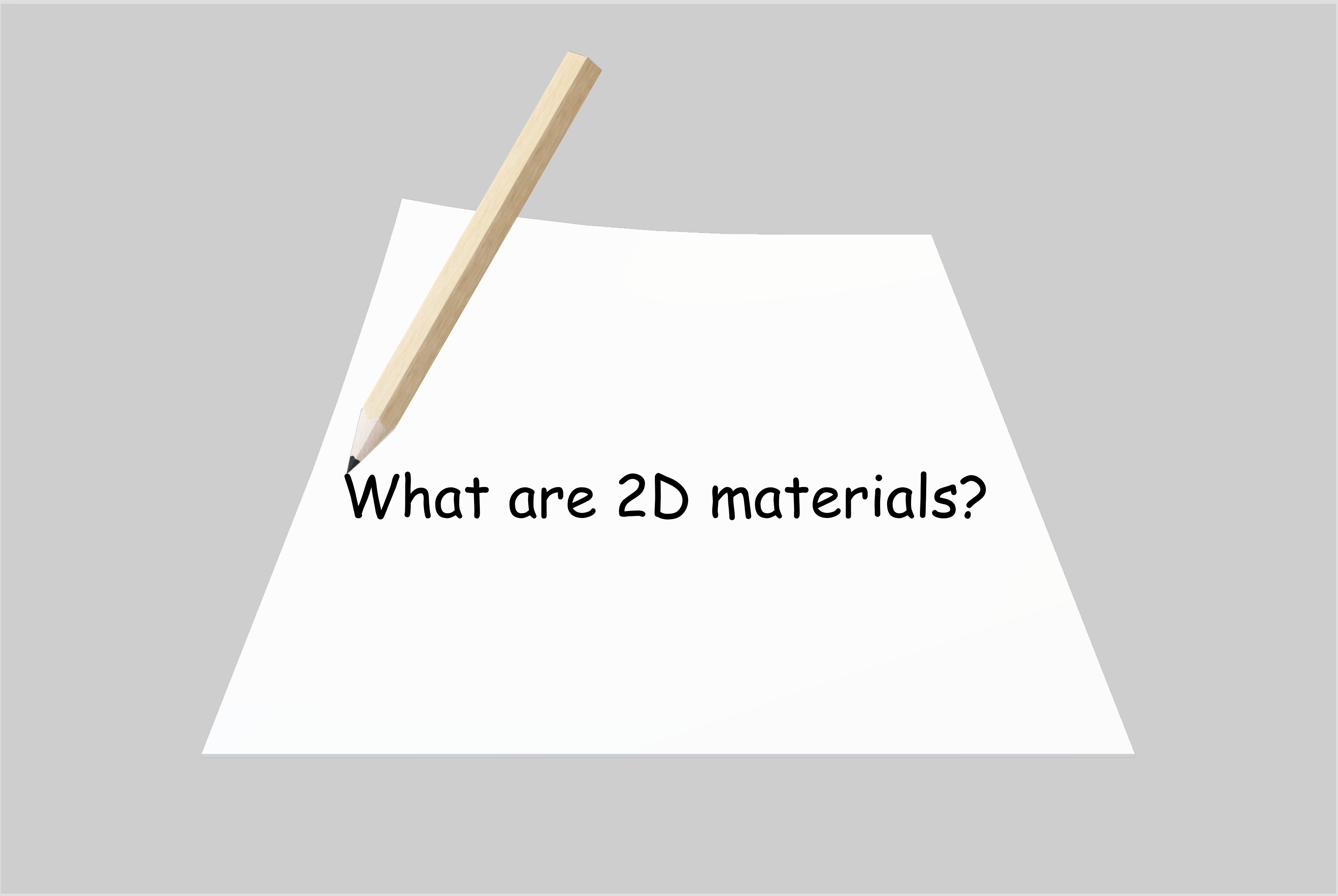


Tiancheng Song

Department of Physics

UW-Madison

# 2D materials with van der Waals bonding

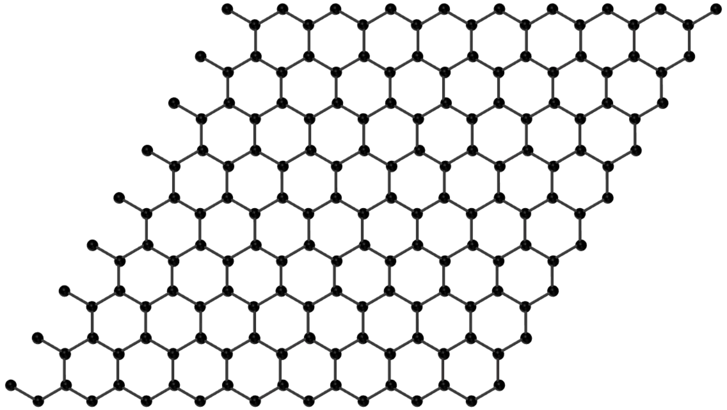


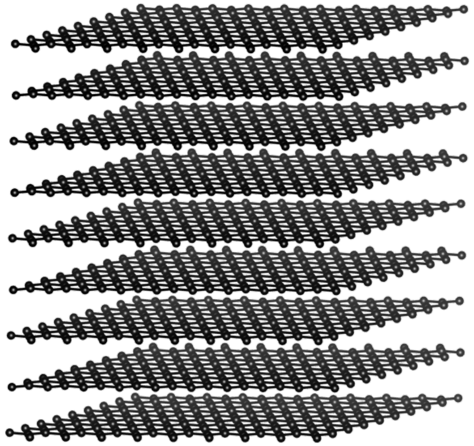
# 2D materials with van der Waals bonding

Single sheet of paper

Stack of paper

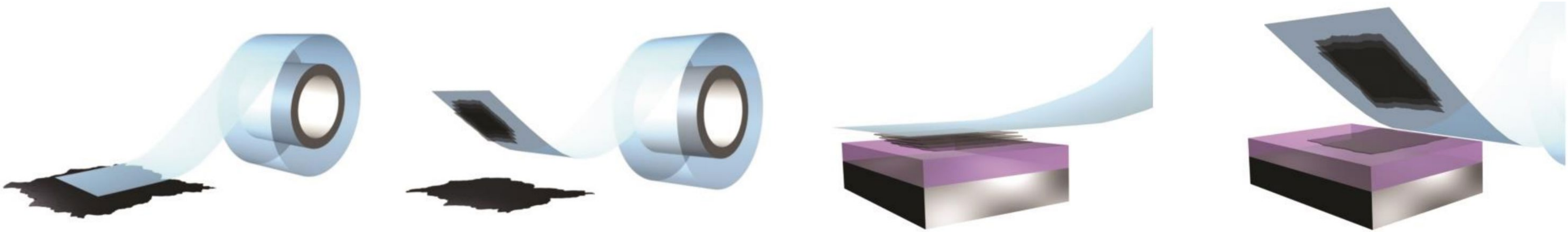
Monolayer graphene





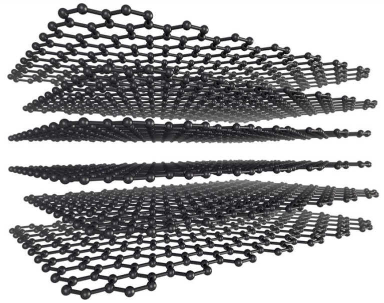
Stack of graphene

# The first 2D material: Graphene!

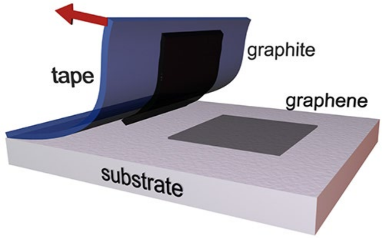


# Evolution of 2D materials

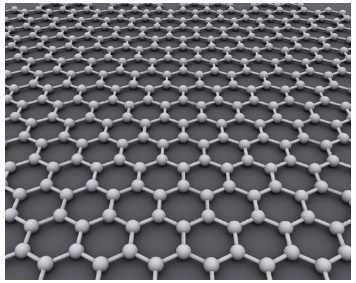
3D bulk crystal graphite



Scotch tape exfoliation




2D monolayer graphene

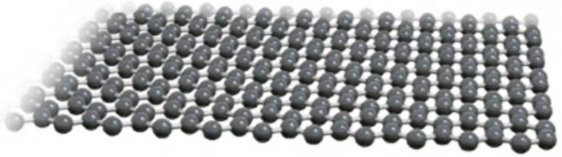


2004

2011

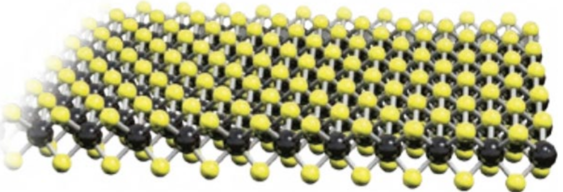
2017

Semimetal



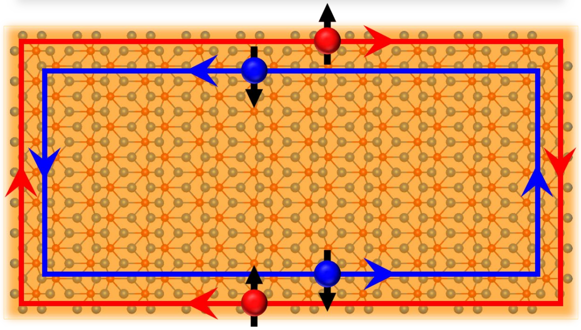
Graphene

Semiconductor

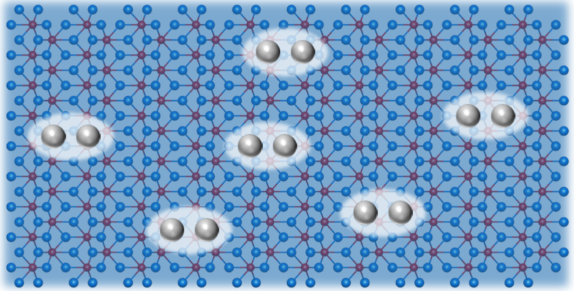


$MoS_2$, $WSe_2$, …
Insulator: h-BN

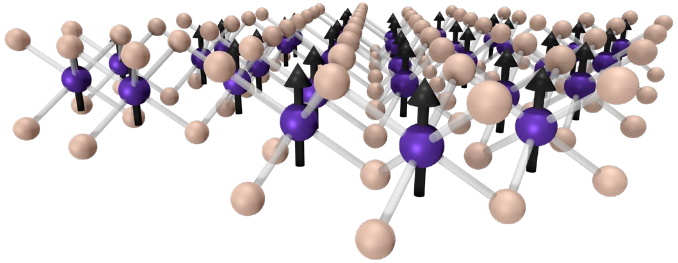
2D topological insulator



2D superconductor

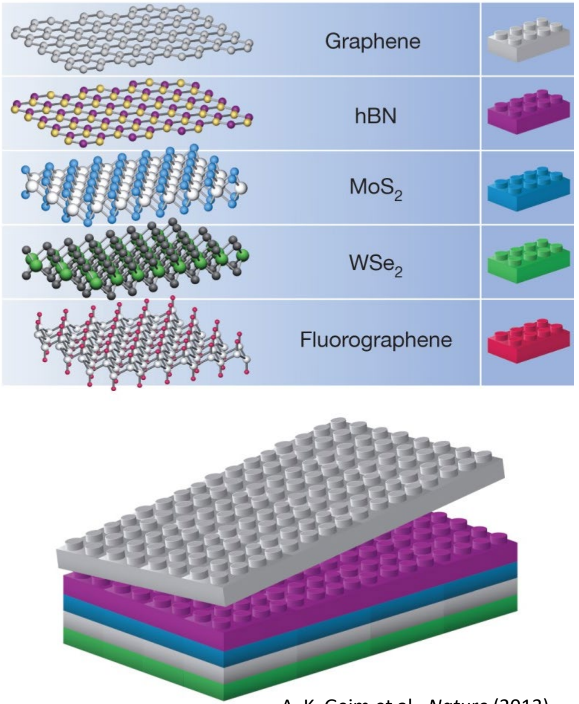


2D magnet



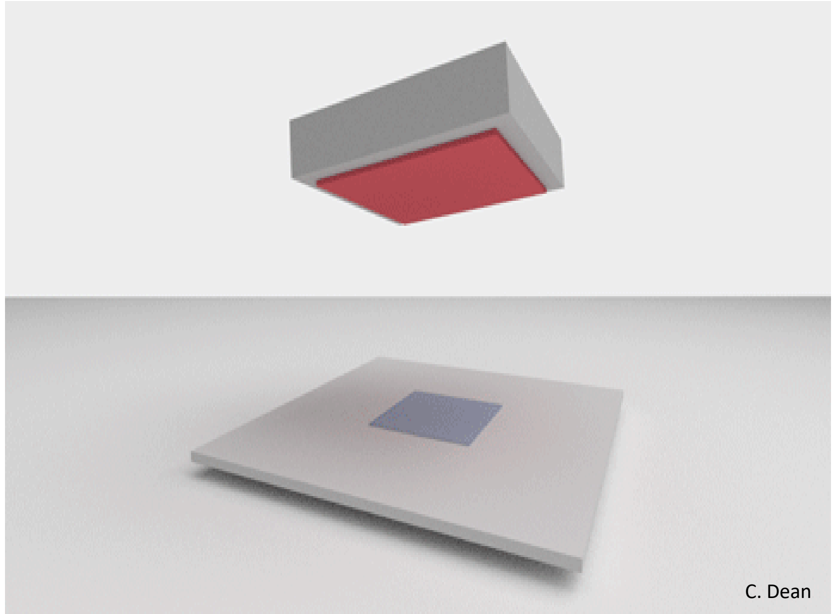
# Play with atomic scale "LEGO"

**"LEGO set" of 2D materials**

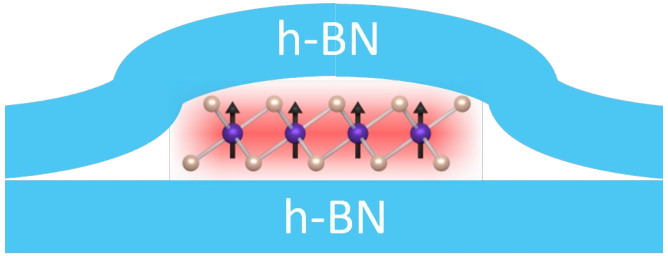



A. K. Geim et al., *Nature* (2013).

**Transfer technique**

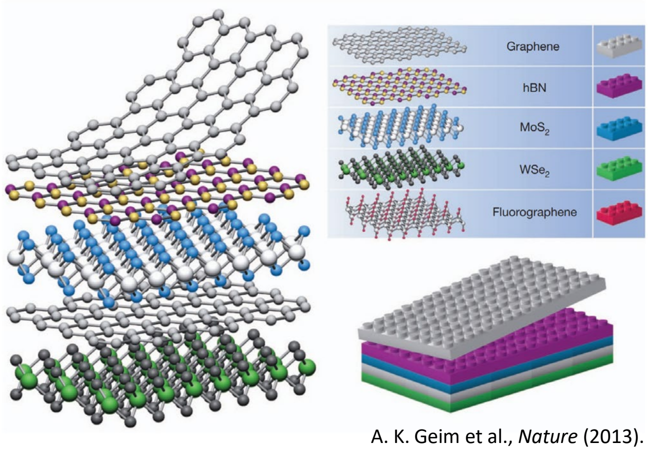



**Van der Waals heterostructures**

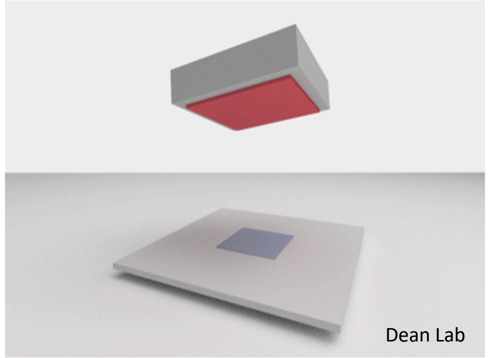


# Versatile platform

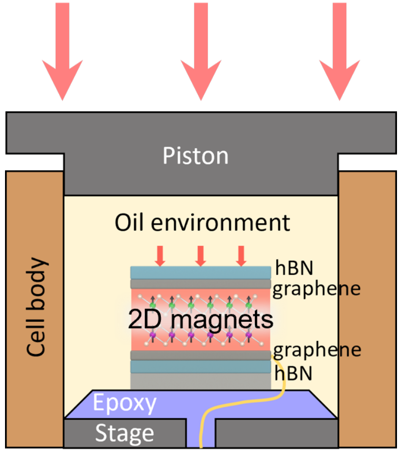
## Building van der Waals heterostructures

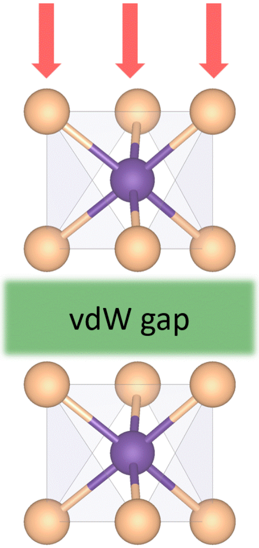



A. K. Geim et al., *Nature* (2013).

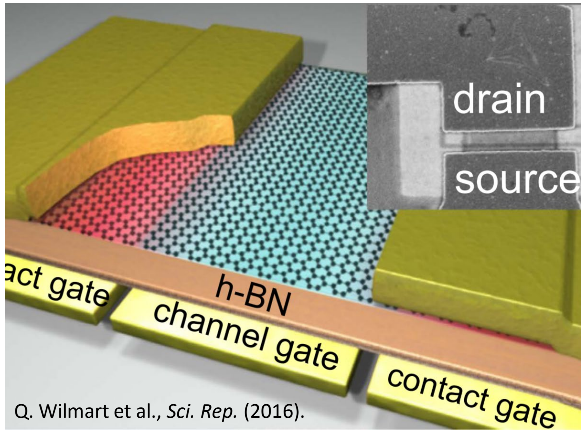



## Applying pressure

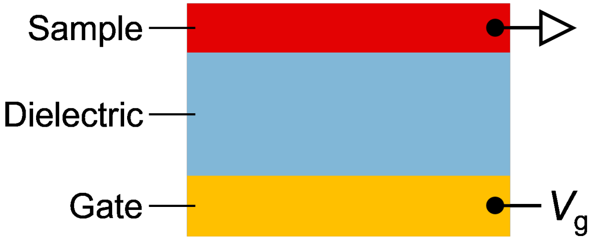


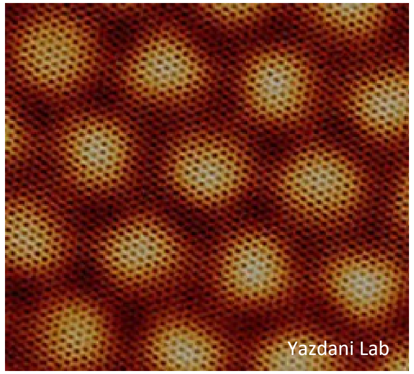

## Tuning carrier density

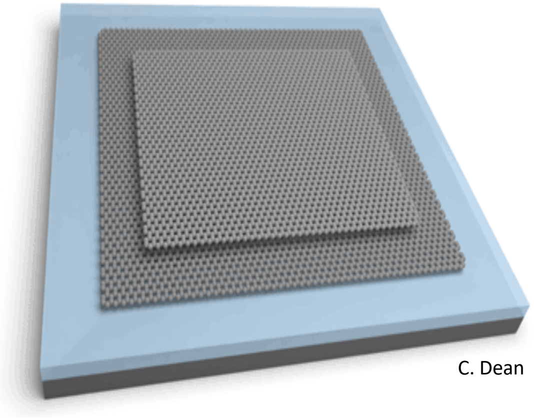



Q. Wilmart et al., *Sci. Rep.* (2016).




## Twisting two layers

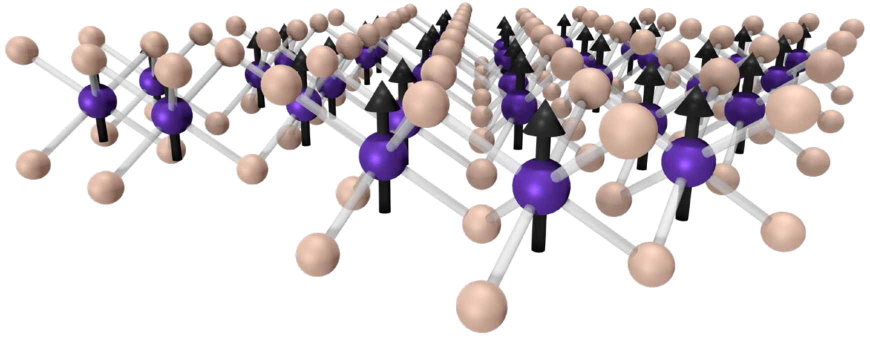


C. Dean

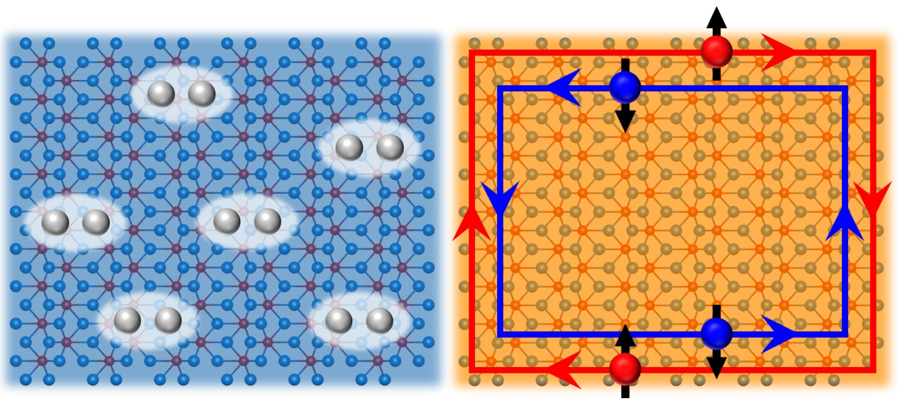
# Outline: two examples

## 2D magnet



- Discovery of **2D magnets**
- Van der Waals **spintronics**
- **Layer stacking**-dependent magnetism
- Twisted 2D magnets → **magnetic moiré**

## 2D superconductor + topological insulator



- 2D **topological insulator**
- Gated-tunable **2D superconductivity**

# Challenge

These new 2D materials are mostly **air-sensitive** (chemical instability)

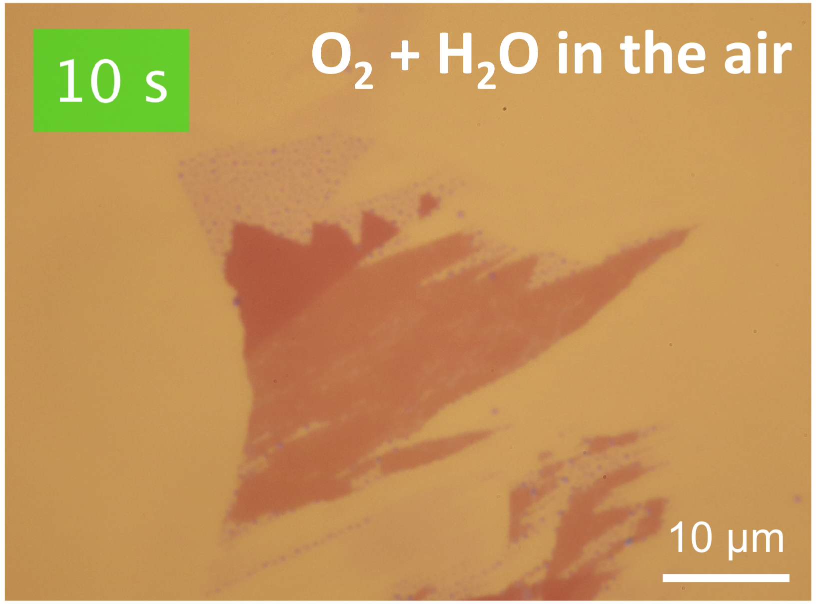
VdW magnet synthesized in **1965**

JOURNAL OF APPLIED PHYSICS VOLUME 36, NO. 3 (TWO PARTS—PART 2) MARCH 1965

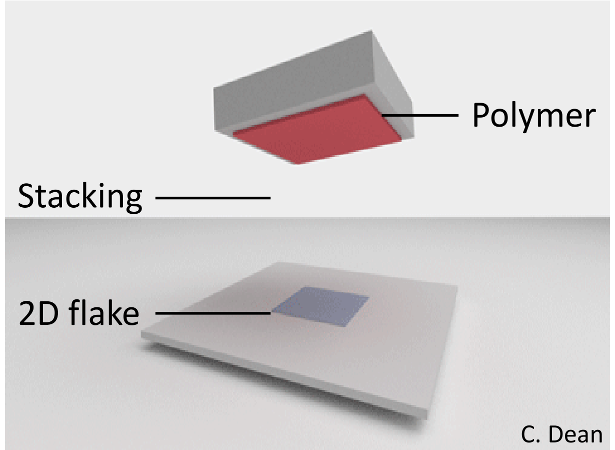
**Magnetization, Resonance, and Optical Properties of the Ferromagnet $CrI_3$**

J. F. DILLON, JR.
*Bell Telephone Laboratories, Murray Hill, New Jersey*
AND
C. E. OLSON
*Los Alamos Scientific Laboratories, Los Alamos, New Mexico*

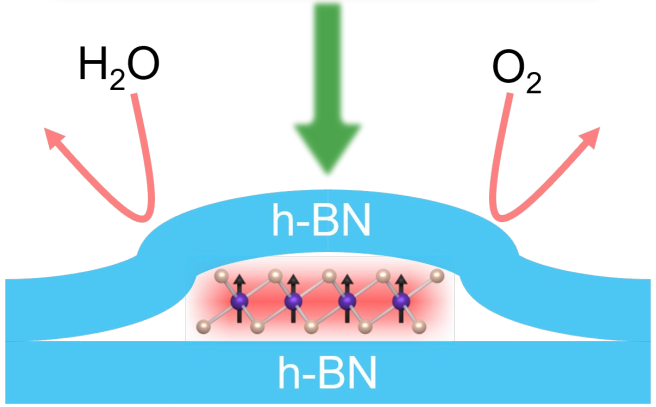
**Extremely** air-sensitive




Solution




Sandwich encapsulation

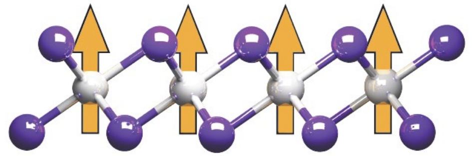


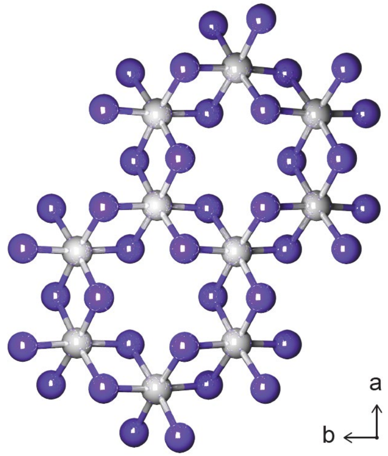

**Air-stable!**

# Discovery of 2D magnets

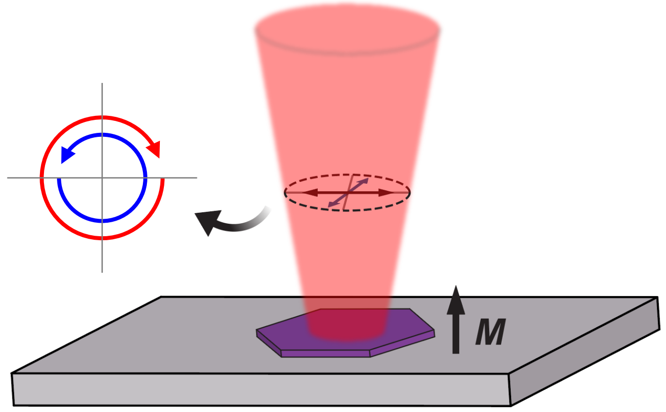
## **First** 2D magnet chromium triiodide ($CrI_3$)

### **Ferromagnetic** monolayer

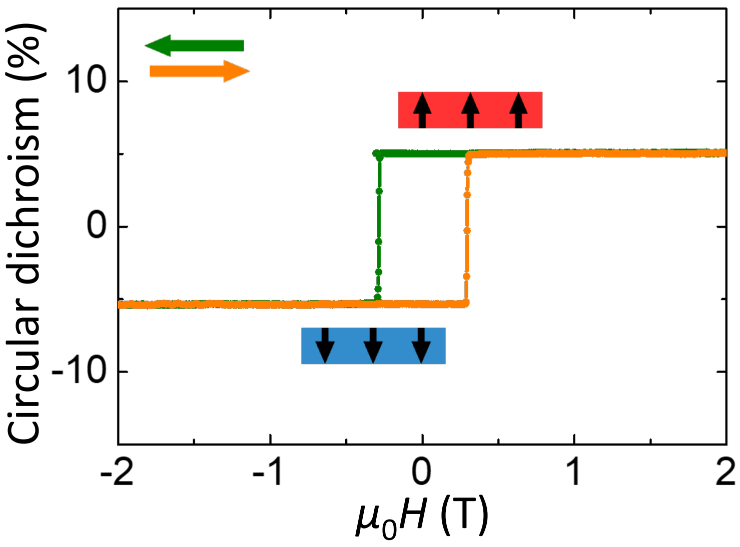




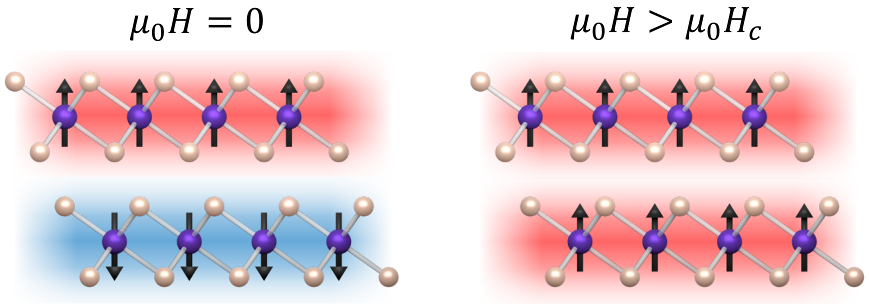

B. Huang, X. Xu et al., *Nature* (2017).
C. Gong et al., *Nature* (2017).

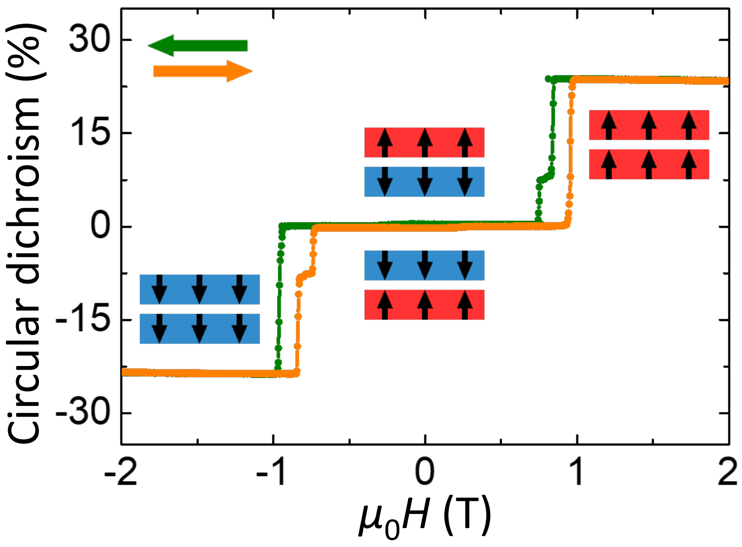



Circular dichroism

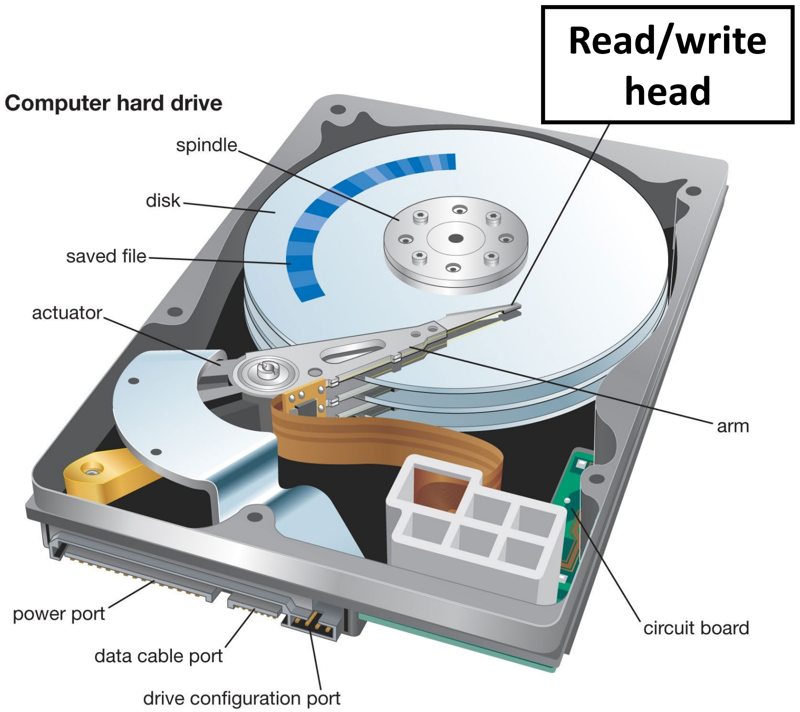


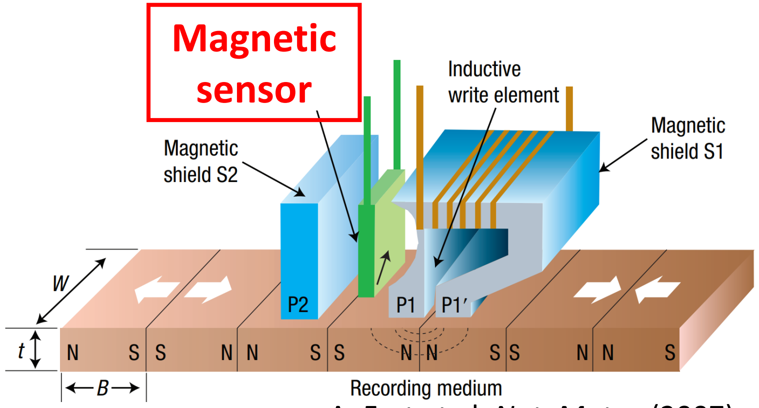

### **Layered-antiferromagnetic** bilayer



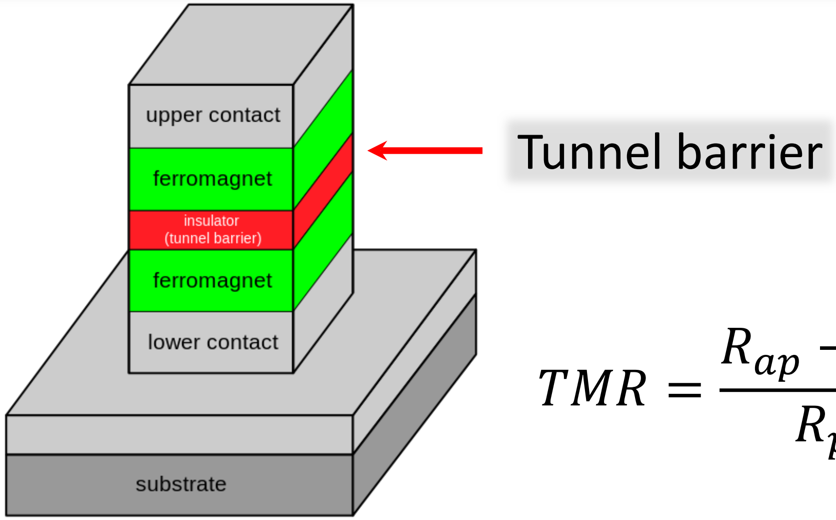
# Spintronic devices




A. Fert et al. *Nat. Mater.* (2007).

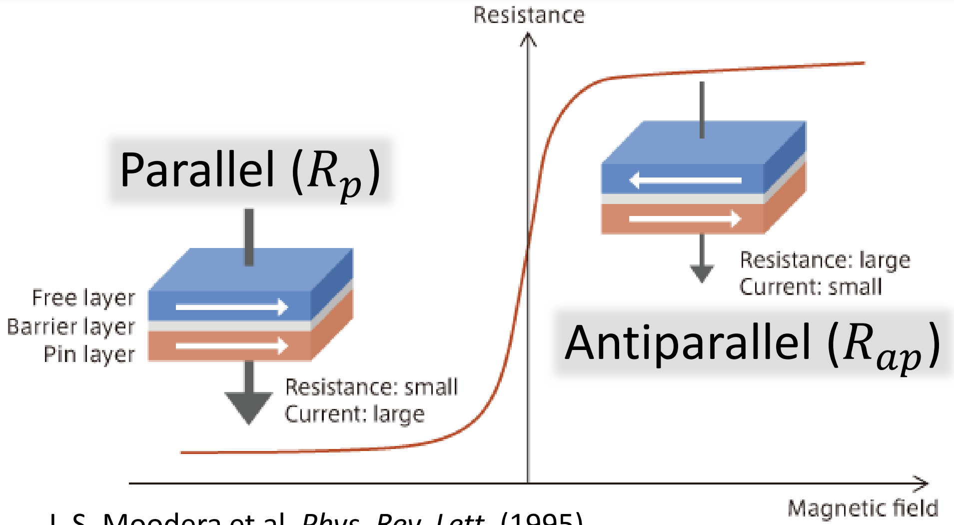
## Magnetic tunnel junction (MTJ): FM/insulator/FM





$$TMR = \frac{R_{ap} - R_p}{R_p}$$

## Tunneling magnetoresistance: 600% (300K) and 1100% (5K)

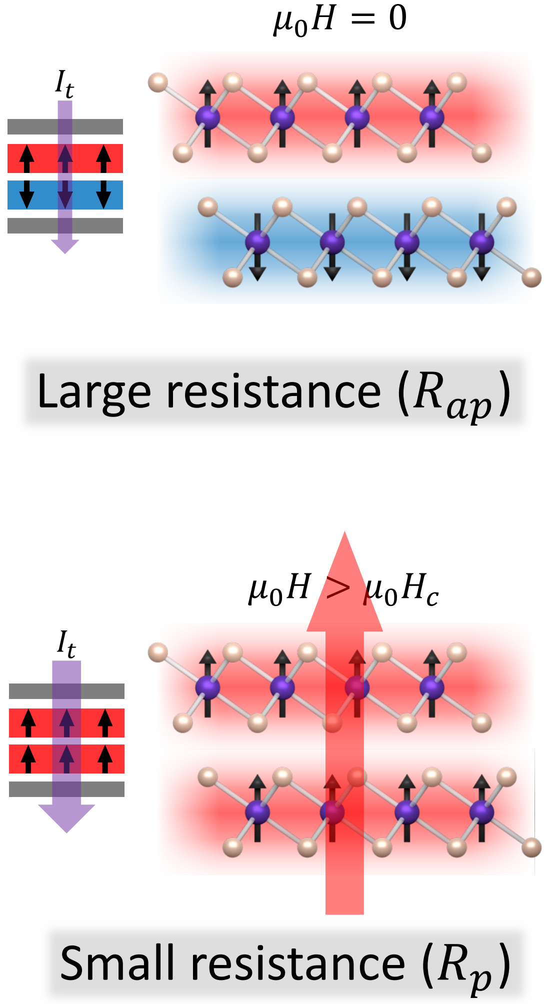


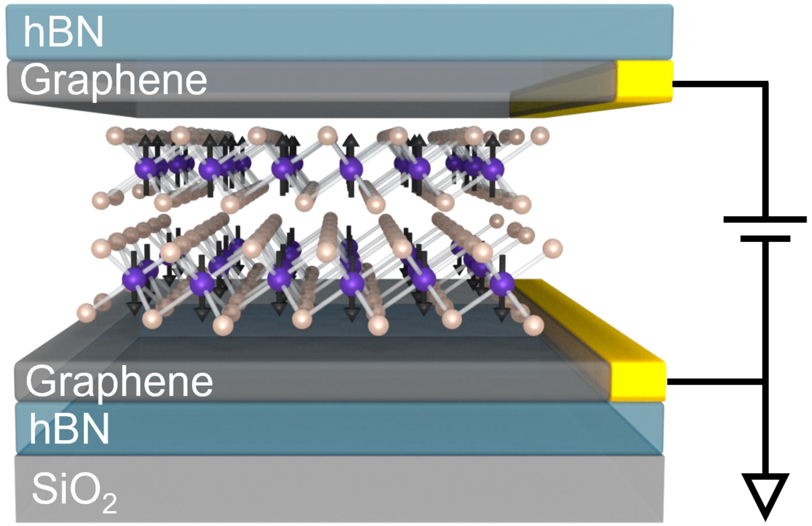

J. S. Moodera et al. *Phys. Rev. Lett.* (1995).
T. Miyazaki et al. *J. Magn. Magn. Mater.* (1995).

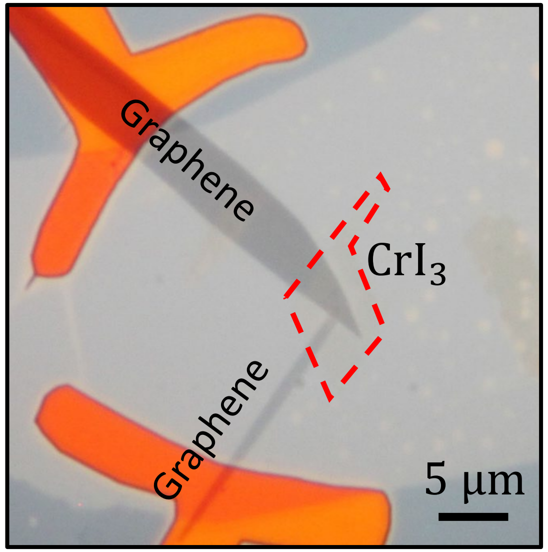
# Van der Waals spintronics?

T. **Song** et al., *Science* (2018).

Bilayer $CrI_3$ is **desirable** for spin-filter MTJ



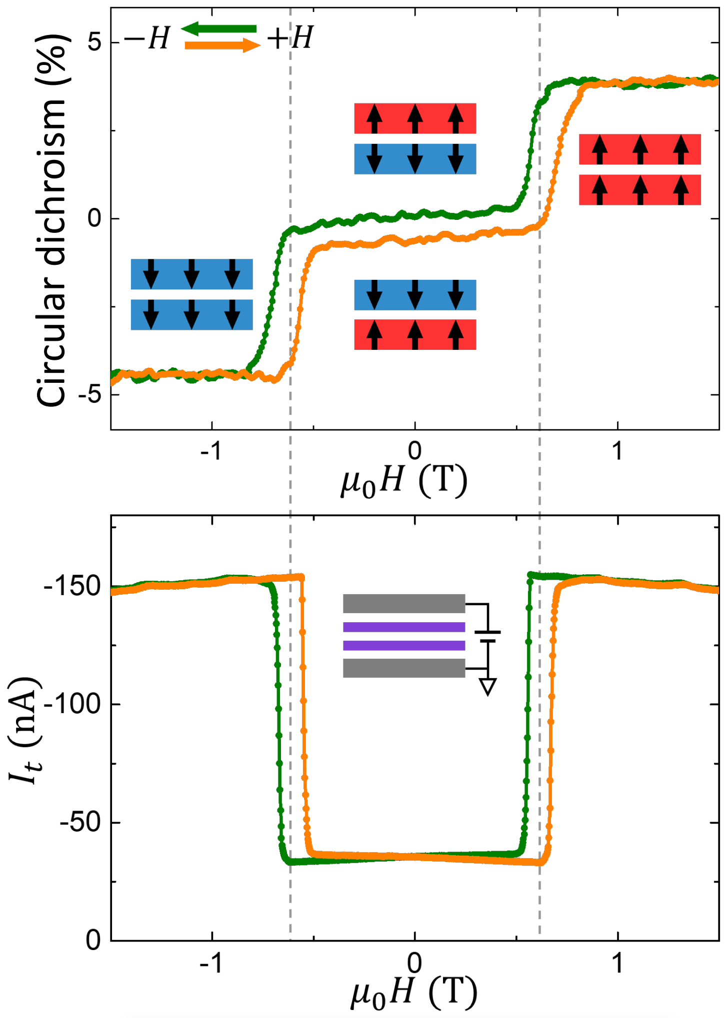



# Atomically thin MTJ

T. **Song** et al., *Science* (2018).

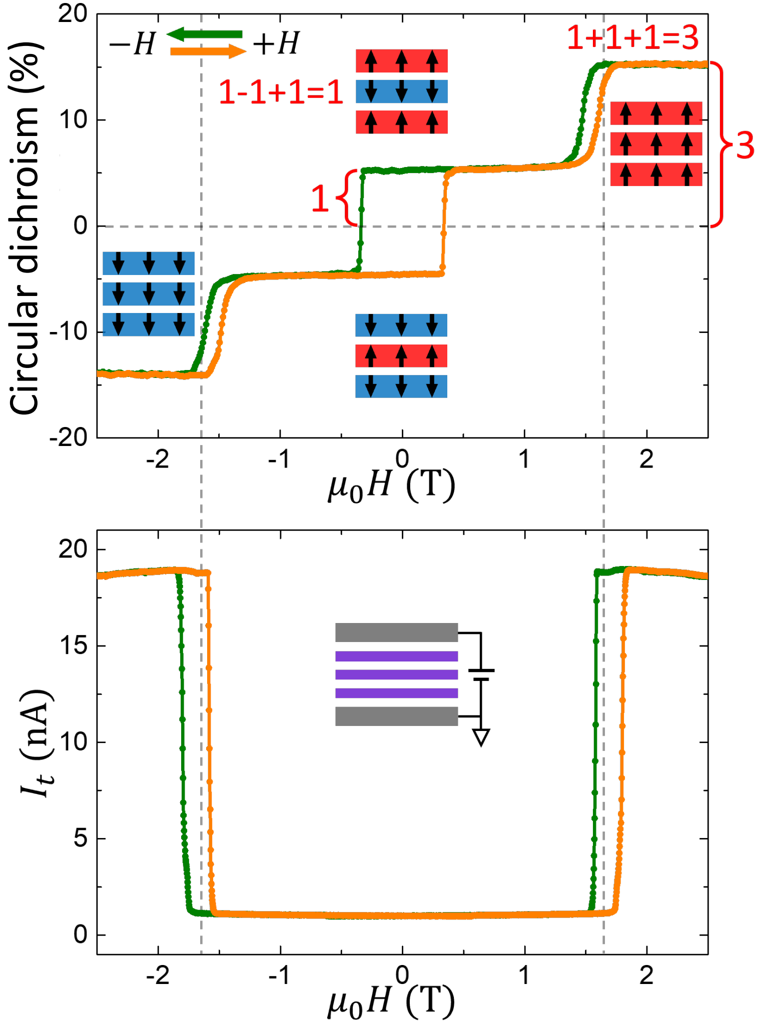
First demonstration of **all-vdW spintronics**

**Thinnest** magnetic tunnel junction




**Bilayer TMR > 300%**

**Enhanced** TMR ratio

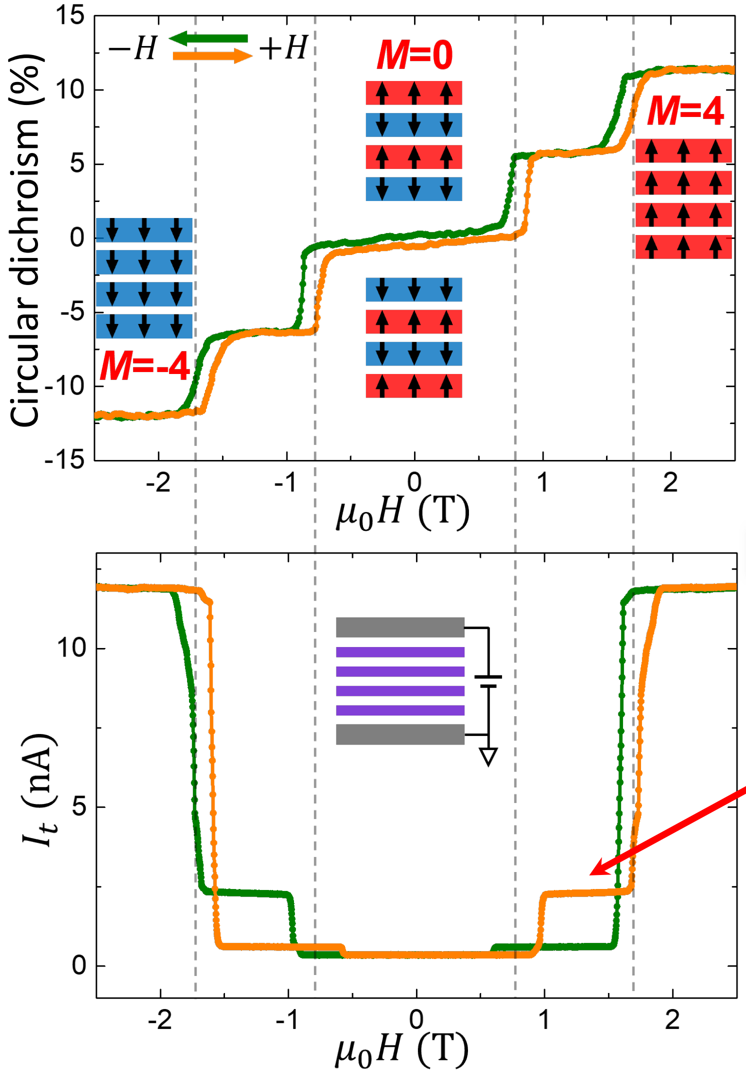



**Trilayer TMR > 2,000%**

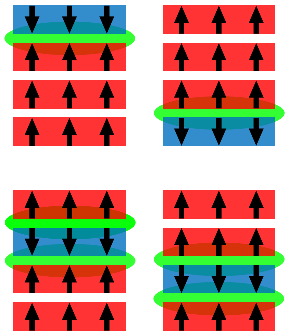
# Record high TMR!

T. Song et al., *Science* (2018).
T. Song et al., *Nano Letters* (2019).

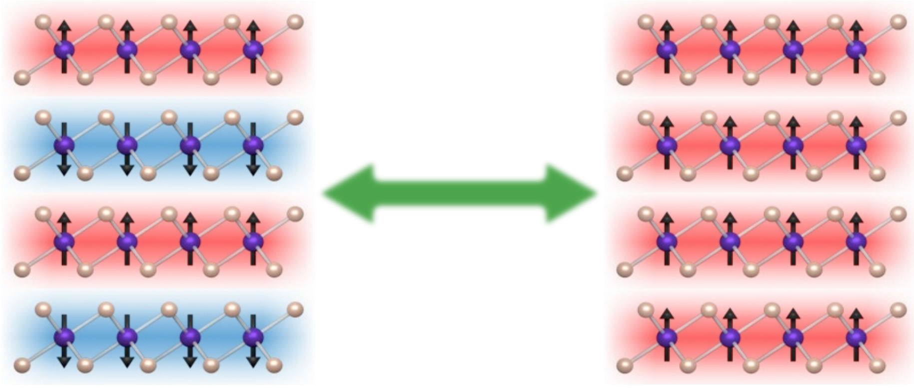
Four-layer TMR > 57,000%, but Why?



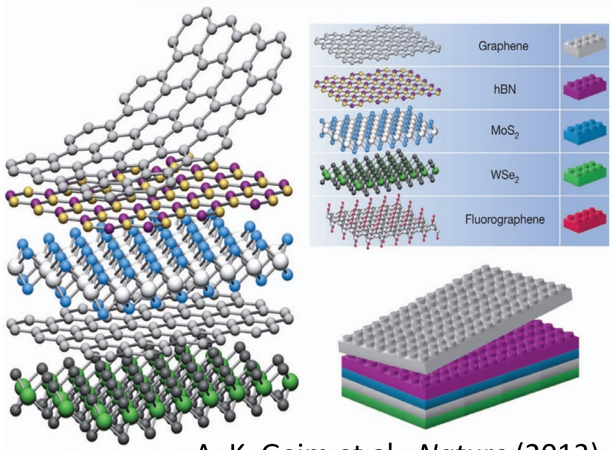

New magnetic states!



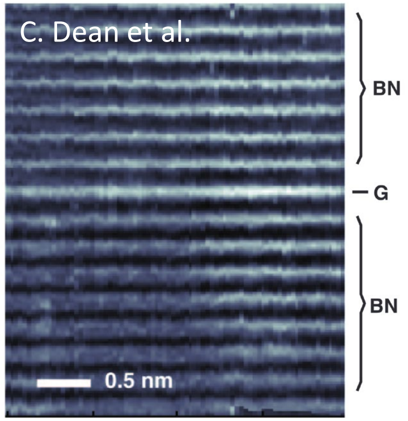
Natural spin filters in series



Atomically sharp vdW interfaces



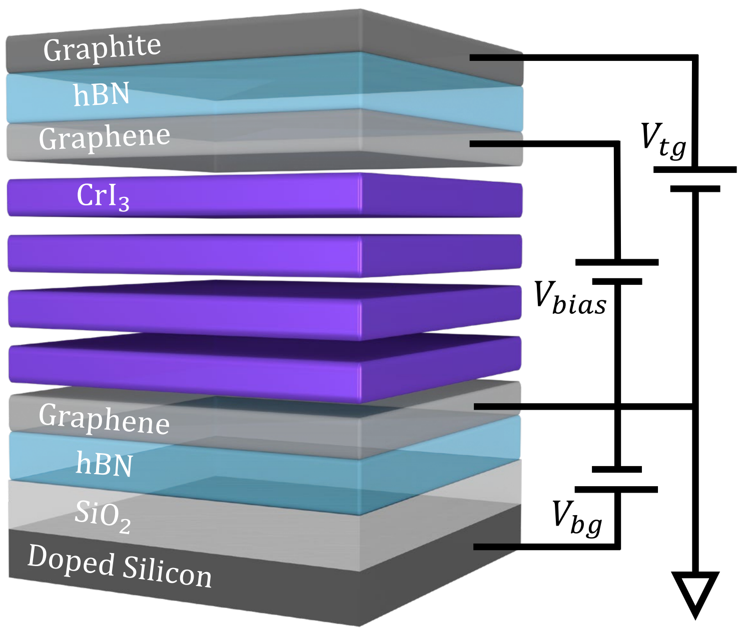

A. K. Geim et al., *Nature* (2013).

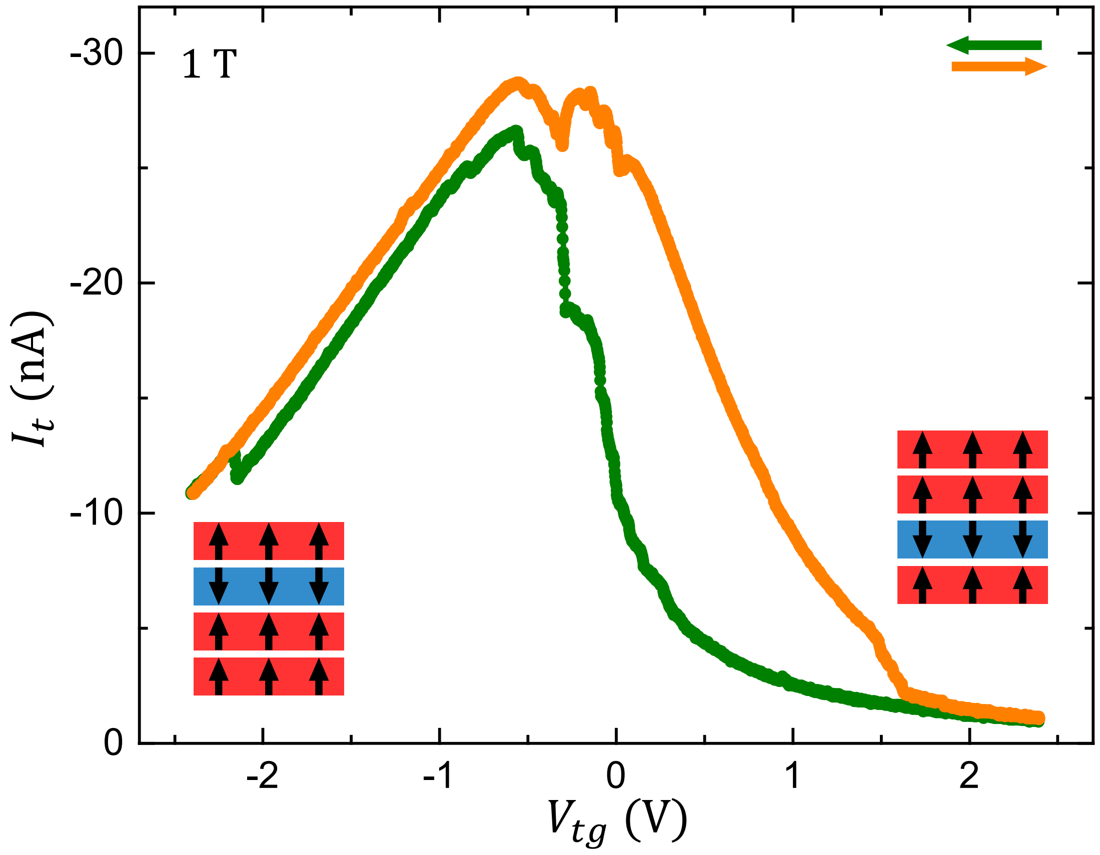


# Electrical switching of bistable states

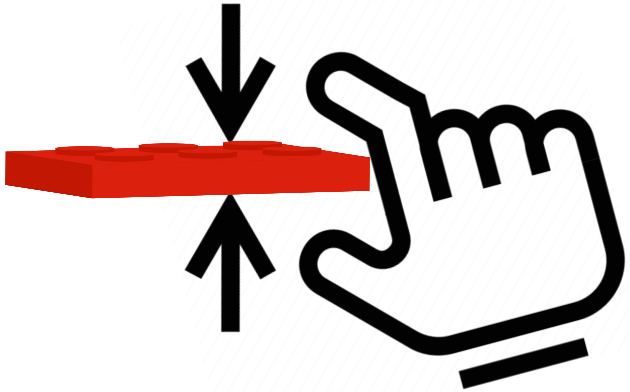
**T. Song** et al., *Nano Letters* (2019).

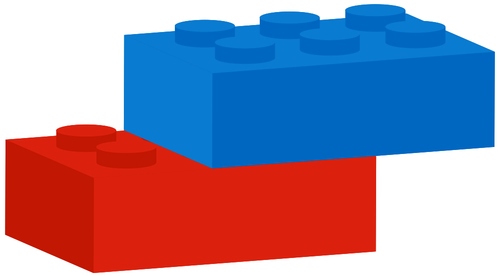
Upgraded with top and bottom gates

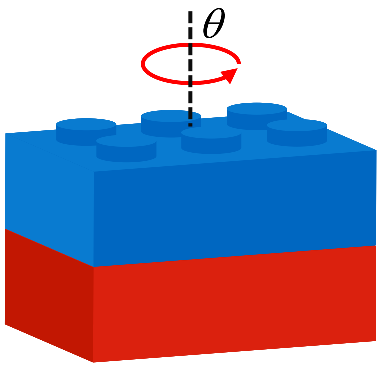


# Blessing of vdW nature



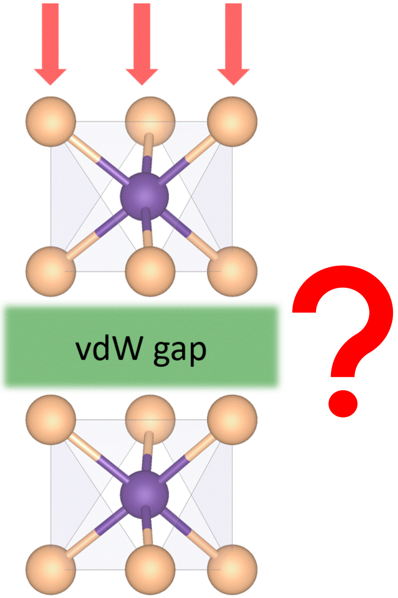


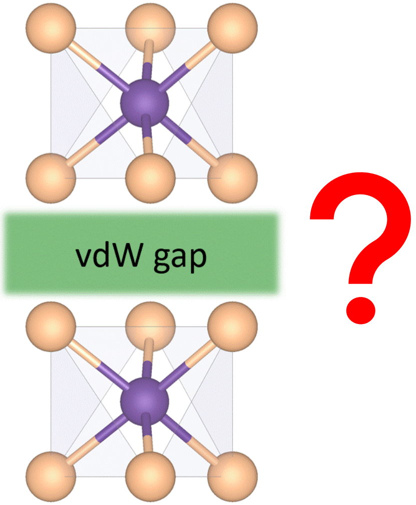


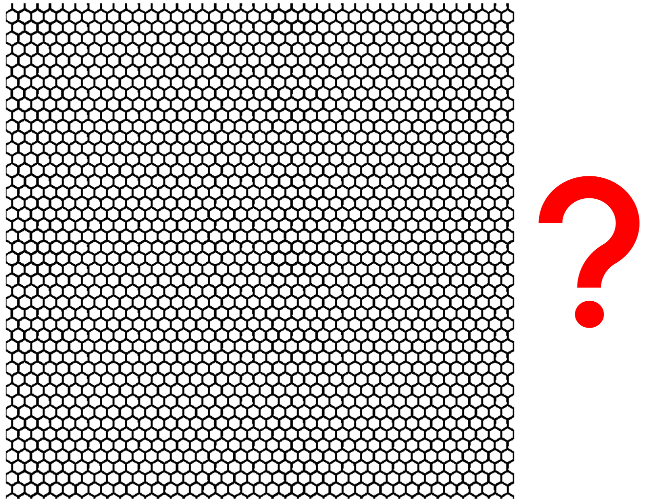

Reduce interlayer **spacing**

Lateral interlayer **shift**

**Twist** two layers



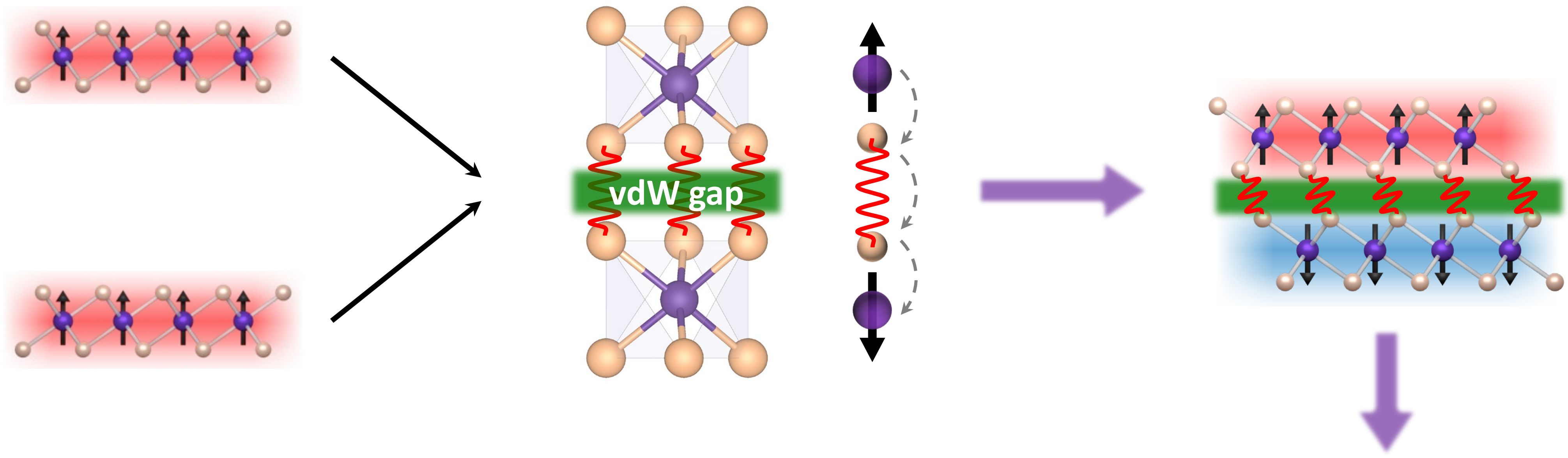



# Why we care about vdW interface?

Two **free** monolayers

**Exchange** interactions

AFM **interlayer** coupling

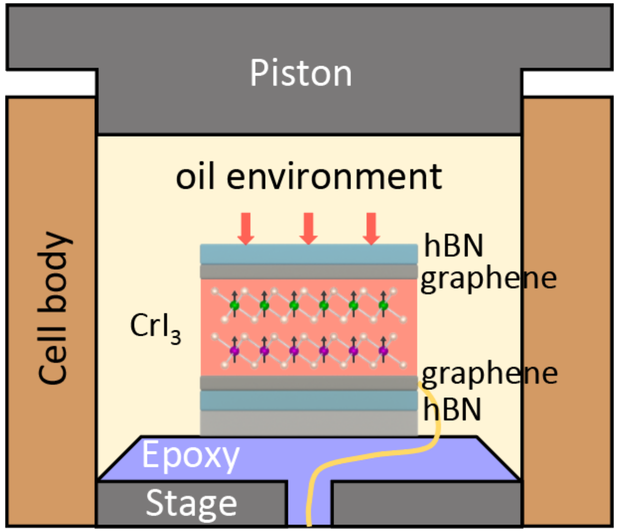



Determined by **vdW interface**

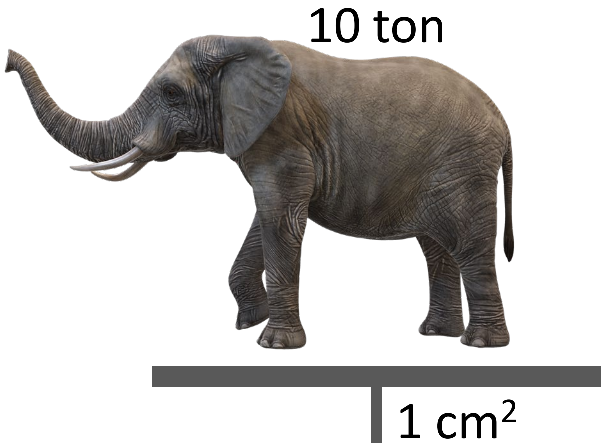
# Pressure tunning

**T. Song** et al., *Nat. Mater.* (2019).
T. Li et al., *Nat. Mater.* (2019).

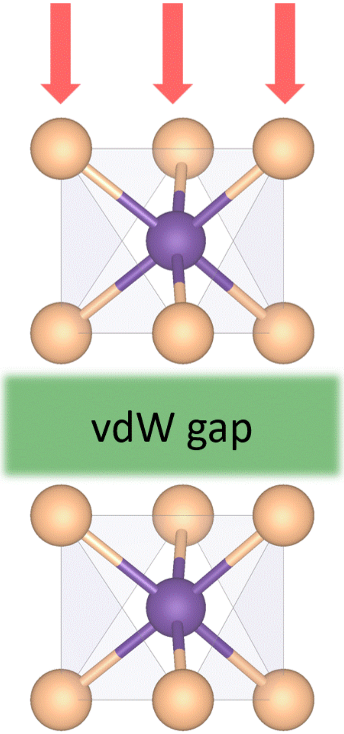
Piston-cylinder pressure cell




Up to 3 GPa

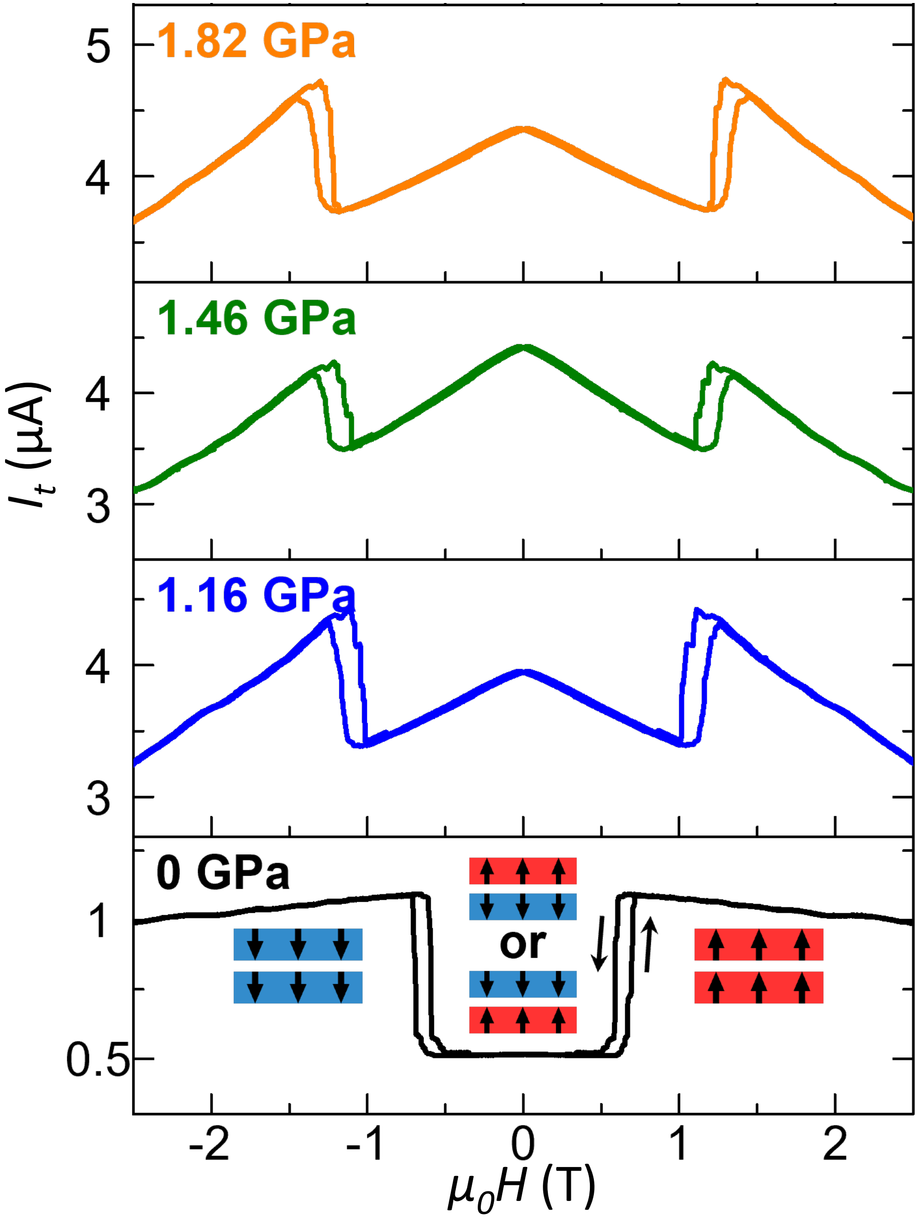


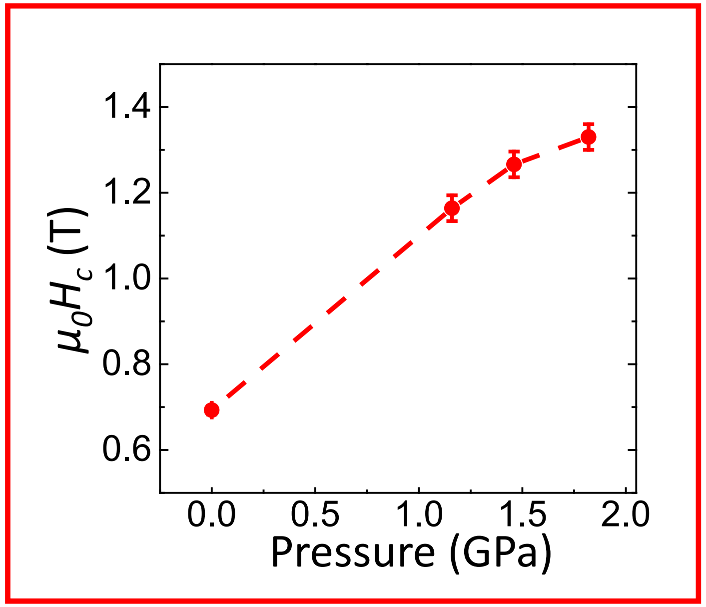

Pressure = 1 GPa



# Enhance AFM interlayer coupling?

**T. Song** et al., *Nat. Mater.* (2019).
T. Li et al., *Nat. Mater.* (2019).

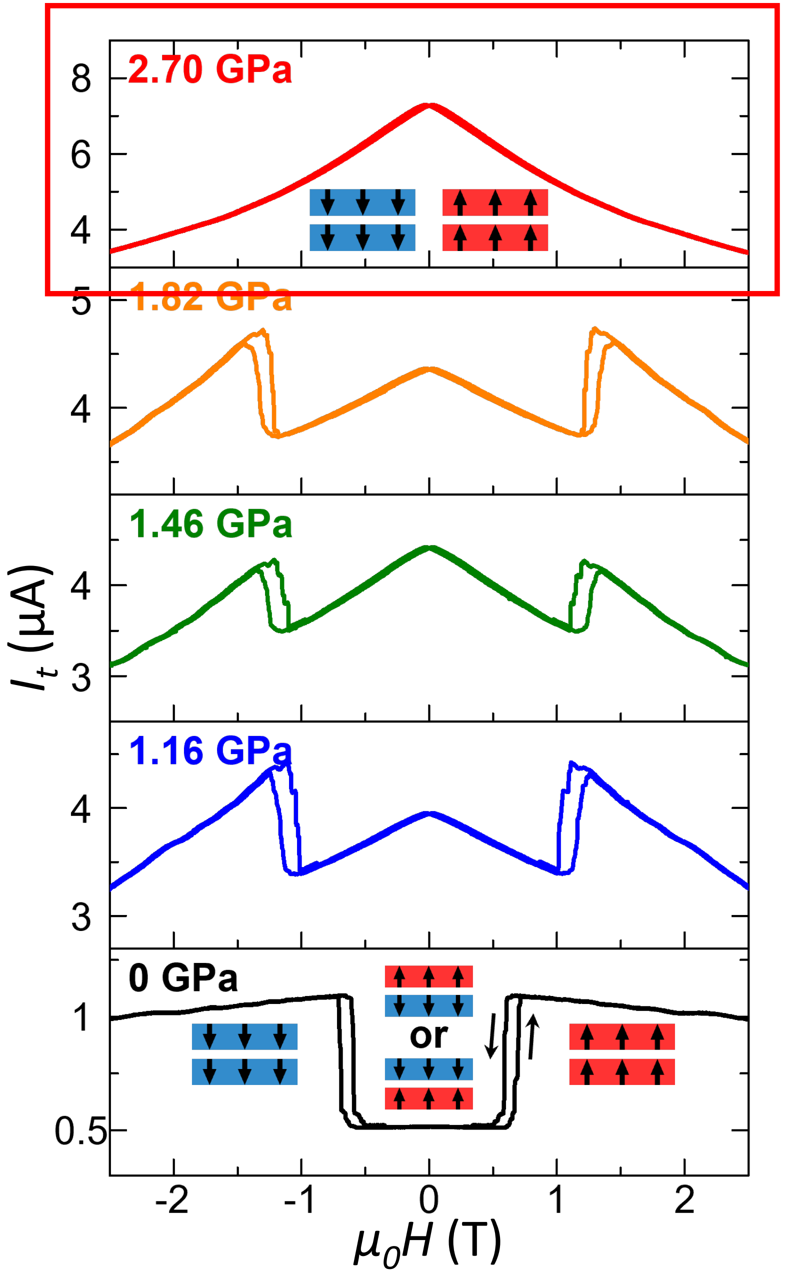


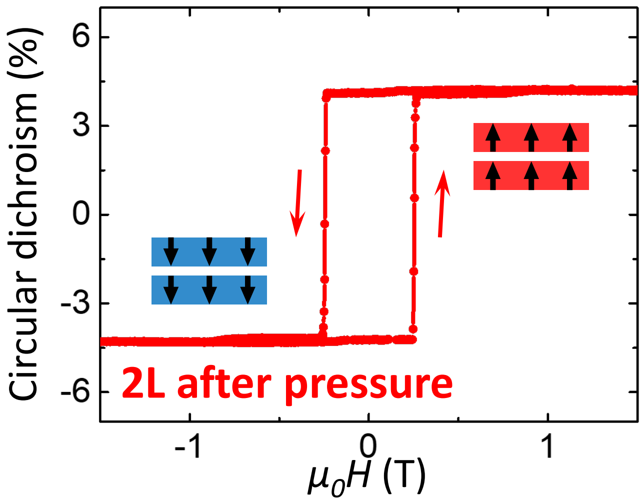
# "Bonus" discovery

**T. Song** et al., *Nat. Mater.* (2019).
T. Li et al., *Nat. Mater.* (2019).

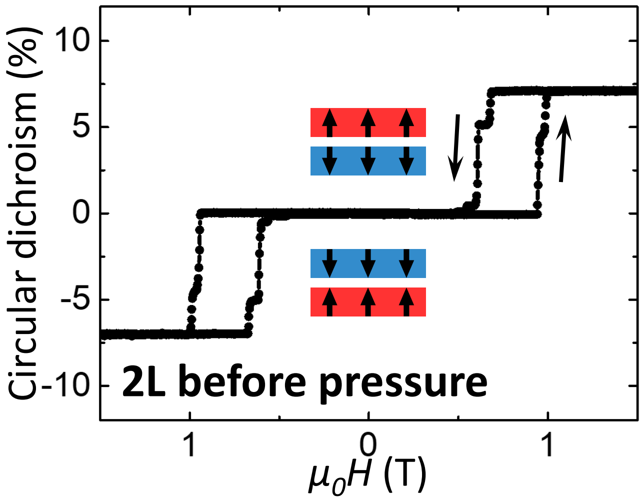
Higher pressure → **absence of AFM** → **FM**



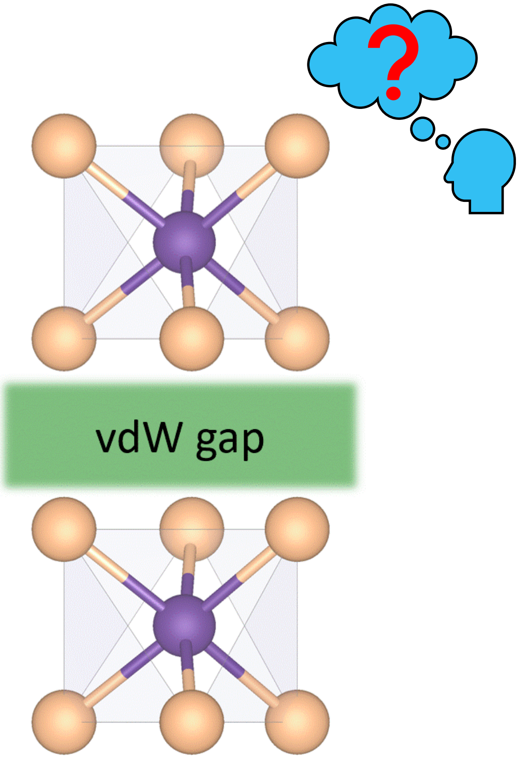

**Ferromagnetic**




**Antiferromagnetic**



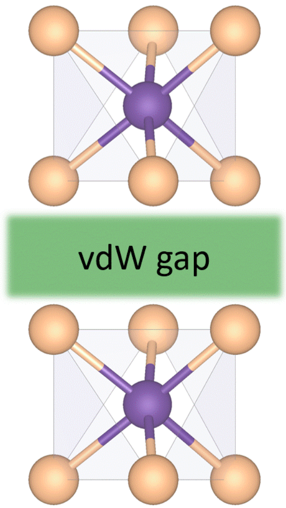

Lateral interlayer **shift**



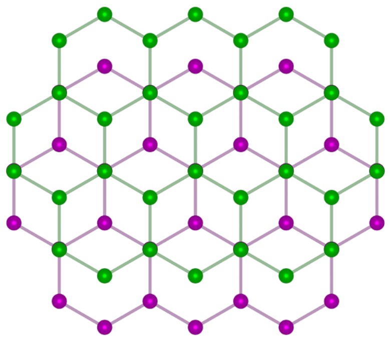
# Mystery solved

T. Song et al., *Nat. Mater.* (2019).
T. Li et al., *Nat. Mater.* (2019).

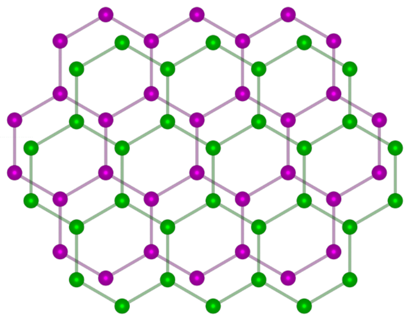
## Layer stacking identified by Raman spectroscopy

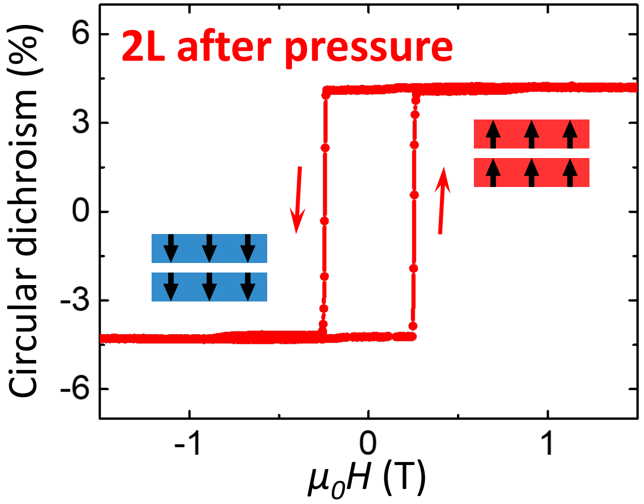


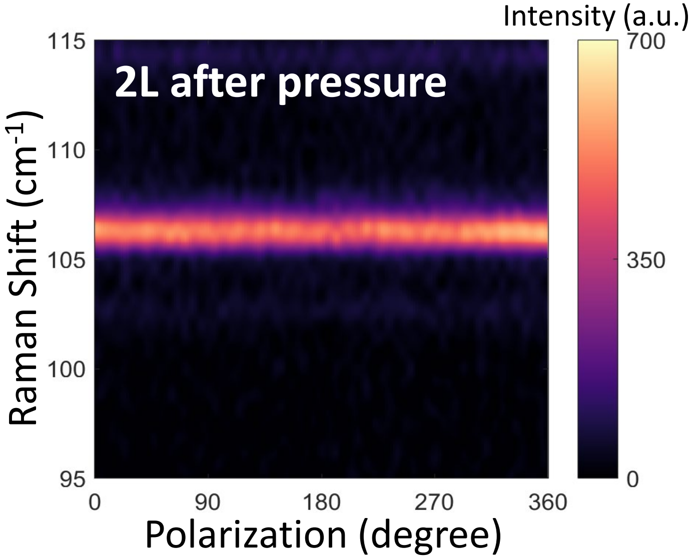

**Rhombohedral**

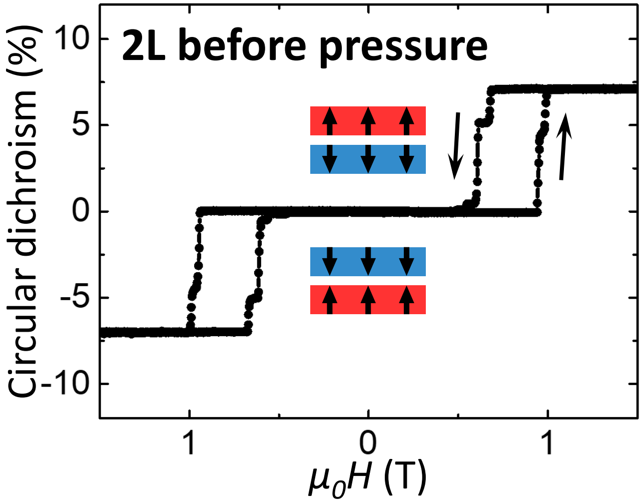


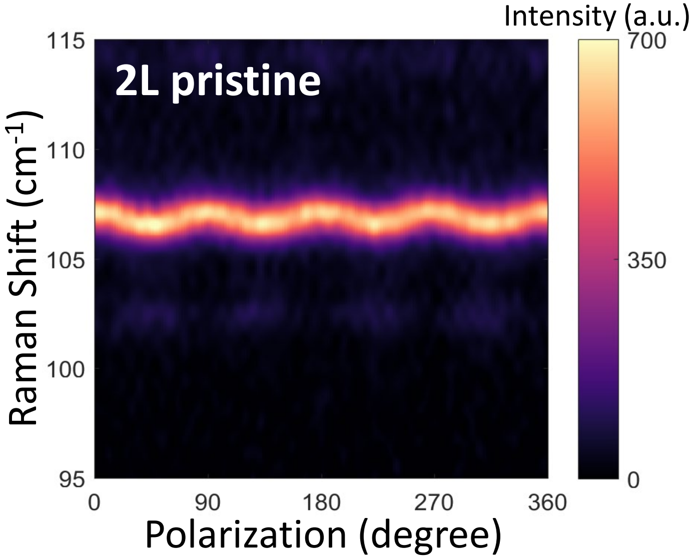
**Monoclinic**





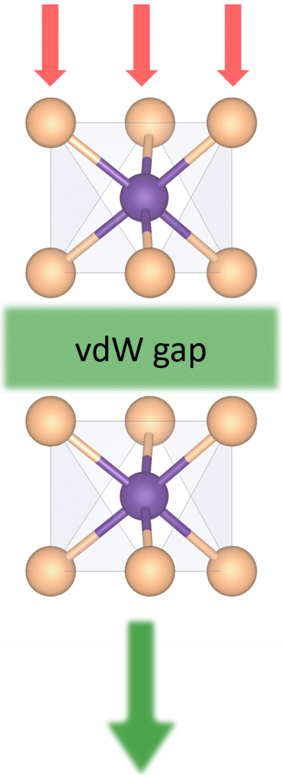



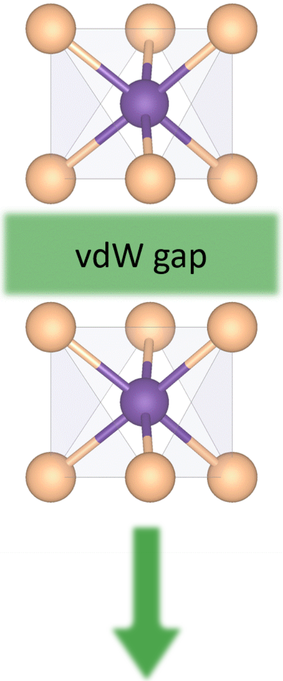
# Summary

Reduce interlayer **spacing**



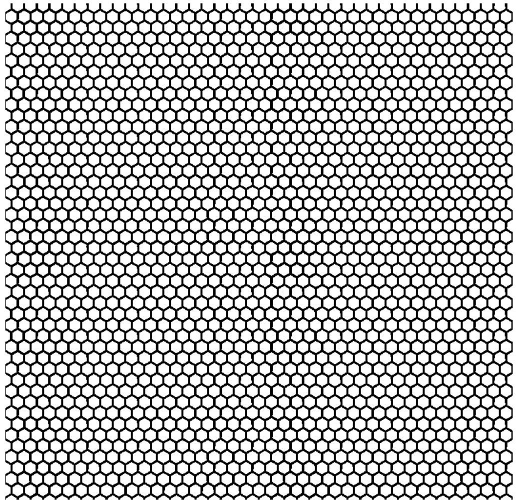

**Enhanced** AFM coupling

Lateral interlayer **shift**




**AFM → FM**

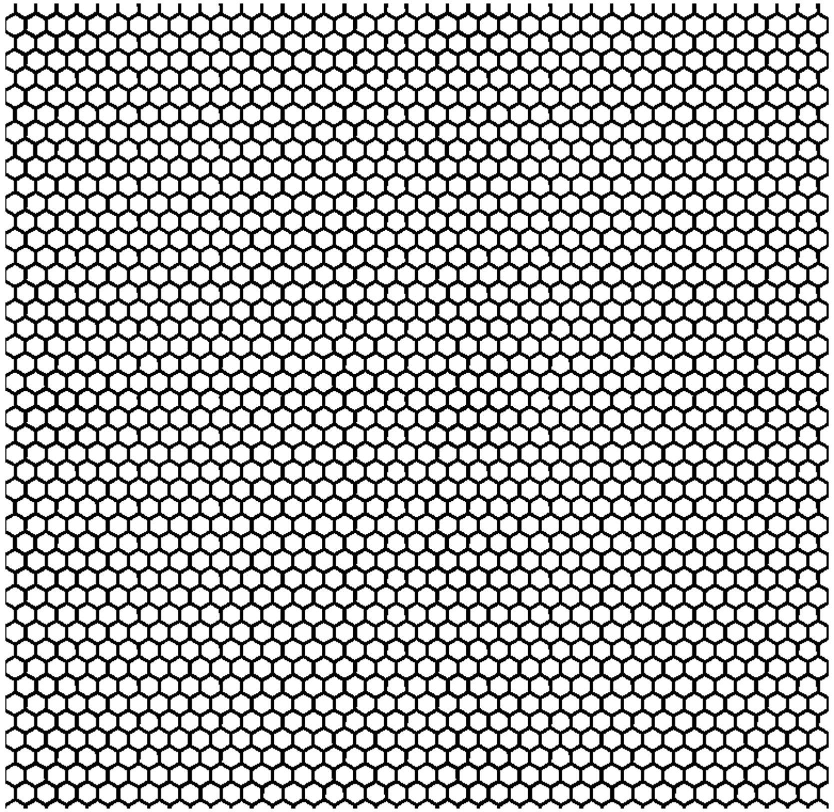
**Twist** two layers



**?**

# Moiré superlattices

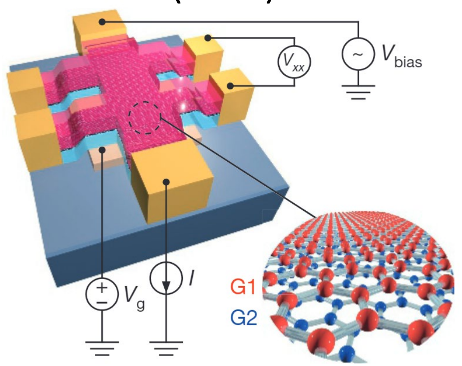
Twist two layers → moiré superlattice



Moiré in everyday life

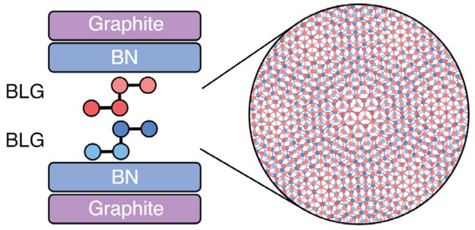
# Emergent phenomena in moiré superlattices

## Twisted bilayer graphene (tBLG)



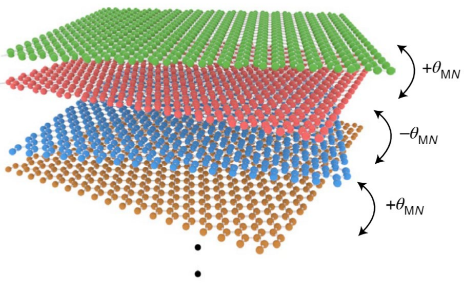

Jarillo-Herrero Group

## Twisted double bilayer graphene (2+2)



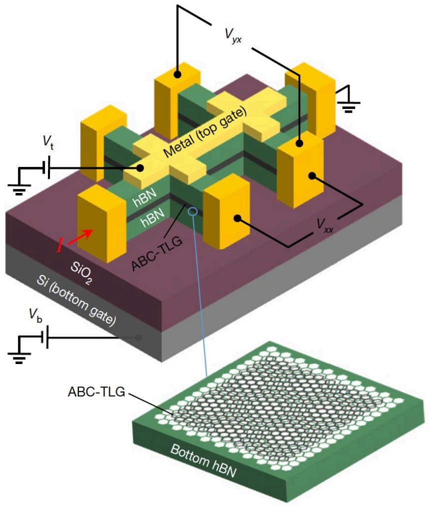

Jarillo-Herrero, Kim, Xu Group

## Twisted multilayer graphene (1+1+1…)



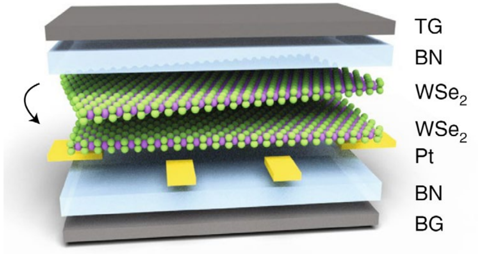

Jarillo-Herrero, Nadj-Perge, Tutuc Group

## Aligned graphene/hBN



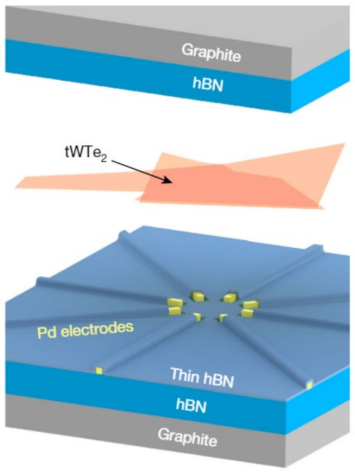

Feng, Young, Dean, Kim Group

## Twisted bilayer $WSe_2$



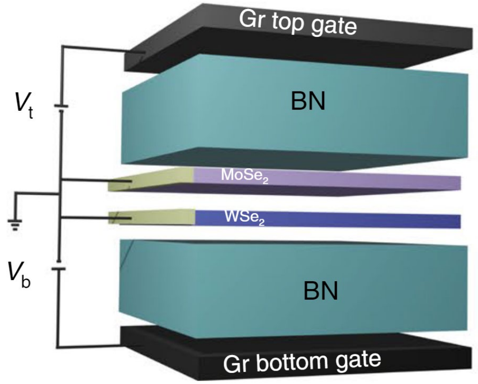

Dean, Pasupathy Group

## Twisted bilayer $WTe_2$



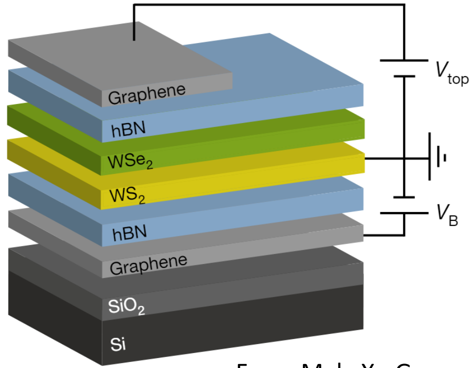

Wu Group

## Twisted $WSe_2/MoSe_2$




Xu, Li, Feng Group

## Twisted $WSe_2/WS_2$

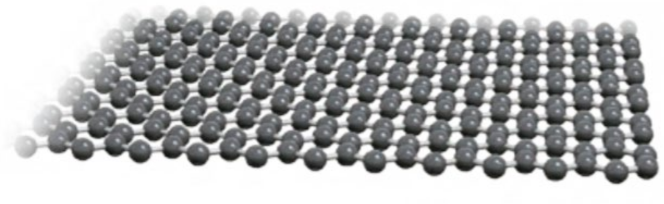



Feng, Mak, Xu Group

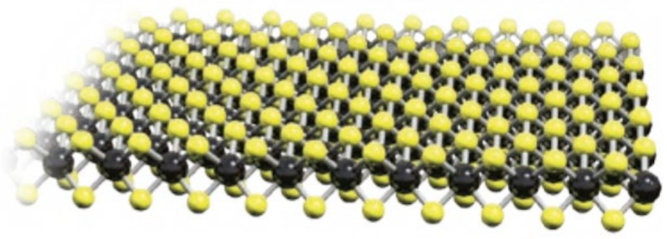
**and many more…**

# Emergent phenomena in moiré superlattices

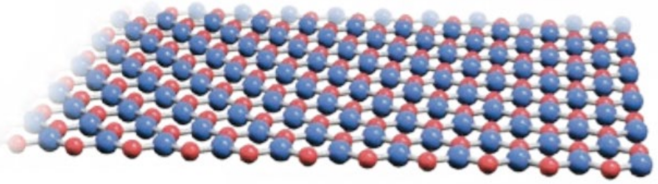
Based on **non-magnetic** materials



Graphene



Dichalcogenides

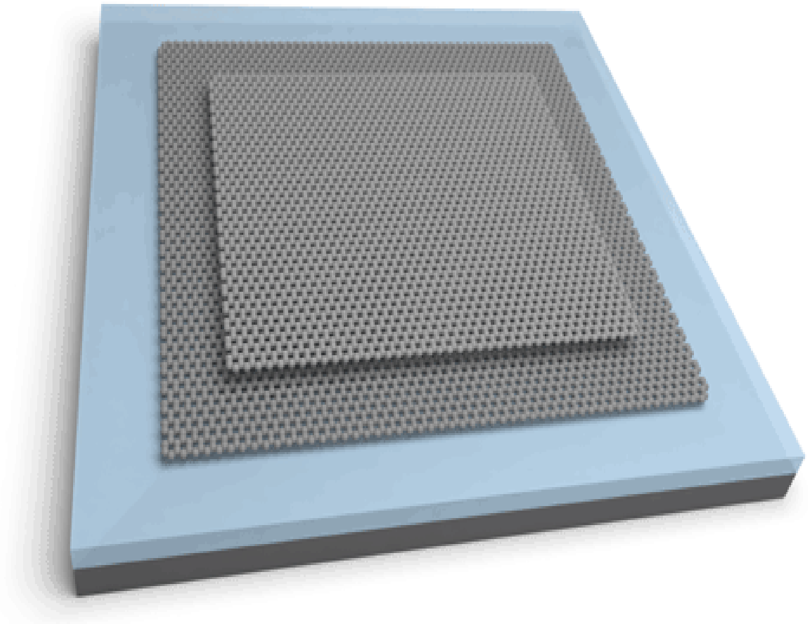


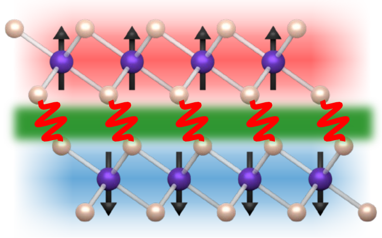
Boron nitride

**What about moiré superlattices formed by twisting 2D magnets**

# Unique opportunity: twist → magnetic moiré

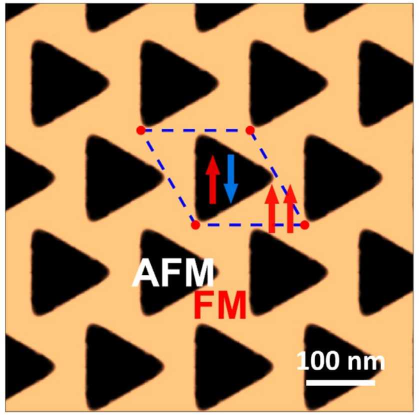
**Twisted** 2D magnet

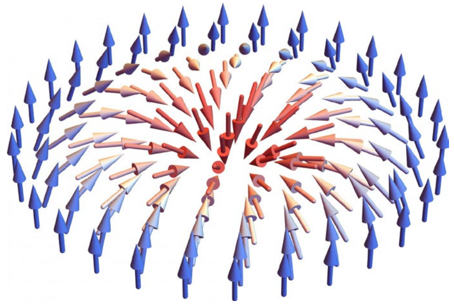


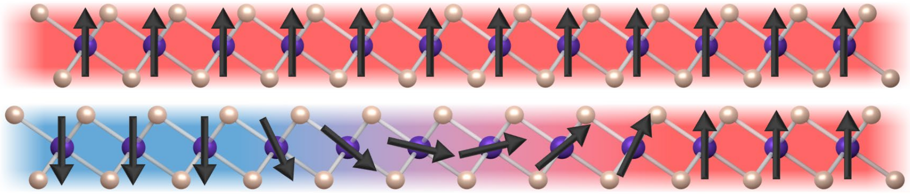


**Atomic registry**

**Nanoscale magnetic moiré**



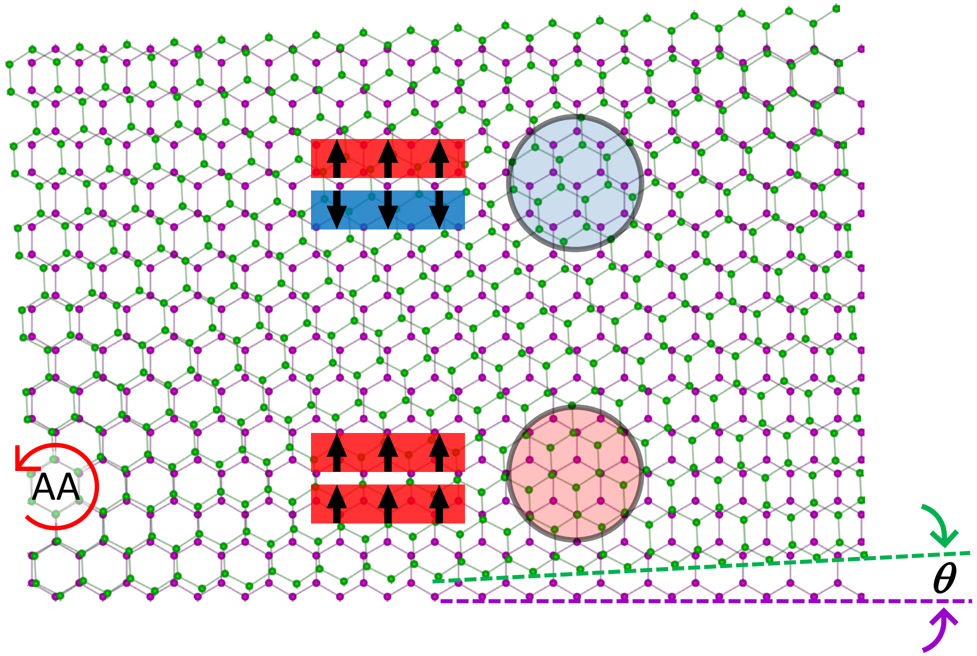


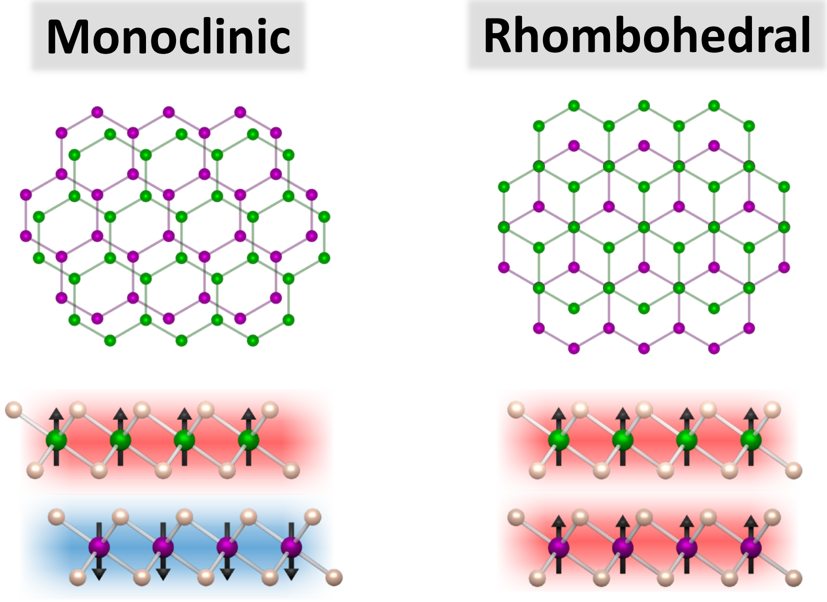
New pathway towards **nanoscale magnetic textures** and **new spintronic devices**

# Unique opportunity: twist → magnetic moiré

**T. Song** et al., *Science* (2021).

Stacking moiré → magnetic moiré



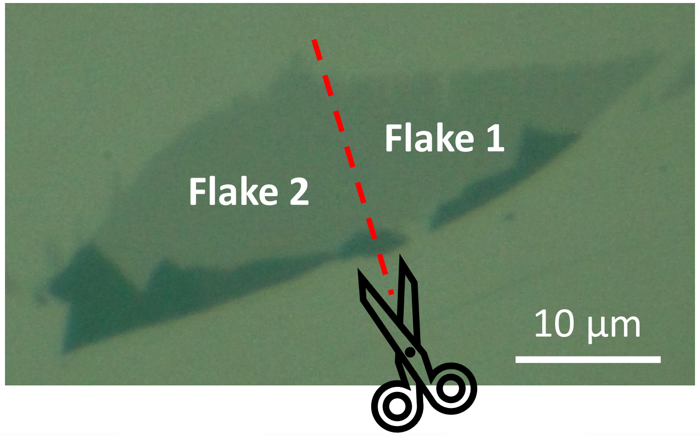



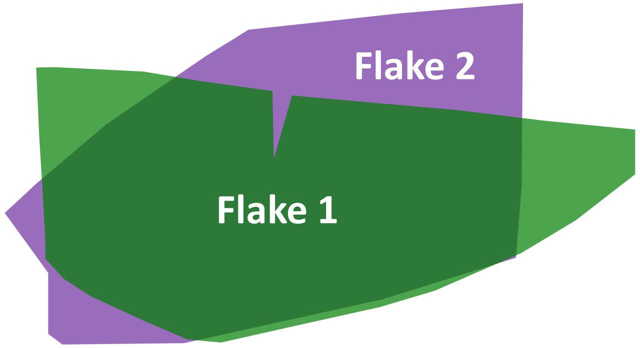
# Twisted 2D magnet

**T. Song** et al., *Science* (2021).

## "Tear and stack" technique

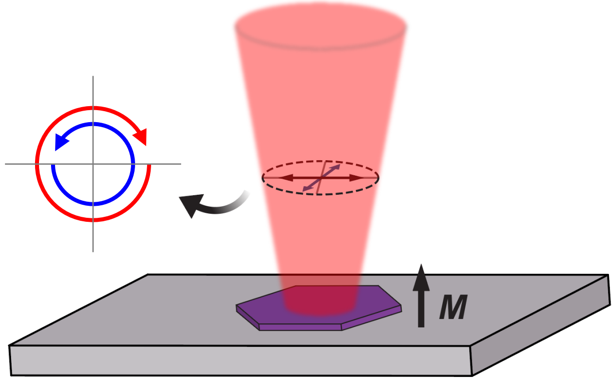
Tutuc Group



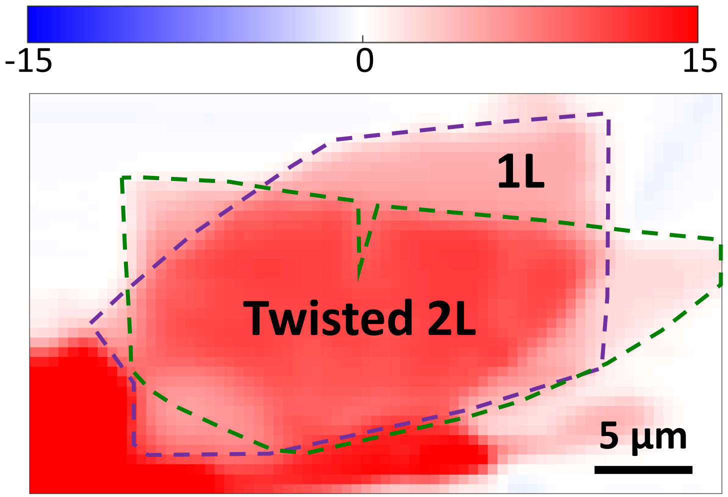



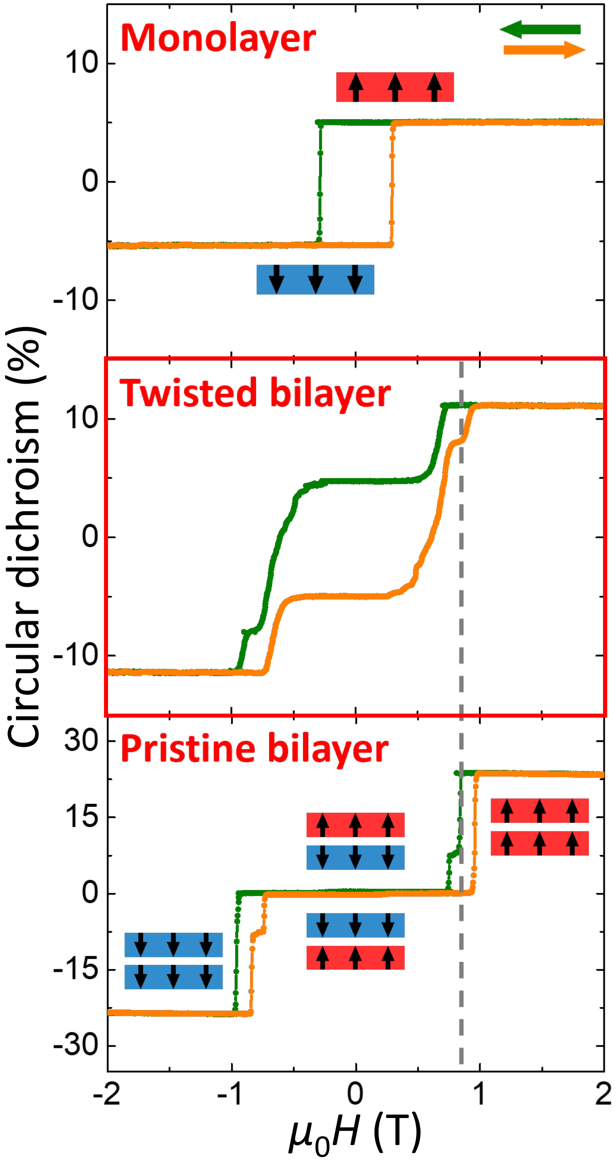

**Tear** → **Rotate** → **Stack**

## Circular dichroism microscopy




## Coexisting AFM and FM interlayer coupling



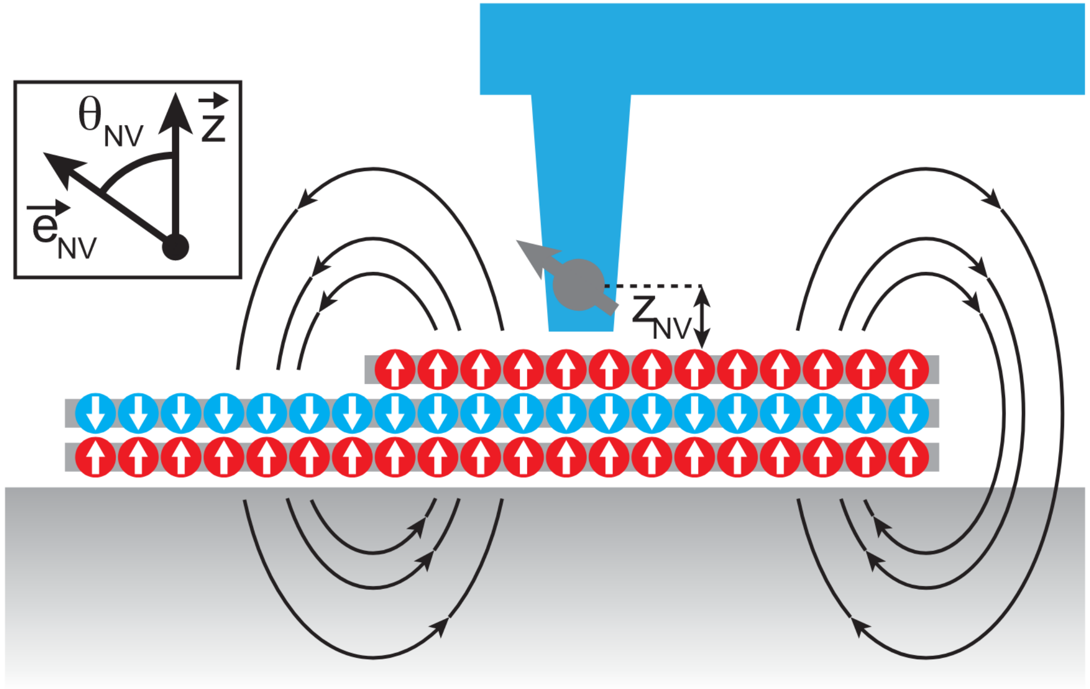
# Scanning NV magnetometry

**Probing magnetism in 2D materials at the nanoscale with single-spin microscopy**

L. Thiel[1], Z. Wang[2,3], M. A. Tschudin[1], D. Rohner[1], I. Gutiérrez-Lezama[2,3], N. Ubrig[2,3], M. Gibertini[2,4], E. Giannini[2], A. F. Morpurgo[2,3], P. Maletinsky[1*]

Spatial resolution
**~50 nm**

Monolayer $CrI_3$ magnetization
**~15 $\mu_B$/nm$^2$**



# Magnetic moiré!

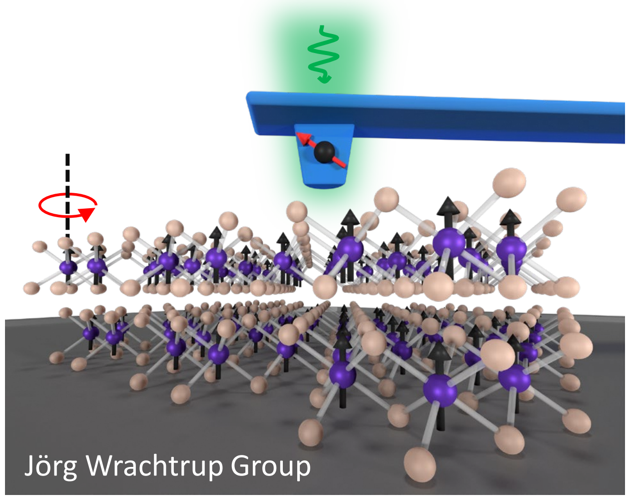
T. Song et al., *Science* (2021).

Periodic **AFM-FM** domains

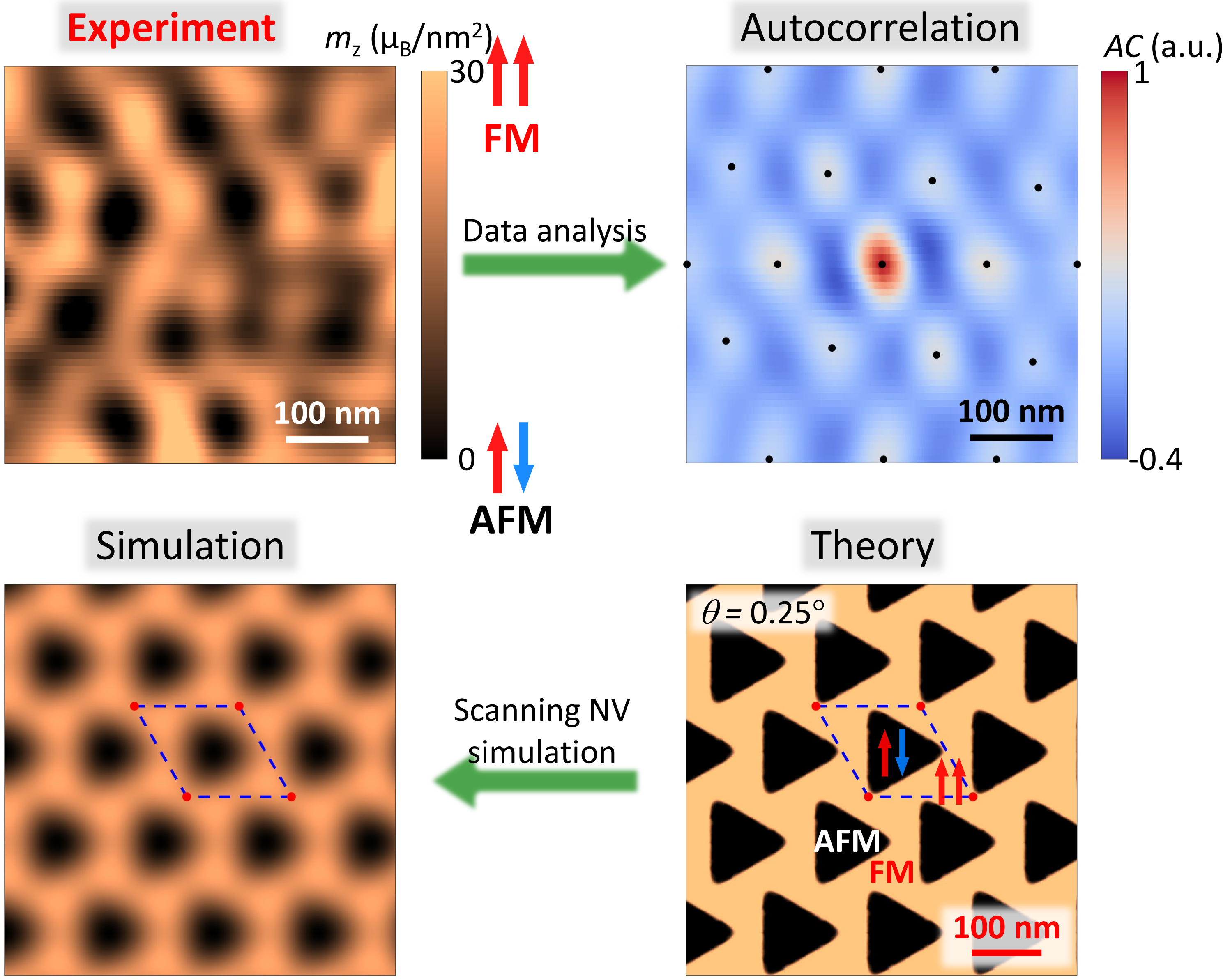
Agree well with the simulation

Moiré periodicity (~150 nm)

Clear **six-fold symmetry**

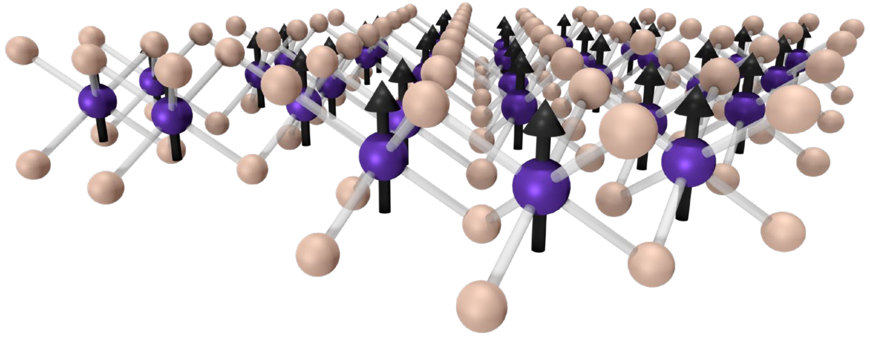



Spatial resolution **~50 nm**



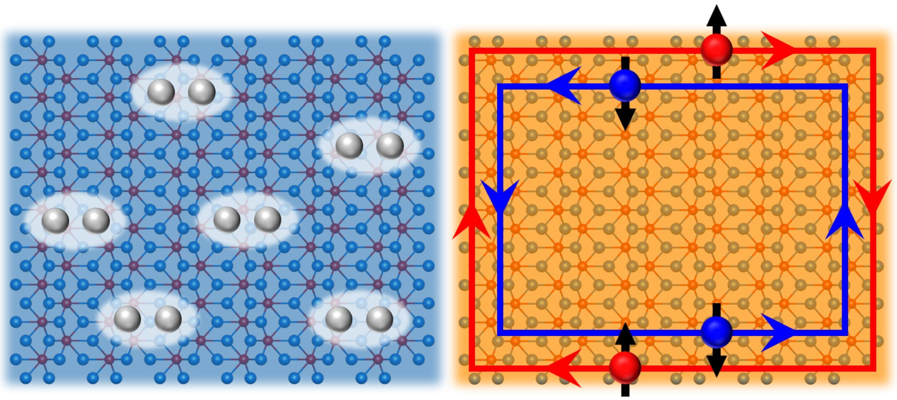
# Outline: two examples

## 2D magnet



- Discovery of **2D magnets**
- Van der Waals **spintronics**
- **Layer stacking**-dependent magnetism
- Twisted 2D magnets → **magnetic moiré**

## 2D superconductor + topological insulator

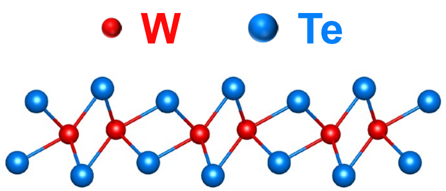


- 2D **topological insulator**
- Gated-tunable **2D superconductivity**

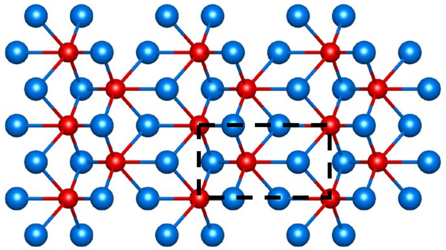
# Many faces of tungsten ditelluride ($WTe_2$)

## Monolayer tungsten ditelluride (Td-$WTe_2$)

Side view

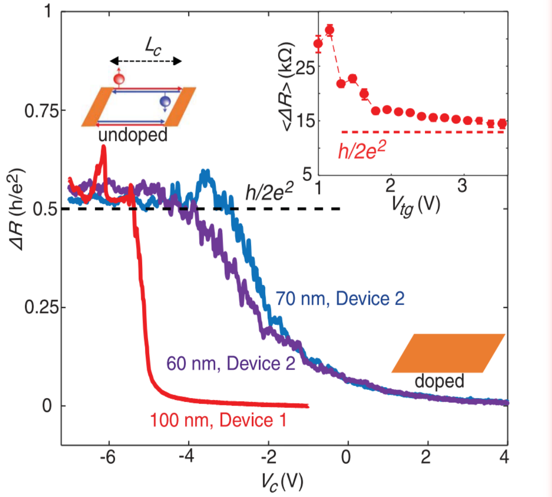



Top view



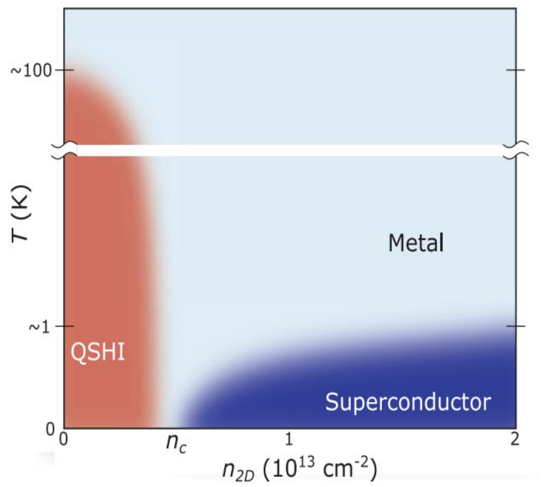
Natural multiple phases coexist in a single material


### 2D topological insulator (quantum spin Hall insulator)



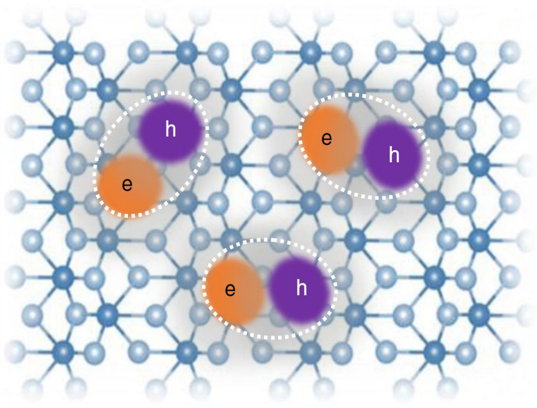

S. Wu et al., *Science* (2018).
Z. Fei et al., *Nat. Phys.* (2017).

### Superconductivity



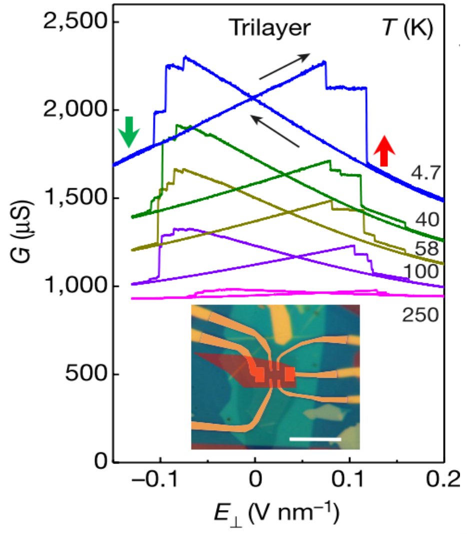

V. Fatemi et al., *Science* (2018).
E. Sajadi et al., *Science* (2018).

### Excitonic insulator




Y. Jia et al., *Nat. Phys.* (2022).
B. Sun et al., *Nat. Phys.* (2022).

### Ferroelectricity

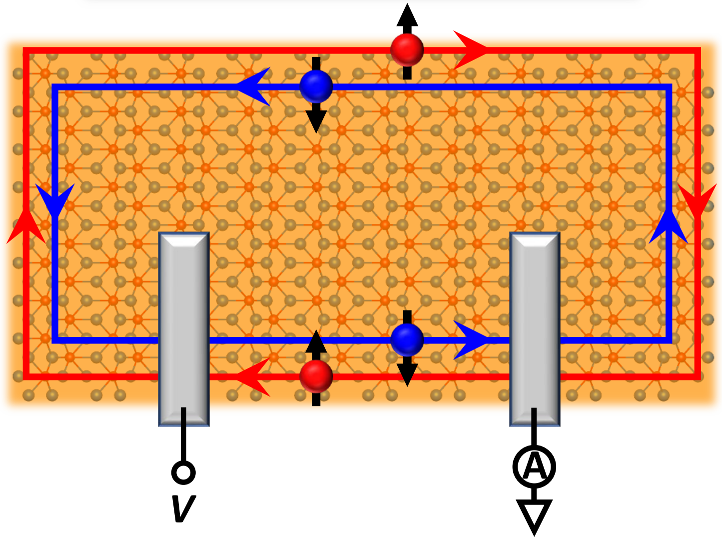


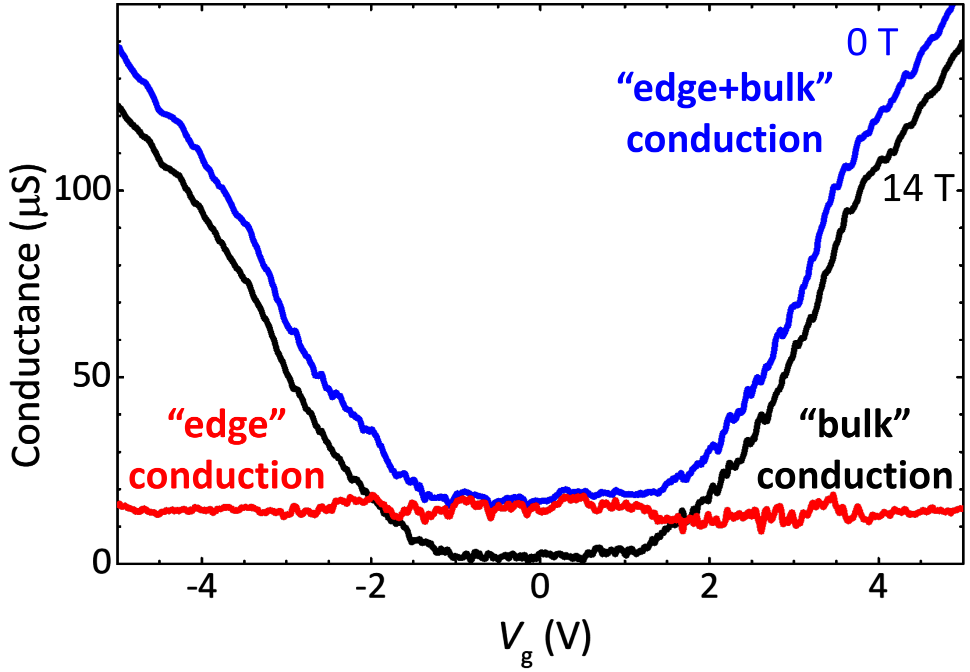

Z. Fei et al., *Nature* (2019).

# 2D topological insulator

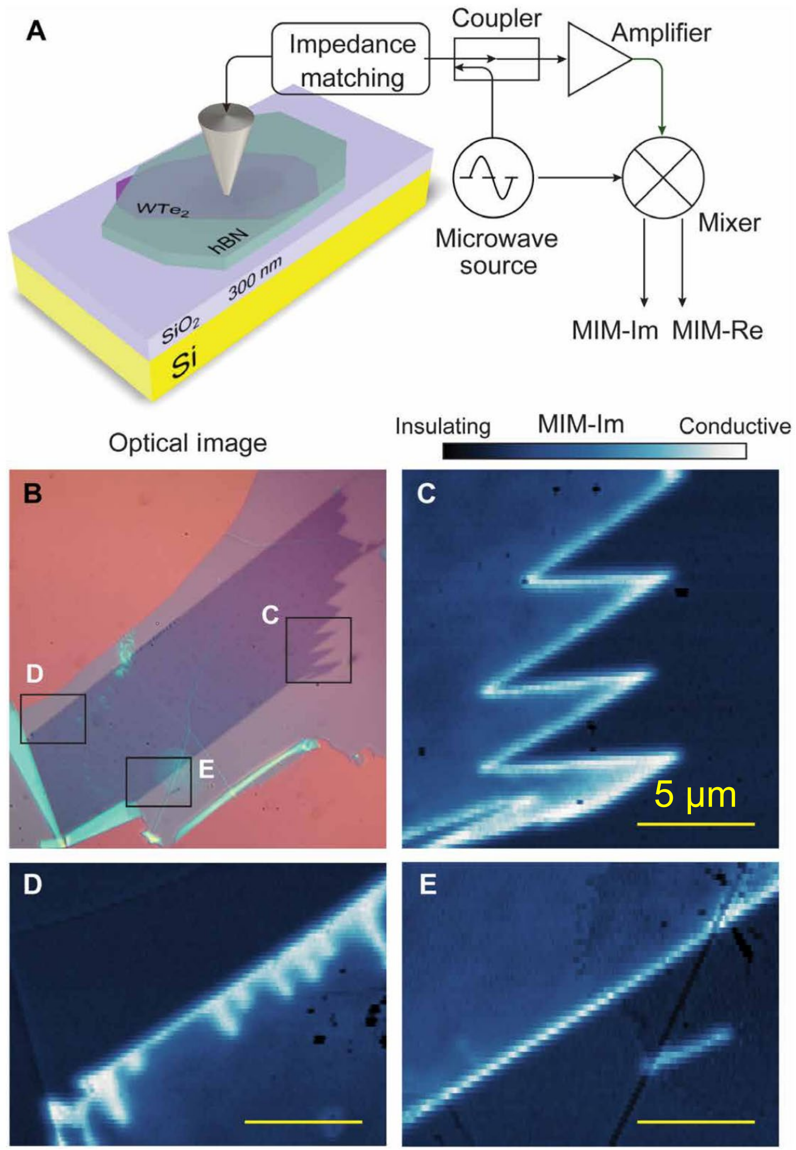
Z. Fei et al., *Nat. Phys.* (2017).
S. Wu et al., *Science* (2018).
Y. Shi et al., *Sci. Adv.* (2019).

## Helical edge modes




## Real-space imaging of 2D TI



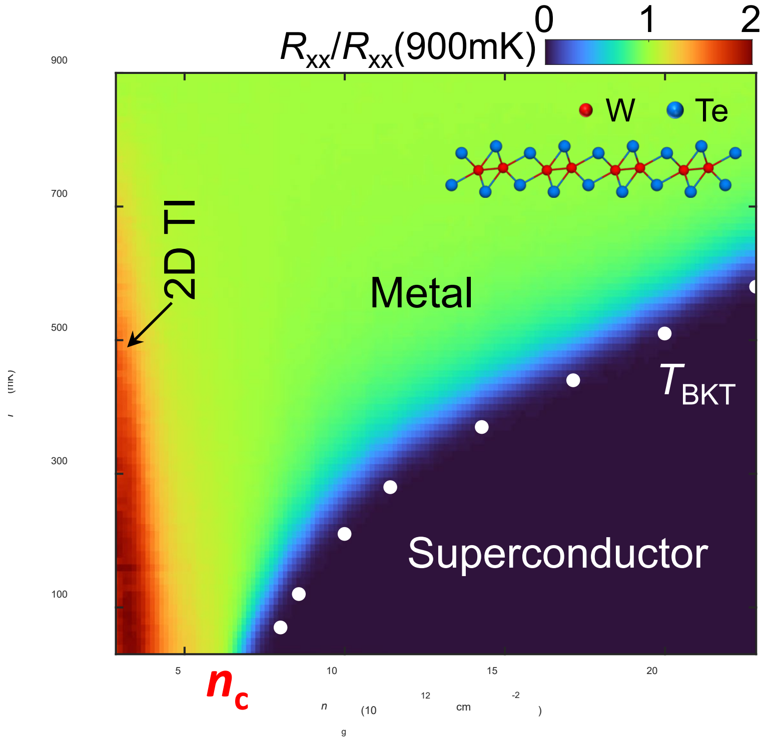
# A big surprise: 2D superconductivity

V. Fatemi et al., *Science* (2018).
E. Sajadi et al., *Science* (2018).
Song et al., *Nat. Phys.* (2024).

**Topology** | **Superconductivity**

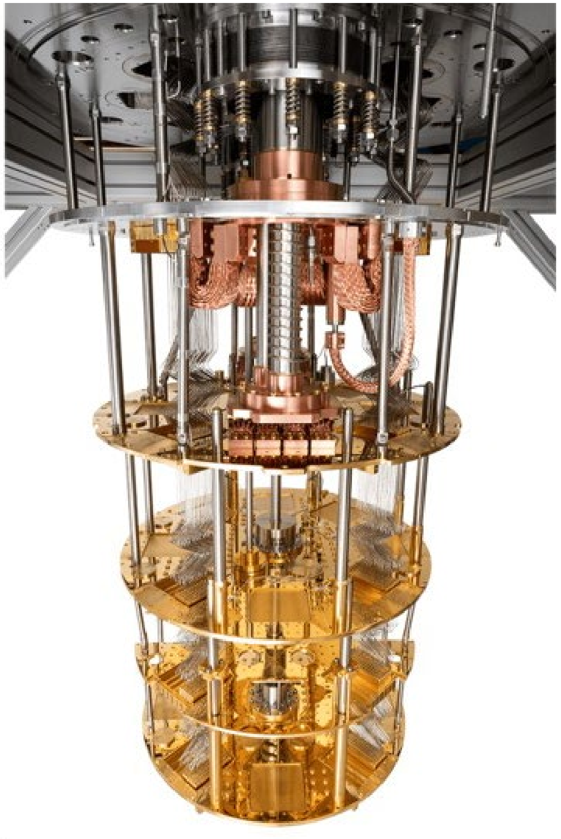
"Insulator" | "Metal"

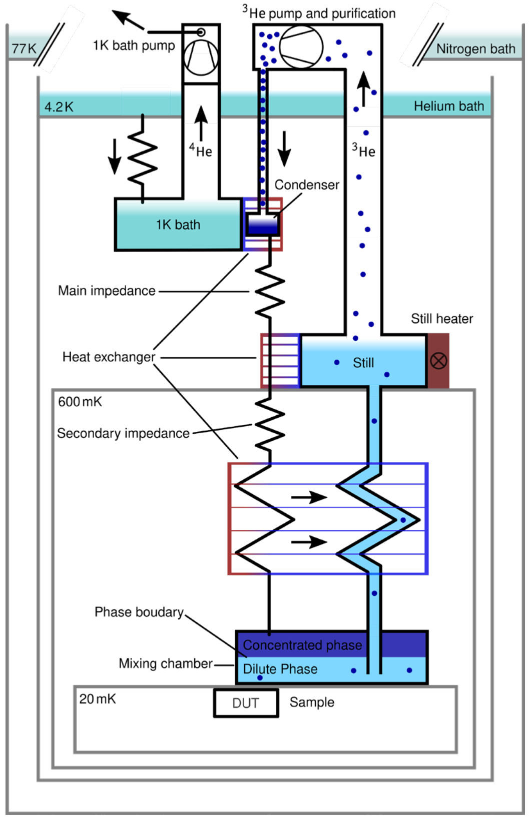
Low carrier density | High carrier density




## Dilution refrigerator

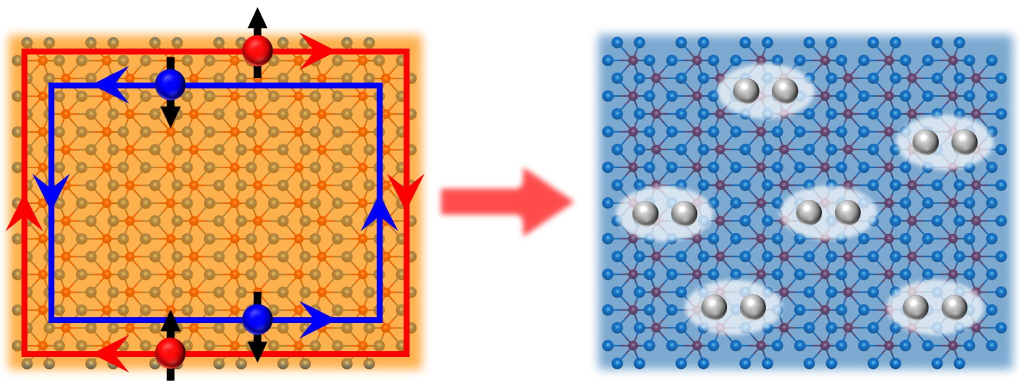
- Base temperature 8 mK
- 9T-1T-1T vector magnet



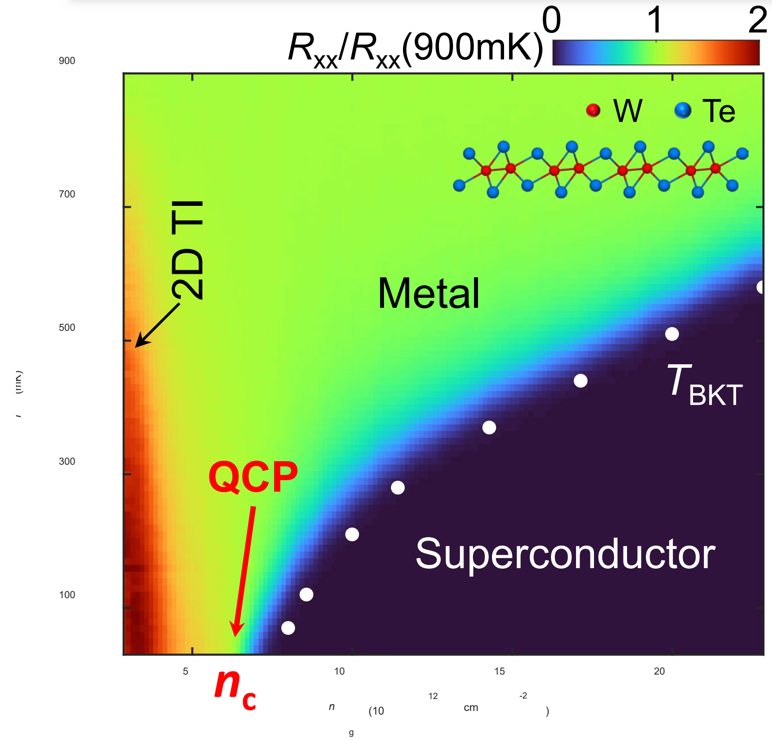


# Surprising 2D superconductivity

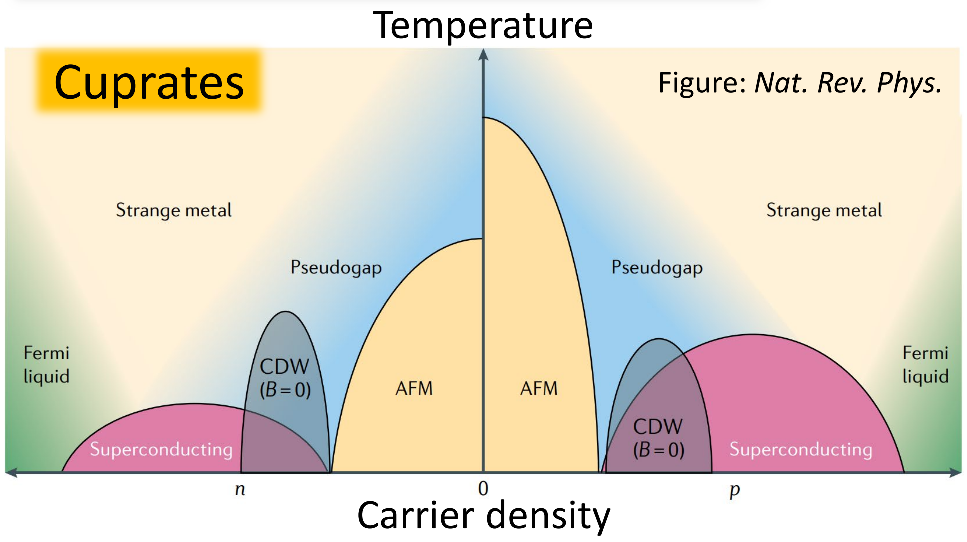
V. Fatemi et al., *Science* (2018).
E. Sajadi et al., *Science* (2018).
Song et al., *Nat. Phys.* (2024).

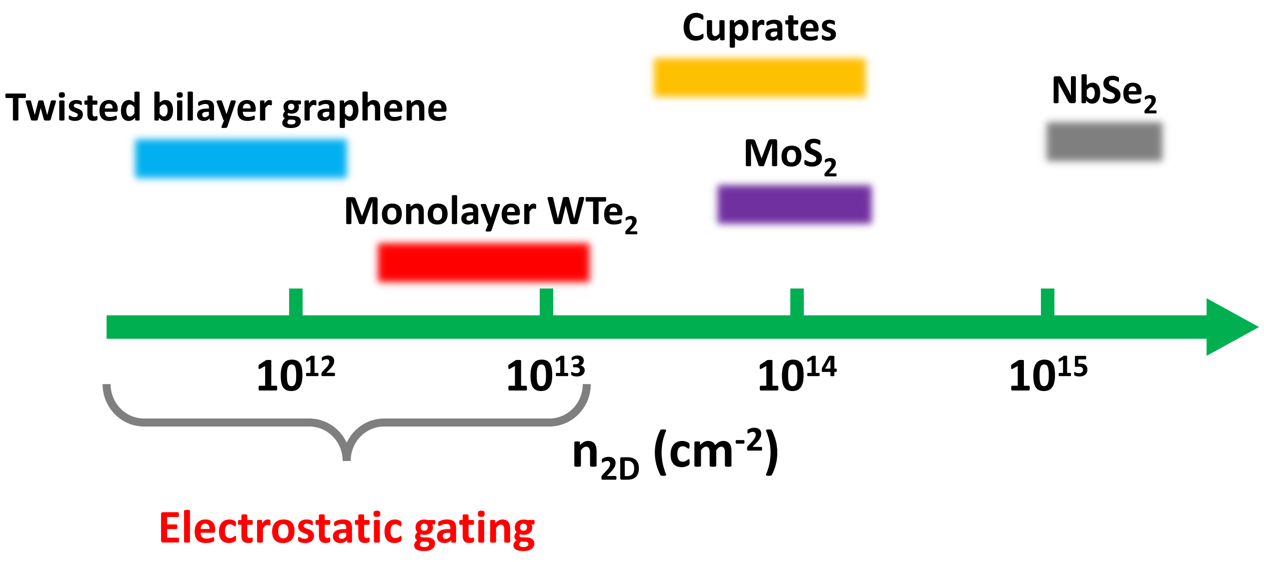


## Electronic phase diagram




## Unconventional superconductors

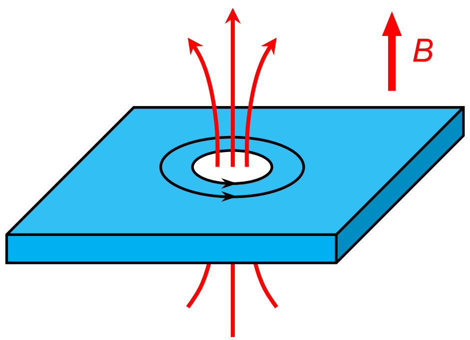


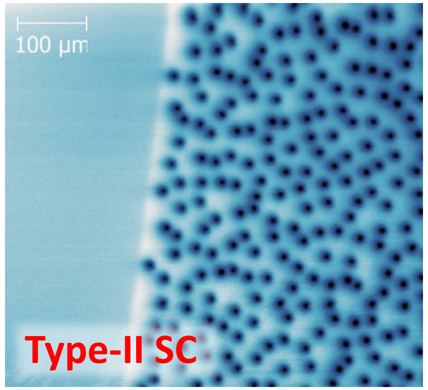



# A new sensitive probe: vortex Nernst effect

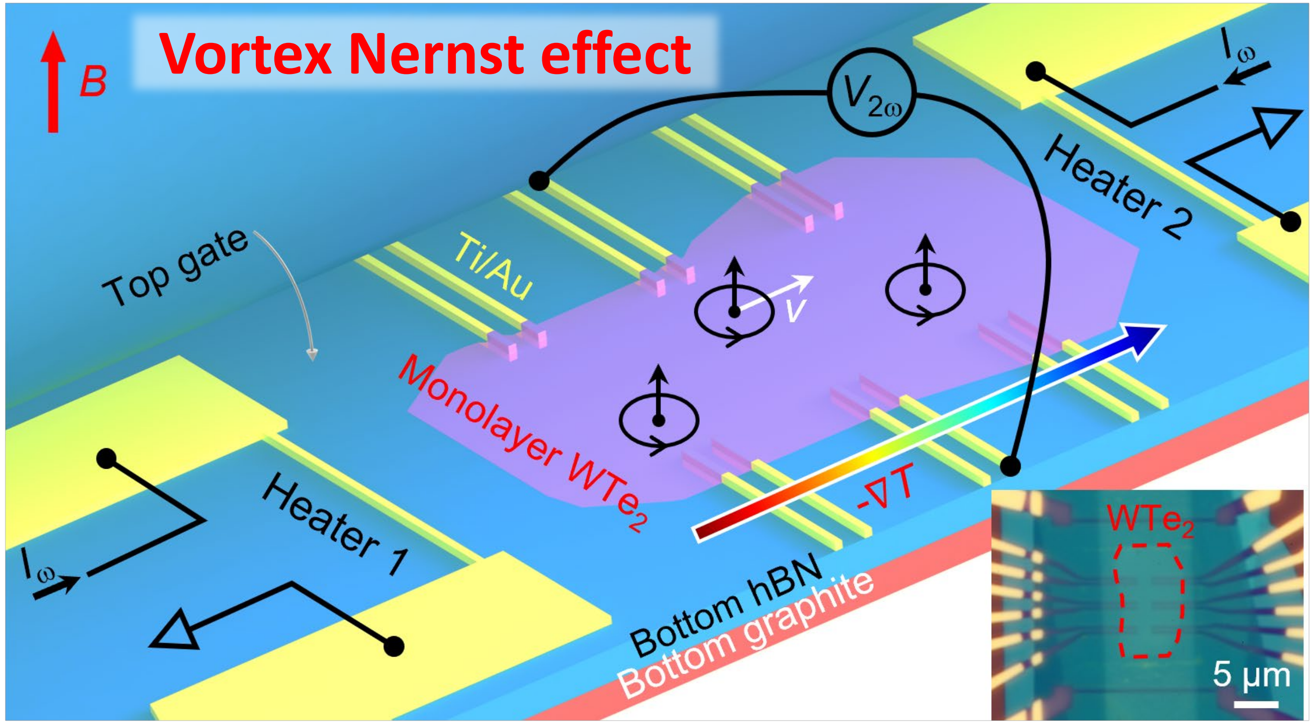
**T. Song** et al., *Nature Physics* (2024).

What is a vortex?



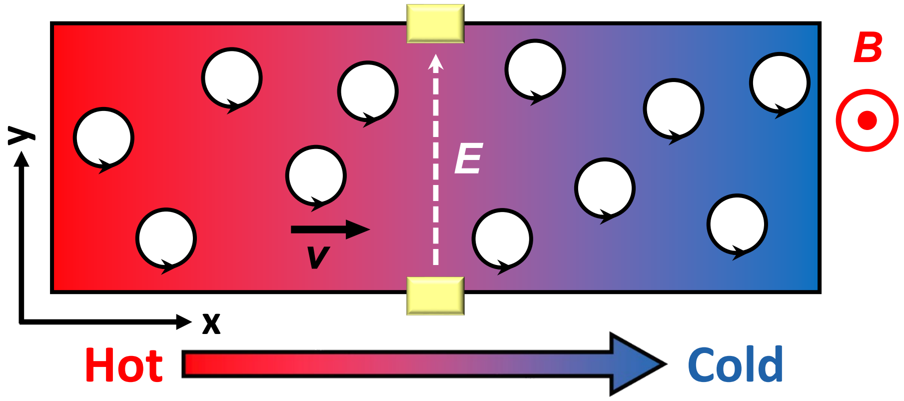

F. Wells et al., *Sci. Rep*. (2014).




Electrical transport → resistance

**Nernst signal → vortex motion**

VdW engineering of -∇*T*
Dual-heater geometry → Down to **~45 mK**

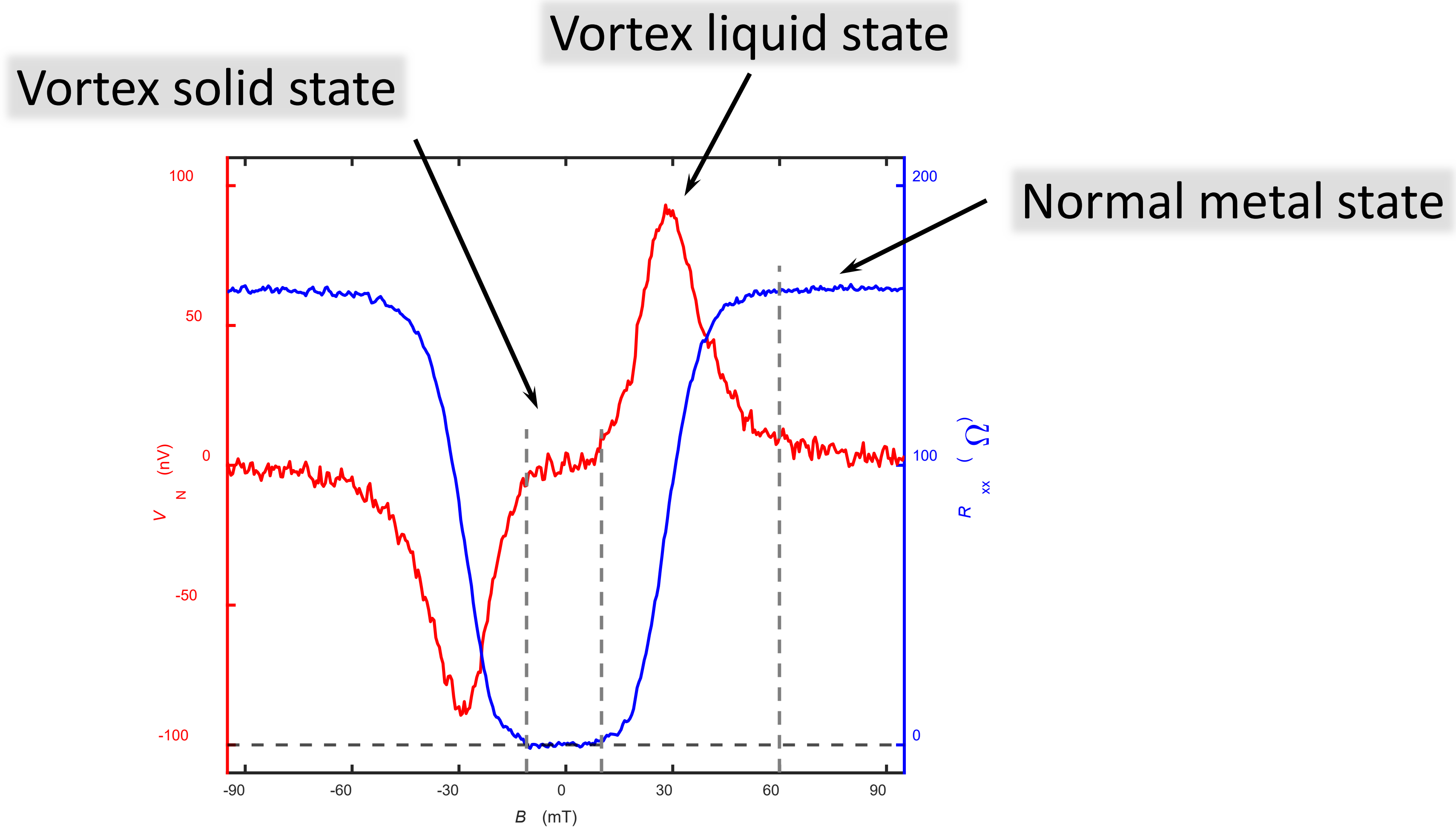



- Vortices are "pin-holes" in the superfluid
- -∇*T* → flow of vortices → phase slippage
- Josephson effect → voltage (Nernst signal)

N. P. Ong et al., *PRB* (2006).

# Vortex Nernst effect

**T. Song** et al., *Nature Physics* (2024).



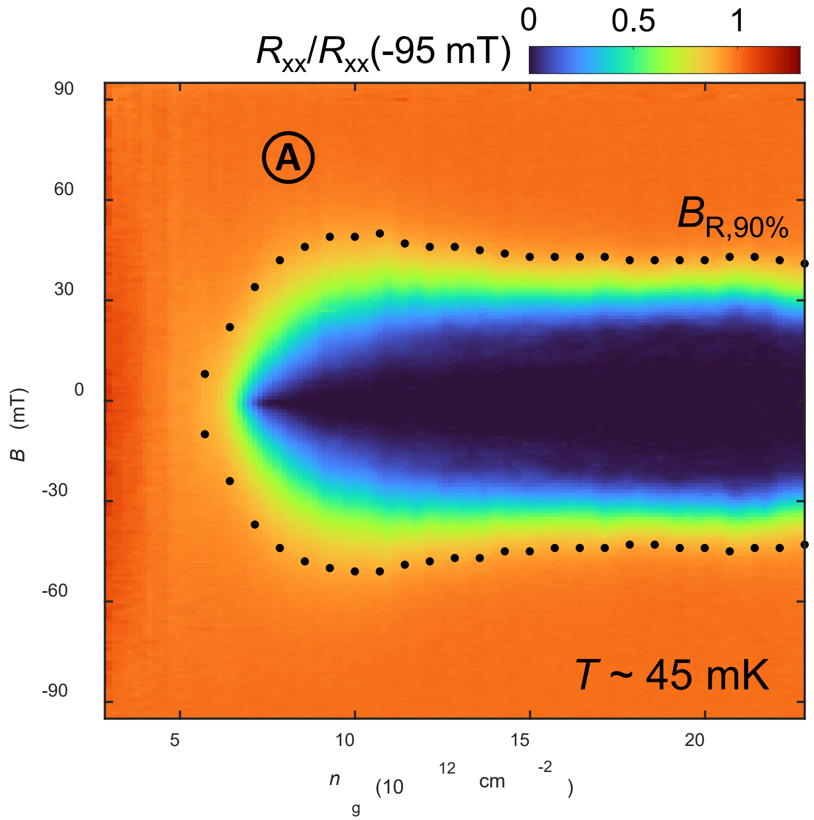

**$V_N$ detects mobile vortices (superconducting fluctuations)**

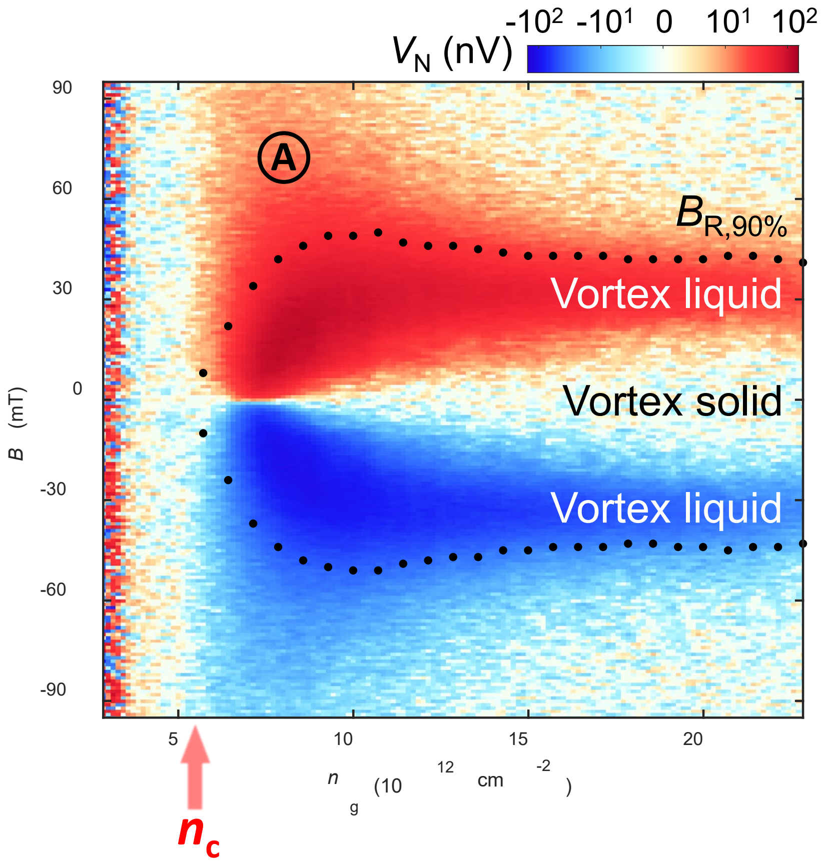
# Resistance vs Vortex Nernst

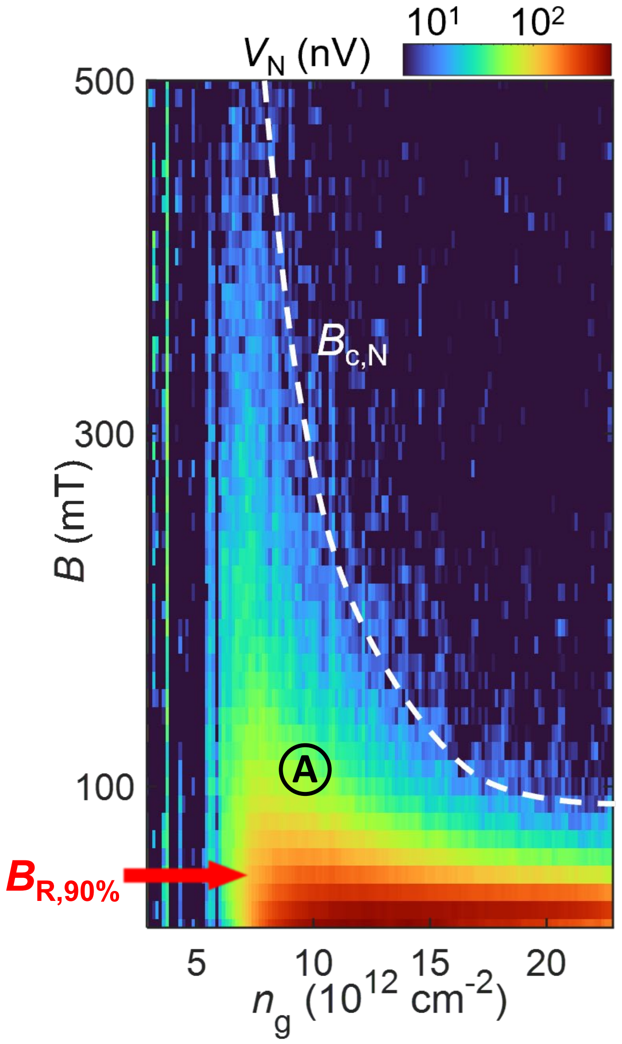
**T. Song** et al., *Nature Physics* (2024).

- **Direct comparison** between resistance and vortex Nernst
- Vortex Nernst **survives well above $B_{R,90\%}$ (?)**

**High magnetic field**



# Resistance vs Vortex Nernst

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Superconducting order parameter $\boldsymbol{\Psi}(\vec{r}) = |\boldsymbol{\Psi}(\vec{r})|e^{i\theta(\vec{r})}$

Amplitude — Phase

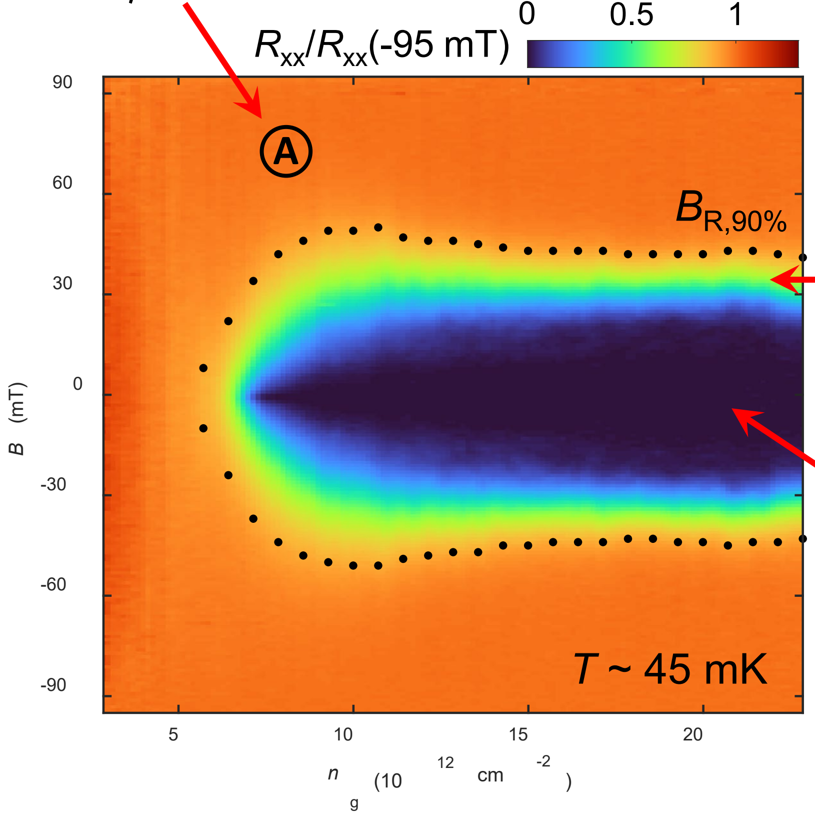
| | Amplitude | Phase |
|---|---|---|
| Resistance | ✓ | ✓ |
| Nernst | ✓ | ✗ |

Phase fluctuations

$\langle e^{i\theta(\vec{r})} \rangle = zero$

Pairing amplitude survives

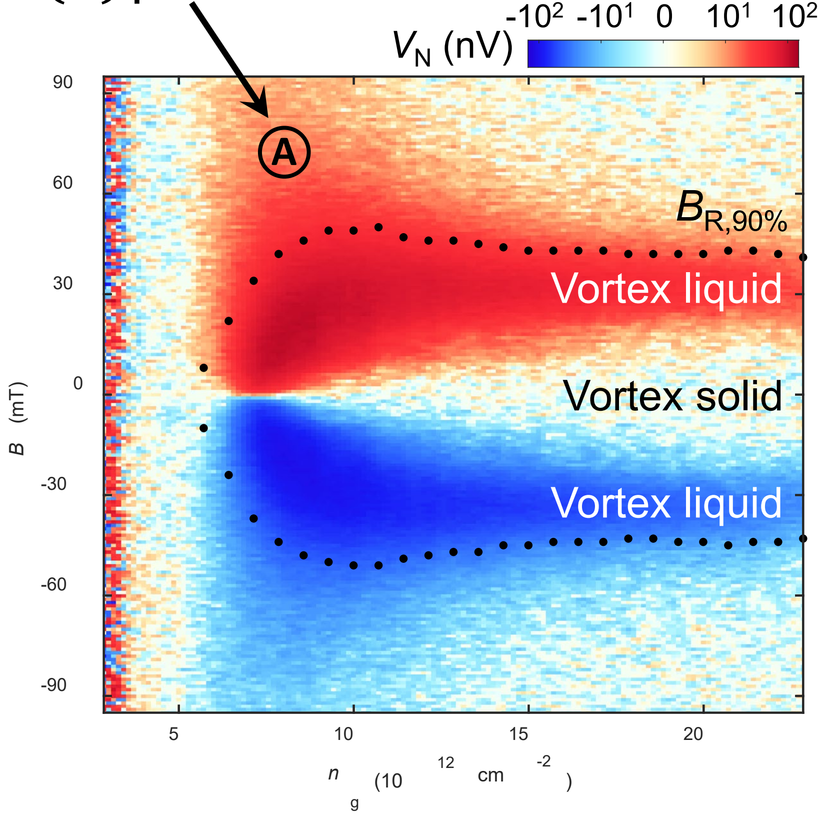
$|\boldsymbol{\Psi}(\vec{r})| \neq zero$




Destroyed by mobile vortices

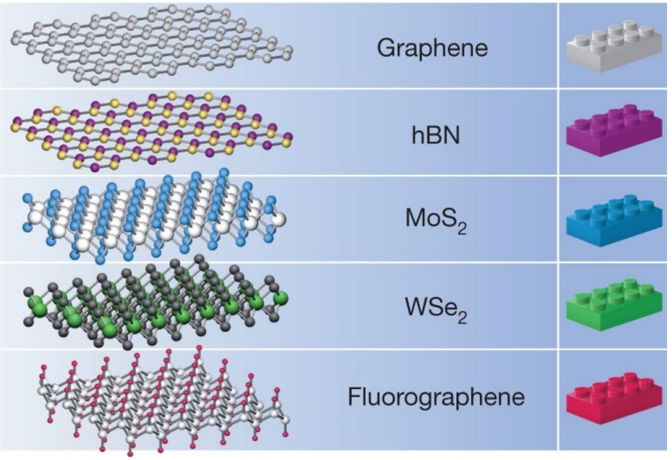
Long-range phase coherence

$e^{i\theta(\vec{r})} = constant$

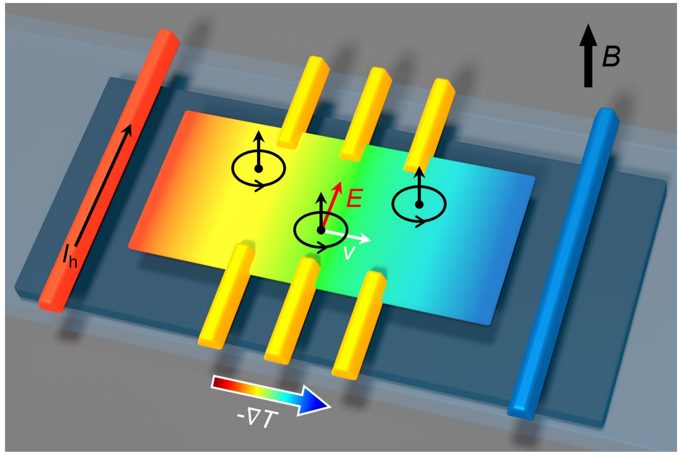


# New opportunities

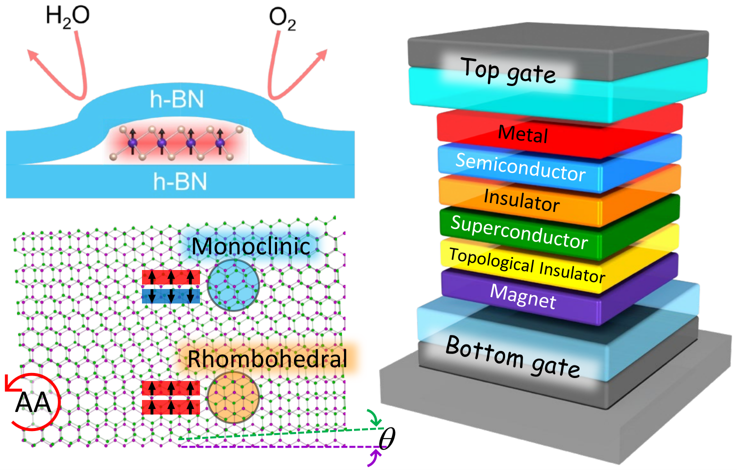
## "LEGO set" of 2D materials



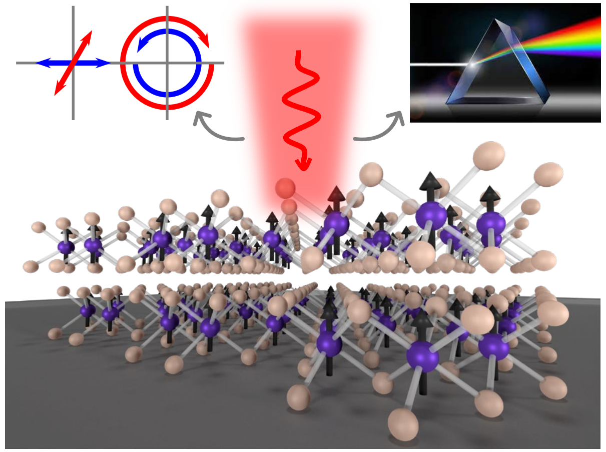

## Thermoelectricity & electrical transport




## Van der Waals fabrications




## Optical spectroscopies & microscopies



# Acknowledgements

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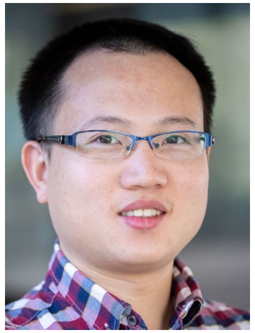
Princeton Physics

Dicke fellowship

## Thank you for your attention!



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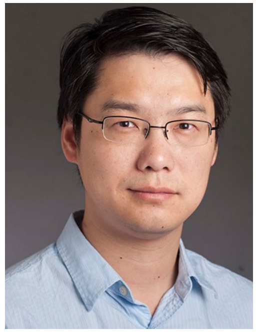

Di Xiao

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Michael McGuire

Experiment: Yanyu Jia, Pengjie Wang, Guo Yu, Yue Tang, Ayelet Uzan, Michael Onyszczak

$WTe_2$ crystal: Ratnadwip Singha, Xin Gui
hBN crystal: Kenji Watanabe, Takashi Taniguchi

Experiment: Qi-Chao Sun, Eric Anderson, Xinghai Cai, Zaiyao Fei

Theory: Ting Cao, Matisse Tu, Chong Wang, Jimin Qian

Pressure study: David Graf, Cory Dean, Matthew Yankowitz

# University of Wisconsin-Madison

- **Top public research** university founded in 1848 (Big Ten).
- UW-Madison Physics Program is **ranked #17** in U.S. News.
- Capital of the state, beautiful and safe city with **five lakes**.
- **High stipend** and guaranteed TA/RA position.

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- **174** Students
- **5.7** Years median time to graduation
- **5** Years guaranteed support as TA
- **51** Credits required to graduate
- **33%** International students
- **20%** Female students
- **4%** Targeted minorities

## UW–MADISON GRADUATE SCHOOL

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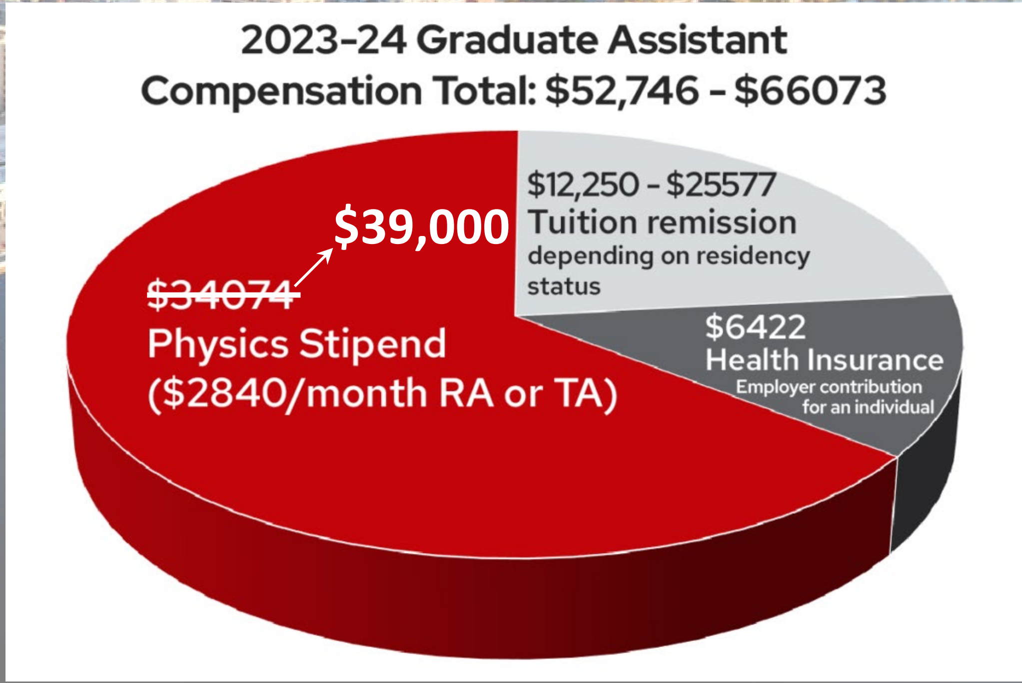
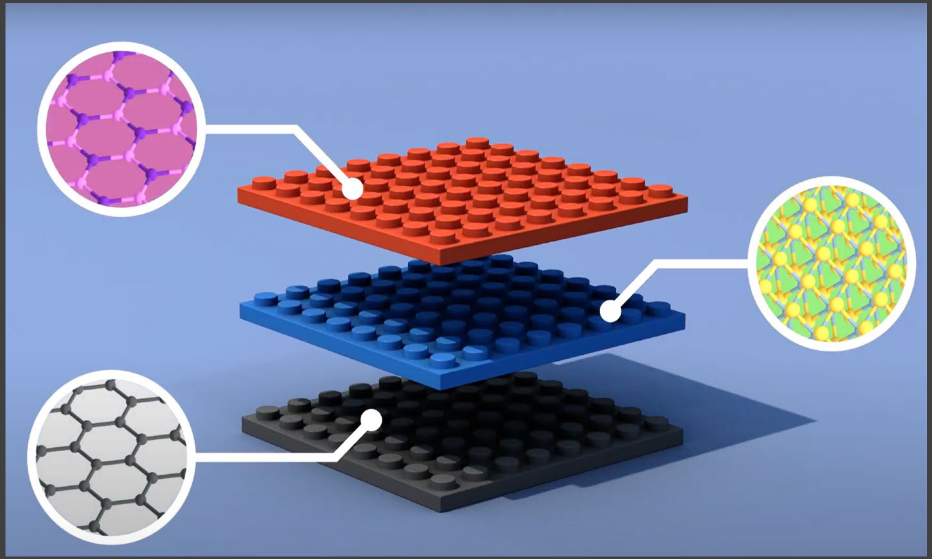
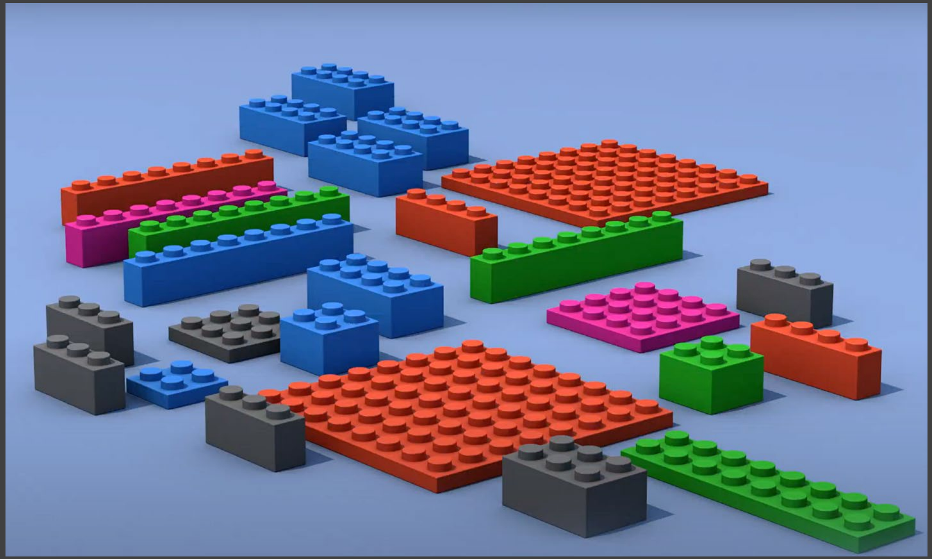
- **#5** In federal support of graduate students
- Professional development opportunities and student support services
- Apply to three programs with one application fee
- Application fee waivers available

**We are hiring!**



# Questions?





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