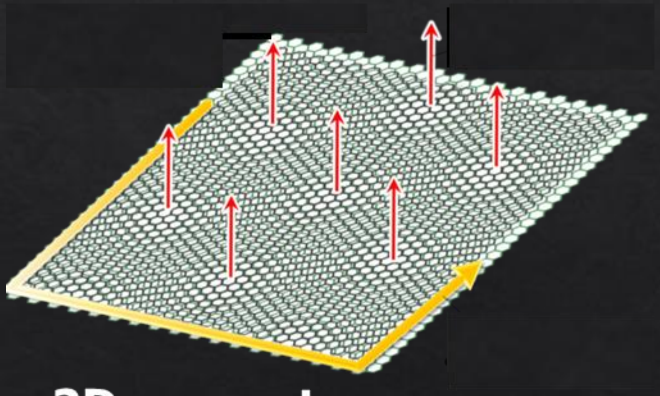
Emergent electronic and magnetic phases in quasi-2D vdW ferromagnet

Atindra Nath Pal
S N Bose National Center for Basic Sciences



Riju Pal   Dr. Buddhadeb Pal

Collaborators

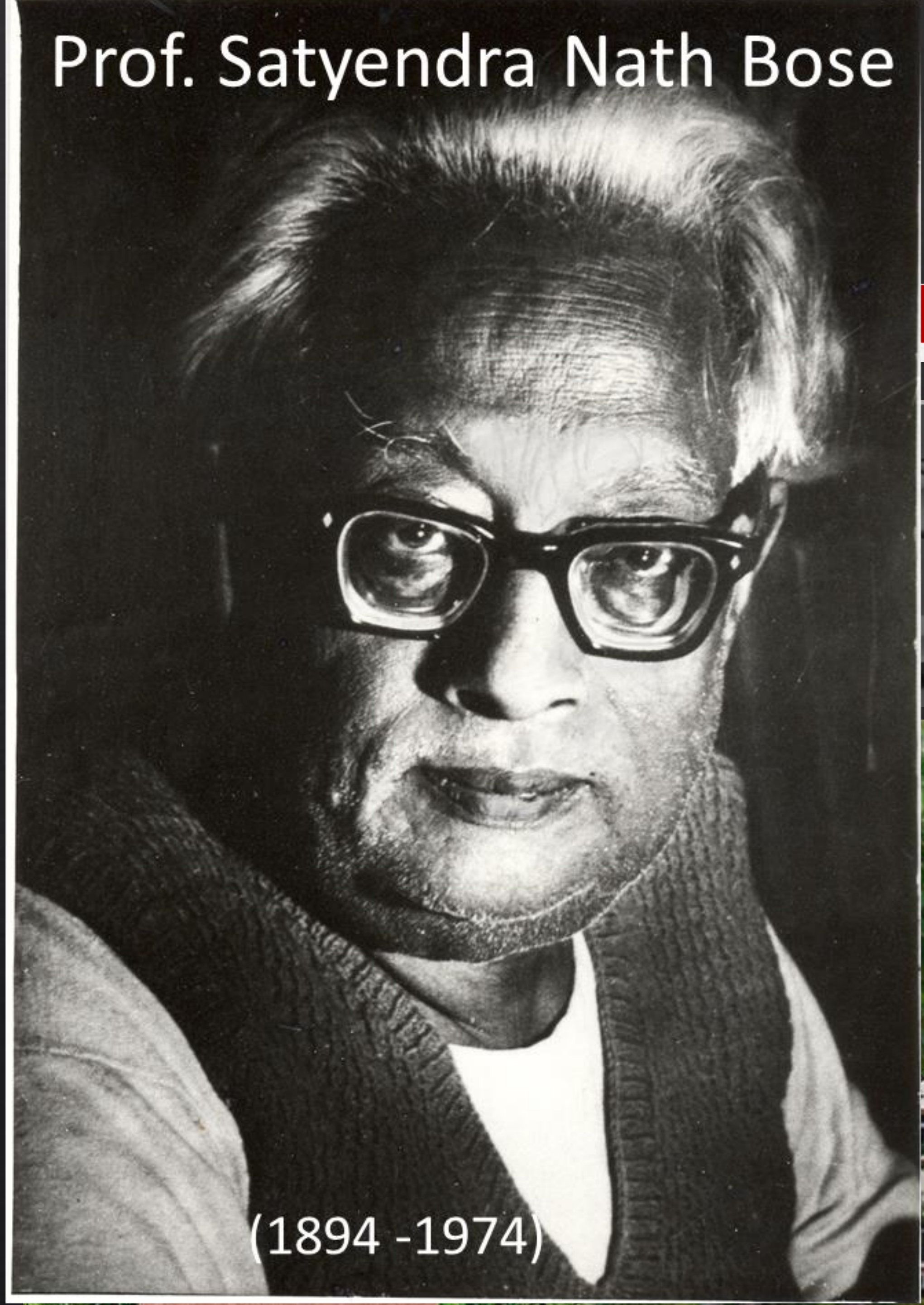
- Prabhat Mandal, SNBNCBS
- Tanmoy Das – Theory
- Alexey Alfansov Vladislav Kataev and Bernd Büchner, IFW Dresden

Engineered 2D Quantum Material, 15th – 26th July 2024

CENTRE FOR BASIC SCIENCES 1986 S. N. BOSE NATIONAL 1894-1974

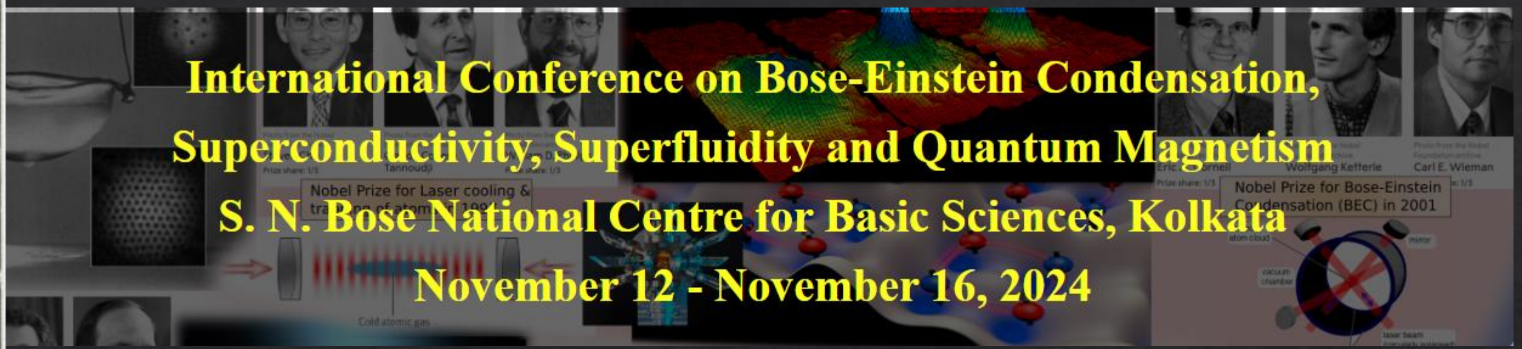
IFW

Department of Science & Technology, Government of India

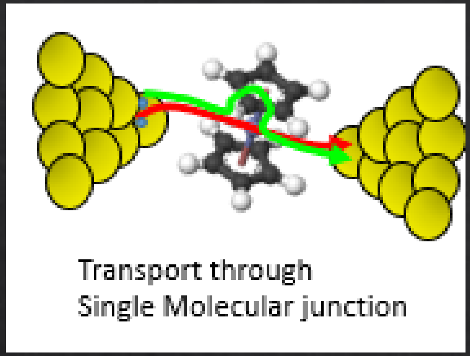


S N Bose National Center for Basic Sciences, Kolkata

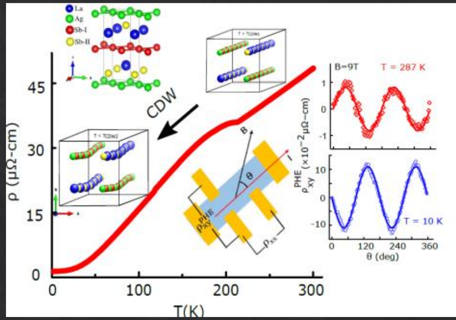
BoseStat@100: Centenary of Bose Statistics



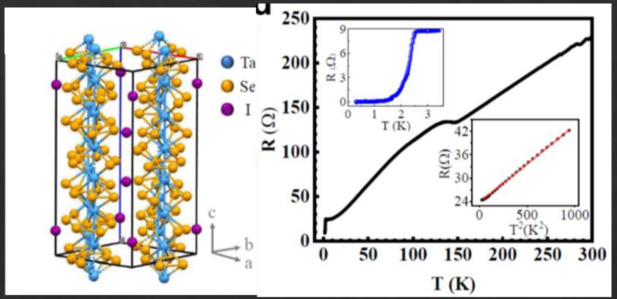
Quantum transport and Devices Lab



PRB (letter), 2021
Nanoscale (2023)
Nano Letter (2023)
Nanoscale (2023)

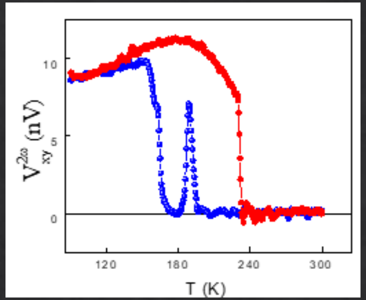


Chiral CDW
Advanced Functional Materials (2023)

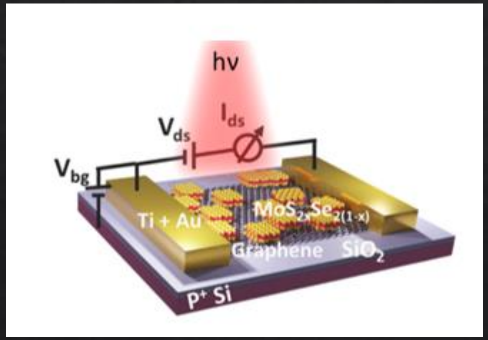


Non-centrosymmetric $Ta(Se_4)_3I$, CDW, superconductivity and magnetism

Arxiv (2021)

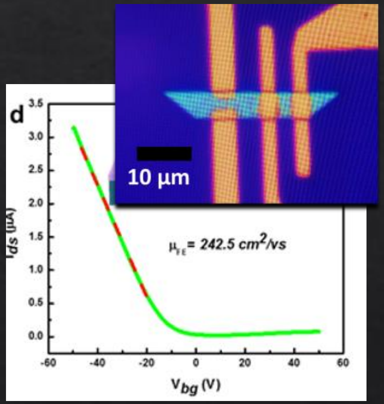


Nonlinear Hall effect in $1T\text{-}TaS_2$
under preparation

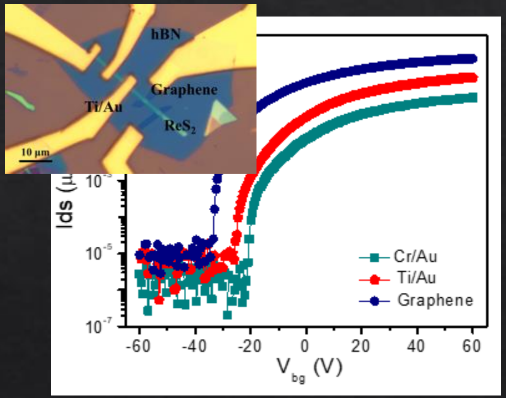


Light matter interaction
Optoelectronics

ACS AMI (2022)
Advanced Optical Material (2022)
ACS AMI (2022)
Phys. Rev. Applied (2023)

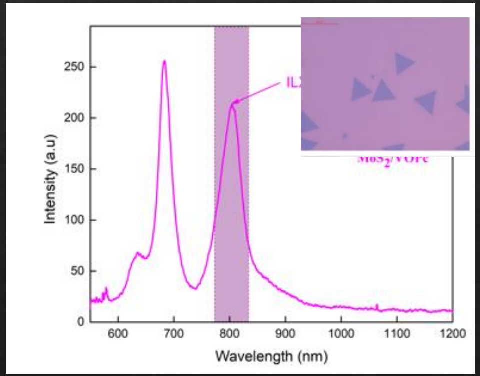


P type Tellurene
under review



n-type ReS_2 contact engineering

submitted



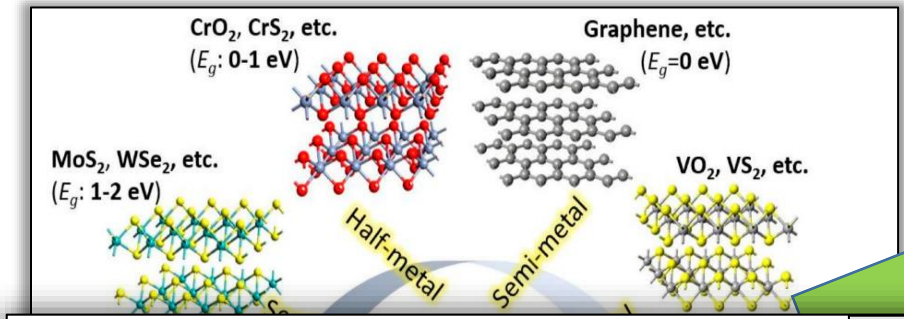
Hybrid 2D /organic Semiconductor
under preparation

Plan

- Unusual magnetism and transport in Fe_4GeTe_2
- ESR measurement in Fe_4GeTe_2

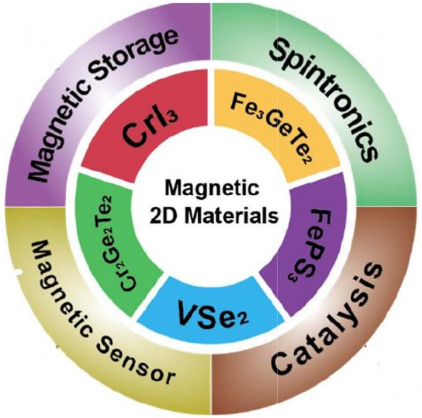
Group picnic

2D Ferromagnet



Missing member till 2016

2D Magnets



CrI$_3$ (2017)

LETTER

doi:10.1038/nature22391

Layer-dependent ferromagnetism in a van der Waals crystal down to the monolayer limit

Bevin Huang[1]*, Genevieve Clark[2]*, Efrén Navarro-Moratalla[3]*, Dahlia R. Klein[3], Ran Cheng[4], Kyle L. Seyler[1], Ding Zhong[1], Emma Schmidgall[1], Michael A. McGuire[5], David H. Cobden[1], Wang Yao[6], Di Xiao[4], Pablo Jarillo-Herrero[3] & Xiaodong Xu[1,2]

Since the discovery of graphene[1], the family of two-dimensional materials has grown, displaying a broad range of electronic properties. Recent additions include semiconductors with spin-valley coupling[2], Ising superconductors[3–5] that can be tuned into a quantum metal[6], possible Mott insulators with tunable charge-

A variety of layered magnetic compounds have recently been investigated to determine whether their magnetic properties can be retained down to monolayer thickness[12–14,26]. Recent Raman studies suggest ferromagnetic ordering in few-layer $Cr_2Ge_2Te_6$ and antiferromagnetic ordering in monolayer $FePS_3$[27,28]. However, no evidence yet exists for

CrGeTe3 (2017)

LETTER

doi:10.1038/nature22060

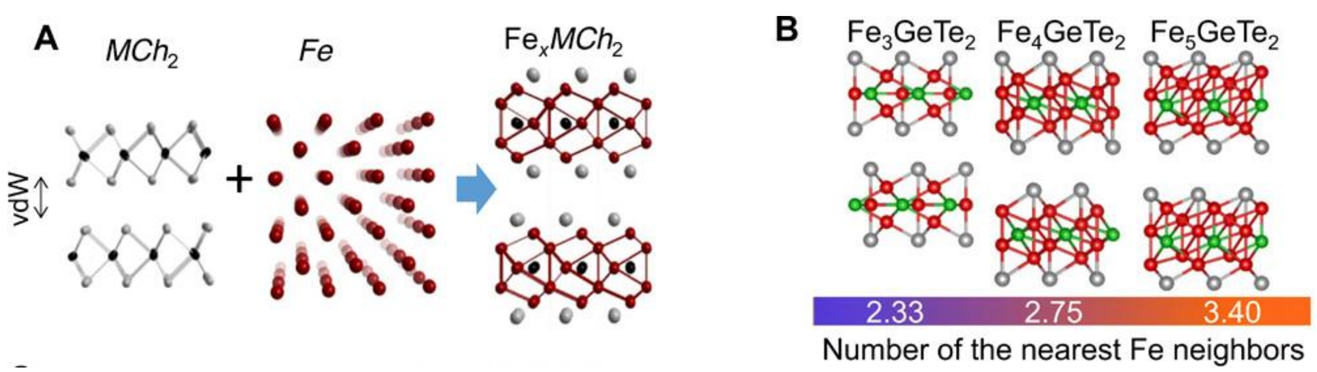
Discovery of intrinsic ferromagnetism in two-dimensional van der Waals crystals

Cheng Gong[1]*, Lin Li[2]*, Zhenglu Li[3,4]*, Huiwen Ji[5], Alex Stern[2], Yang Xia[1], Ting Cao[3,4], Wei Bao[1], Chenzhe Wang[1], Yuan Wang[1,4], Z. Q. Qiu[3], R. J. Cava[5], Steven G. Louie[3,4], Jing Xia[2] & Xiang Zhang[1,4]

In this case, the magnetic anisotropy of the system, stabilized the long range magnetic order down to monolayer and suppresses the effect of thermal fluctuations#.

#M. Bander and D. L. Mills, Phys. Rev. B ***38****, 12015–12018 (1988).*

Strategy to increase T_C



Seo et al., Sci. Adv. , 6, (2020)

RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 38, NUMBER 16 1 DECEMBER 1988

Ferromagnetism of ultrathin films

Myron Bander and D. L. Mills

Institute for Surface and Interface Science and Department of Physics, University of California, Irvine, California 92717

(Received 10 August 1988)

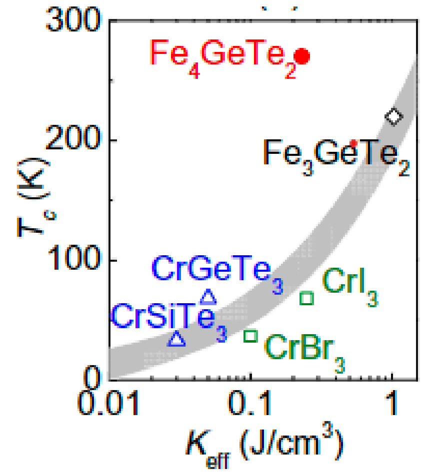
We discuss the nature of ferromagnetism in ultrathin films of magnetic ions, here regarded as two-dimensional Heisenberg ferromagnets subject to uniaxial anisotropy with the easy axis normal to the film. We show that a phase transition to ferromagnetism occurs always for arbitrarily small anisotropy. Renormalization-group scaling relations for the transition temperature and the temperature variation of the correlation length are obtained. Implications of these results are discussed.

$$T_c \sim J/\ln(3\pi J/4\,K)$$

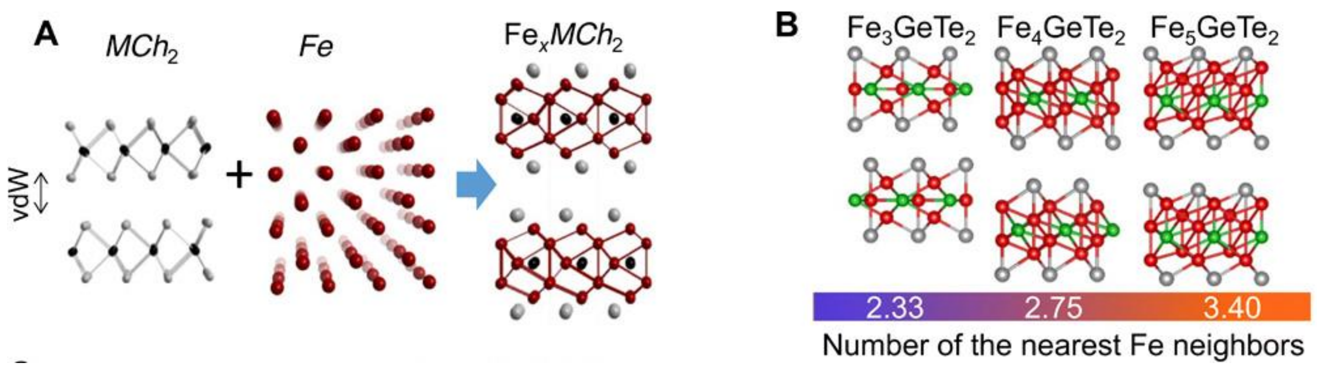
3D like exchange interaction keeping the vdW structure intact!

2D FM Metal: **Fe_nGeTe_2** where, n = 3, 4, 5

Fe_3GeTe_2 (T_c - 210K), Fe_4GeTe_2 (270K), Fe_5GeTe_2 (310K)



Strategy to increase T_C



Seo et al., Sci. Adv. , 6, (2020)

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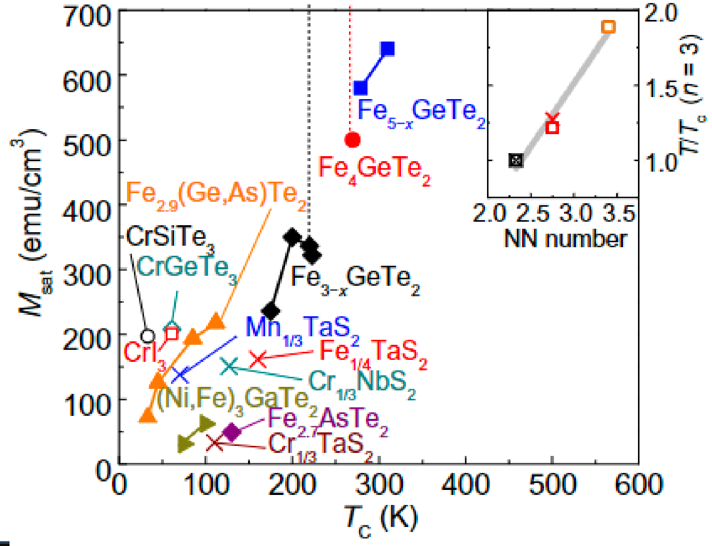
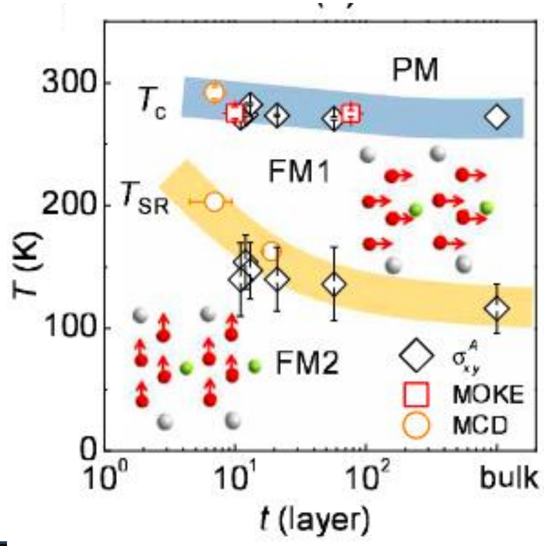




3D like exchange interaction keeping the vdW structure intact!

2D FM Metal: **Fe_nGeTe_2** where, n = 3, 4, 5

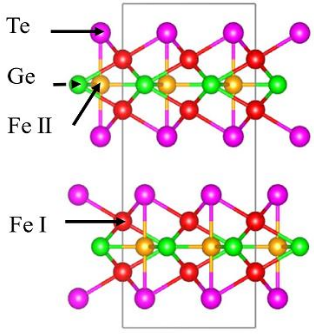
Fe_3GeTe_2 (T_c - 210K), Fe_4GeTe_2 (270K), Fe_5GeTe_2 (310K)

Fe-rich vdW ferromagnets are promising spin-source materials.

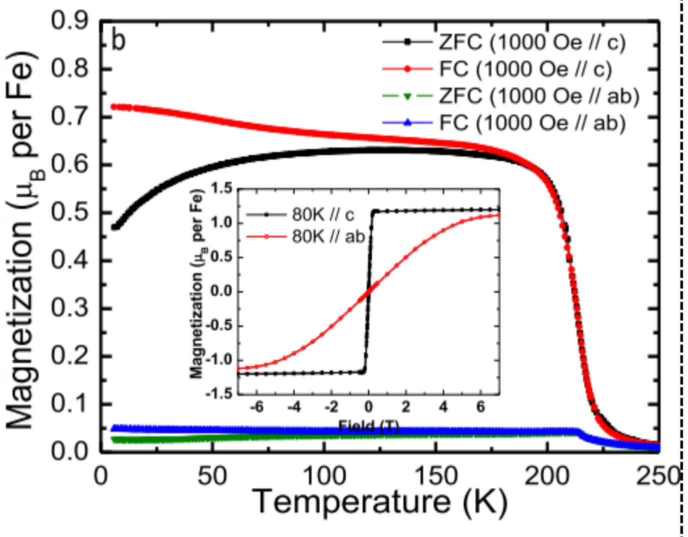
- High T_C, High M_{SAT}, Metallic-easier for spin injection.
- Less anisotropy.

Fe_nGeTe_2 family (n = 3, 4, 5)

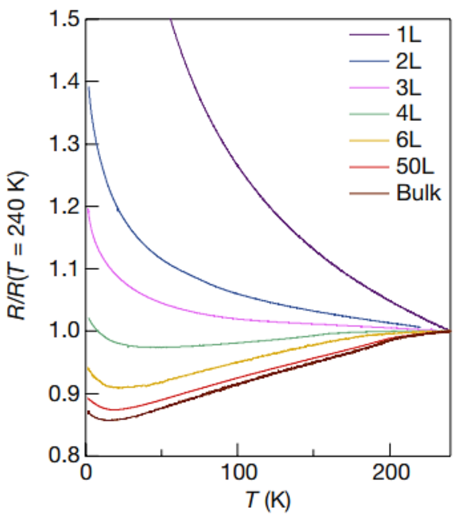
Fe_3GeTe_2 (2018)



Side view[1] of the atomic lattice of bilayer F3GT

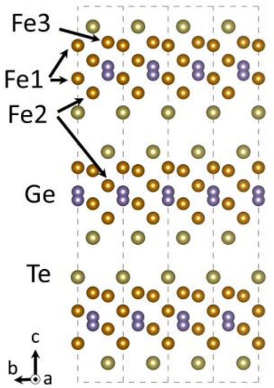


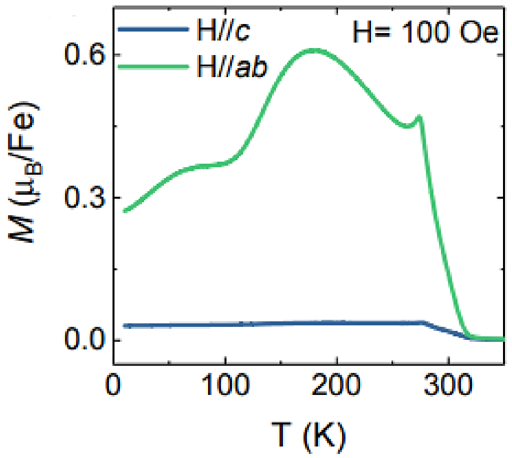
- Hexagonal structure ($P6_3/mmc$ space group) with **T_C ~ 210 K** (bulk F3GT)[1,2]
- **Easy-axis FM** with a large perpendicular magnetic anisotropy down to monolayer[2].
- Co-existence[2] of AFM+FM (152-214K)
- Thickness dependent[3] T_C (130K for monolayer (0.8 nm))

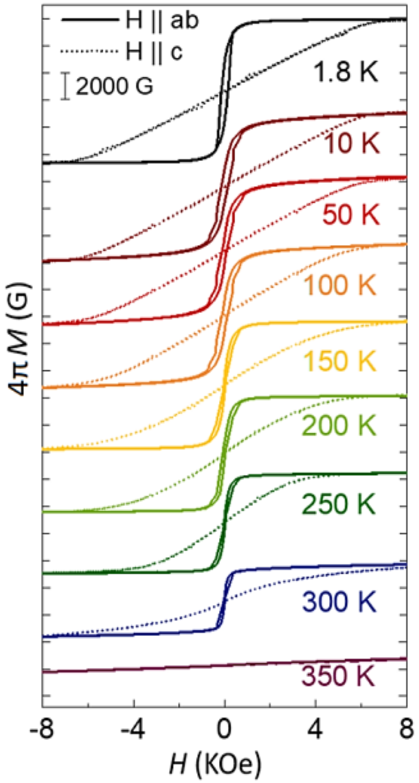


[1]Jieyu, Yi et. al., 2D Mater. **4** 011005 (2017)
[2]Fei, Z. Y. et. al., Nat. Mater. **17**, 778–782 (2018)
[3]Deng, Y., et al., Nature **563,** 94–99 (2018)

Fe_5GeTe_2 (2019)



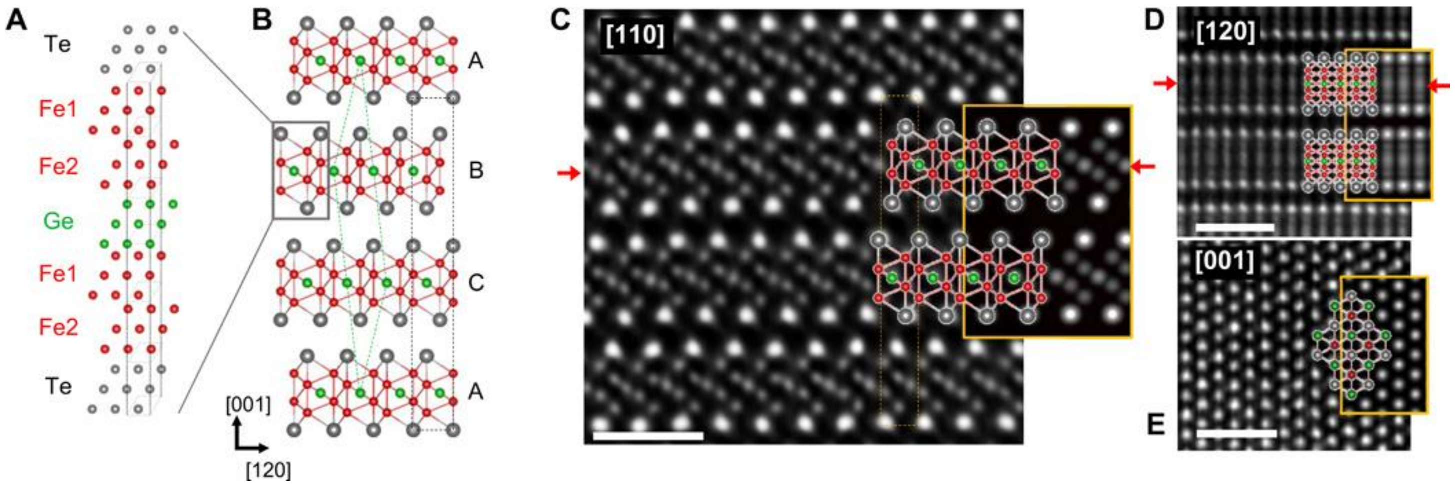




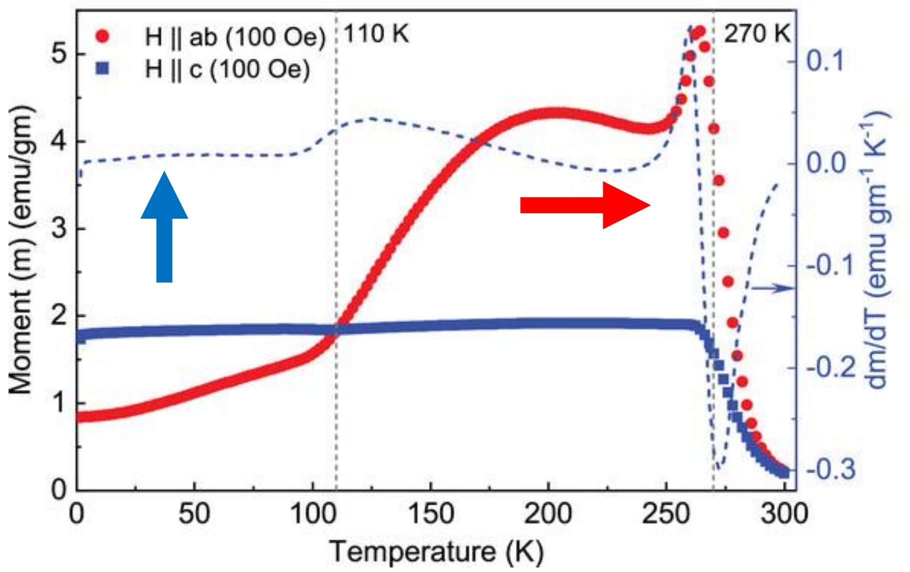
- Rhombohedral structure (space group $R\bar{3}m$) with **T_C ~ 310 K** (bulk F5GT)[4]
- **Easy-plane FM** with in-plane magnetic anisotropy[4] in bulk.
- Complex ferromagnetic to ferrimagnetic transition at 275 K, and then to glassy clusters state[5] below 110 K (in bulk).
- Switch of magnetic anisotropy from in-plane to out-of-plane for bilayer F5GT, frozen spin-glass-like behavior for monolayer F5GT[6].

[4]May, A. F. et. al., ACS Nano **13**, 4436 (2019)
[5]Zhang, H. et. al., Phys. Rev. B **102**, 064417 (2020)
[6]Deng, Y. et. al., Nano Lett. **22**, 9839–9846 (2022)

Crystal Structure, Magnetization of Fe_4GeTe_2



Seo et al., Sci. Adv. , 6, (2020)

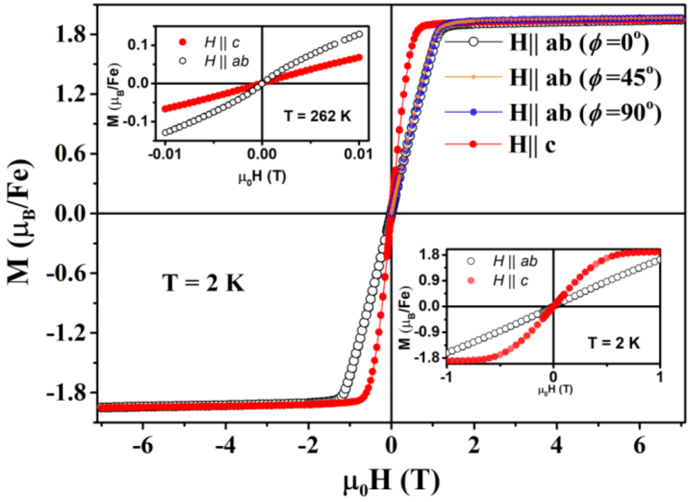


Rhombohedral structure
space group R‾3m (No.166).

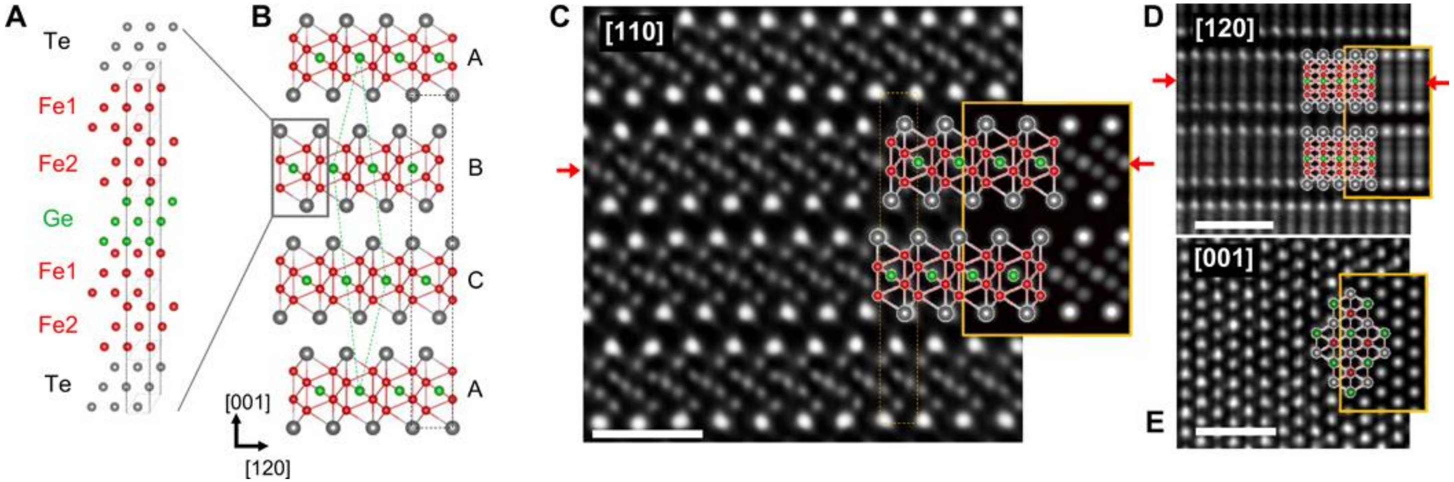
Single crystal:
Prabhat Mandal (SINP), currently Emeritus Professor at SNBNCBS

S Mondal,…,P. Mandal, PRB 104, 094405 (2021)

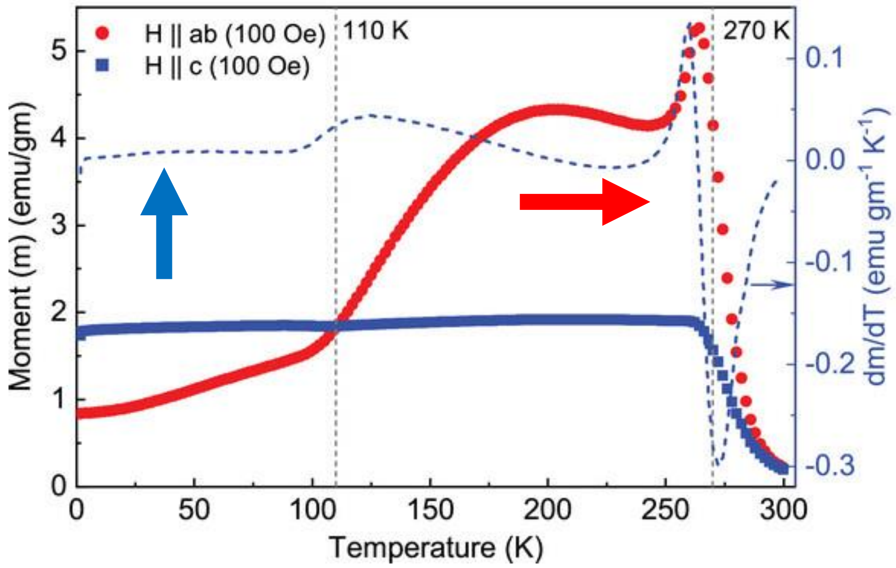
Competition between shape and magneto-crystalline anisotropy?



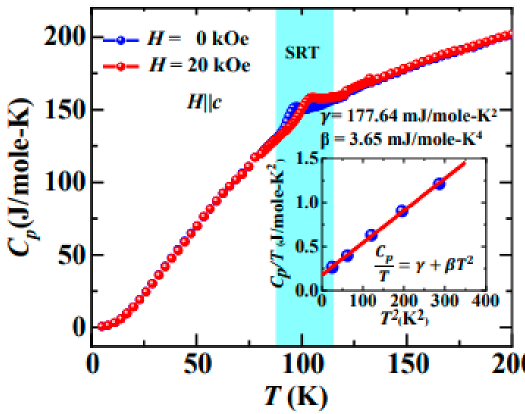
Crystal Structure, Magnetization of Fe_4GeTe_2



Seo et al., Sci. Adv. , 6, (2020)



Thermodynamic transition



Rhombohedral structure
space group $R\bar{3}m$ (No.166).

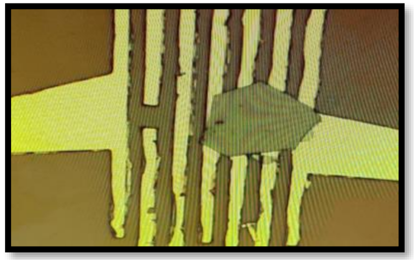
Single crystal:
Prabhat Mandal (SINP), currently Emeritus Professor at SNBNCBS

S Mondal,…,P. Mandal, PRB 104, 094405 (2021)

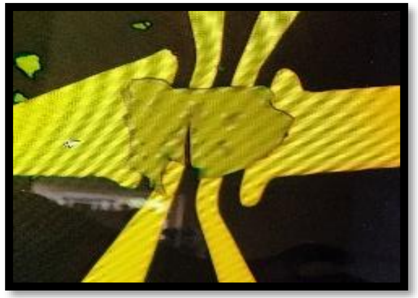
S Bera, …, AN Pal, M Mondal, JMMM (2022)

With Mintu Mandal (IACS, Kolkata)

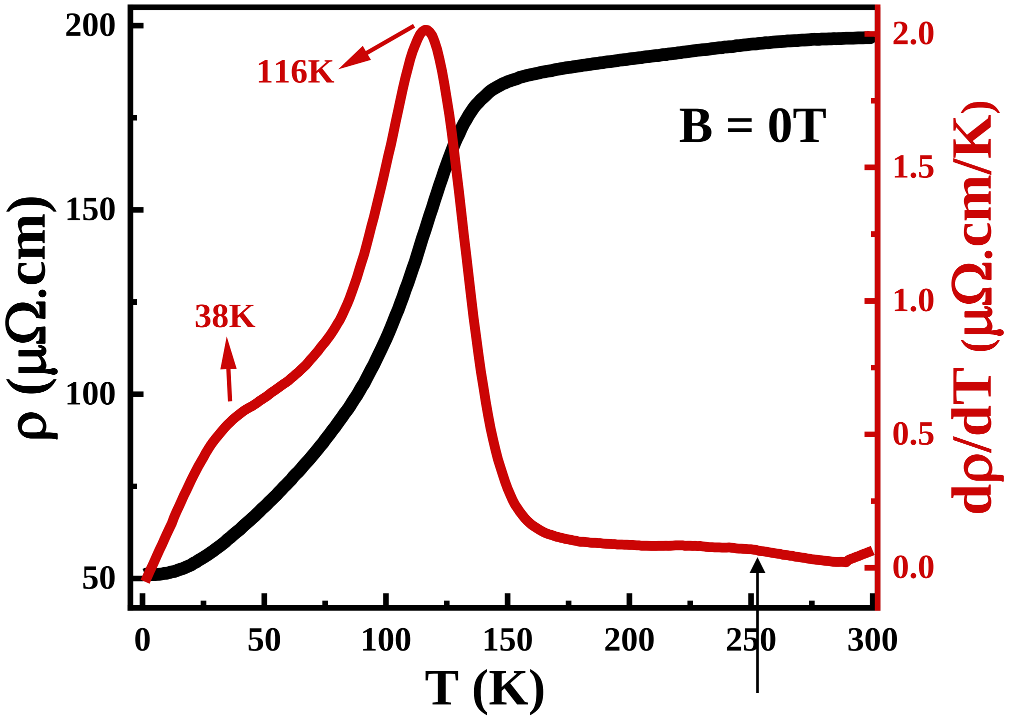
Resistivity: Temperature dependence



Device 1 (16 nm)



Device 2 (95 nm)



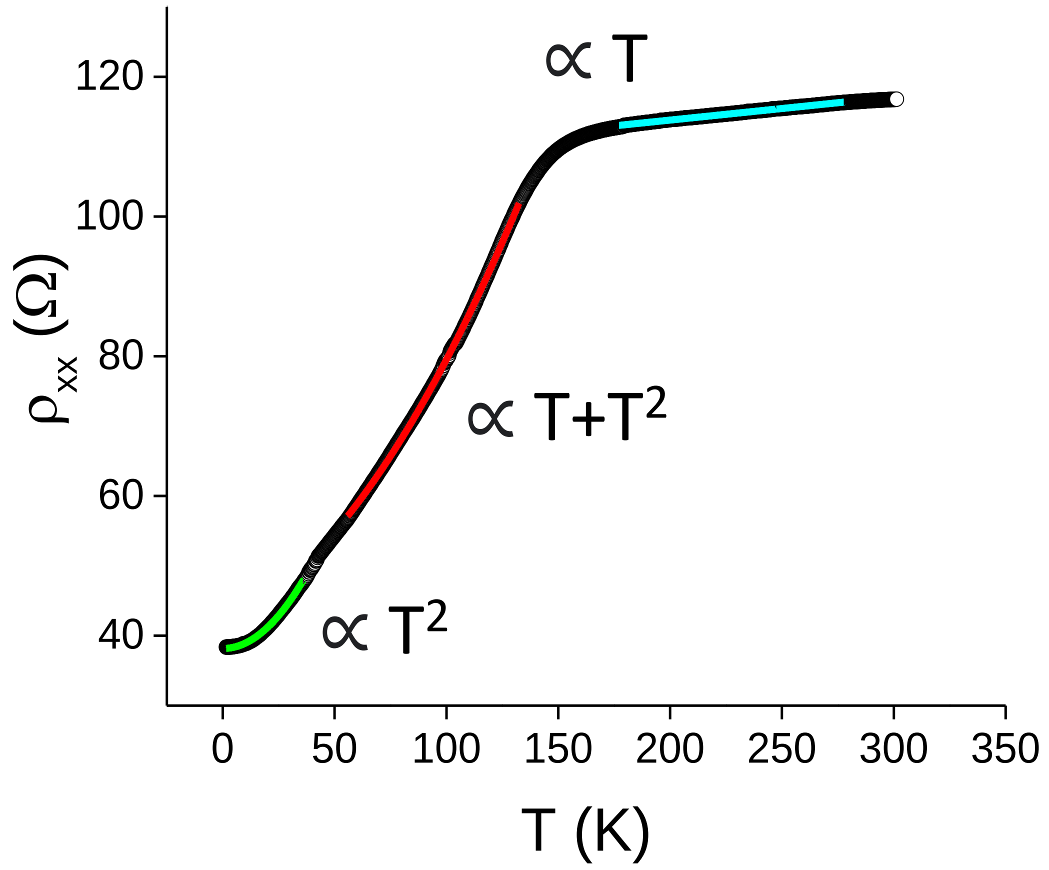
FM transition

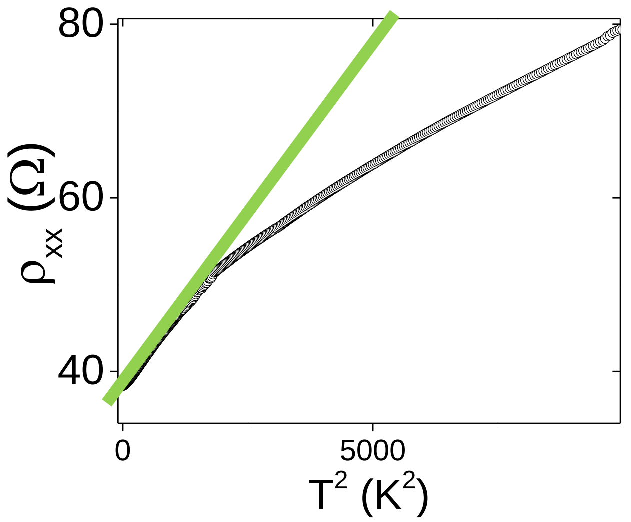
Dramatic effect near spin reorientation transition

Resistivity: Temperature dependence

$$\rho_{total} = \rho_{res} + \rho_{e\text{-}e}(T) + \rho_{e\text{-}ph}(T) + \rho_{e\text{-}m}(T)$$

PRB, 66, 024433 (2002)

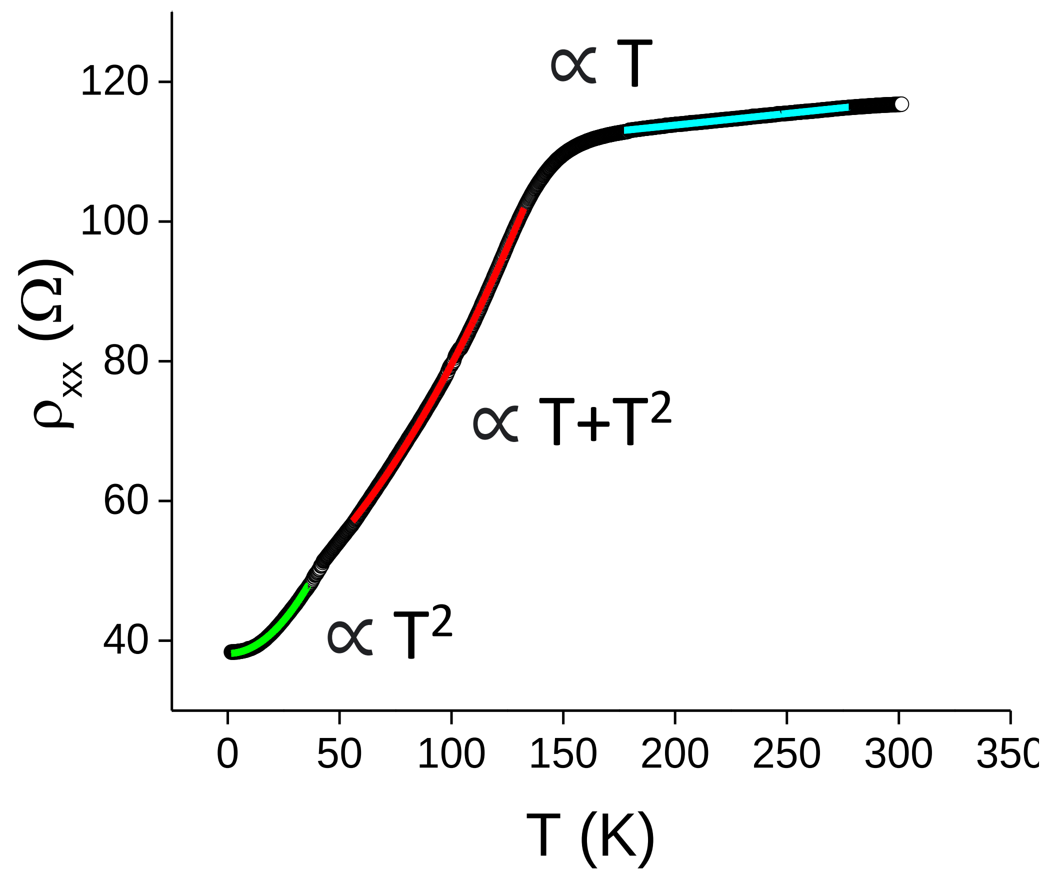


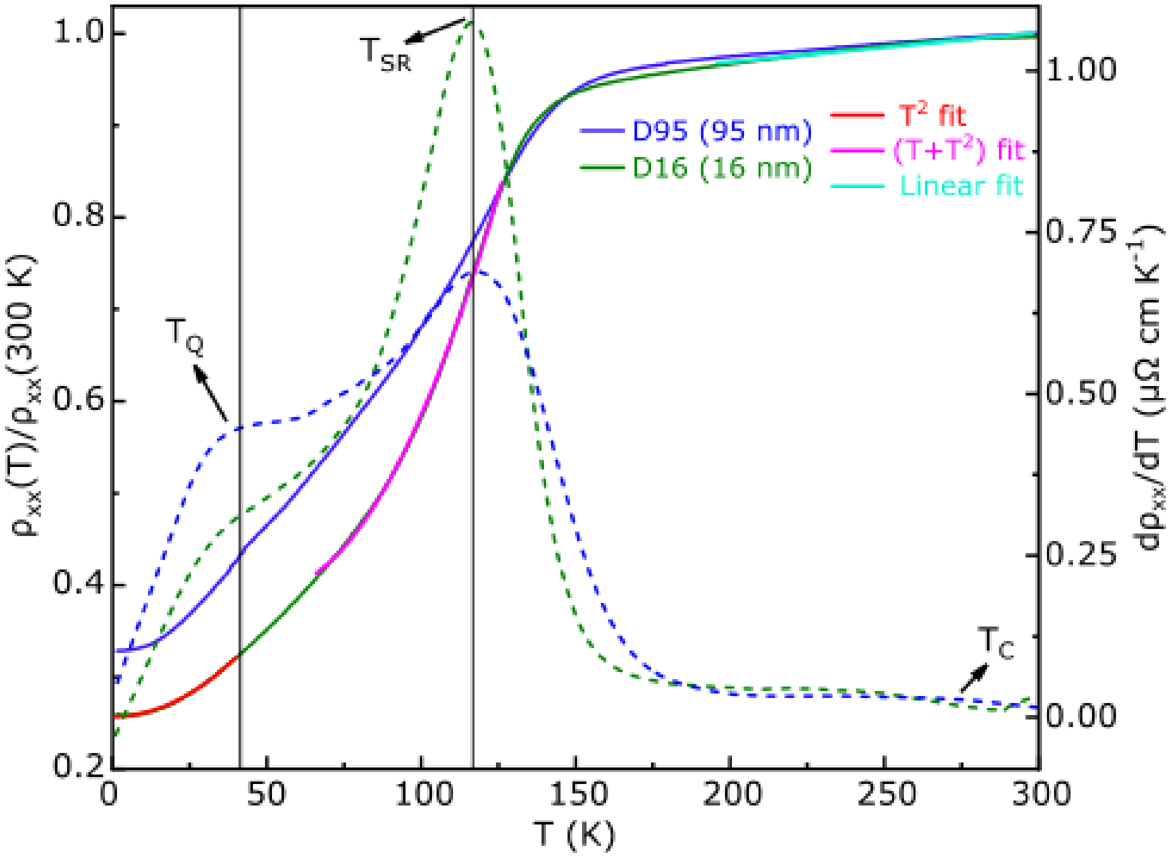


Resistivity: Temperature dependence

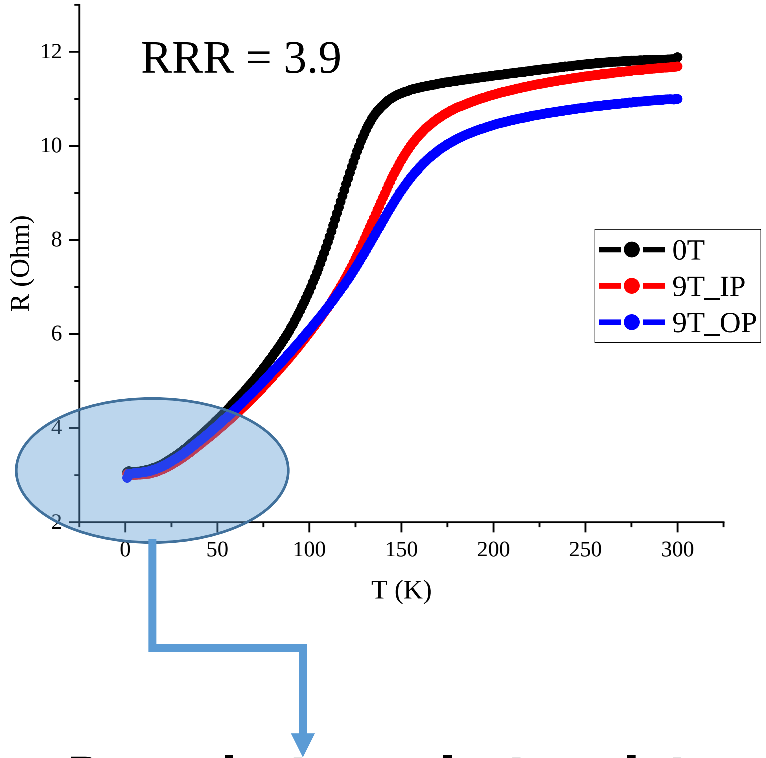
$$\rho_{total} = \rho_{res} + \rho_{e\text{-}e}(T) + \rho_{e\text{-}ph}(T) + \rho_{e\text{-}m}(T)$$

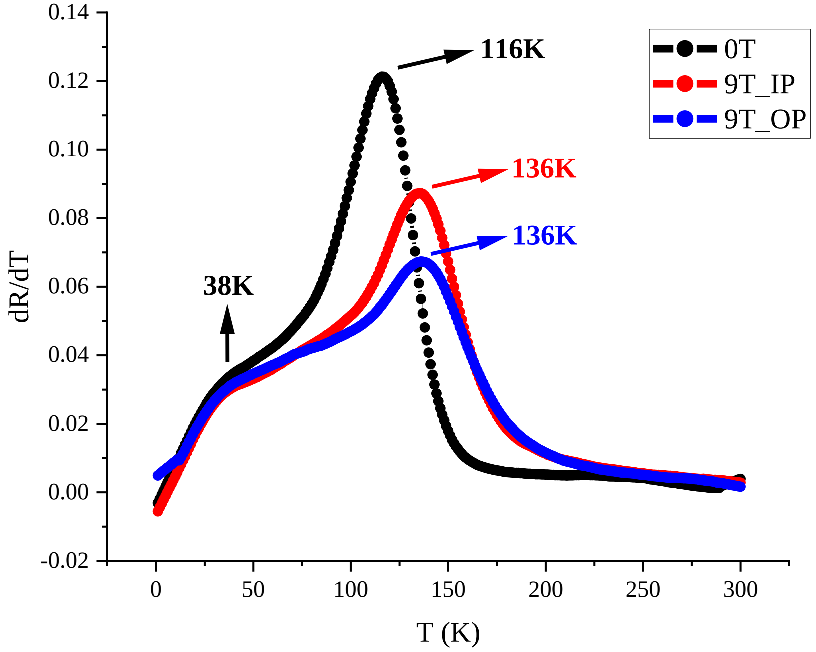
PRB, 66, 024433 (2002)





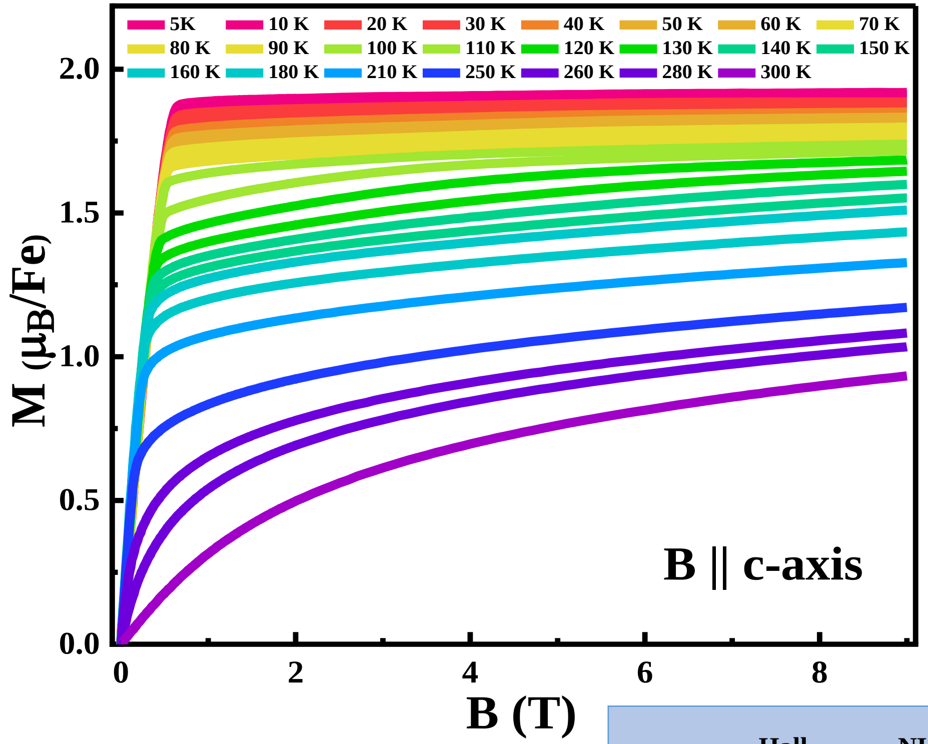
Resistivity: Temperature dependence

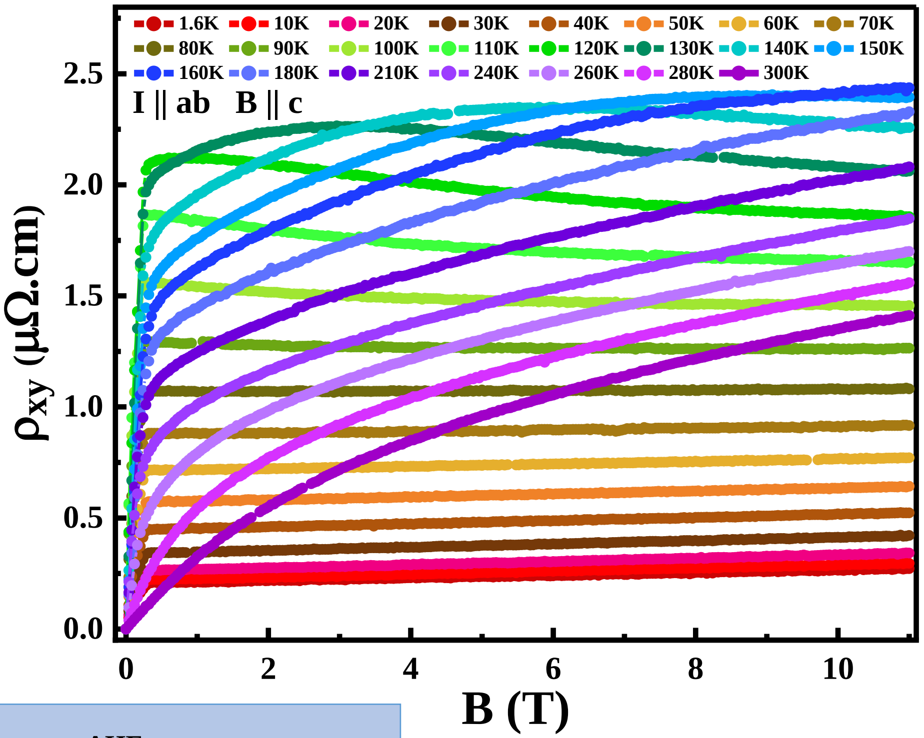




Pure electron-electron interaction, does not change with magnetic field, Behaves like metal

Hall effect





$\rho_{xy}^{Hall} = \rho_{xy}^{NH} + \rho_{xy}^{AHE}$

$= R_o B + \mu_0 R_s M$

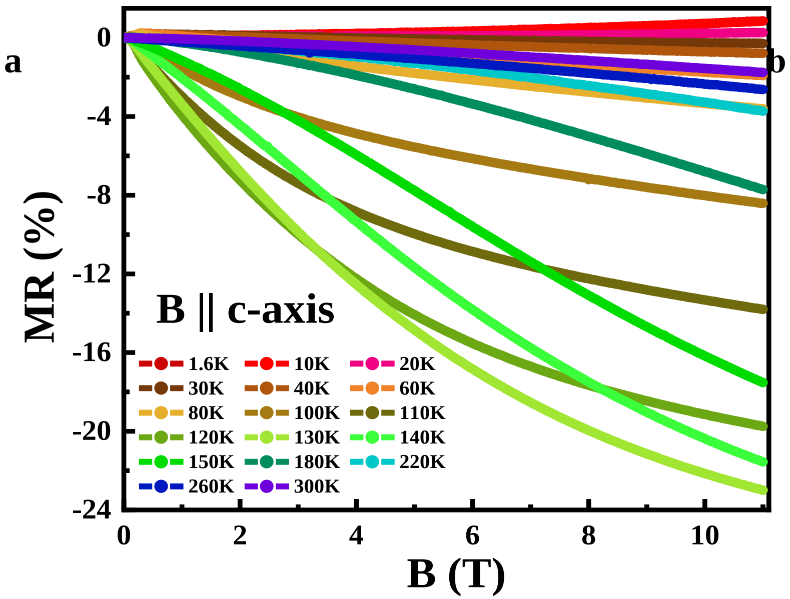
$\rho_{xy}^{Hall}/B = R_o + \mu_0 R_s (M/B)$

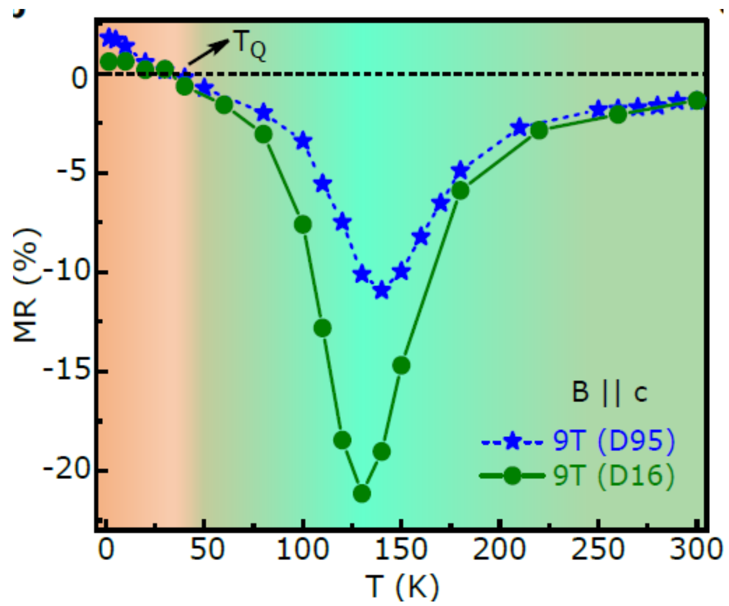
$Y = c + mX$

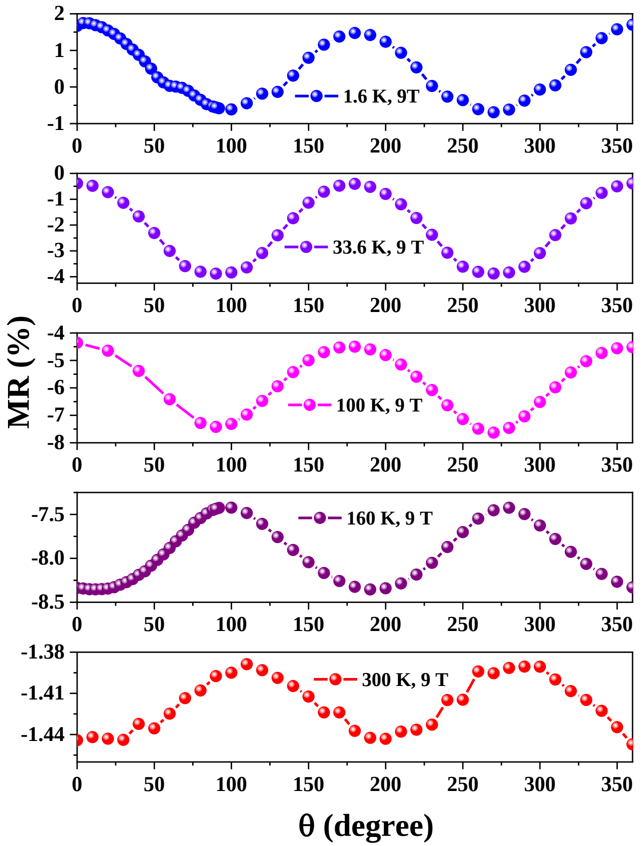
Nagaosa et al., REVIEWS OF MODERN PHYSICS, 82, 1539, (2010)

Here, R_0 and R_s are the ordinary & Anomalous Hall coefficient respectively

Magnetoresistance



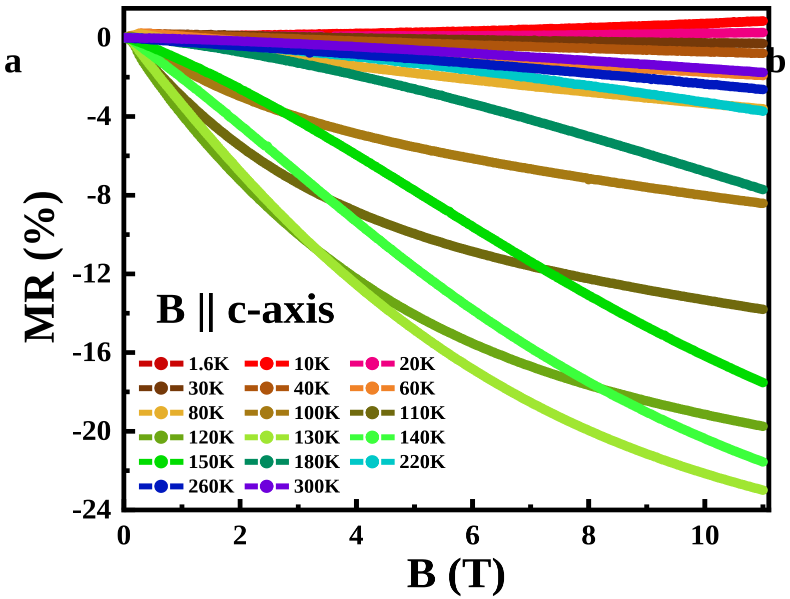


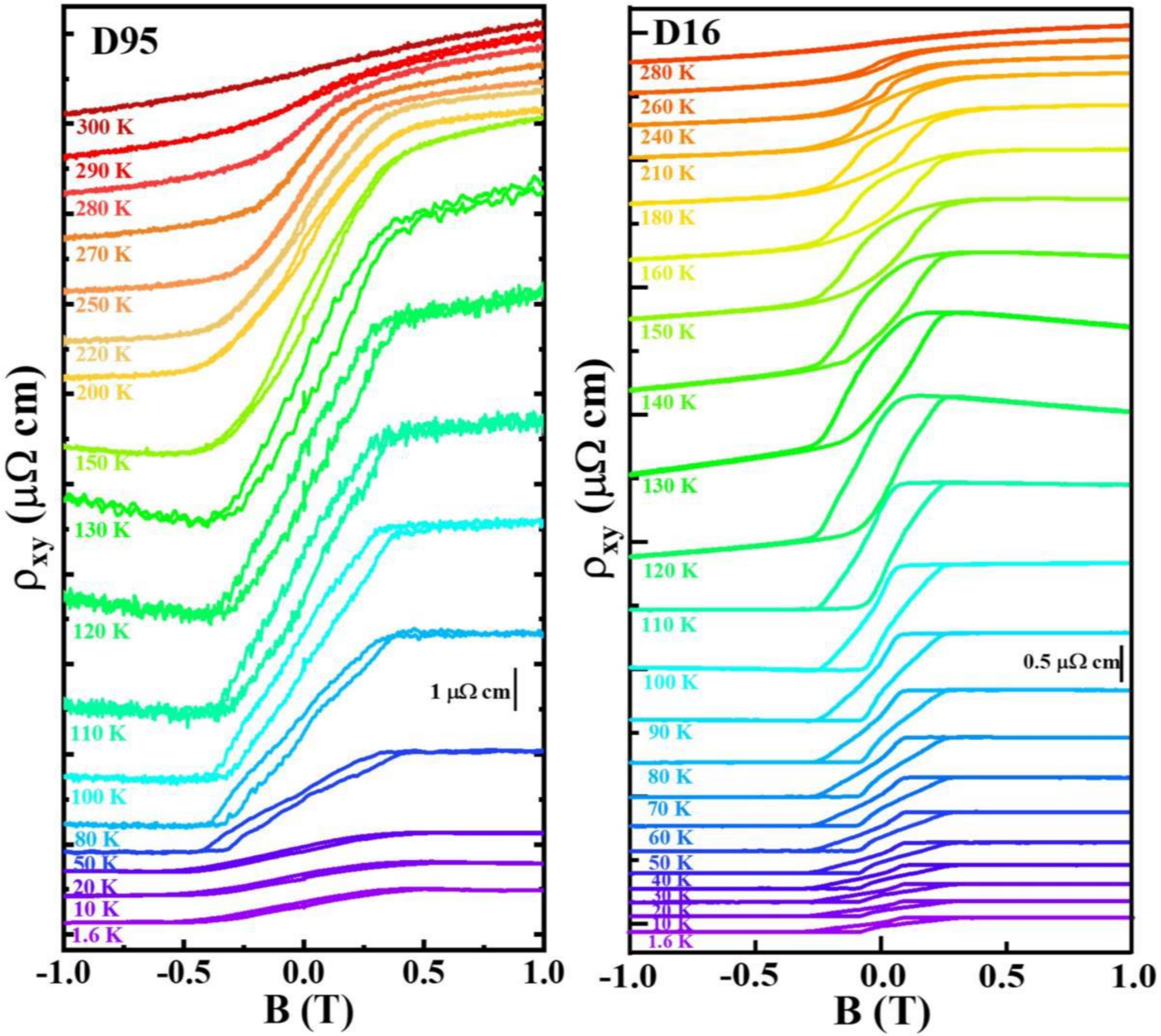


Positive MR: orbital contribution

Negative MR: electron-magnon scattering

Magnetoresistance

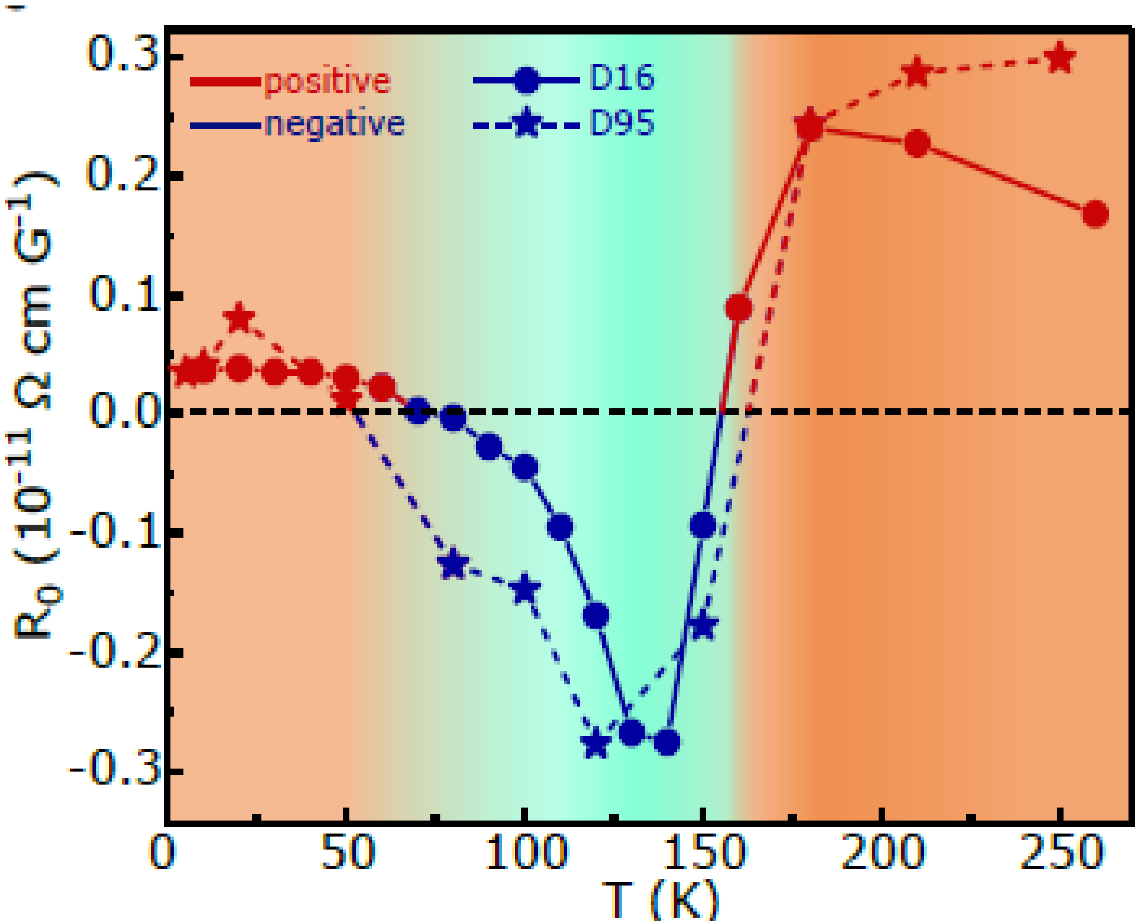




Positive MR: orbital contribution

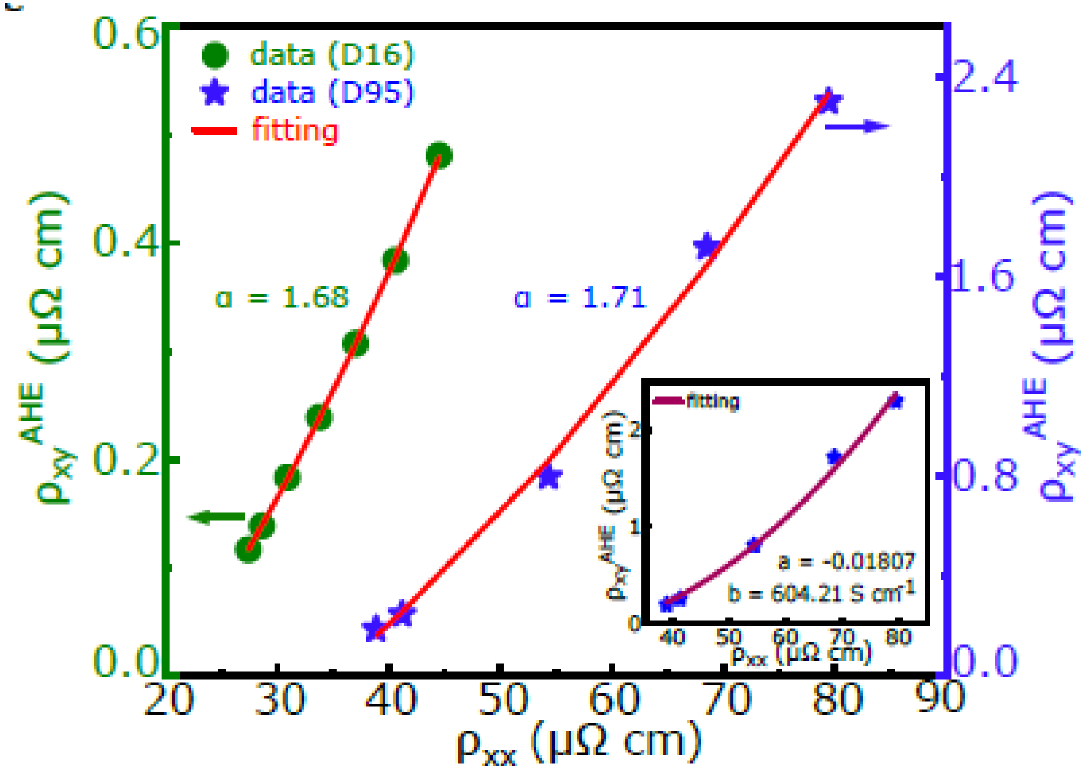
Negative MR: electron-magnon scattering

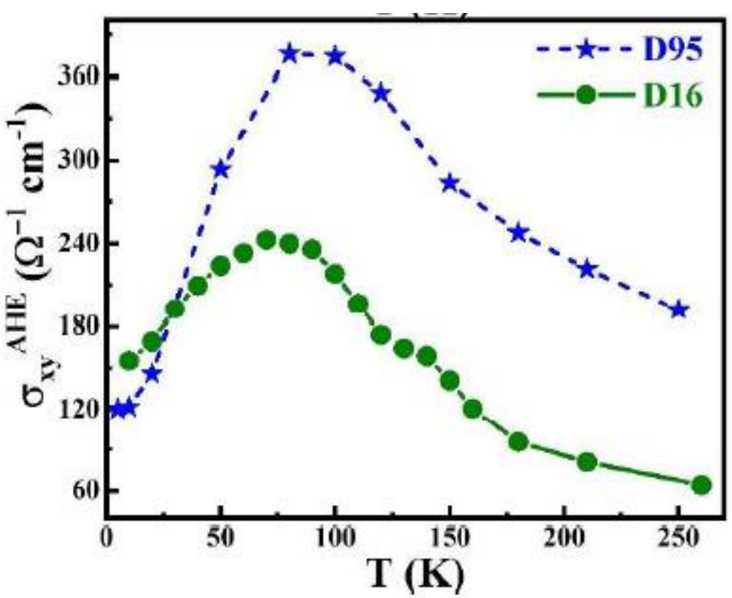
Ordinary Hall Effect: Temperature dependence



Fermi surface reconstruction near SRT

Scaling behaviour and AHC plot





Origin: interplay between magnetism and topology

IOP Publishing Phys. Scr. 98 (2023) 125916 https://doi.org/10.1088/1402-4896/ad0698

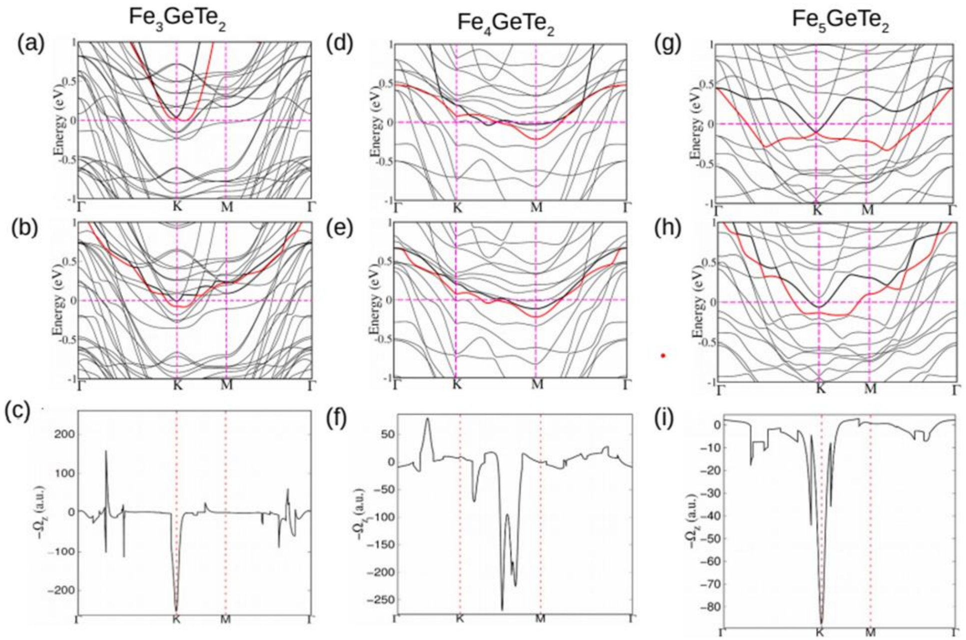
Physica Scripta

PAPER

RECEIVED 14 April 2023
REVISED 19 October 2023

Topological properties and anomalous transport in van der Waals ferromagnets Fe_nGeTe_2: a comparative study

Jyotirmoy Sau[1], S R Hassan[2,3], Nitesh Kumar[1] and Manoranjan Kumar[1]



Mostly intrinsic origin

npj computational materials www.nature.com/npjcompumats

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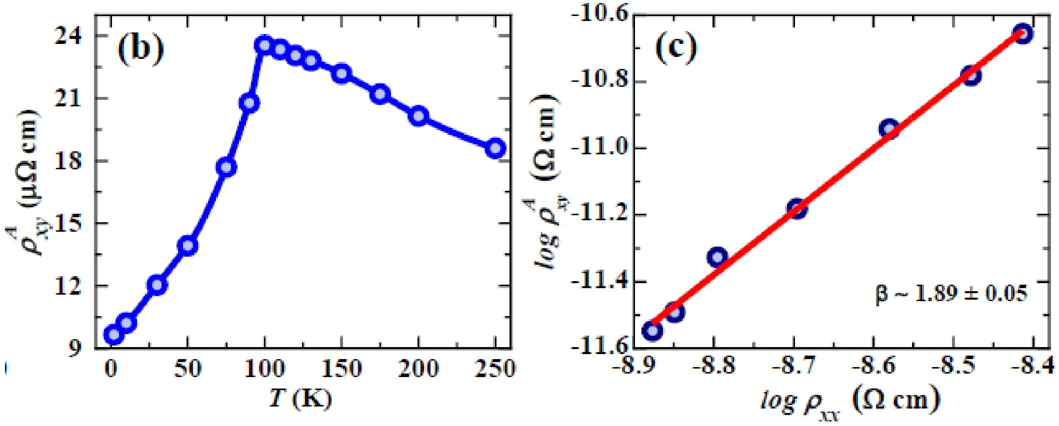
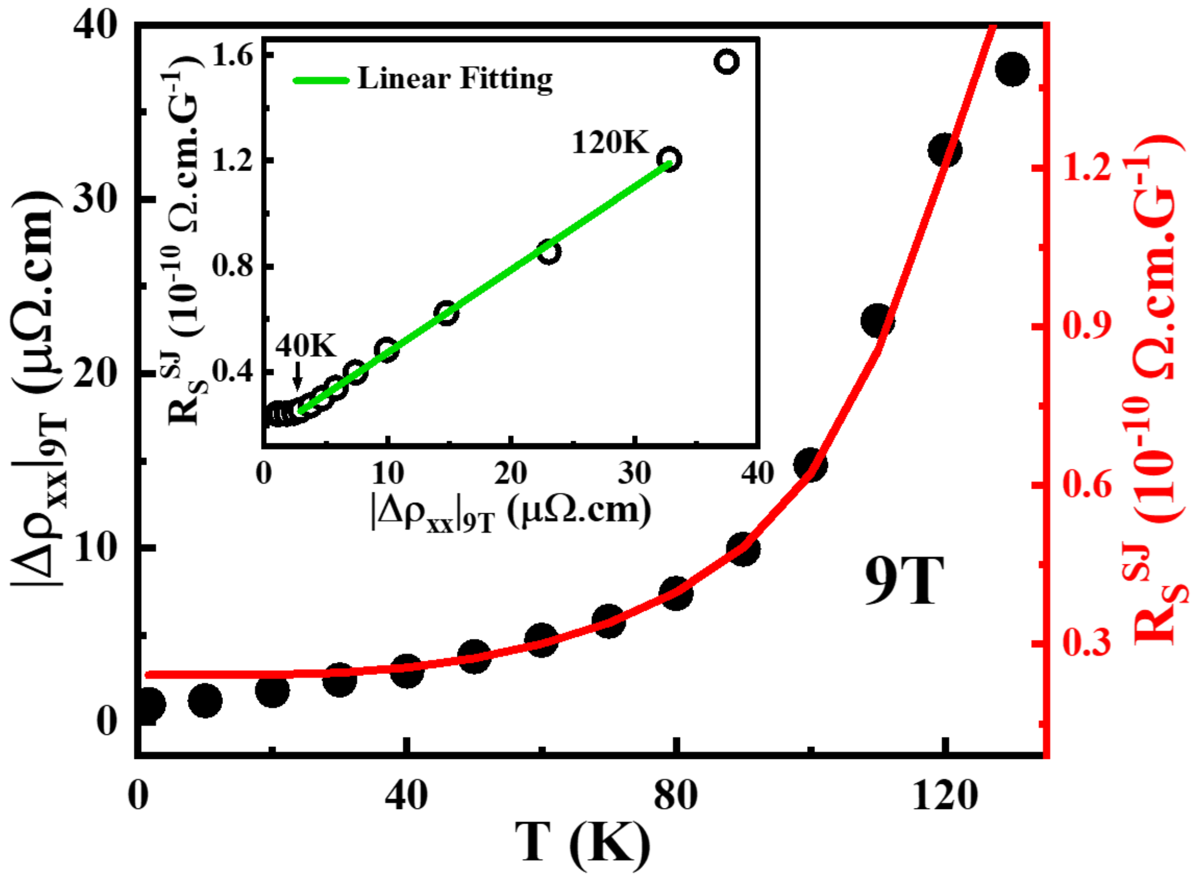
Unraveling effects of electron correlation in two-dimensional Fe_nGeTe_2 ($n = 3, 4, 5$) by dynamical mean field theory

Sukanya Ghosh[1], Soheil Ershadrad[1], Vladislav Borisov[1] and Biplab Sanyal[1]



S Bera, …A Nandy and M Mondal
Phys. Rev. B 108, 115122 (2023)

Origin: interplay between magnetism and topology



$$\rho_{xy}^{AHE} = a\rho_{xx} + b\rho_{xx}^2$$

$(\rho_{xx0} + \rho_{xxT})$

$$= (\alpha_0\rho_{xx0} + \alpha_1\rho_{xxT}) + \beta_0\rho_{xx0}^2 + \gamma\rho_{xx0}\rho_{xxT} + \beta_1\rho_{xxT}^2.$$

Skew-scattering term

Intrinsic K-L mechanism term

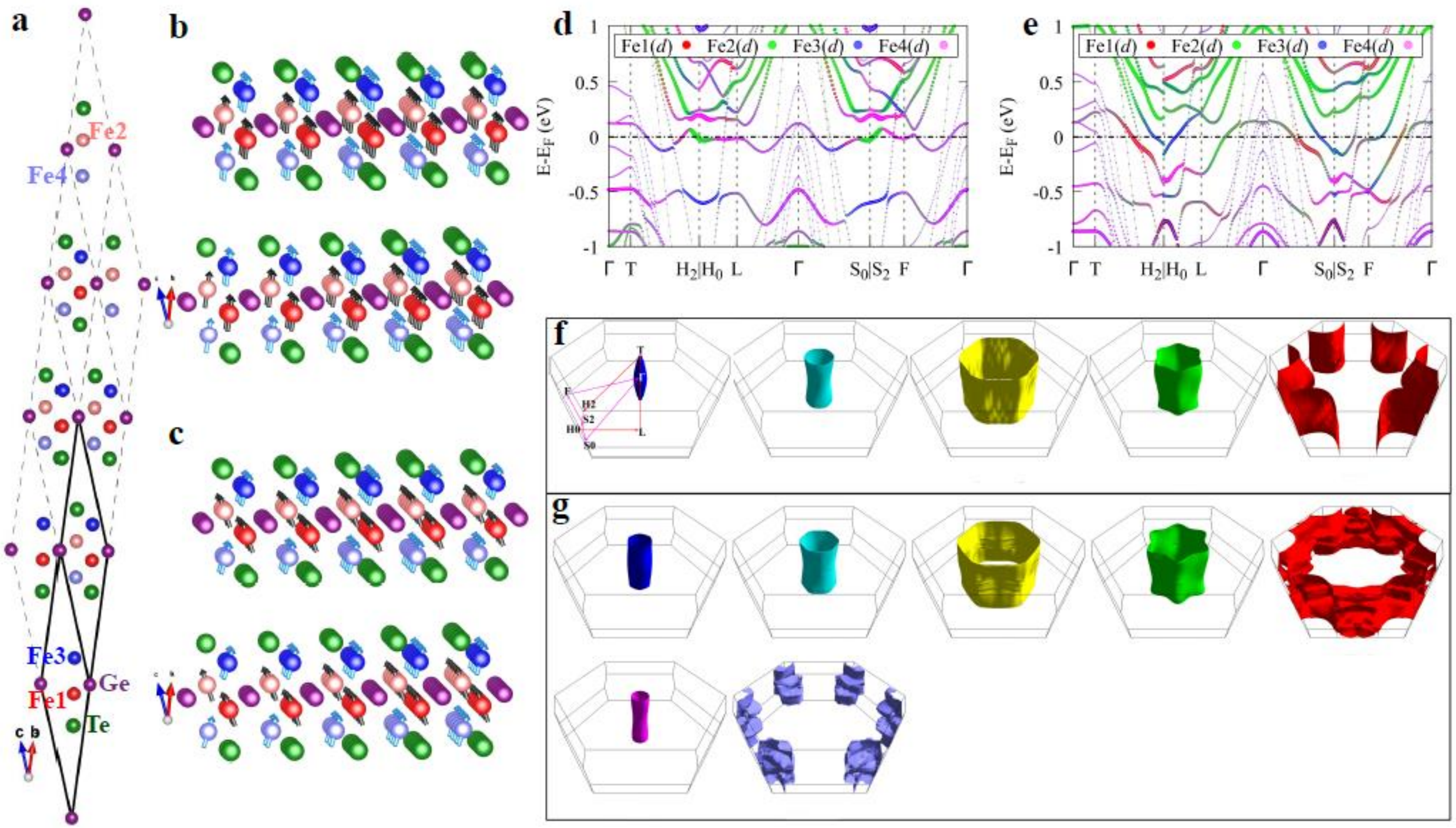
Side-jump contribution term (R_S^{SJ})

can be affected by the electron-magnon scattering, gives rise to the T-dependence of AHC.

- Temperature-dependent side-jump mechanism mainly originated from the spin-flip electron-magnon scattering in between 40K-120K.

Jena, R. P. et al. Journal of Physics: Condensed Matter **32**, *365703 (2020)*

Origin: interplay between magnetism and topology



Theory: Rajesh Sharma and Tanmoy Das, IISc

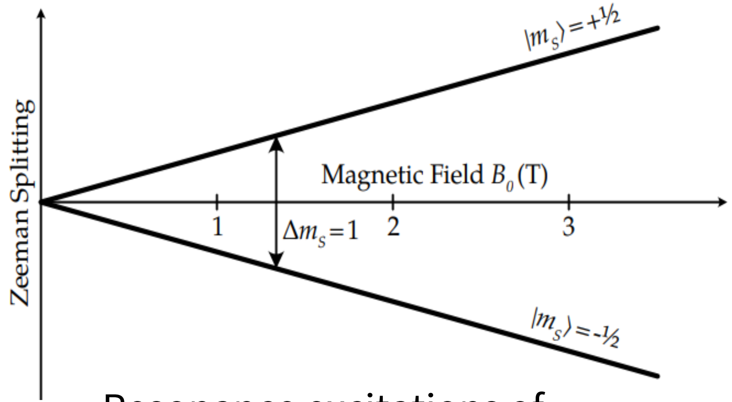
Electron Spin Resonance (ESR) Study of Fe_4GeTe_2

Electron Spin Resonance Group
Institute for Solid State Research
IFW Dresden, Helmholtzstraße 20
01069 Dresden, Germany

Electron Spin Resonance

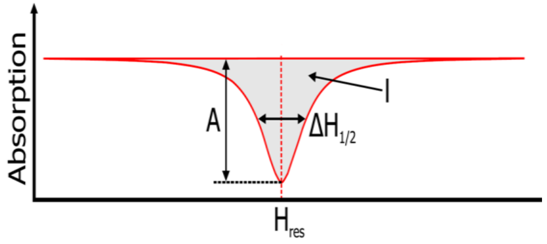
- ESR: A spectroscopic tool, mainly focuses on the spin of the electrons.

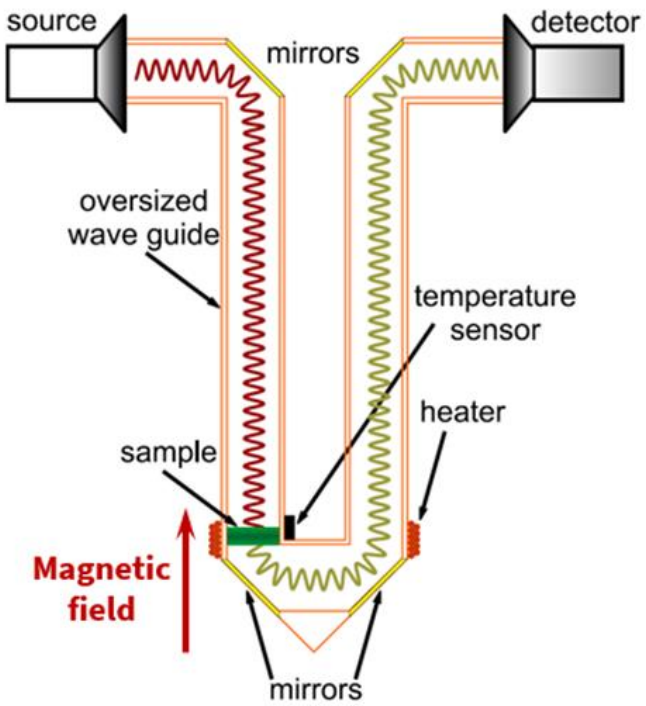
- Paramagnetic resonance condition: $h\nu = g\mu_B B_0$



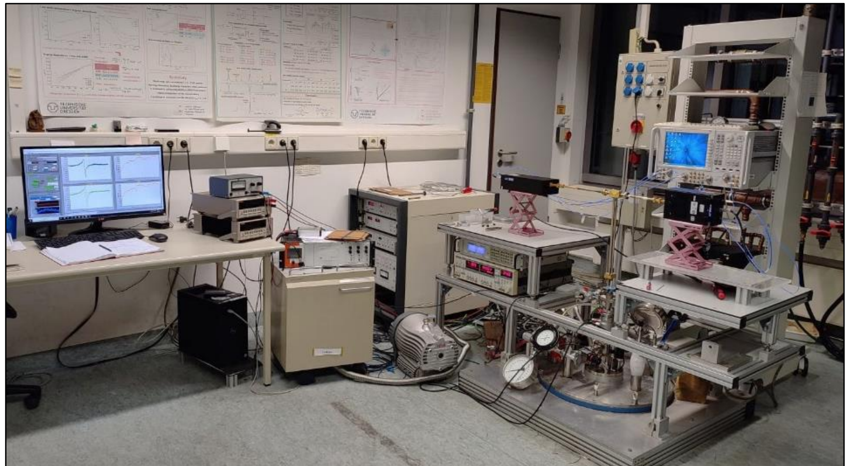
Resonance excitations of individual paramagnetic spins

Magnetic field is swept at a constant microwave frequency





- Determine g-factor
- Characterization of magnetic anisotropy, easy-axis, magnon excitation gap.
- Determination of spin-spin correlations, spin-dynamics above T_C


HFESR Lab at IFW Dresden, Germany

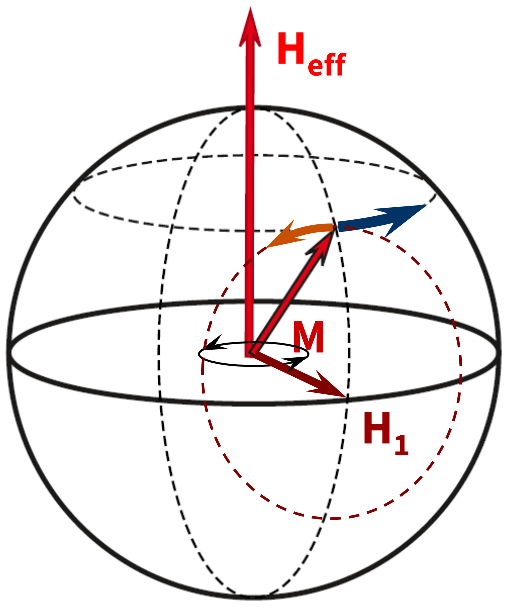
Equipment at the ESR lab:

Frequency (MW) range: 10 GHz – 950 GHz (0.04 meV – 3.9 meV)

Magnetic field: up to 16 T

Temperature: T = 2K - 300K

ESR resonance field: ordered state



coordinate system rotating with Larmor frequency

Landau-Lifshitz equation

$$\frac{dM}{dt} = -|\gamma|(M \times H_{eff})$$

Isotropic ferromagnet:

$H_{eff} = H_{ext}$

$2\pi\nu = \omega = -|\gamma| H_{ext}$

Larmor frequency

With anisotropy:

$H_{eff} = H_{ext} + H_a$

H_a is the total anisotropy field

It causes the shift of the line from the position given by the g factor.

Analytical expressions of the spin-wave energies for a uniaxial ferromagnet:

(a) For easy-axis FM:

$\mathbf{H} \parallel c$: $h\nu = g\mu_B\mu_0(H + |H_a|)$

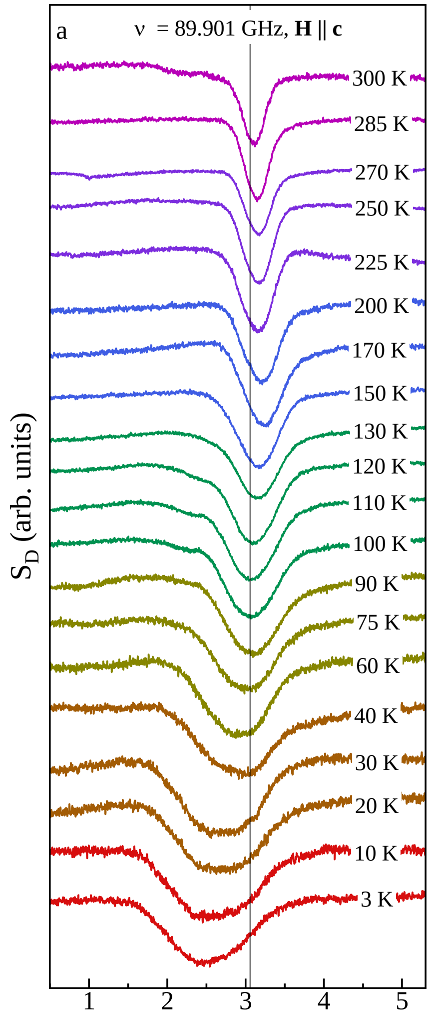
$\mathbf{H} \parallel ab$: $h\nu = g\mu_B\mu_0\sqrt{H(H - |H_a|)}$

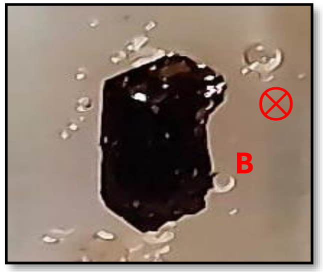
(b) For easy-plane FM:

$\mathbf{H} \parallel c$: $h\nu = g\mu_B\mu_0(H - |H_a|)$

$\mathbf{H} \parallel ab$: $h\nu = g\mu_B\mu_0\sqrt{H(H + |H_a|)}$

The temperature evolution of the ESR spectra: out-of-plane

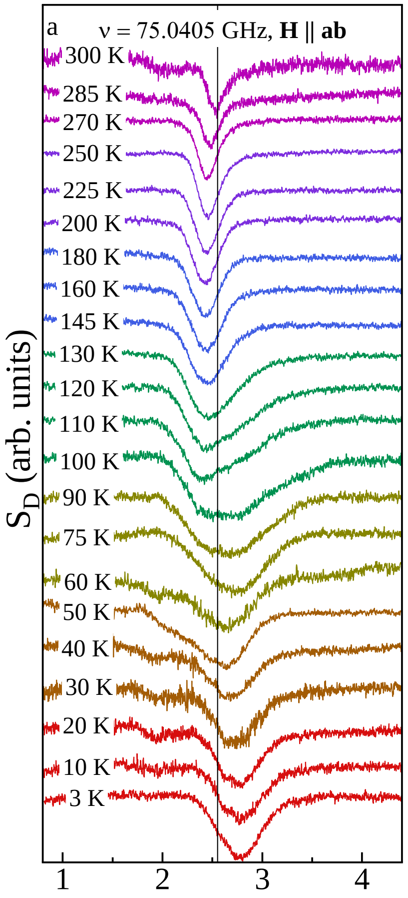


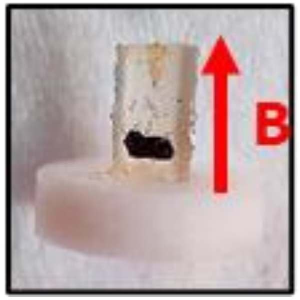


Observations:

- In the ordered state: ESR line is shifted to the left from the expected PM line (anisotropy).
- Line is shifting towards right with increasing T.
- Crosses the PM line near T_{SR}, reaches max^{m} at 200K.
- Does not reach to the PM line even at $T>T_{C}$.

The temperature evolution of the ESR spectra: in-plane

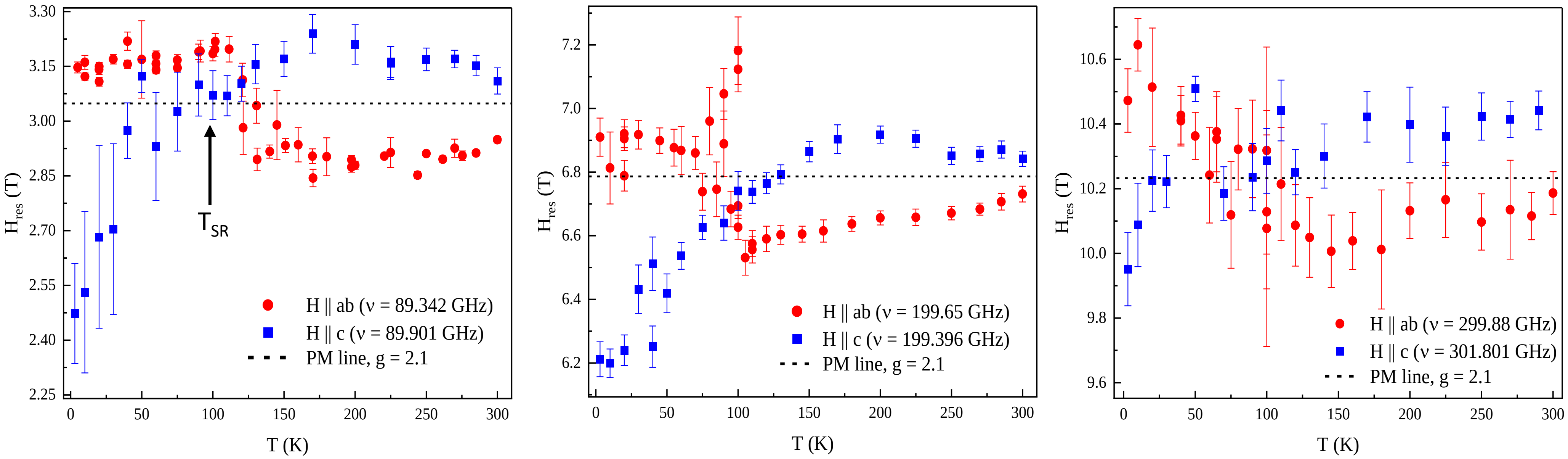




Observations:

- Situation is completely opposite compared to OP
- Line shape is also changing from low T to 110K
- Does not reaches to the PM line even at $T>T_C$

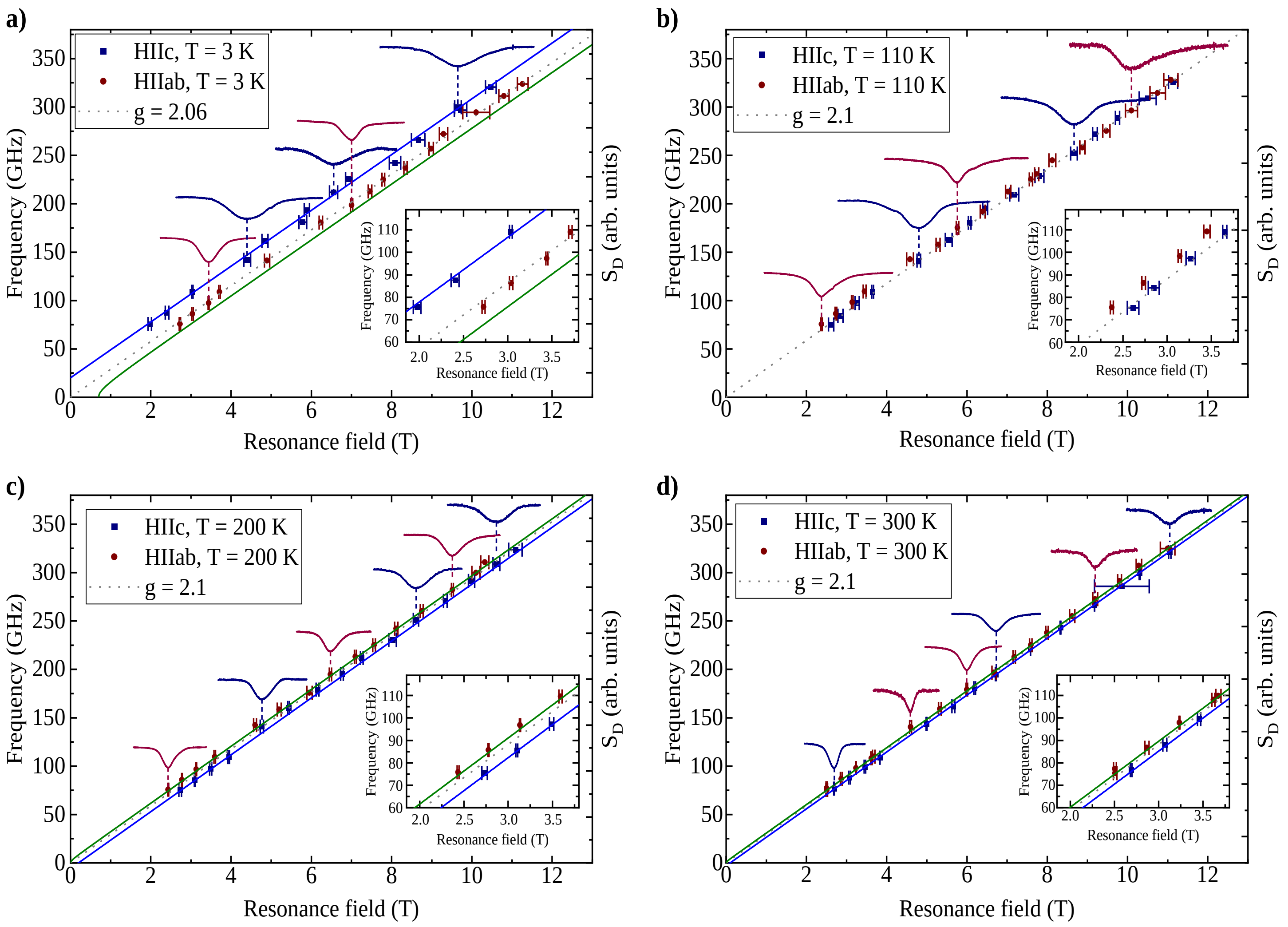
H_{res} vs. T Plot (both field orientations)



Observations:

- ESR line/ H_{res} is changing its position w.r.to the PM line, for both field orientations
- There is a change of the type of anisotropy near T_{SR}
- Above T_C, at 300 K, H_{res} does not reach the expected PM line position.

Frequency dependence of the resonance field



Spin-wave energy expressions

(a) For easy-axis FM:

$\mathbf{H} \parallel c$: $h\nu = g\mu_B\mu_0(H + |H_a|)$

$\mathbf{H} \parallel ab$: $h\nu = g\mu_B\mu_0\sqrt{H(H - |H_a|)}$

(b) For easy-plane FM:

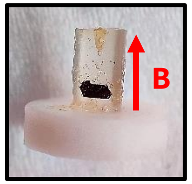
$\mathbf{H} \parallel c$: $h\nu = g\mu_B\mu_0(H - |H_a|)$

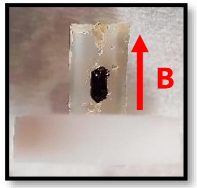
$\mathbf{H} \parallel ab$: $h\nu = g\mu_B\mu_0\sqrt{H(H + |H_a|)}$

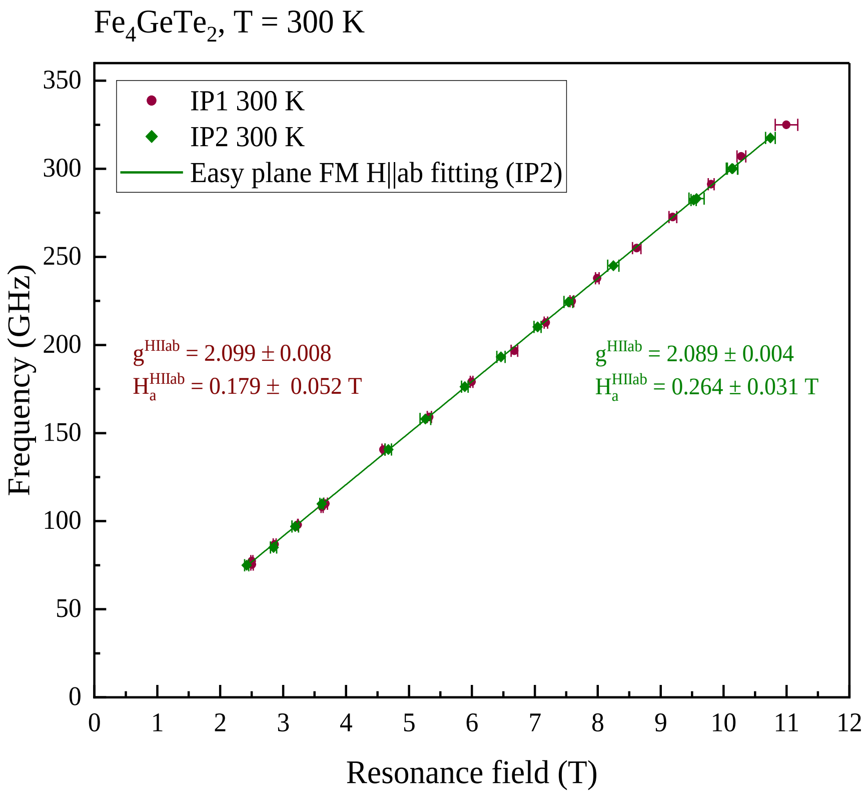
Observations:

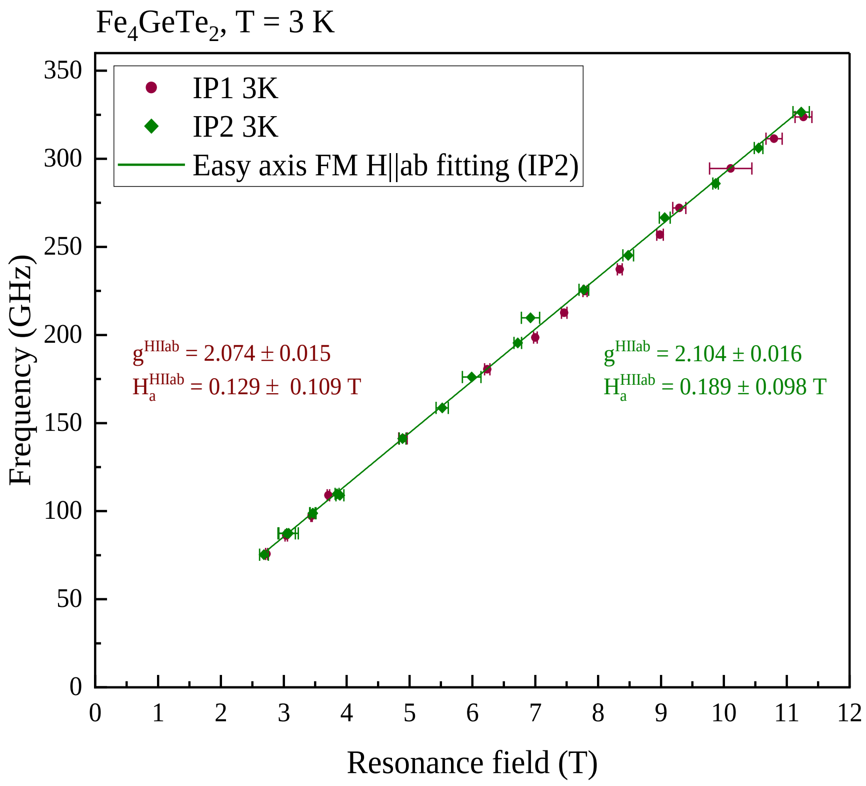
- g factor is about 2.1 almost everywhere except below 70 K.
- We can fit the data by the **easy-plane FM eq^n** above 110K.
- At 110K, almost isotropic behavior.
- At 3K, for H||c, well fitted by **easy-axis FM** eq^n.
- for H||ab, as well, but the anisotropy is reduced, as the shift is smaller.

In-plane anisotropy









Observations: No in-plane anisotropy within the error bar.

Summary: Temperature dependent anisotropy!

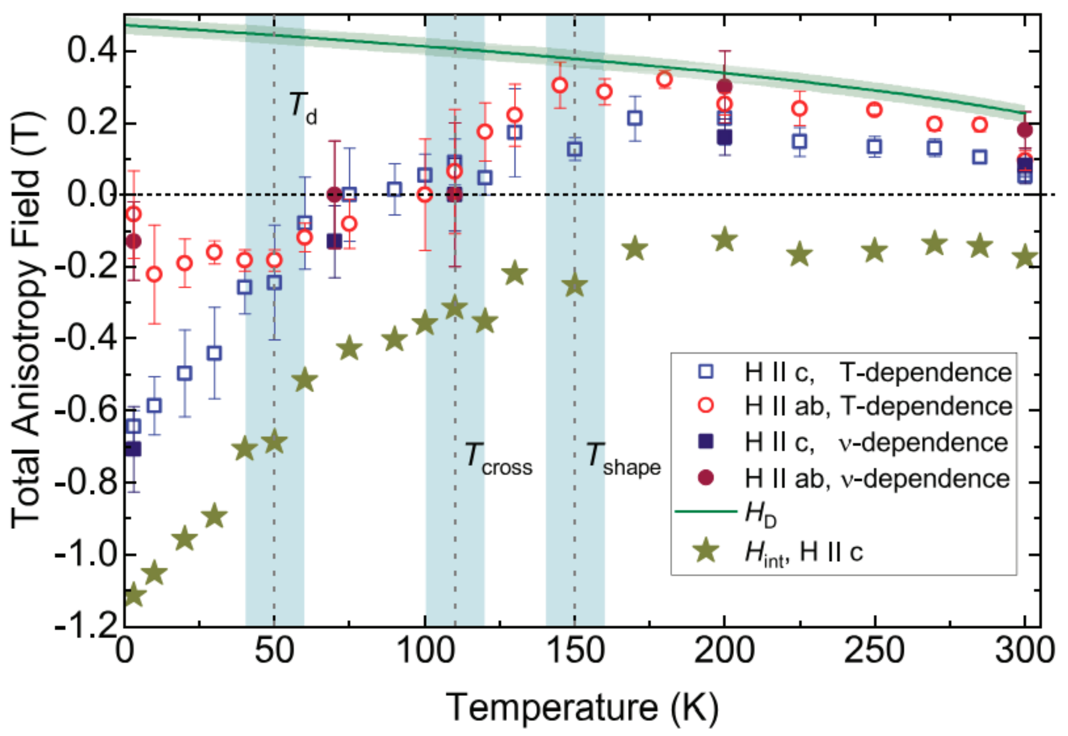
Total anisotropy field

$$H_a = H_U + H_D$$

Magneto-crystalline anisotropy
(SOC, dipole-dipole int. - Intrinsic property, due to the crystal structure)

Shape anisotropy
(Demagnetizing Field, H_d) Stray field, self de-magnetization. (Due to the sample shape)

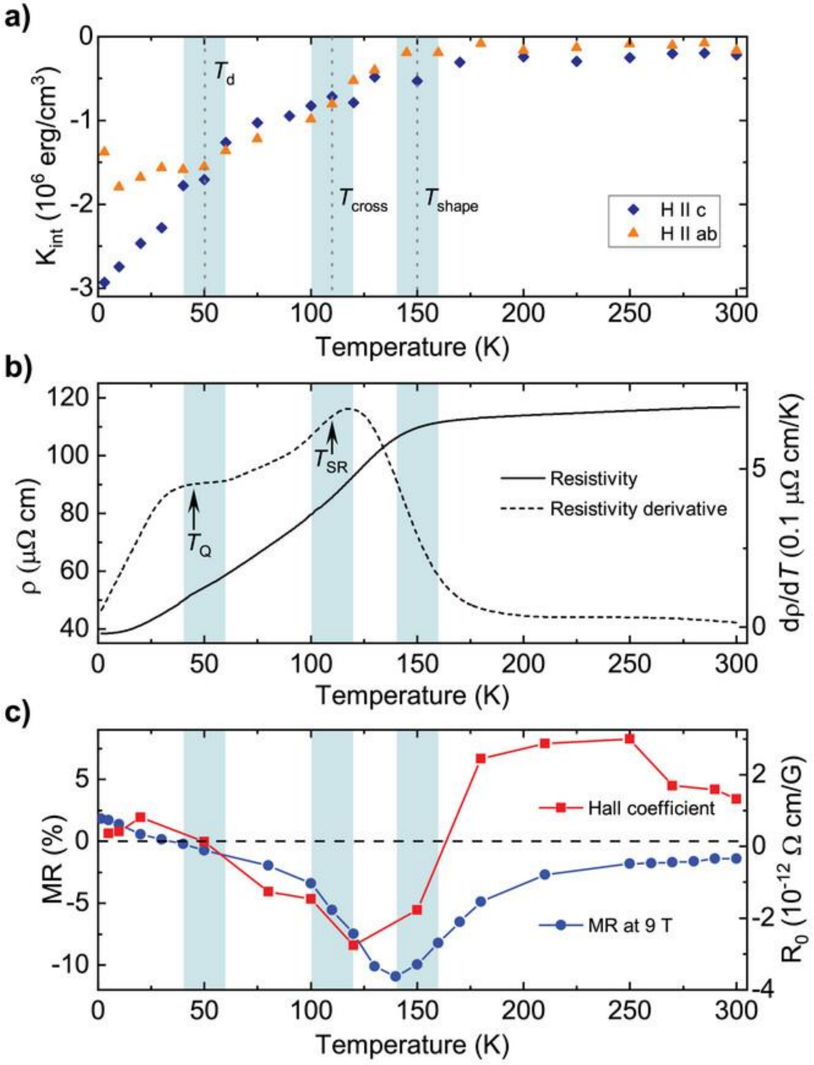
❖Shape anisotropy extracted from M_s-T plot
($H_D = 4\pi M_s$) *(Considering 2D limit, H_D maximum)*

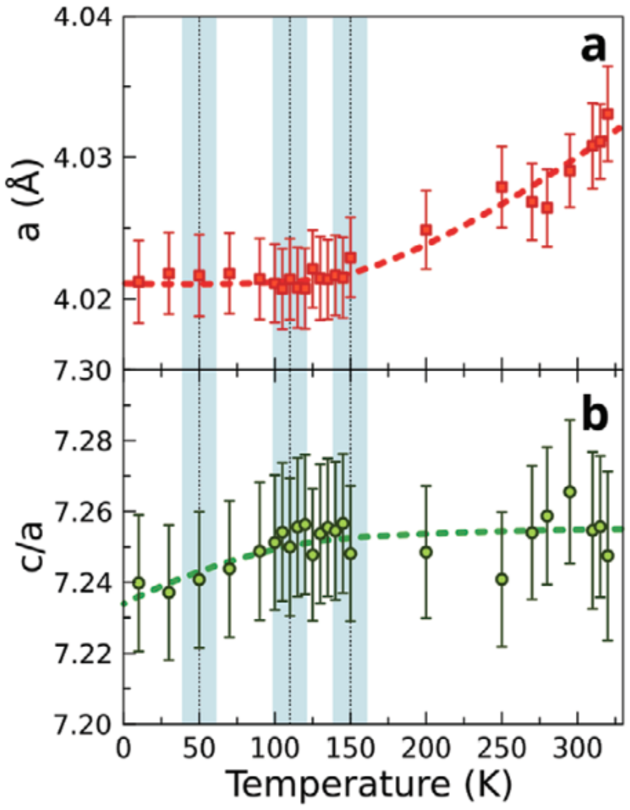


Observations

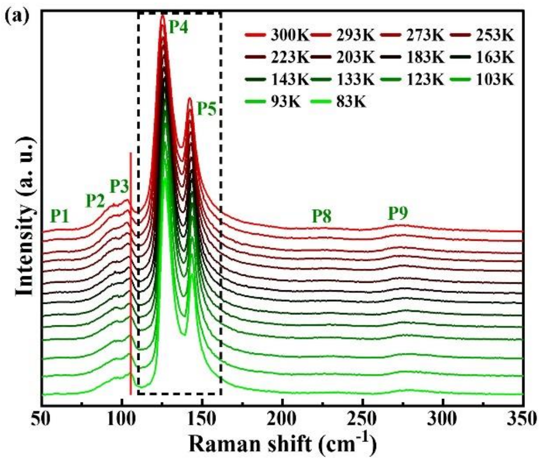
- Main contribution above 150 K is due to the **shape anisotropy.** *(Standard easy-plane FM above 150K, H_a is positive)*
- H_a ~ 0 near T_{SR}
- Below T_{SR}, easy-axis type of anisotropy, *H_a is negative*.
- Below 50K, H_a is dependent on the field orientations (contradicts standard SW equations).

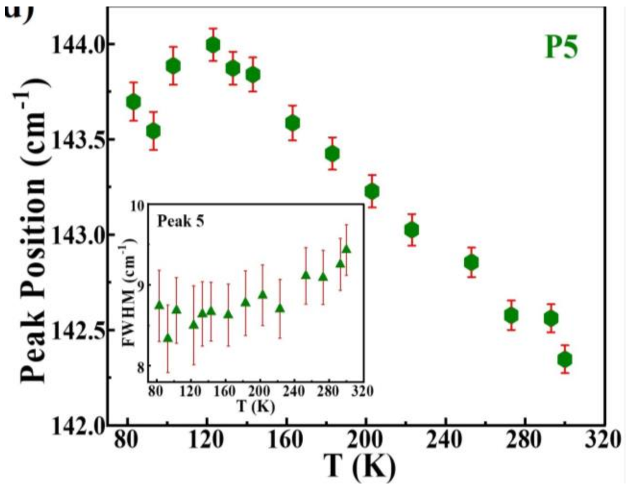
Summary: Temperature dependent anisotropy!





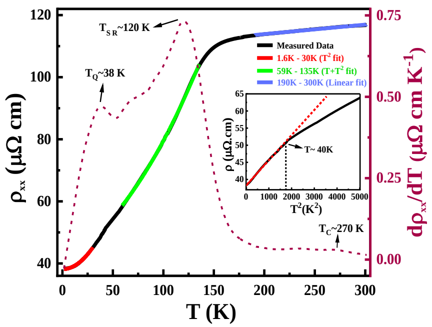
XRD: IFW Dresden



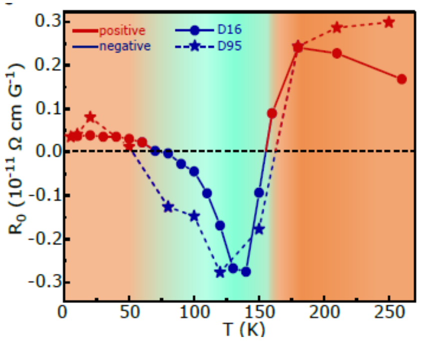


Raman: Achintya Singha, Bose Institute

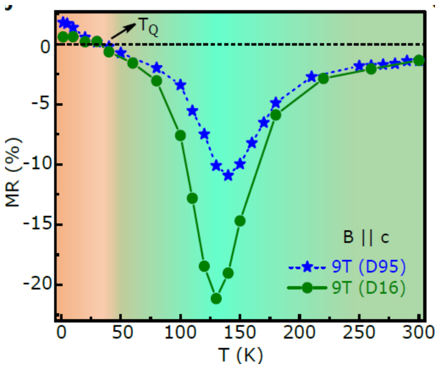
Take home message



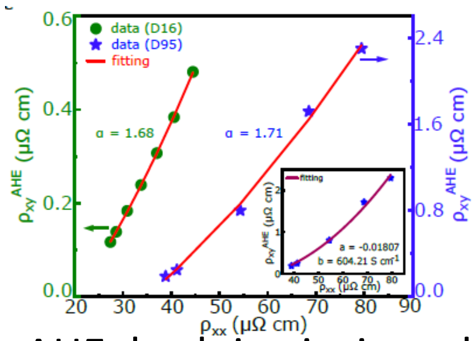
Resistivity: scattering process



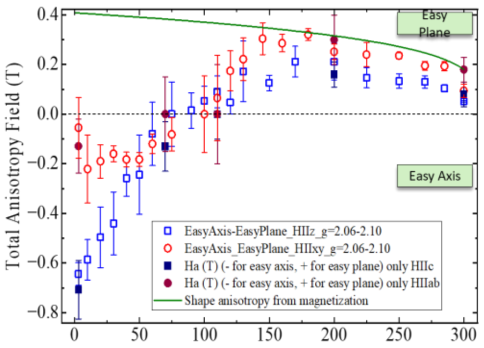
Ordinary Hall: Fermi surface reconstruction



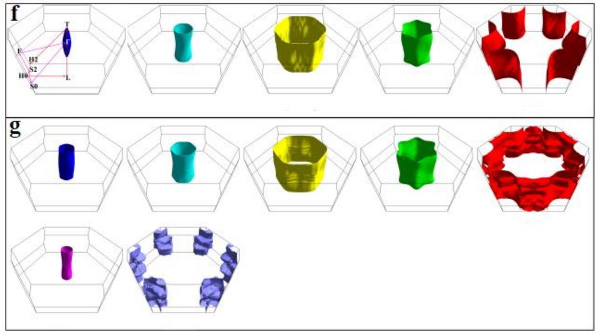
Magnetoresistance



AHE: both intrinsic and extrinsic origin



ESR: change of competing anisotropy



DFT: collinear and non-colinear phases

- Unusual transport features due to spin reorientation
- Low temperature phases below 40K

Riju Pal, …., ANP, **npj 2D Mater Appl** 8, 30 (2024).

Riju Pal, ..ANP…Alexey Alfonsov, **Advanced Functional Materials, 2402551,** (2024)

Thank you