Dimension-Dependent Critical Scaling Analysis and Emergent Interaction Scales in a 2D Van der Waals magnets

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https://sites.google.com/view/molabiitk

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Suprotim Saha et al (submitted 2024)

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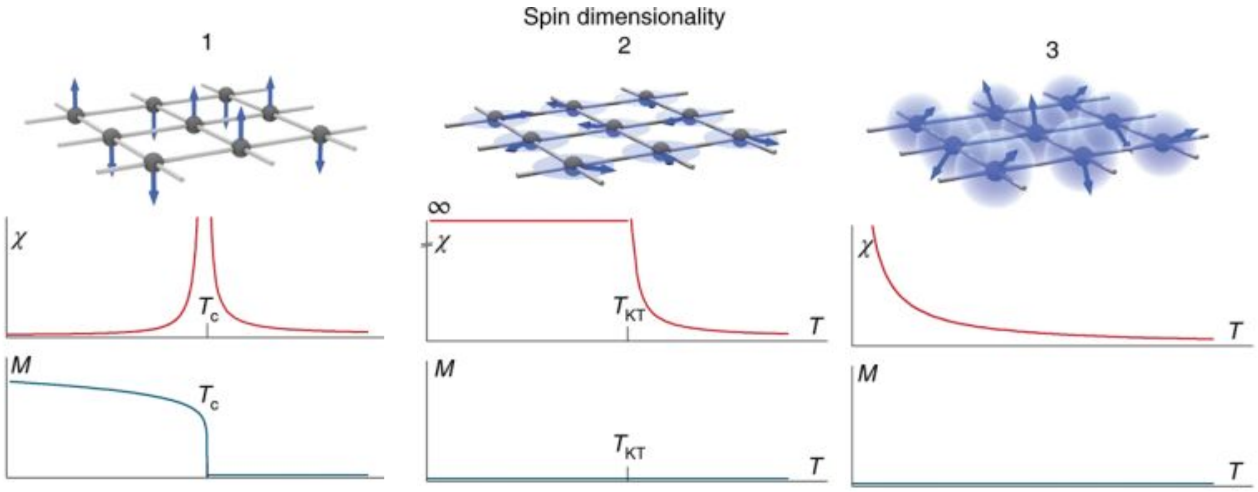
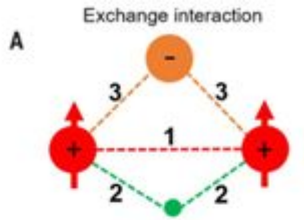
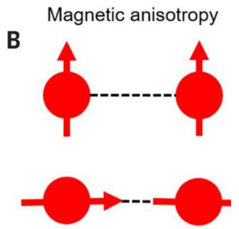
Magnetism at low Dimensions

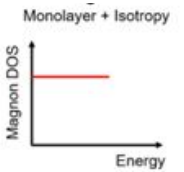


Xu *et al. Microstructures* 2022;2:2022011

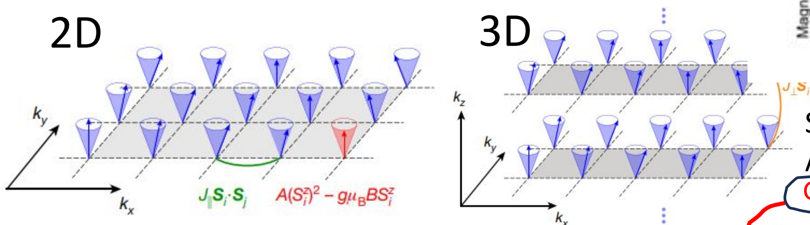




Through SOC,
Shape Anisotropy
Stress Anisotropy

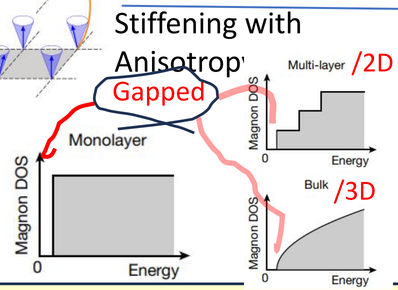


Gapless Magnon Excitation (no Anisotropy) Expected in monolayer



Exchange vs Anisotropy: Spin wave excitation
In 2D and 3D, role of anisotropy

Stiffening with Anisotropy



- **Gapless excitation of long λ spin waves destroys Long magnetic order**

- Magnetic anisotropy creates a Gap in the magnon DOS, which Prevents thermal excitation of Spin waves and Long-range order Can be preserved even in 2D

- Magnetic order can emerge even in 2D systems with magnetic anisotropy

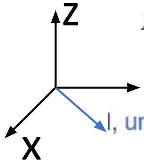
Gong and Zhang, Science363, eaav4450 (2019)
Nature **volume 546**, 265–269 (2017)

2D-Ising model n=1

$$H(\sigma) = -\sum_{\langle ij \rangle} J_{ij}\sigma_i\sigma_j$$

PM → Magnetically ordered state transition, with diverging Susceptibility, Specific heat, divergence spin – spin Correlation lengths

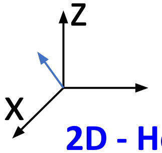
n=2, 2D- xy model



$$H(\mathbf{s}) = -\sum_{i \neq j} J_{ij}\,\mathbf{s}_i \cdot \mathbf{s}_j$$

unit-length vector $\mathbf{s}_j = (\cos\theta_j, \sin\theta_j)$

In 2D BKT transition, with quasi long range magnetic order, power law decay Of spin – spin correlation



$\vec{s}_i \in \mathbb{R}^3, |\vec{s}_i| = 1$

$$\mathcal{H} = -\sum_{i,j} \mathcal{J}_{ij}\vec{s}_i \cdot \vec{s}_j$$

2D - Heisenberg Model Cannot sustain Long range Magnetic order Mermin Wagner theorem

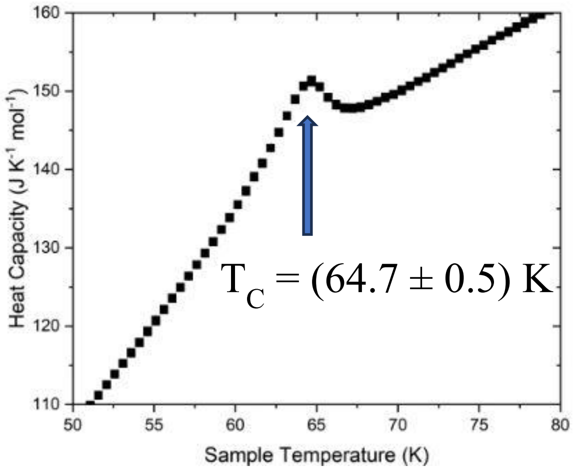
For engineering devices : 1. Material choice

Why we choose to work with a 2D vdw material Cr2Ge2Te6

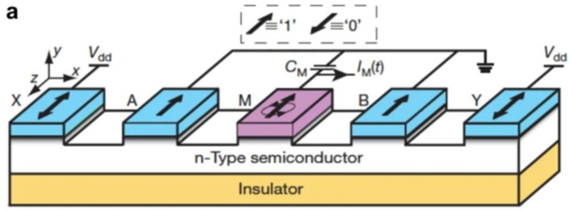
$Cr_2Ge_2Te_6$ (CGT) : FM semiconductor and vdW material

(Bulk properties)

PHYSICAL REVIEW B 100, 134437 (2019)



One possibly can make 2D spintronic devices



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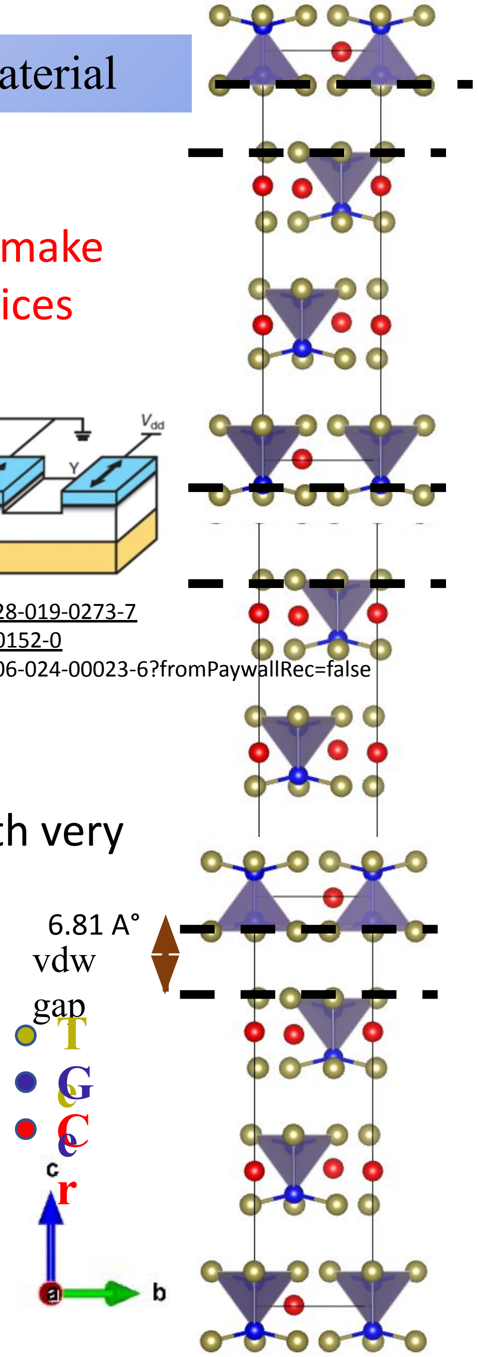
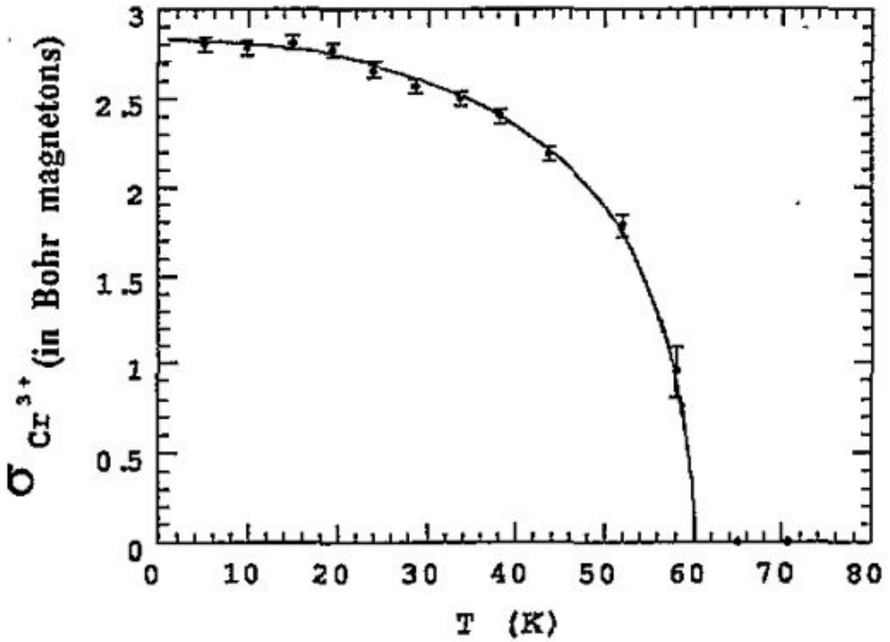
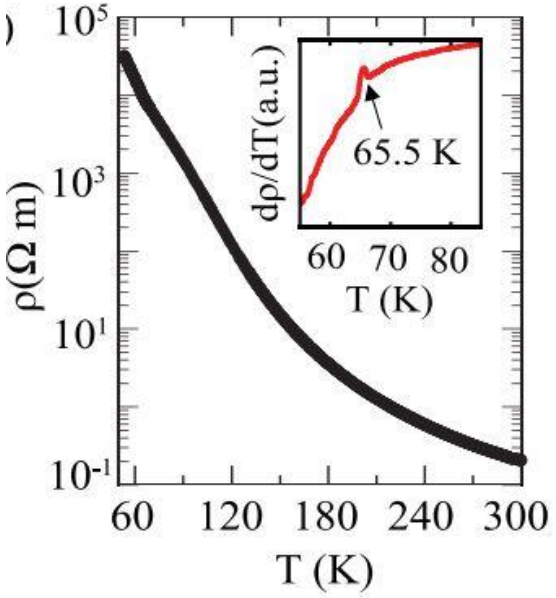




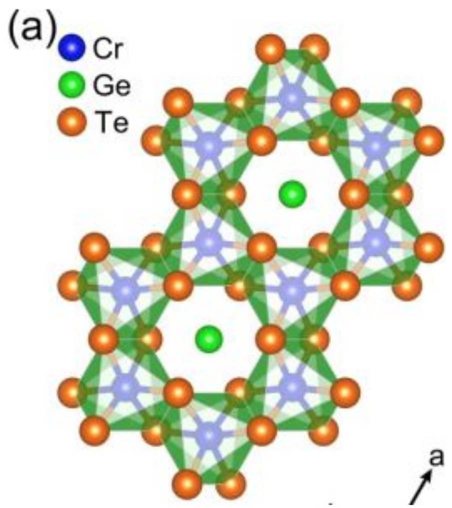
V. Carteaux et al.
JPCM 7, 69−87 (1995)



Adv. Mater. **2021**, *33*, 2008586

VDW material with very Weak interlayer coupling

$$\rho = \rho_0 \exp\left(\frac{E_g}{2k_BT}\right)$$



ACS Appl. Electron. Mater. 20
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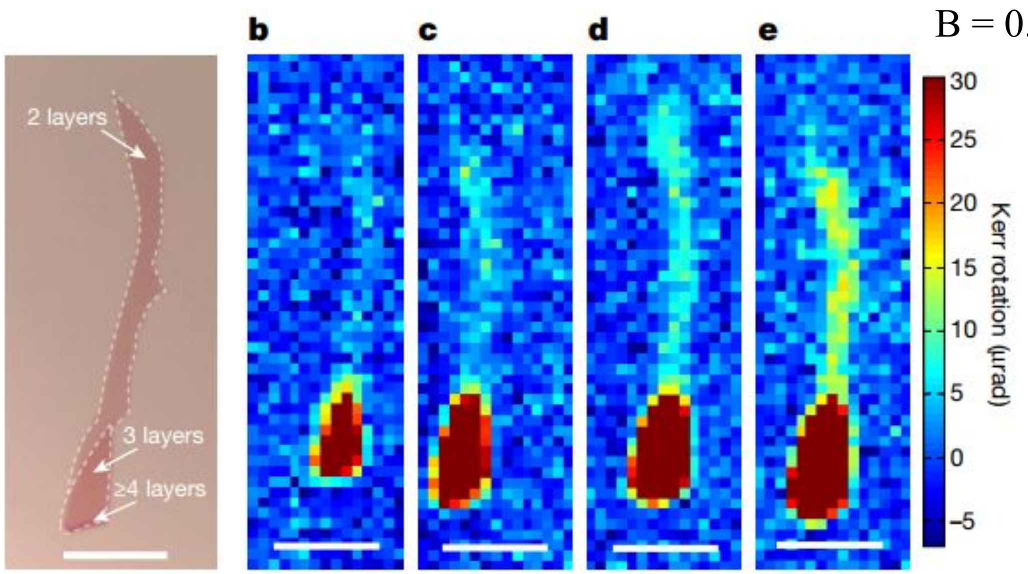
Bandgap ~ 0.4 to 0.7 eV indirect, and 1.2 eV direct (PRB 98 (12), 125127 (2018))

What happens if you thin down the system to few layers

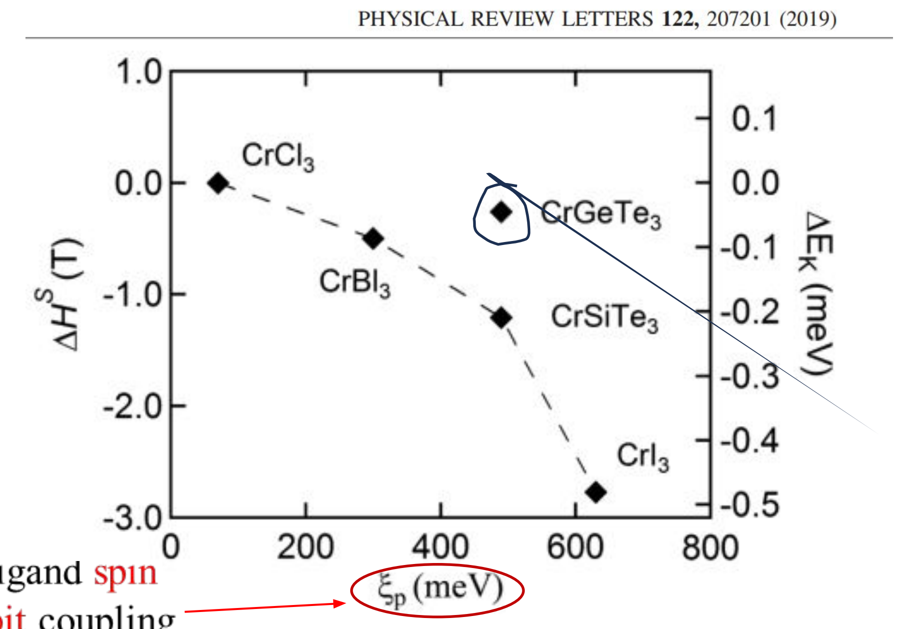


Scale: 10μm

Nature **volume 546**, 265–269 (2017)

FM Retained

(a critical transition in still seen to persist in few layer CGT)



Ligand spin orbit coupling strength $(\xi_p L S)$

$$\Delta H^S = H_c^S - H_{ab}^S$$

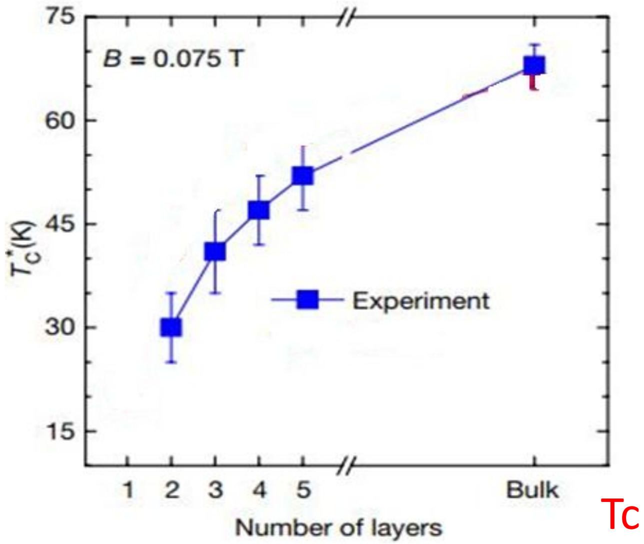
$$\Delta E_K = 3\mu_B \Delta H^S$$

Ligand p-Spin orbit coupling controls

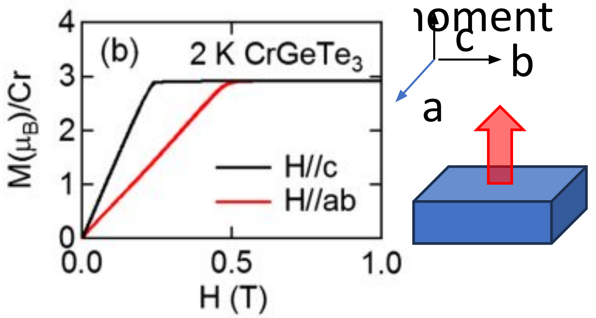
Anisotropy associated something else also plays a role in CGT for

reduced anisotropy fields and energy i

Cr^{+3} moment



Tc seems to drop



Magnetic Anisotropy

For engineering devices : One peels the system down to 2D layers

How do you quantitatively evaluate that things remain/do not remain the same as you go down to 2D layers

Magnetism

Thickness reduction from 3D

In 2D
Significant enhancement in the effects
Of thermal fluctuations

Complex magnetic phenomena at low D
Dependent on the Dimensionality of spins (n)

They were pages in book form
Should remain as pages when torn out?

Critical exponents around a 2nd Order phase transition point

Reduced Temp : $t = (T - T_c)/T_c$

Specific heat exponent : $C \sim t^{-\alpha}$

Order Parameter exponent $< |\phi| > \sim t^{\beta}$

Correlation length exponent: $\xi \sim t^{-\nu}$

Susceptibility exponent: $\chi \sim t^{-\gamma}$

Can be used to quantify which properties remain Unchanged as you thin down the sample.

Universality Classes

Table 5.4.2. *Some critical exponents from theory and experiment.*

Exponent	α	β	γ	ν	η
Property	specific heat	order parameter	susceptibility	coherence length	correlation function
Definition	$C \sim t^{-\alpha}$	$\langle\phi\rangle \sim t^{\beta}$	$\chi \sim t^{-\gamma}$	$\xi \sim t^{-\nu}$	$G(q) \sim q^{-2+\eta}$
Mean-field	0	0.5	1	0.5	0
3D theory					
$n = 0$ (SAW)	0.24	0.30	1.16	0.59	
$n = 1$ (Ising)	0.11	0.32	1.24	0.63	0.04
$n = 2$ (xy)	−0.01	0.35	1.32	0.67	0.04
$n = 3$ (Heisenberg)	−0.12	0.36	1.39	0.71	0.04
Experiment					
3D $n = 1$	$0.11^{+.01}_{-.03}$	$0.32^{+.16}_{-.04}$	$1.24^{.16}_{-.04}$	$0.63^{+.04}_{-.04}$	0.03 - 0.06
3D $n = 3$	$0.1^{+.05}_{-.04}$	$0.34^{+.04}_{-.04}$	$1.4^{+.07}_{-.07}$	$0.7^{+.03}_{-.03}$	
2D $n = 1$	$0.0^{+.01}_{-.003}$	$0.3^{+.04}_{-.04}$	$1.82^{+0.7}_{-.07}$	$1.02^{+.07}_{-.07}$	

Experiments on 3D $n = 1$ compiled from liquid-gas, binary fluid, ferromagnetic, and antiferromagnetic transitions.
Experiments on 3D $n = 3$ transitions compiled from some ferromagnetic and antiferromagnetic transitions.
Experiments on 2D $n = 1$ complied from some antiferromagnetic transitions.

From P. Chaikin and T Lubensky "Principles of Condensed Matter Physics"

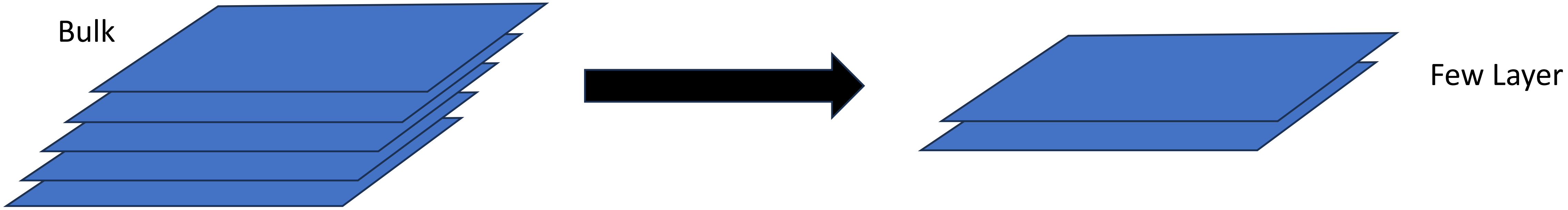
Theory suggests that the class (i.e. set of exponents) depends on spatial dimensionality, symmetry of the order parameter and interaction (and range of the latter as well) but not on the detailed form or strength of the interactions

Questions

Bulk

In CGT (2D layered bulk), is the experimentally identified Tc associated with critical phenomena?

If so what is the universality class for bulk CGT magnetic system (Ising like, or XY, …)?



Few layer thick CGT

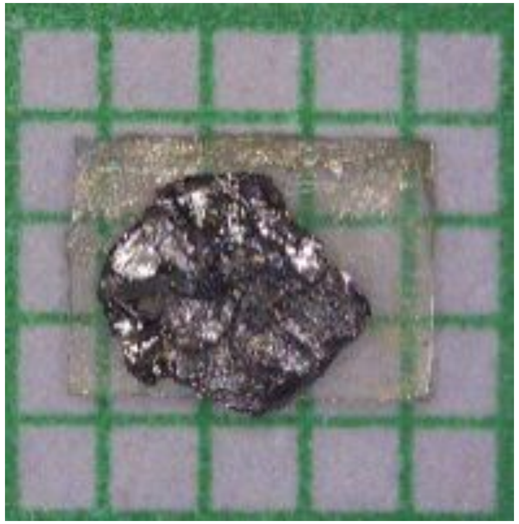
What happens to Tc as one lowers thickness ?

Lowering the thickness of the vdW material, **naively,** we expected to preserve this Universality class, since the bulk system is anyway weakly coupled, does one observe this ?

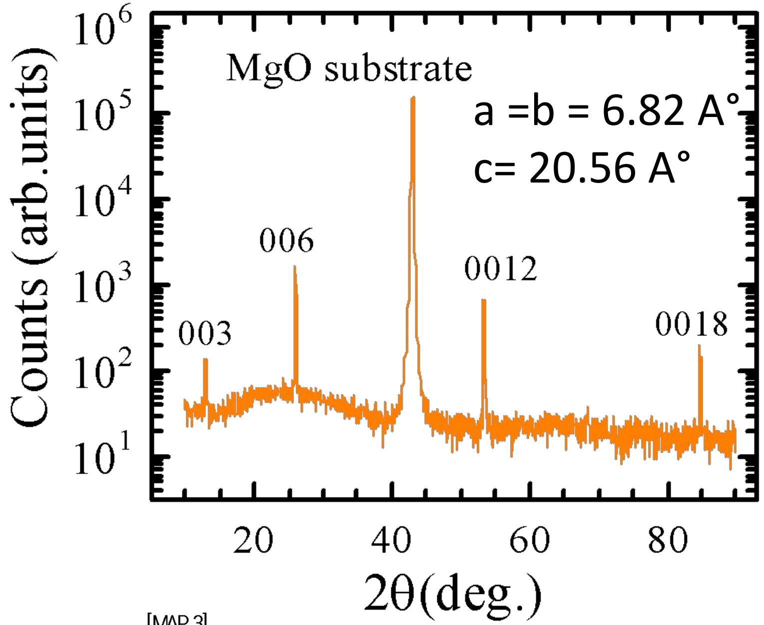
What happens to magnetic anisotropy ?

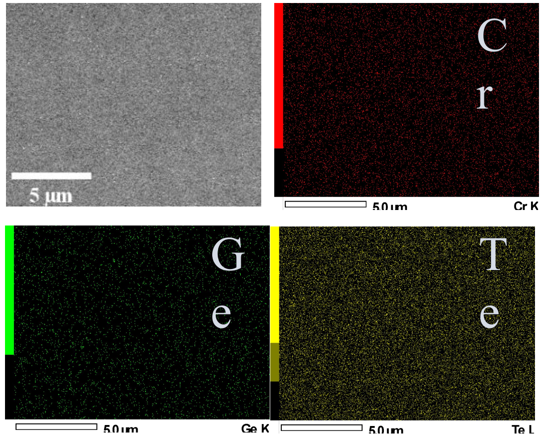
Magnetic configuration ?

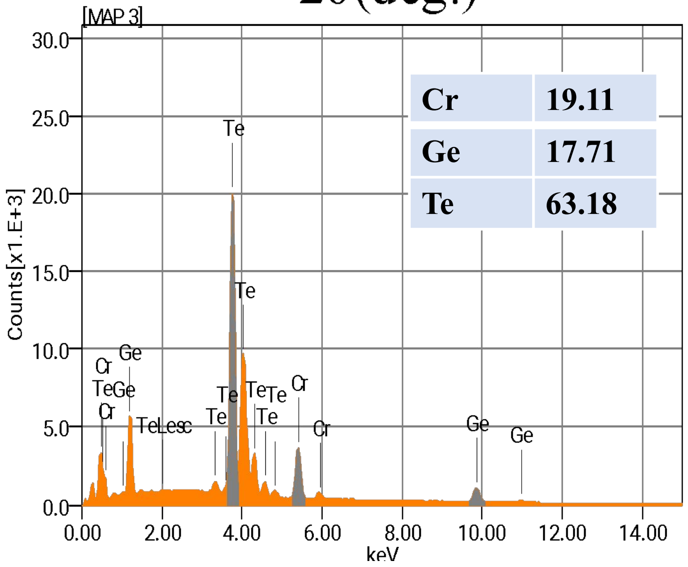
Bulk $Cr_2Ge_2Te_6$ (CGT)



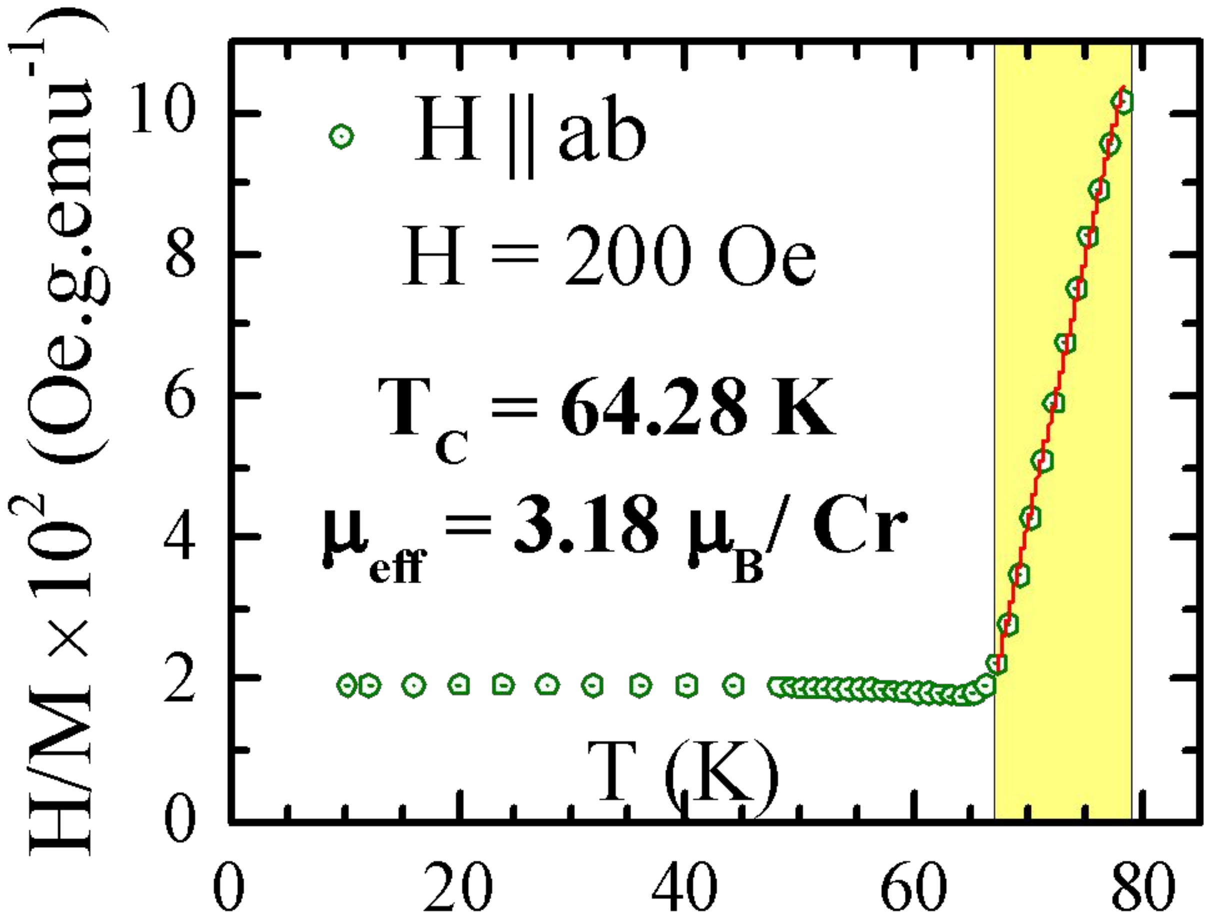
2.5 mm x 2.3 mm x 0.24 mm

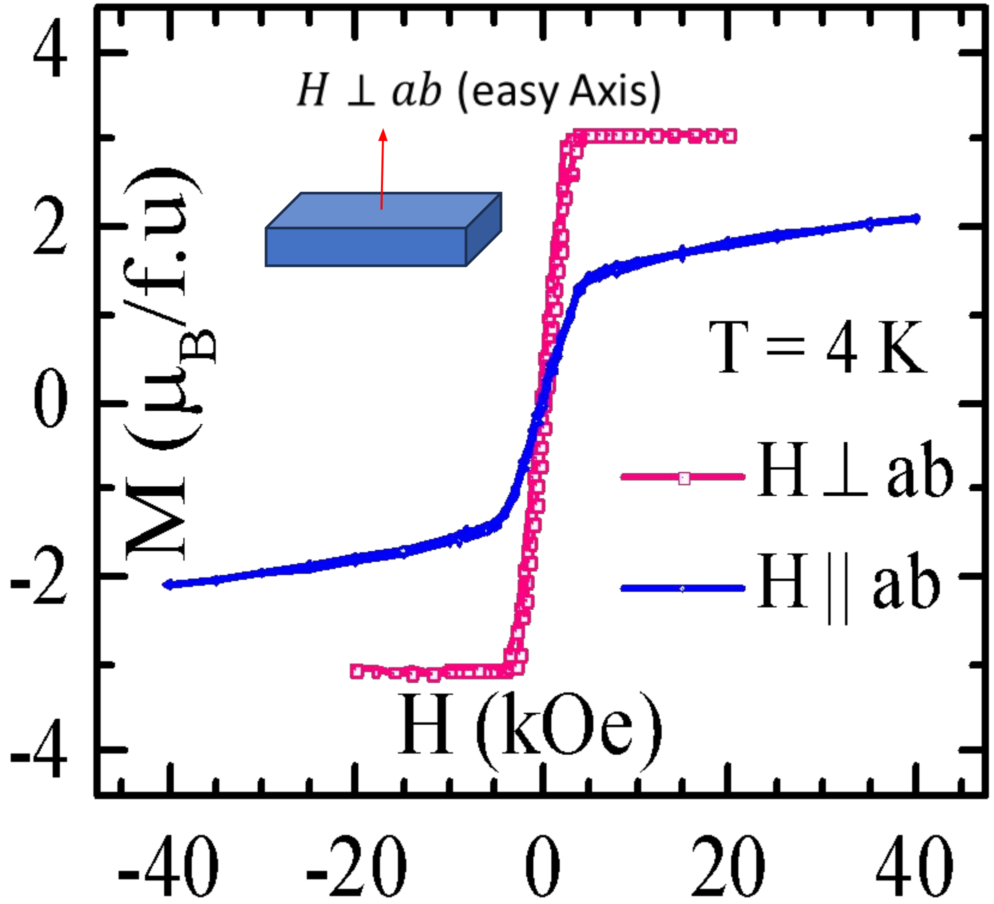




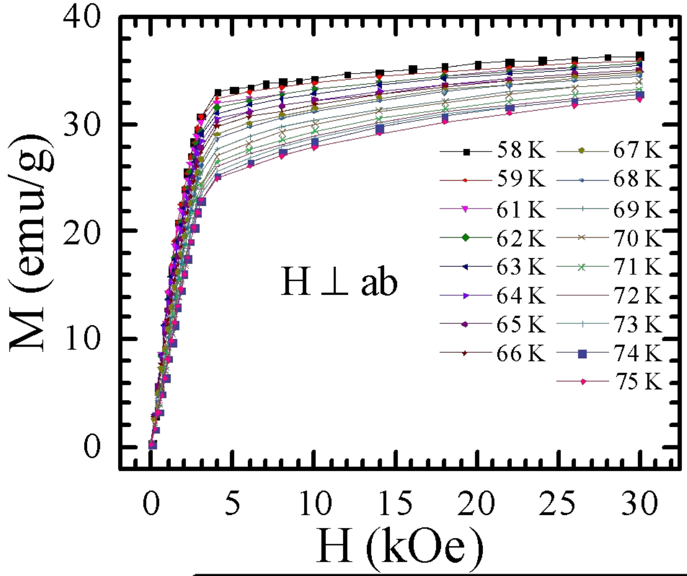


Magnetic properties of bulk CGT





Critical Scaling of M(H) for bulk CGT with 2D-Ising like spins



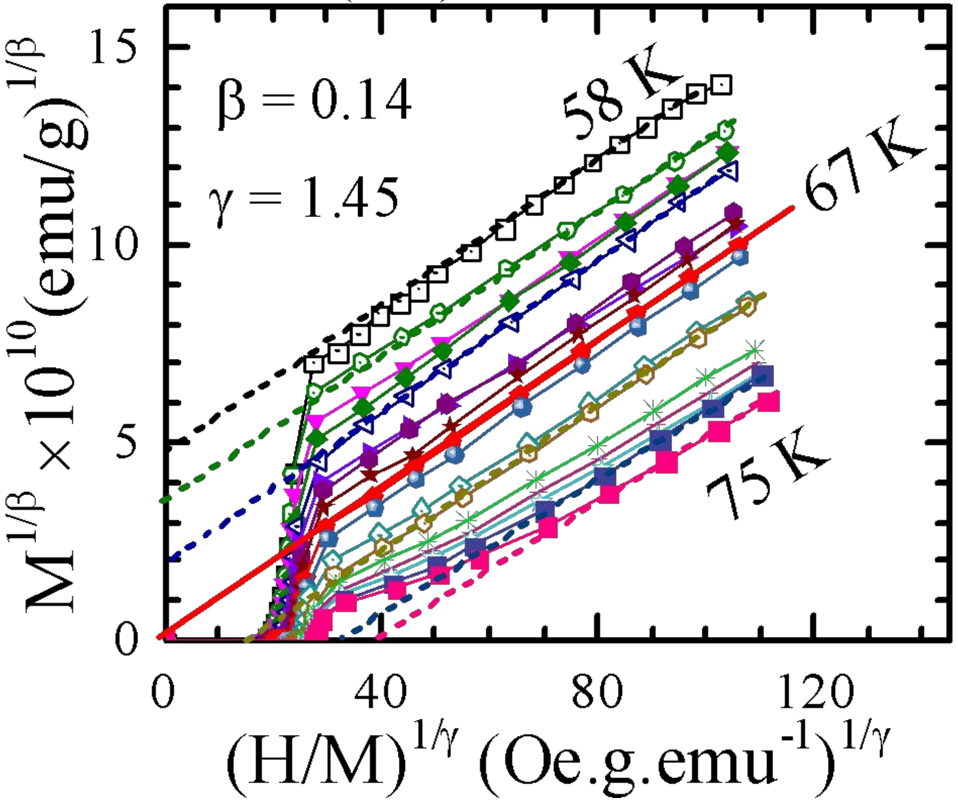
$$\varepsilon = (T - T_C)/T_C$$

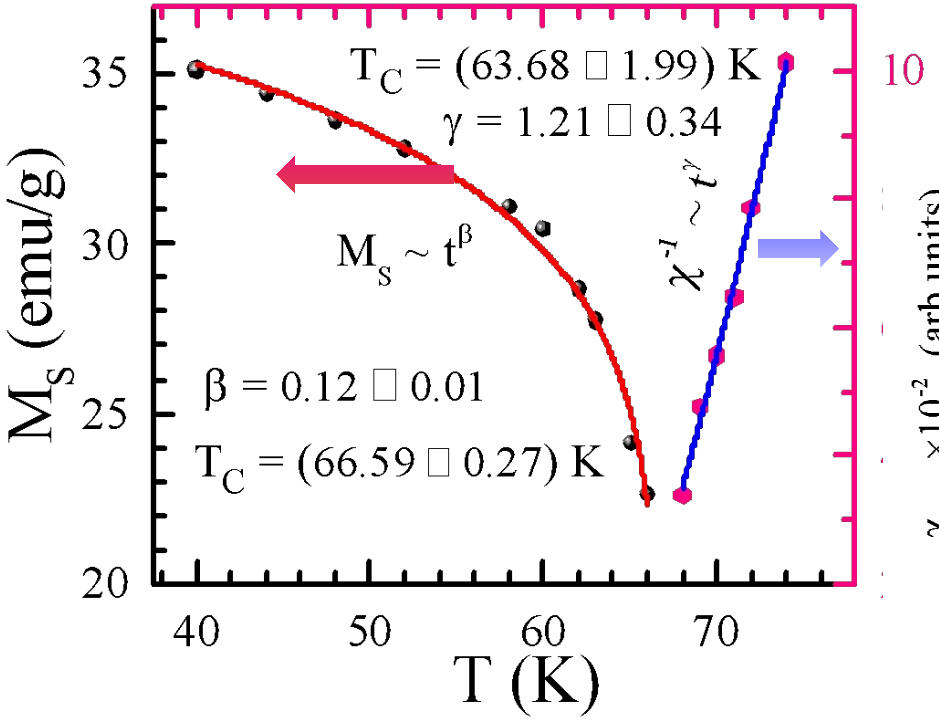
$$M_S(T) = M_0(-\varepsilon)^{\beta},\ \varepsilon < 0, T < T_C$$

$$\chi_0^{-1}(T) = (h_0/M_0)\varepsilon^{\gamma},\ \varepsilon > 0, T > T_C$$

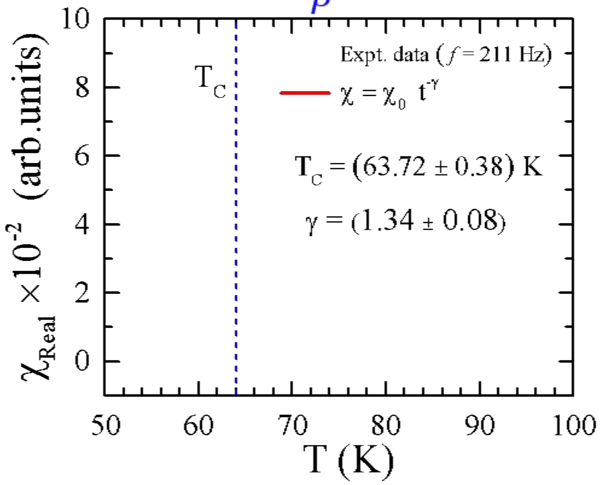
$$M = DH^{1/\delta},\ \varepsilon = 0, T = T_C$$

Equation of State
Modified Arrotts plot

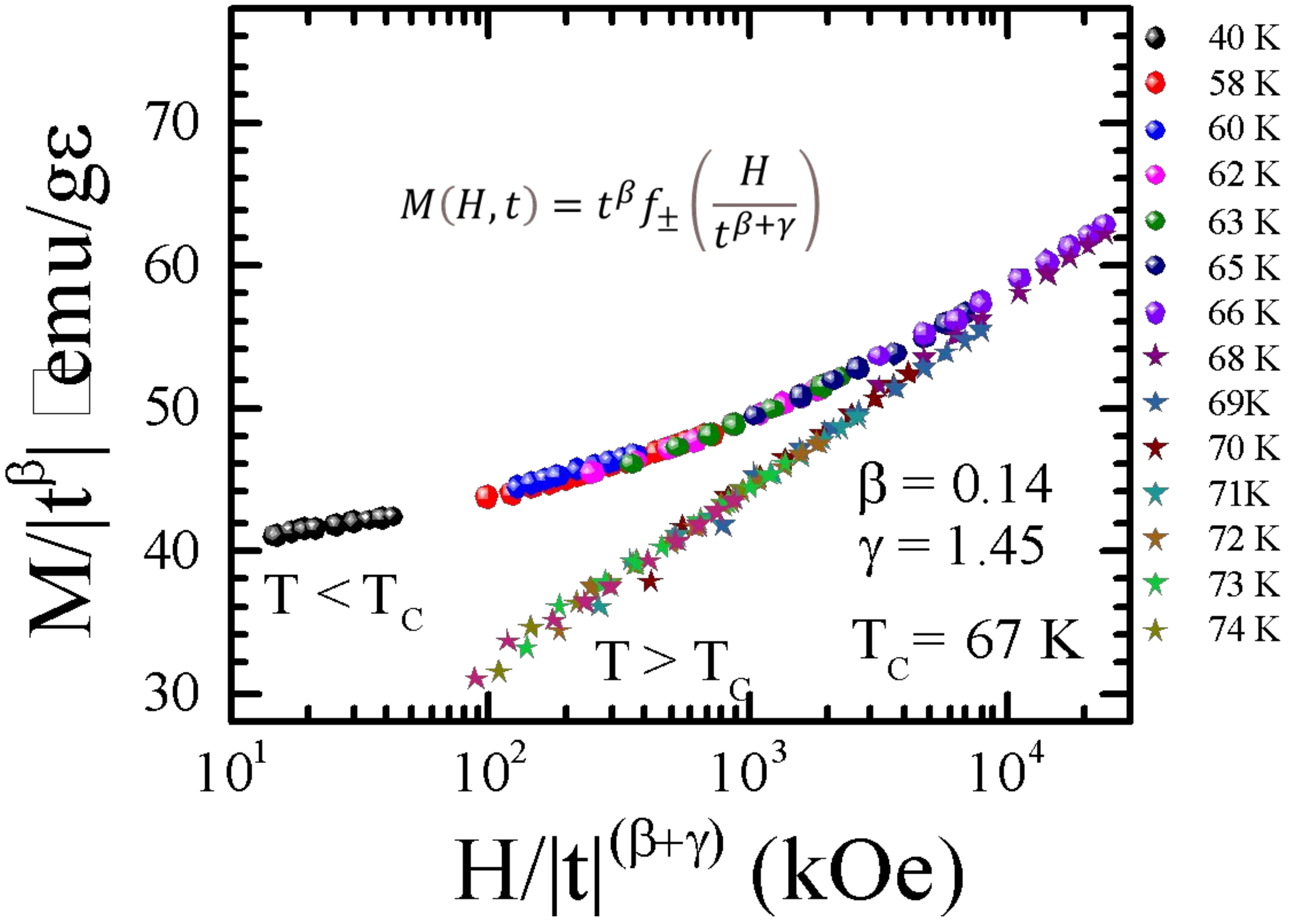


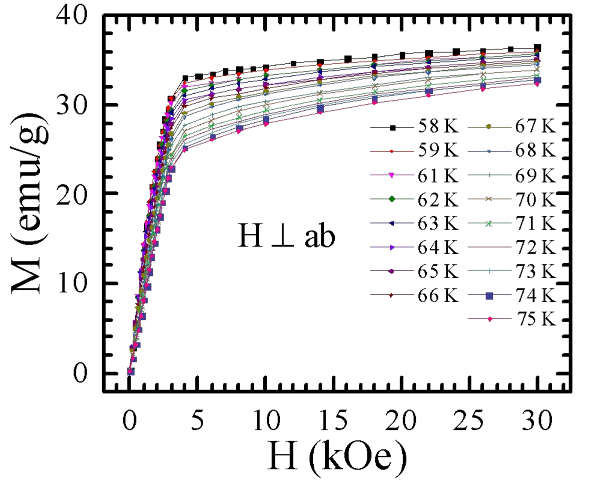


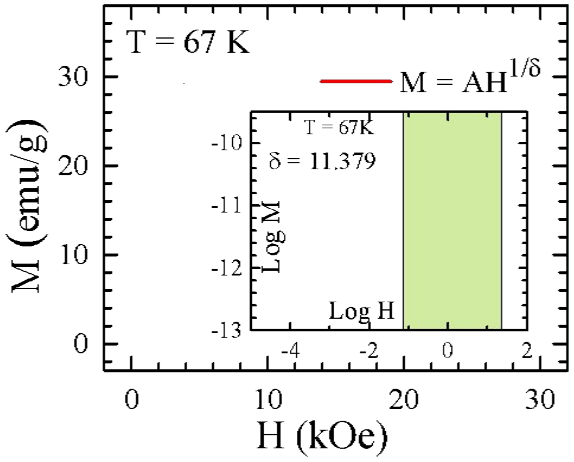
$$\delta = 1 + \frac{\gamma}{\beta} = 11.35$$



Bifurcation into two scaled curves above and below Tc

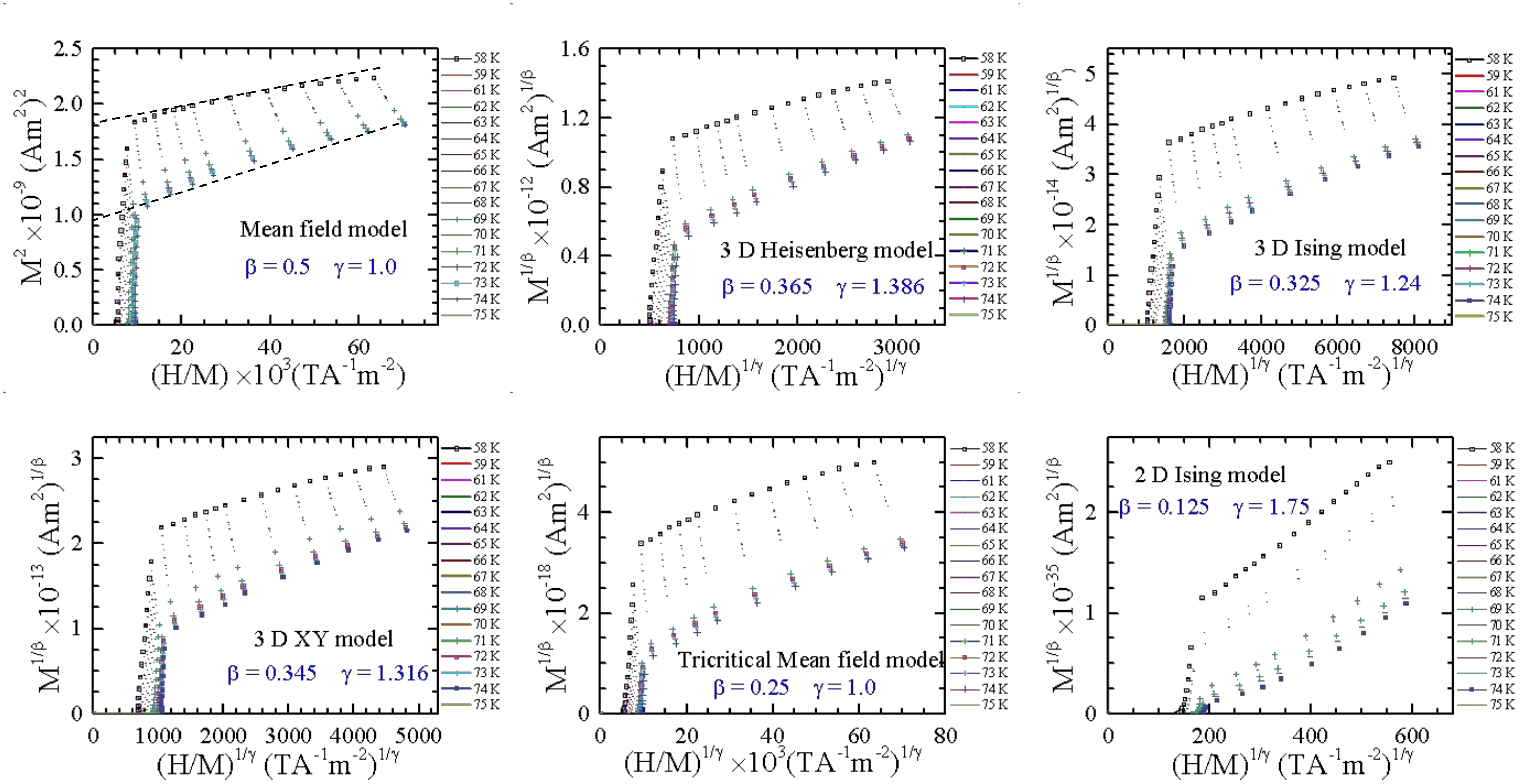






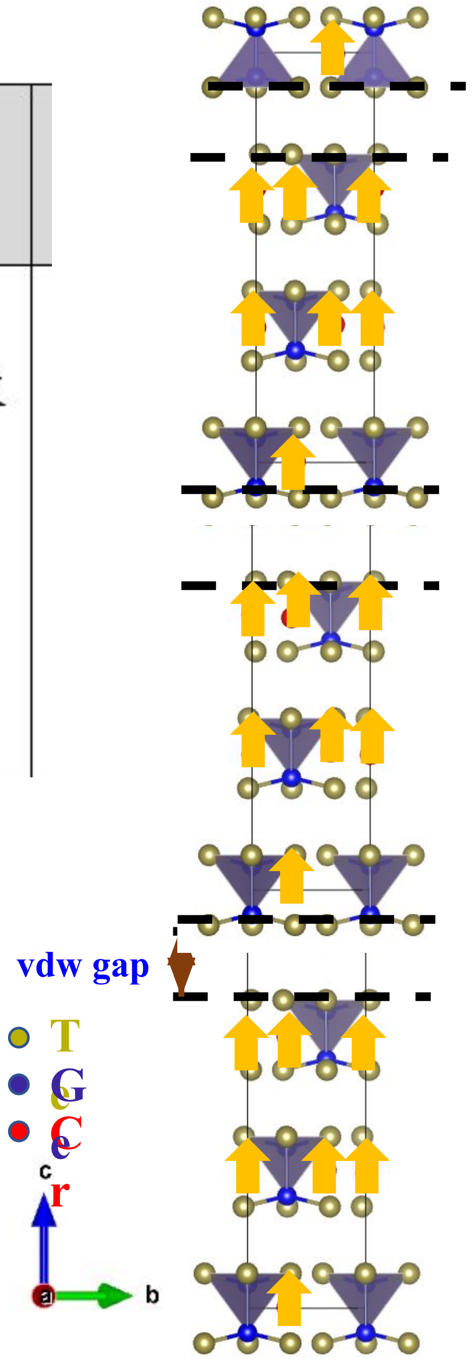
$$\delta = 1 + \frac{\gamma}{\beta} = 11.35$$

Validity of the scaling

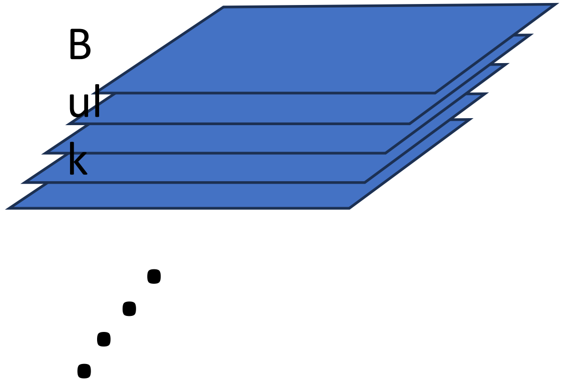


Universality Class for bulk CGT

Sample	μ_{eff}	β	β (refs.[33, 34])	γ	γ (refs.[33, 34])	δ	δ (refs.[33, 34])	T_c
CGT bulk crystal	$3.18\mu_B$ per Cr	0.14 (Fig.2(b)) 0.12 (Fig.2(d))	2d-Ising 0.17 to 0.2	1.45 (Fig.2(b)) 1.21 (Fig.2(d))	2d-Ising 1.75 to 1.28	11.37 (Fig. S7(a)) 11.35 (Widom)	2d-Ising 10.87 to 7.96	64.28K (Fig.2(a)) 63.68K - 66.59 K (Fig.2(d)) 67 K (Fig.2(b), (c))

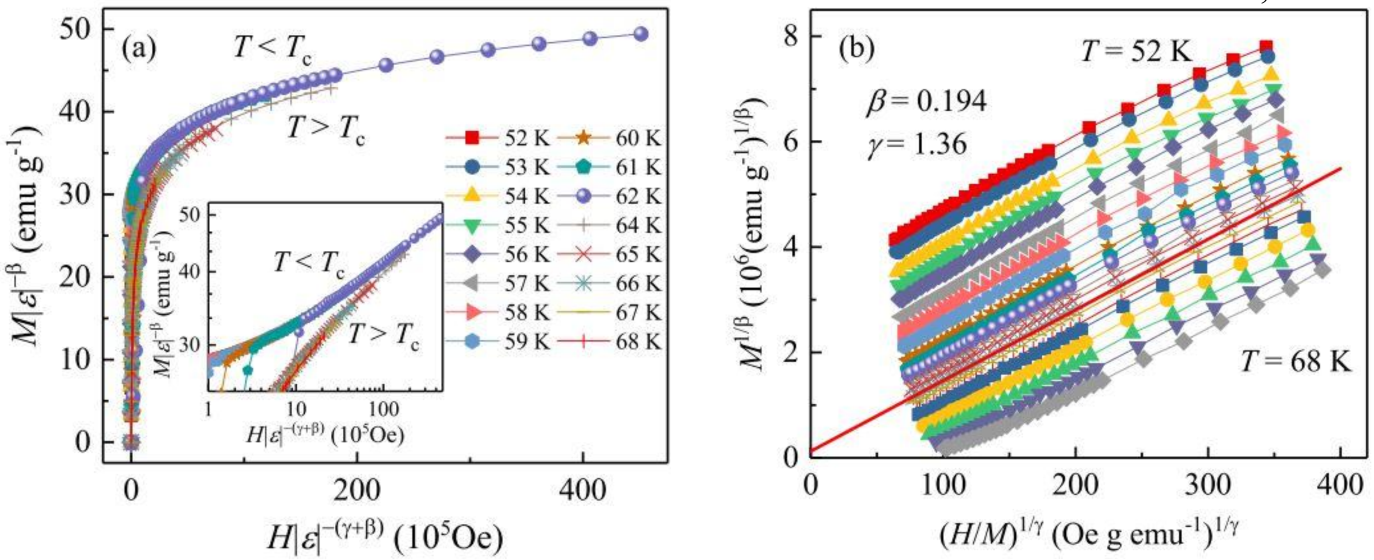


Our Analysis confirms
2D Ising universality class
For bulk CGT



Past Critical scaling study in bulk CGT

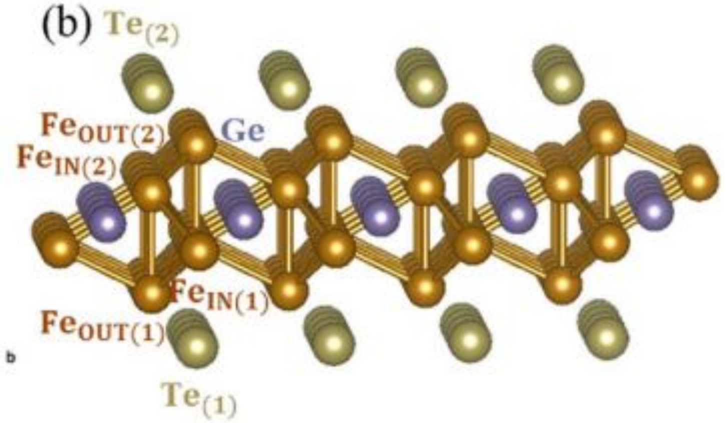
PHYSICAL REVIEW B **96**, 054406 (2017)

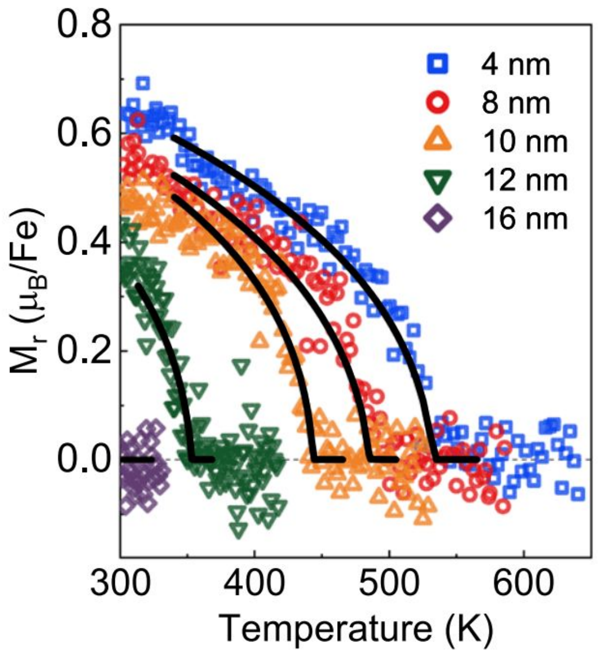


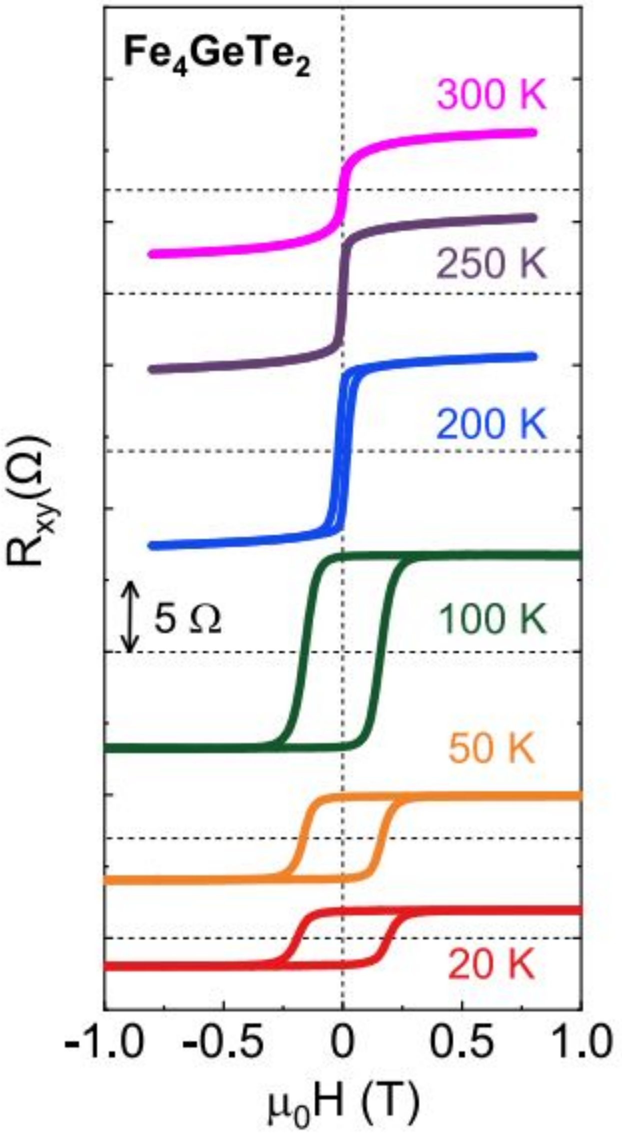
Scaling analysis for bulk CGT
<u>2D Ising universality class</u>

Is this uniform across other bulk 2D systems

Brief Introduction to Fe_4GeTe_2 (F4GT)

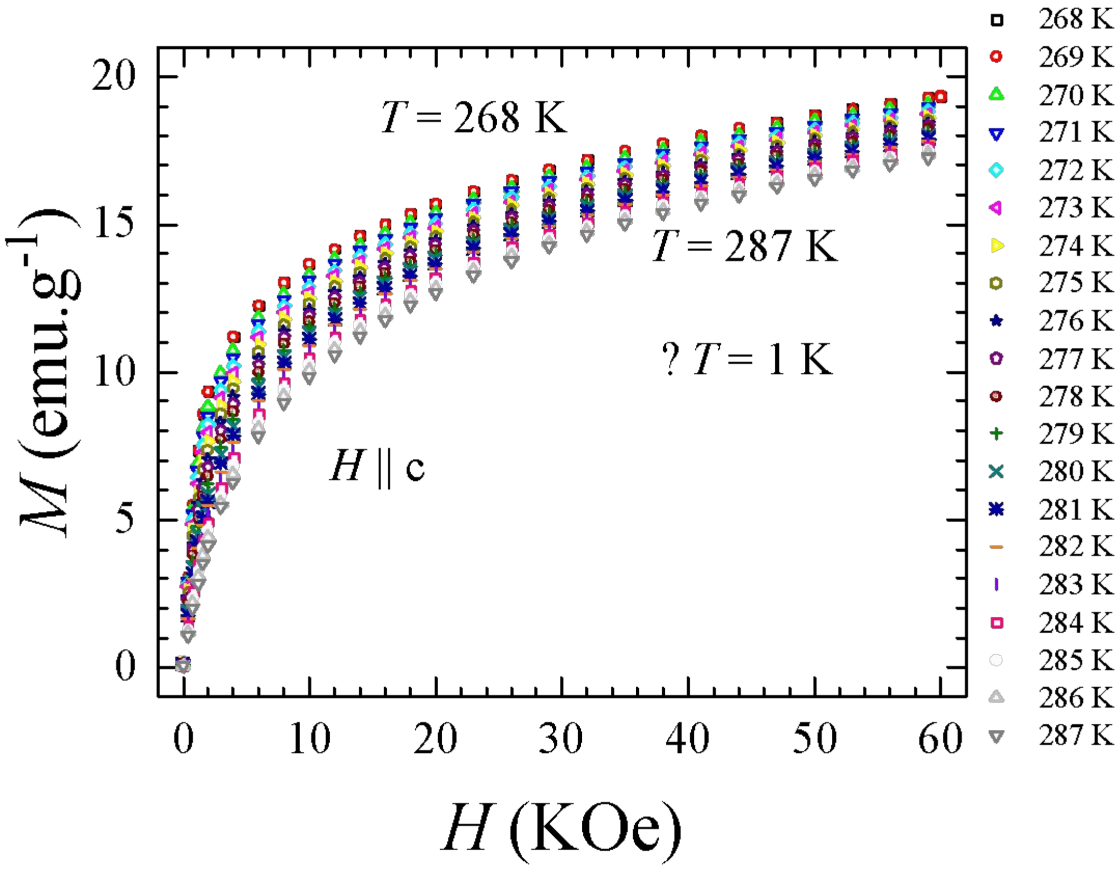


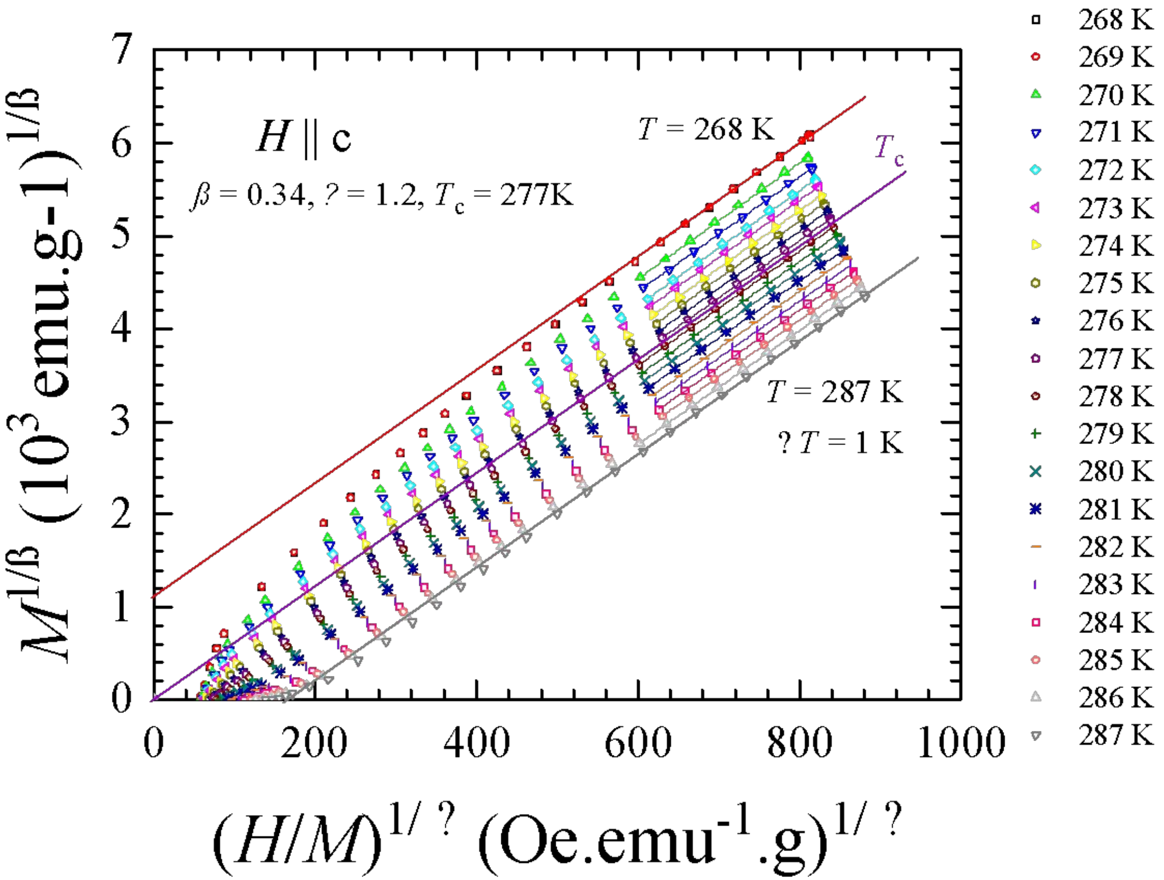




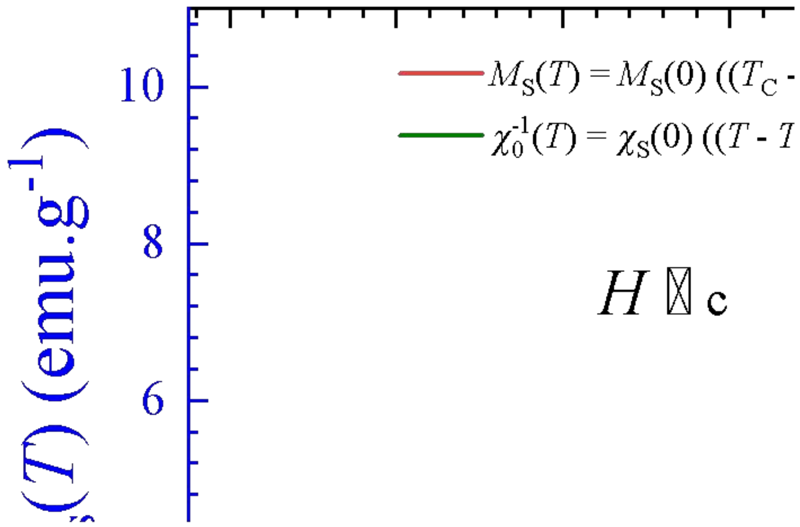
- F4GT is a metallic vdW ferromagnet with a rhombohedral structure
- Has a nearly room temperature transition from para to ferromagnet

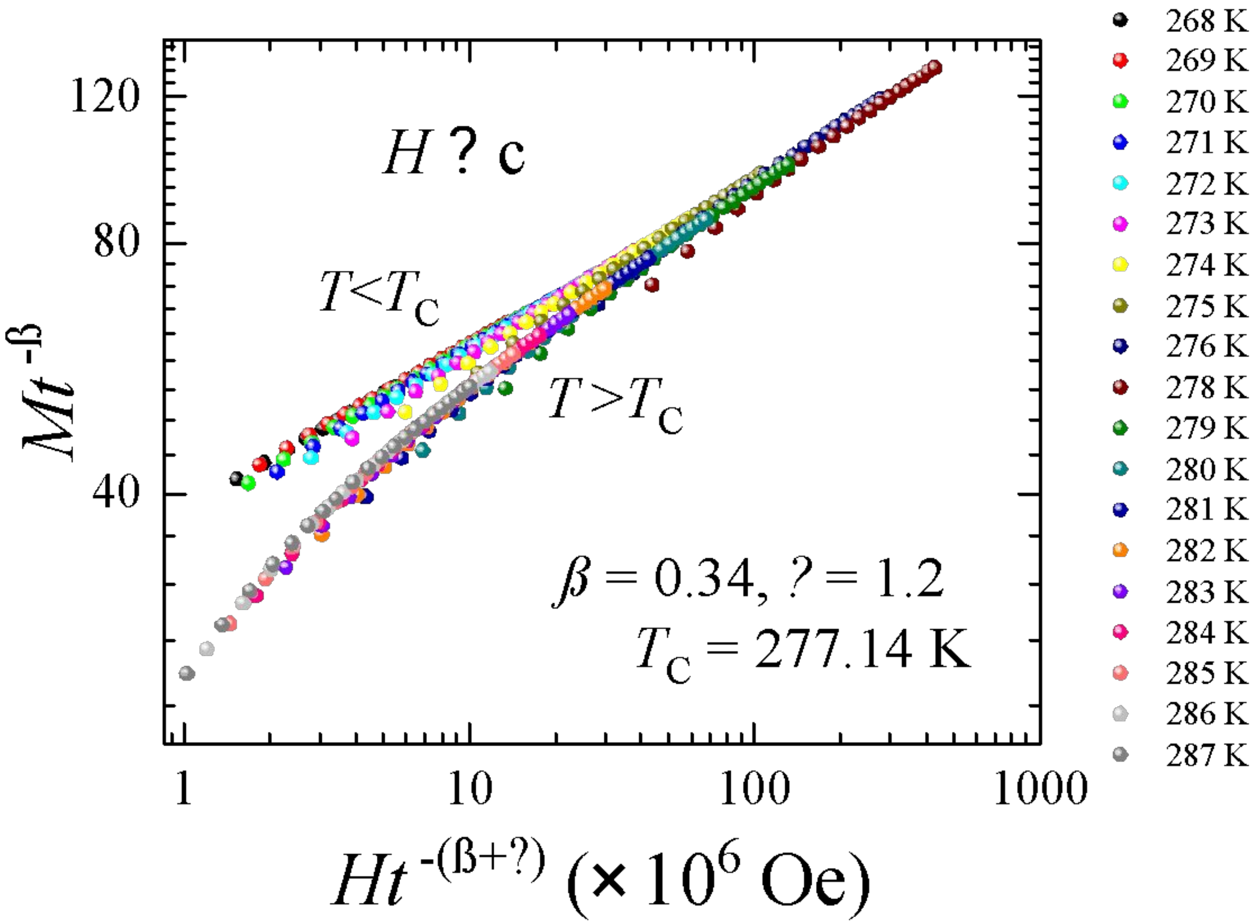
Scaling Analyses on bulk Fe_4GeTe_2





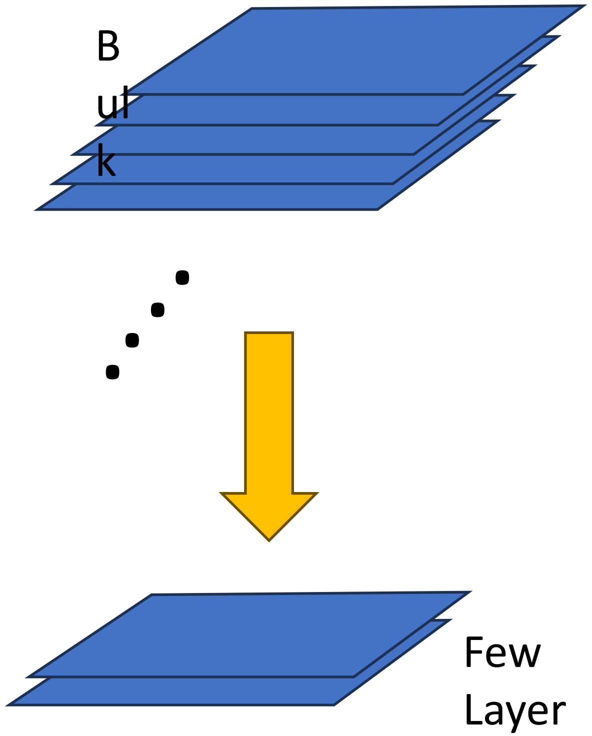
Suprotim Saha et al. (MS, submitted 2024)





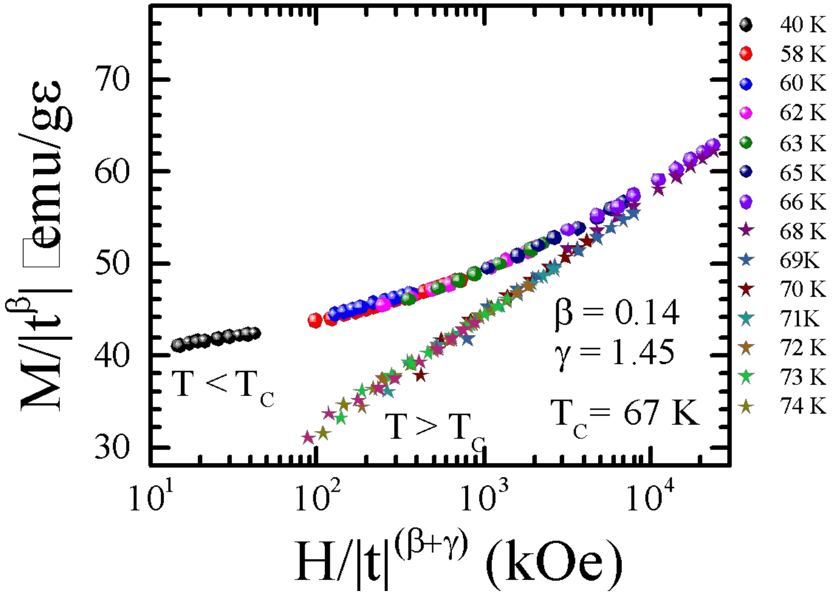
3D Heisenberg system

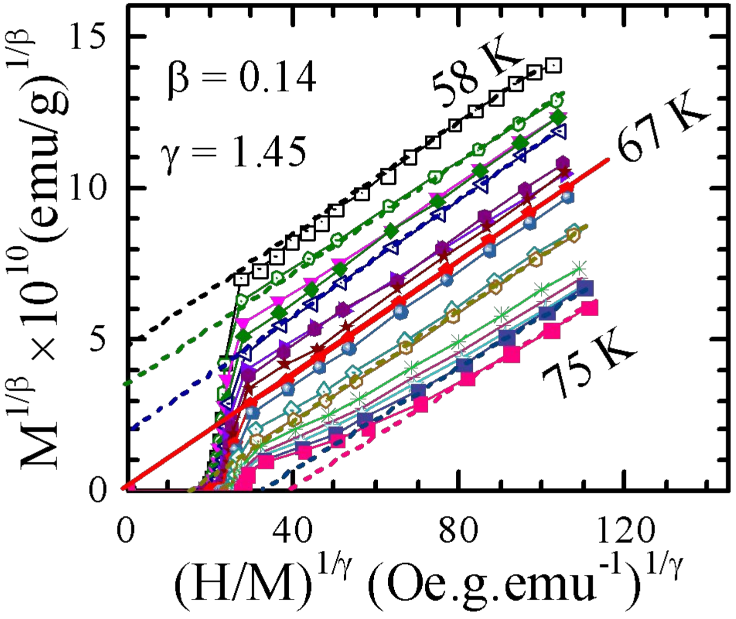
Our Analysis confirms 2D Ising universality class For bulk CGT



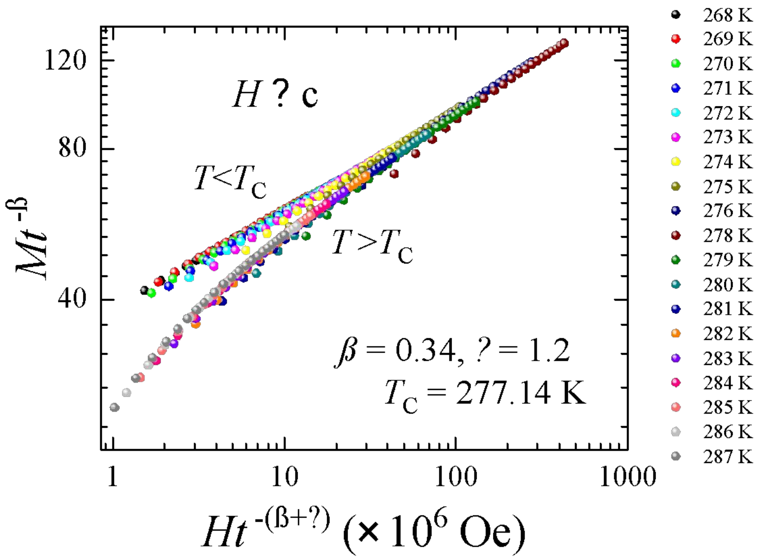
No critical scaling Studies on low dimensional CGT

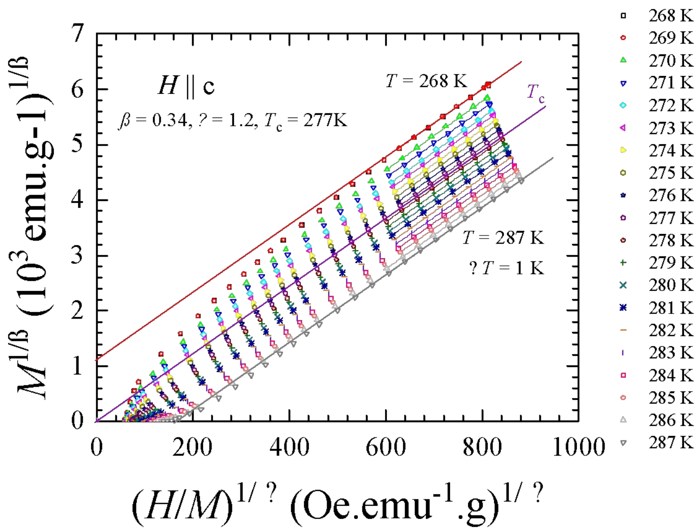
Critical scaling study in bulk CGT: 2D Ising Universality Class



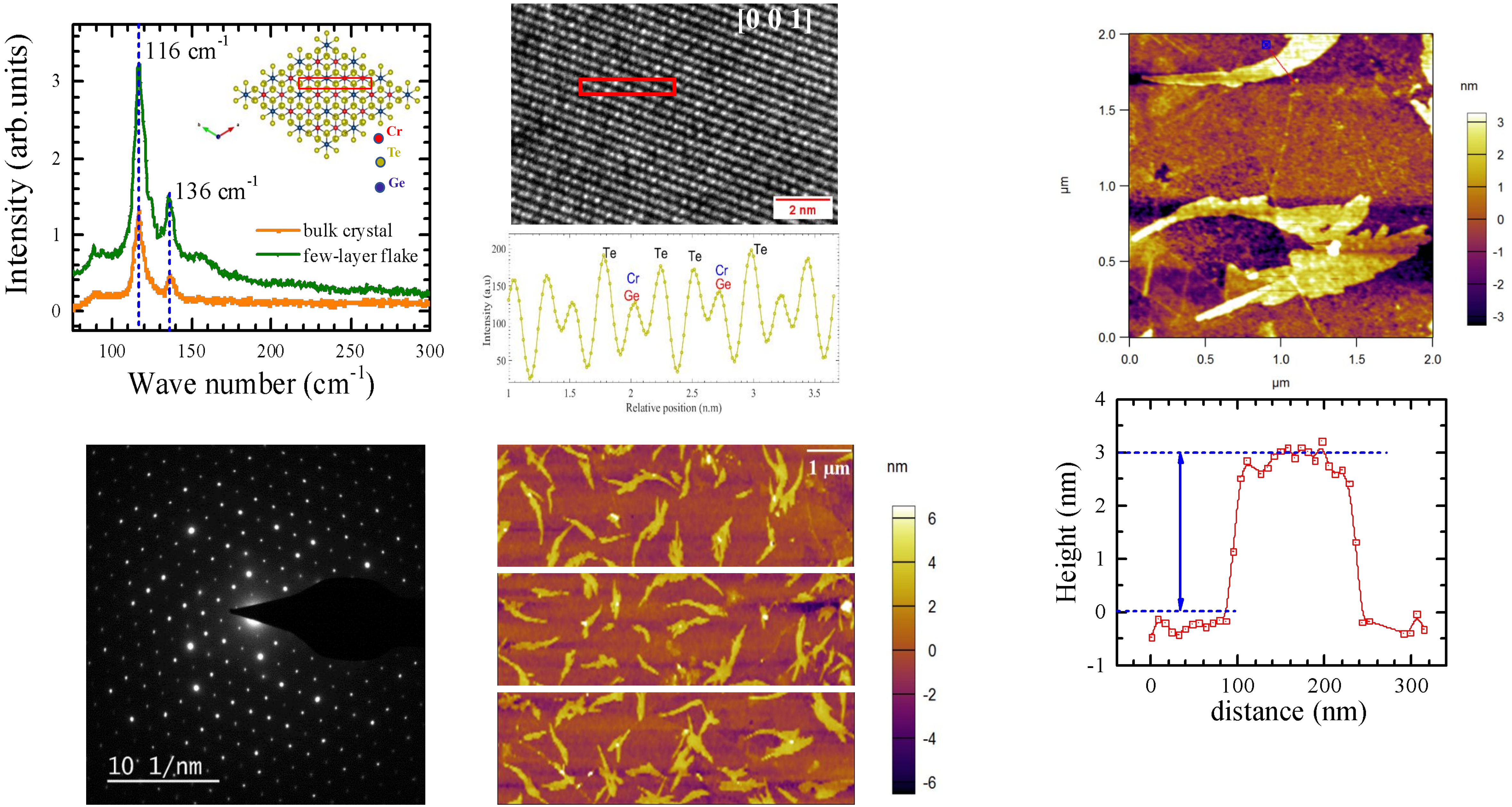


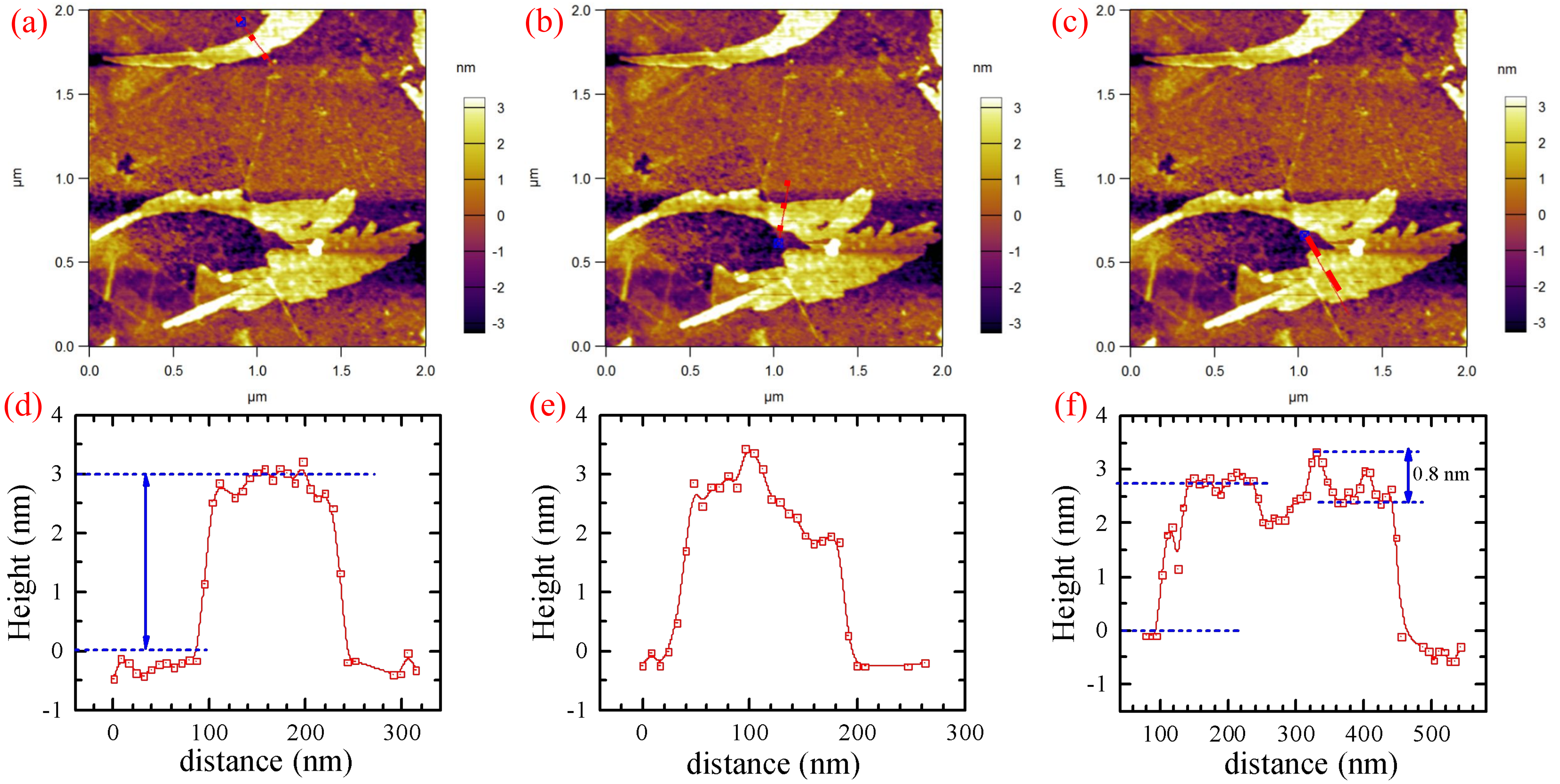
Critical scaling study in bulk F4GT: 3D Heisenberg Universality



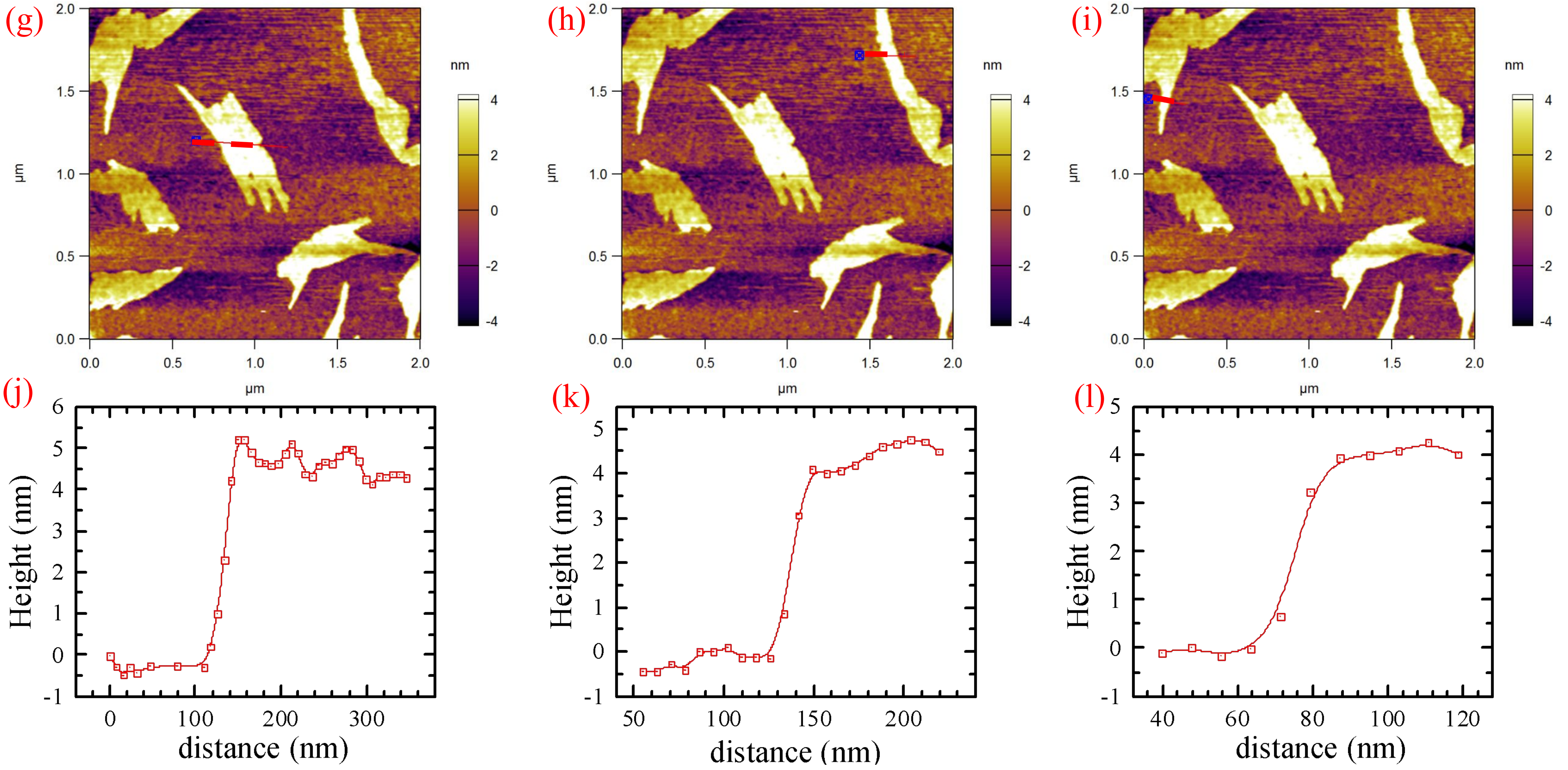


Few layer thick CGT on HBN



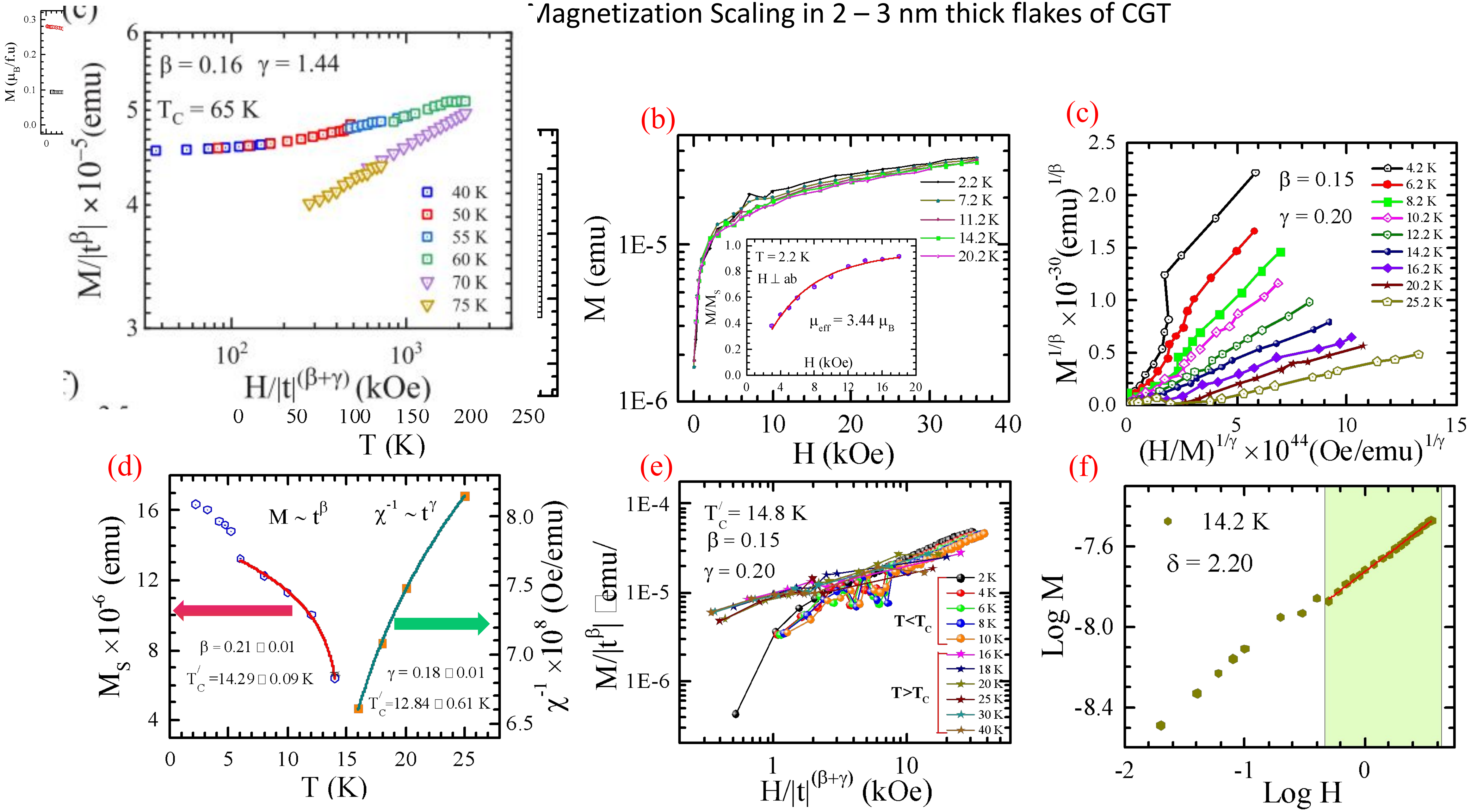


(a)
(b)
(c)
(d)
(e)
(f)
µm
nm
Height (nm)
distance (nm)
0.8 nm



(g)
(h)
(i)
(j)
(k)
(l)
nm
μm
Height (nm)
distance (nm)

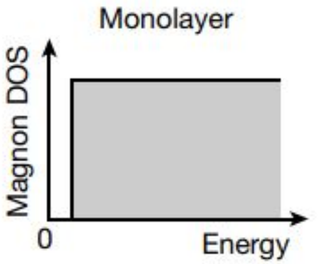
Magnetization Scaling in 2 – 3 nm thick flakes of CGT



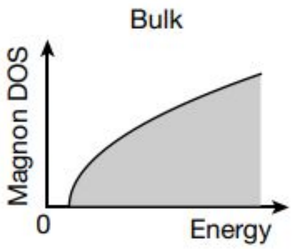
Universality Class for bulk and few layer CGT

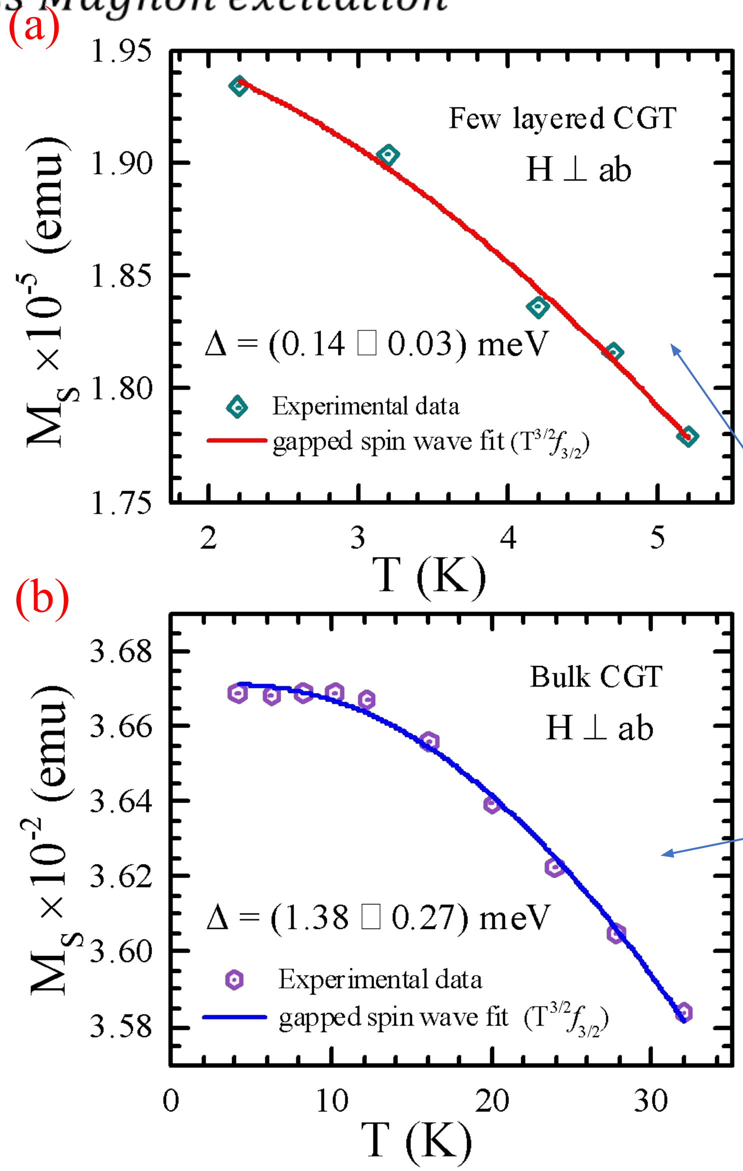
Sample	μ_{eff}	β	β (refs.[33, 34])	γ	γ (refs.[33, 34])	δ	δ (refs.[33, 34])	T_c	T'_c
CGT bulk crystal	$3.18\mu_B$ per Cr	0.14 (Fig.2(b)) 0.12 (Fig.2(d))	2d-Ising 0.17 to 0.2	1.45 (Fig.2(b)) 1.21 (Fig.2(d))	2d-Ising 1.75 to 1.28	11.37 (Fig. S7(a)) 11.35 (Widom)	2d-Ising 10.87 to 7.96	64.28K (Fig.2(a)) 63.68K - 66.59 K (Fig.2(d)) 67 K (Fig.2(b), (c))	--
CGT flake ensemble	$3.44\mu_B$ per Cr	0.15 (Fig.3(c)) 0.21 (Fig.3(d))	--	0.20 (Fig.3(c)) 0.18 (Fig.3(d))	--	2.20 (Fig.3(f)) 2.33 (Widom)	--	66.42 K (Fig.3(a))	14.29 K - 12.84 K (Fig.3(d)) 14.80 K (Fig.3(e))

$E = Dq^2$: *Gapless Magnon excitation*



$$E = \Delta + Dq^2$$





$$M(T,H) = M(0,H) - g\mu_B \left(\frac{k_B T}{4\pi D}\right)^{\frac{3}{2}} f_{\frac{3}{2}}\left(\frac{\Delta'}{k_B T}\right)$$

$f_{\frac{3}{2}}(y)$ is the Bose-Einstein integral function

$\Delta' = (0.14 \pm 0.03)$ meV for **few-layered flake ensemble**

$\Delta = (1.38 \pm 0.27)$ meV for **bulk CGT**

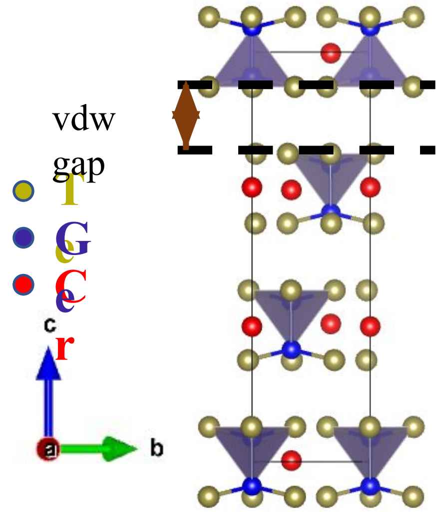
Clearly there is a change in anisotropy with lowering of Dimensionality of CGT.

DFT Calculations
Prasenjit Sen
(HRI, IISER Tirupati)
& Sourav Mal)

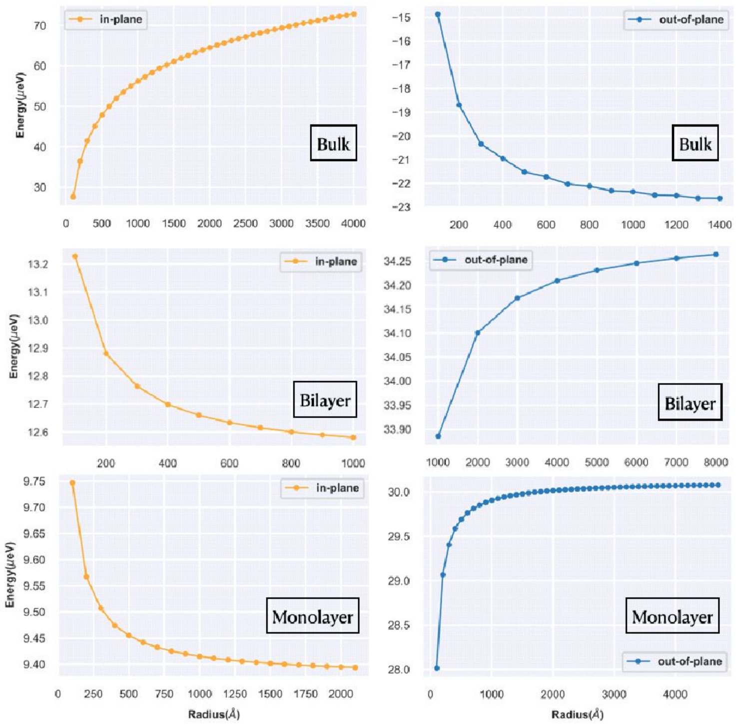
Structural Distortions with reducing Dimensionality

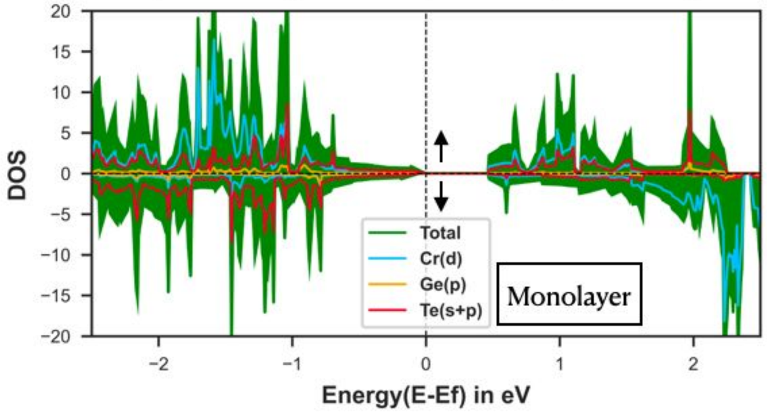
System	a(Å)	b(Å)	Cr-Cr(Å)	Cr-Te(Å)	Te-Te(Å)	∠Cr-Te-Cr
Bulk	6.892	6.892	3.98	2.78	3.765 ± 0.035	91.29°
Bilayer	6.882	6.882	3.97	2.78	3.755 ± 0.015	90.96° ± 0.12°
Monolayer	6.875	6.875	3.97	2.79	3.75	90.72°

0.02A



System	MAE (meV/f.u.)	DAE (meV/f.u.)	ϵ_{ani} (meV/f.u.)
Bulk	0.3	0.095	0.395
Bilayer	0.143	-0.022	0.121
Monolayer	0.066	-0.021	0.045





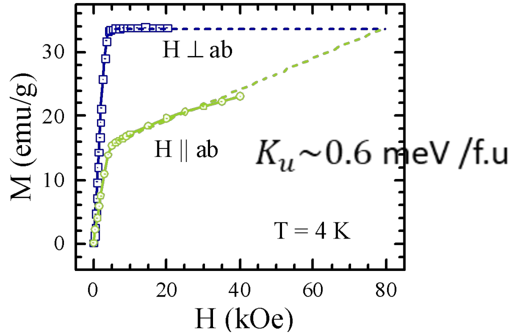
$$U = \frac{\mu_0}{4\pi}\frac{1}{r^3}\left[\vec{m}_1.\vec{m}_2 - 3\left(\vec{m}_1.\hat{r}\right)\left(\vec{m}_2.\hat{r}\right)\right]$$

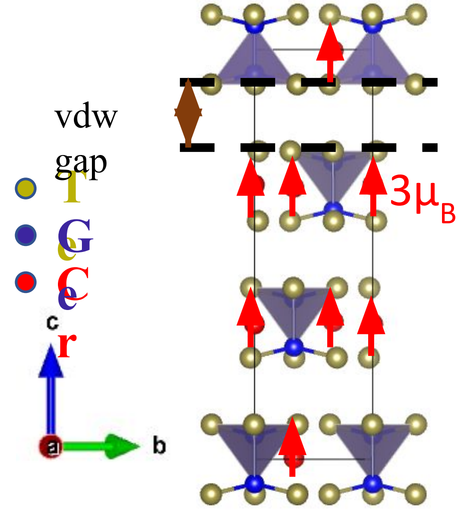
$$\mathrm{MAE} = \left[\left(E_{n_2,\mathrm{up}} + E_{n_2,\mathrm{dn}}\right) - \left(E_{n_1,\mathrm{up}} + E_{n_1,\mathrm{dn}}\right)\right]$$

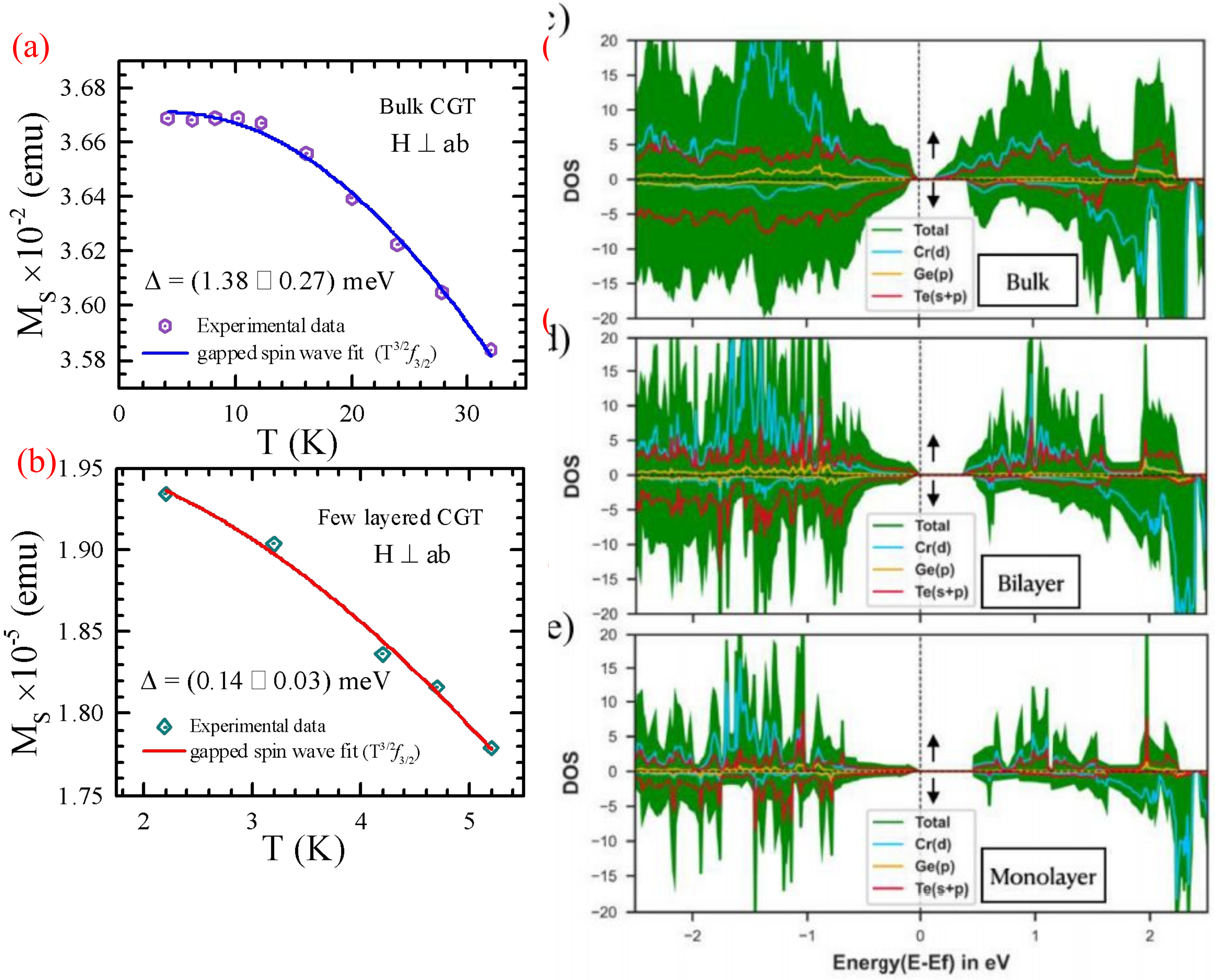
Where n_1 and n_2 are the easy and hard direction of magnetization.

Incorporates SOC

DAE = (OOP, U)-(IP, U)

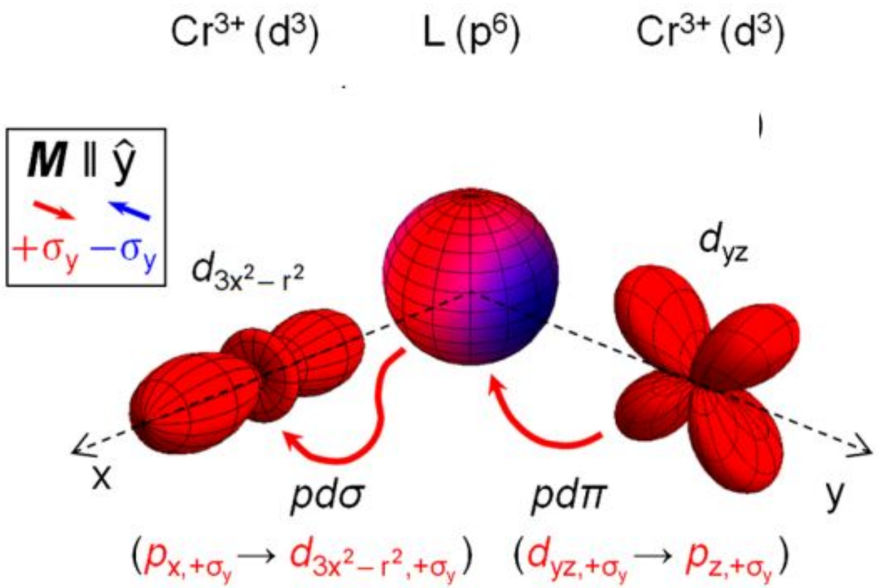






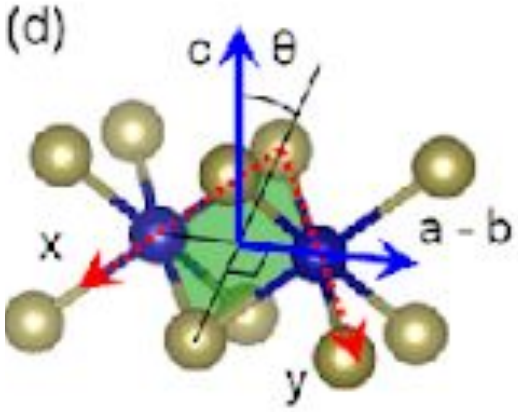
A drop in contribution of Te
To the DOS at Ef,
Results in drop in Anisotropy Gap

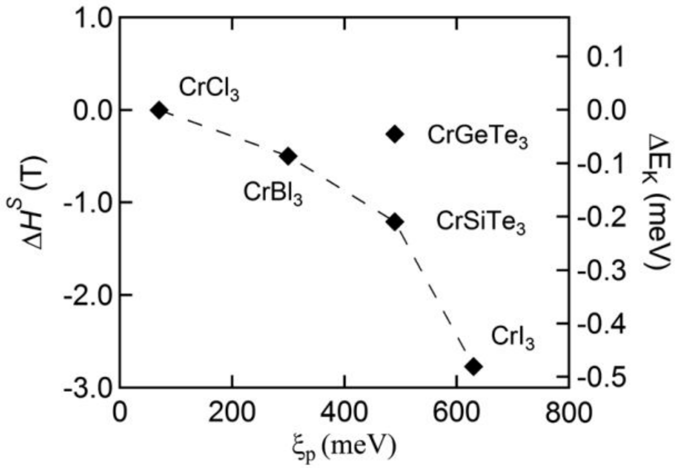
Another competing effect
Is the structural distortions
Affect the Hopping in
Xy plane and
Thereby affect anisotropy
Even further.



System	a(Å)	b(Å)	Cr-Cr(Å)	Cr-Te(Å)	Te-Te(Å)	∠Cr-Te-Cr
Bulk	6.892	6.892	3.98	2.78	3.765 ± 0.035	91.29°
Bilayer	6.882	6.882	3.97	2.78	3.755 ± 0.015	90.96° ± 0.12°
Monolayer	6.875	6.875	3.97	2.79	3.75	90.72°

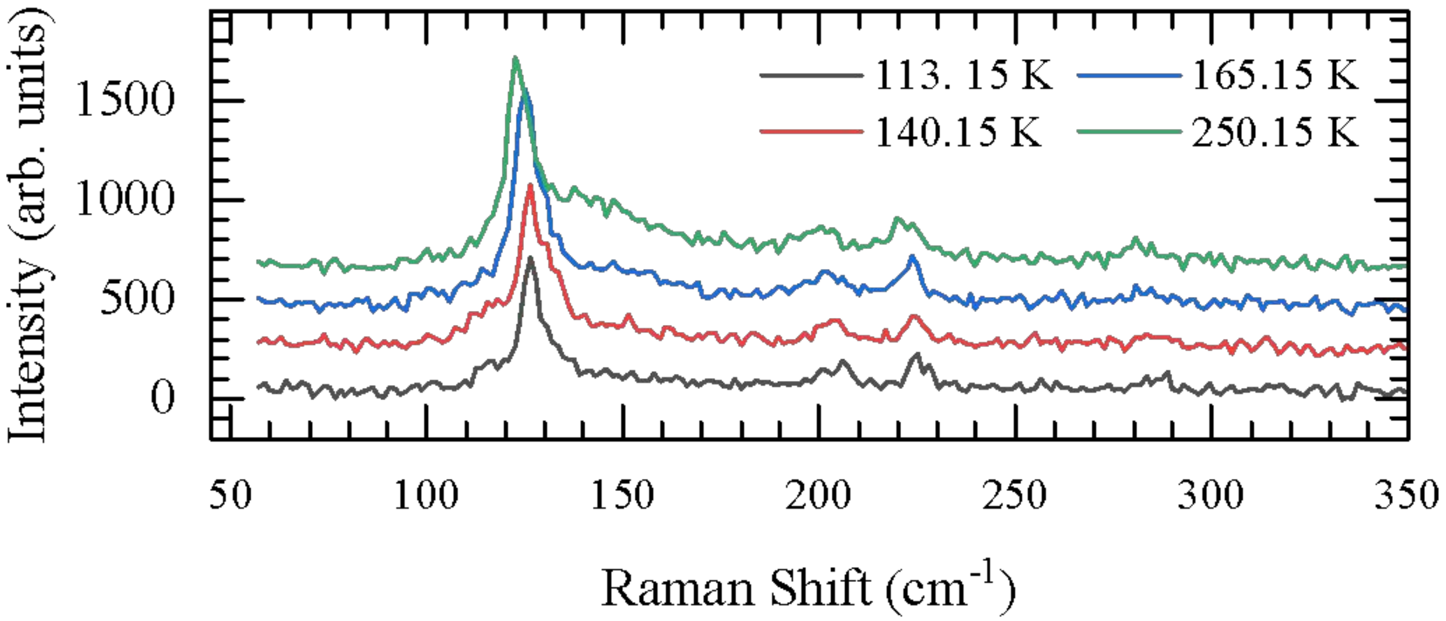
0.02A





Studying magnetoelastic coupling from T-dependent Raman spectroscopy in surface of CGT

*Vertically offset for clarity

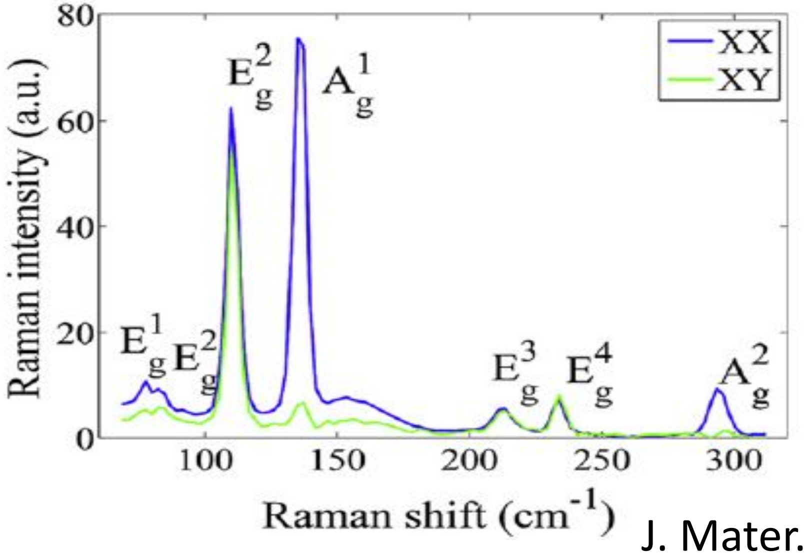


The peaks show blue-shift with decreasing T.

The peak position (cm^{-1}) vs T (K) were fit with Boltzman sigmoidal model which describes the anharmonic dependence of phonon frequency

$$\omega(T) = \omega_1 + \frac{\omega_0 - \omega_1}{1 + \exp\left(\frac{T - T_0}{\Delta T}\right)}$$

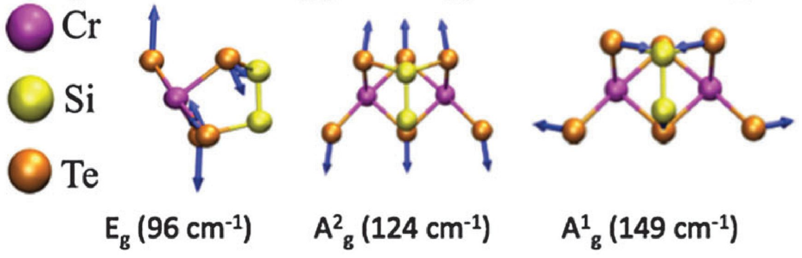
ω_0 = top (i.e. lowest T) freq.
ω_1 = bottom (i.e. highest T) freq.
T_0 = central temperature
ΔT = width of the curve

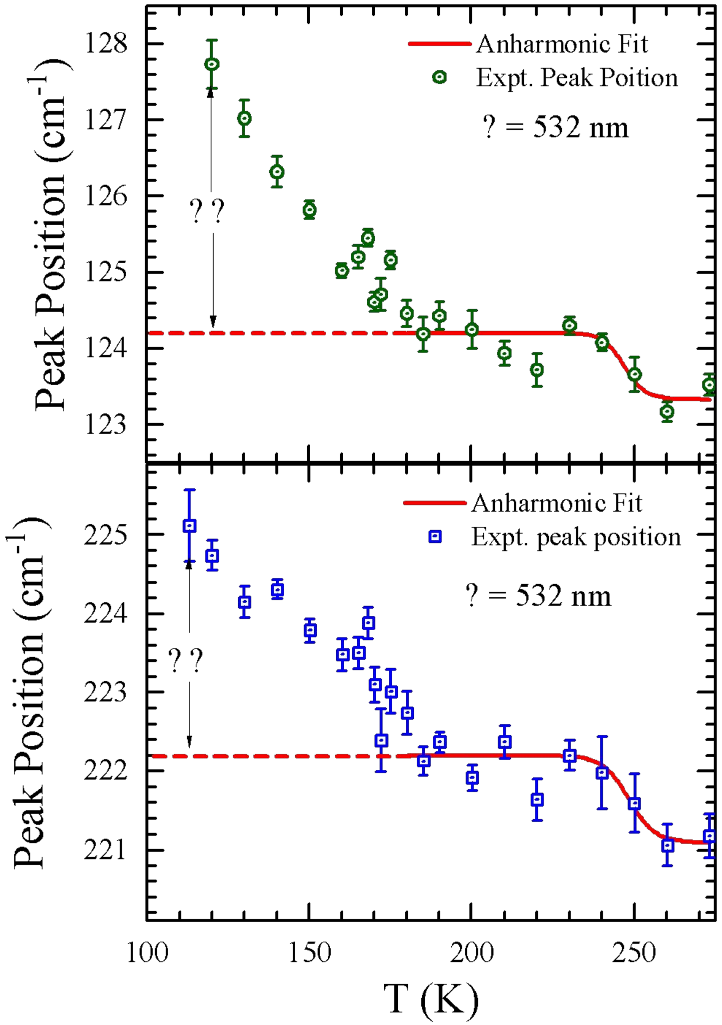


J. Mater. Chem. C, 2016, 4,315

P C Mahato et al. (MS under preparation)

Approximate comparison with a related system Cr2Si2Te6





The spin-phonon coupling strength λ represents how strongly the magnetic ordering influences the atomic vibrations of the non-magnetic sublattice via the superexchange pathway.

$$\lambda = \frac{1}{\mu}\frac{\partial^2 J}{\partial u^2}$$

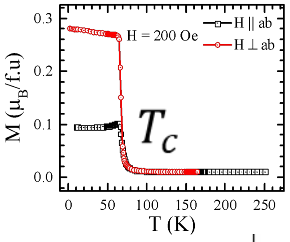
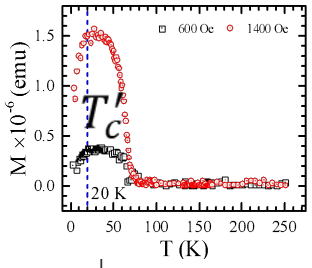
μ is the reduced mass
u = atomic displacement
J = N.N exchange energy

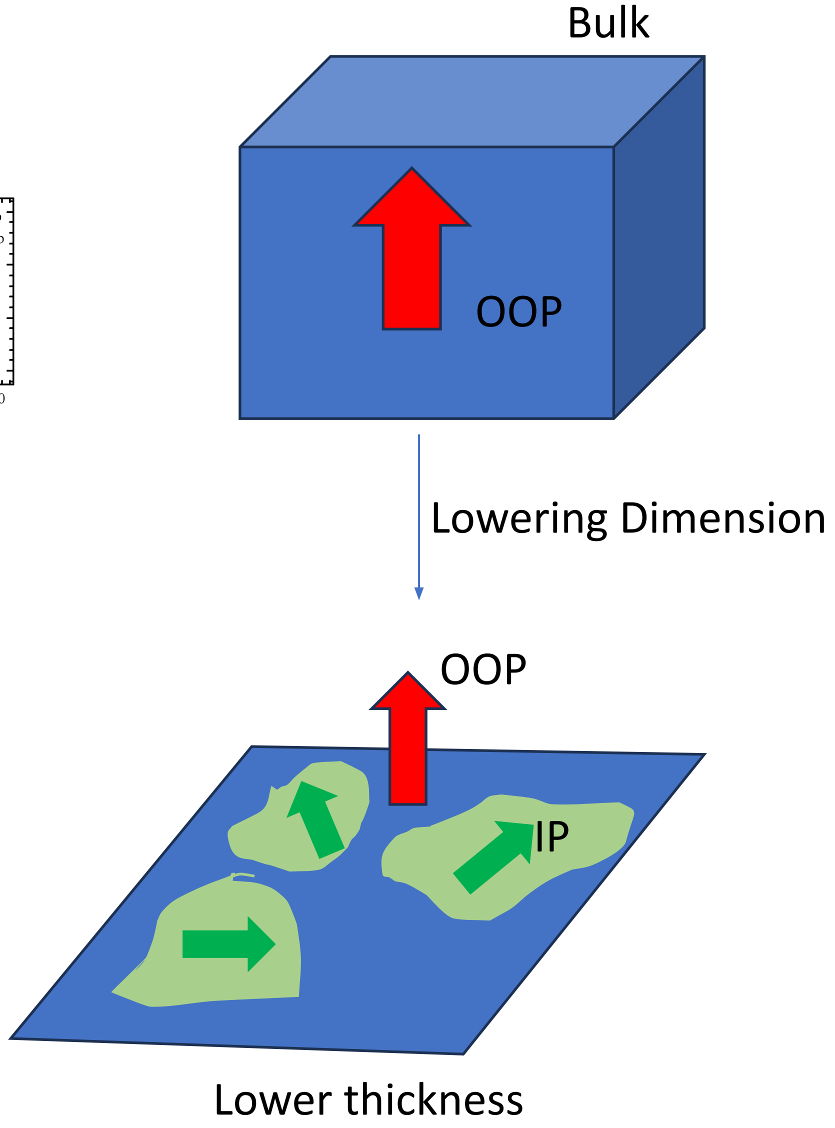
To extract the spin-phonon coupling parameter (λ) we use, $\omega \approx \omega_0 + \lambda' \langle \overrightarrow{S_i}.\overrightarrow{S_j} \rangle$

$\lambda' = \frac{\Delta\omega}{\langle \overrightarrow{S_i}.\overrightarrow{S_j} \rangle}$ ($\Delta\omega$ is the shift in wave number from the sigmoidal model (at the lowest T))

λ' = 1.09 and 1.28 (assuming S = 3/2) for the peak centered at 122.8 cm^{-1} and 221 cm^{-1}, respectively

Competing Inplane and Out of Plane Anisotropy

Sample	μ_{eff}	β	β (refs.[33, 34])	γ	γ (refs.[33, 34])	δ	δ (refs.[33, 34])	T_c	T'_c
CGT bulk crystal	$3.18\mu_B$ per Cr	0.14 (Fig.2(b)) 0.12 (Fig.2(d))	2d-Ising 0.17 to 0.2	1.45 (Fig.2(b)) 1.21 (Fig.2(d))	2d-Ising 1.75 to 1.28	11.37 (Fig. S7(a)) 11.35 (Widom)	2d-Ising 10.87 to 7.96	64.28K (Fig.2(a)) 63.68K - 66.59 K (Fig.2(d)) 67 K (Fig.2(b), (c))	
CGT flake ensemble	$3.44\mu_B$ per Cr	0.15 (Fig.3(c)) 0.21 (Fig.3(d))	--	0.20 (Fig.3(c)) 0.18 (Fig.3(d))	--	2.20 (Fig.3(f)) 2.33 (Widom)			14.29 K - 12.84 K (Fig.3(d)) 14.80 K (Fig.3(e))



$$\mathcal{H} = \mathcal{H}_{int,z} + \mathcal{H}'_{xy}$$

$$\mathcal{H}_{int,z} = -J_z \sum_{\langle ij\rangle} s_z^i s_z^j + K$$

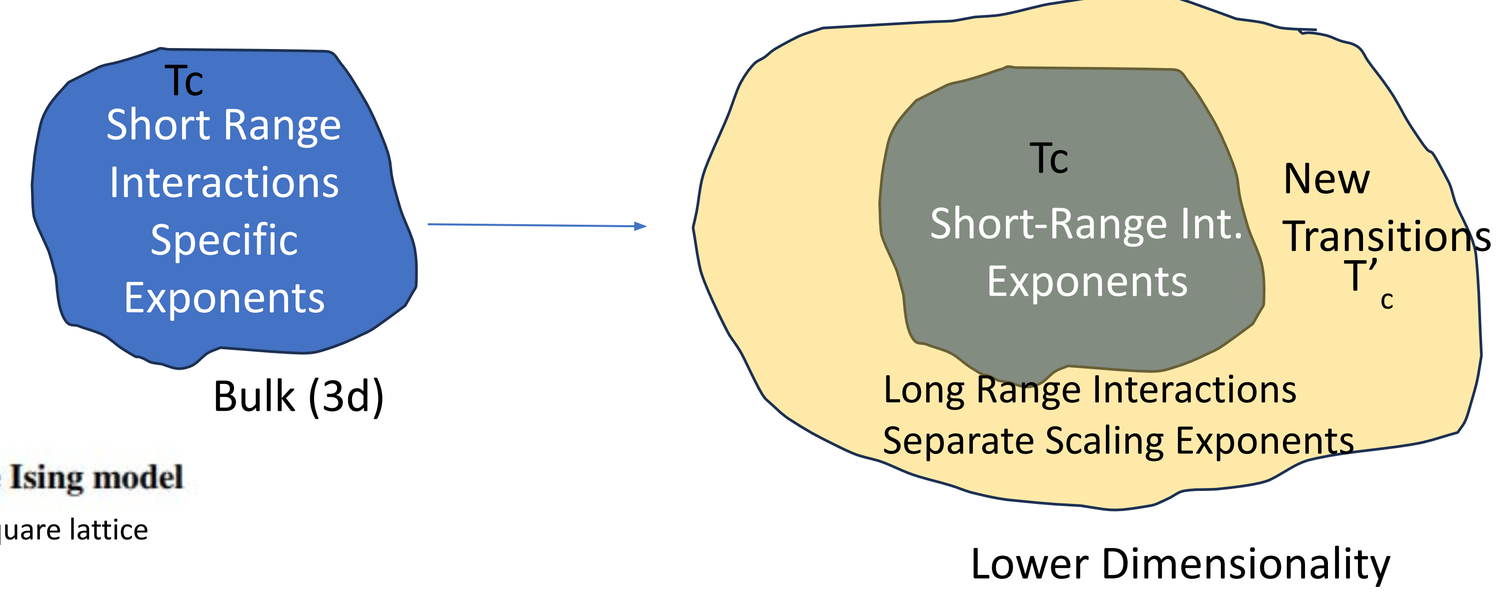
Dominates in bulk, Critical Fluctuations along the z - direction
Promotes OOP Anisotropy
2d Ising Universality Class

$$H'_{xy} = -J'_d \sum_{<ij>} \left[s_x^i s_x^j + s_y^i s_y^j\right] + H_{dip}$$

Lowering thickness, J'_d (Hopping) w.r.t $J_z \downarrow$, *critical fluctuations released along xy*
SOC + MAE decreases
Inplane DAE becomes active

Competition

Critical Crossovers



PHYSICAL REVIEW B **84**, 054433 (2011)

Crossover between a short-range and a long-range Ising model

Short range interaction between neighbouring Ising spins on a square lattice

$$\mathcal{H}_{\rm IS} = -J \sum_{\langle i,j \rangle} \sigma_i \sigma_j ,$$ ➡ Tc ~ 2.267… J

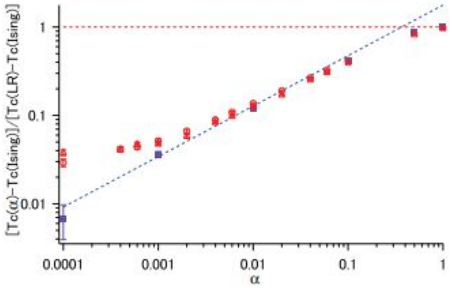
Long range interaction between any Ising spins

$$\mathcal{H}_{\rm HT} = -\frac{4J_0}{2N} \sum_{i=1}^{N} \sum_{j=1}^{N} \sigma_i \sigma_j$$ ➡ Tc ~ 4 J

$$\mathcal{H} = (1-\alpha)\mathcal{H}_{\rm IS} + \alpha \mathcal{H}_{\rm HT}, \quad 0 \leqslant \alpha \leqslant 1 . \quad (5)$$

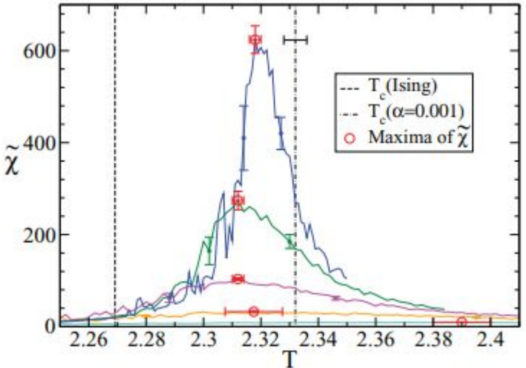
Here, α controls the relative strength of the long-range interaction.

The Tc shifts



$$\frac{1}{N}(\langle M^2 \rangle - \langle M \rangle^2)$$

Susceptibility Still Diverges



Pure shortrange: Cluster Size Divergence

Pure Longrange: No cluster size Divergence

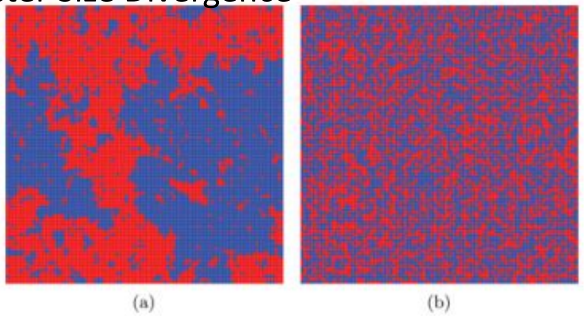


FIG. 5. (Color online) (a) Spin configurations for Ising model at $T_c^{\rm IS} = 2.269J$, $L = 100$; (b) Husimi-Temperley model at $T_c^{\rm HT} = 4J_0$.

Mixed Model

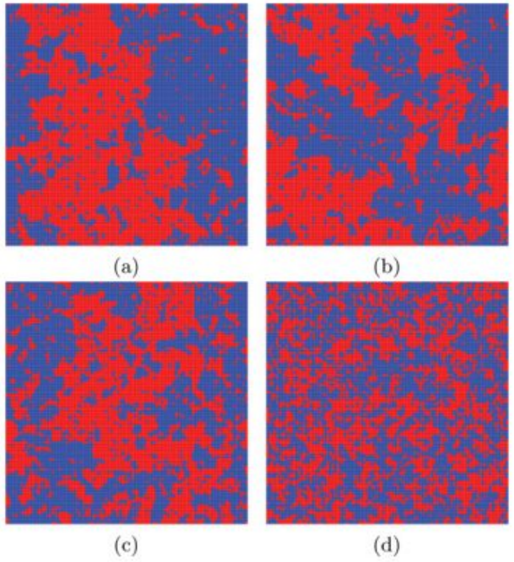
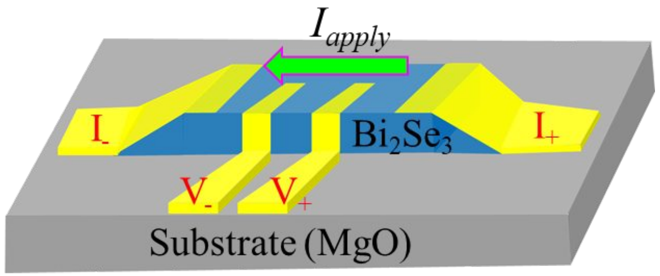


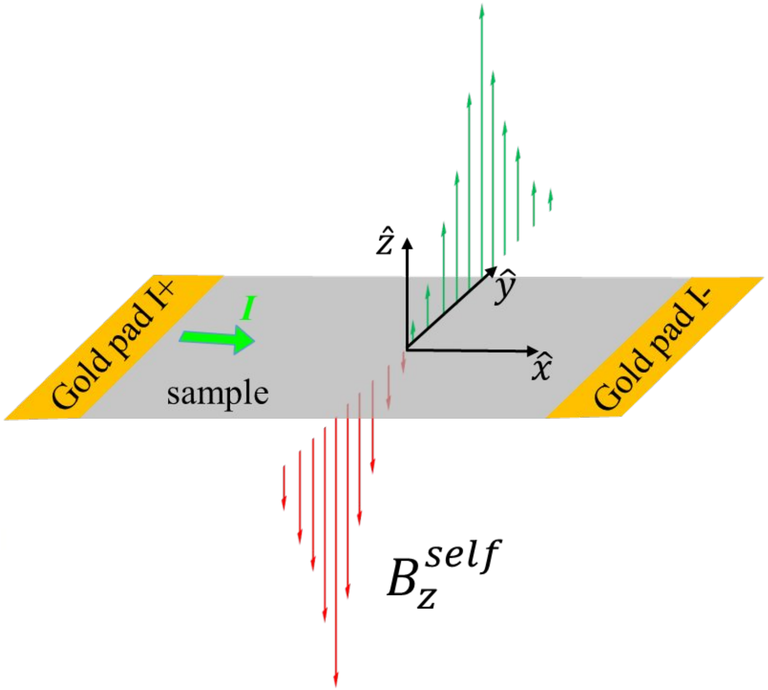
FIG. 7. (Color online) Typical configurations of the hybrid model at the critical temperature $T_c(\alpha)$ for (a) $\alpha = 0.0001$, (b) $\alpha = 0.001$, (c) $\alpha = 0.01$, and (d) $\alpha = 0.1$.

Current imaging technique – A modified magneto-optical imaging technique

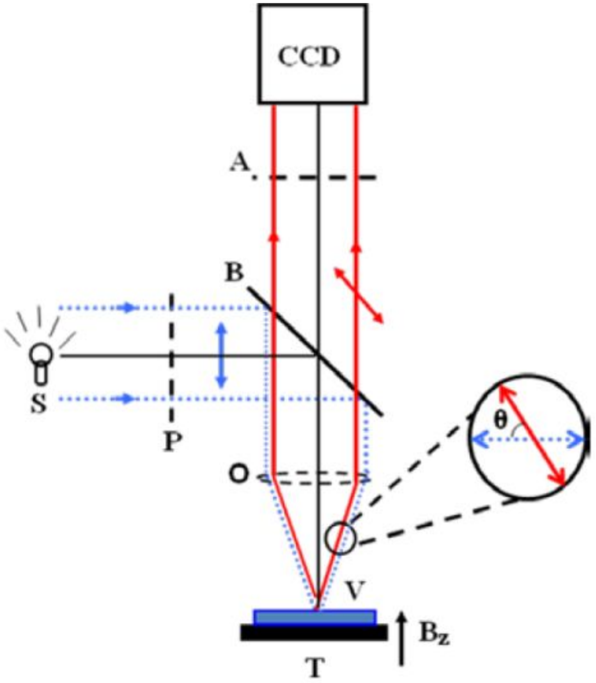
Self field imaging



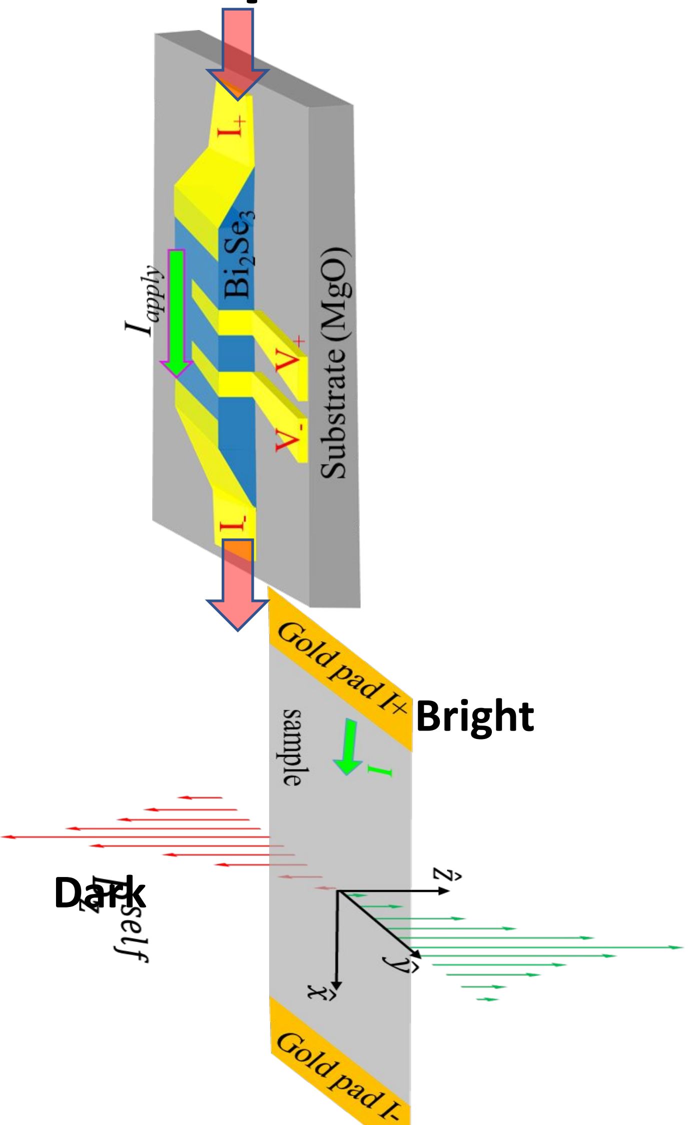
Differential MOI = I(+i) – I(-i)



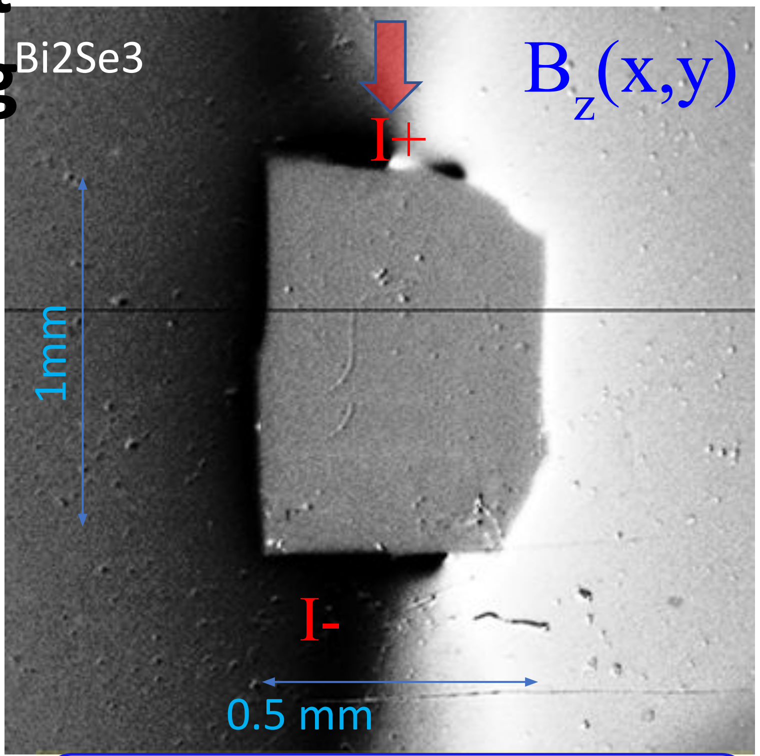
Normal Faraday rotation



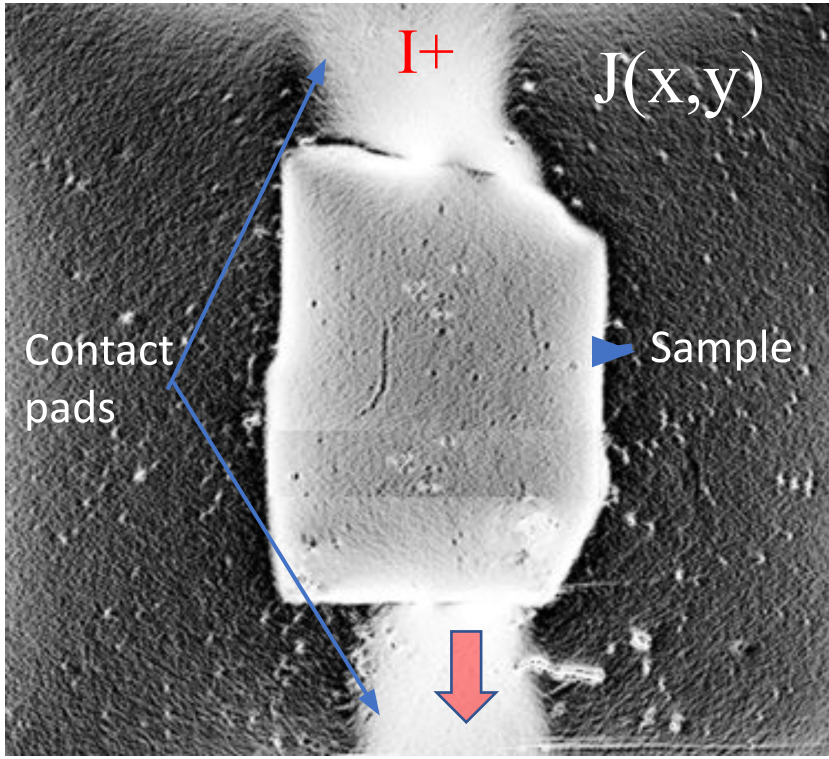
Developed a Current distribution Imaging technique



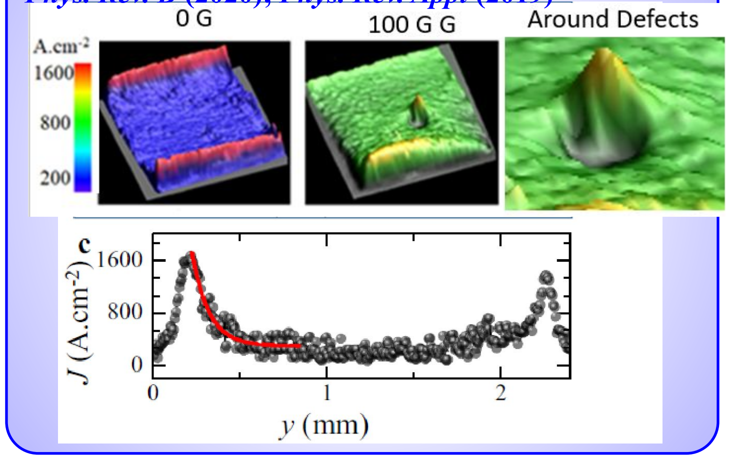
Field distribution image



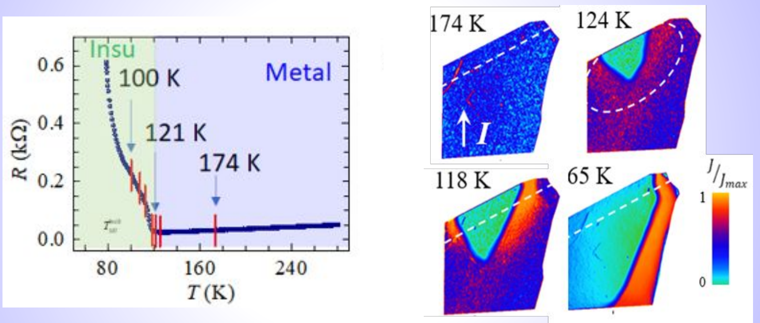
Current distribution image



Imaging current distribution in a topological insulator Bi_2Se_3: A. Jash *et. al.* Sci. Rep. (2021) *Phys. Rev. B* (2020); *Phys. Rev. Appl* (2019)

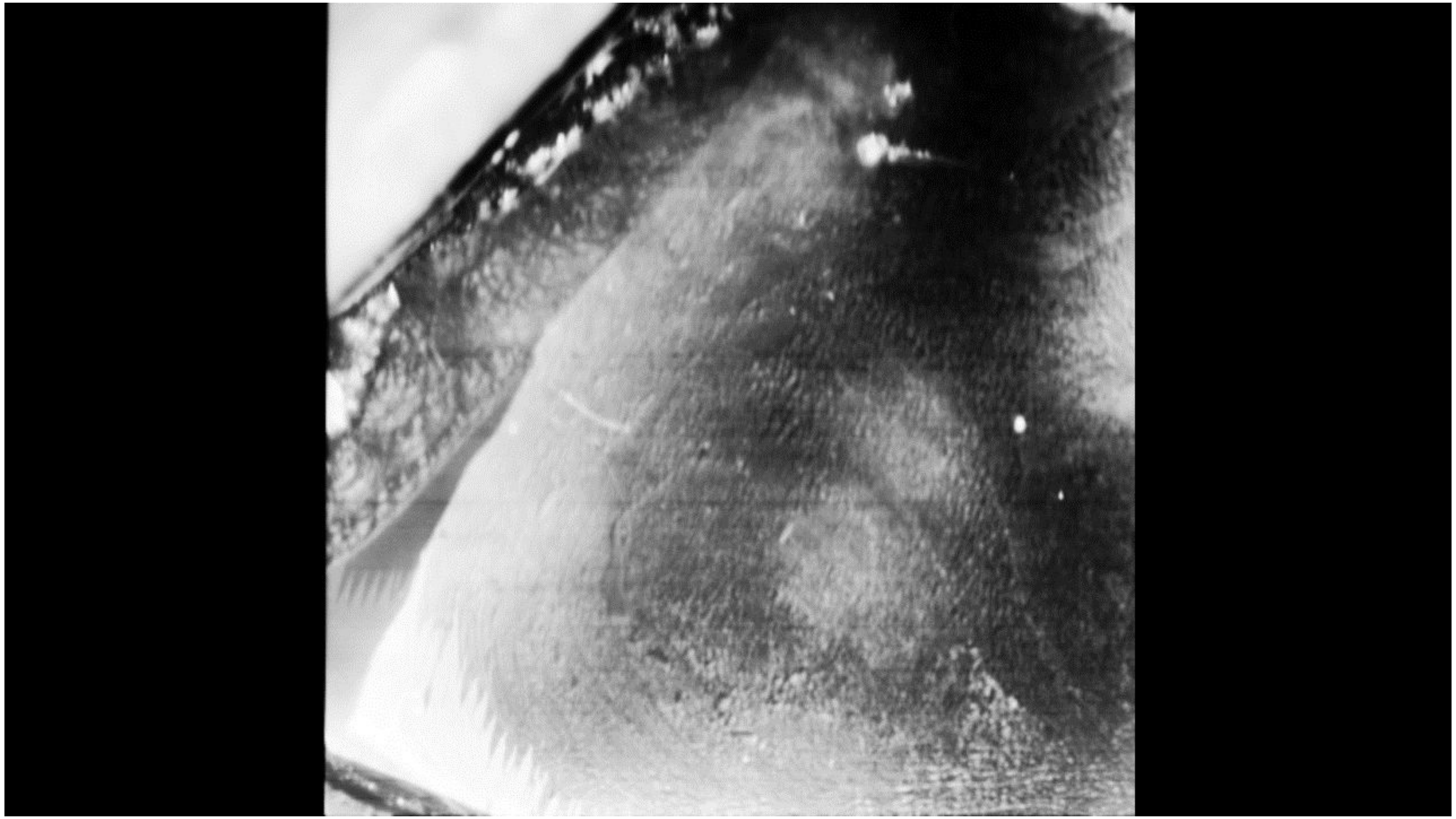


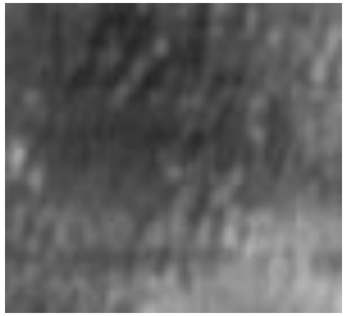
Imaging electric current across MIT in $NdNiO_3$ films: N. Roy *et. al.* Phys. Rev. B (2022)



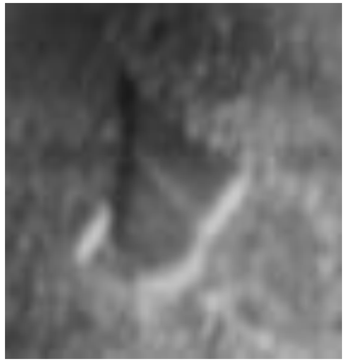
Magneto-optical imaging in CGT

Fine graining at lower T





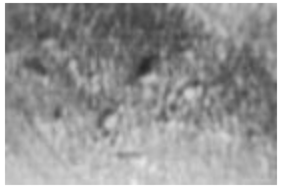
100 K



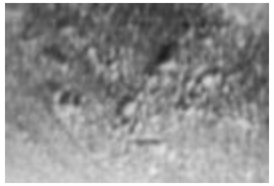
Visible

> 62.8 K

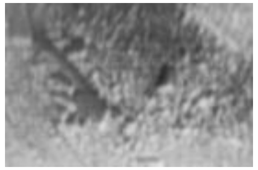
>55.85K



>57.84K



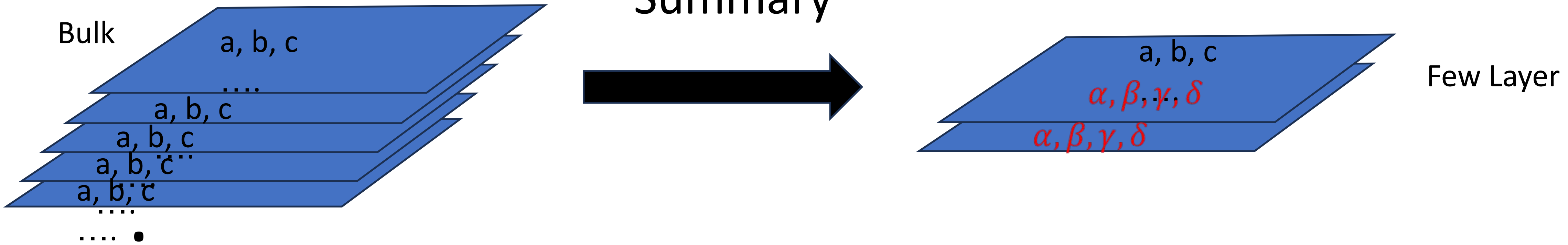
>58.84K



>59.84 K

PC Mahato (MS under Preparation, 2024)

Summary

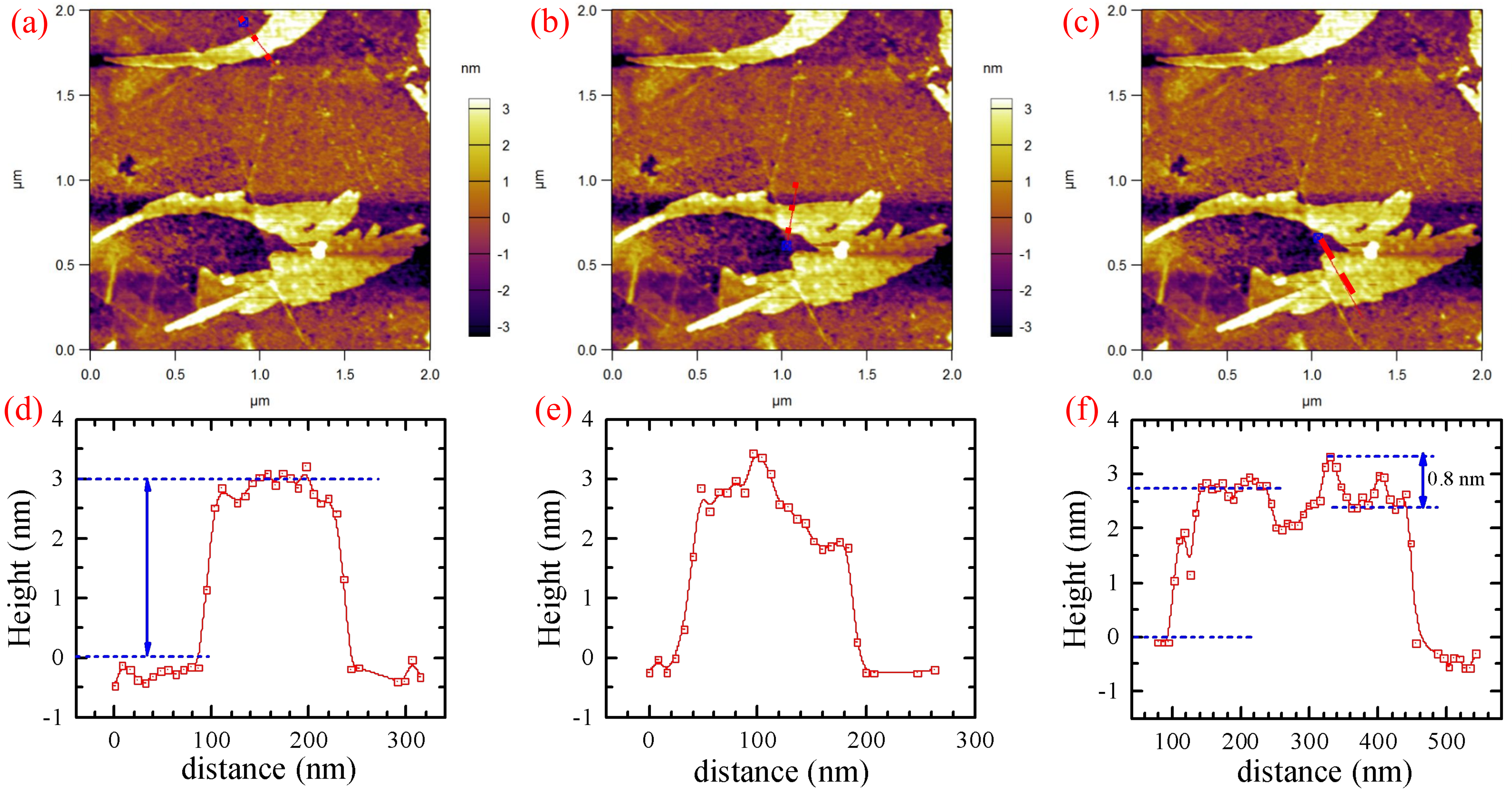


- Lowering of dimensionality in CGT is not simply cleaving the vdW layers
- The universality class is not preserved.
- Subtle changes in structure seem to trigger changes in SOC
- This leads to a competition between Magneto –crystalline anisotropy and Dipolar Anisotropy energy
- Effective magnetic anisotropy is weakened with lowering dimensionality of the vdW, a lower Tc emerges
- Lowered dimensionality there is a complex competition between shortrange interactions (exchange) and long range dipolar interaction
- Role of local fluctuations affecting magnetic order at longer length scales may become quite important
- Some of the Above maybe quite general features of magnetic vdW material

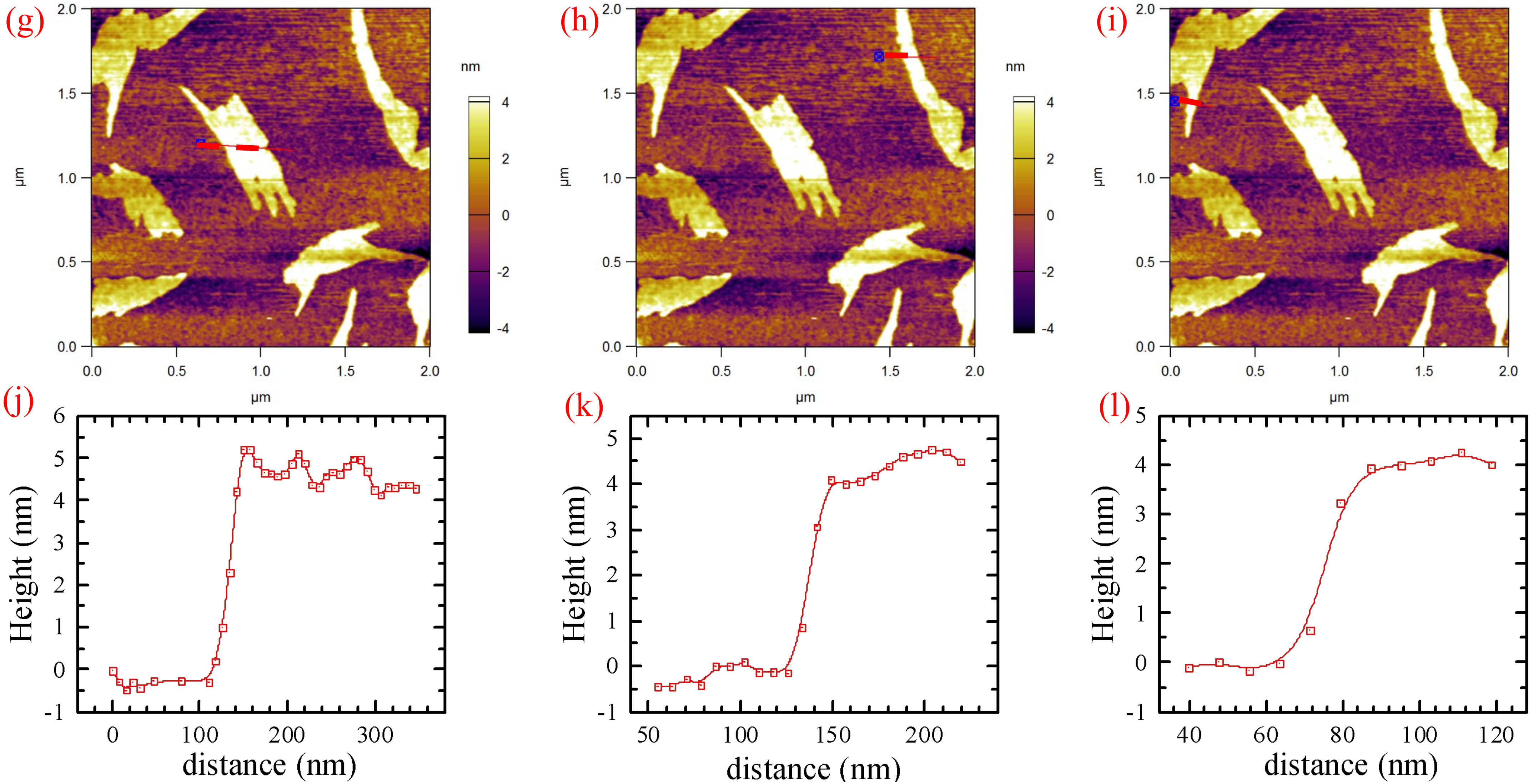
Thankyou

EXTRA SLIDES

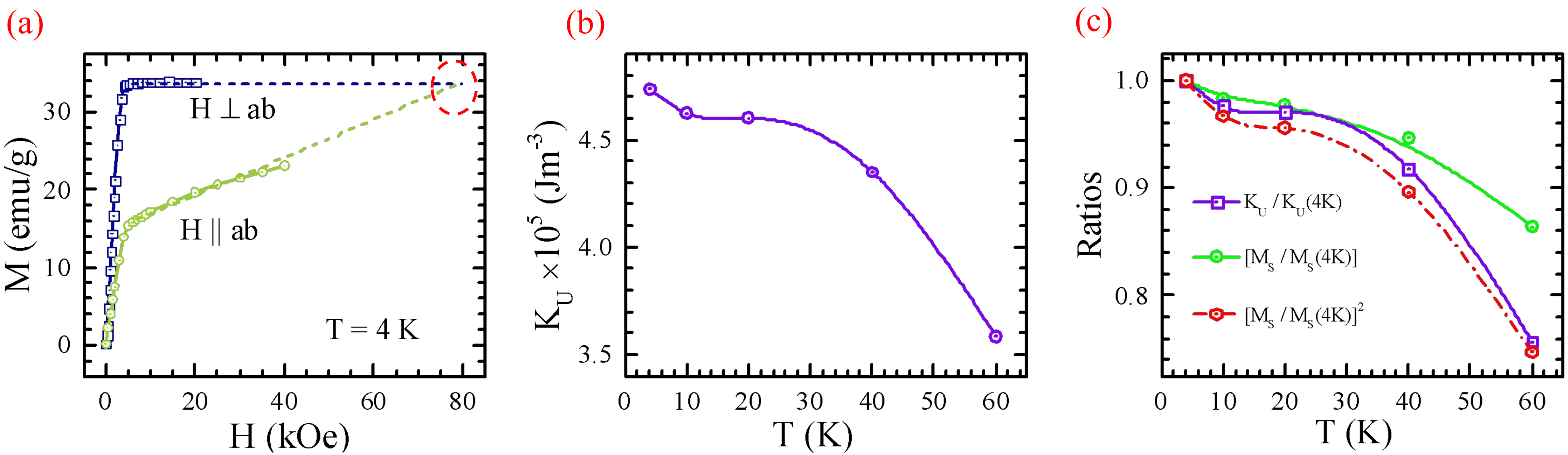
First principles DFT calculations were performed using the Vienna ab initio simulation package (VASP)19, 20. Electronic wave functions were expressed in terms of a plane wave basis set with an energy cutoff of 500 eV. Projector augmented wave (PAW)21 potentials were used to represent the interaction between the valence electrons and the ion cores. The generalised gradient approximation (GGA) as proposed by Perdew, Burke and Ernzerhof (PBE)22 was used to treat the exchange-correlation energy. The GGA+U23 method was used to treat the strong correlations of Cr 3d electrons. It was previously reported9 that the appropriate range of U should be within 0.2 < U < 1.7 eV in order to reproduce the correct experimental magnetic ground state. Hence U = 1eV was taken in our calculation. DFT-D3 method of Grimme24 was used to treat the van der Waals interactions between the CGT layers. The structures were optimized by changing both the ionic positions and lattice parameters of the simulation cell until the energy and force on each atom converged to less than 10^(-7) eV and 10^(-6) eV/Å, respectively.



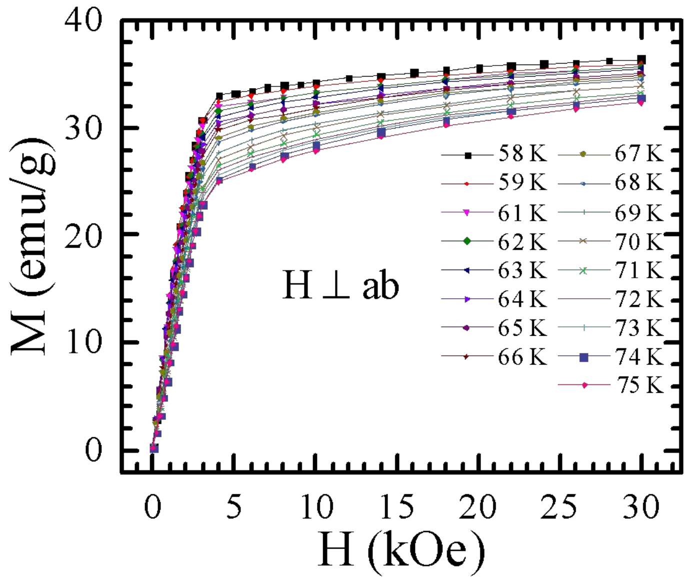
(a)
(b)
(c)
(d)
(e)
(f)
μm
nm
Height (nm)
distance (nm)
0.8 nm



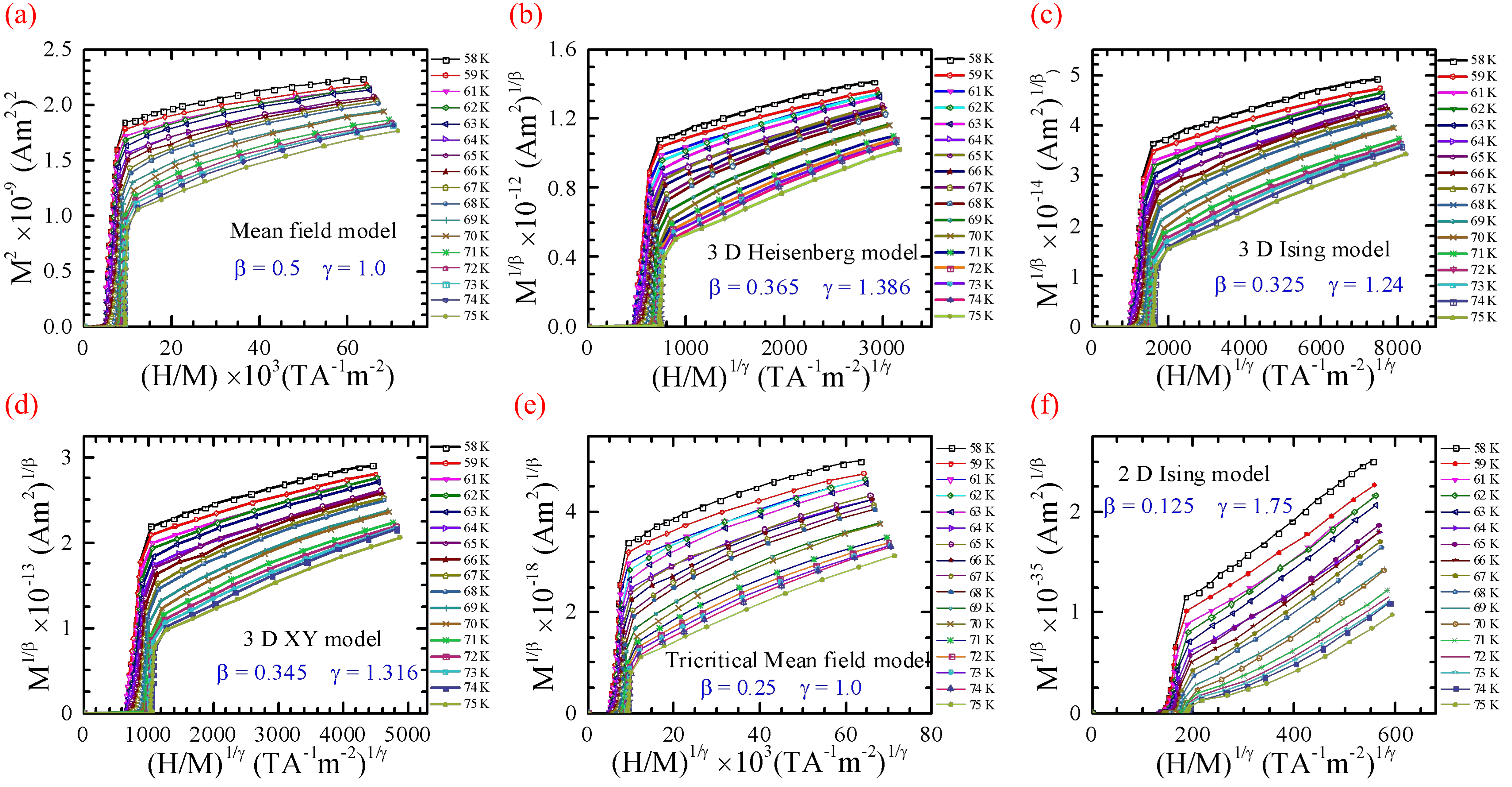
(g)
(h)
(i)
µm
nm
4
2
0
-2
-4
0.0
0.5
1.0
1.5
2.0
(j)
(k)
(l)
Height (nm)
distance (nm)



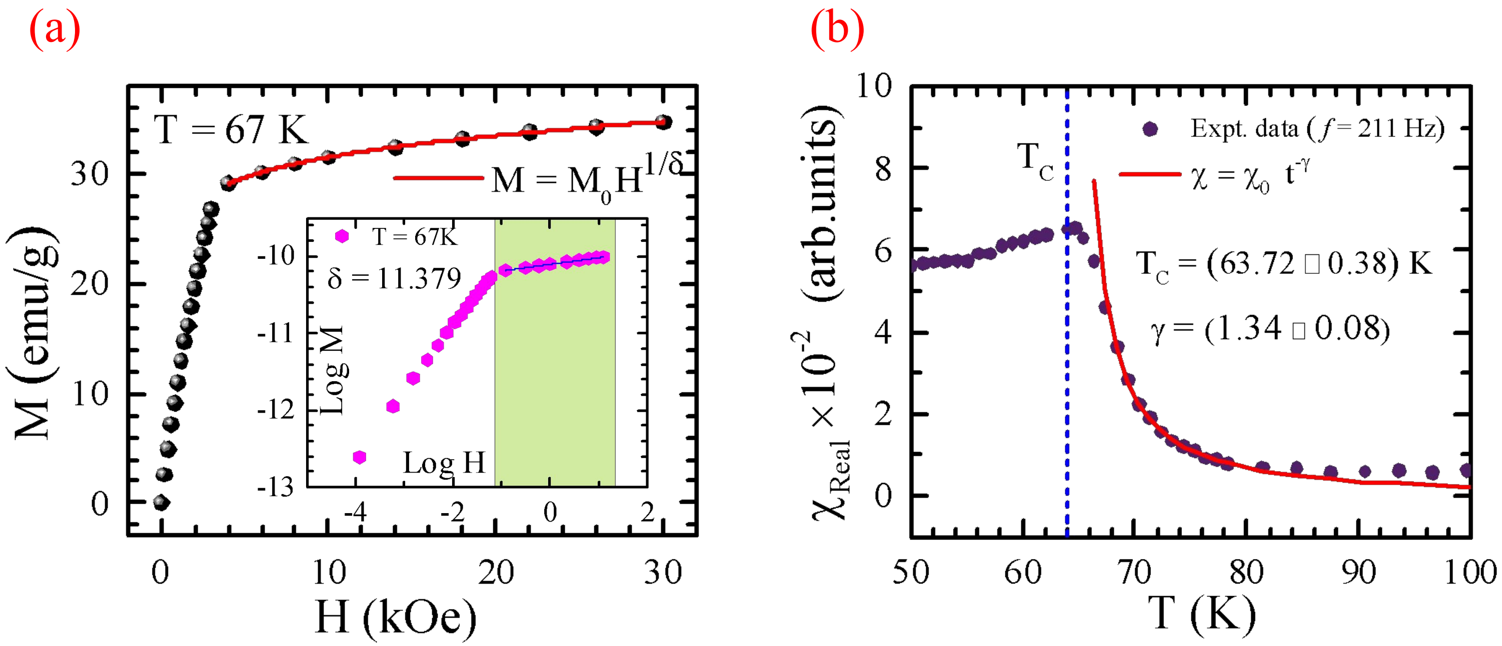
(a)
M (emu/g)
H (kOe)
H ⊥ ab
H || ab
T = 4 K
(b)
K_U ×10^5 (Jm^-3)
T (K)
(c)
Ratios
T (K)
K_U / K_U(4K)
[M_S / M_S(4K)]
[M_S / M_S(4K)]^2



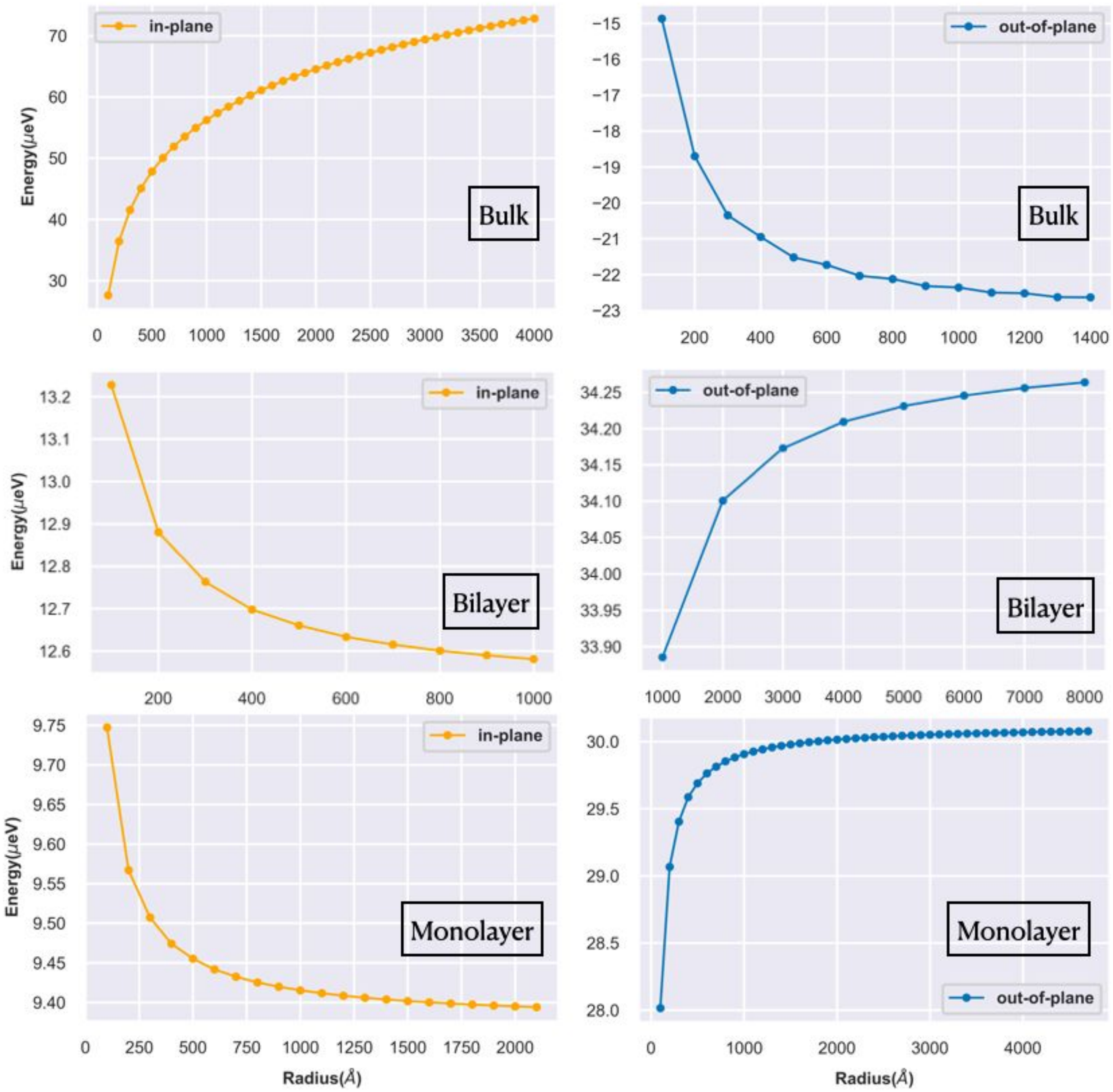
M (emu/g)
H (kOe)
H ⊥ ab
58 K
59 K
61 K
62 K
63 K
64 K
65 K
66 K
67 K
68 K
69 K
70 K
71 K
72 K
73 K
74 K
75 K

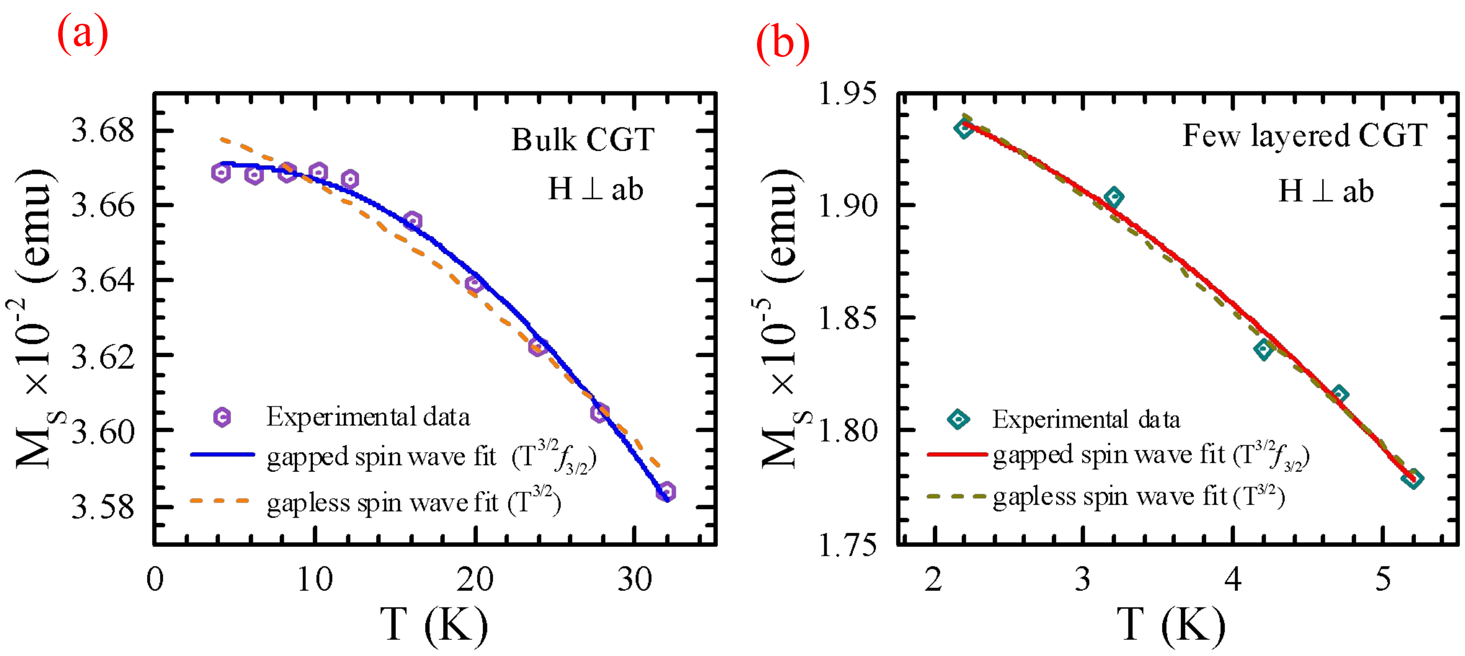


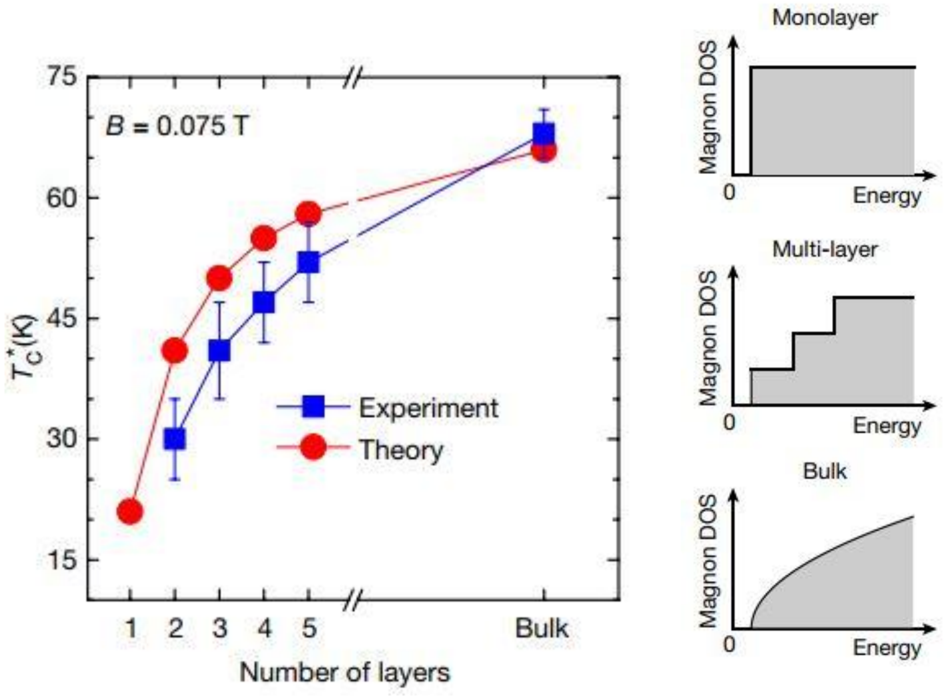
(a)
Mean field model
β = 0.5 γ = 1.0
$M^2 \times 10^{-9}$ $(Am^2)^2$
$(H/M) \times 10^3 (TA^{-1}m^{-2})$
(b)
3 D Heisenberg model
β = 0.365 γ = 1.386
$M^{1/\beta} \times 10^{-12}$ $(Am^2)^{1/\beta}$
$(H/M)^{1/\gamma}$ $(TA^{-1}m^{-2})^{1/\gamma}$
(c)
3 D Ising model
β = 0.325 γ = 1.24
$M^{1/\beta} \times 10^{-14}$ $(Am^2)^{1/\beta}$
$(H/M)^{1/\gamma}$ $(TA^{-1}m^{-2})^{1/\gamma}$
(d)
3 D XY model
β = 0.345 γ = 1.316
$M^{1/\beta} \times 10^{-13}$ $(Am^2)^{1/\beta}$
$(H/M)^{1/\gamma}$ $(TA^{-1}m^{-2})^{1/\gamma}$
(e)
Tricritical Mean field model
β = 0.25 γ = 1.0
$M^{1/\beta} \times 10^{-18}$ $(Am^2)^{1/\beta}$
$(H/M)^{1/\gamma} \times 10^3 (TA^{-1}m^{-2})^{1/\gamma}$
(f)
2 D Ising model
β = 0.125 γ = 1.75
$M^{1/\beta} \times 10^{-35}$ $(Am^2)^{1/\beta}$
$(H/M)^{1/\gamma}$ $(TA^{-1}m^{-2})^{1/\gamma}$
58 K
59 K
61 K
62 K
63 K
64 K
65 K
66 K
67 K
68 K
69 K
70 K
71 K
72 K
73 K
74 K
75 K



(a)
T = 67 K
M = M0H1/δ
M (emu/g)
H (kOe)
T = 67K
δ = 11.379
Log M
Log H
(b)
Expt. data (f = 211 Hz)
χ = χ0 t-γ
TC
TC = (63.72 □ 0.38) K
γ = (1.34 □ 0.08)
χReal ×10-2 (arb.units)
T (K)


in-plane
Bulk
out-of-plane
Bulk
in-plane
Bilayer
out-of-plane
Bilayer
in-plane
Monolayer
out-of-plane
Monolayer
Energy(μeV)
Radius(Å)


(a)
Bulk CGT
H ⊥ ab
Experimental data
gapped spin wave fit ($T^{3/2}f_{3/2}$)
gapless spin wave fit ($T^{3/2}$)
$M_S \times 10^{-2}$ (emu)
T (K)
(b)
Few layered CGT
H ⊥ ab
Experimental data
gapped spin wave fit ($T^{3/2}f_{3/2}$)
gapless spin wave fit ($T^{3/2}$)
$M_S \times 10^{-5}$ (emu)
T (K)

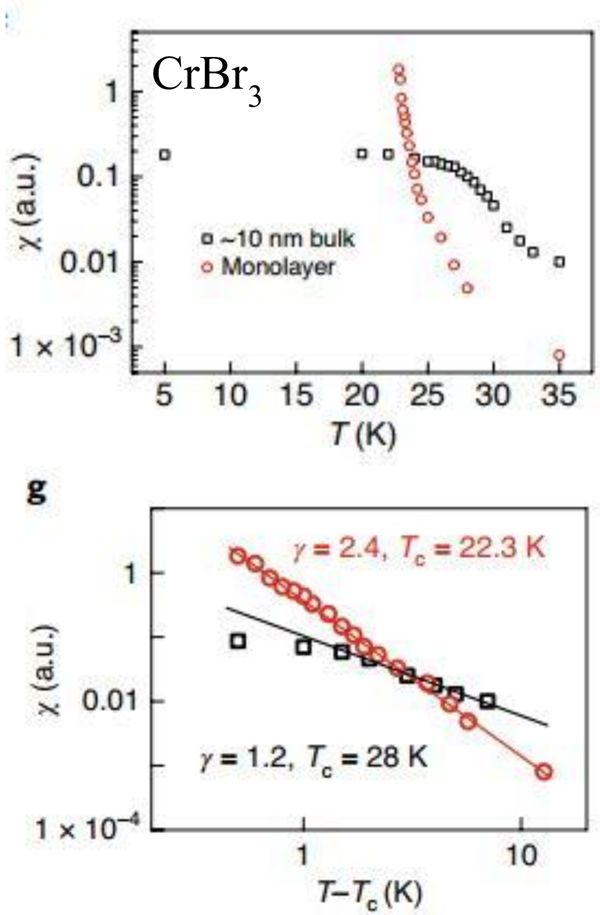


Strong dimensionality (or layer) dependence of T_C

Nature **volume 546**, 265–269 (2017)

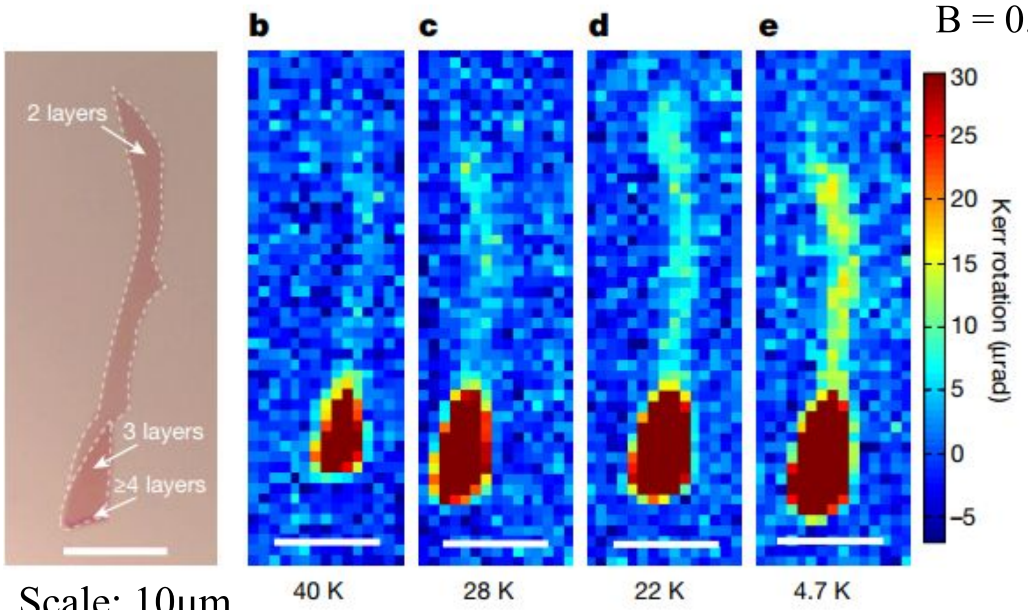
$$H=\frac{1}{2}\sum_{i,j} J_{ij}\mathbf{S}_i\cdot\mathbf{S}_j+\sum_i A(S_i^z)^2-g\mu_B\sum_i BS_i^z$$

Effect of dimensionality on critical exponents or, nature of interactions



Dimensional reduction causes a crossover from Mean field like behavior to a 2D Ising like scenario.

Nature Materials **volume 19**,1290–1294 (2020)



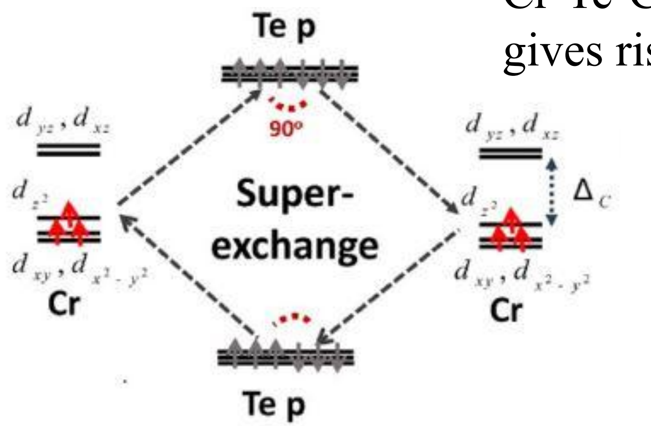
B = 0.075 T

Discovery of 2D magnetism in 2L CGT from MOKE study

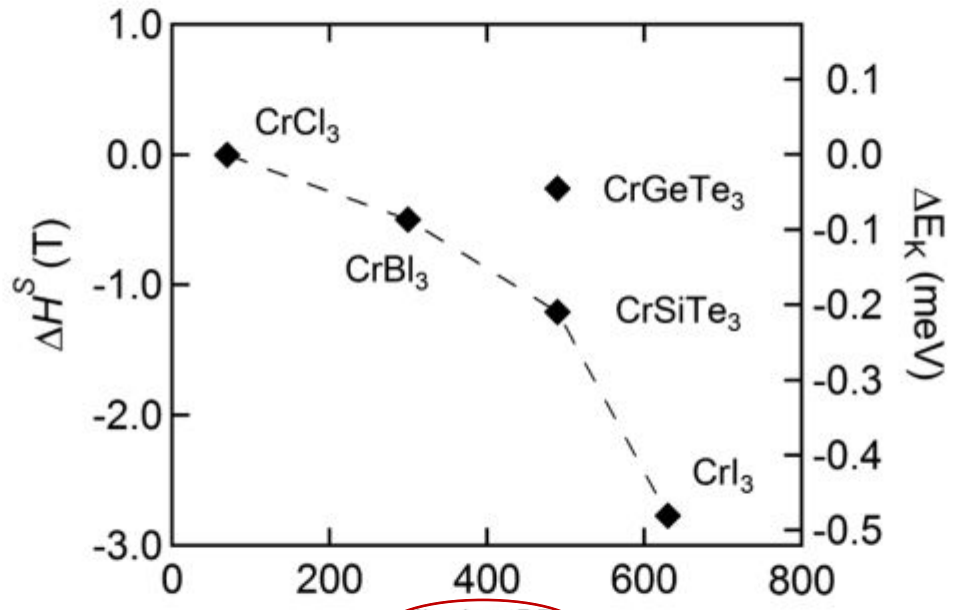
Scale: 10µm

Nature **volume 546**, 265–269 (2017)

Cr-Te-Cr 90^0 superexchange pathway gives rise to robust 2D-FM order



J. Am. Chem. Soc. 2019, 141, 17166-17173

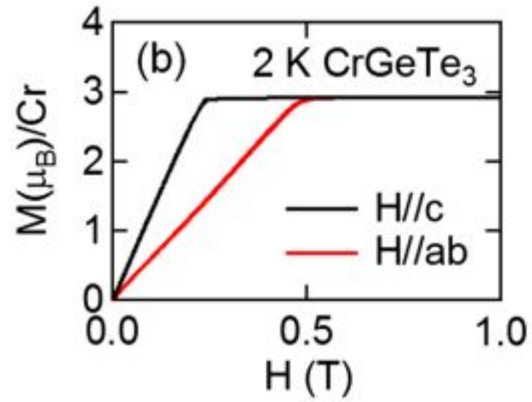


Ligand p spin orbit coupling strength

$$\Delta H^S = H_c^S - H_{ab}^S$$

$$\Delta E_K = 3\mu_B \Delta H^S$$

Cr^{+3} moment



PHYSICAL REVIEW LETTERS **122,** 207201 (2019)

Critical Exponents

Table 5.4.1. *Definition of critical exponents and amplitudes.*

Susceptibility	$\chi = \Gamma t^{-\gamma}$	$t > 0$
	$\chi = \Gamma'(-t)^{-\gamma'}$	$t < 0$
Specific heat	$C = \frac{A}{\alpha} t^{-\alpha}$	$t > 0$
	$C = \frac{A'}{\alpha}(-t)^{-\alpha'}$	$t < 0$
Correlation length	$\xi = \xi_0 t^{-\nu}$	$t > 0$
	$\xi = \xi_0'(-t)^{-\nu'}$	$t < 0$
Order parameter	$\langle\phi\rangle = B(-t)^{\beta}$	$t < 0$
	$\langle\phi\rangle = D_c^{-1} h\lvert h\rvert^{(1-\delta)/\delta}$	$t = 0$
Correlation function	$G(q) = D_\infty q^{-(2-\eta)}$	$t = 0$

From P. Chaikin and T Lubensk
"Principles of Condensed Matte
Physics"

Universality Class

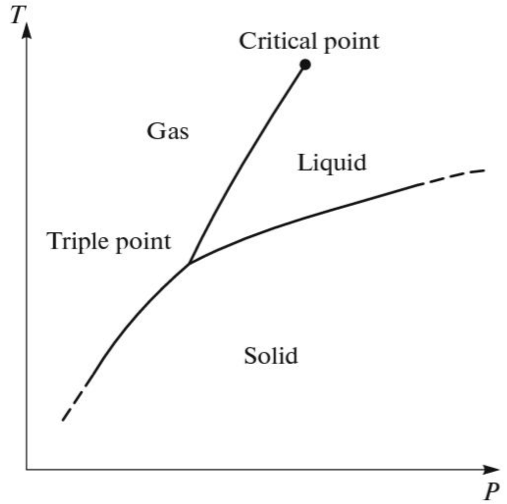
Table 5.4.2. *Some critical exponents from theory and experiment.*

Exponent	α	β	γ	ν	η
Property	specific heat	order parameter	susceptibility	coherence length	correlation function
Definition	$C \sim t^{-\alpha}$	$\langle\phi\rangle \sim t^{\beta}$	$\chi \sim t^{-\gamma}$	$\xi \sim t^{-\nu}$	$G(q) \sim q^{-2+\eta}$
Mean-field	0	0.5	1	0.5	0
3D theory					
$n = 0$ (SAW)	0.24	0.30	1.16	0.59	
$n = 1$ (Ising)	0.11	0.32	1.24	0.63	0.04
$n = 2$ (xy)	−0.01	0.35	1.32	0.67	0.04
$n = 3$ (Heisenberg)	−0.12	0.36	1.39	0.71	0.04
Experiment					
3D $n = 1$	$0.11^{+.01}_{-.03}$	$0.32^{+.16}_{-.04}$	$1.24^{.16}_{-.04}$	$0.63^{+.04}_{-.04}$	0.03 - 0.06
3D $n = 3$	$0.1^{+.05}_{-.04}$	$0.34^{+.04}_{-.04}$	$1.4^{+.07}_{-.07}$	$0.7^{+.03}_{-.03}$	
2D $n = 1$	$0.0^{+.01}_{-.003}$	$0.3^{+.04}_{-.04}$	$1.82^{+0.7}_{-.07}$	$1.02^{+.07}_{-.07}$	

Experiments on 3D $n = 1$ compiled from liquid-gas, binary fluid, ferromagnetic, and antiferromagnetic transitions.
Experiments on 3D $n = 3$ transitions compiled from some ferromagnetic and antiferromagnetic transitions.
Experiments on 2D $n = 1$ complied from some antiferromagnetic transitions.

From P. Chaikin and T Lubensky "Principles of Condensed Matter Physics"

Theory suggests that the class (i.e. set of exponents) depends on spatial dimensionality, symmetry of the order parameter and interaction (and range of the latter as well) but not on the detailed form or strength of the interactions

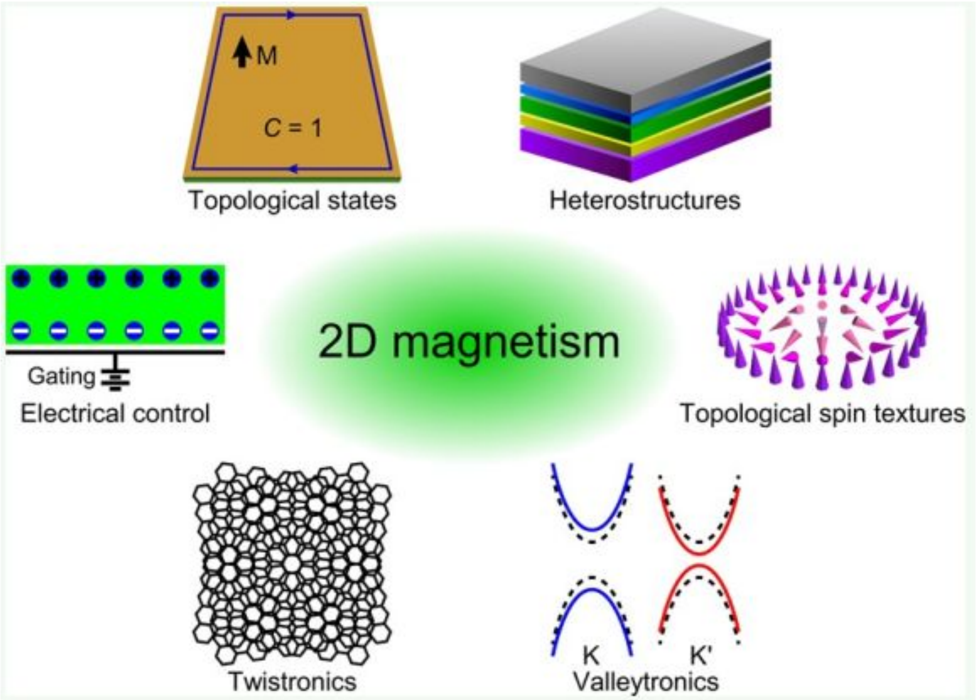


Reduced Temp : $t = (T - T_c)/T_c$

Specific heat exponent : $C \sim t^{-\alpha}$

Order Parameter exponent $< |\phi| > \sim t^{\beta}$

Emergent rich phenomena in 2D system



Magnetism

Dimensional reduction from 3D

↓

<u>In 2D</u>
Significant enhancement in the effects
Of thermal fluctuations

↓

Complex magnetic phenomena at low D
Dependent on the Dimensionality of spins (n)

Plan

- A brief discussion about relevant features of 2D magnetism for our work
- Why we choose the materials
- What are we interested in exploring
- Magnetic Features in the bulk system
- Features as you lower dimensionality
- Some understanding of our results and its consequences
- Ongoing work
- Summary