Oak Ridge National Laboratory

Revealing Magnetic and Topological Quantum States with Scanning Tunneling Microscopy (STM)

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US QIS Summer School

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Outline

- Introductions on STM technique and the STM group in ORNL
- Image and manipulate magnetic skyrmion bubbles in a van der Waals ferromagnet Fe_3GeTe_2 using SP-STM
- Detect spin-momentum locked conductance through topological surface states on Bi_2Te_2Se using SP-4-Probe STM

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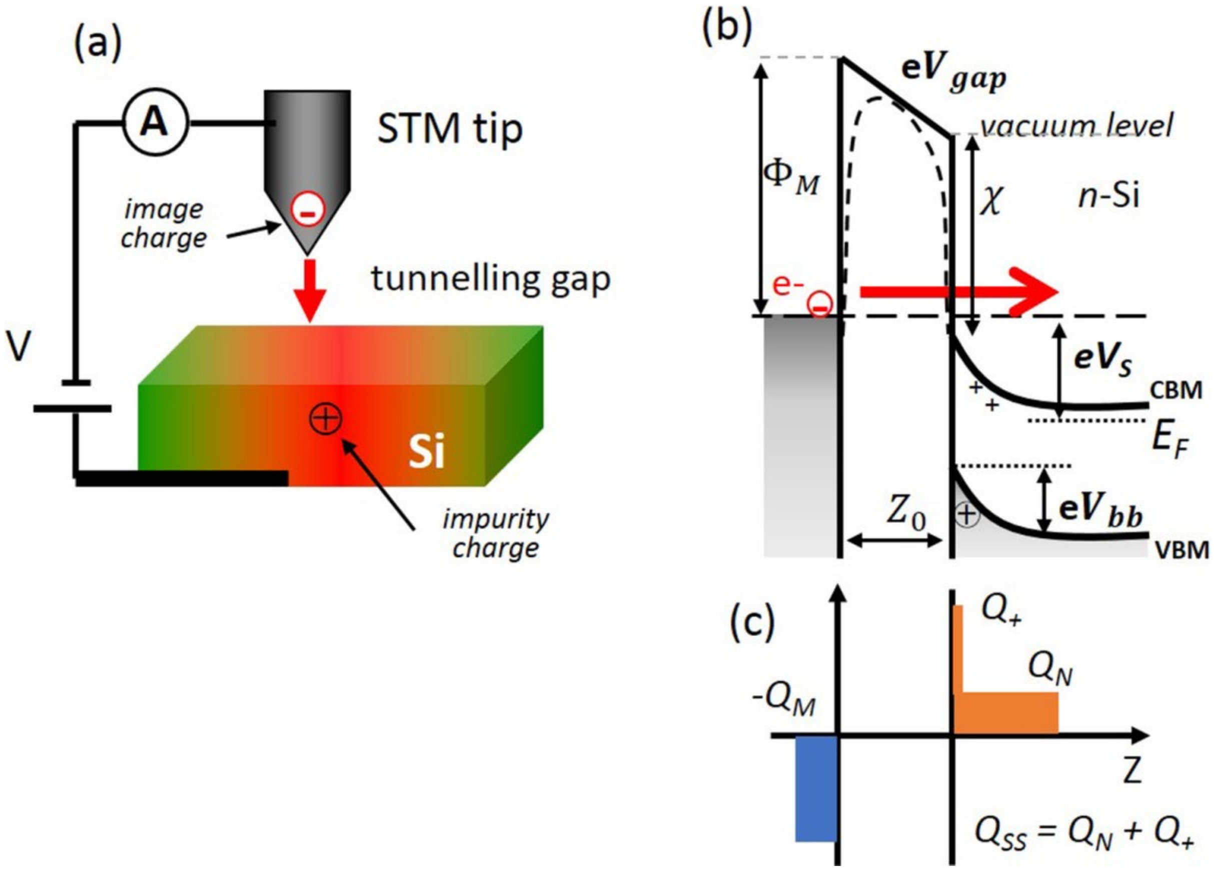
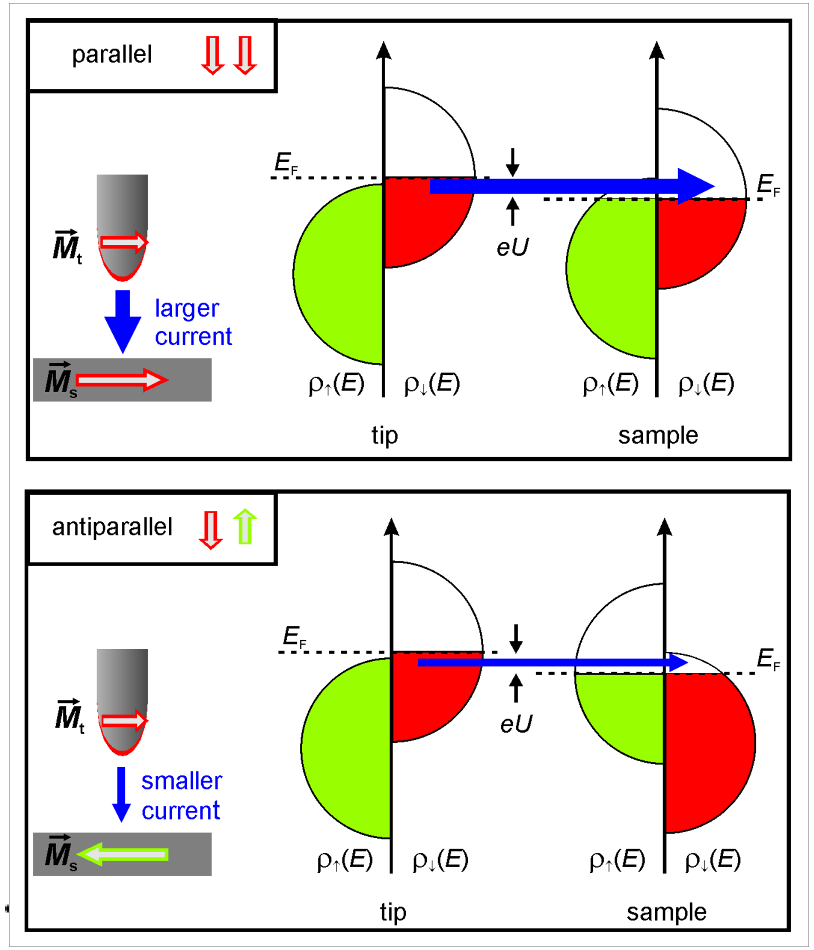
Electronic tunneling spectroscopy

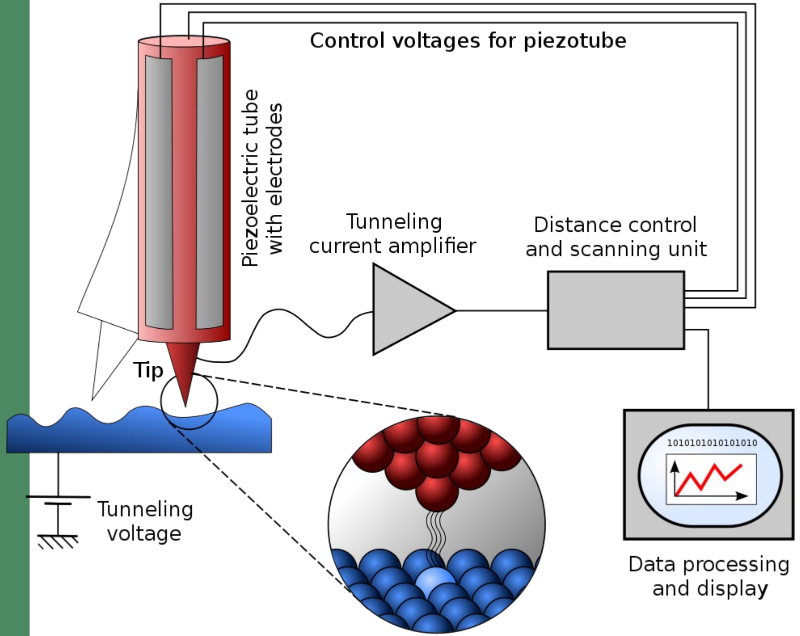




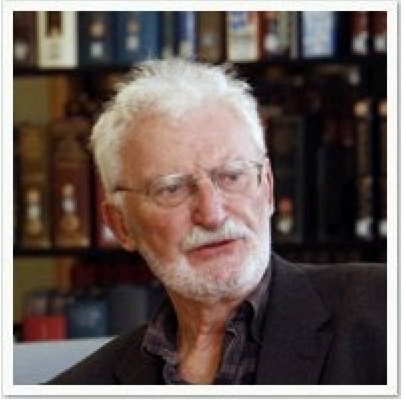
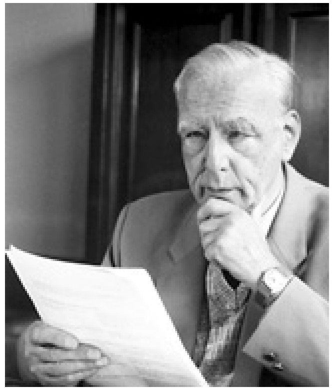
$$I_t = \frac{4\pi e}{\hbar}\int_0^{eV} \rho_{\mathrm{S}}(E_{\mathrm{F}} - eV + \varepsilon)\,\rho_{\mathrm{T}}(E_{\mathrm{F}} + \varepsilon)\,|M|^2\, d\varepsilon.$$

Scanning Tunneling Microscopy (STM)

Developed in 1981



Nobel Prize in Physics 1986 for STM and electron microscopy

Gerd Binnig | Heinrich Rohrer | Ernst Ruska

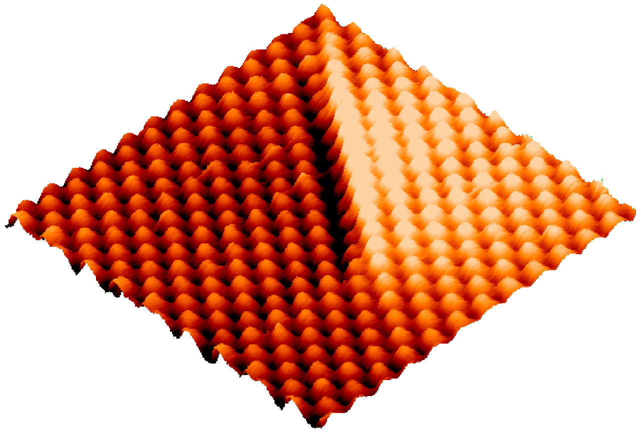
STM | EM

STM is the instrument for imaging surfaces at the atomic level

History of STM at ORNL and STM Group Science

STM History at ORNL:

Topography



Dislocation, J. Wendelken, ORNL, 1995

Electronic density of states

Charge density wave,
W. Plummer, ORNL, 1996

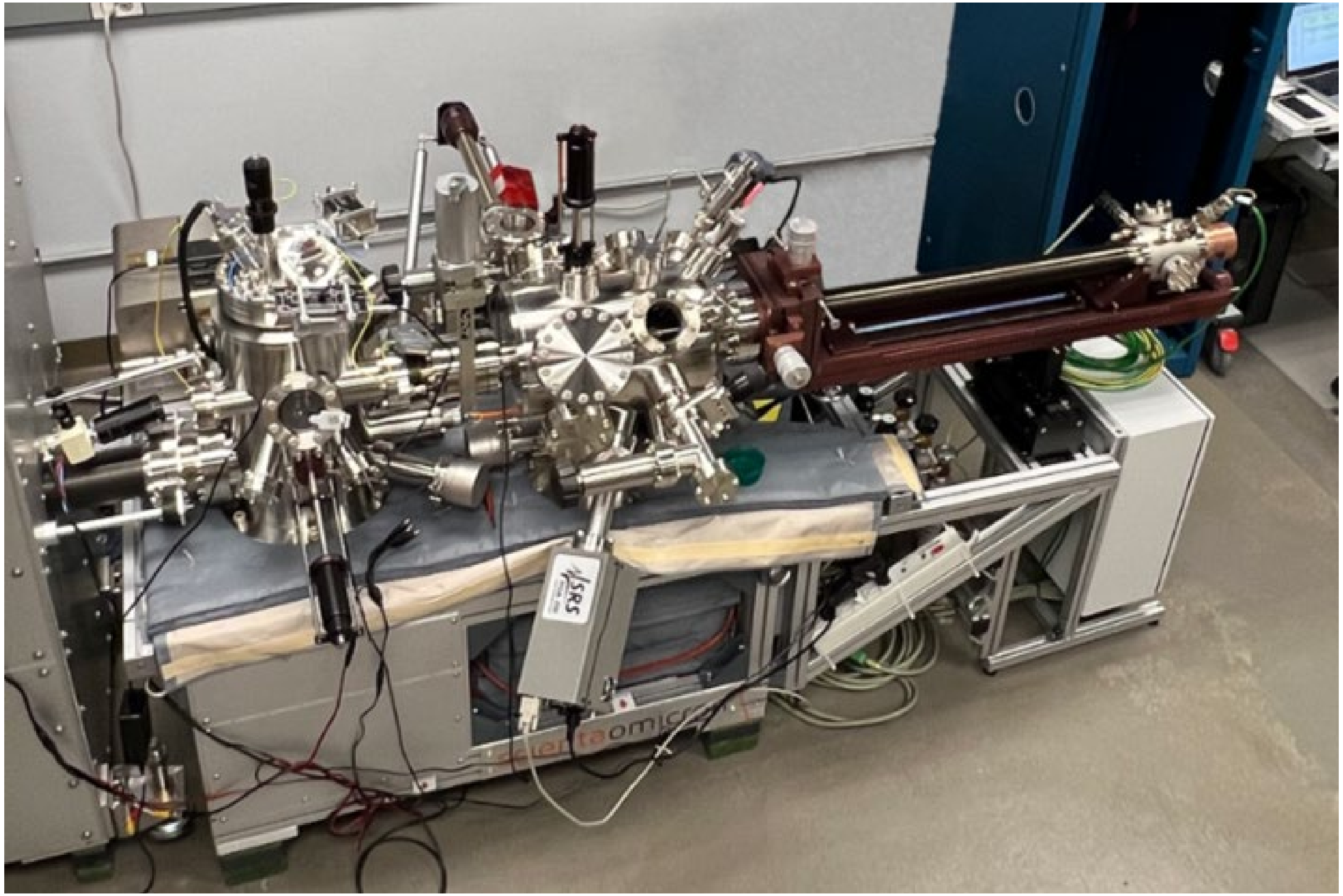
Manipulation

Atom manipulation,
W. Plummer, ORNL, 1995

STM Group Science:

Observing, characterizing, and manipulating atoms and nanostructures on materials to elucidate complex correlations of the atomic constituents and their fundamental electronic properties

A typical STM



Features of ORNL Infinity LT-STM

- STM/STS from 10-300 K with atomic resolution
- q-Plus AFM
- Nanonis control system with CNMS developed control and analysis tools
- High temperature tip cleaning
- Ar ion sample sputtering annealing to 800 K
- MBE deposition sources
- Controlled gas exposures
- Low Energy Electron Diffraction (LEED)

ORNL STM Group

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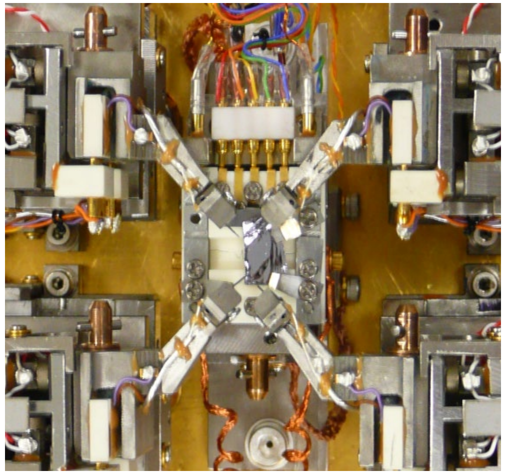
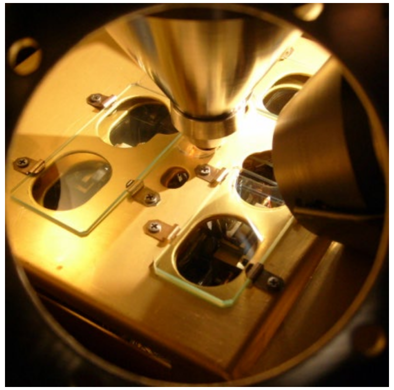
https://www.ornl.gov/group/scanning-tunneling-microscopy

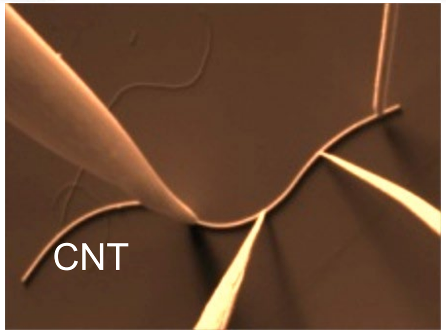
Cryogenic Four-Probe STM

ORNL 4-Probe STM
Installed in 2006



Unisoku/RHK QuadraProbe STM



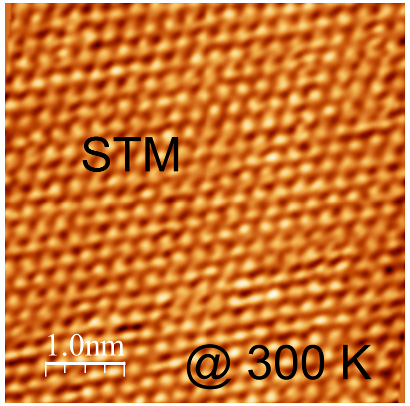


- Cryogenic (10-300 K)
- Atomic resolution imaging, spectroscopy
- Nanoscale transport
- Nanomanipulation and nanofabrication
- SEM/MBE/SAM
- Tuning fork-AFM/STM

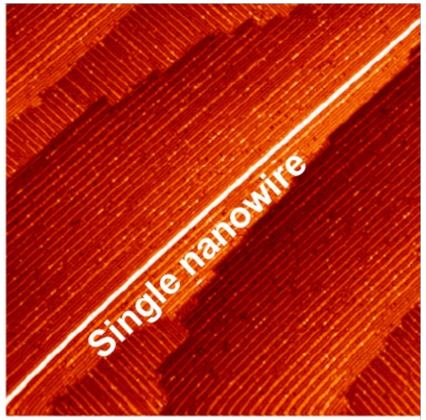
Kim et al., Rev. Sci. Instrum. 78, 12370 (2007)

Imaging, spectroscopy, and manipulation

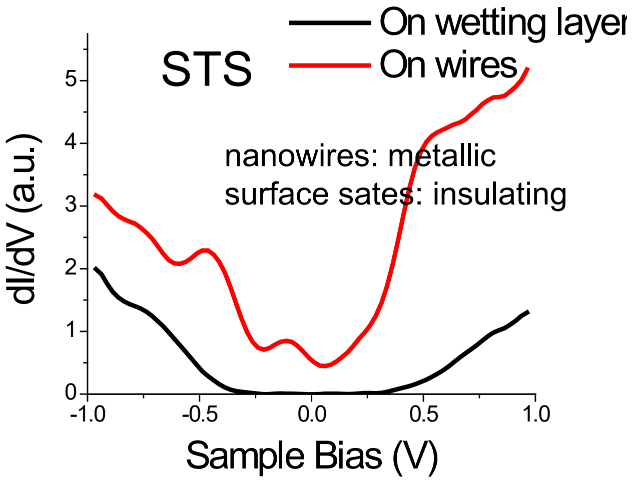
HOPG

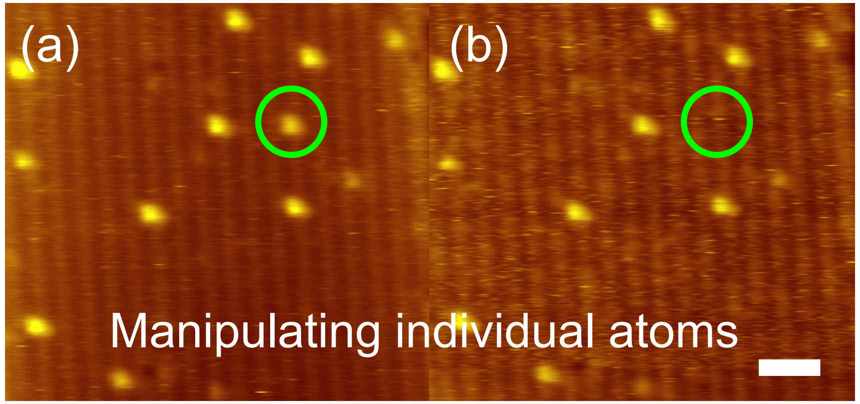


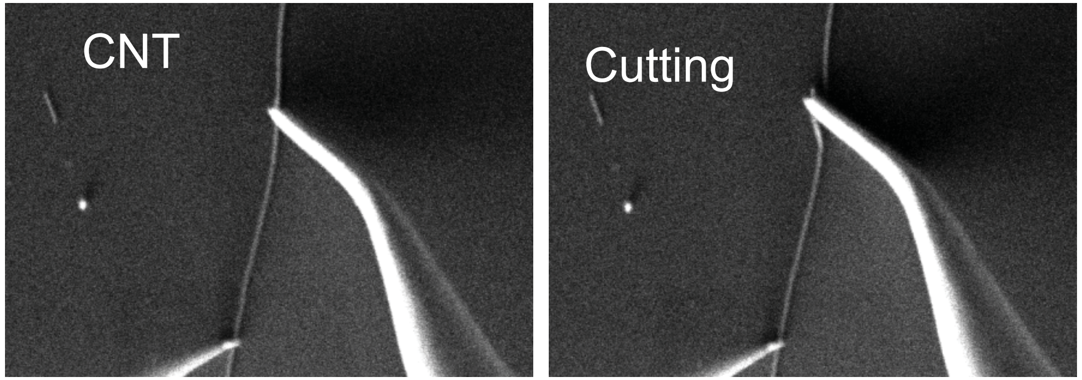
GdSi$_2$ nanowires

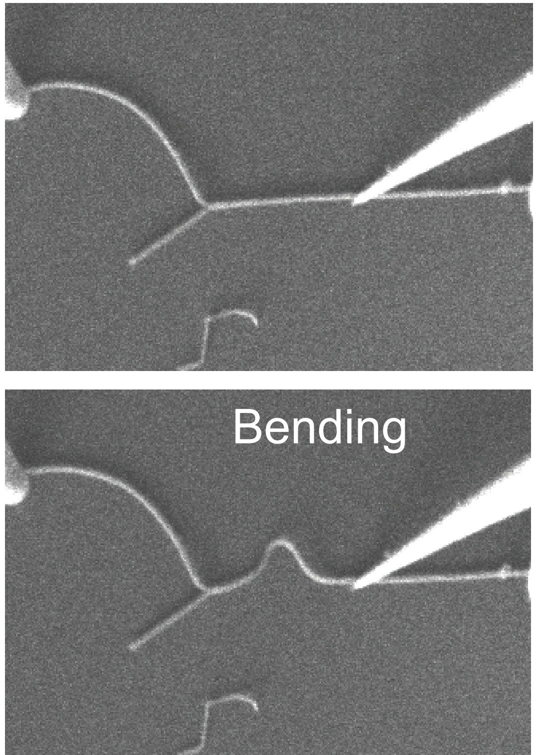


GdSi$_2$ nanowires



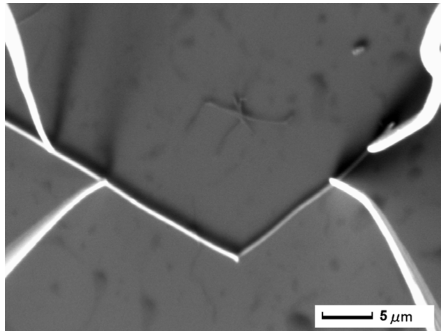


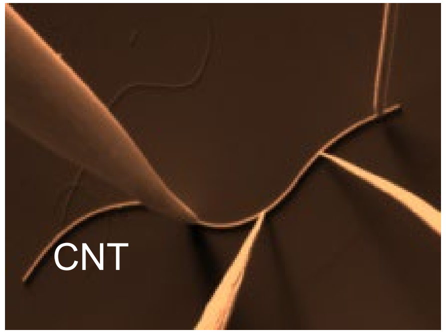


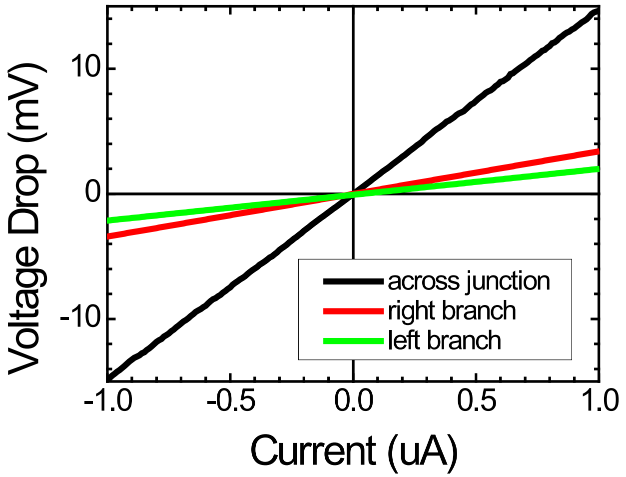


Electrical transport for nanomaterials

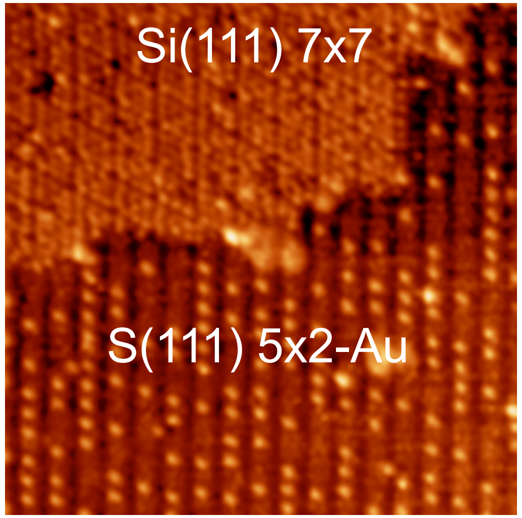
Transport in 1D

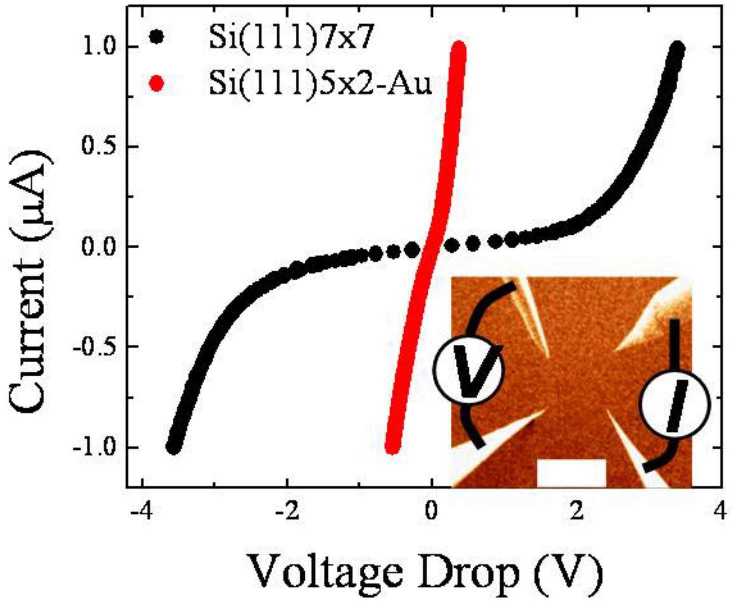






Transport in 2D





A suite of STM at CNMS of ORNL for atomic-scale imaging, spectroscopy, and manipulation

Single-probe STM

- Omicron-VT (30 – 600K) STM/AFM + MBE (metal and semiconductors) – J-G43 (8610)
- Home-built cryogenic STM + MBE (organic molecules) – J-G43 (8610)
- Home-built HB (9T) LT (2K) STM (superconductors) – J-G43 (8610)
- Omicron VT STM/AFM + PLD (oxides) – J-G47 (8610)
- Unisoku 40mK vector field (9-2-2T) STM (spin detection) – L120 (4515)
- Omicron Infinity Close-Cycle LT (10 – 420 K) STM (atom/molecule manipulations) – L120 (4515)
- SPECS Joule-Thomson STM/AFM (1.2 – 300 K) (Josephson tunneling/molecules) – 112 (3516)

Four-probe STM

- Customized RHK/Unisoku LT (10K) 4-probe STM (transport) – J-G47 (8610)
- Omicron LT (4.5K) 4-probe STM/AFM (atomic imaging, transport, manipulation) – L121 (4515)

Magneto-meter

- SQUID Magnetometer – Magnetic Property Measurement System – J-155 (8610)
- Scanning NV Magnetometer – C141(4100)

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Materials of interest

Nanostructured Materials

- Heterogeneities in atomic lattices
  - Defect structures and their multiple degrees of freedom (electronic, magnetic, ..)
  - Structure-property relationship
- Controlled positioning and manipulation of individual dopants/defects
- Confined boundary states
  - Interaction of 2D vdw layers
  - Interfacial states

Published Reviews on Topic:
Adv. Func. Mater. **29**, 1903770(2019);
Prog. Surf. Sci. **92**, 176(2017)

Quantum Materials

- Quasiparticle states in topological matter
  - Topological surface/edge states
  - Magnetic skyrmions
  - Majorana bound states
- Dissipationless edge current
  - Spin-momentum locked current
  - Quantum anomalous Hall effect
- Superconductivity
  - Competing order parameters in superconductors
  - Topological superconductivity

Adv. Mater. **35**, 2106909 (2023)

Molecular Materials

- On-surface synthesis of graphene nanoribbons with atomic precision
  - Long GNR arrays
  - In situ assessment of intrinsic electronic, magnetic, and transport properties
- Atomic precise molecular heterostructures for single photon emission
  - Large quantity of "identical" heterojunctions
  - Correlation of atomic/electronic structures with optical behaviors
- Directed molecular motions and reactions

Nat. Rev. Phys. **3**, 791 (2021)

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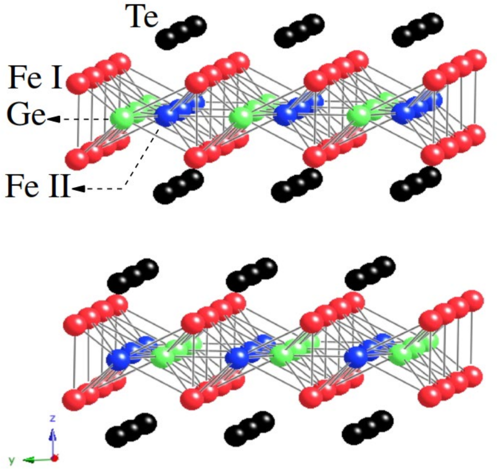
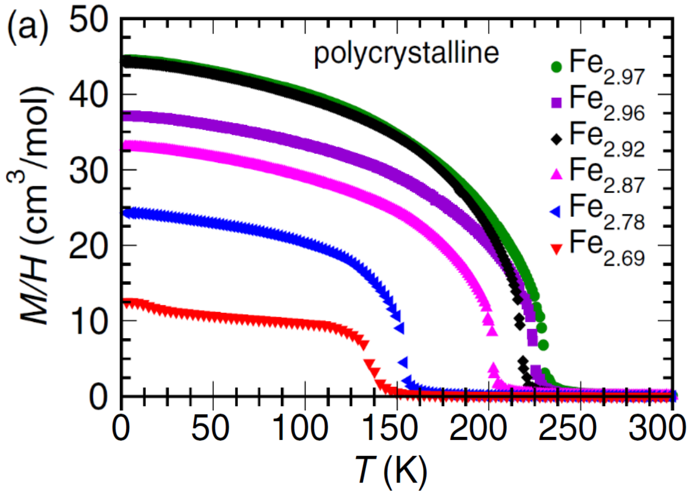
Image and manipulate magnetic skyrmion bubbles in a van der Waals ferromagnet Fe_3GeTe_2

Crystal structure



B. Chen *et al., J. Phys. Soc. Jpn* **(2013)**

Ferromagnetic FGT



High bulk magnetic transition temperature Tc>200K

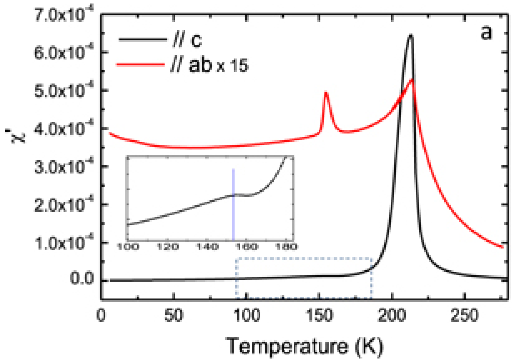
A. F. May et al, PRB **93**, 014411 (2016).

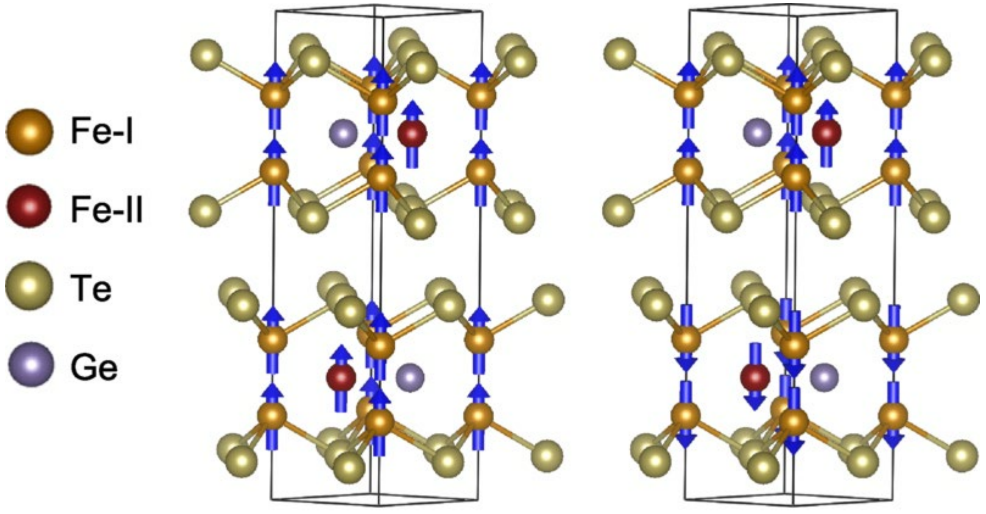
Ferromagnetic interlayer coupling:

H.-J. Deiseroth et al, Eur. J. Inorg. Chem. **2006**, 1561 (2006).
B. Chen et al, J. Phys. Soc. Jpn. **82**, 124711 (2013).
V. Y. Verchenko et al, Inorg. Chem. **54**, 8598 (2015).
N. León-Brito et al, J. Appl. Phys. **120**, 083903 (2016).
A. F. May et al, Phys. Rev. B **93**, 014411 (2016).
J.-X. Zhu et al, Phys. Rev. B **93**, 144404 (2016).
H. L. Zhuang et al, Phys. Rev. B **93**, 134407 (2016).
Y. Liu *et al.*, Phys. Rev. B **96**, 144429 (2017).

Antiferromagnetic interlayer coupling:

J. Yi et al, 2D Mater. **4**, 011005 (2016).





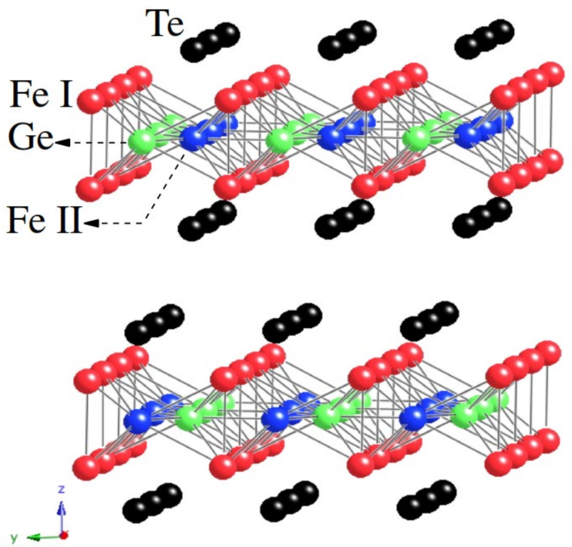
(a) Interlayer FM (b) Interlayer AFM

- **What is the nature of interlayer coupling?**

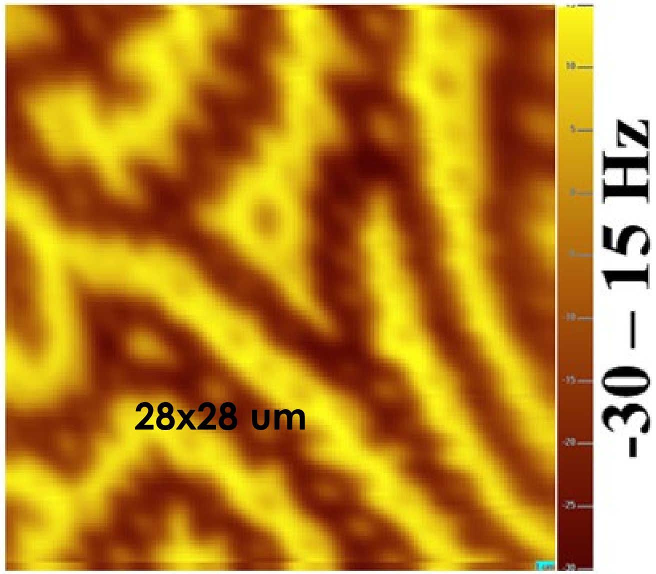
Magnetic domains in a van der Waals ferromagnet Fe_3GeTe_2

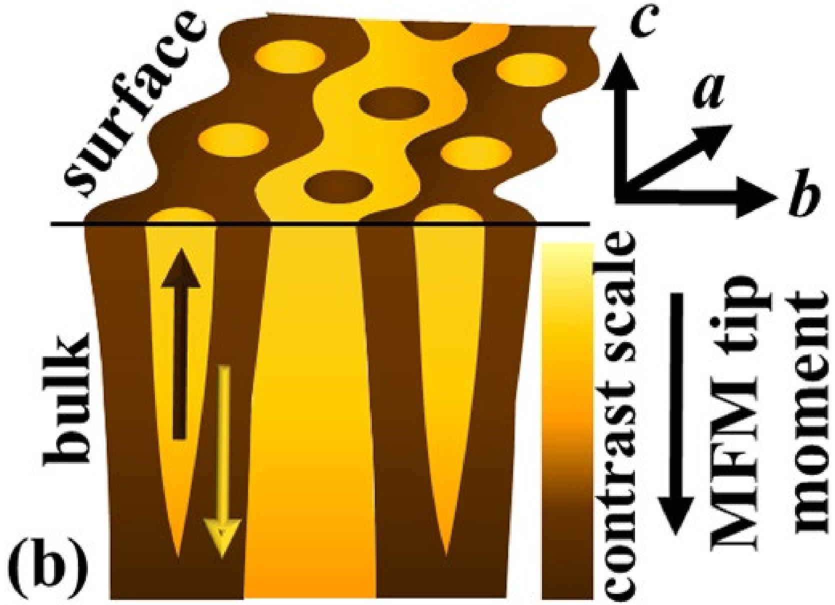
Magnetic ordering temperature (140K<Tc<230K) and magnetic anisotropy are reduced with increasing concentration of Fe-II vacancies

A. F. May et al, PRB **93**, 014411 (2016).



MFM imaging

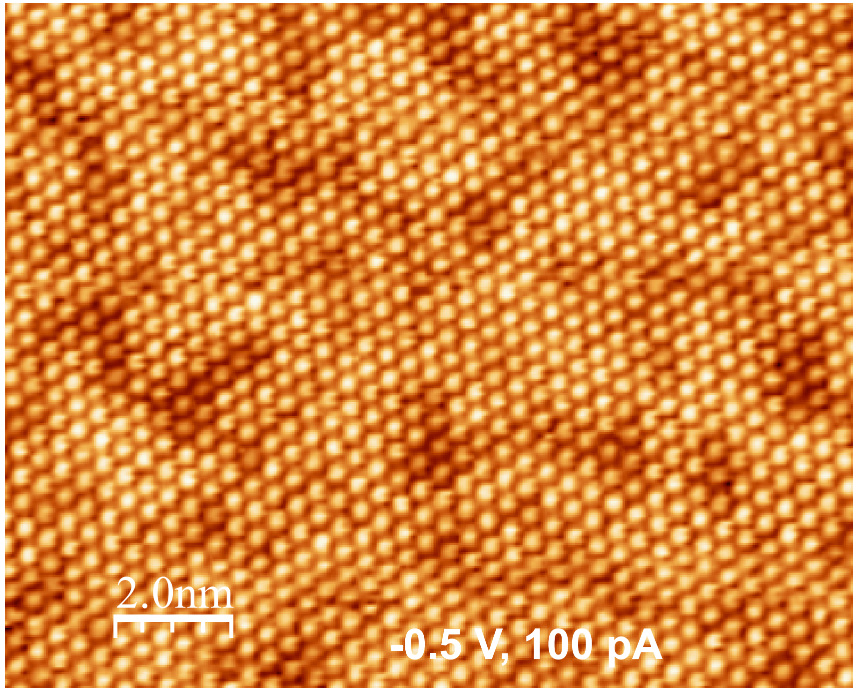


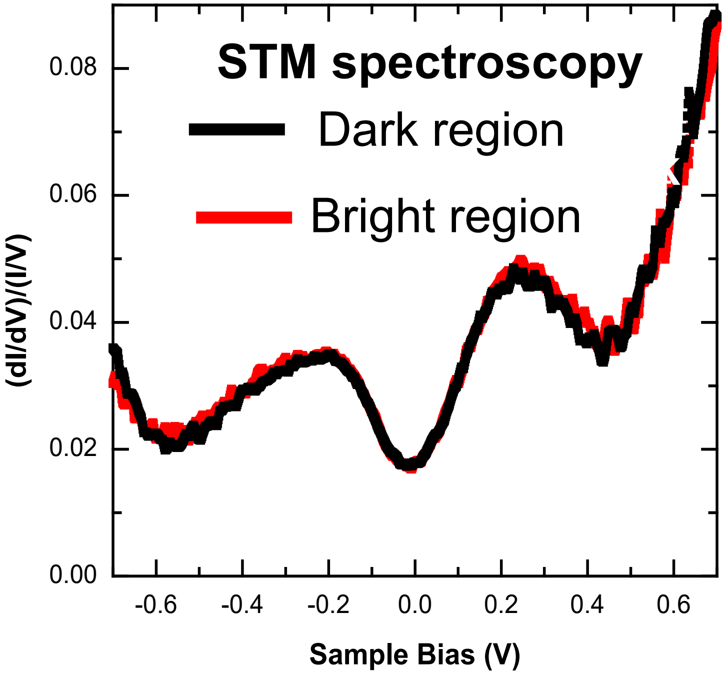


N. Leon-Brito *et al., J. Appl. Phys.* 120, 083903(2016)

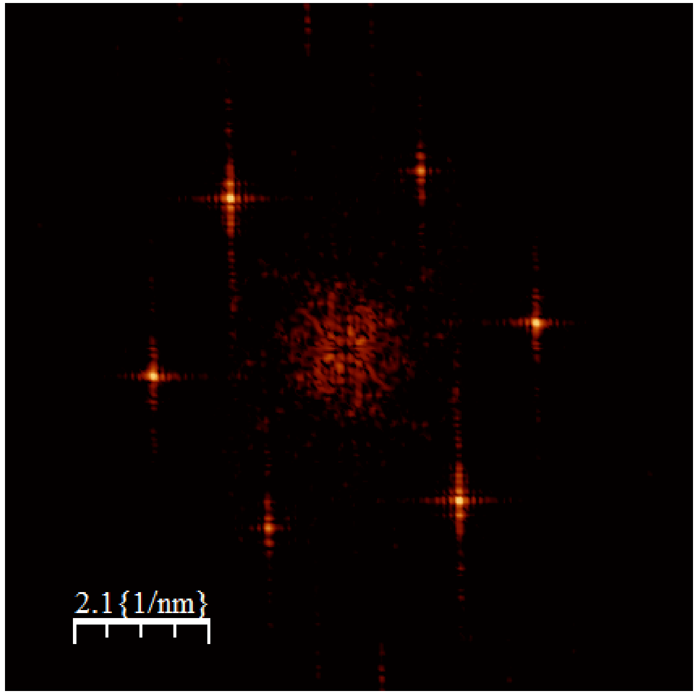
- **What is the nature of magnetic domains?**
- **What role does the Fe-II vacancy play?**

STM image of Fe_3GeTe_2





FFT



Atomic domain: depressed atoms
Depressed height: 13 to 50 pm
Area density: 23 ± 4%

Atomic scale domains without long range order

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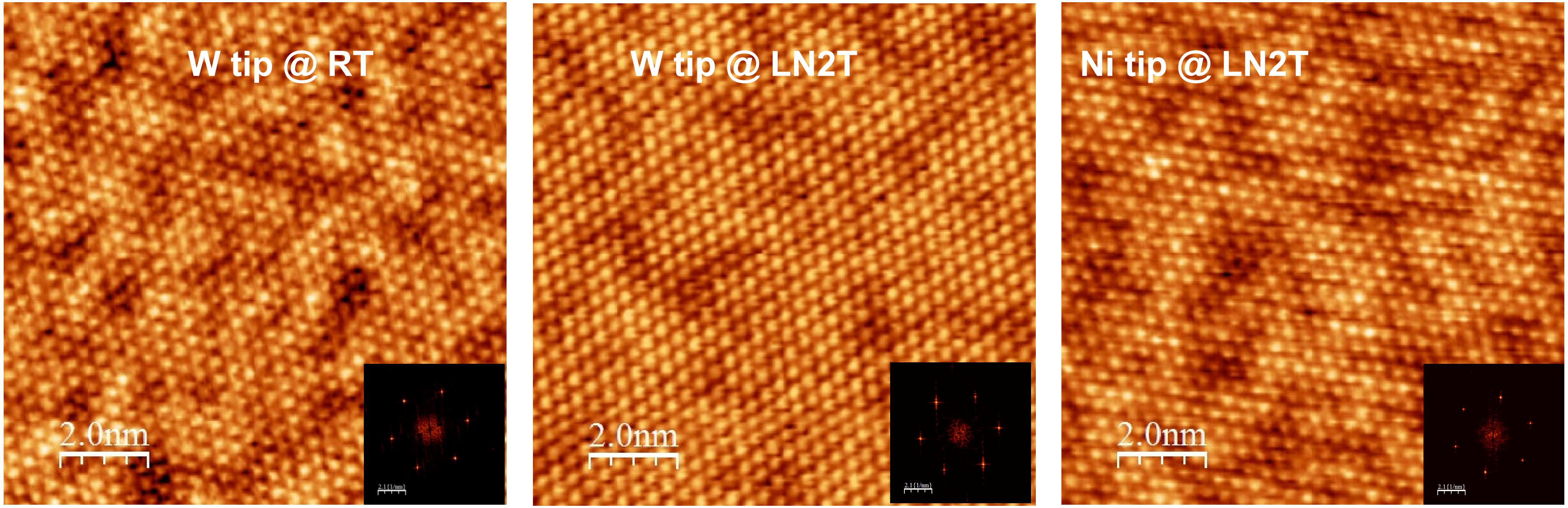
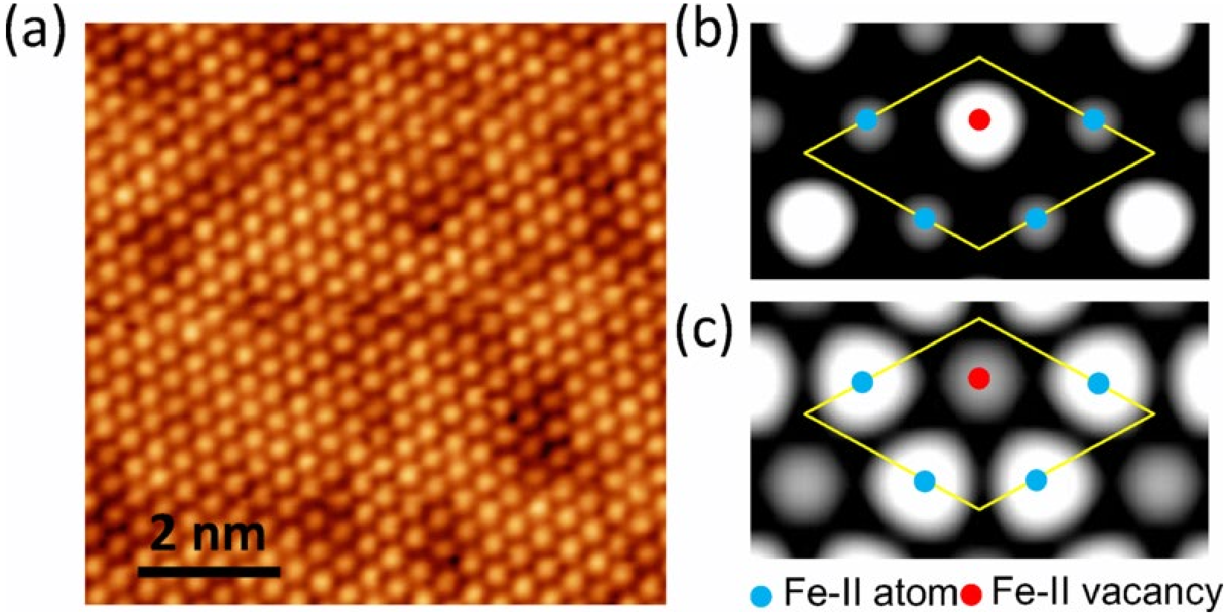
SP-STM at the atomic scale



No magnetic contrast at the atomic scale

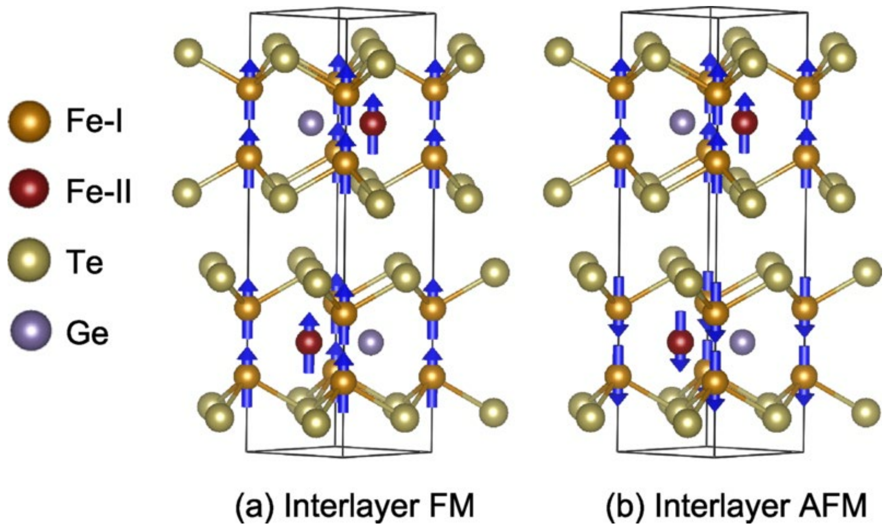
Atomic domains are not the magnetic domains observed at a larger length scale

Interlayer coupling of Fe3GeTe2

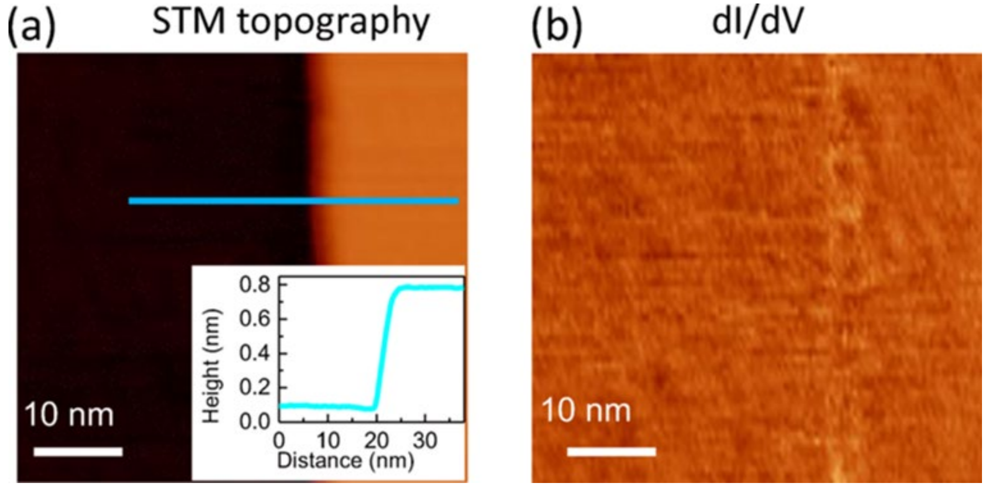


X. Kong et al, Phys. Rev. Materials **4**, 094403 (2020)

Atomic domains come from Fe-II vacancies which show inhomogeneous distributions



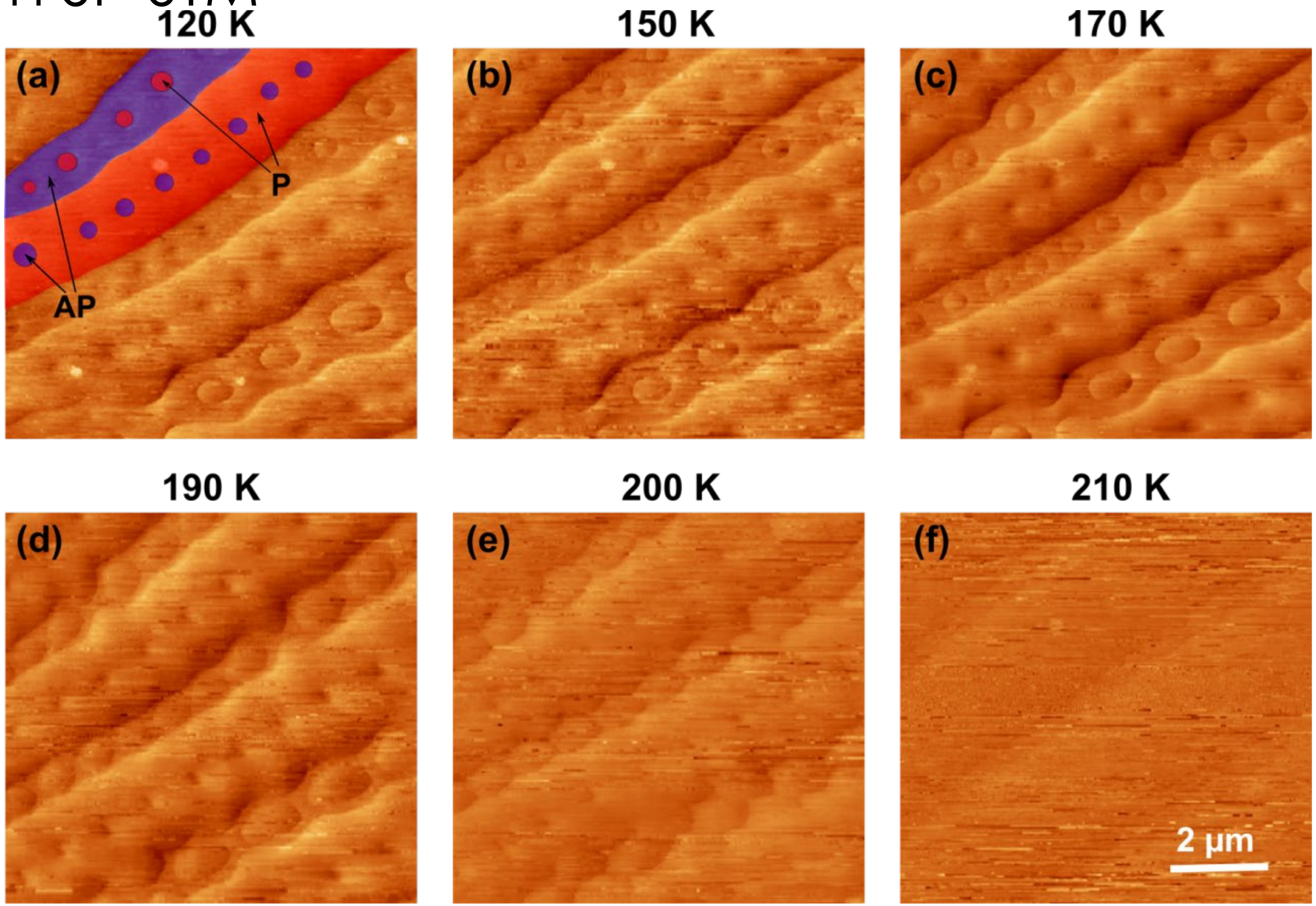
Te atoms move down by 13 pm

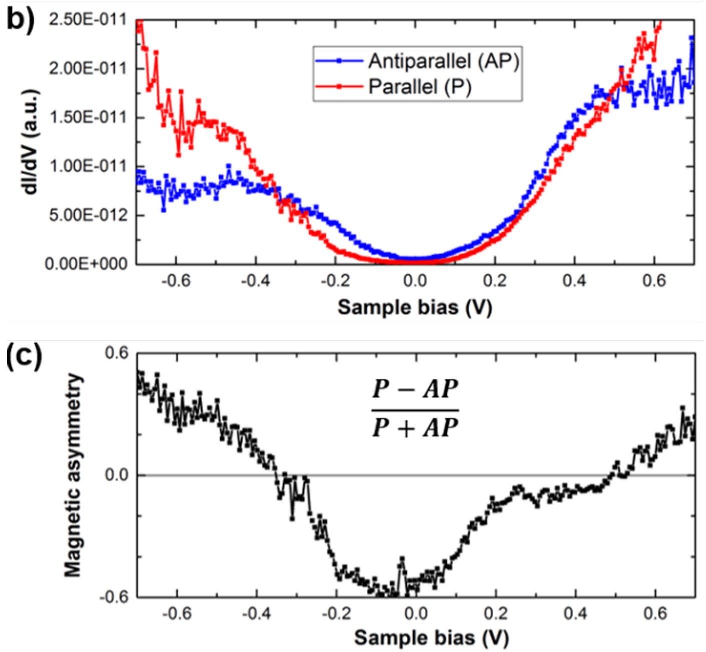


SP-STM

Layers of FGT are coupled ferromagnetically

Temperature dependence of ZFC magnetic domains revealed with SP-STM





✓ **Transition temperature: 200-210 K**

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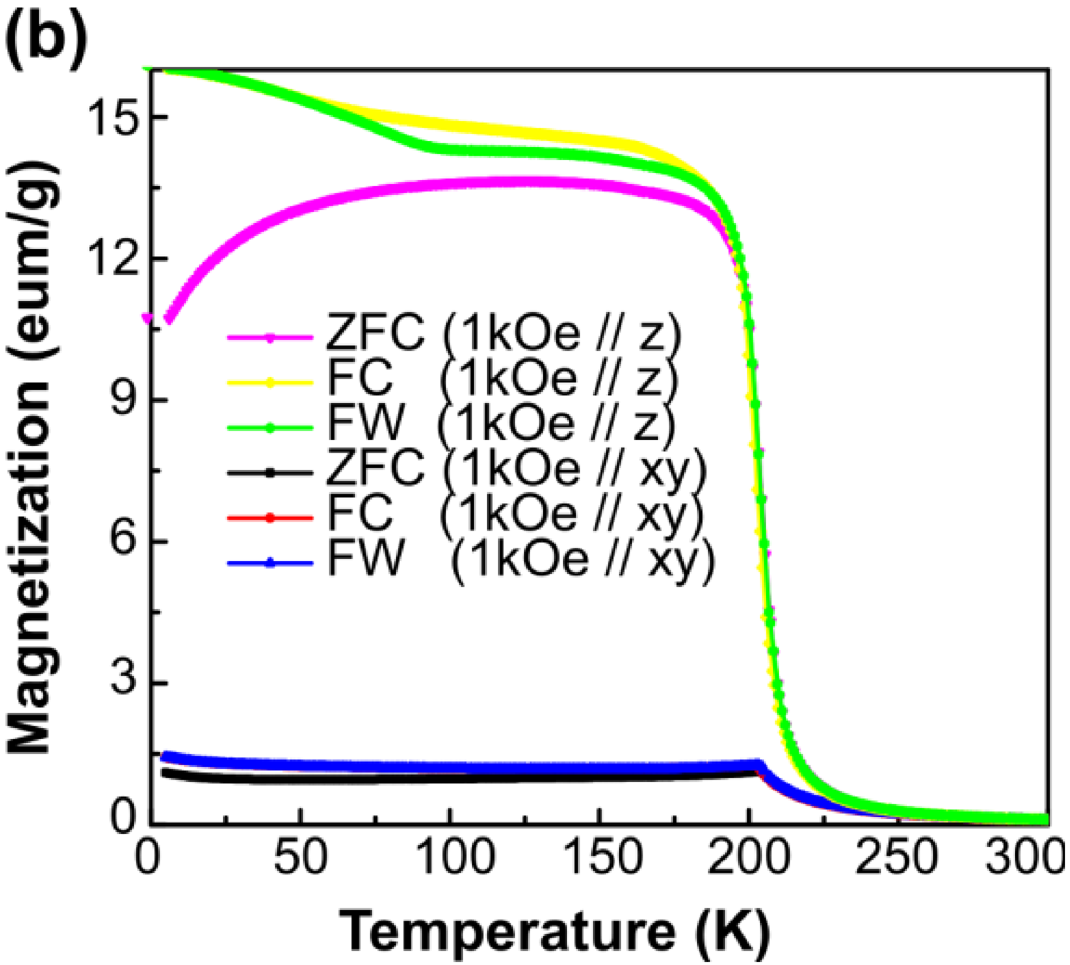
SQUID measurement



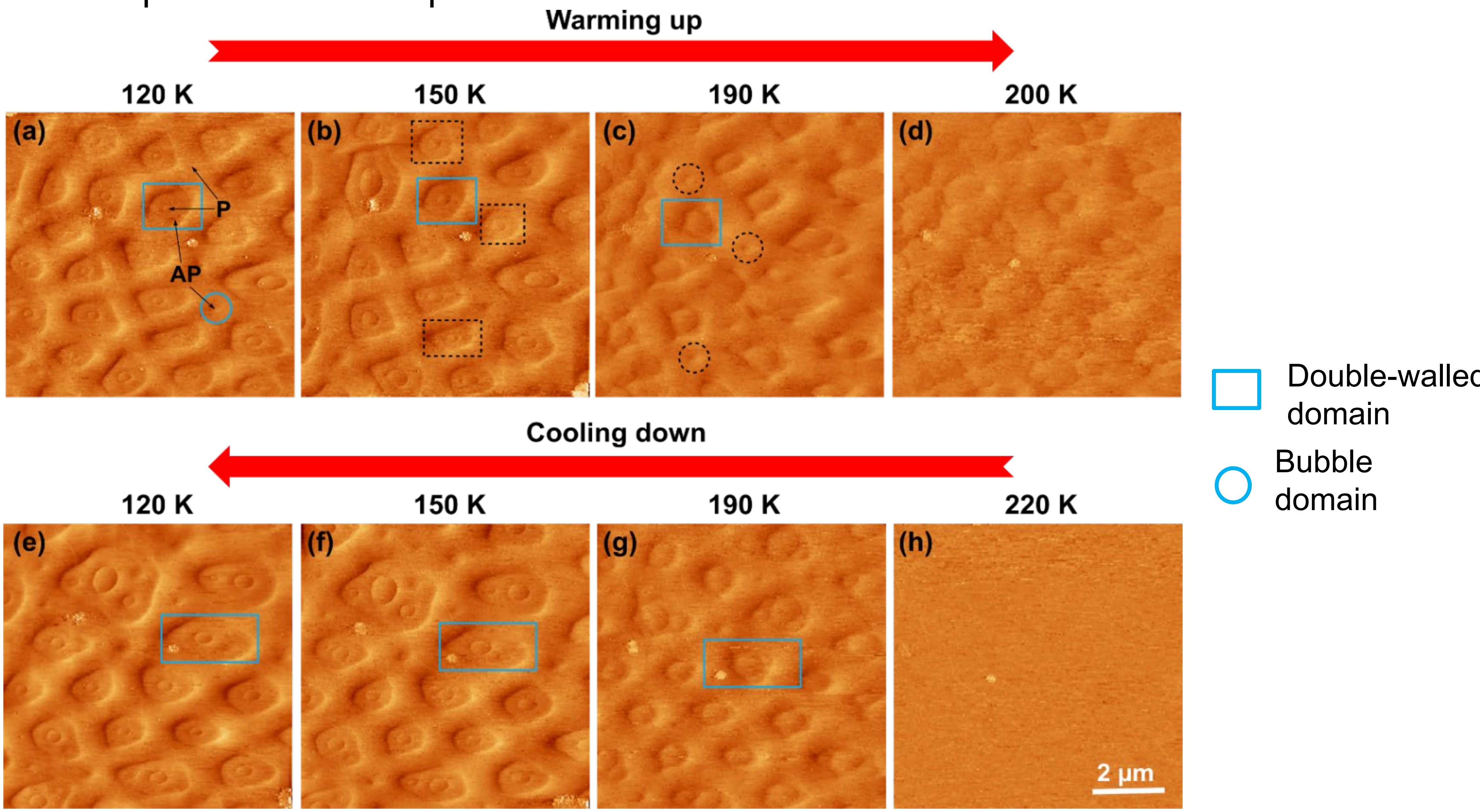
ZFC: Zero field cooled

FC: Field cooled

FW: Field Warmed

Transition temperature: 205 K

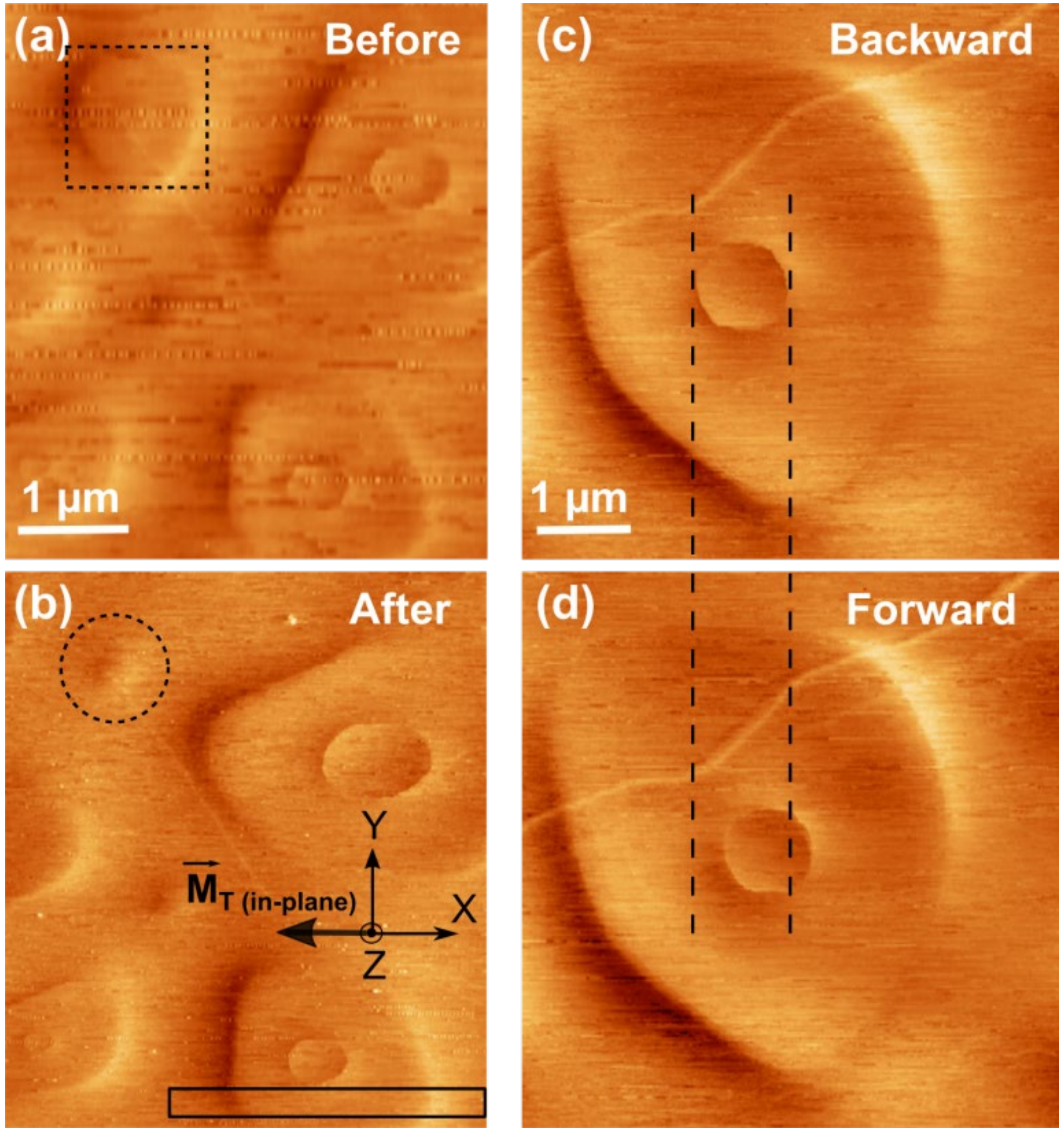
Temperature dependence of FC domains



Tip manipulation of FC domain

Double walled domain

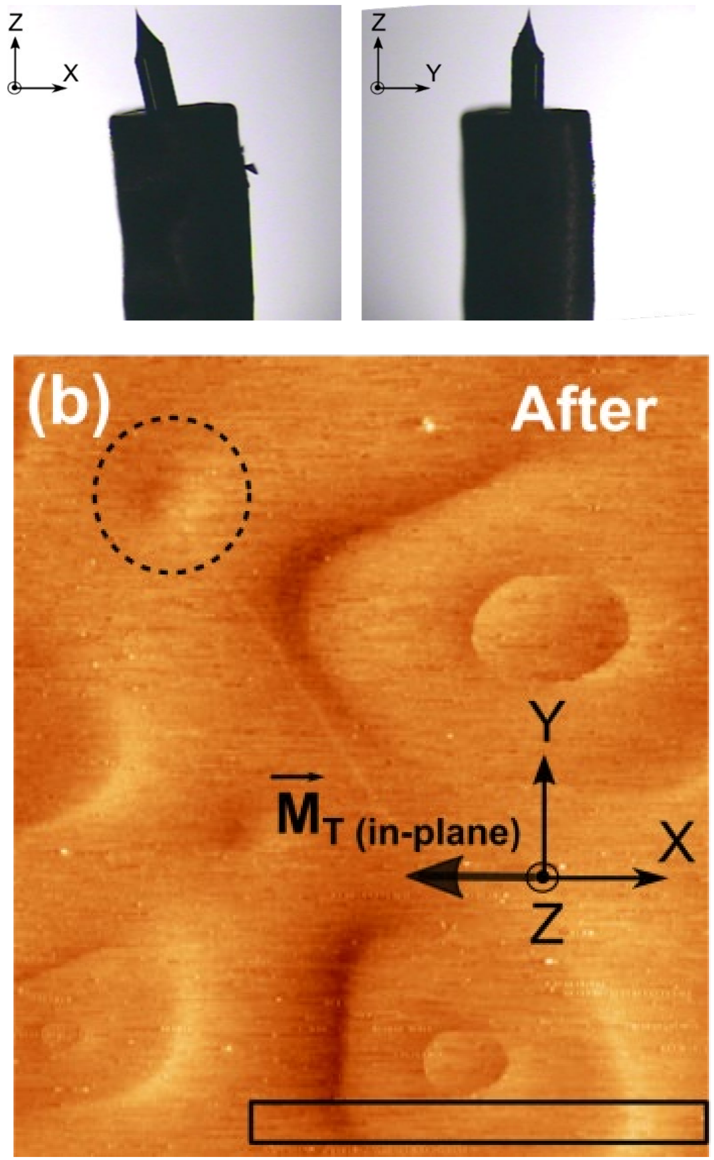
Bubble domain

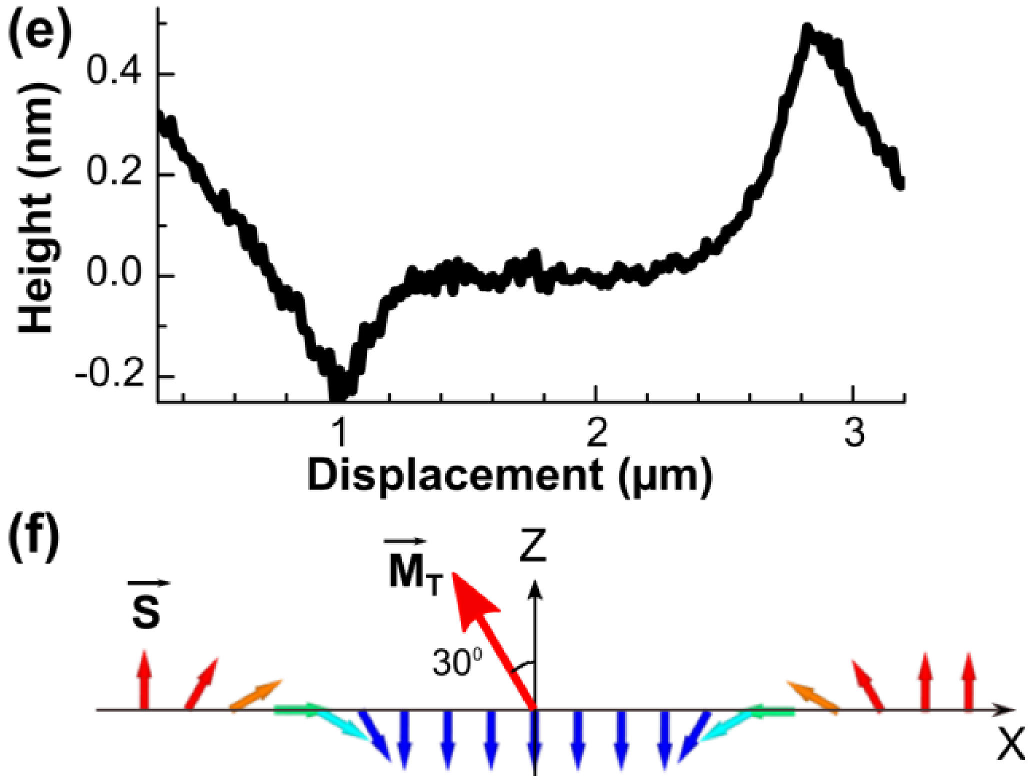


Domain displacement

Neel type domain wall structure

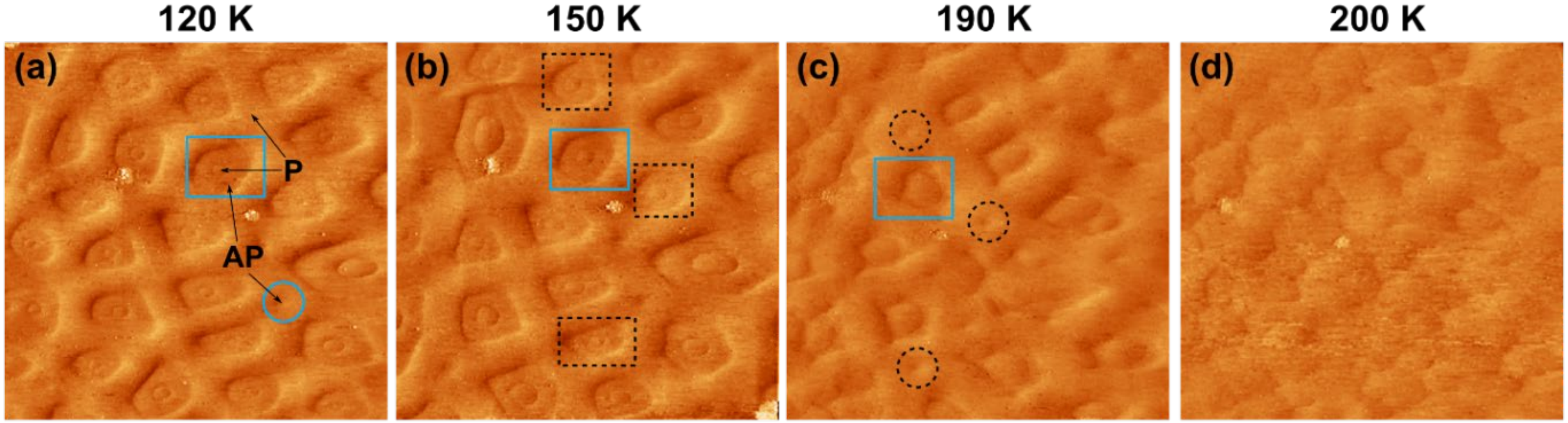
Images of the SP-STM tip





360∘-Neel wall

Neel-type skyrmion observed in FGT



- Spin texture with 360∘ rotation across the domain.
- Emergence of the circular domain structure from stripy domain shows that a magnetic field promotes the formation of skyrmion phase from a helical phase.
- Size of the skyrmion bubble (100 nm to 1μm) due to a competition of long-range dipolar interactions with shape anisotropy.

G.D. Nguyen et al, Physical Review B 97, 014425 (2018)

Mechanisms of skyrmion formation

- Dzyaloshinskii–Moriya exchange interaction (DMI) in magnets without inversion center and a weak magnetic anisotropy
  - Observed skyrmion structure in FGT, inconsistent with its presumed centrosymmetric structure

- Interfacial DMI could account for the observation of chiral Néel skyrmions in an otherwise centrosymmetric compound
  - Observed skyrmion structure in FGT, inconsistent with its presumed bulk structure

- Fe vacancies break the inversion symmetry?

- “further investigation is required to confirm if the magnetic domain structures observed here have any topologically protected nature of skyrmions”.

G.D. Nguyen et al, Physical Review B 97, 014425 (2018)

Lorentz transmission electron microscopy (LTEM)

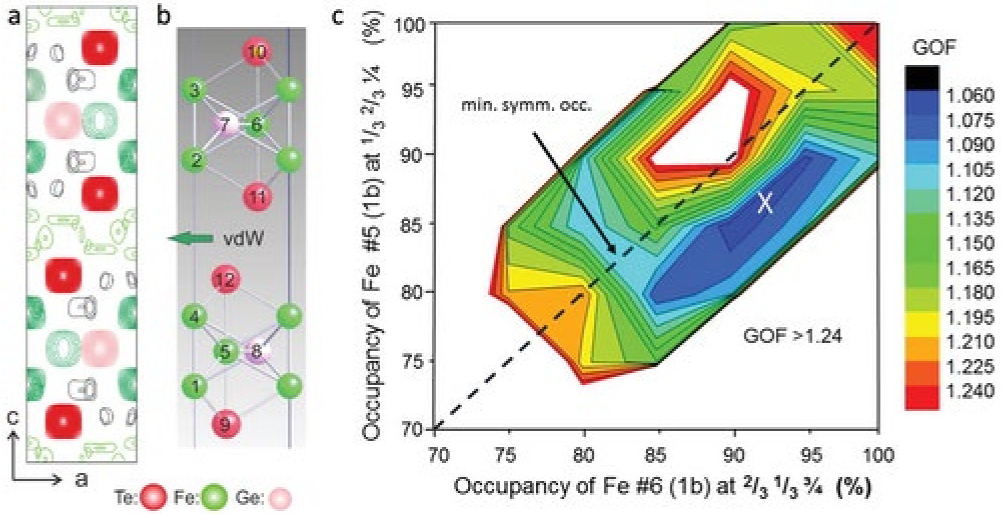
ADVANCED MATERIALS

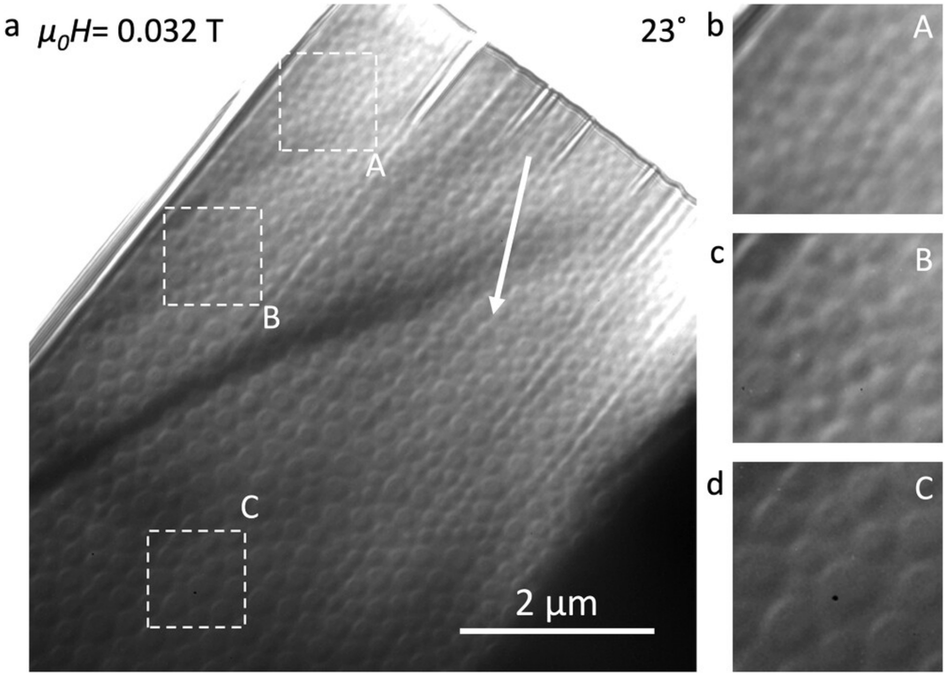
Research Article | Open Access

Magnetic Skyrmions in a Thickness Tunable 2D Ferromagnet from a Defect Driven Dzyaloshinskii–Moriya Interaction

Anirban Chakraborty, Abhay K. Srivastava, Ankit K. Sharma, Ajesh K. Gopi, Katayoon Mohseni, Arthur Ernst, Hakan Deniz, Binoy Krishna Hazra, Souvik Das, Paolo Sessi, Ilya Kostanovskiy, Tianping Ma, Holger L. Meyerheim, Stuart S. P. Parkin ... See fewer authors

First published: 19 January 2022 | https://doi.org/10.1002/adma.202108637 | Citations: 17





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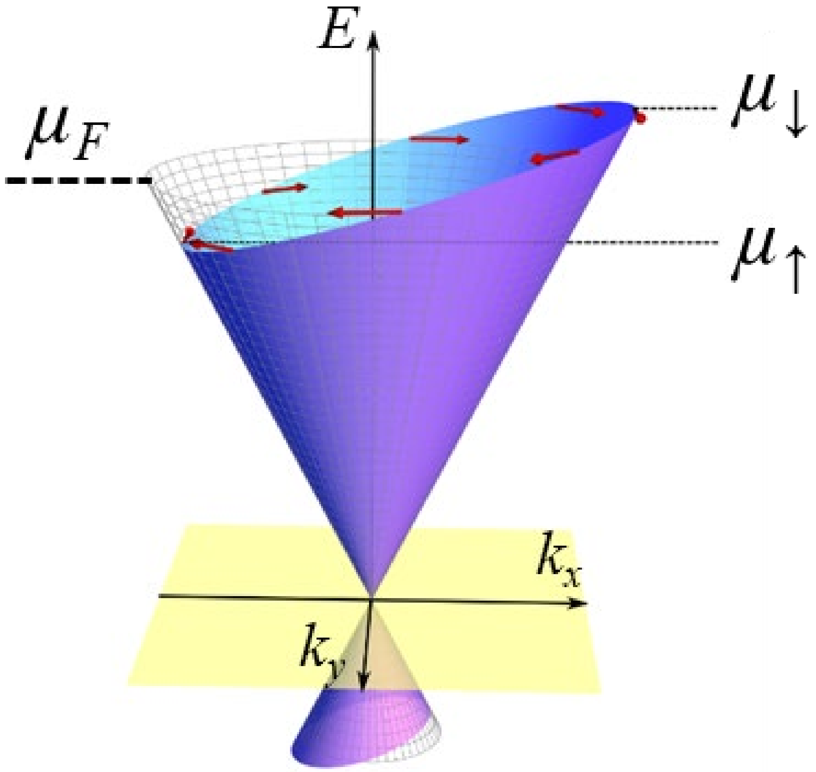
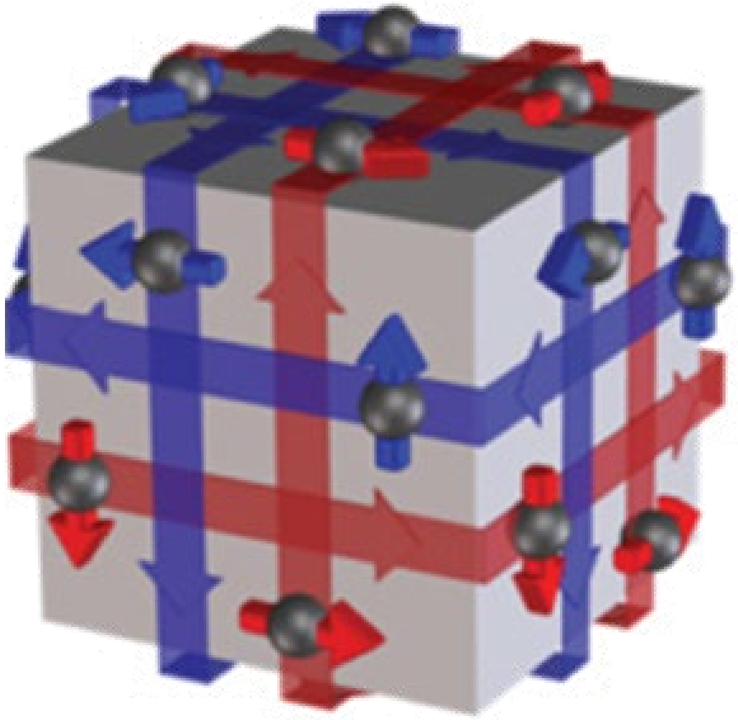
Spin–momentum- locked conduction in topological insulators

- Spin-dependent transport carried by topological surface states.

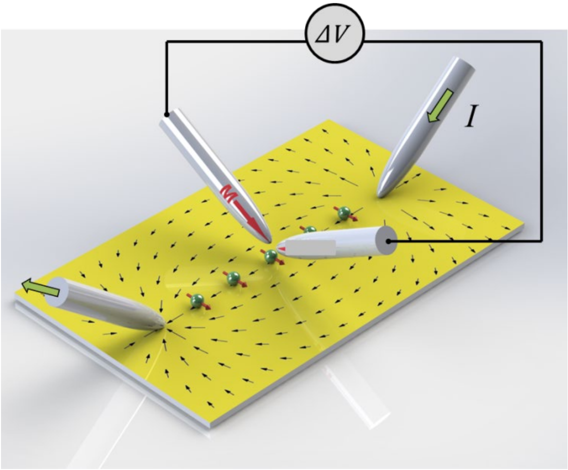




Spin-dependent transport in topological insulators

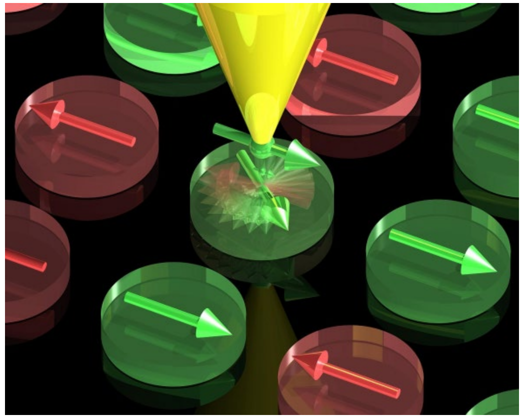
- 4-probe STM transport measurement

Four-Probe STM



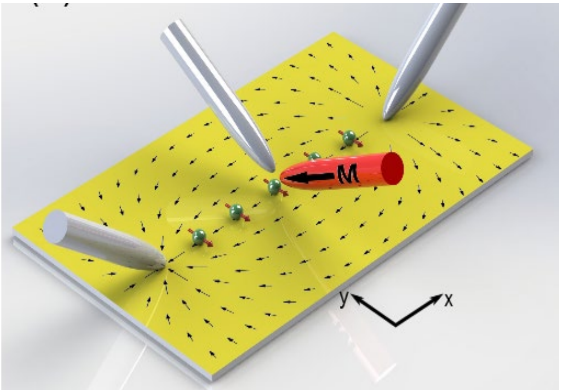
T. H. Kim et al, RSI 78, 123701 (2007)

Spin-Polarized STM



Stefan Krause, Universität Hamburg

Spin-Polarized Four-Probe STM



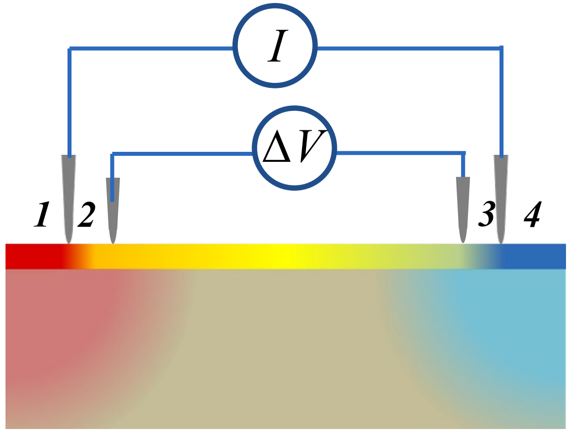
S. M. Hus *et al.*, *PRL* **119** 137202 (2017)

Advantage:

- UHV in situ cleaved surface
- Clean and minimal stray field from electrical contacts
- Distinguish and quantify the 2D and 3D conductance
- Differentiate the spin polarized and spin averaged currents

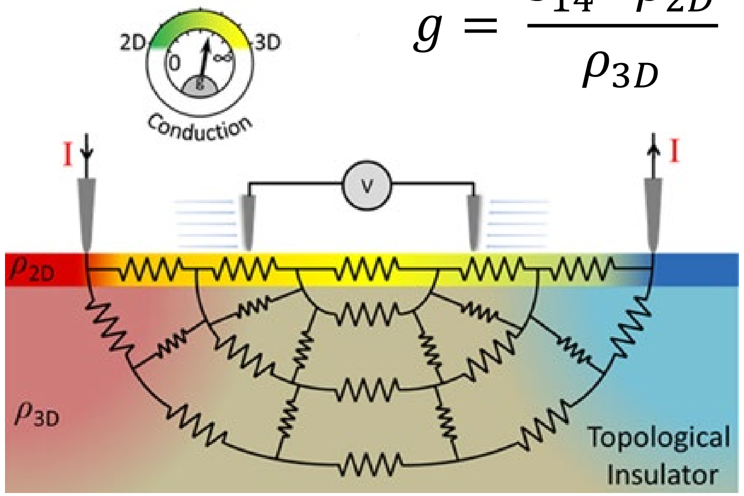
Four-probe STM studies on topological insulators

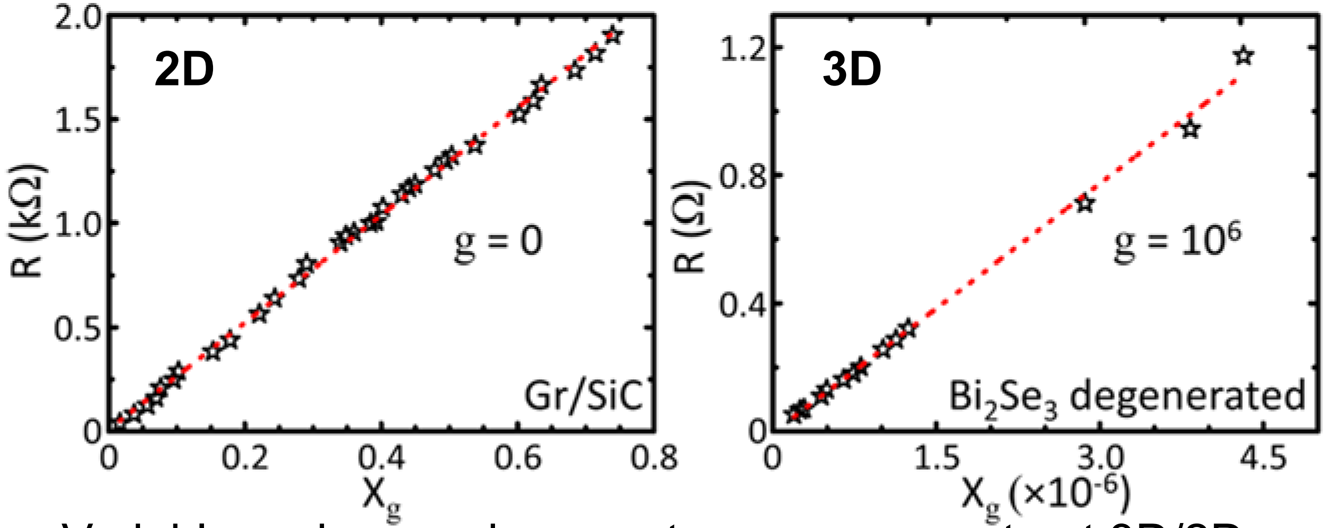
- Differentiating bulk and surface conduction



$$R = \frac{\Delta V}{I} = \rho_{2D} \cdot \frac{1}{2\pi} \ln \left[\frac{\left(g + \frac{s_{14}}{s_{12}}\right)\left(g + \frac{s_{14}}{s_{34}}\right)}{\left(g + \frac{s_{14}}{s_{13}}\right)\left(g + \frac{s_{14}}{s_{24}}\right)} \right] = \rho_{2D} \cdot X_g$$

$$g = \frac{s_{14} \cdot \rho_{2D}}{\rho_{3D}}$$



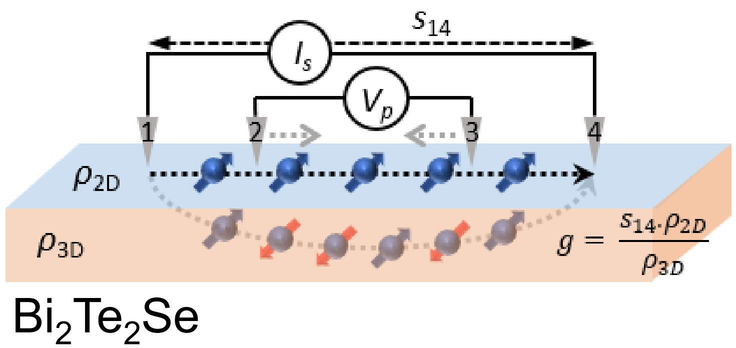


Variable probe-spacing spectroscopy can extract 3D/2D conductivity ratio g

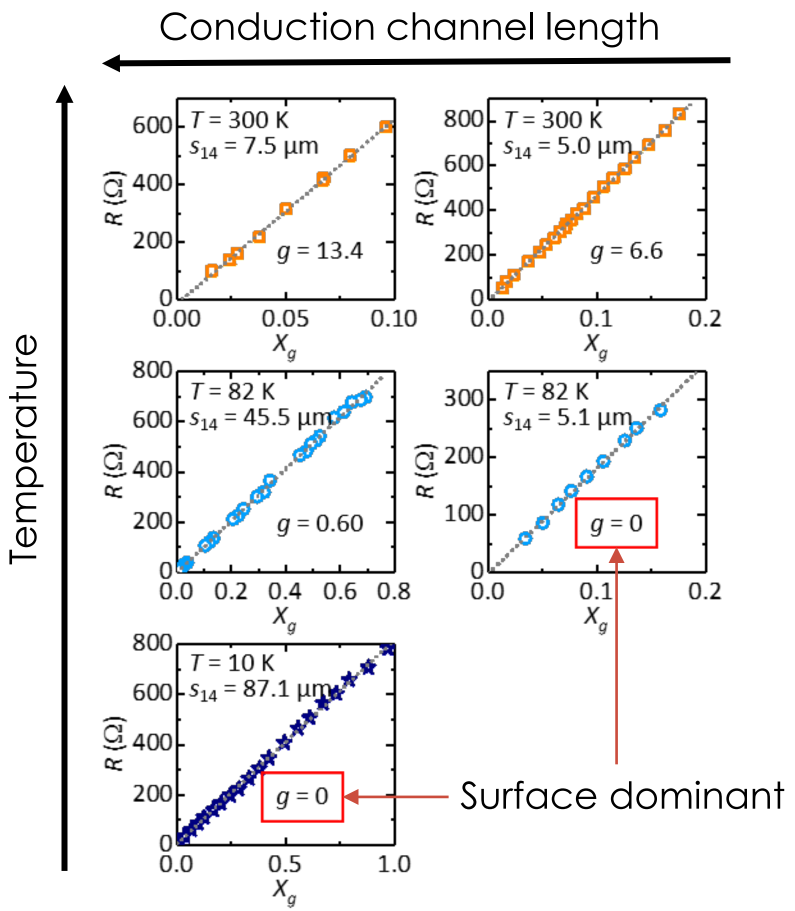
C. Durand *et al.*, *Nano Lett.* **16** 2213 (2016)

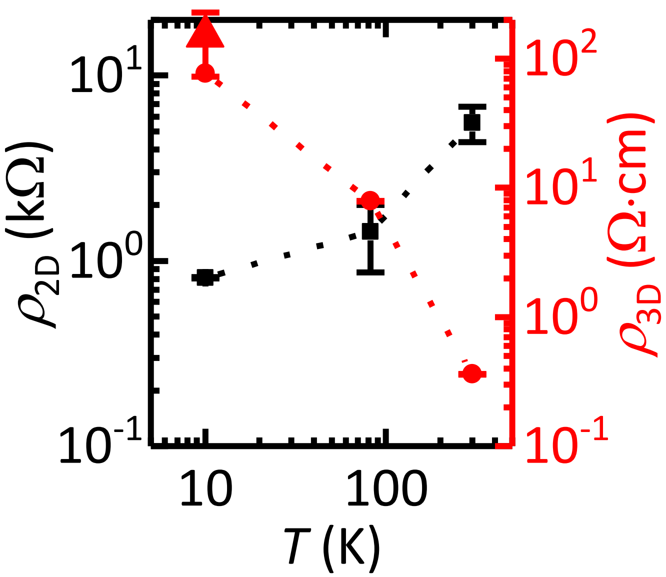
Multiprobe STM studies on topological insulators

- Crossover of bulk-surface conduction in Bi_2Te_2Se



$$g(T, s_{14}) = \frac{s_{14} \cdot \rho_{2D}(T)}{\rho_{3D}(T)}$$





W. Ko *et al.*, *PRL* 121 176801 (2018)

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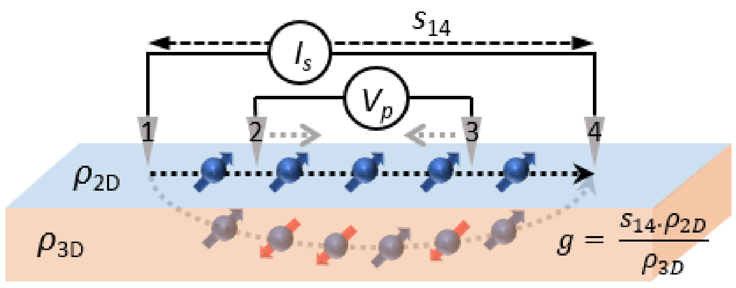
Multiprobe STM studies on topological insulators

- High surface mobility from topological protection

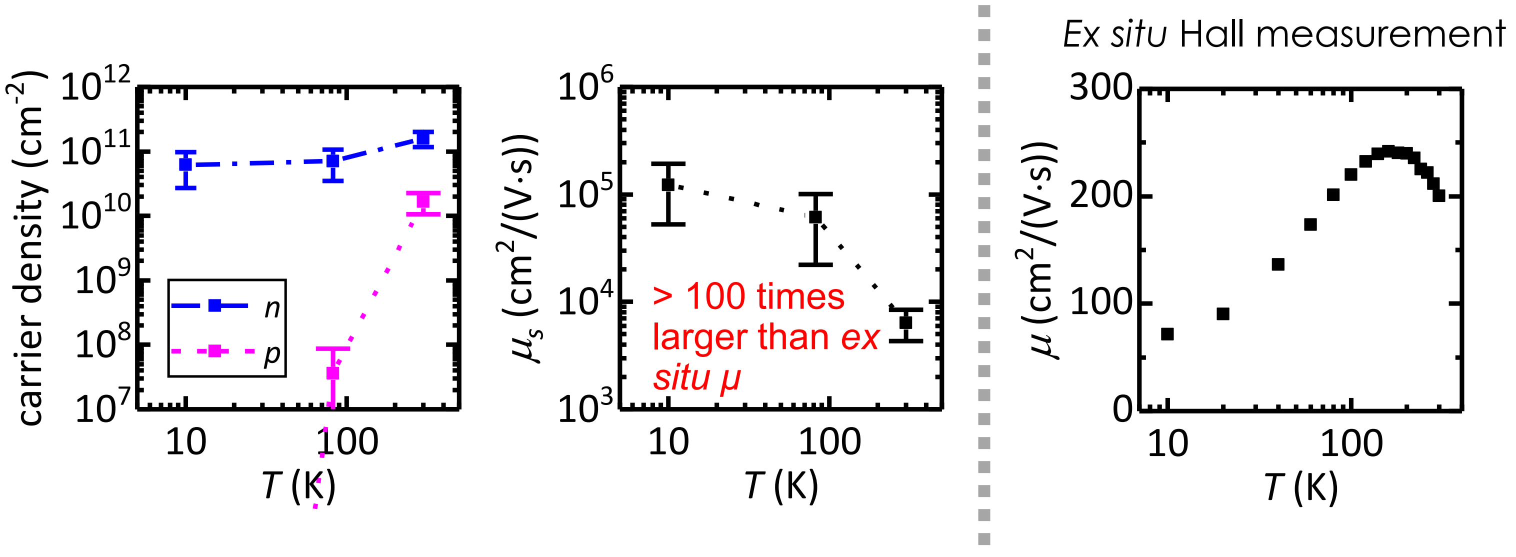


Bi_2Te_2Se

$$g(T, s_{14}) = \frac{s_{14} \cdot \rho_{2D}(T)}{\rho_{3D}(T)}$$

$$H = \hbar v_F \boldsymbol{\sigma} \cdot \boldsymbol{k} + E_D$$

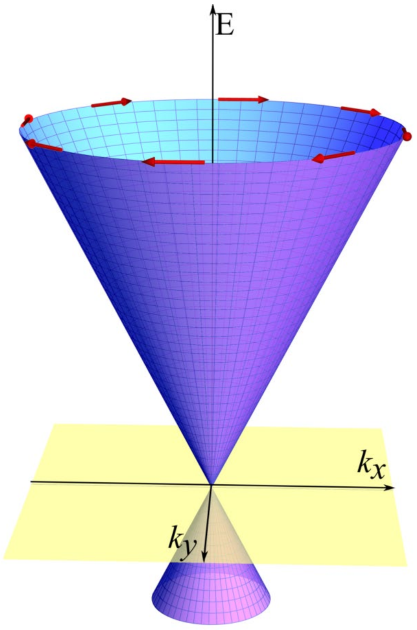
(E_D = -35 ± 10 meV, v_F = (6 ± 1.5)×10^5 m/s)

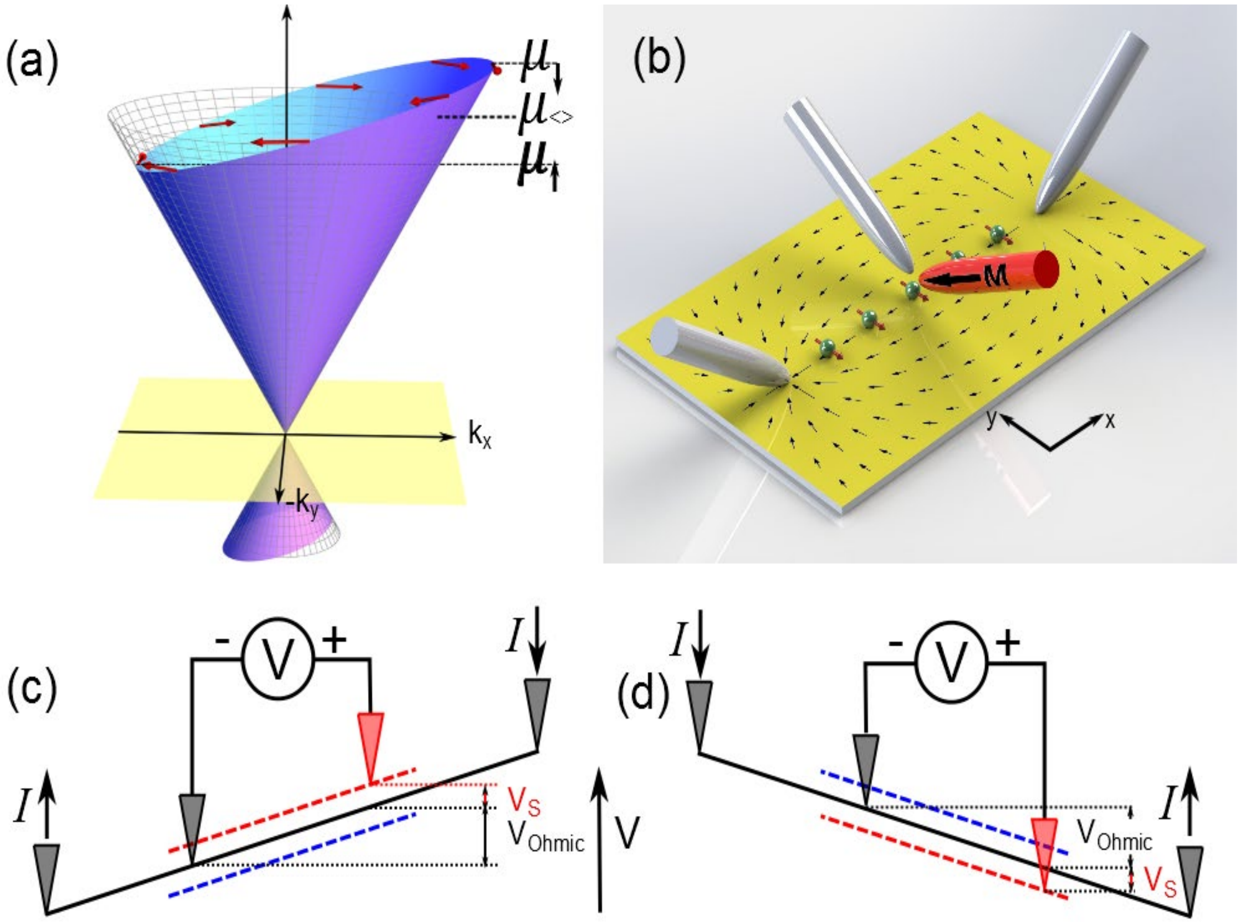


W. Ko *et al.*, *PRL* **121** 176801 (2018)

Multiprobe STM Studies on Topological Insulators

- Spin-dependent transport measurements

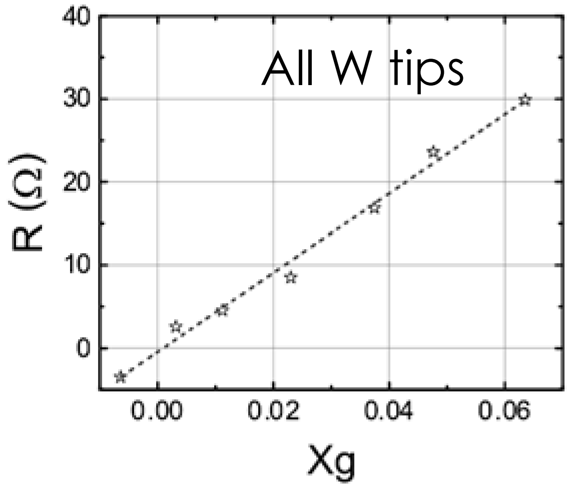


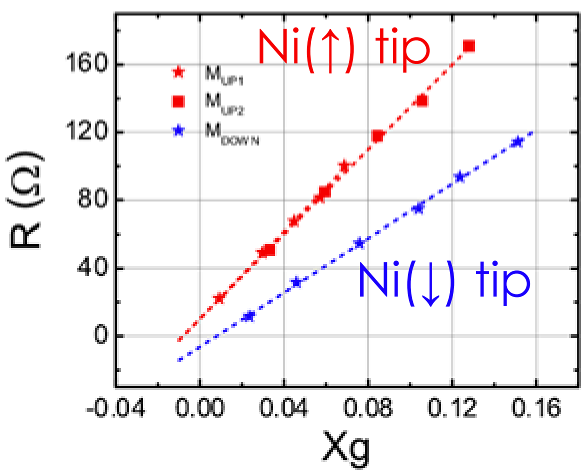


Hus, et al., PRL 119, 137202 (2017)

Multiprobe STM Studies on Topological Insulators

- Spin-dependent transport measurements on Bi_2Te_2Se





S. M. Hus *et al.*, *PRL* **119** 137202 (2017)

Offset in *R* at $X_g = 0$ from spin potential

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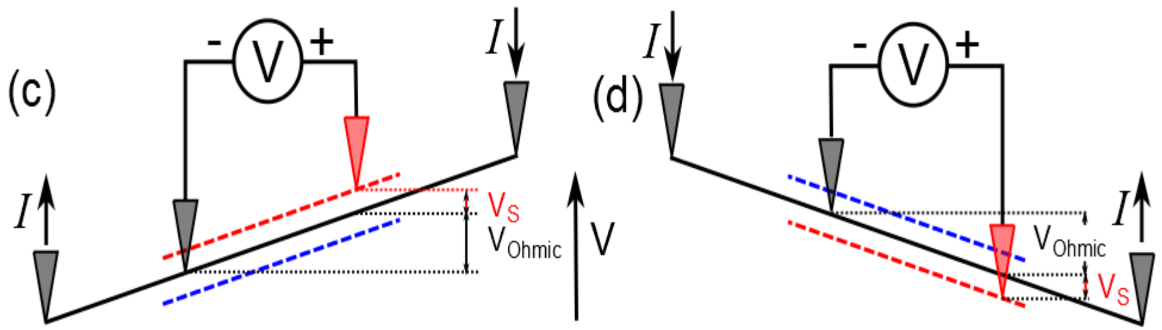
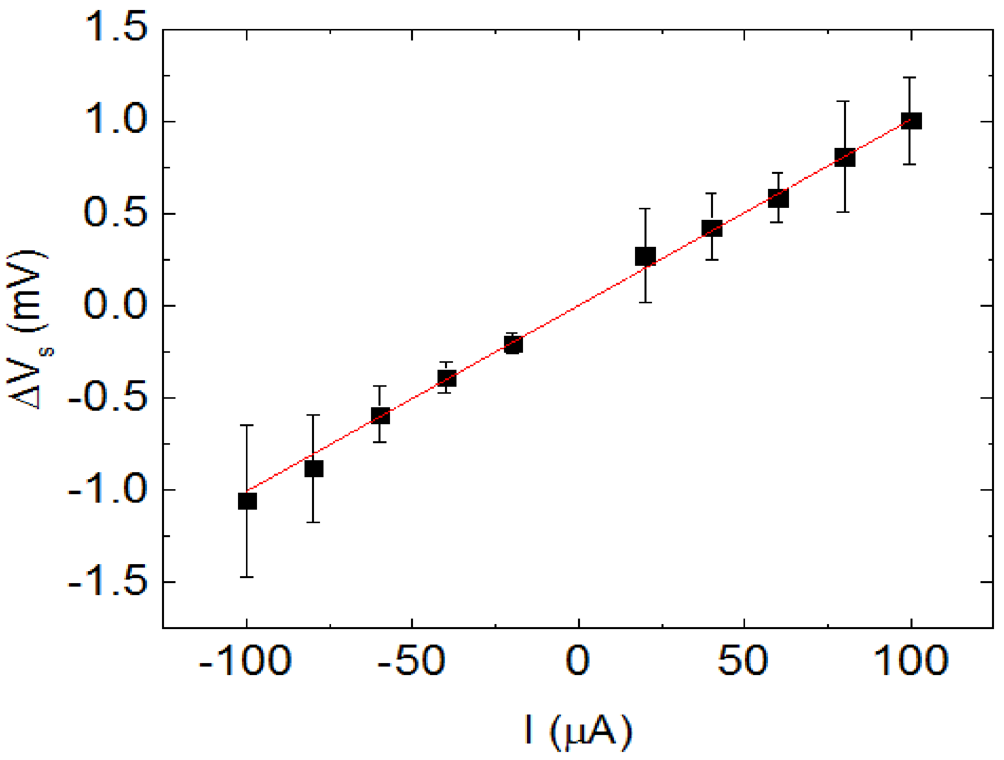
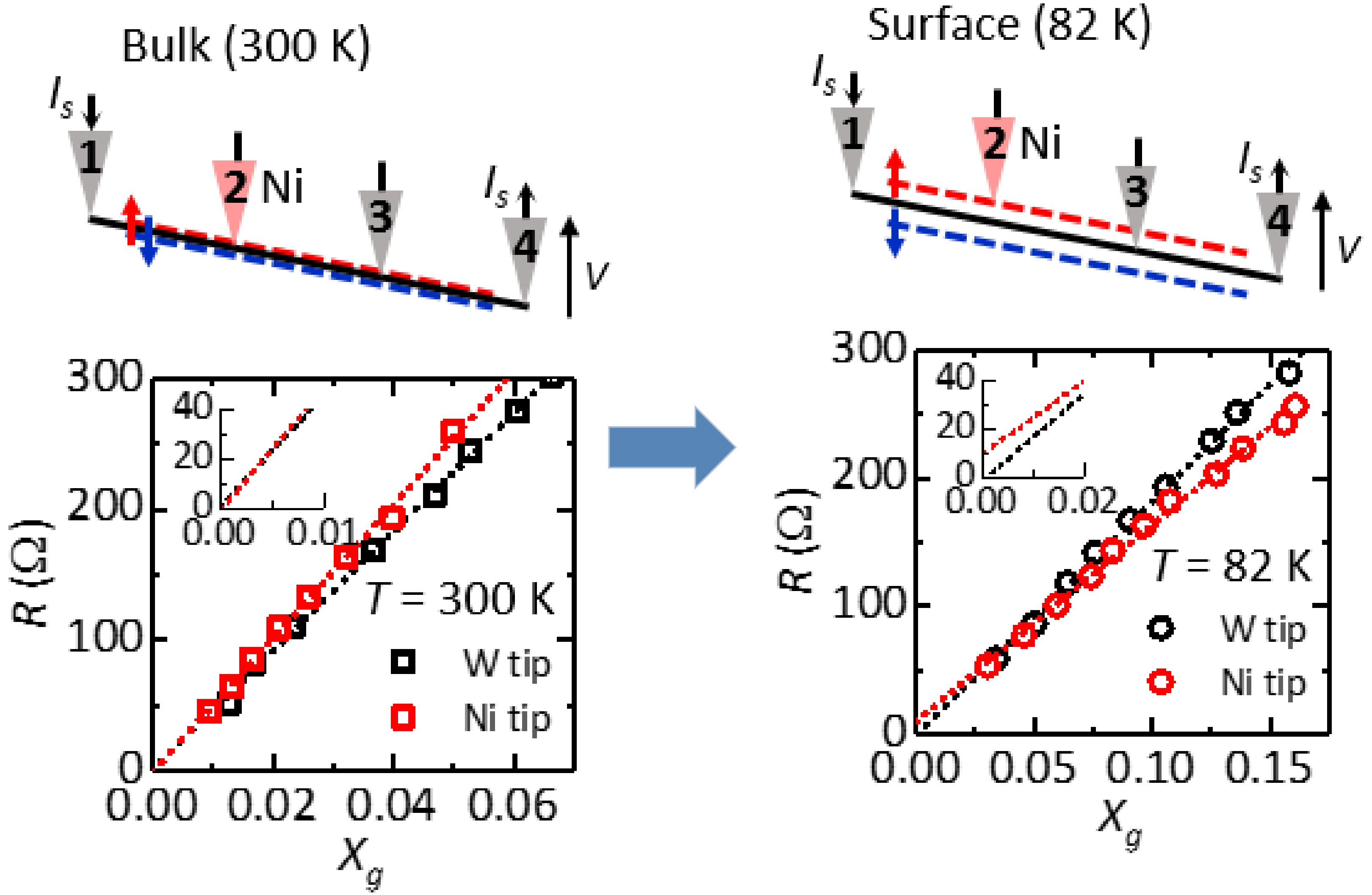
Spin-momentum-locking

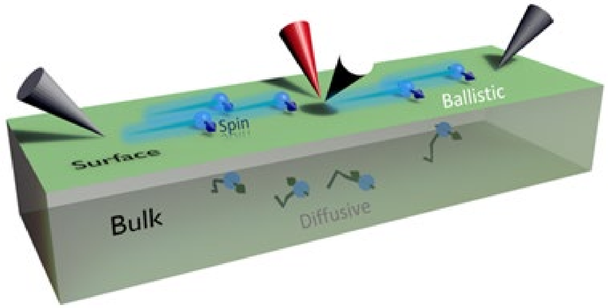
- Spin-dependent transport measurements on Bi_2Te_2Se





Spin-momentum-locking





Spin polarization $P = 72\ \%$ comparable to the theoretical limit for topological surface states

Mean-free-path $\ell = 1.4\ \mu m$

W. Ko *et al.*, *PRL* 121 176801 (2018)

Summary

- Scanning tunneling microscopy (STM) allows understanding electronic properties and their relationship with atomic structures.
- Magnetic skyrmions are revealed for the first time in a van der Waals ferromagnet Fe_3GeTe_2 with Spin-polarized-STM
- Current-induced spin polarization and spin-momentum-locked conductance are detected with the first spin-polarized 4-probe-STM