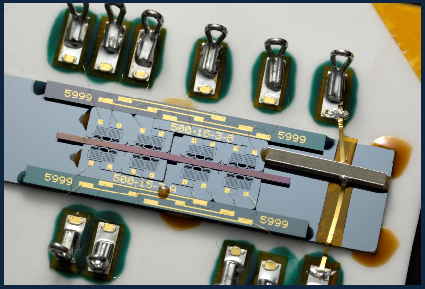
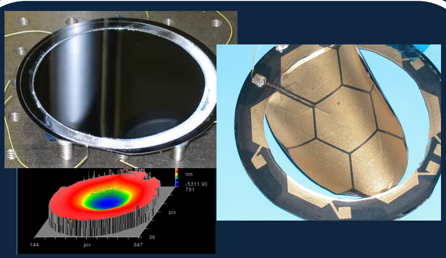
Air Stability, Doping, and Magnetism in Transition Metal Dichalcogenides

Dr. EH Yang
Professor, Mechanical Engineering Department
Stevens Institute of Technology

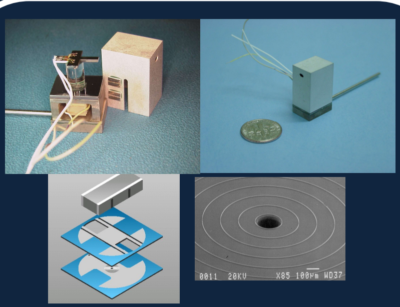
MEMS Technologies for Space Applications



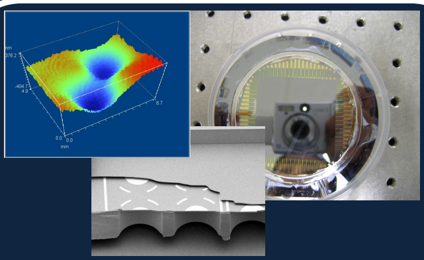
Self-Latched Linear Microactuator



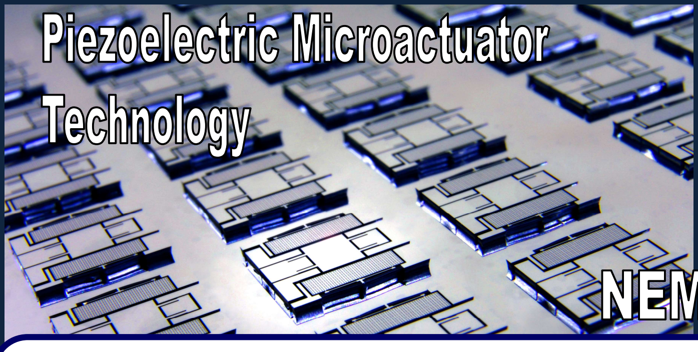
Electroactive Polymer Actuated Mirror



High-Pressure, Leak-Tight Piezoelectric Microvalve

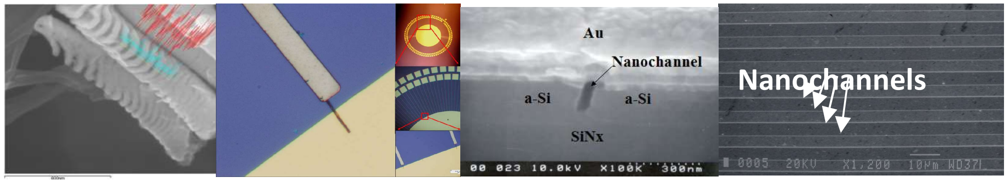


Large-Stroke, Large-Area MEMS Deformable Mirror

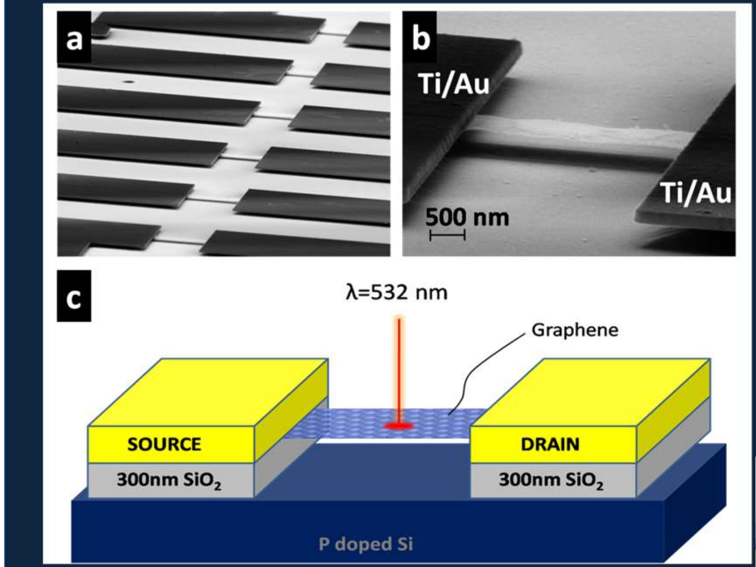


Piezoelectric Microactuator Technology

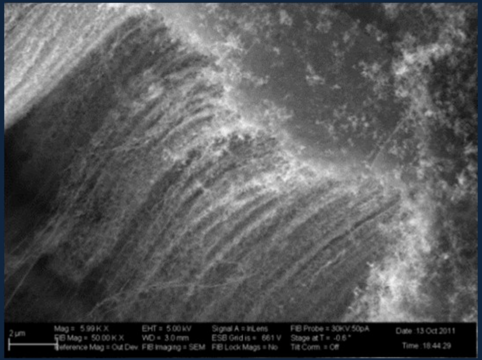
NEMS Technology



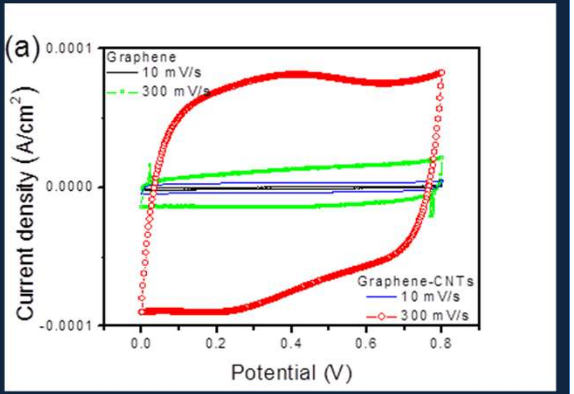
Graphene Photodetector, Supercapacitor and Flexible Substrate



Scientific Reports, (3) 2791 (2013)

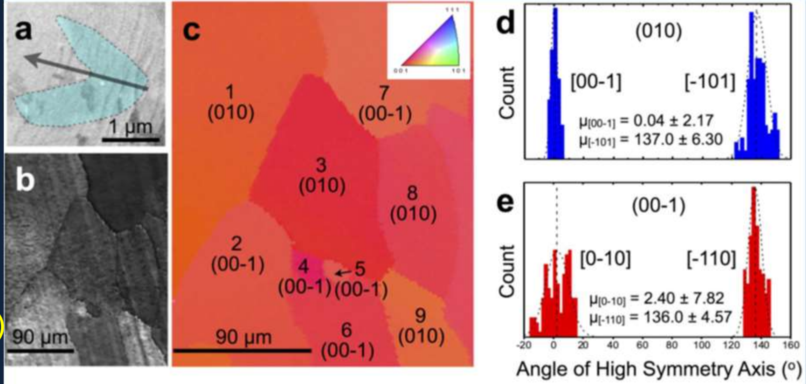


Chemistry of Materials, 25(19), 3874 (2013)

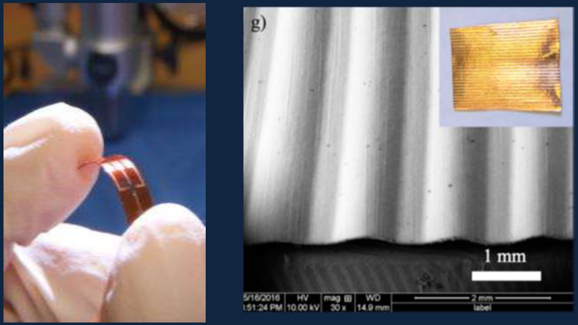


Nanotechnology, 23, 015301 (2012)

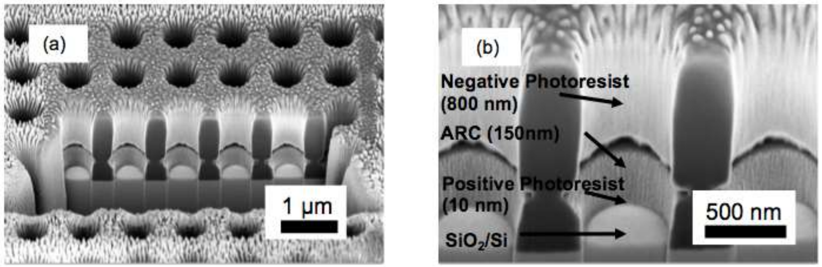
Nano Letters, 11(3), 1254 (2011)
Nano Letters, 11, 4874 (2011)



Scientific Reports, (3) 2571 (2013)



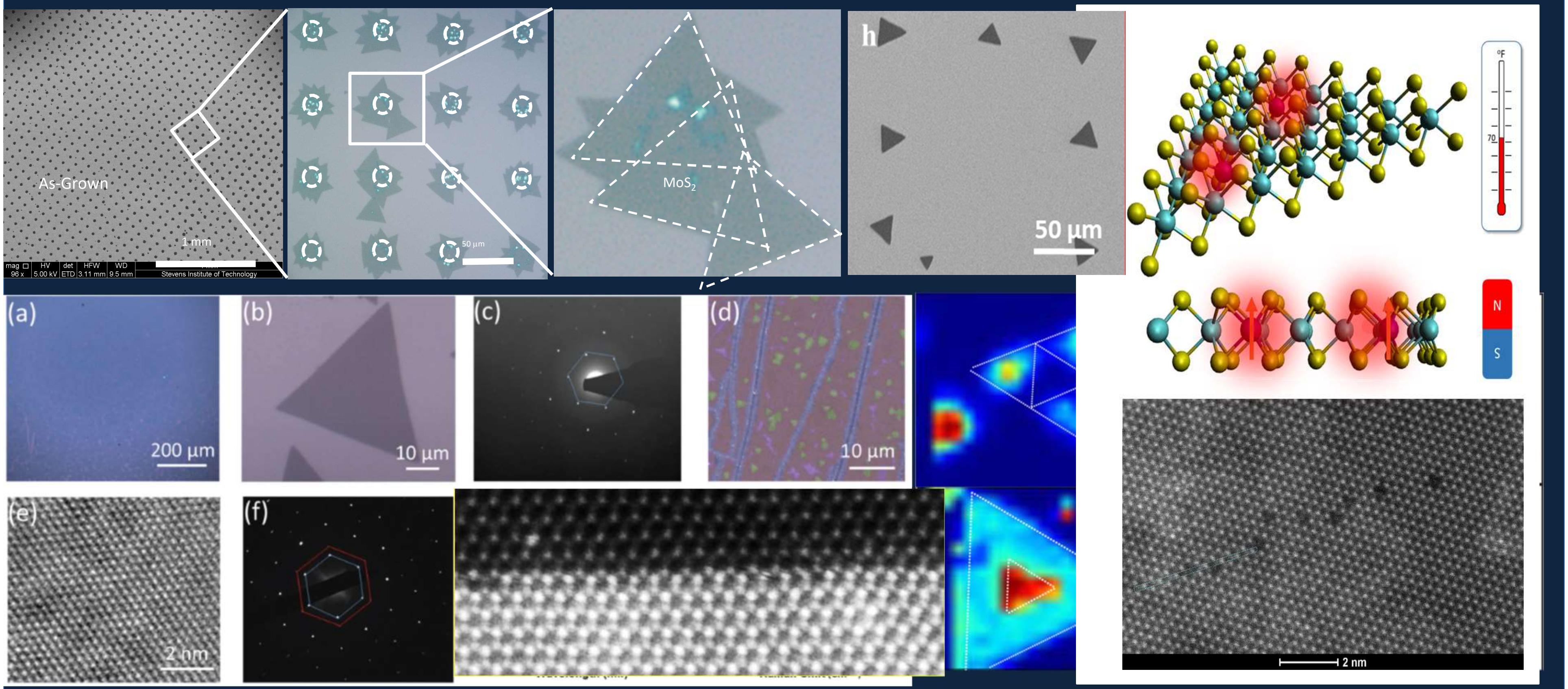
JVST B, 34(5), 051205 (2016)



Carbon, (64), 35, (2013)

JVST B, 32(6), 06FF01 (2014)

TMD Growth, Characterization and Applications



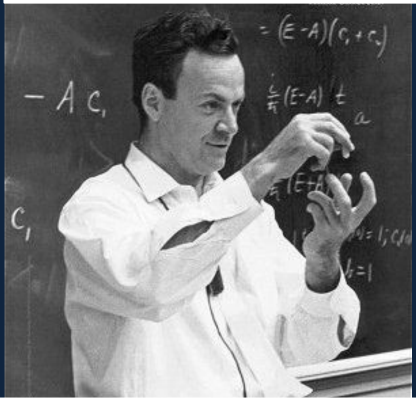
Nature Communications, 11, 2034 (2020)
Scientific Reports, Nanotechnology (2020) *in print*
Chemistry of Materials, 30, 5148 (2018)
Advanced Materials, 18, 1603898 (2017)
2D Materials, 4, 025093 (2017)

Scientific Reports, 10, 1648 (2020)
Annalen der Physik., 1800507 (2019)
Journal of Physical Chemistry C, 123(35), 21813 (2019)
2D Materials, 4, 025045 (2017)
Scientific Reports, 7, 17798 (2017)

Layered Materials

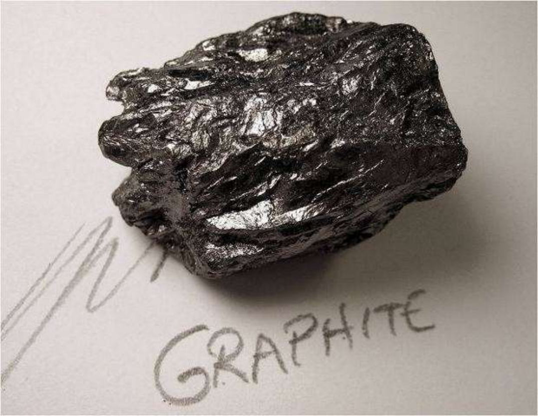
“What could we do with layered structures with just the right layers? What would the properties of materials be if we could really arrange the atoms the way we want them…… I can hardly doubt that when we have some control of the arrangement of things on a small scale, we will get an enormously greater range of possible properties that substances can have…….”

R. P. Feynman
There is Plenty of Room at the Bottom
December 29, 1959

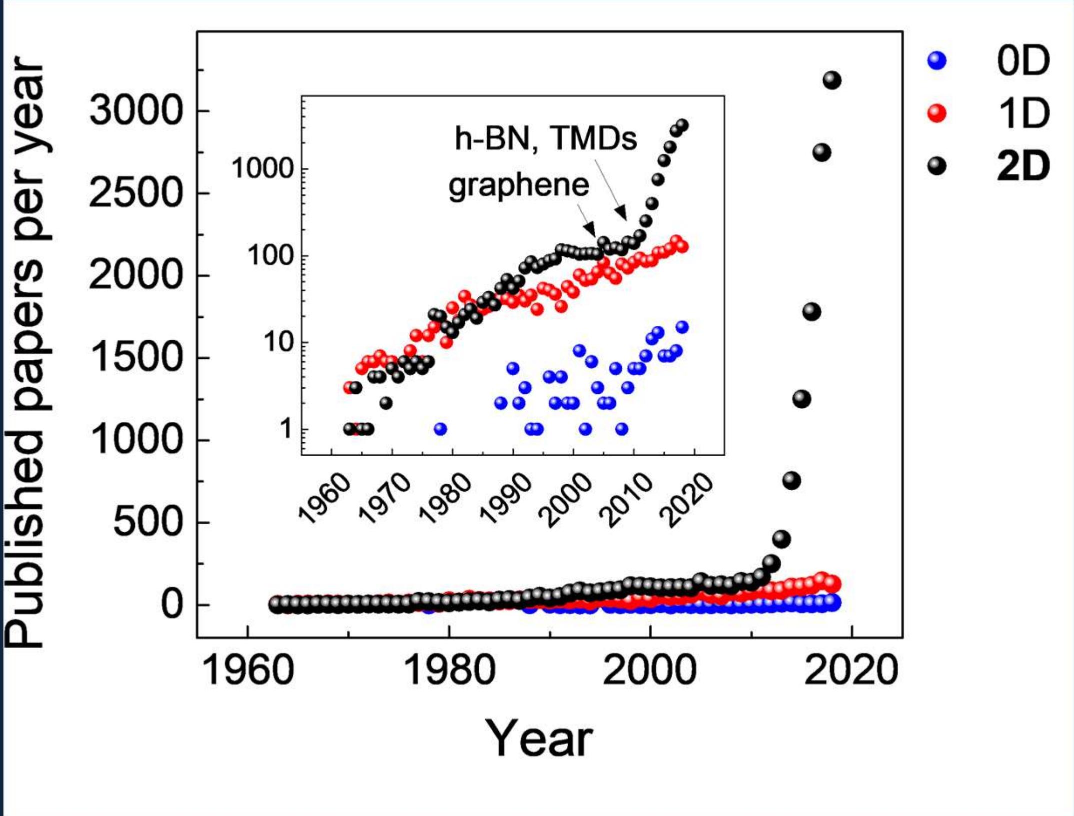


Discovery of Graphene

- Graphene theory first explored by P.R. Wallce (1947)
- Although scientists knew graphene existed, no-one had worked out how to extract it from graphite.
- That was until it was isolated in 2004 by Geim and Novoselov who received Nobel Prize (2010)



Exponential Growth



https://www.mdpi.com/journal/crystals/special_issues/2d_materials_structure_function

- There has been an exponential growth in the research on the increasingly broad portfolio of 2D materials.
- Since 2010, the number of publications per year on 2D materials jumped exponentially from 139 in 2010 to 3189 in 2018.

No Bandgap

NATURE NANOTECHNOLOGY DOI: 10.1038/NNANO.2010.89 REVIEW ARTICLE

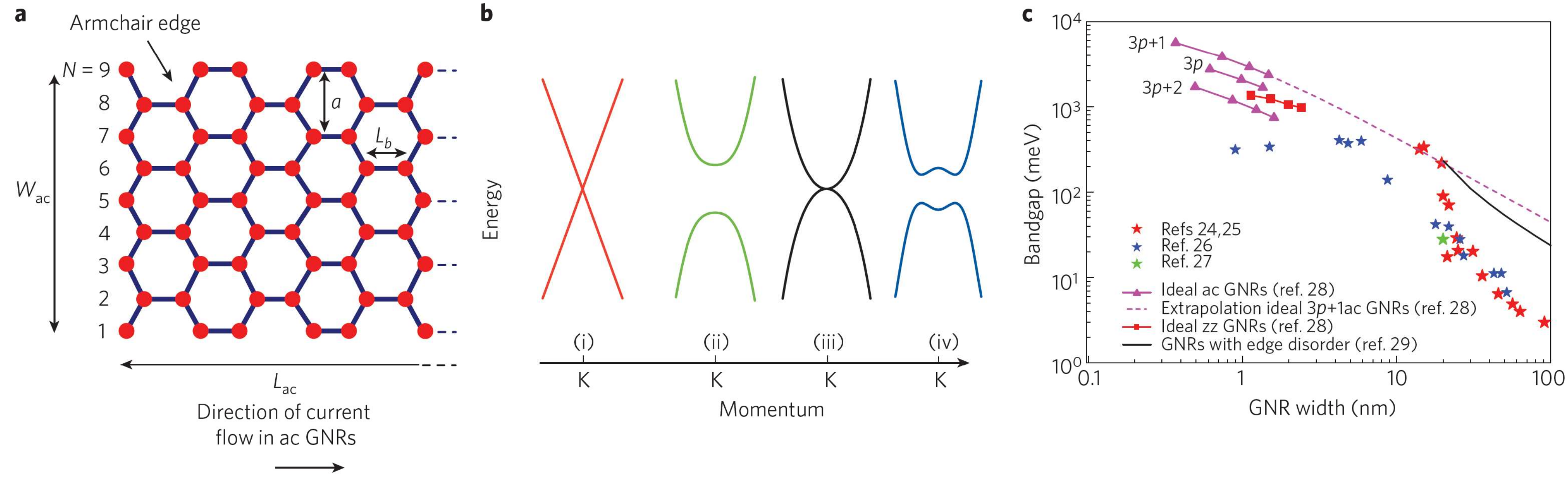
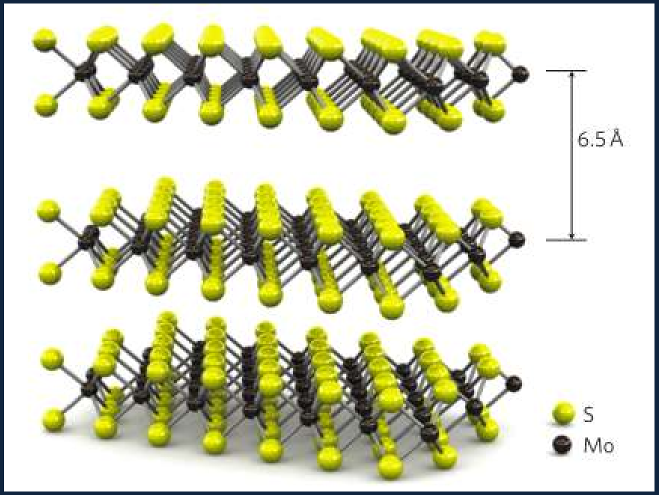


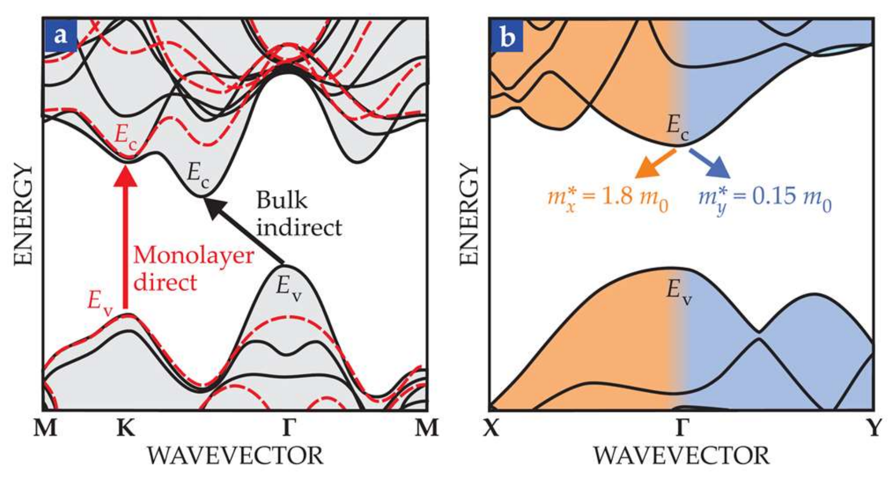
Figure 4 | Properties of graphene and graphene nanoribbons. a, Schematic of an armchair (ac) graphene nanoribbon (GNR) of length L_{ac} and width W_{ac}. The nanoribbon shown here has $N = 9$ carbon atoms along its width and thus belongs to the $3p$ family, where p is an integer. **b**, Band structure around the K point of (i) large-area graphene, (ii) graphene nanoribbons, (iii) unbiased bilayer graphene, and (iv) bilayer graphene with an applied perpendicular field. Large-area graphene and unbiased bilayer graphene do not have a bandgap, which makes them less useful for digital electronics. **c**, Bandgap versus nanoribbon width from experiments[24–27] and calculations[28,29]. By comparison, the bandgap of Si is above 1 eV. zz: zigzag.

2D Materials beyond Graphene

- The discovery of graphene shows how new physical properties emerge when a bulk crystal is thinned down to one atomic layer.
- Transition metal dichalcogenides (TMDs) (MX_2 where M=Ti, Zr, Hf, V, Nb, Ta, **Mo**, **W**, and X = **S**, **Se**, Te) handle just like graphene.
- Some of TMD monolayers have a *direct bandgap*, and can be directly used in electronics and optics, complementing graphene.



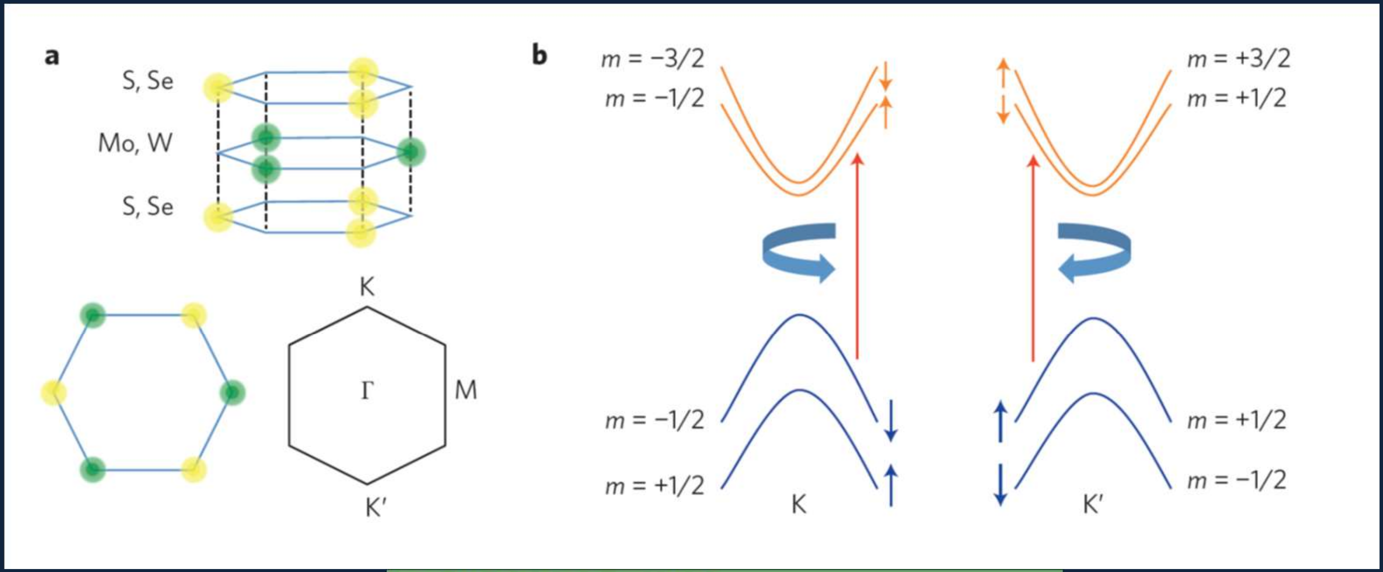
Fuhrer and Hone, *Nature Nanotechnology*, 8(3), 147, (2013)



Ajayan, Kim, Benerjee, *Phys. Today 69, 38, (2016)*

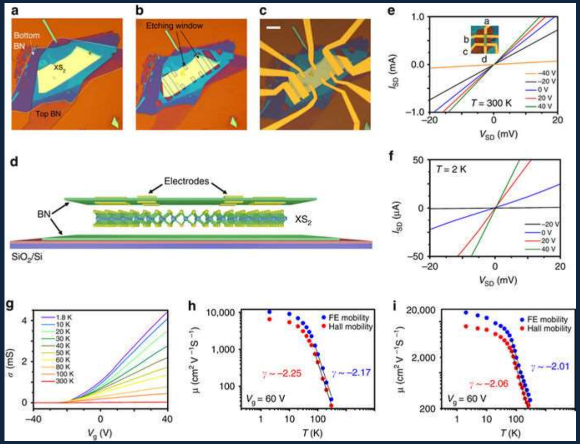
Transition Metal Dichalcogenides

- The discovery of the direct bandgap makes the work on TMD monolayers an emerging research and development field.
- The TMD monolayer crystal structure allows to open up a new field of physics.
- TMDs are often combined with other 2D materials like graphene and hexagonal boron nitride to make van der Waals heterostructure devices.

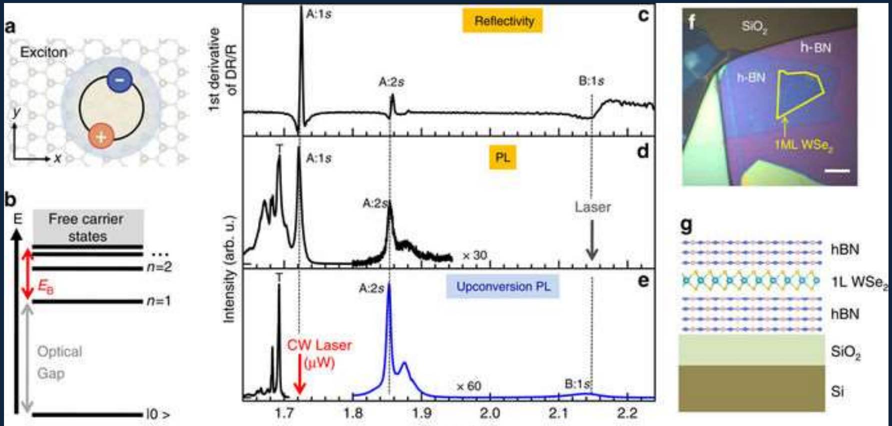


NATURE PHOTONICS DOI: 10.1038/NPHOTON.2015.282

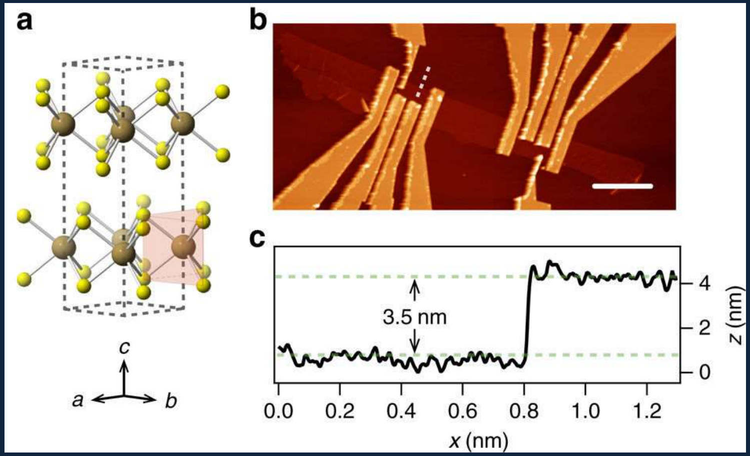
Exploring exotic properties from exfoliated flakes



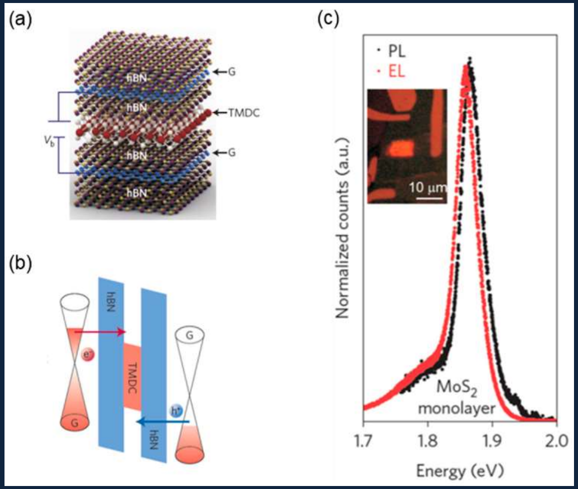
Wu, *et al., Nature Communications* 7, 12955 (2016)



Manca, *et al., Nature Communications* 8, 14927 (2017)

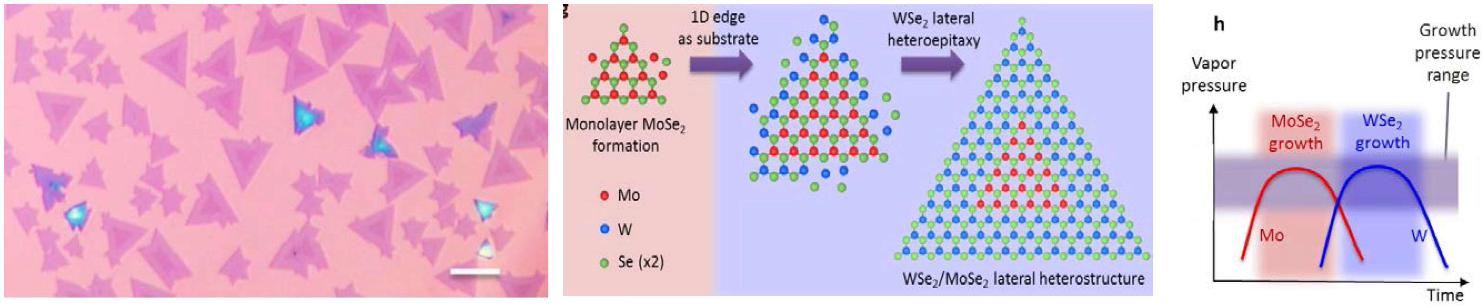


Navarro-Moratalla, *et al., Nature Communications* 7, 11043 (2016)

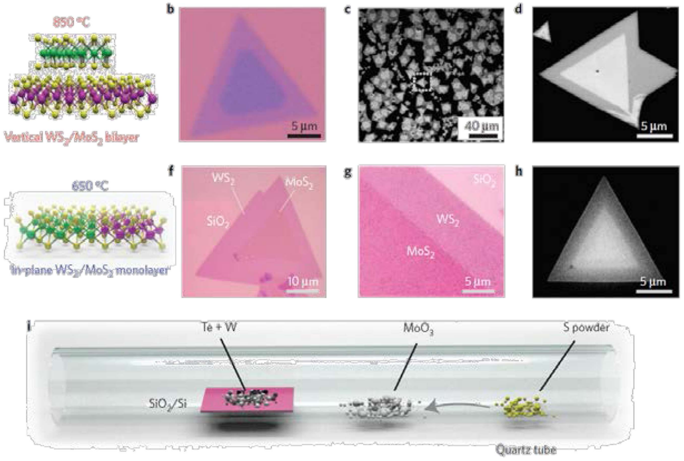


Novoselov *et al., Nature Materials 14*, 301 (2015)

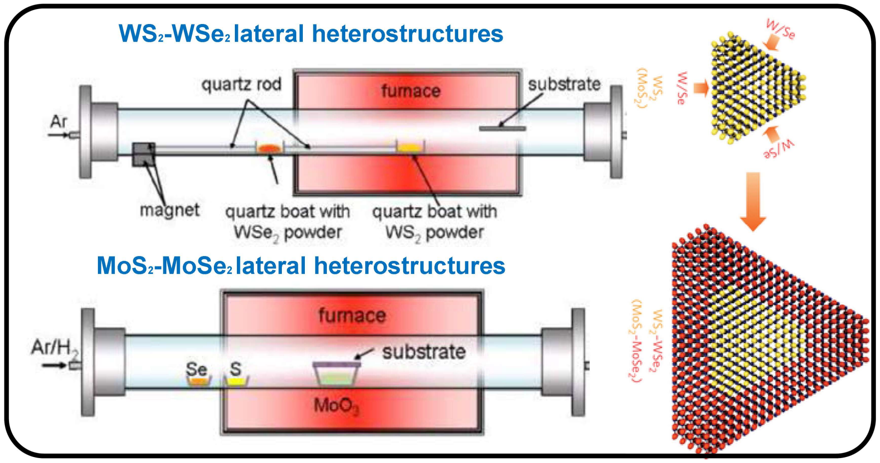
Heterostructures Growth via LPCVD



Xu, *et al., Nature Materials* 13, 1096 (2014)

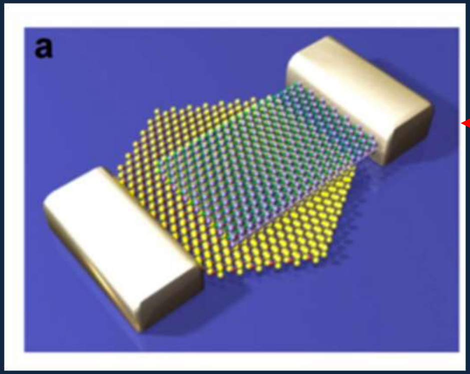


Ajayan, *et al., Nature Materials* 13, 1135 (2014)



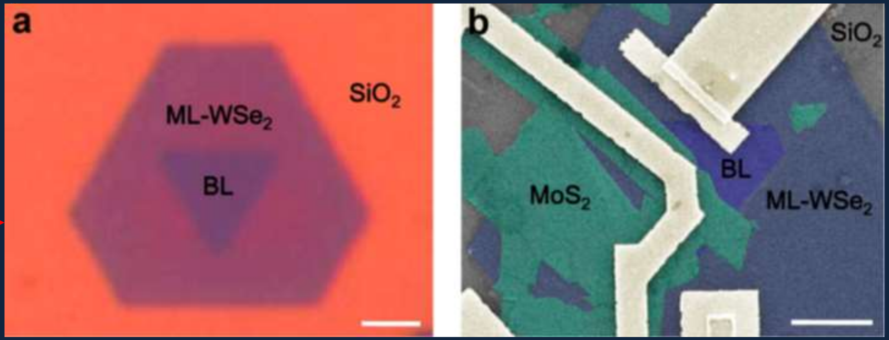
Duan, *et al., Nature Nanotechnology* 9, 1024 (2014)

How to cheaply produce large, uniform, high-quality layers and heterostructures?

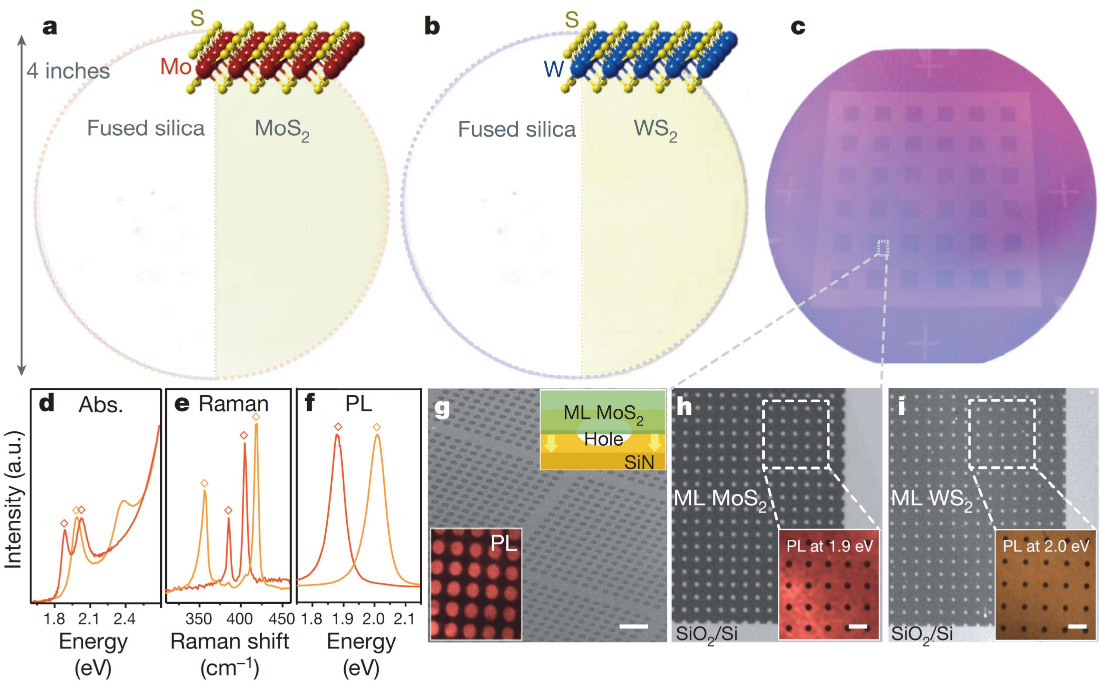


Ideal

Reality



Cheng *et al.*, *Nano Lett.*, 14, 5590 (2014)

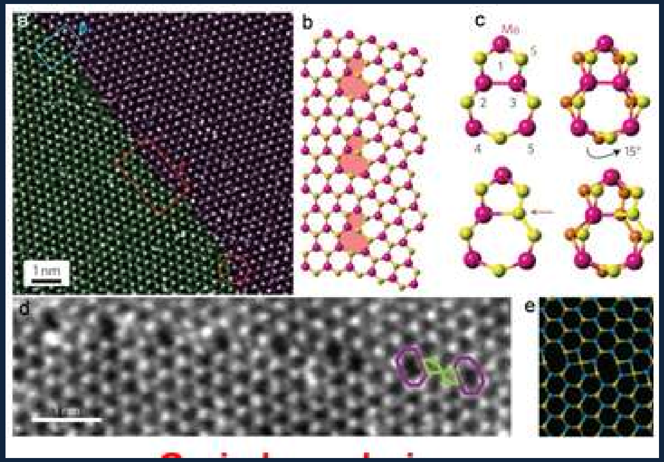


Park, *et al.*,
Nature 520, 656 (2015)

Key Challenges Facing TMD Research

- Scaling up
- Controlled growth
- Control of grain boundaries
- Control of defects
- Air stability
- Control of doping (alloys)

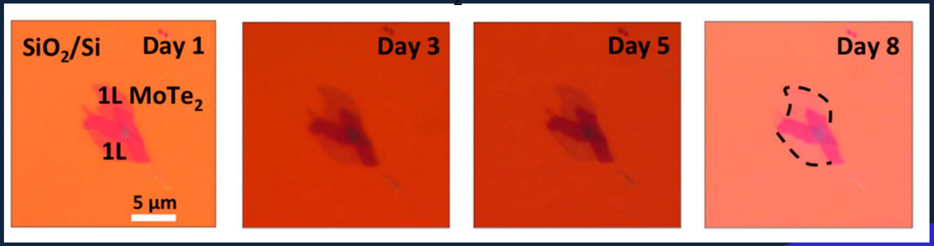
Najmaei, *et al., Nature Materials*, 12, 754 (2013)



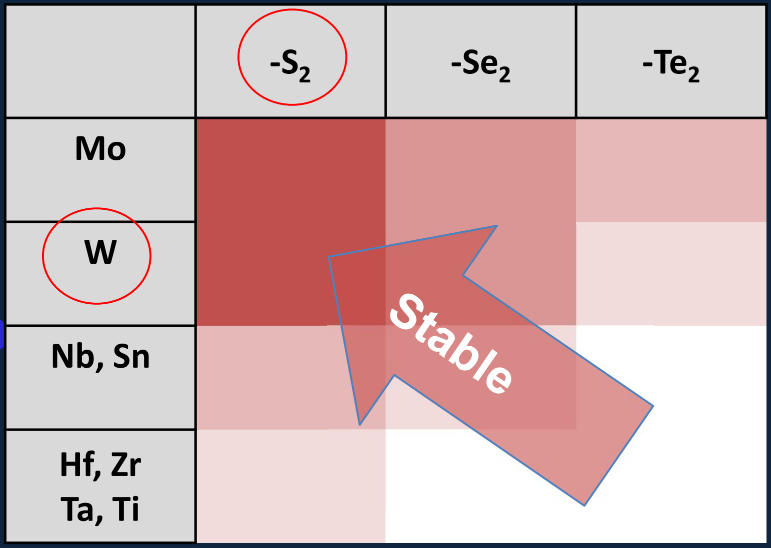
Useful parallels can be drawn with GaAs, where control over materials' quality was achieved over decades of work.

Air Stability of 2D Materials

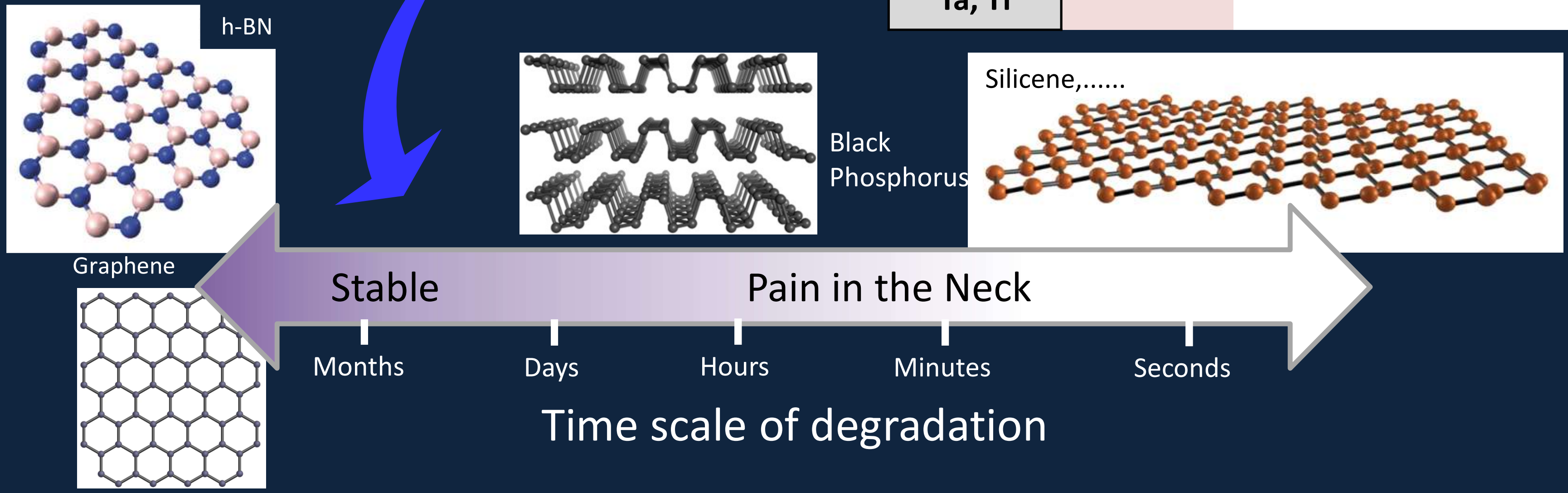
MoTe$_2$



B. Chen et al. *ACS nano* 9.5326 (2015)

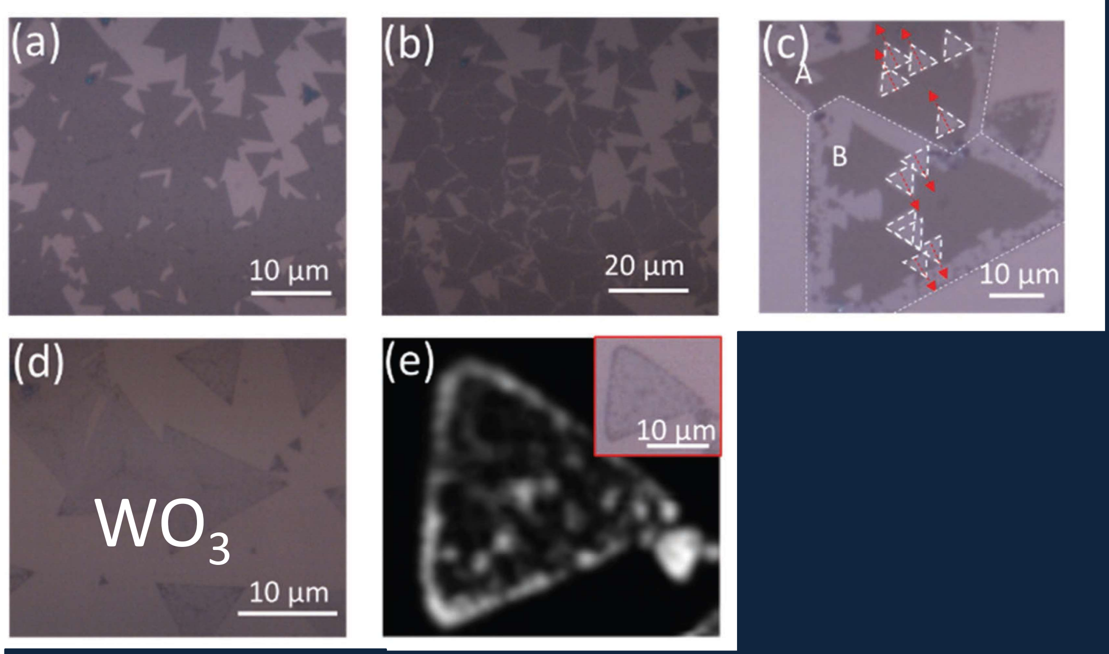


	-S$_2$	-Se$_2$	-Te$_2$
Mo			
W			
Nb, Sn			
Hf, Zr Ta, Ti			



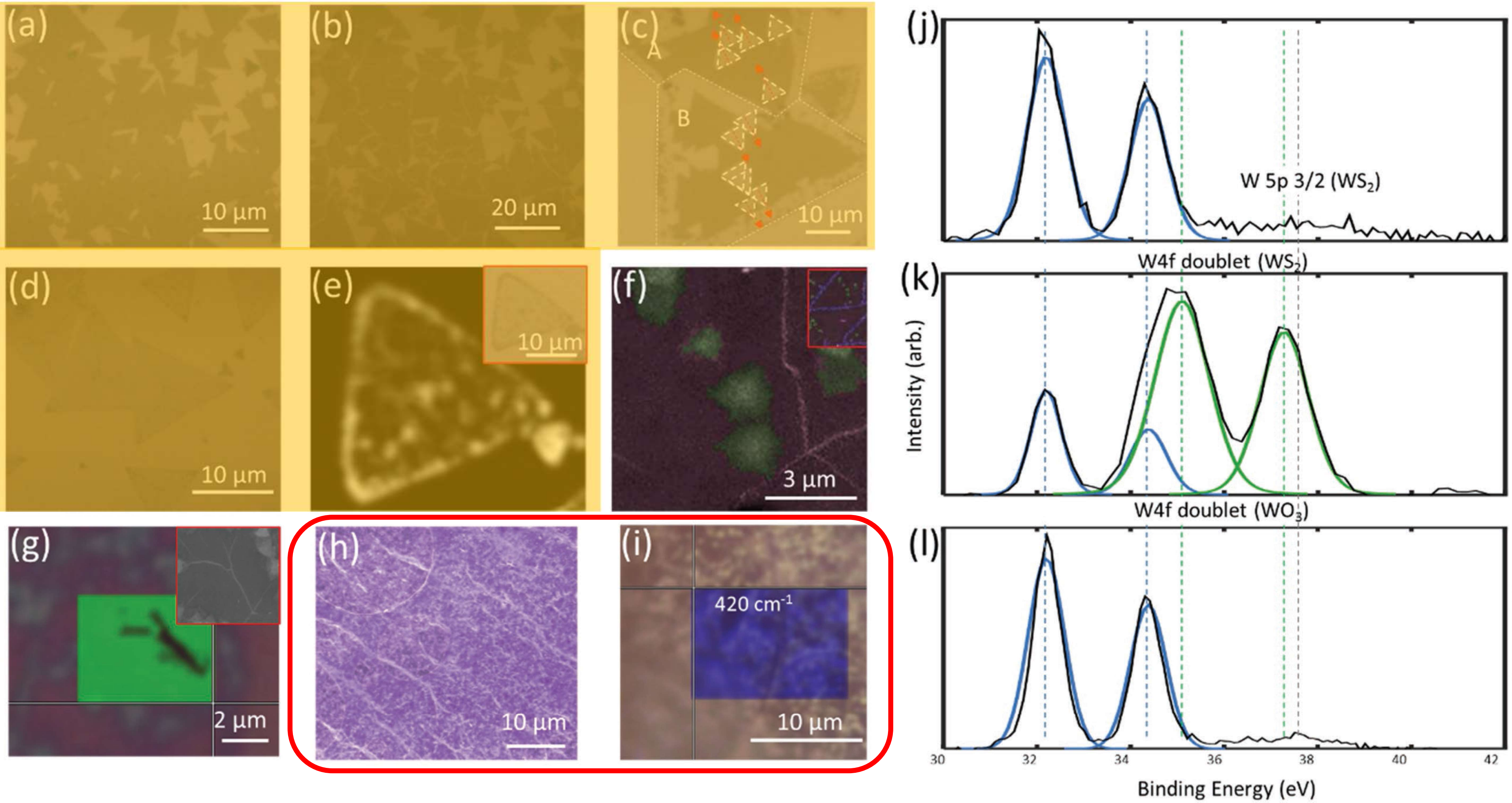
Time scale of degradation

In Air Oxidation of WS_2 Monolayers on SiO_2



Advanced Materials, 18, 1603898 (2017)

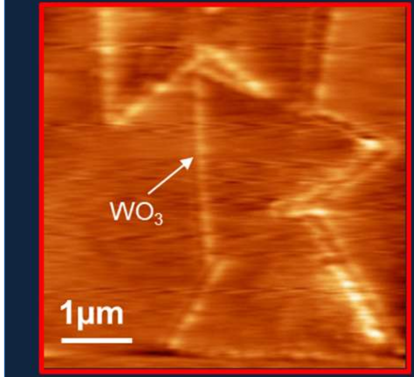
Enhanced Air Stability: WS_2 Grown on Graphene

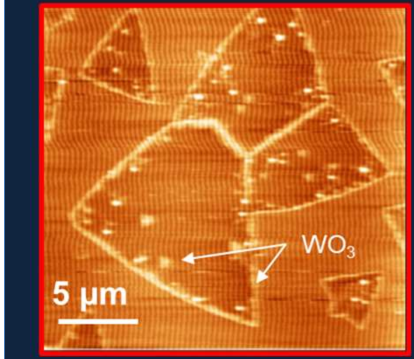


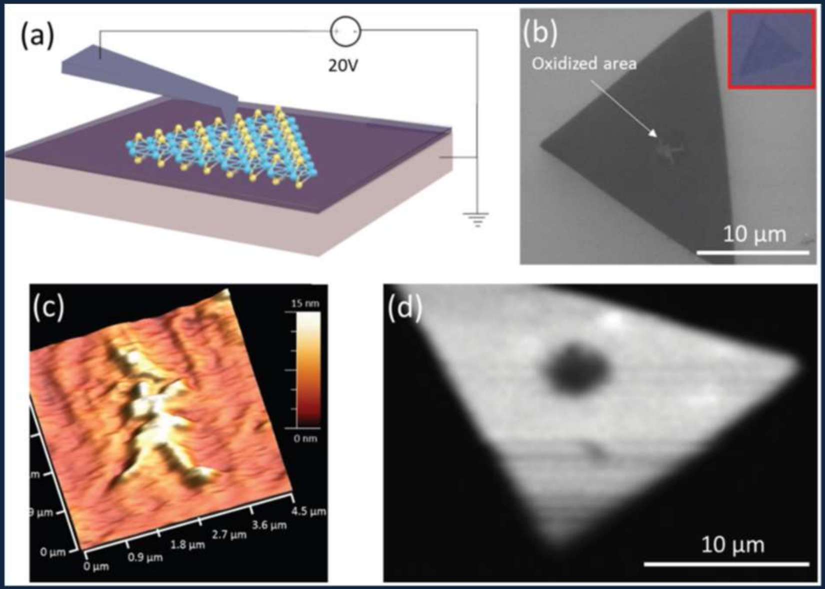
Advanced Materials, 18, 1603898 (2017)

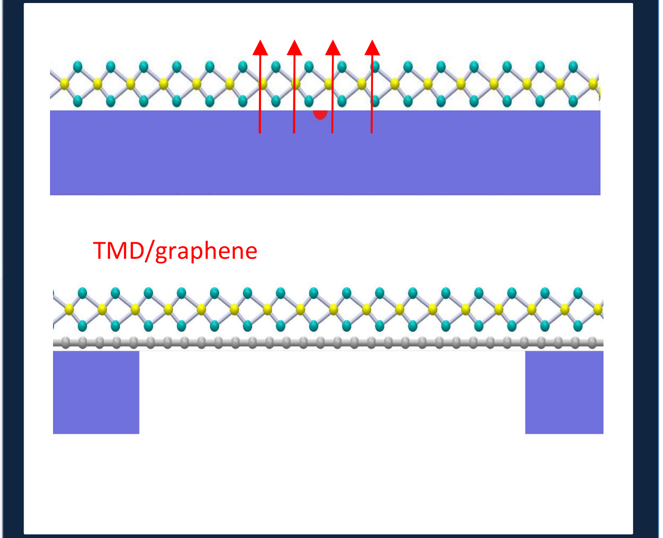
Surface Electric Field and Oxidation

The charges or dipoles in the Si substrate generate electric fields on the SiO_2 surface.









The surface electric fields reinforce the adsorbate - TMD interaction by inducing an electronic charge transfer, affecting the rate of WS_2 oxidation.

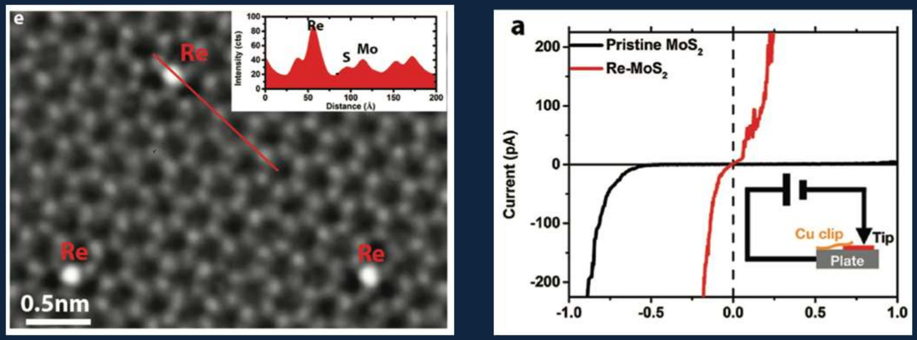
Advanced Materials, 18, 1603898 (2017)

Key Challenges Facing TMD Research

- Scaling up
- Controlled growth
- Control of grain boundaries
- Control of defects
- Air stability
- Control of doping (alloys)

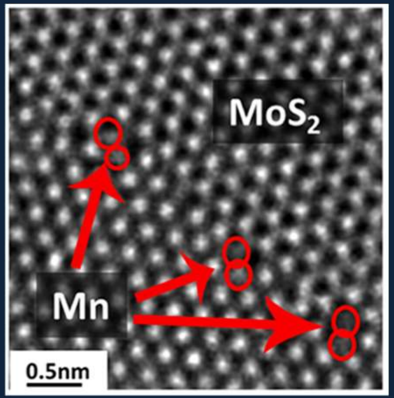
2D magnets

Re:MoS_2



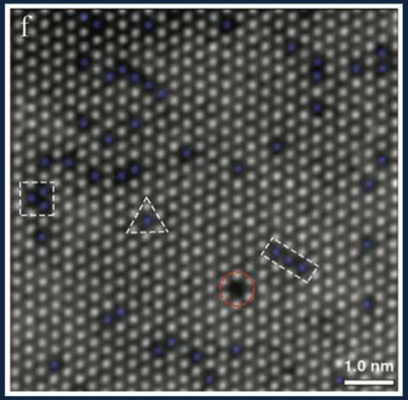
Zhang *et al.*, *Adv. Funct. Mater.*, 2018, **28**, 1706950

Mn:MoS_2



Zhang *et al.*, *Nano Lett.*, 2015, **15**, 6586

Nb:WS_2



Gao *et al.*, *Adv. Mat.*, 2016, **28**, 9735

INTRODUCTION

What Don't We Know?

Special Section

At *Science*, we tend to get excited about new discoveries that lift the veil a little on how things work, from cells to the universe. That puts our focus firmly on what has been added to our stock of knowledge. For this anniversary issue, we decided to shift our frame of reference, to look instead at what we *don't* know: the scientific puzzles that are driving basic scientific research.

We began by asking *Science*'s Senior Editorial Board, our Board of Reviewing Editors, and our own editors and writers to suggest questions that point to critical knowledge gaps. The ground rules: Scientists should have a good shot at answering the questions over the next 25 years, or they should at least know how to go about answering them. We intended simply to choose 25 of these suggestions and turn them into a survey of the big questions facing science. But when a group of editors and writers sat down to select those big questions, we quickly realized that 25 simply wouldn't convey the grand sweep of cutting-edge research that lies behind the responses we received. So we have ended up with 125 questions, a fitting number for *Science*'s 125th anniversary.

First, a note on what this special issue is not: It is not a survey of the big societal challenges that science can help solve, nor is it a forecast of what science might achieve. Think of it instead as a survey of our scientific ignorance, a broad swath of questions that scientists themselves are asking. As Tom Siegfried puts it in his introductory essay, they are "opportunities to be exploited."

We selected 25 of the 125 questions to highlight based on several criteria: how fundamental they are, how broad-ranging, and whether their solutions will impact other scientific disciplines. Some have few immediate practical implications—the composition of the universe, for example. Others we

- Suggest questions that point to critical knowledge gaps.
- Ended up with 125 questions, a fitting number for *Science*'s 125th anniversary.

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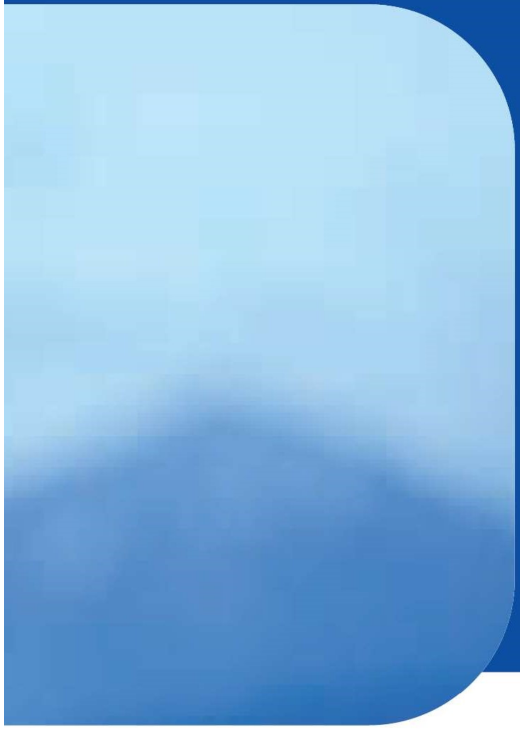
vith microwaves
isible light.



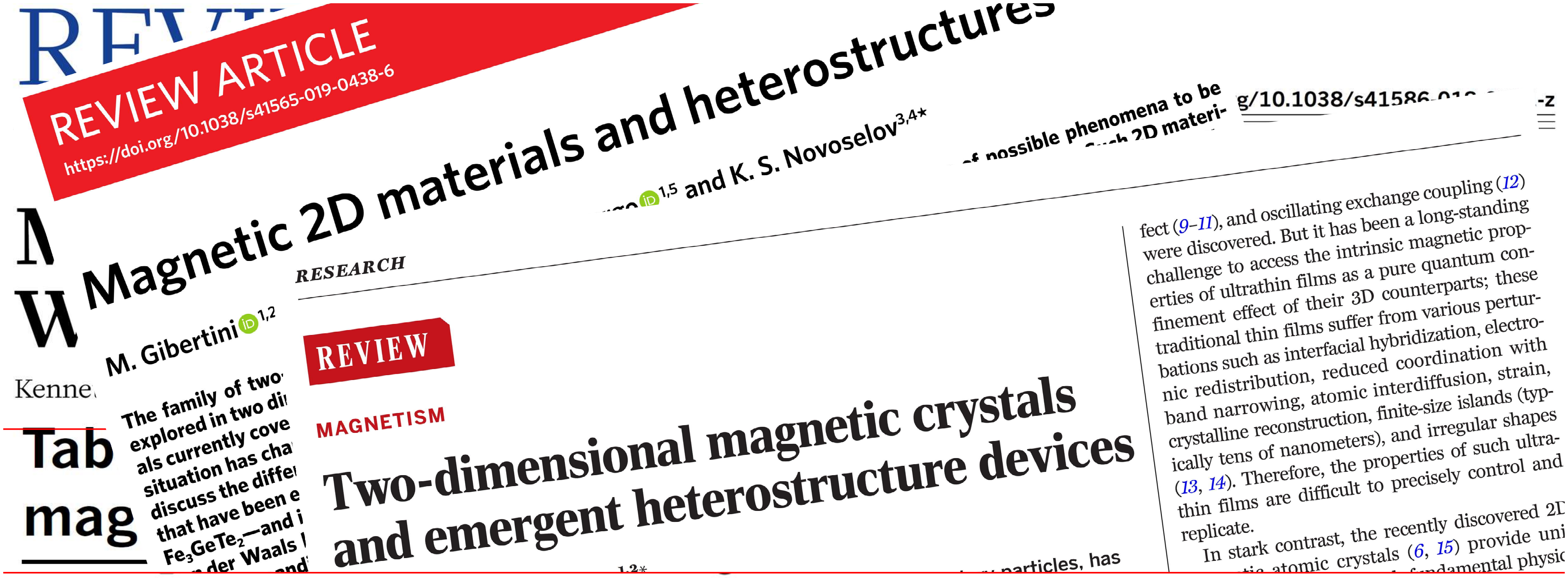
JUPITER IMAGES

Is it possible to create magnetic semiconductors that work at room temperature?

Such devices have been demonstrated at low temperatures but not yet in a range warm enough for spintronics applications.

What is the pairing behind high-tempe superconductivity

Electrons in supercon
together in pairs. Afte
intense study, no one
holds them together i
high-temperature ma



REVIEW ARTICLE
https://doi.org/10.1038/s41565-019-0438-6

Magnetic 2D materials and heterostructures

M. Gibertini 1,2 … 1,5 and K. S. Novoselov 3,4*

The family of two… explored in two di… als currently cove… situation has cha… discuss the diffe… that have been e… that have been i… Fe_3GeTe_2—and i… der Waals …

…of possible phenomena to be … such 2D materi…

…g/10.1038/s41586-01…-z

RESEARCH

REVIEW

MAGNETISM

Two-dimensional magnetic crystals and emergent heterostructure devices

fect (9–11), and oscillating exchange coupling (12) were discovered. But it has been a long-standing challenge to access the intrinsic magnetic properties of ultrathin films as a pure quantum confinement effect of their 3D counterparts; these traditional thin films suffer from various perturbations such as interfacial hybridization, electronic redistribution, reduced coordination with band narrowing, atomic interdiffusion, strain, crystalline reconstruction, finite-size islands (typically tens of nanometers), and irregular shapes (13, 14). Therefore, the properties of such ultrathin films are difficult to precisely control and replicate.

In stark contrast, the recently discovered 2D … atomic crystals (6, 15) provide uni… fundamental physic…

Kenne…

Tab…

mag…

The considerations here show that there are many different directions for future work. The results reported over the past year or two represent the starting point of a new field in which major developments should be expected. At this stage, the key questions that are being addressed are of fundamental nature, but as soon as 2D magnetic materials can be reliably synthesized with sufficiently high critical temperatures, the potential for technological impact is enormous.

issues

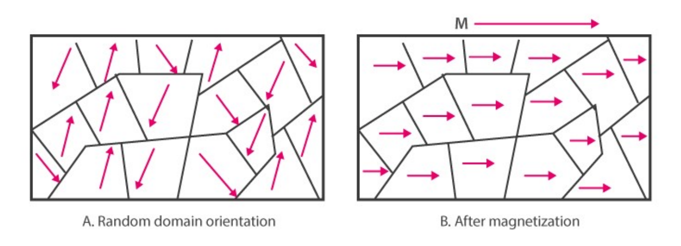
phenomena may … gin. Magnetism is fundamentally roo… charges and spin of elementary par… as the electron

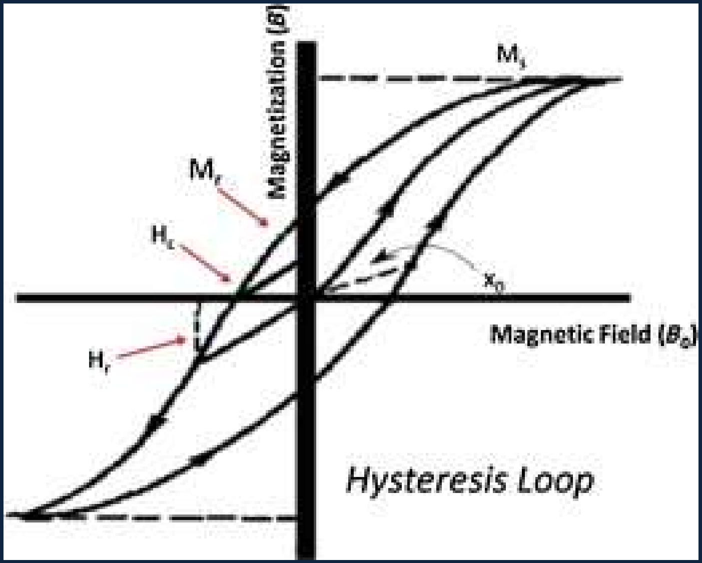
dispersio… to that in the 3D counterpar… to an abrupt onset of magnon density of states … and thus an ease of thermal … mag…

netic coupling. In hete… and photonic materials, the seamle… intricate interplay of distin…

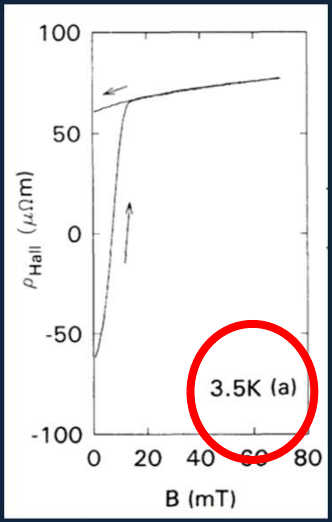
Ferromagnetism

- Ferromagnetism is a form of magnetic ordering in which the intrinsic magnetic dipole moment on each crystal lattice all align in the same direction.
- When a field is applied and then removed, the magnetization does not return to its original value (hysteresis).
- When heated to the Curie point, ferromagnetic materials lose their characteristic properties, but they become ferromagnetic again on cooling.

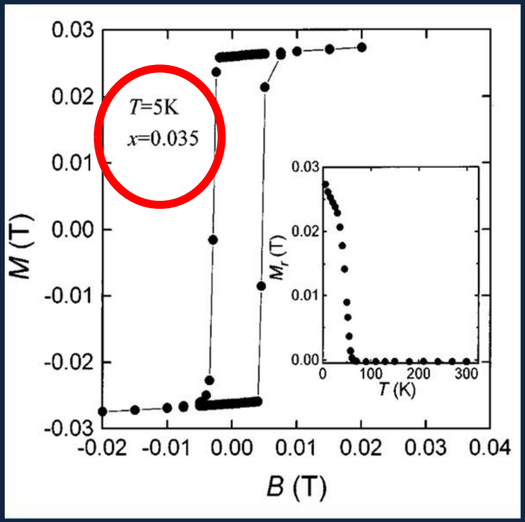




Ferromagnetism in Transition Metal-doped Compound Semiconductors

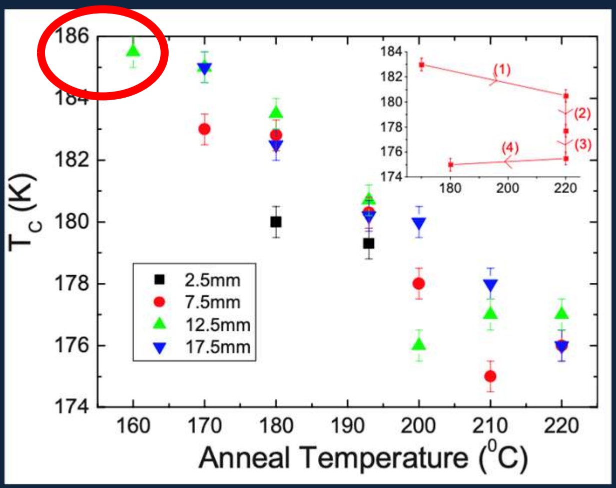


Anomalous Hall effect in (In,Mn)As

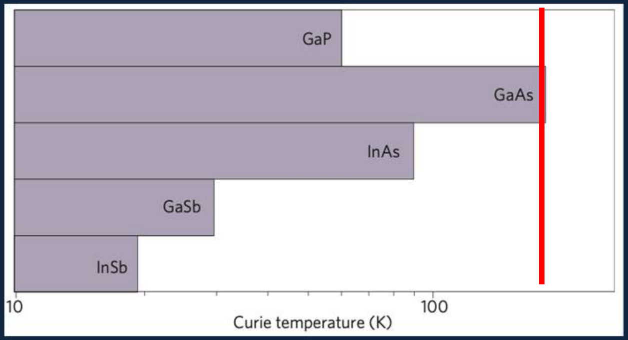


Ferromagnetism in (Ga,Mn)As

Ohno, *et al. Phys. Rev. Lett.* **68**, 2665 (1992)
Ohno, *et al. Appl. Phys. Lett.* **69**, 364 (1996)



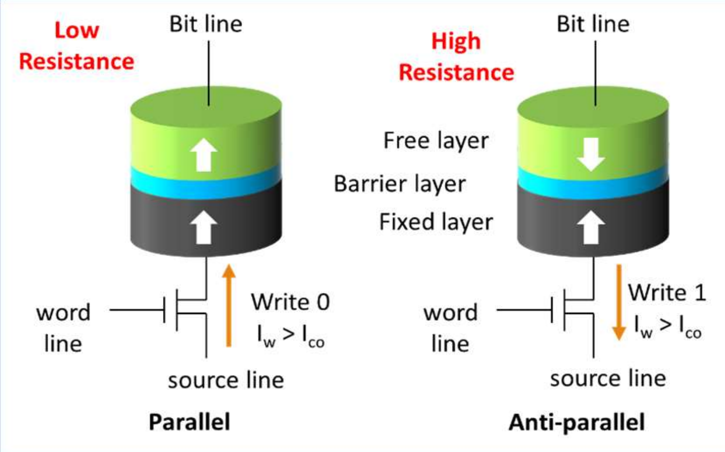
Wang, *et al. Appl. Phys. Lett.* **93**, 132103 (2008)



Dietl, *Nat. Mater.* **9**, 967 (2010)

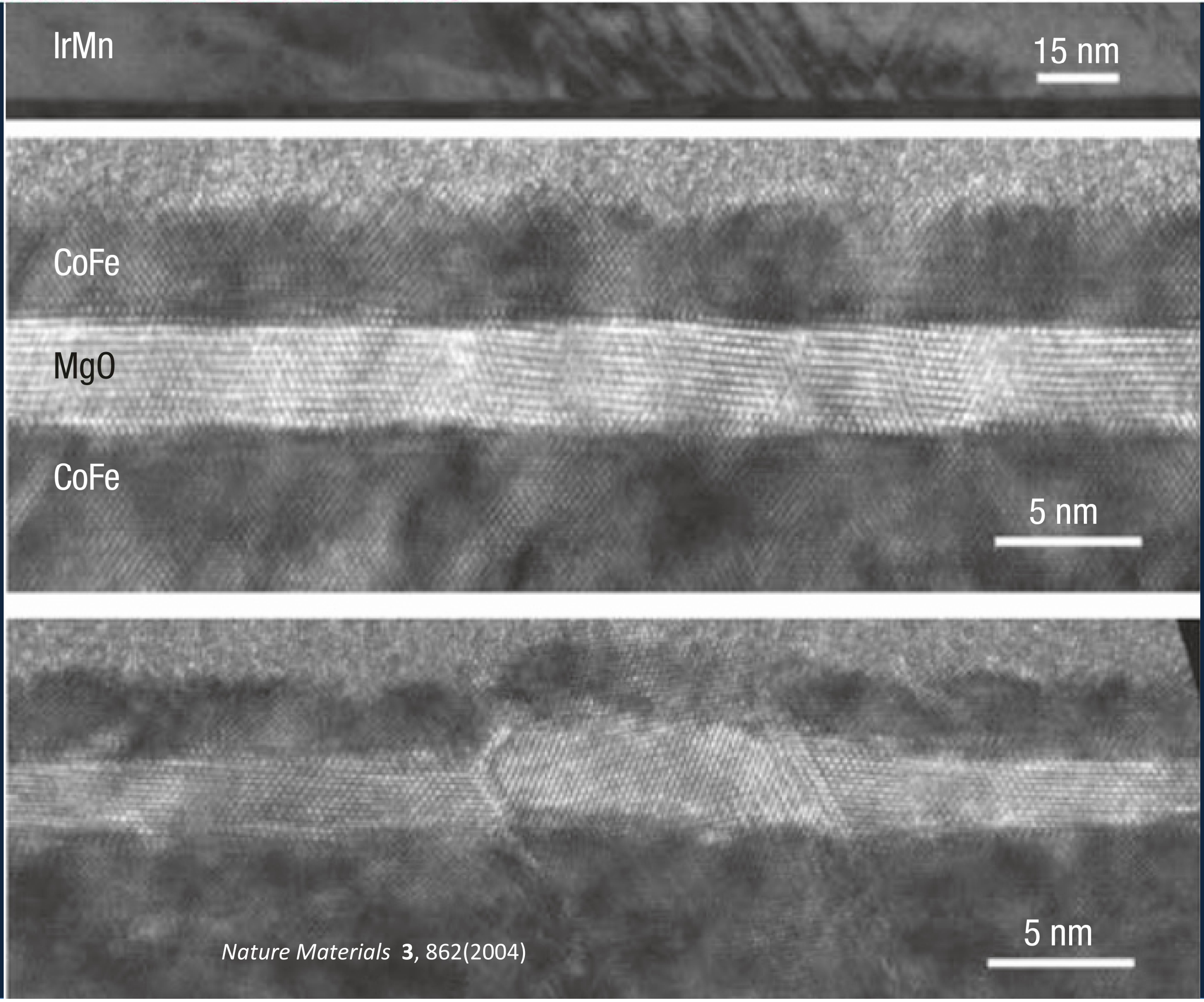
Magnetic Tunnel Junction (MTJ)

- The free layer stores information, and the fixed layer provides a reference frame required for reading and writing.
- Electrons flow through the MTJ to transfer spin angular momentum between the magnetic layers, which results in a torque on the magnetization of the free layer.
- Sufficiently strong torque enables the magnetic state of the free layer to be changed, and thus information can be written.

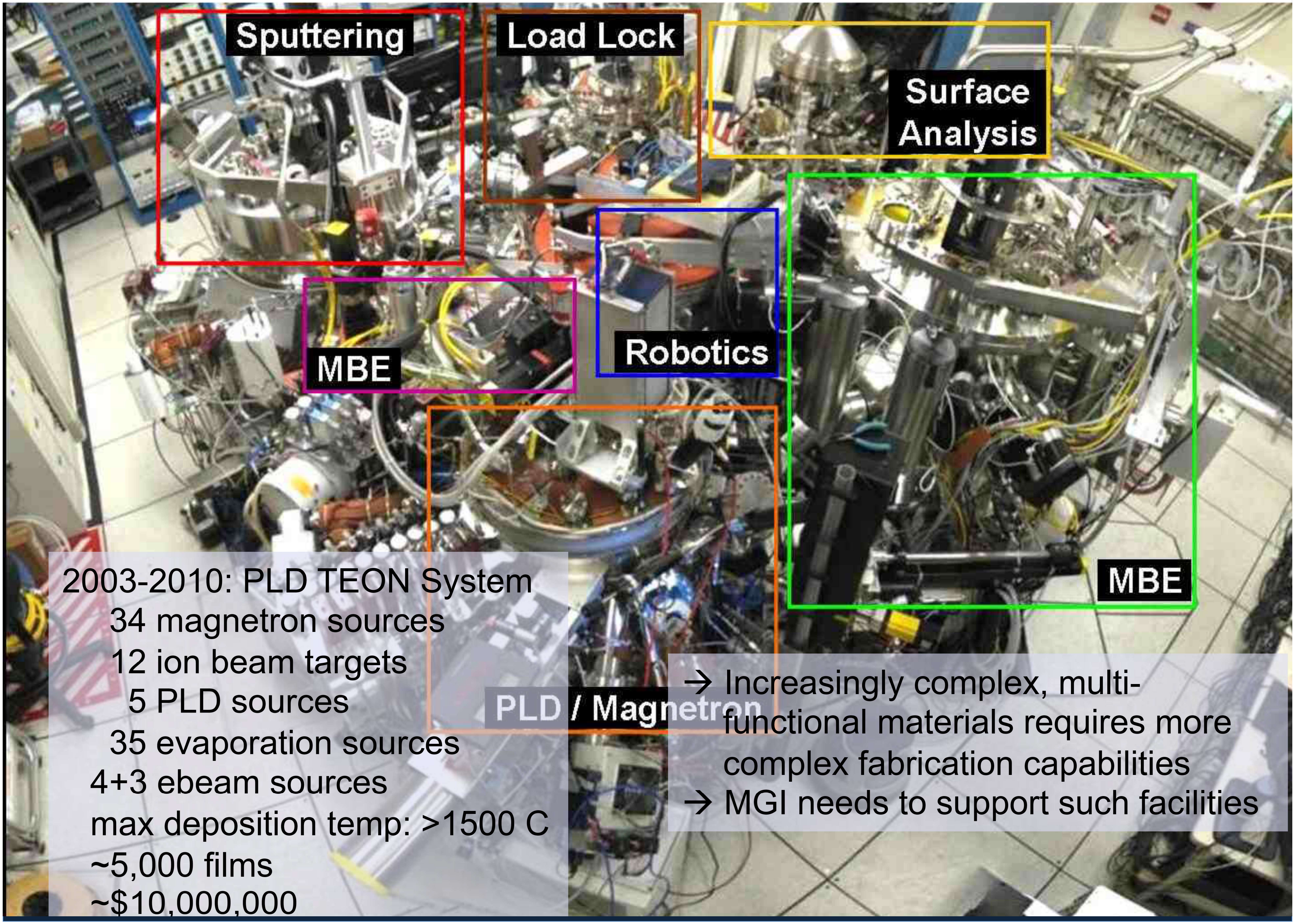


Giant tunnelling magnetoresistance at room temperature with MgO (100) tunnel barriers

STUART S. P. PARKIN[1]*, CHRISTIAN KAISER[1], ALEX PANCHULA[1], PHILIP M. RICE[1], BRIAN HUGHES[2], MAHESH SAMANT[1] AND SEE-HUN YANG[1]



Nature Materials **3**, 862(2004)



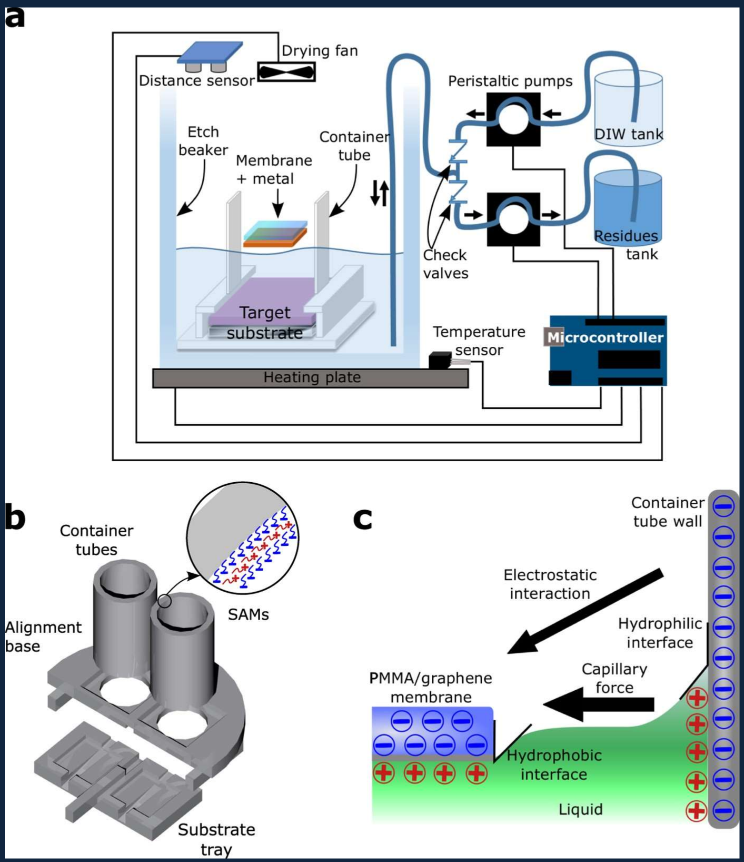
2003-2010: PLD TEON System

34 magnetron sources
12 ion beam targets
5 PLD sources
35 evaporation sources
4+3 ebeam sources
max deposition temp: >1500 C
~5,000 films
~$10,000,000

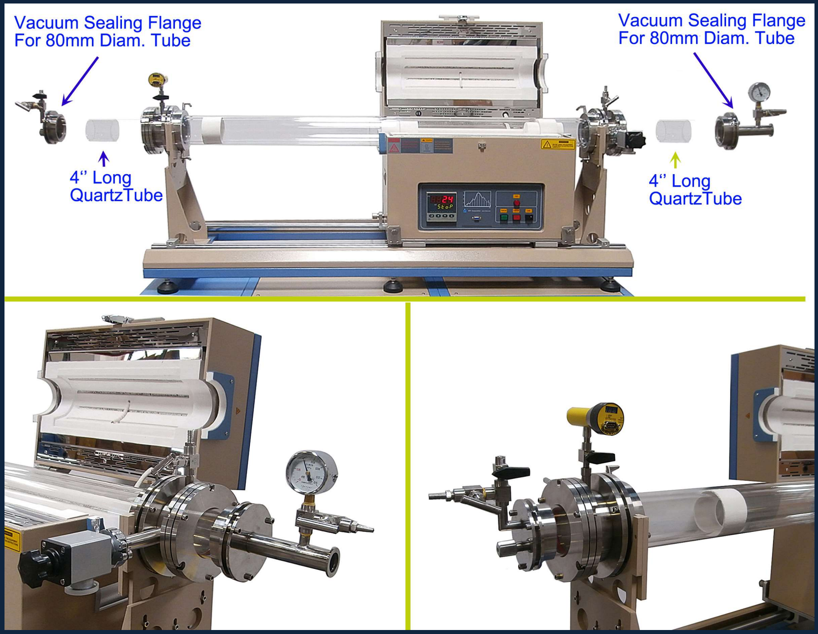
→ Increasingly complex, multi-functional materials requires more complex fabrication capabilities

→ MGI needs to support such facilities

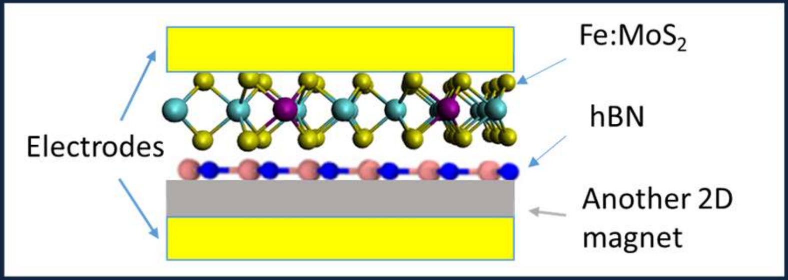
2D magnets would need….



Bosca, et al., *Scientific Reports*, **6**, 21676 (2016)



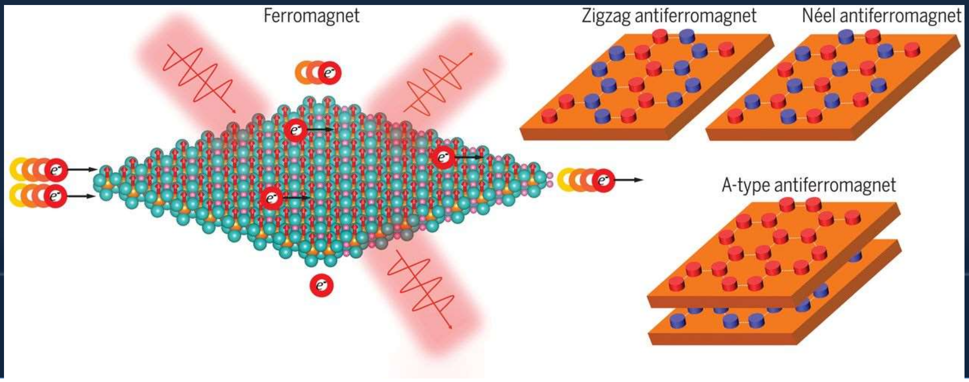
http://www.premier-sols.com/



Provisional patent app: U.S. 63/034,812

Ferromagnetism in 2D Crystals

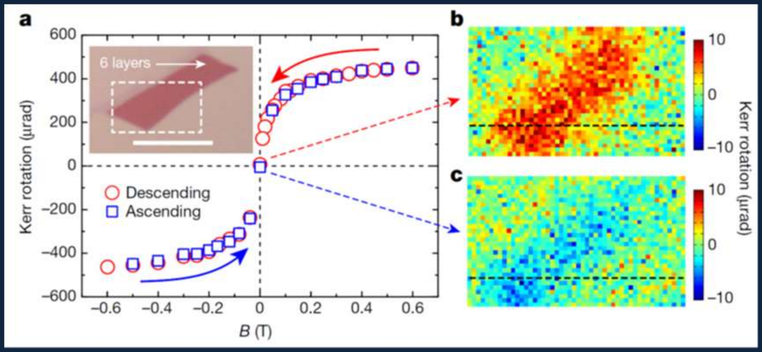
- The ferromagnetism in 2D crystals, combined with their rich electronics and optics, could lead to new discoveries and applications.
- 2D materials largely decouple from the substrates, allow electrical control, are mechanically flexible, and are open to chemical functionalization.
- 2D magnets are accessible, engineerable, and integrable into emergent heterostructures, where the interplay of distinct physical properties could give rise to emergent interfacial phenomena.
- The sensitive responses of 2D magnets allow the development of miniaturized, lightweight, flexible, and biocompatible devices based on magnetoresistive, magnetoelectric, magnetostrictive, magneto-optical, and magnetobiological effects.



Gong and Zhang, Science, 363(6428), eaav4450 (2019)

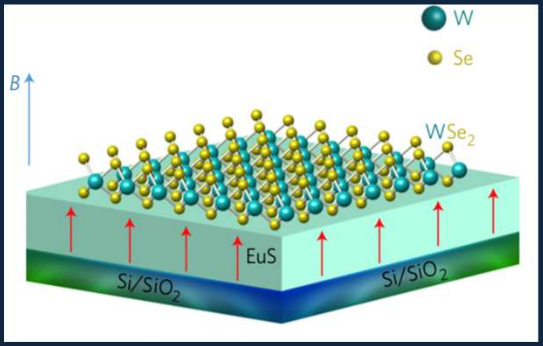
Ferromagnetism in 2D Crystals

- In early 2017, the first observations of ferromagnetism was reported. (insulator or conductor, unstable in air)
- Enhanced valley splitting was demonstrated in monolayer WSe_2 on an EuS substrate.
- 2D ferromagnet was formed by monolayer WSe_2 and 10 nm CrI_3.



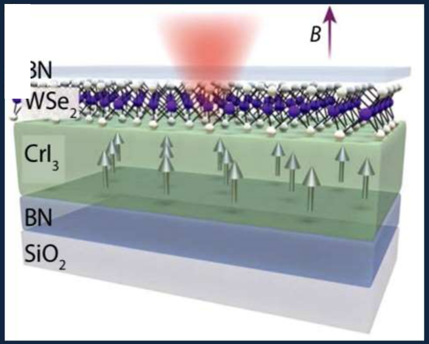
CrI_3 on EuS ➔ T_c ~ 45K

Gong, *et al.*, *Nature* **546**, 265 (2017)



WSe_2 on EuS ➔ T_c ~ 16.5K

Zhao *et al*., *Nat. Nano*., **12**, 757 (2017)



WSe_2/CrI_3 ➔ T_c ~ 61K

Zhong *et al*., *Sci. Adv*., **3**, 6586 (2017)

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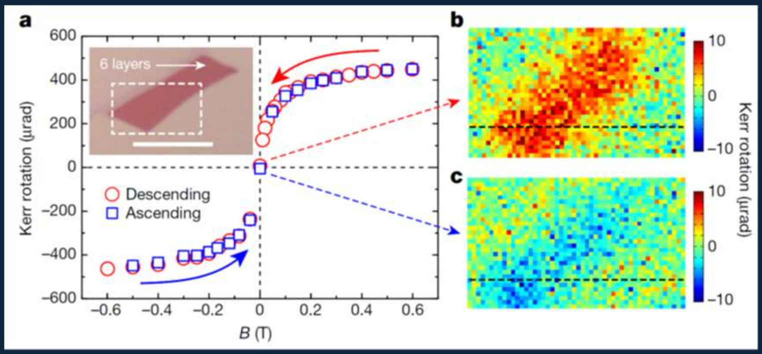

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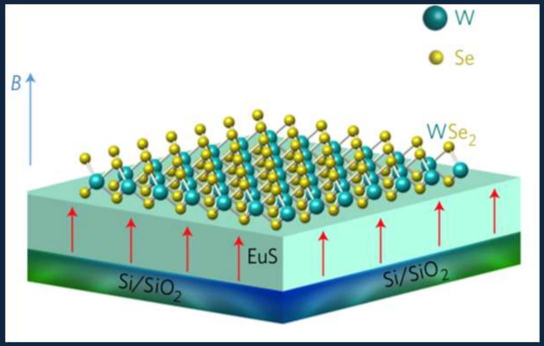

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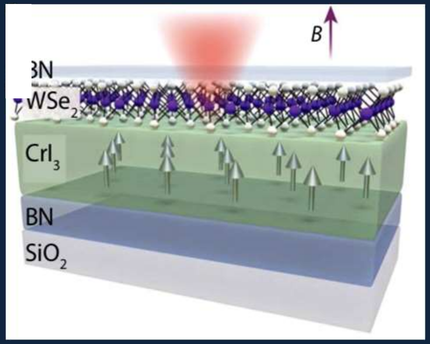
Zhao *et al.*, *Nat. Nano.*, **12**, 757 (2017)



WSe_2/CrI_3 ➔ T_c ~ 61K

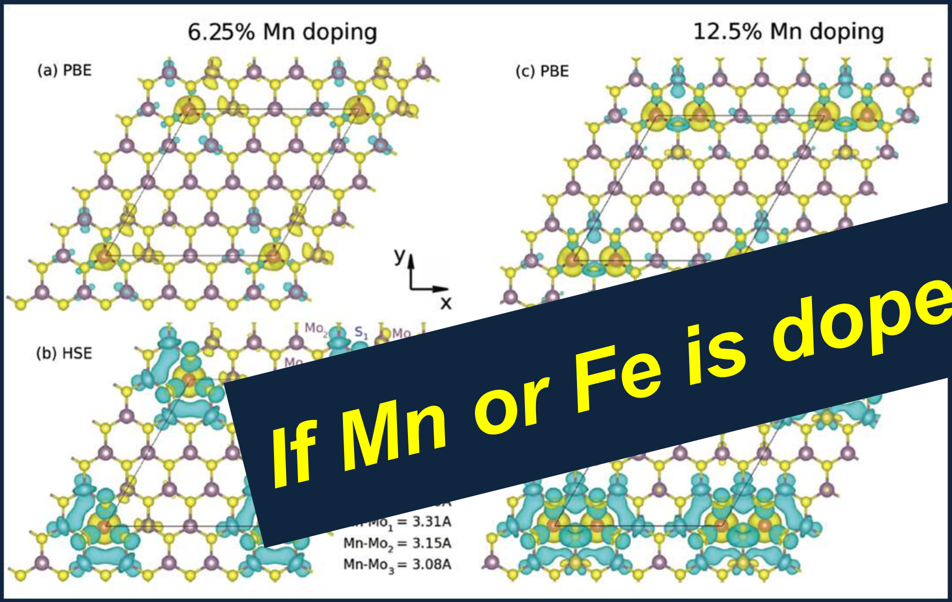
Zhong *et al.*, *Sci. Adv.*, **3**, 6586 (2017)

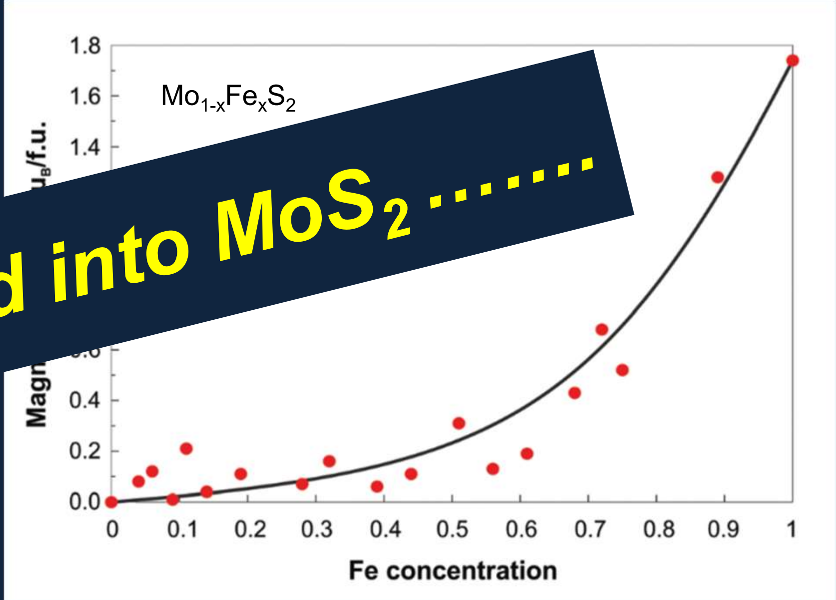
Critical Factors

- 2D Dilute Magnetic Semiconductor…
- Curie Temperature above RT…..
- Scalability (uniformity)………

Nonexistent before 2020

Theoretical Studies Predicted 2D DMS with Curie Temp above RT





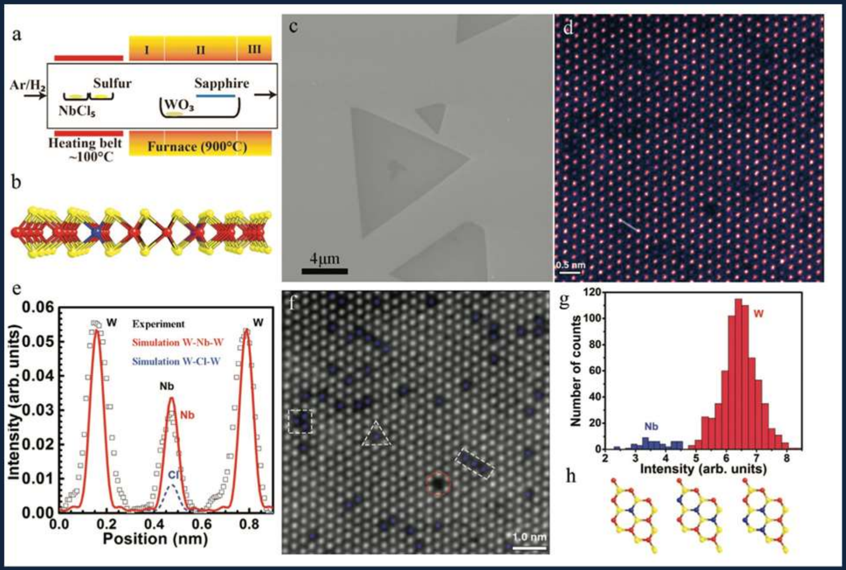
If Mn or Fe is doped into MoS_2 ……

Ramasubramaniam, Phys Rev. B. ***87***, 195201 (2013)

Antipina *et al.*, *Phys. Chem. Chem. Phys.*, **18**, 26956 (2016)

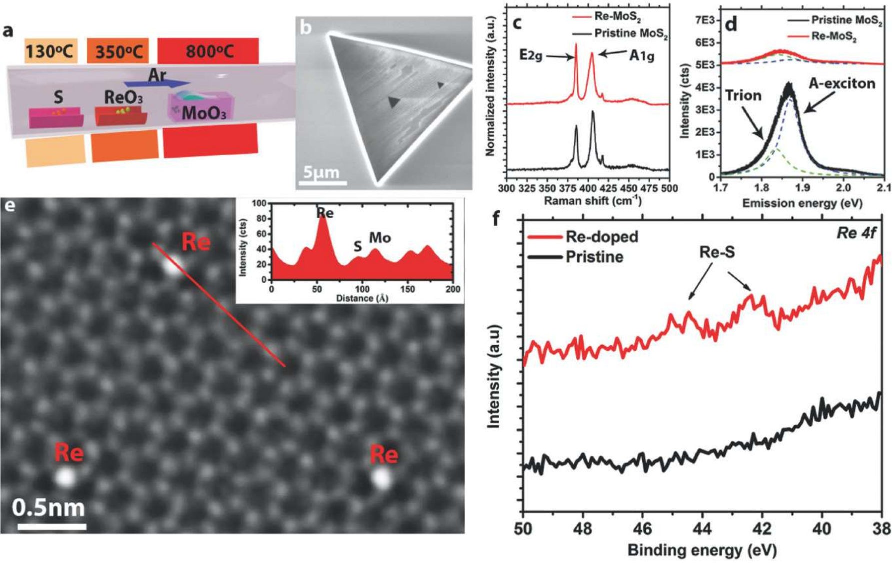
First-principles studies predict that doping of transition metal ions into TMD monolayers is a promising way to realize a 2D DMS with a **Curie temperature at or above room temperature**.

in situ doping of TMDs via CVD growth



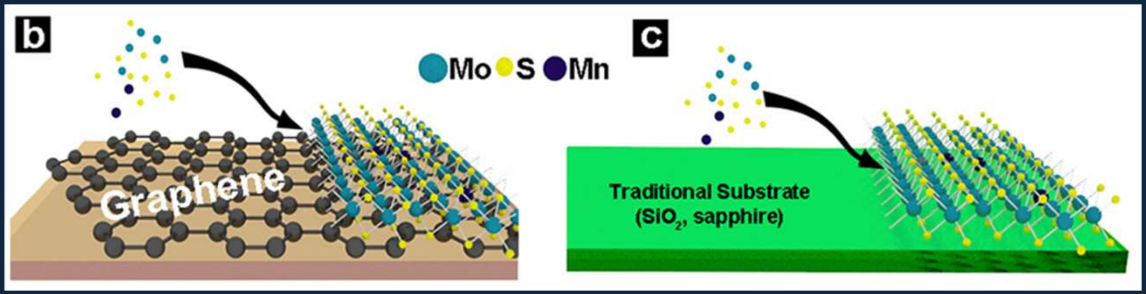
Gao *et al.*, *Adv. Mat.*, 2016, **28**, 9735-9743

6.7 at% Nb doping of WS_2 monolayer



Zhang *et al.*, *Adv. Funct. Mater.*, 2018, **28**, 1706950

1 at% Re doping of MoS_2 monolayer



Zhang *et al.*, *Nano Lett.*, 2015, **15**, 6586-6591

Enhanced 2 at% Mn doping of MoS_2 monolayer
via Graphene as a substrate

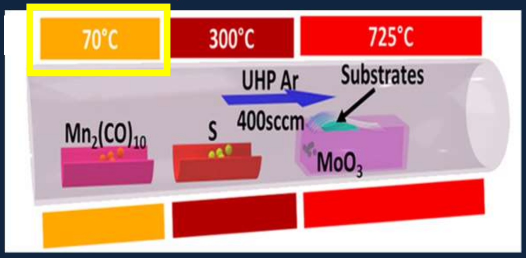
Ferromagnetism NOT observed

in situ doping of TMDs via CVD growth

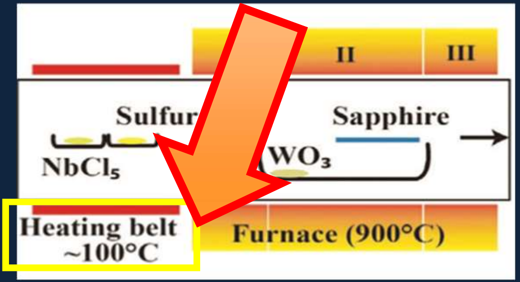
Dopant	Mn	Nb	Re
Source	$Mn_2(CO)_{10}$	$NbCl_5$	ReO_3
Melting point	154 °C	204.7°C	400 °C
Heating Temp.	70 °C	100 °C	350 °C 500 °C

5	6	7	8
23 V	24 Cr	25 Mn	26 Fe
41 Nb	42 Mo	43 Tc	
73 Ta	74 W	75 Re	

Iron oxide melting point 1,597°C



Zhang *et al., Nano Lett.*, 2015, **15**, 6586-6591



Gao *et al., Adv. Mat.*, 2016, **28**, 9735-9743

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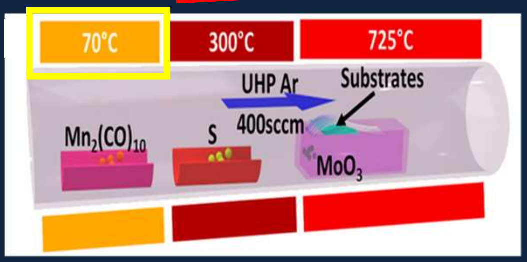
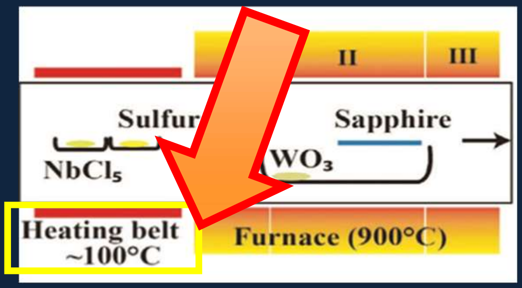


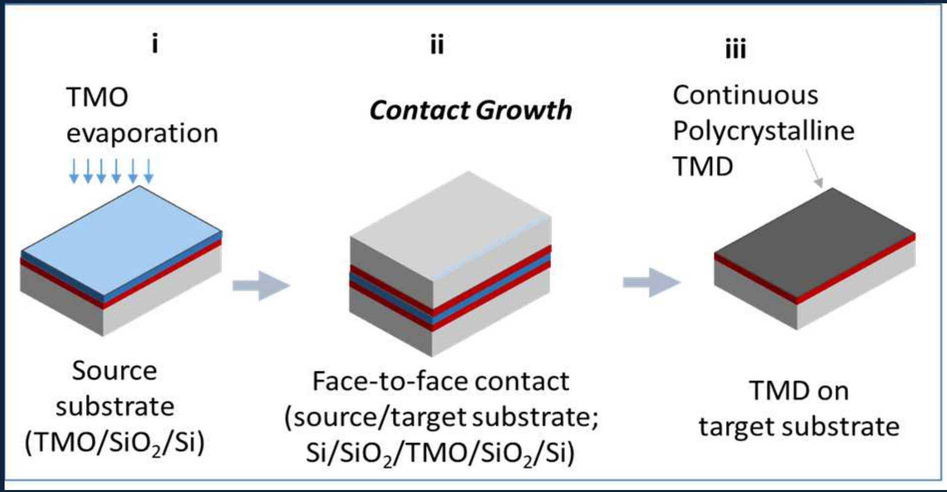
Zhang *et al., Nano Lett.*, 2015, **15**, 6586-6591

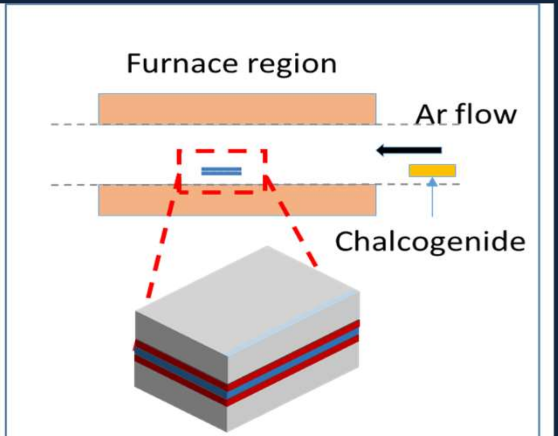


Gao *et al., Adv. Mat.*, 2016, **28**, 9735-9743

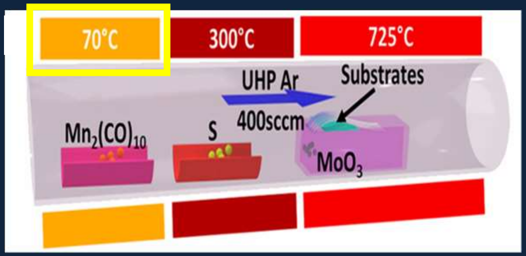
Zhang *et al., Adv. Funct. Mater.*, 2018, **28**, 1706950

Contact-Growth of TMDs

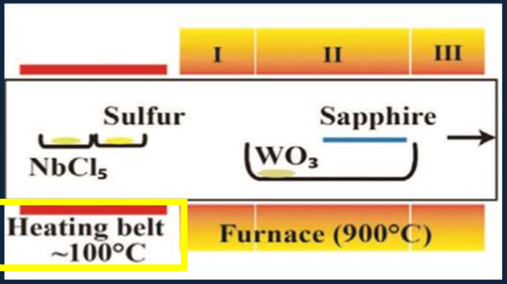




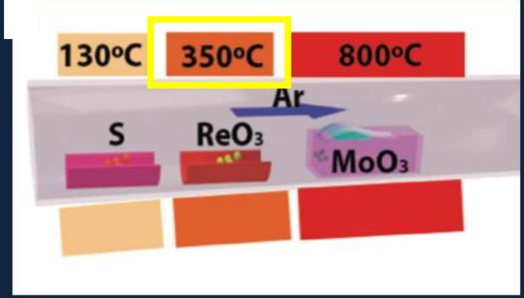
US Patent US 9,640,391 (2017)



Zhang *et al., Nano Lett.*, 2015, **15**, 6586-6591

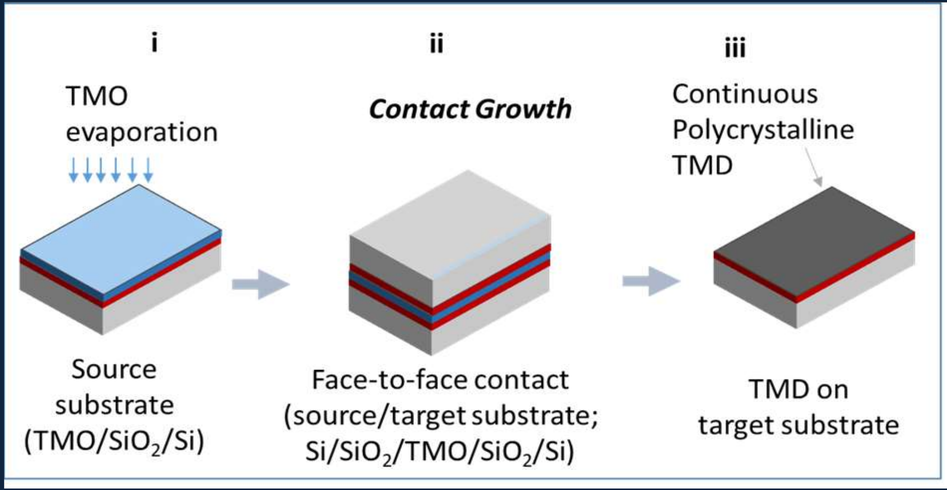


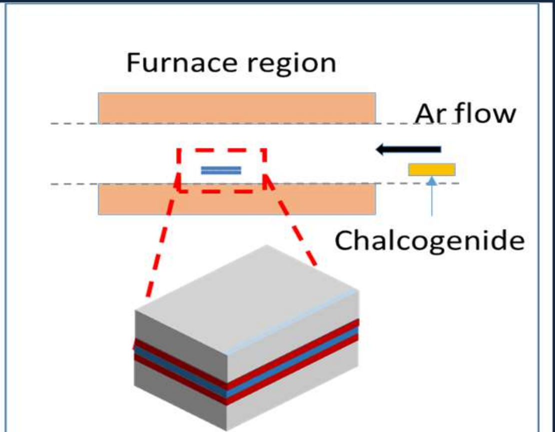
Gao *et al., Adv. Mat.*, 2016, **28**, 9735-9743



Zhang *et al., Adv. Funct. Mater.*, 2018, **28**, 1706950

Growth Mechanism

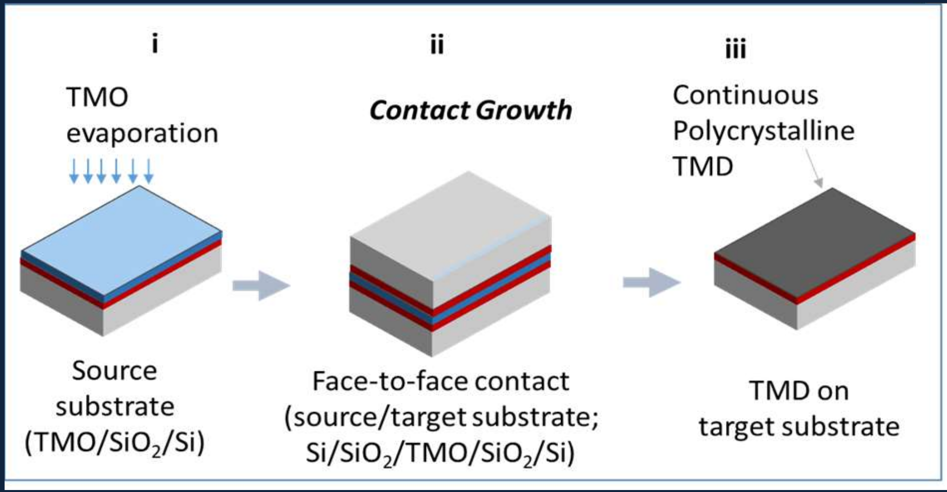


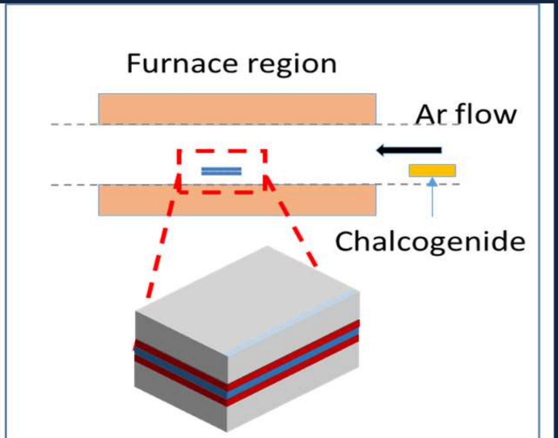


US Patent US 9,640,391 (2017)

- The TMD grow is based on the reaction of transition metal oxide and chalcogen powder in the vapor phase.
- For the MoS_2 growth, the initial reaction between MoO_3 and S produces intermediate volatile $MoO_{3-x}S_y$, and then further sulfuration of $MoO_{3-x}S_y$ on the substrate lead to a complete conversion into MoS_2 grains.

Our TMD Synthesis via LPCVD (1)

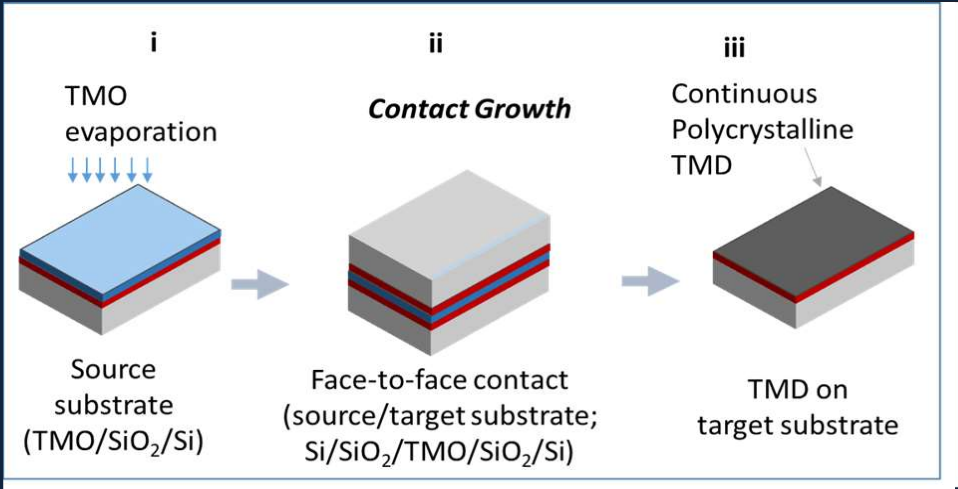


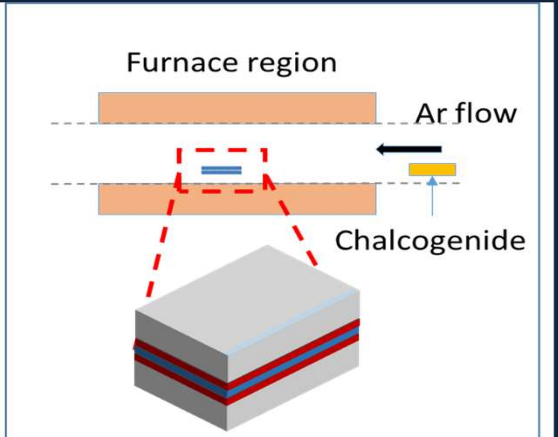


US Patent US 9,640,391 (2017)

- A thin film of MoO_3 is deposited onto a SiO_2/Si substrate, which then contacts another SiO_2/Si substrate face-to-face.
- The sandwiched sample is loaded into the tube with sulfur powder placed in the upstream of the growth area for controlled sublimation, after which the furnace is heated to grow TMDs.
- The ambient gas is purged out to the base pressure of 850 mTorr.
- Ar gas is introduced from 150 ˚C to remove ambient gas and H_2 gas is supplied from 650˚C (increasing temperature) to 700˚C (decreasing temperature).

Our TMD Synthesis via LPCVD (2)

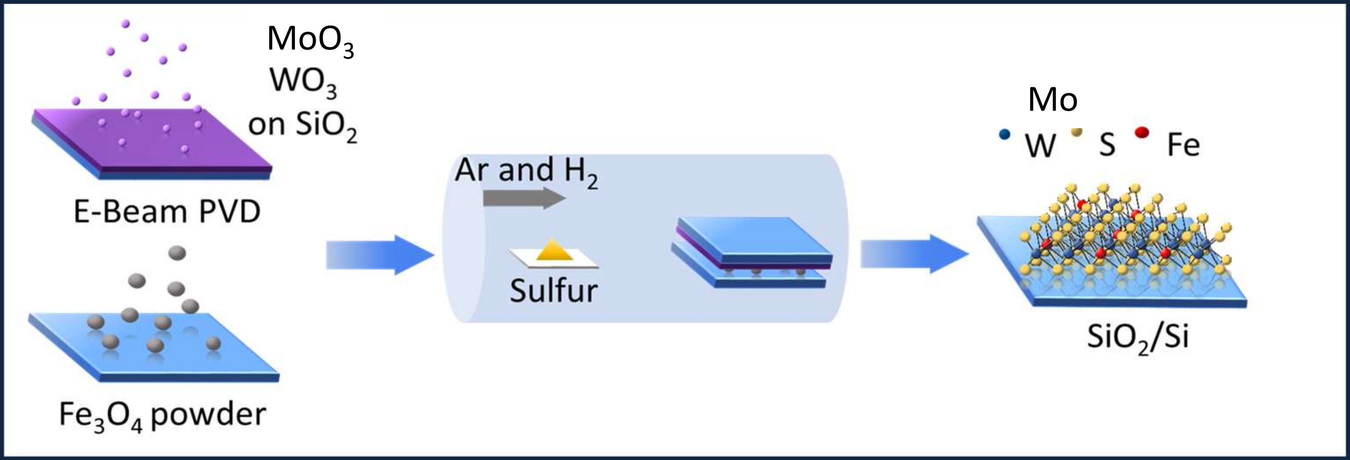




US Patent US 9,640,391 (2017)

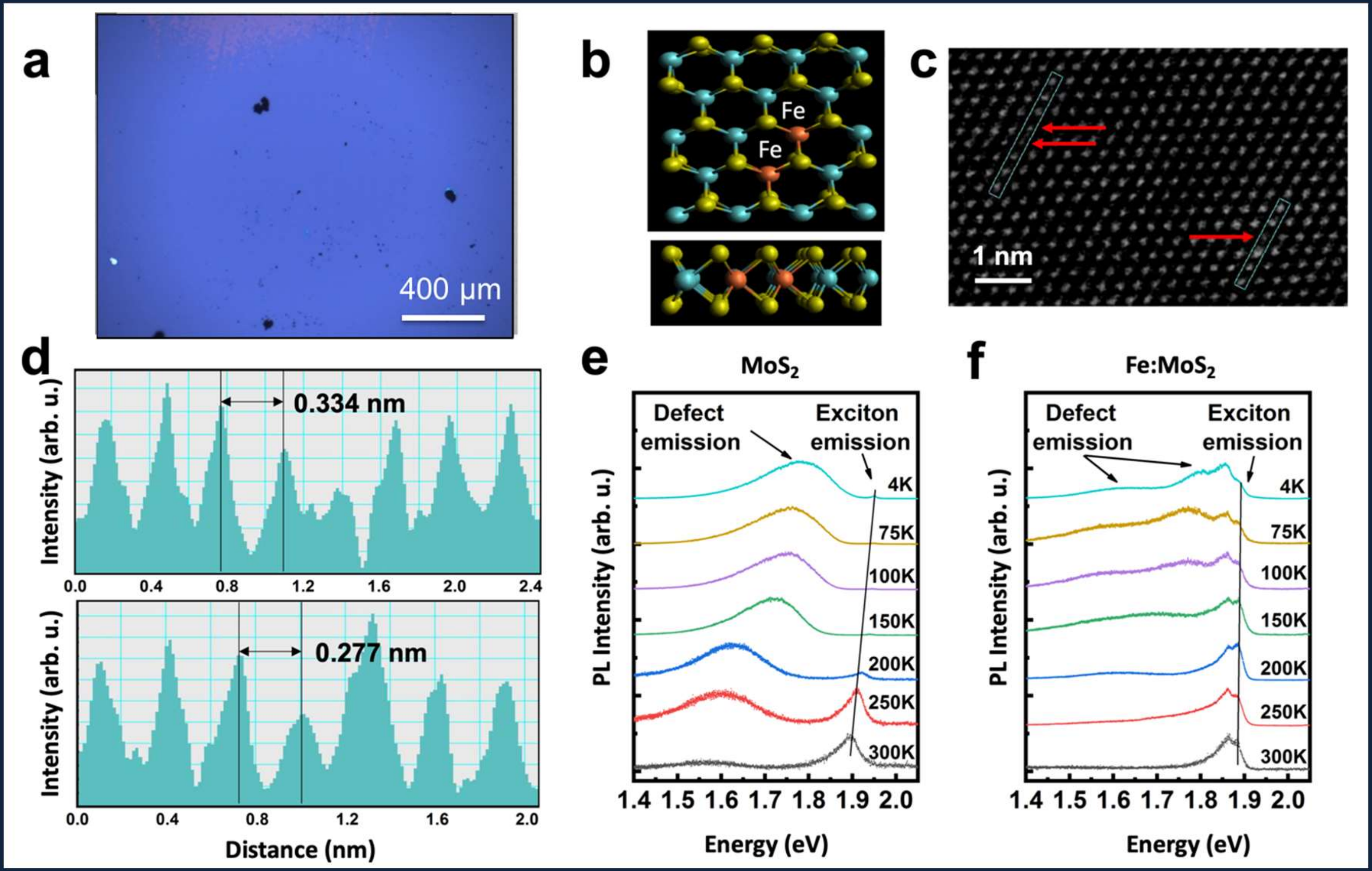
- As the furnace is ramped in temperature, the reaction proceeds via reduction of MoO_3 by hydrogen and subsequent sulfurization.
- The growth temperature is up to 850˚C.
- At an optimized location, the sulfur powder starts vaporizing at 830°C furnace temperature and sulfur powder is exhausted in 30 minutes.

In situ Fe Doping Process



- Fe_3O_4 particles are cast onto an SiO_2/Si substrate, which then contacts the MoO_3 film-deposited substrate face-to-face.
- Ar (30 sccm) and H_2 (15 sccm) are delivered at 300˚C and 760˚C, respectively.
- Sulfur is supplied when the temperature reaches 790˚C.
- Furnace is held at 850 ˚C for 20 min and cooled down to the room temperature, where Fe:MoS_2 monolayers (~ mm size) are obtained.

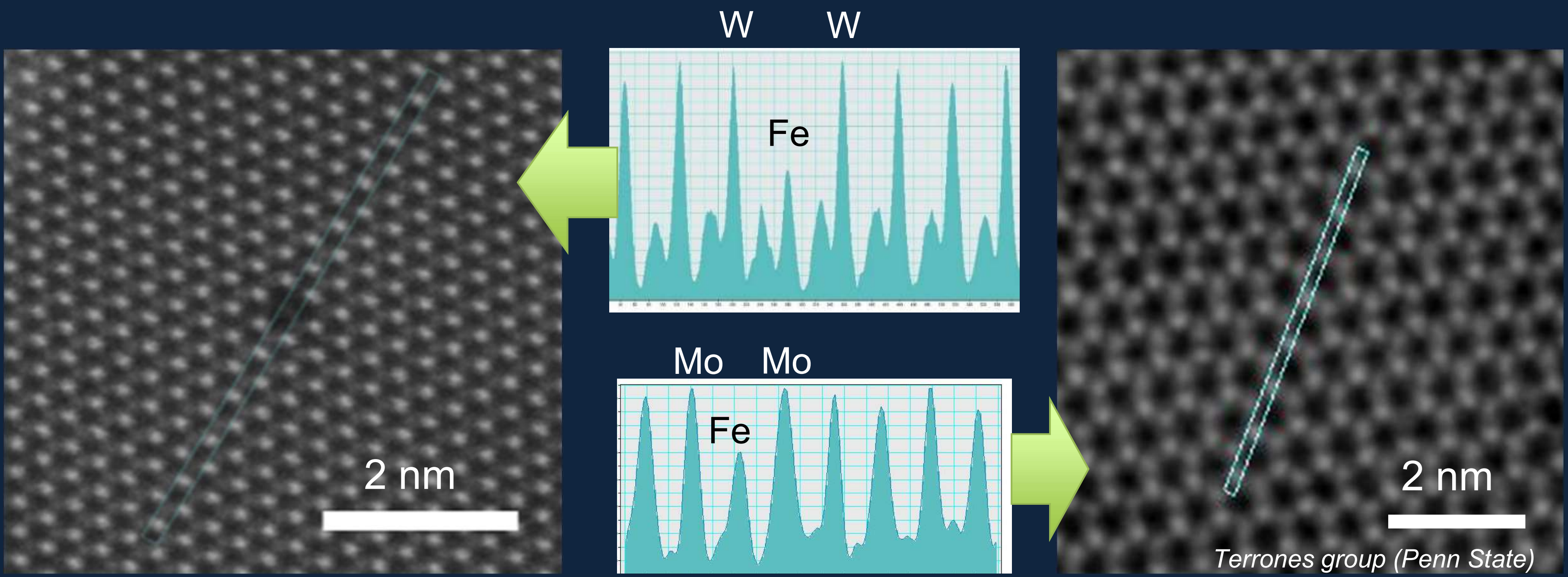
RT ferromagnetic 2D semiconductor ($Fe:MoS_2$)



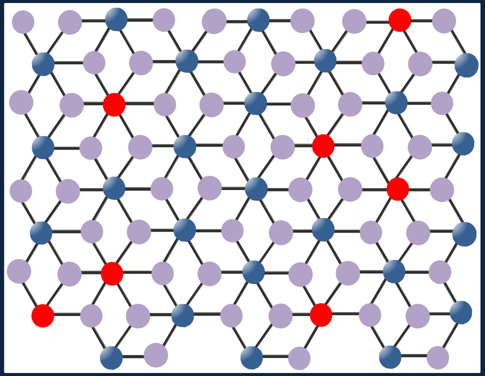
Collaboration: *Strauf (Stevens); Pasupathy (Columbia); Meunier (RPI); Vamivakas (U. Rochester); Zhang (DOE-BNL)*

Fu, *et al., Nature Communications*, 11, 2034 (2020)

Substitutional Doping of Irons



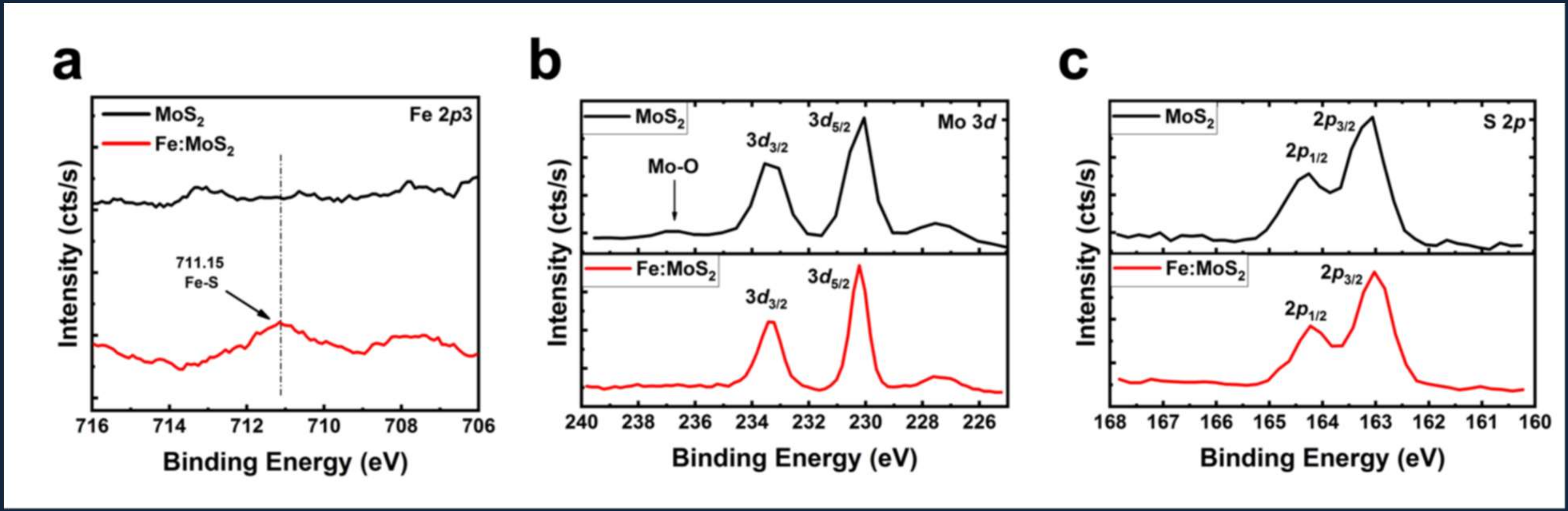
Terrones group (Penn State)



(left, right) Contrast-corrected STEM images of Fe:WS$_2$ and Fe:MoS$_2$ monolayers.
(center) STEM intensity spectra of the selected areas, where Fe atoms exhibit approximately 40% lower intensity.

Manuscript in 2nd review

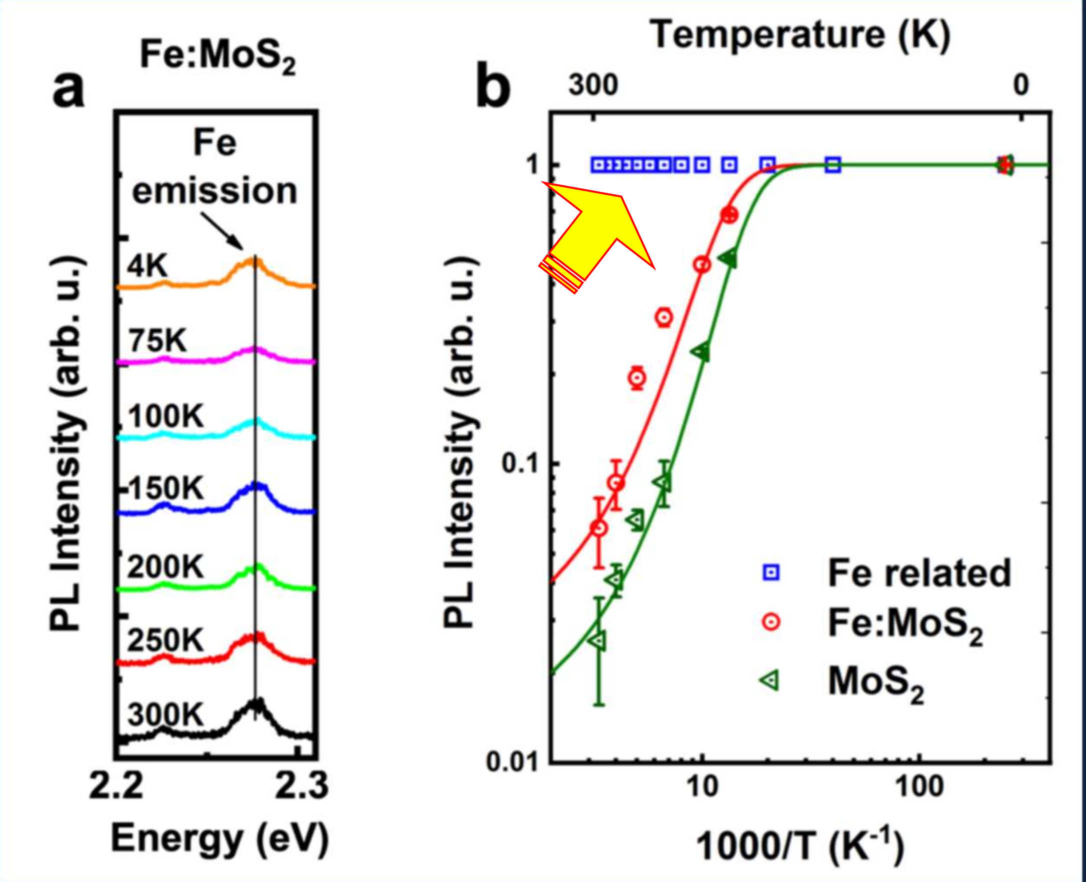
X-ray Photoelectron Spectroscopy (XPS)



(a) Fe 2*p*3 peaks; (b) Mo 3*d* peaks. The reduced intensity of the Mo-O bond in Fe:MoS_2 (237 eV) as compared to MoS_2 is indicative of an underlying reduction of the sulfur vacancy concentration upon doping; (c) S 2*p* peaks.

Fu, *et al.*, *Nature Communications*, 11, 2034 (2020)

Temperature dependent PL spectra of Fe related emission



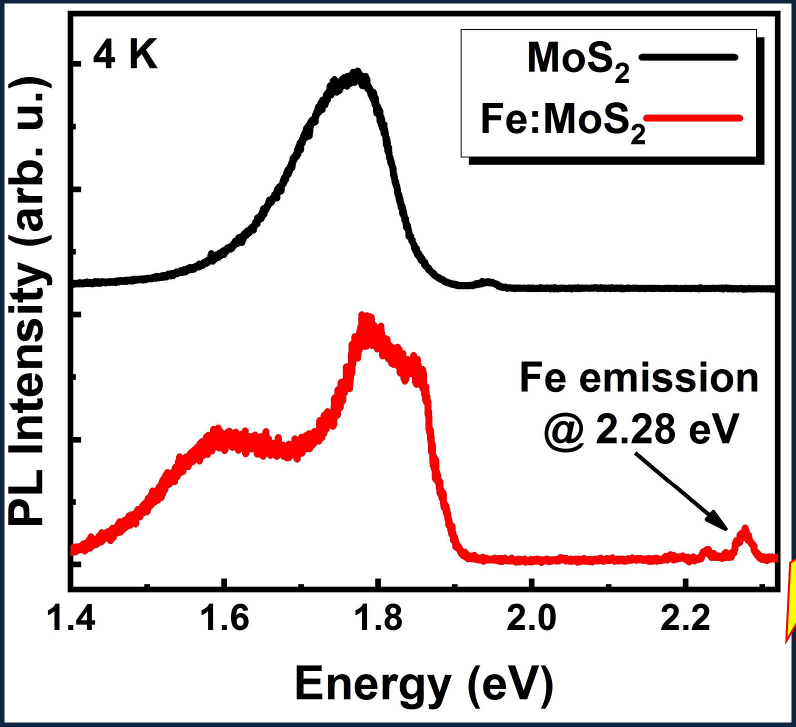
Measured at Strauf Lab (Stevens)

- Integrated PL for the bandgap emission in MoS_2 (green triangle), Fe:MoS_2 (red circle) and for the Fe-related emission (blue square).
- The solid red and green lines are standard Arrhenius fits for the exciton emission.

Fu, *et al.*, *Nature Communications*, 11, 2034 (2020)

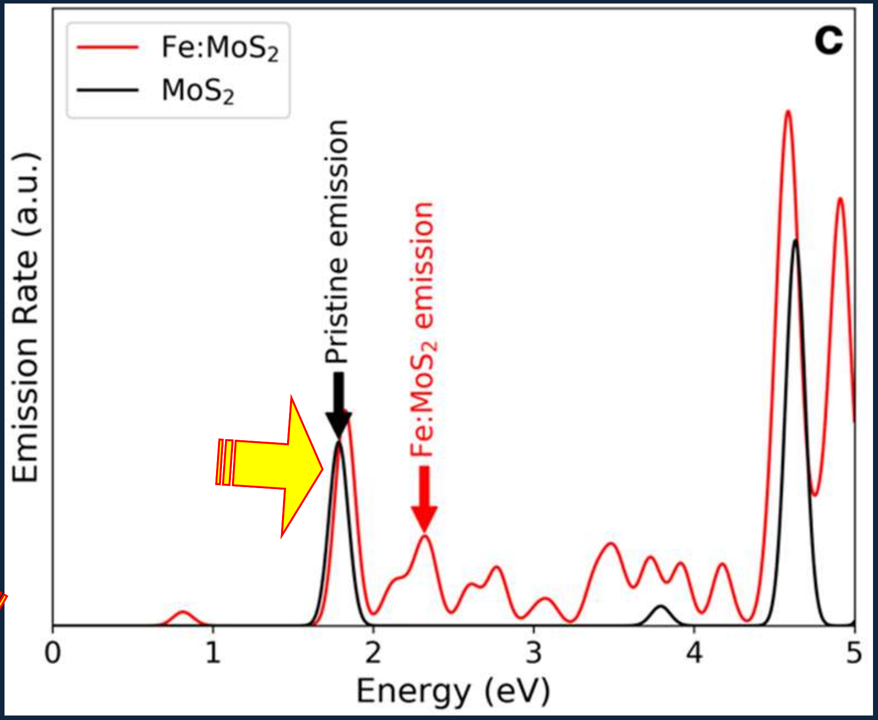
Fe-related Emission from Fe:MoS$_2$

PL spectra of MoS$_2$ and Fe:MoS$_2$



Measured at Strauf Lab (Stevens)

DFT calculations of dipole-allowed transitions

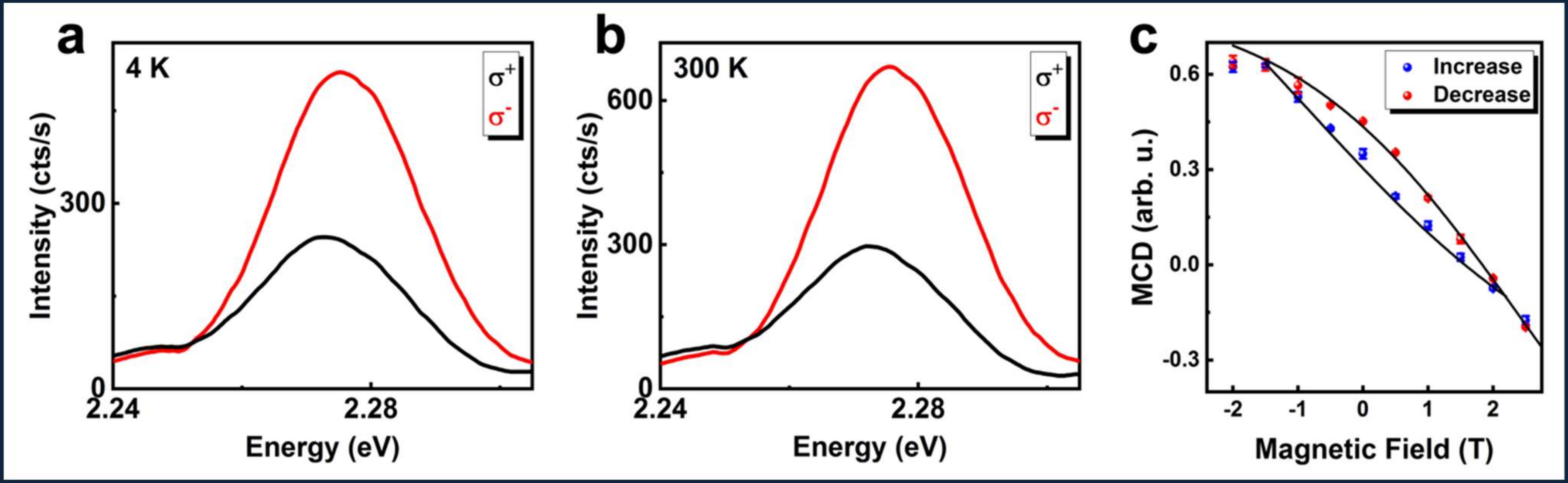


Meunier group (RPI)

Fu, *et al., Nature Communications*, 11, 2034 (2020)

Magneto-Photoluminescence Measurements

- The transition metals' luminescence loses its CD above T_C ➔ CD at 300K suggests that Fe:MoS_2 is ferromagnetic at RT.
- The light absorption is closely related to the Zeeman shifts - pronounced hysteresis loop ➔ ferromagnetic nature of Fe-related emission

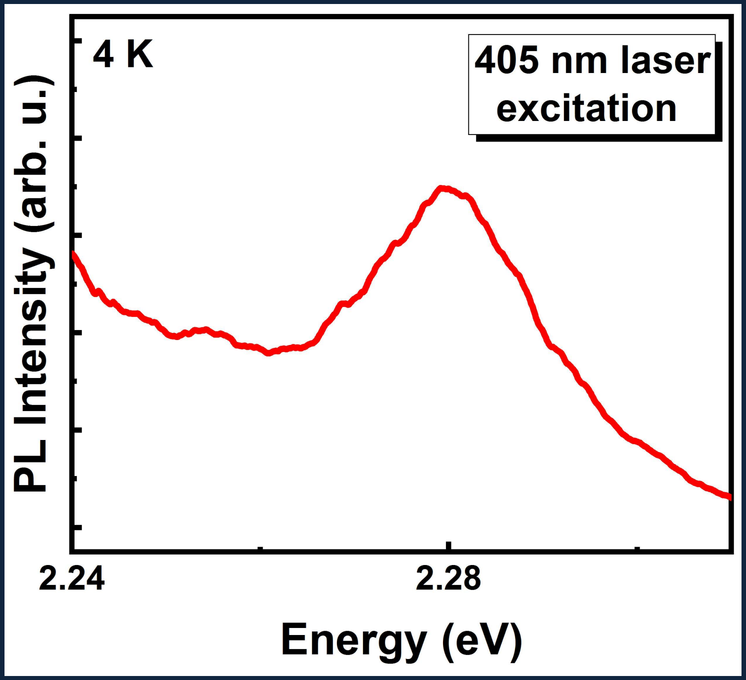


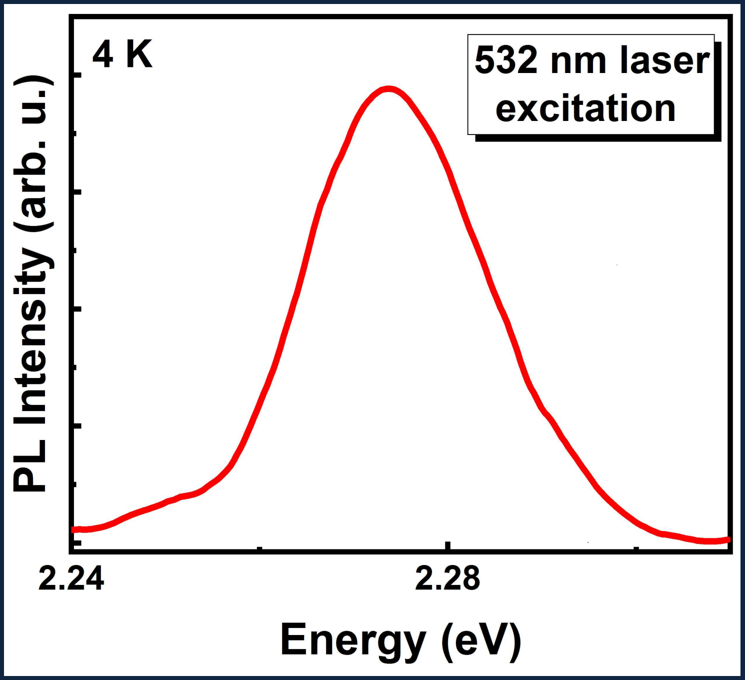
Measured at Strauf Lab (Stevens)

Fu, *et al.*, *Nature Communications*, 11, 2034 (2020)

Fe-related Emission from Fe:MoS$_2$

Strauf group (Stevens)

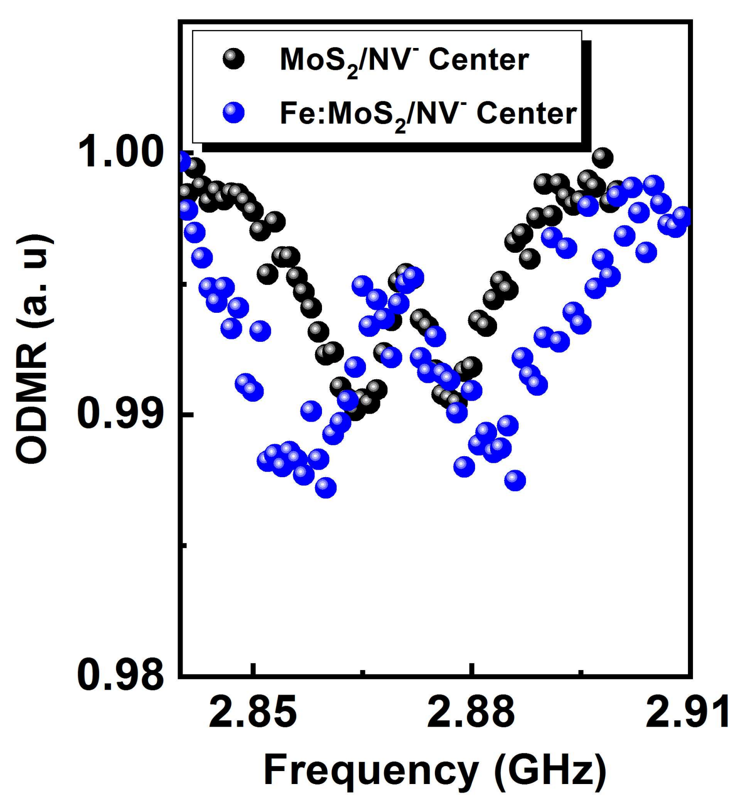




This Fe-related emission peak is consistent at different excitation wavelengths, showing that this emission is not related to a Raman peak.

Local Magnetic Field of Fe:MoS_2 Monolayers

ODMR spectra of the NV^- centers



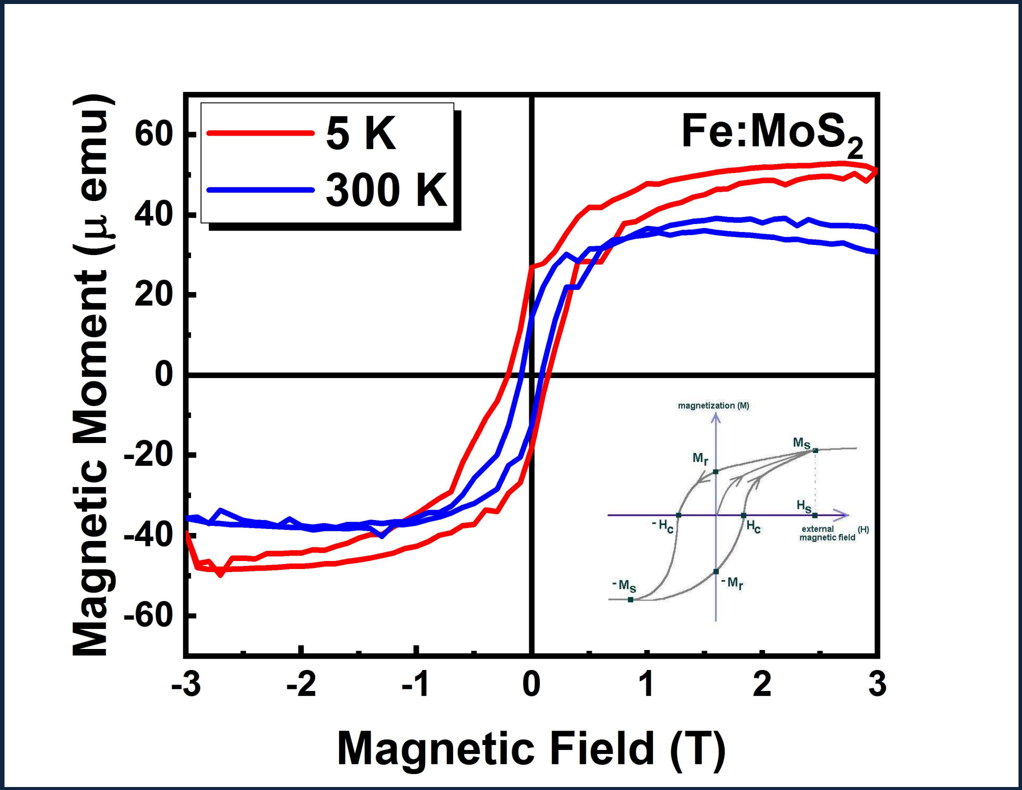
Vamivakas group (U. Rochester)

- We detect the ODMR of electron spins of NV^- centers manipulated by simultaneous MW radiation.
- The increase of Zeeman splitting from 10 MHz to 21 MHz in the vicinity of Fe:MoS_2 monolayers indicates that a local magnetic field presents in the monolayers at RT.
- The local magnetic field was up to 0.5 mT, comparable to the values measured in 2D CrI_3 and $CrBr_3$ at cryogenic temperature.

Fu, *et al., Nature Communications*, 11, 2034 (2020)

Superconducting Quantum Interference Device Measurement

SQUID Results

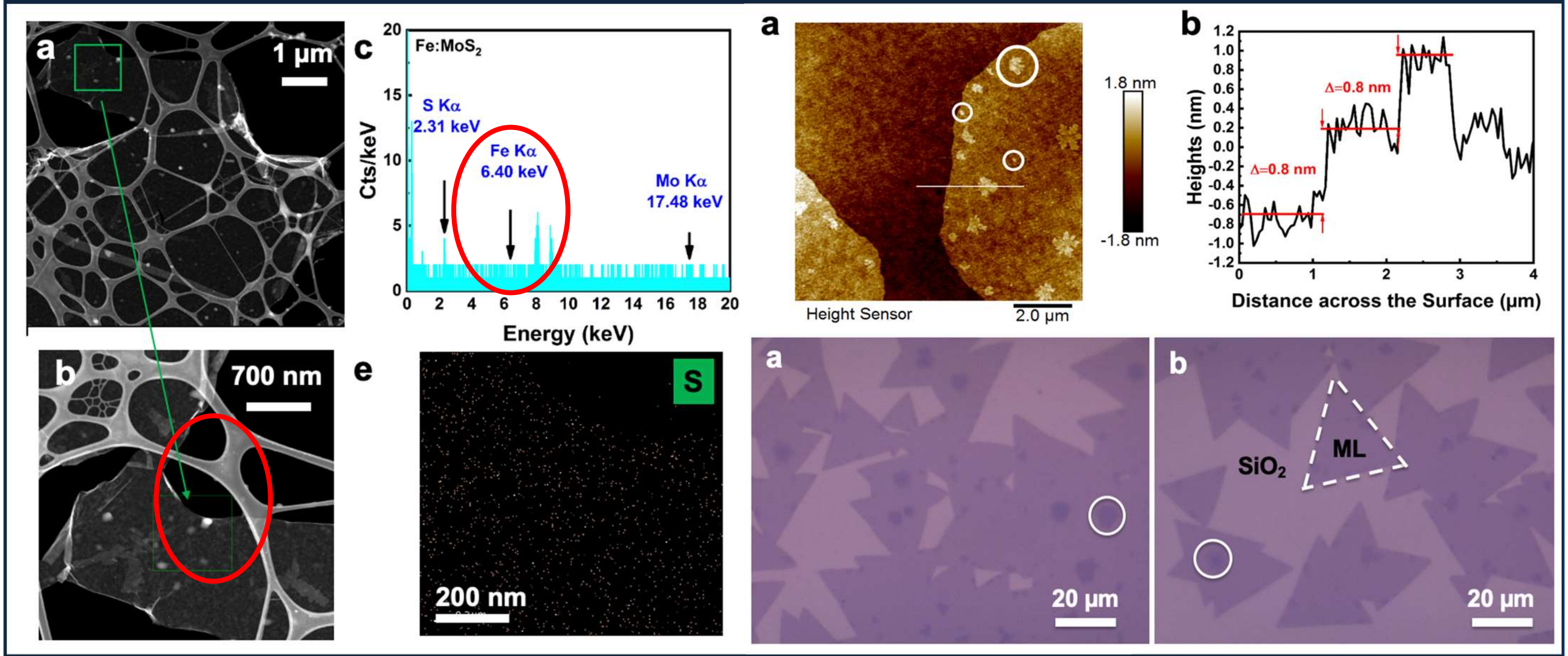


Pasupathy group (Columbia)

- Fe:MoS$_2$ monolayers exhibit a pronounced *M-H* hysteresis loop at both cryogenic and room temperatures.
- The magnitude of the hysteresis loop decreases with increasing temperature, while the T_C has not been reached at 300 K.

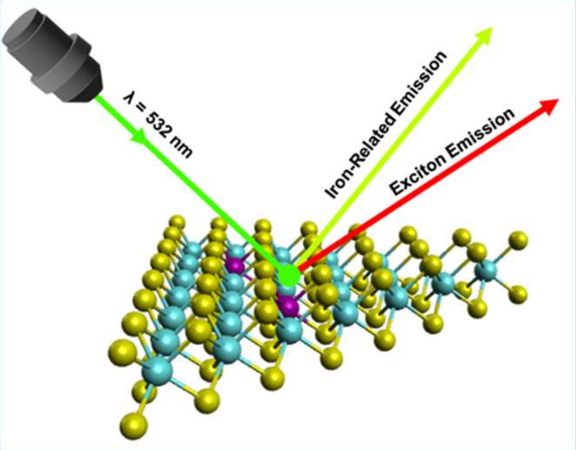
Fu, *et al., Nature Communications*, 11, 2034 (2020)

Confirmation of Substitutional Doping – I

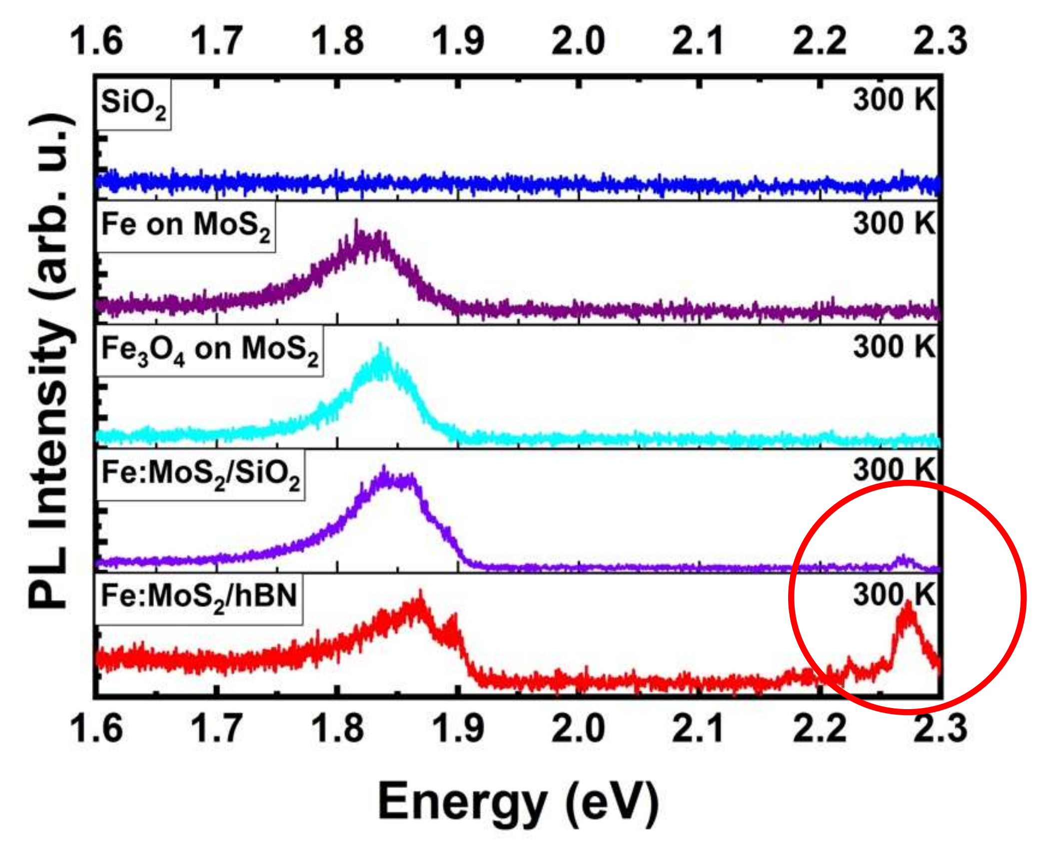


Fu, *et al., Nature Communications*, 11, 2034 (2020)

Confirmation of Substitutional Doping – II

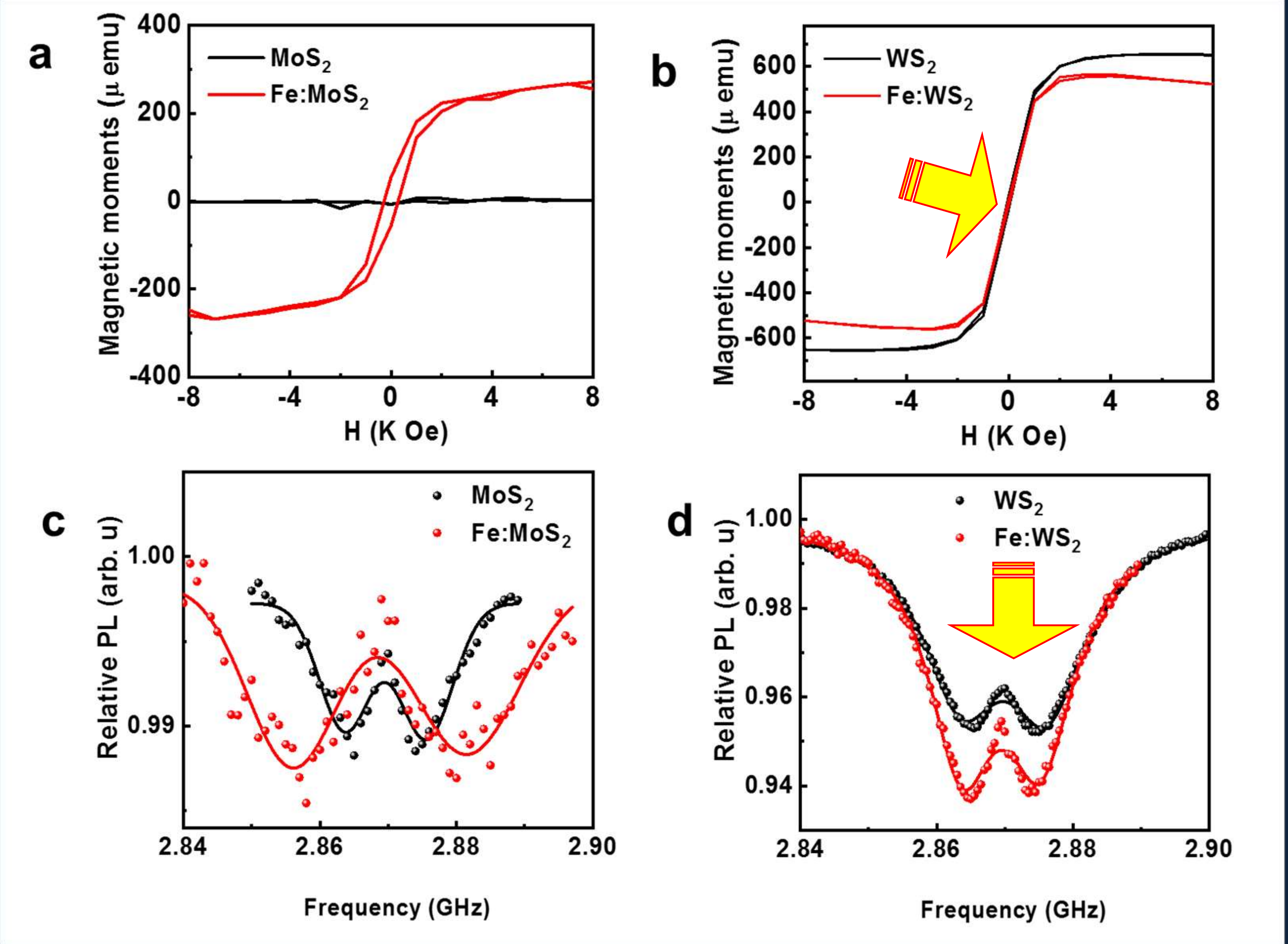


Measured at Strauf Lab (Stevens)



Fu, *et al., Nature Communications*, 11, 2034 (2020)

Confirmation of Substitutional Doping – III

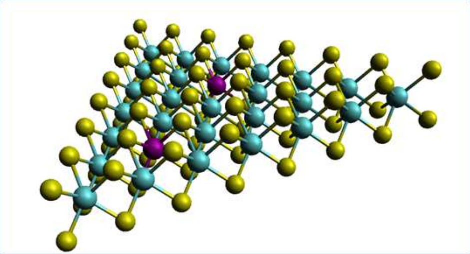


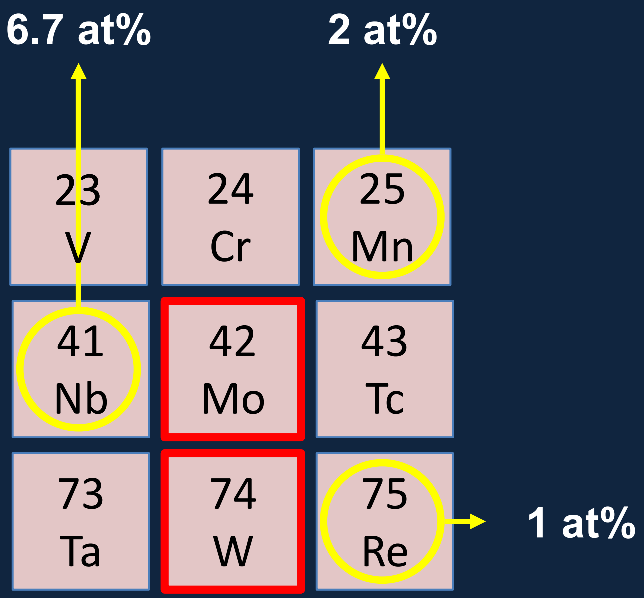
Measured at Argonne National Lab (Oakland University)

Vamivakas group (U. Rochester)

Manuscript in 2nd review

Low-Doping Concentration in 2D Materials




6.7 at%

2 at%

23 V

24 Cr

25 Mn

41 Nb

42 Mo

43 Tc

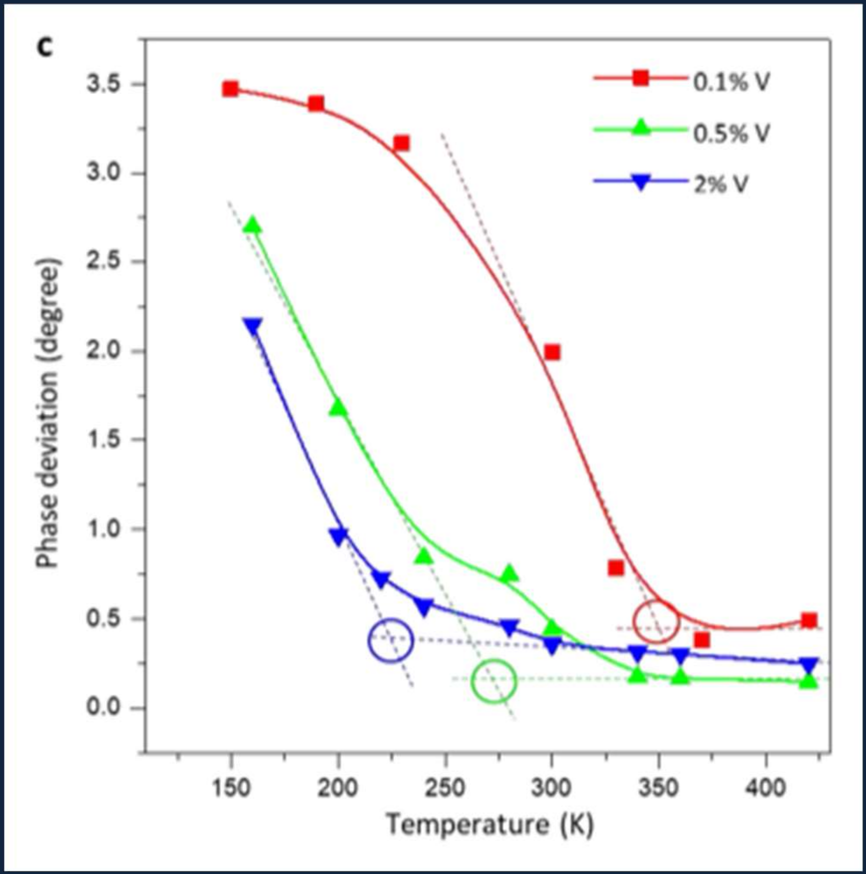
73 Ta

74 W

75 Re

1 at%

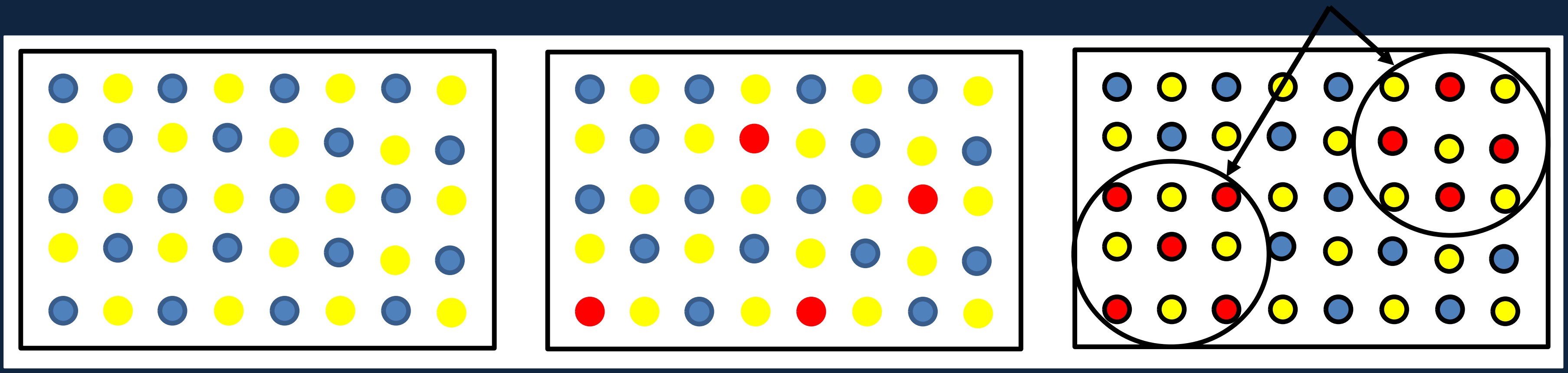
Zhang *et al., Nano Lett.* **15**, 6586-6591 (2015)
Gao *et al., Adv. Mater.* **28**, 9735-9743 (2016)
Zhang *et al., Adv. Funct. Mater.* **28**, 1706950 (2018)



Vanadium-doped WSe_2

Yun, *et al., Adv. Sci.* 1903076 (2020)
Duong, *et al., Appl. Phys. Lett.* **115**, 242406 (2019)

Segregation of Impurities



Are they (ZnMn)O or MnO?

ZnO (Semiconductor)

(ZnMn)O (DMS) ?

Segregation of impurities

“Ferromagnetic behavior of ZnO lightly doped with Mn coincides with the presence of MnO nanoparticles, whereas cluster-free Mn doped ZnO behaves paramagnetically.”

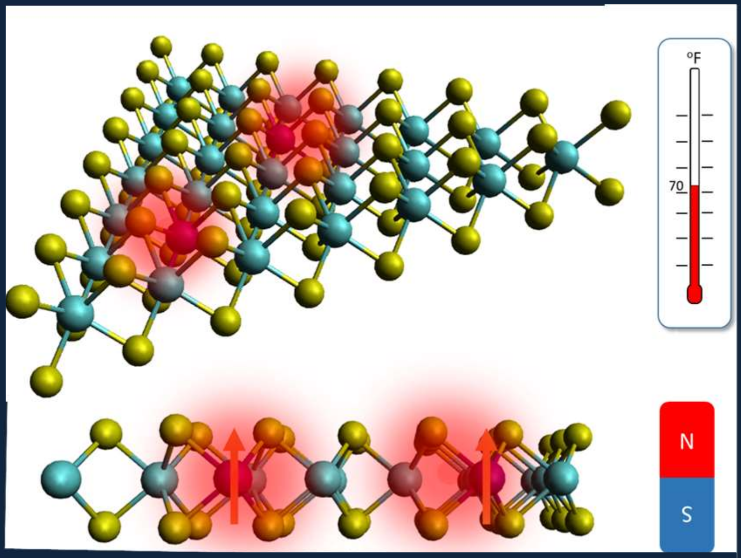
Lancon, *et al. Appl. Phys. Lett.* 109, 252405 (2016)

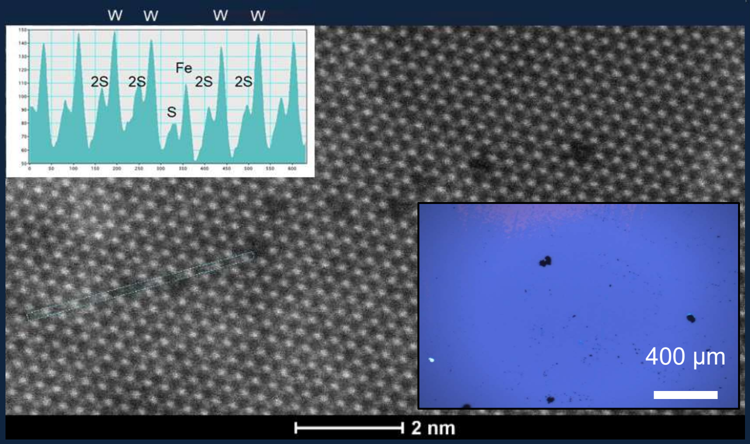
Next Experiments in Line

- Anomalous Hall Effects
- STT-MRAM
- Gate modulation of ferromagnetism
- …….

Perspective

- The magnetic field strength in this material is 0.5 mT at room temperature.
- While it cannot pick up a paper clip, it can alter the spin of electrons!
- 2D magnetic semiconductors will allow the development of transistors to control of the spin of electrons, while the whole device remains lightweight, flexible and transparent.
- This work could lead to a new era of information technologies with exciting applications in computing, sensing, and data storage





Summary

- We investigate chemical vapor deposition (CVD)-growth, including MoS_2, WS_2, WSe_2 and $MoSe_2$, as well as their heterostructures.
- We illuminate the role of dissimilar 2D substrates, uncovering the conditions for anti-oxidation.
- We demonstrate ferromagnetism with a Curie temperature above room temperature in monolayer MoS_2 via *in situ* iron-doping and measured local magnetic field strength up to 0.5±0.1 mT.
- This research unlocks new opportunities towards atomically thin magneto-optical and magnetoelectric devices for ultracompact spintronics, on-chip optical communications, and quantum computing.

Acknowledgements

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Collaborators: *Strauf, Huang, Zhang, Wang (Stevens); Rahimi-Iman (Philipps-Universitat Marburg, Germany); Pasupathy (Columbia); Datta (NJIT); Terrones (PSU); Meunier (RPI); Vamivakas (U. Rochester); Zhang (DOE-BNL)*