Magnetic Skyrmion Materials

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THE UNIVERSITY OF WARWICK

SPICE Quantum Functionalities of Nanomagnets

June 2025

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EPSRC
Engineering and Physical Sciences Research Council

THE SKYRMION PROJECT

Durham University

WARWICK THE UNIVERSITY OF WARWICK

UNIVERSITY OF OXFORD

UNIVERSITY OF CAMBRIDGE

UNIVERSITY OF Southampton

Science & Technology Facilities Council ISIS Neutron and Muon Source

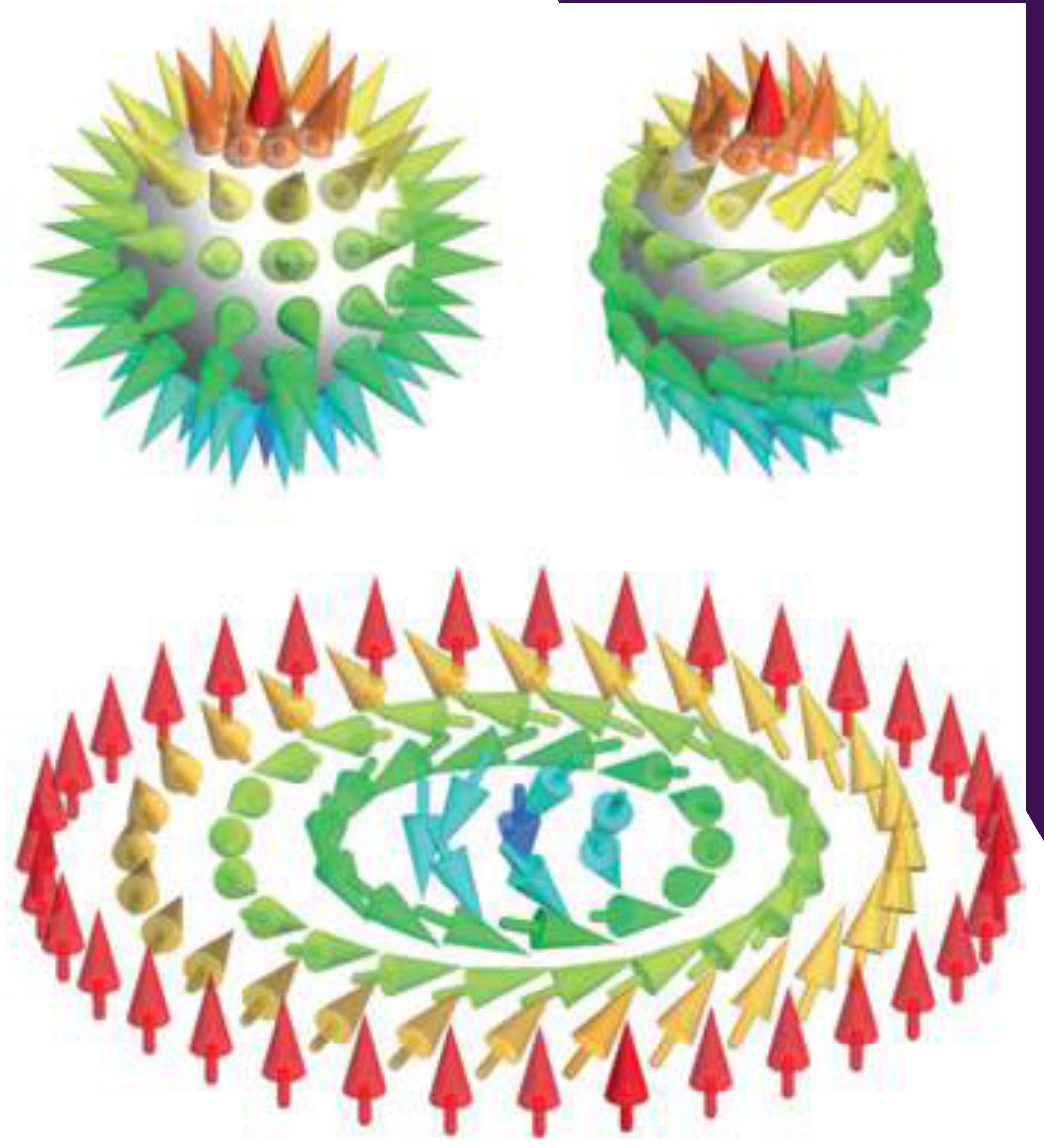
NEUTRONS FOR SOCIETY

https://go.warwick.ac.uk/supermag

SPICE 2025

Skyrmionic materials

Material	Symmetry	Type	Tc (K)	
MnSi	B20-*P*213	**Chiral**	**30**	**Metal**
FeGe	B20-*P*213	**Chiral**	**278**	**Metal**
Cu_2OSeO_3	B20-*P*213	**Chiral**	**58**	**Insulator**
Co-Zn-Mn	*P*4132	**Chiral**	**280-400**	**Metal**
GaV_4S_8	*C*3v	**Polar**	**13**	**Insulator**

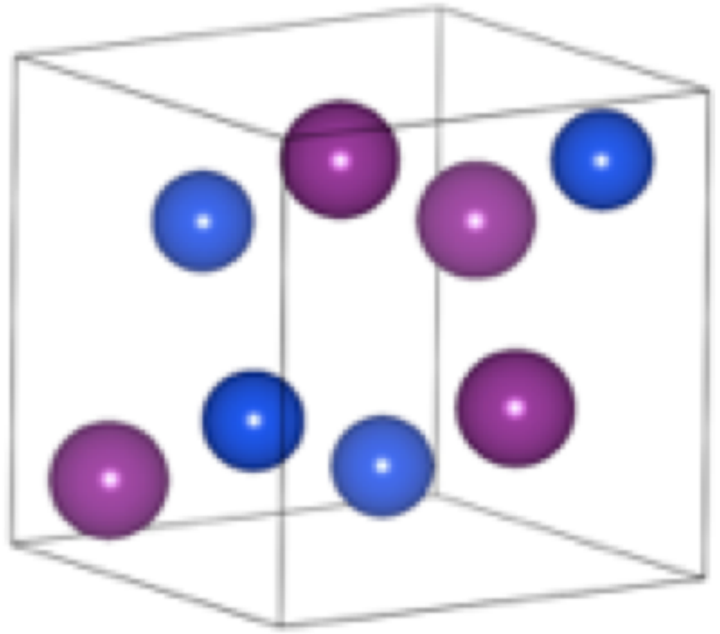


-The interplay between the *Dzyaloshinskii-Moriya interaction* (DMI) and the short range exchange interaction cants the spins on an intermediate length scale
-Produces an extraordinary spin texture comprising a chiral, topologically stable magnetic whirl.

Skyrmions in Chiral Cubic lattices

B20 Alloys (Metal)

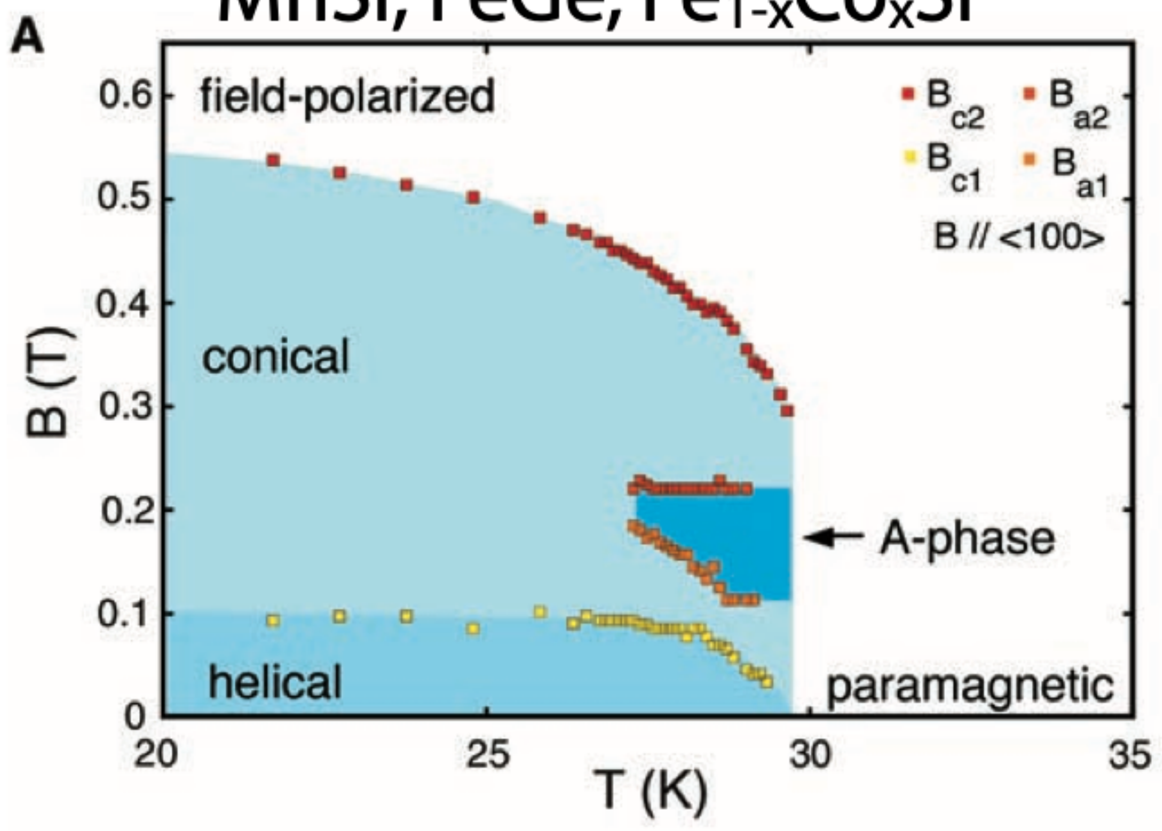
$P2_13$



T_c ~ 280 K

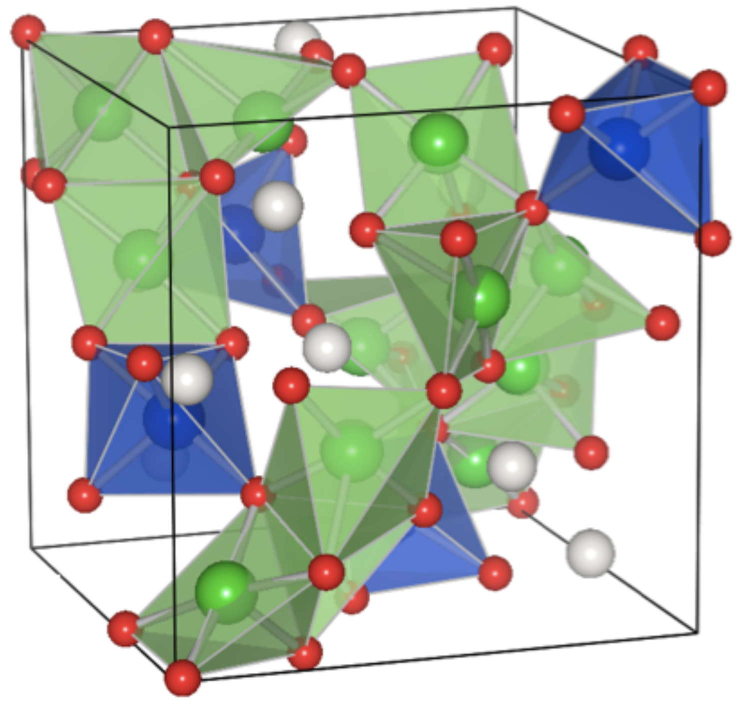
Muhlbauer *et al.*, Science (2009).
X. Z. Yu *et al.*, Nature (2010).

MnSi, FeGe, $Fe_{1-x}Co_xSi$



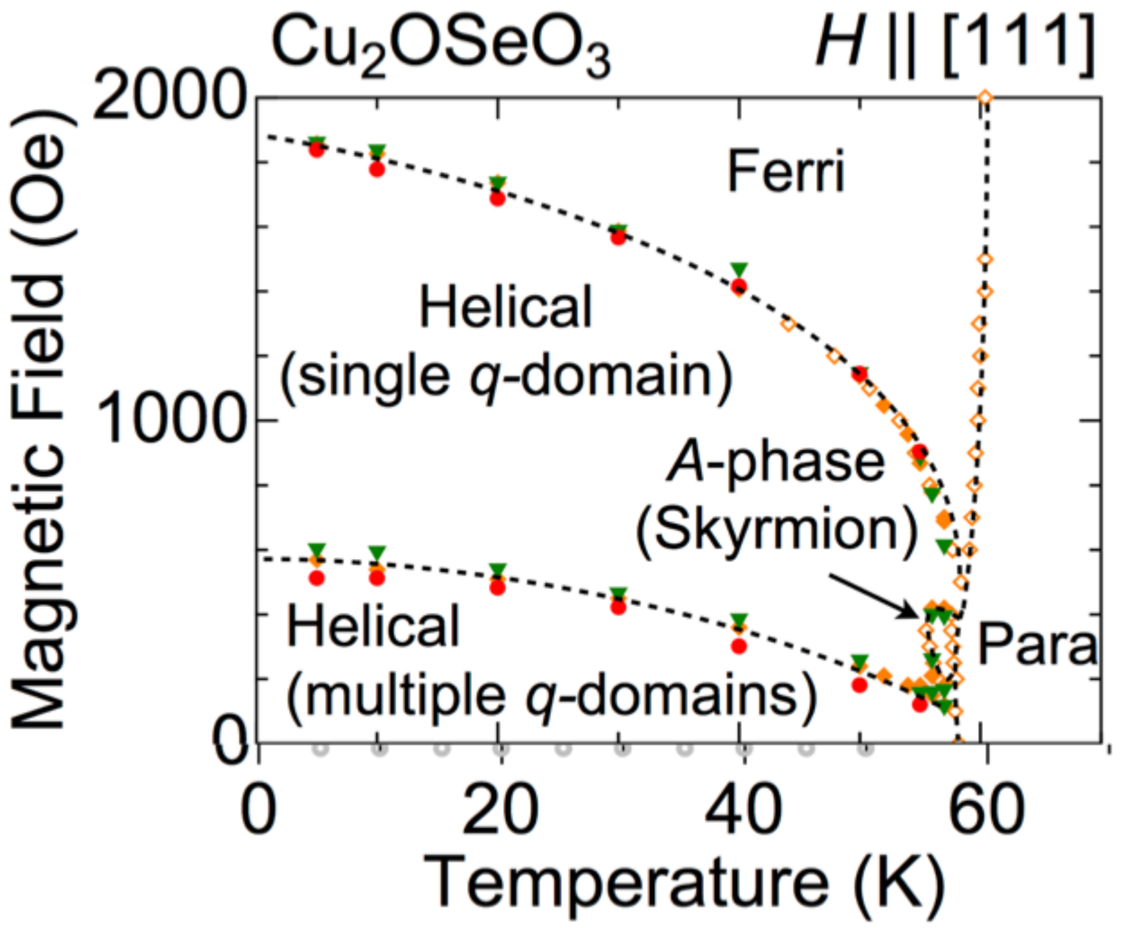
Cu_2OSeO_3 (Insulator)

$P2_13$



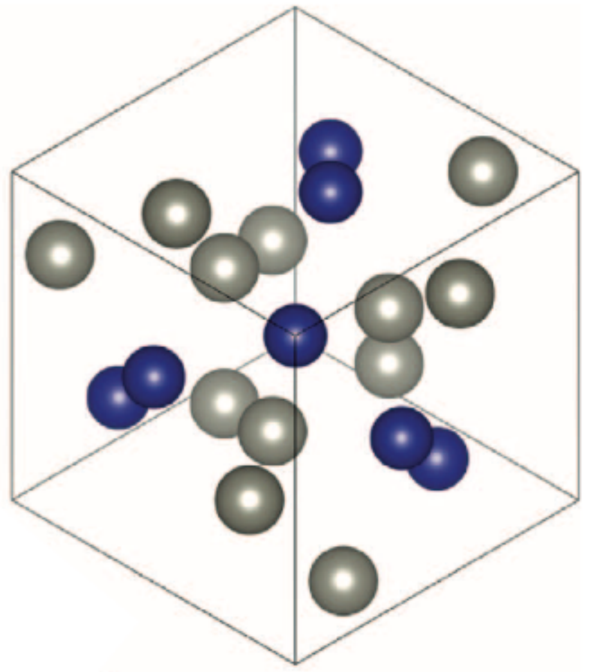
T_c ~ 58 K

Seki *et al.*, Science (2012).



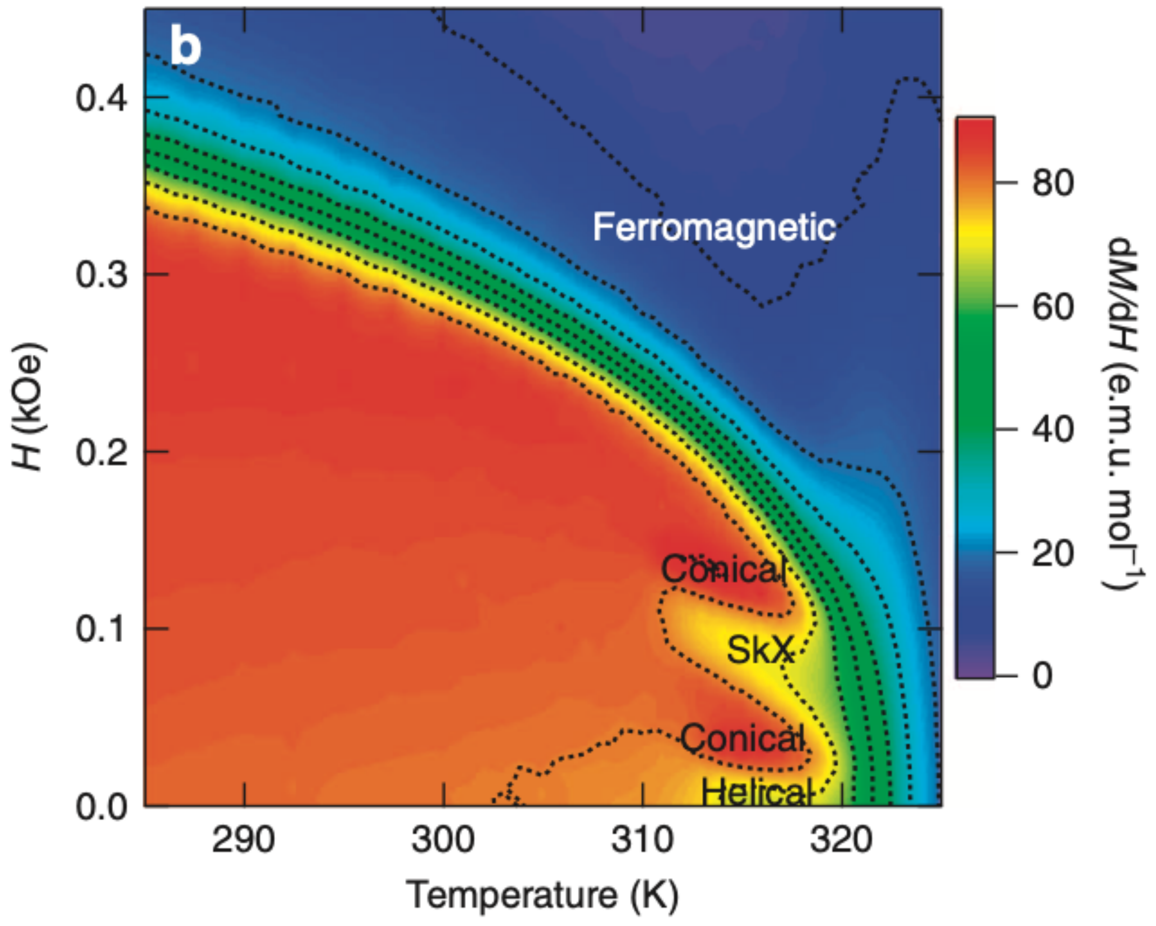
Co-Zn-Mn Alloys (Metal)

$P4_132$



T_c ~ 400 K

Y. Tokunaga *et al.*,
Nat. Comm. (2015).



S. Seki *et al.*, *Science* **336** (2012) 198–201

Materials of Interest

Frustrated/intermetallic magnets

RE_2PdSi_3 (RE=Gd,Tb..)
$GdRu_2Si_2$
$GdRu_4Al_{12}$

Layered/2D magnets

Fe_3GeTe_2, Fe_5GeTe_2, Fe_3GaTe_2

Cu_2OSeO_3 Nanoparticles

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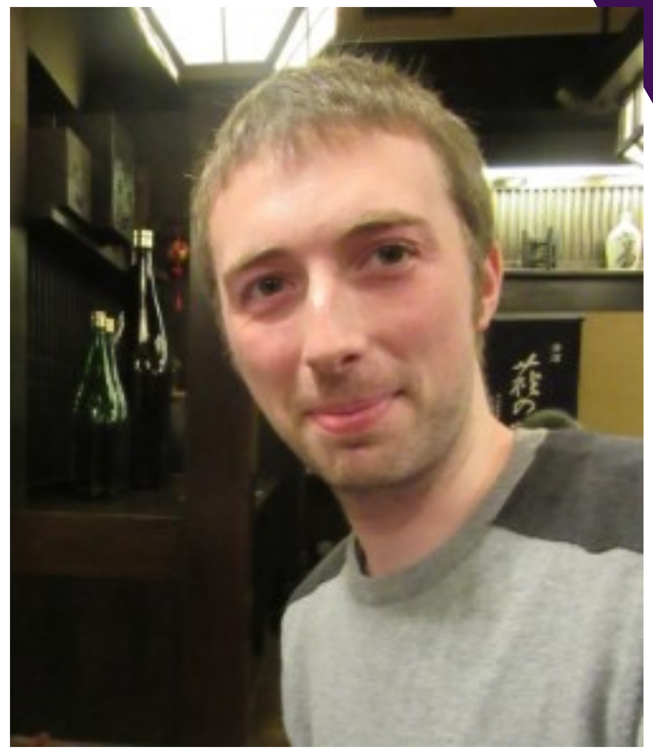
Frustrated/intermetallic magnets

RE_2PdSi_3 (RE=Gd,Tb..)
$GdRu_2Si_2$
$GdRu_4Al_{12}$

George Wood

Sam Holt



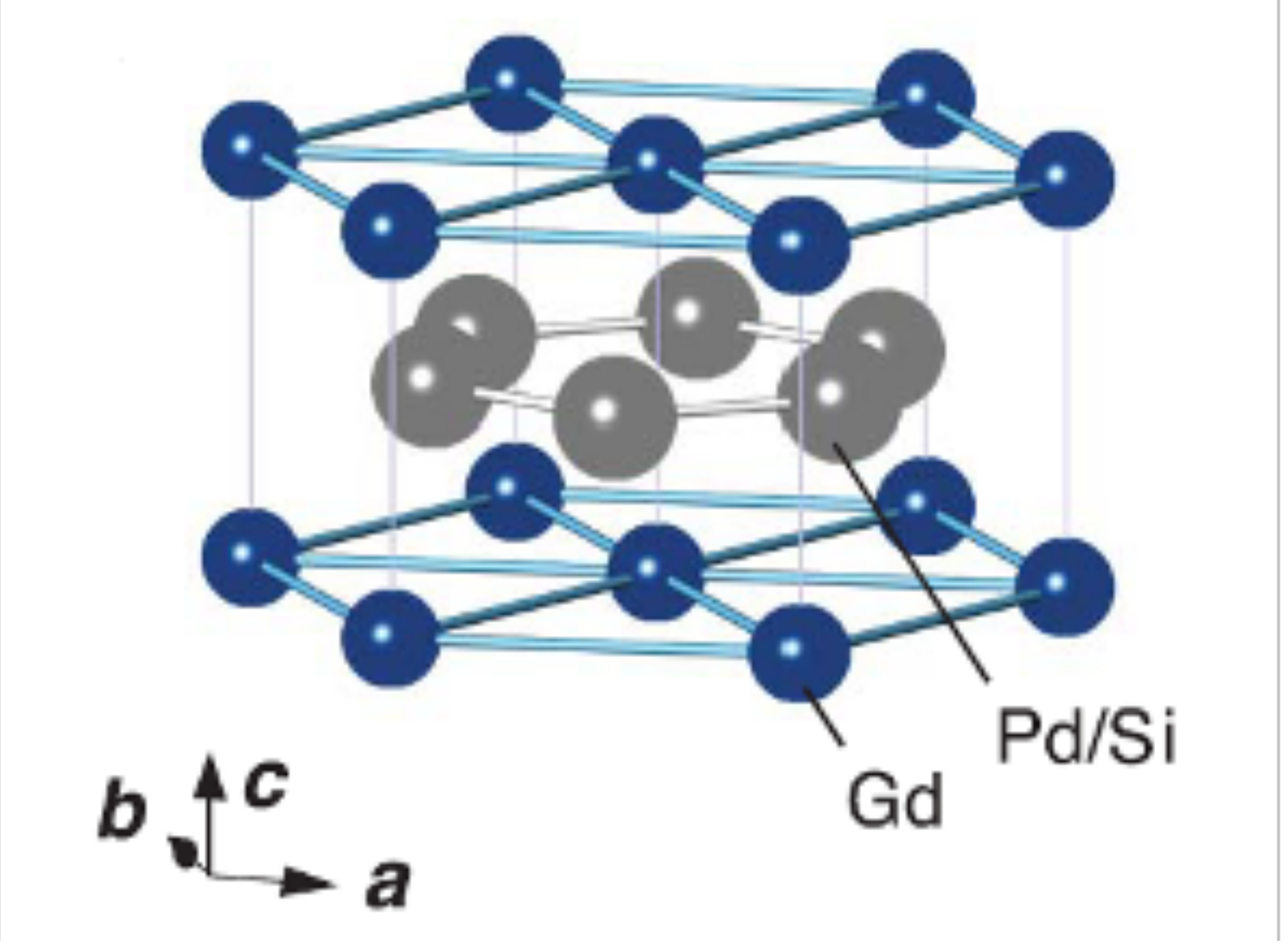
Ales Stefancic

Daniel Mayoh

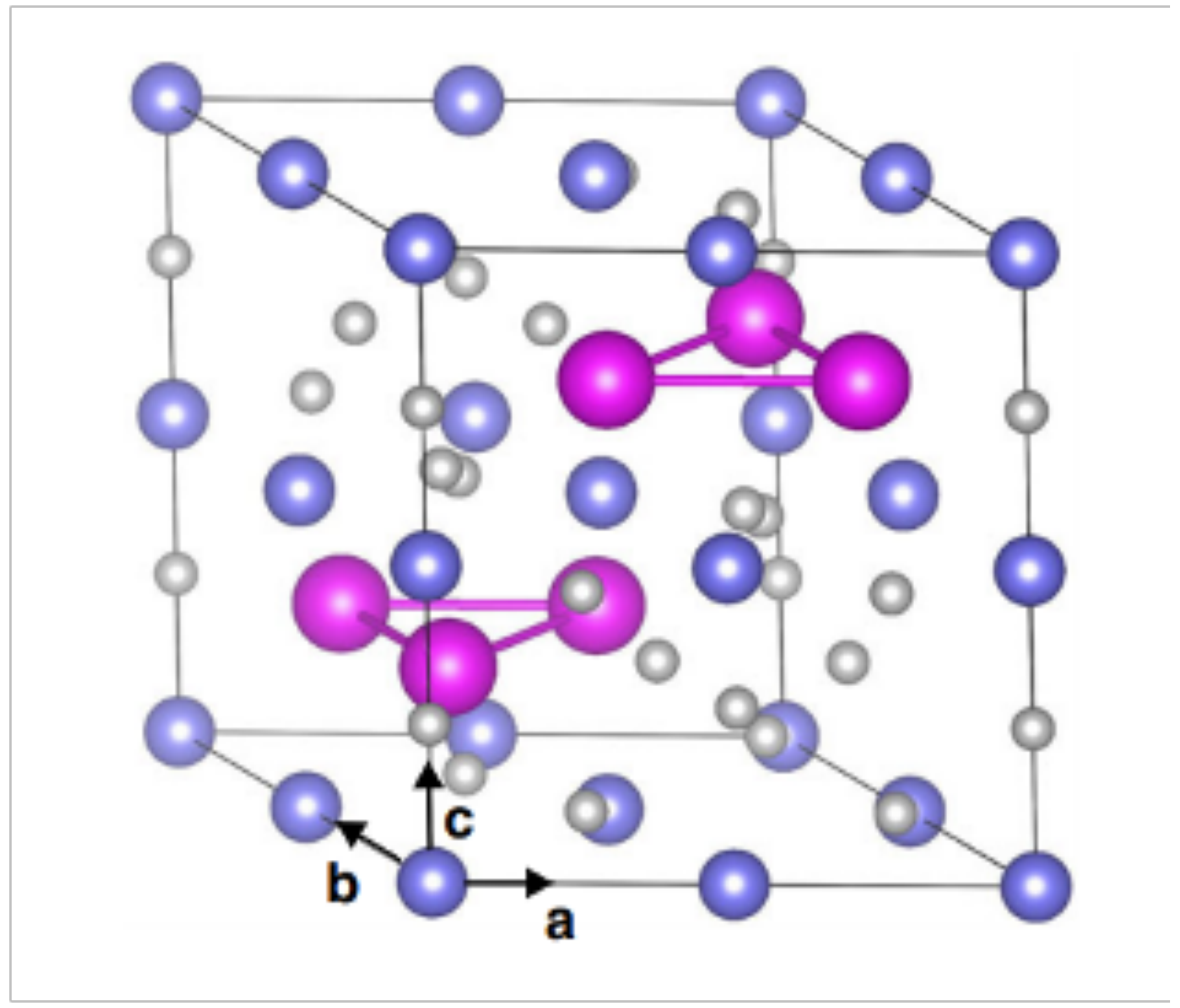
And UG project students

Skyrmions: Intermetallic Magnetic materials

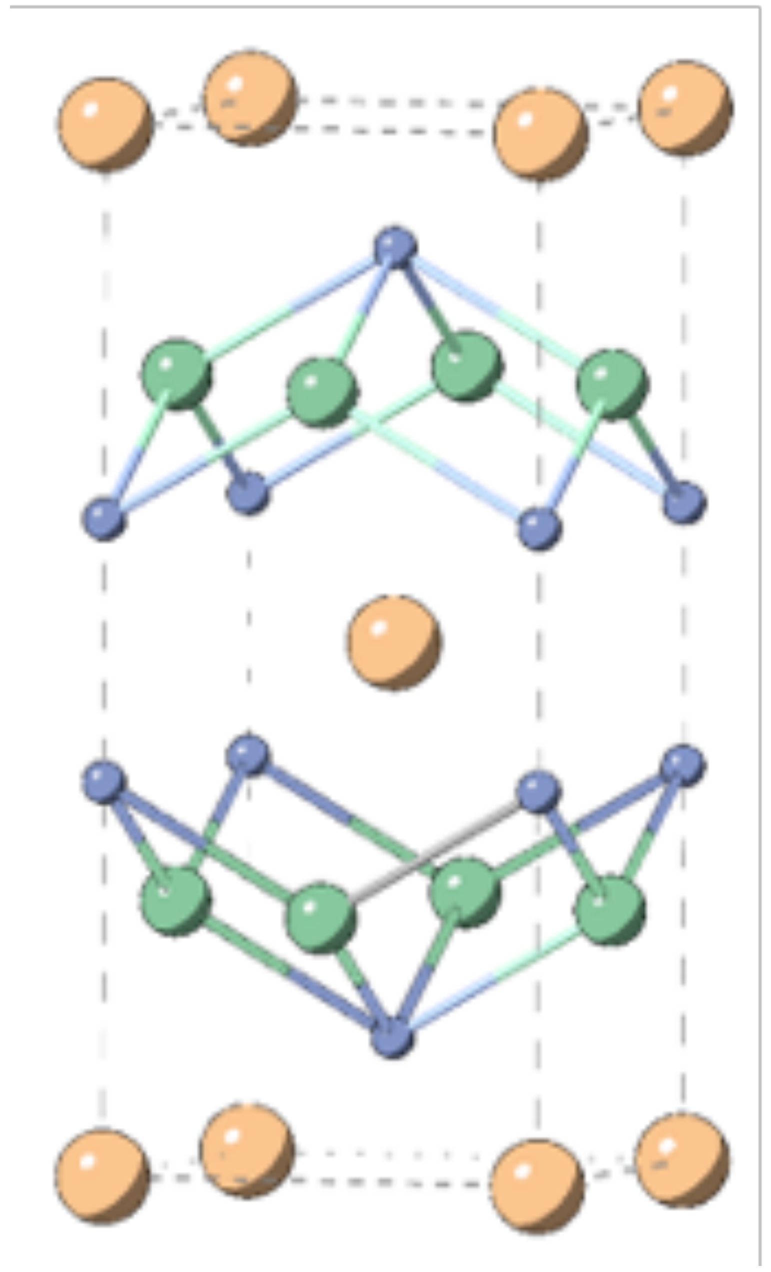
Gd_2PdSi_3



$Gd_3Ru_4Al_{12}$



$GdRu_2Si_2$



Possible Mechanism:

1.FM and AFM interactions in geometrical frustration.

A. Leonov and M. Mostovoy, Nat. Commun. 6, 8275 (2015).

2.RKKY and four spin interaction mediated by itinerant electrons.

R. Ozawa, S. Hayami, Y. Motome, PRL 118, 147205 (2017).

3.Inter-orbital frustration between Gd 5d and Gd 4f.

T. Nomoto, T. Koretsune, R. Arita, PRL 125, 117204 (2020).

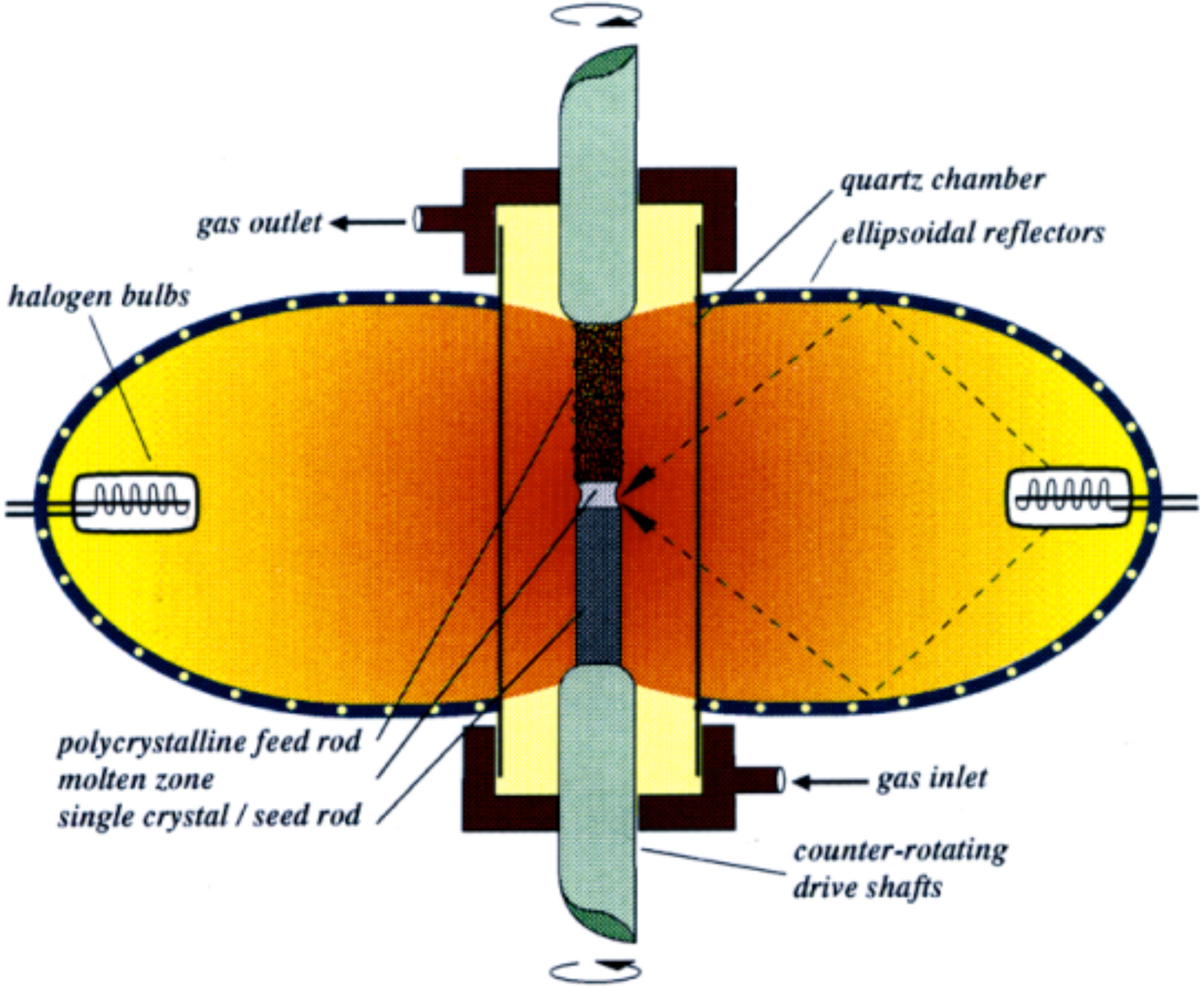
Crystal Growth using Optical Furnaces

Floating Zone Technique

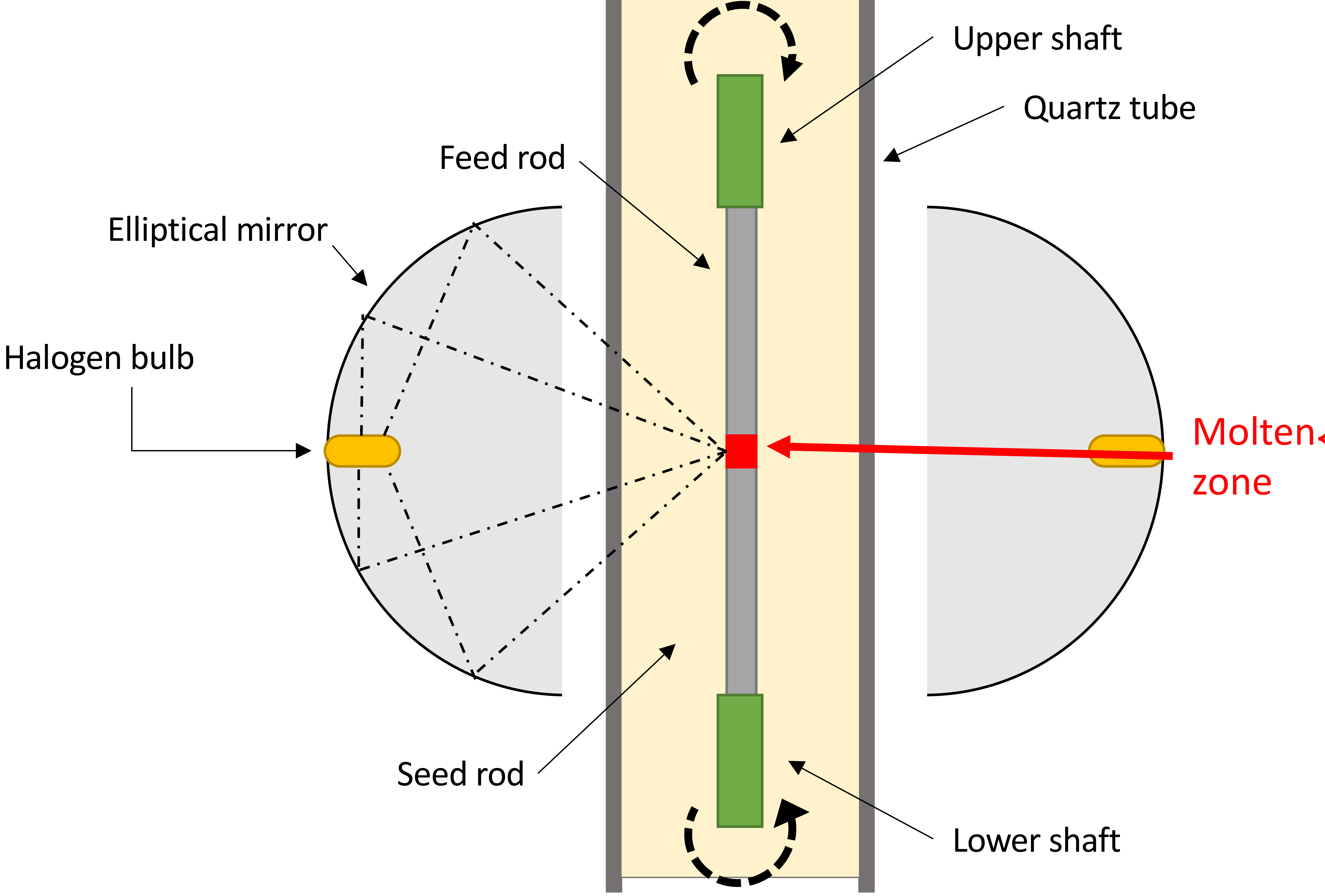
2 Mirror Halogen Lamp Furnace
4 Mirror Halogen Lamp Furnace
4 Mirror Xenon Lamp Furnace

Temperature ranges covered:

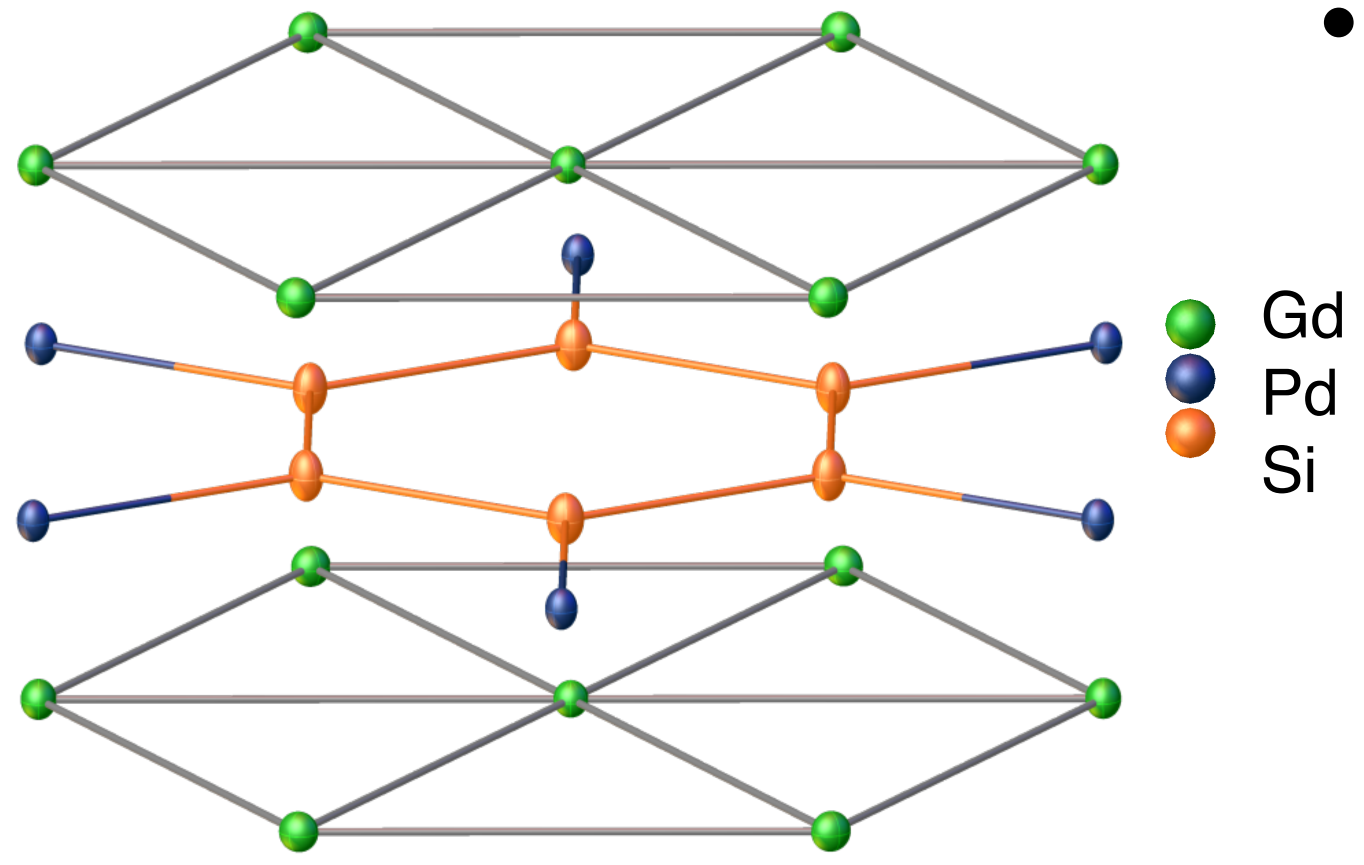
Maximum of 2150°C to 2800°C



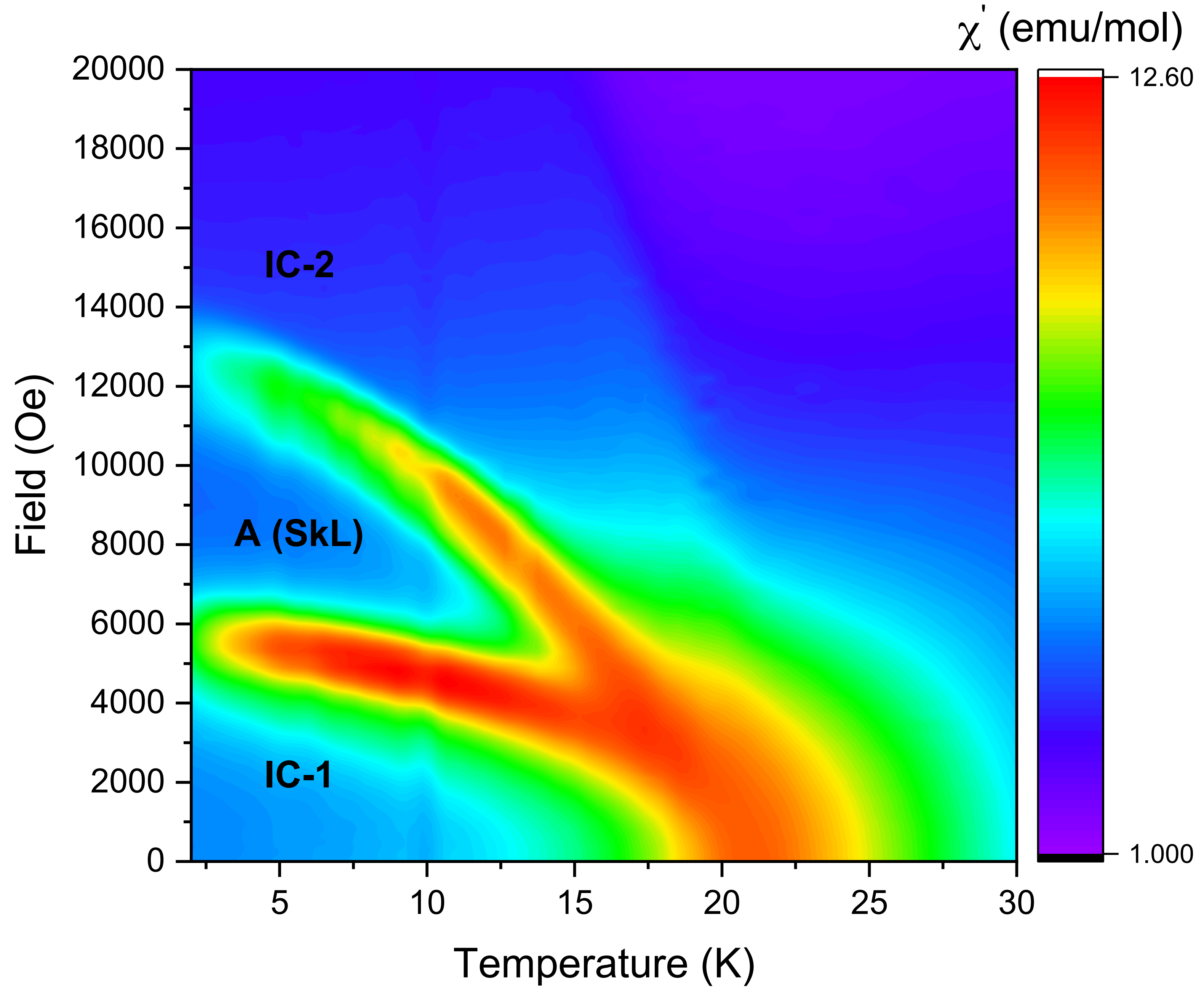
Floating Zone

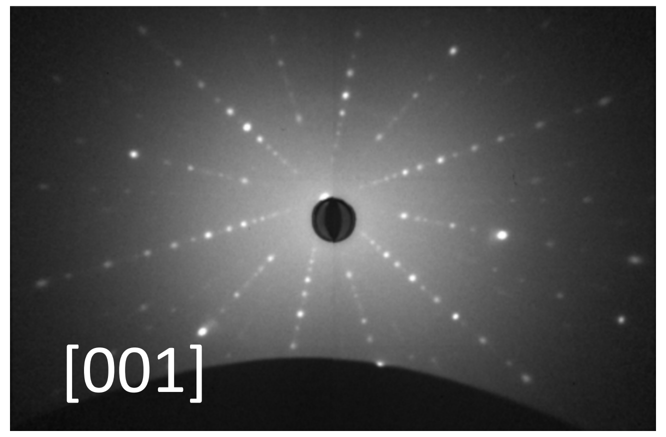


Gd_2PdSi_3 Crystal Growth, Frustration and Skyrmions



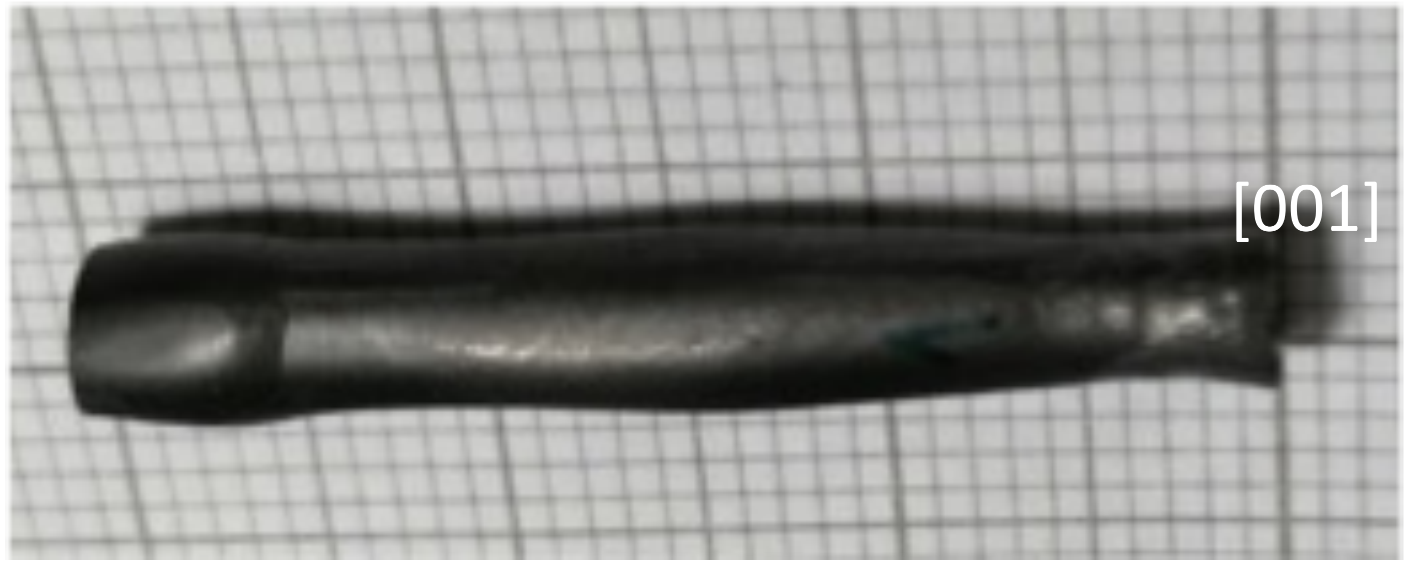
- Centrosymmetric materials
  - Triangular ordering of the magnetic rare earth atoms leads to strong magnetic frustration.
    - First known material where skyrmions are stabilised by magnetic frustration
      - Skyrmion sizes are very small in frustrated magnets (~2-3 nm)
        - Skyrmions stabilised over a large H-T region (< 20 K and 0.5 to 1.5 T)

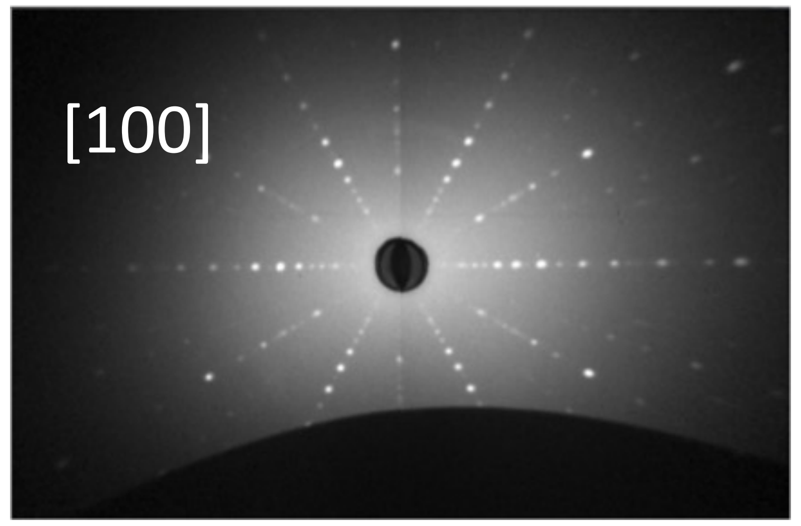




Kurumaji et al Science 365 914-918 (2019)

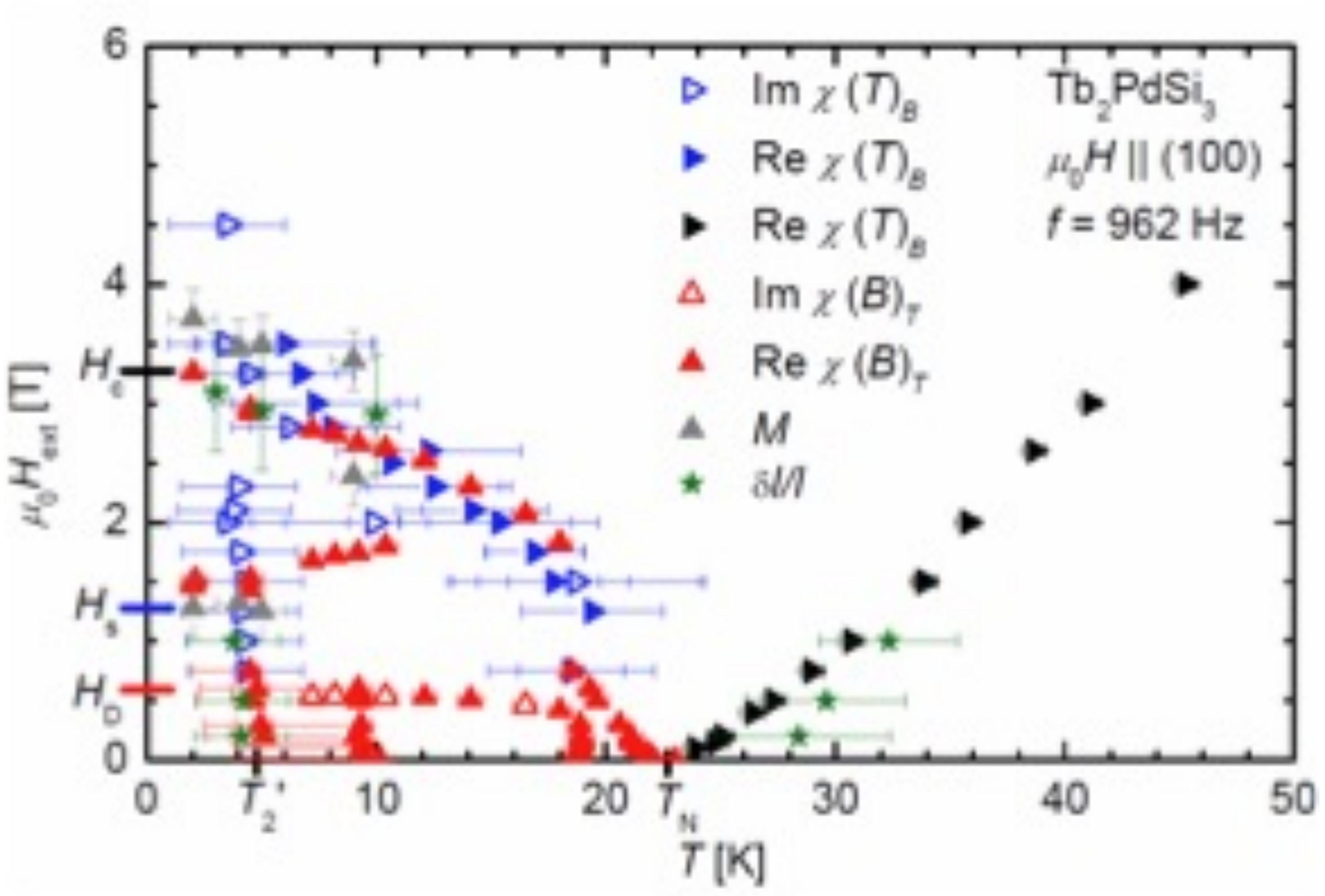
RE_2PdSi_3 (RE= Tb)



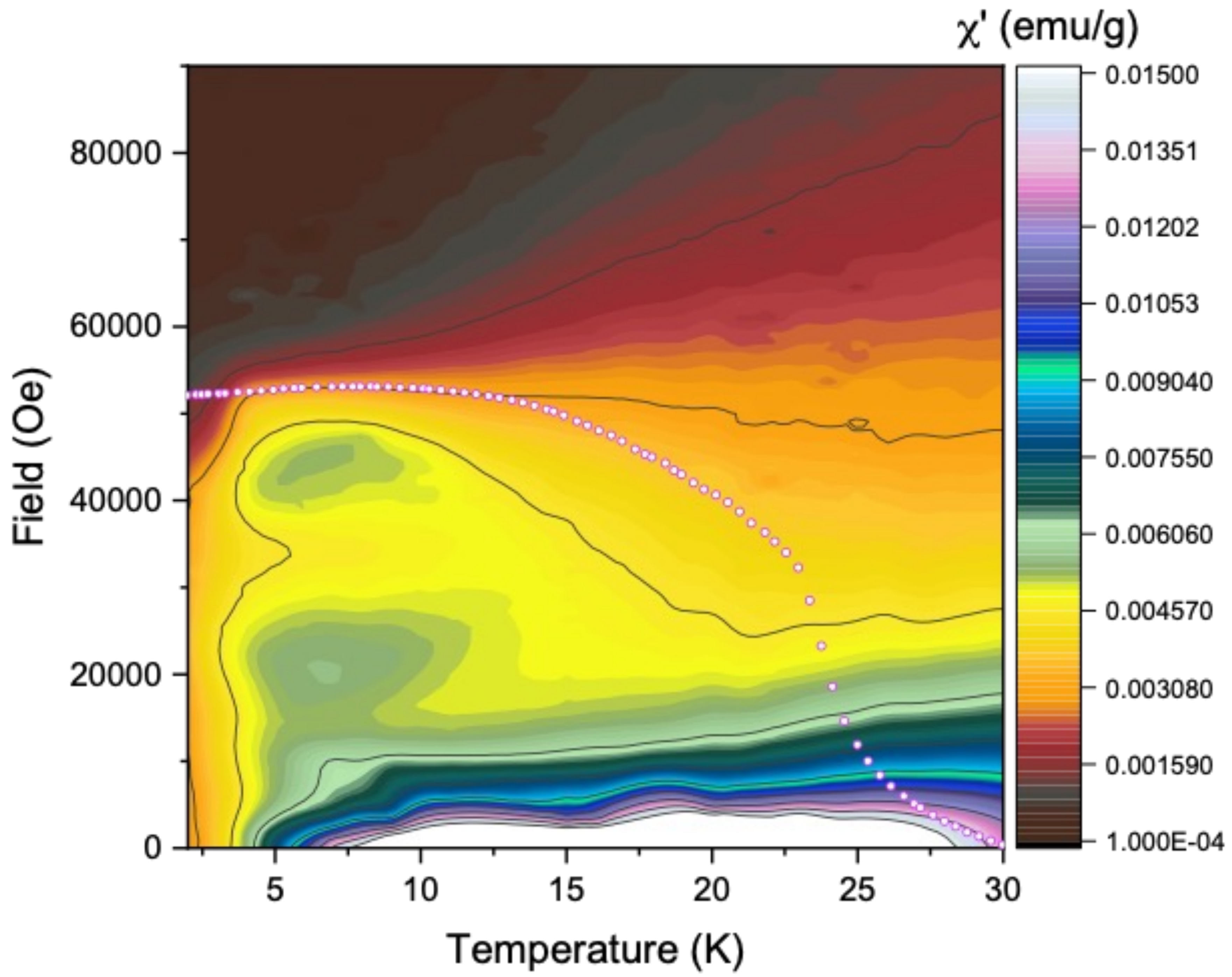


Large high quality single crystals of Tb_2PdSi_3 grown using the optical floating zone technique

Tb_2PdSi_3



A. Szytula *et al.* J. Magn. Magn. Mater. **202** (1999)



T_N = 24 K with 8 distinct states reported to exist at varying temperatures and magnetic fields below T_N

R_2PdSi_3 (R= Gd, Tb, Nd, Ho, Ce and Er)

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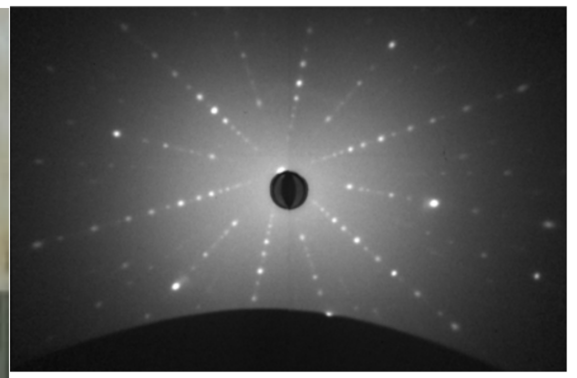
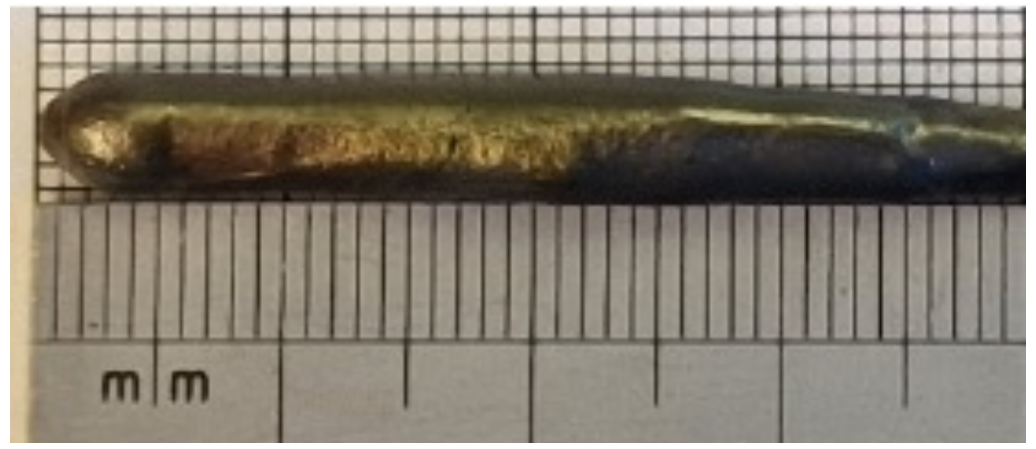
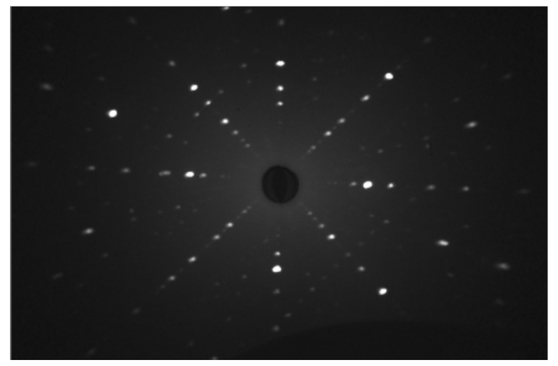
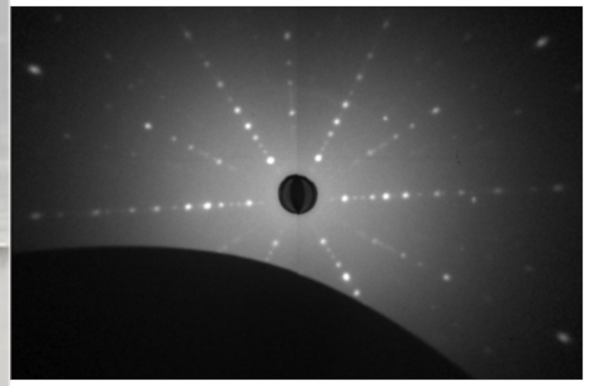


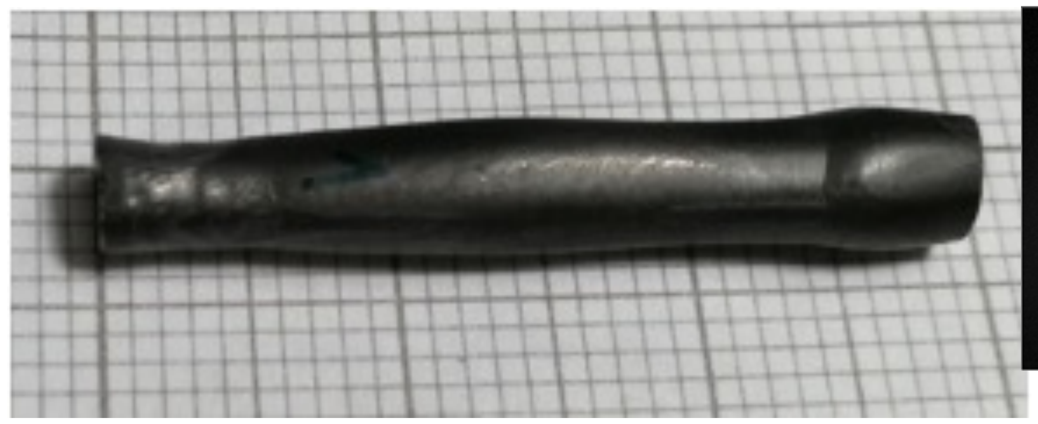
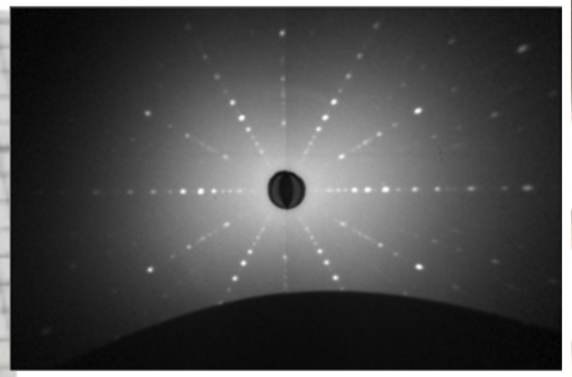
Gd_2PdSi_3

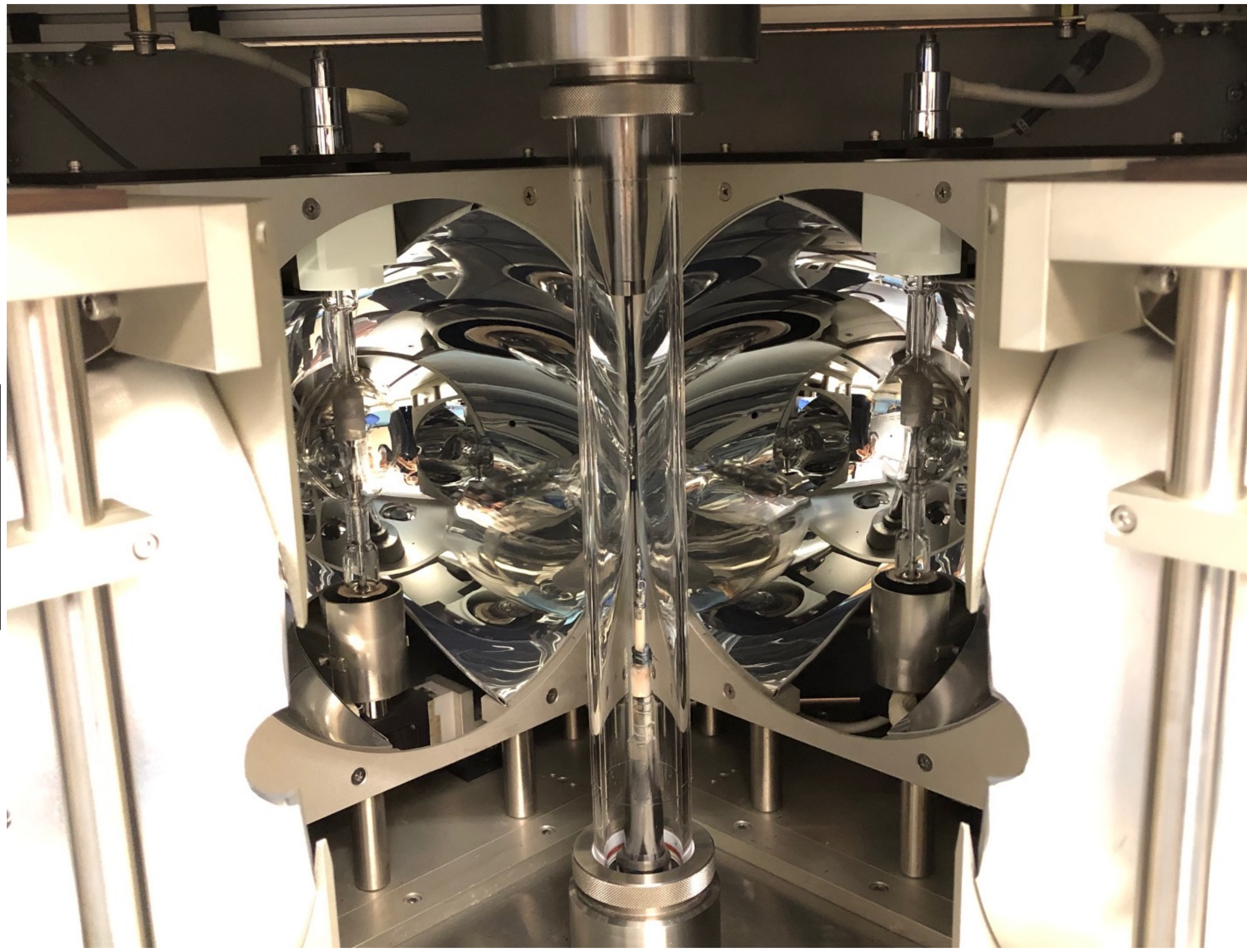
Ce_2PdSi_3

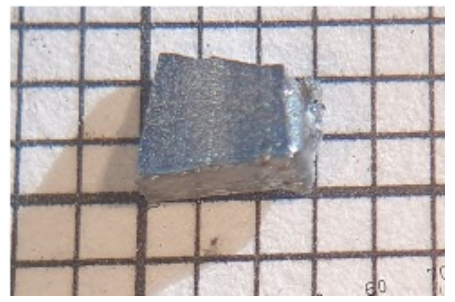
Ho_2PdSi_3

Tb_2PdSi_3



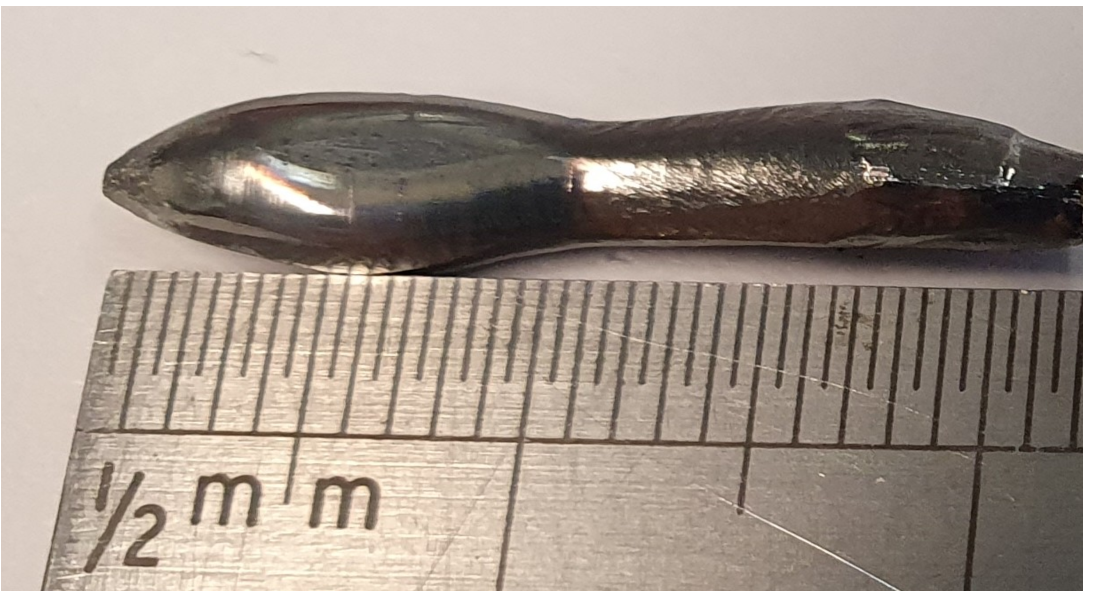
Nd_2PdSi_3

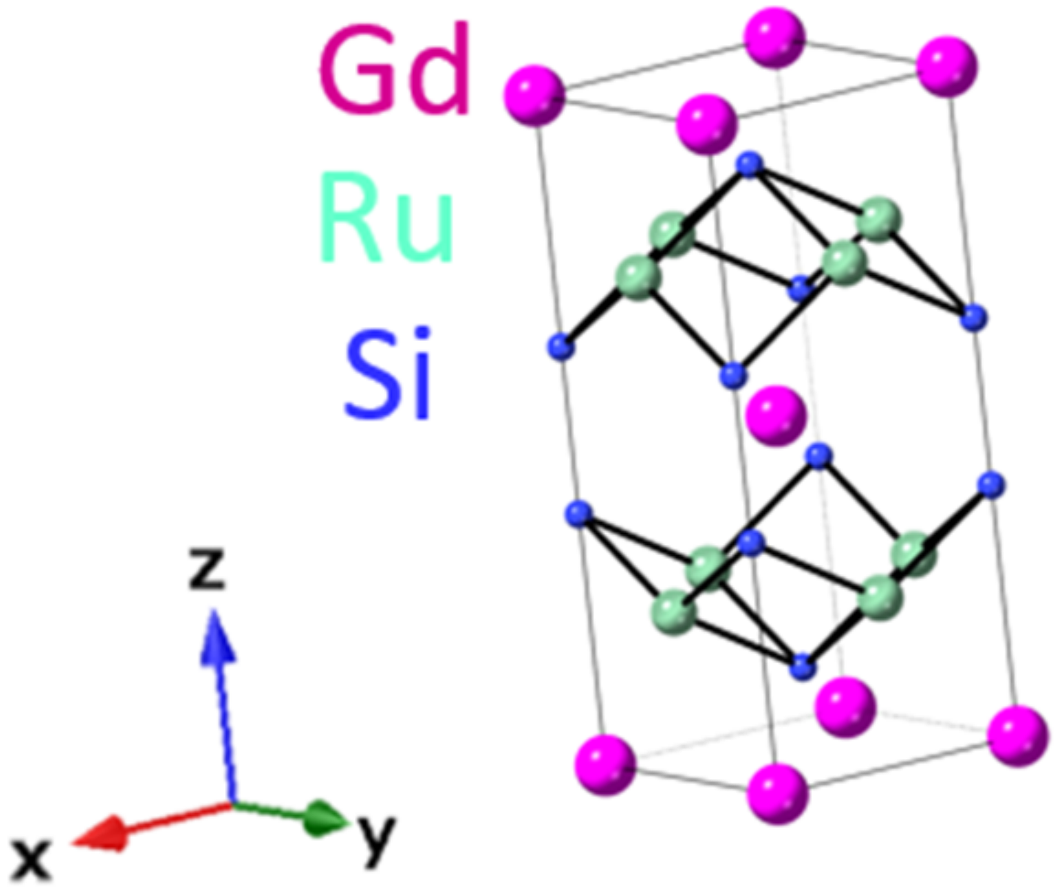



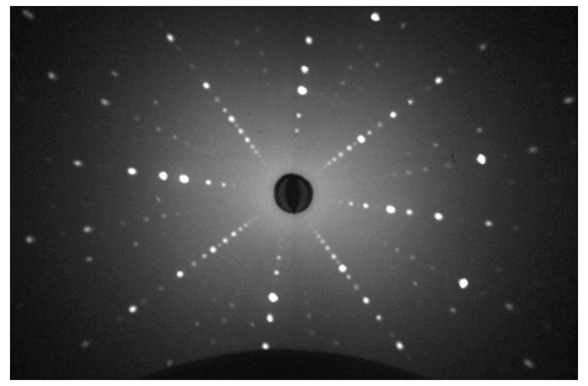
Er_2PdSi_3

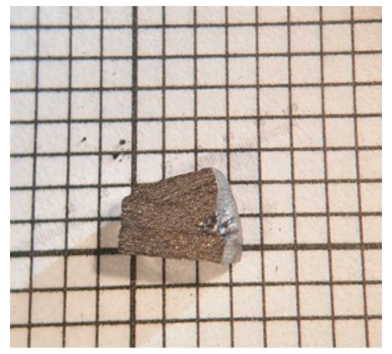
D A Mayoh et al Journal of Crystal Growth 642 (2024) 127774

GdRu$_2$Si$_2$

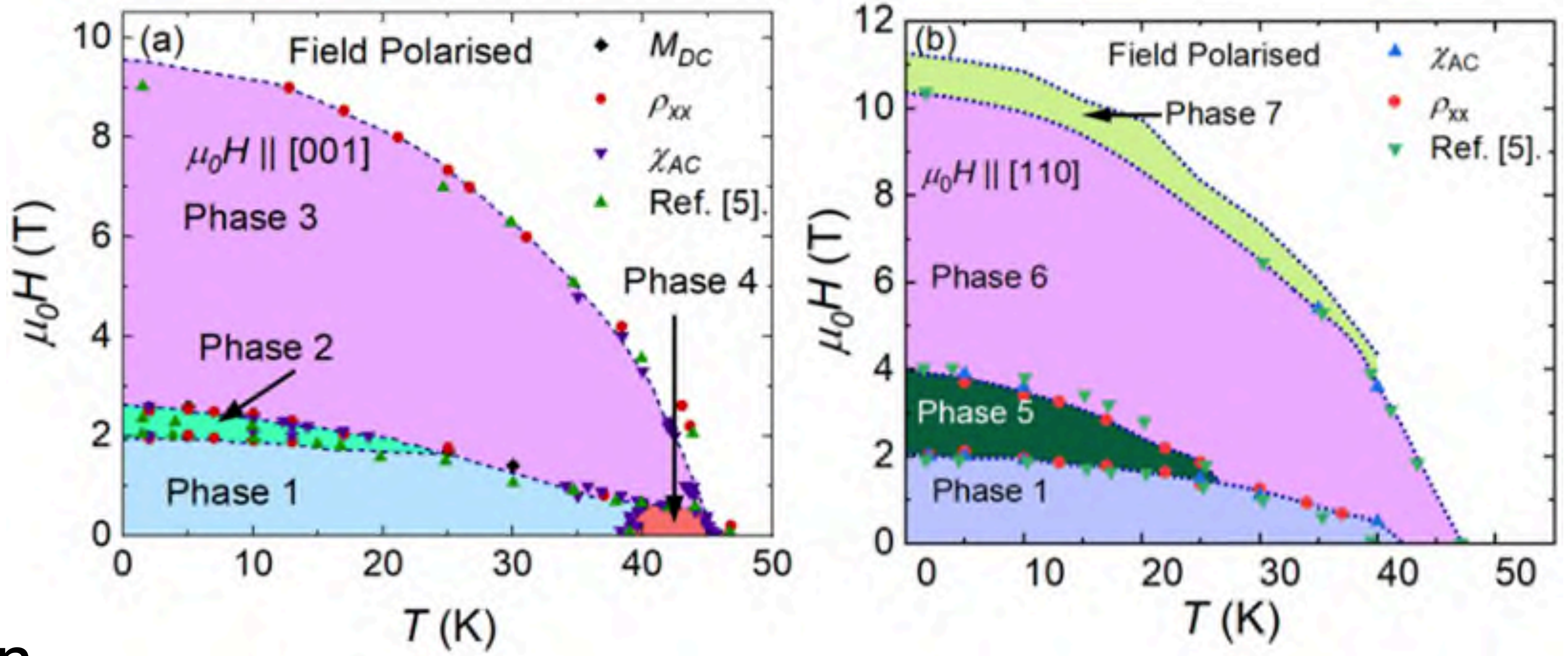








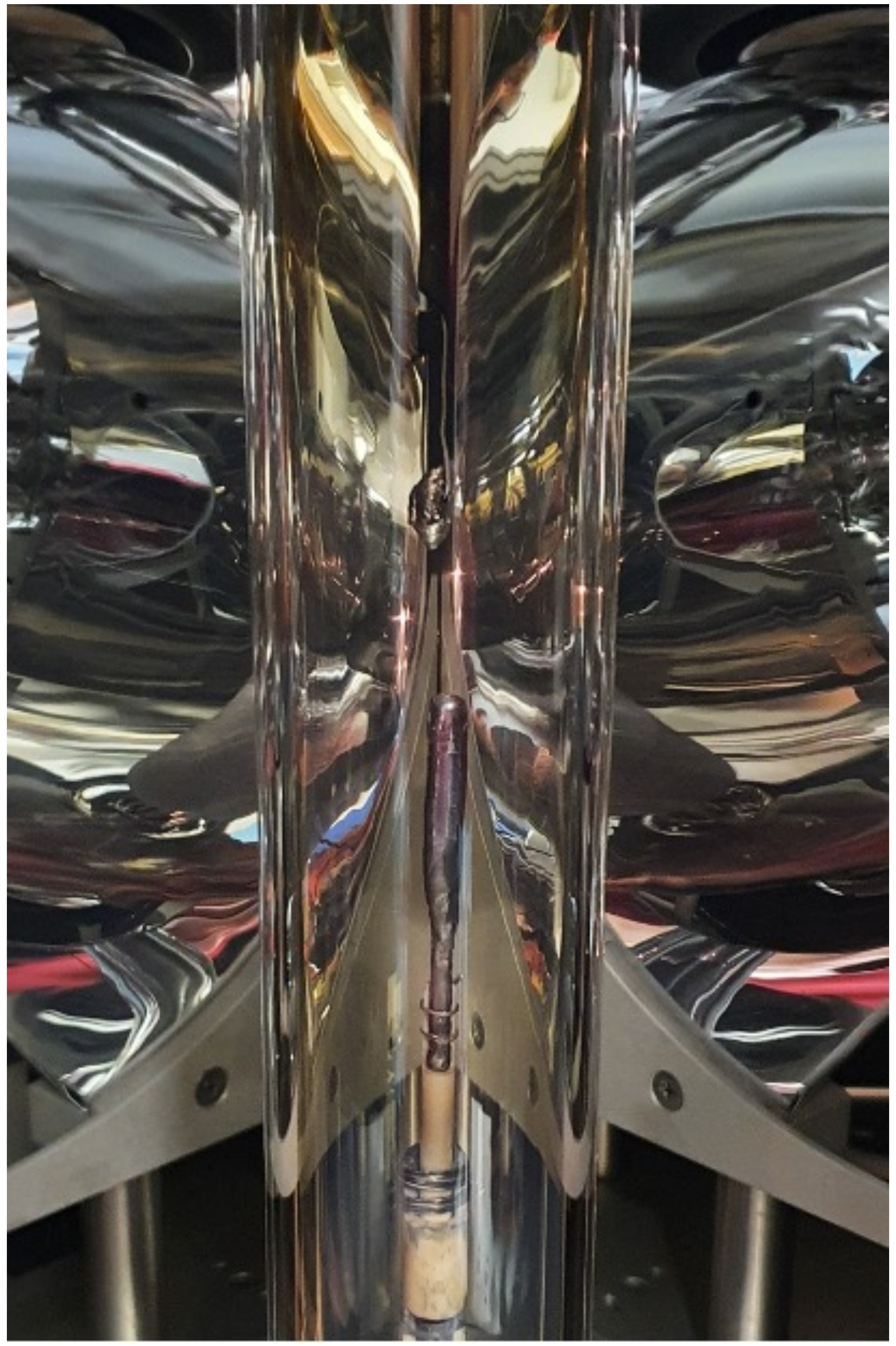
Space group I4/mmm

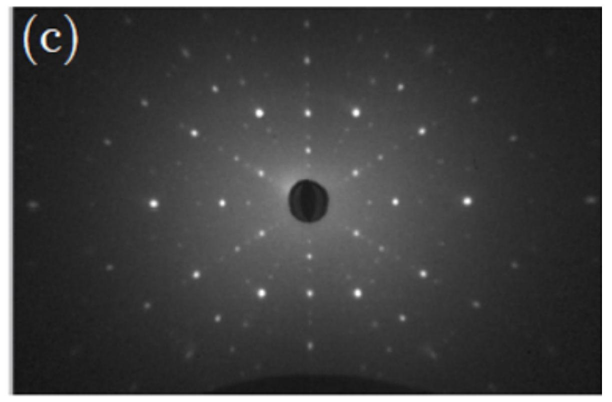


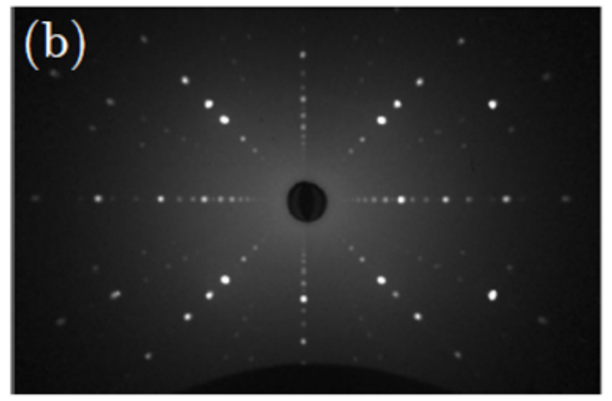
G.A. Wood PhD Thesis, Warwick 2024
Khanh et al Advanced Science, 2105452 (2022);
Nat. Nano. **15**, 444 (2020)

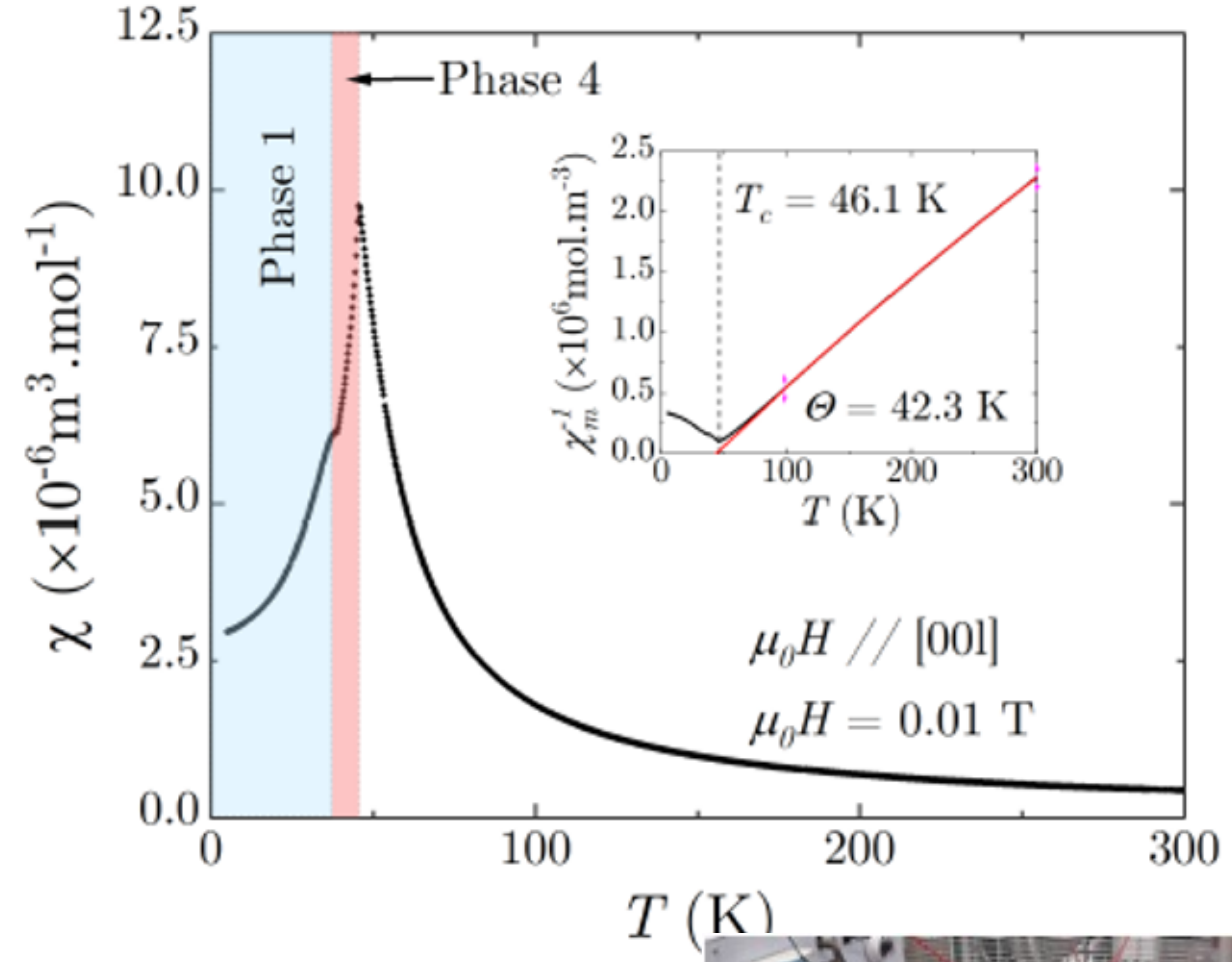
- Centrosymmetric, no magnetic frustration
- Alternating square lattice Gd layers and Ru$_2$Si$_2$ layers
- Skyrmions result due to the RKKY interaction
- Skyrmions stabilised for field along [001]
- *H-T* Phase diagrams (produced at Warwick) confirm that it is very different for field along [110]
- New Phase (4) identified for field along [001]

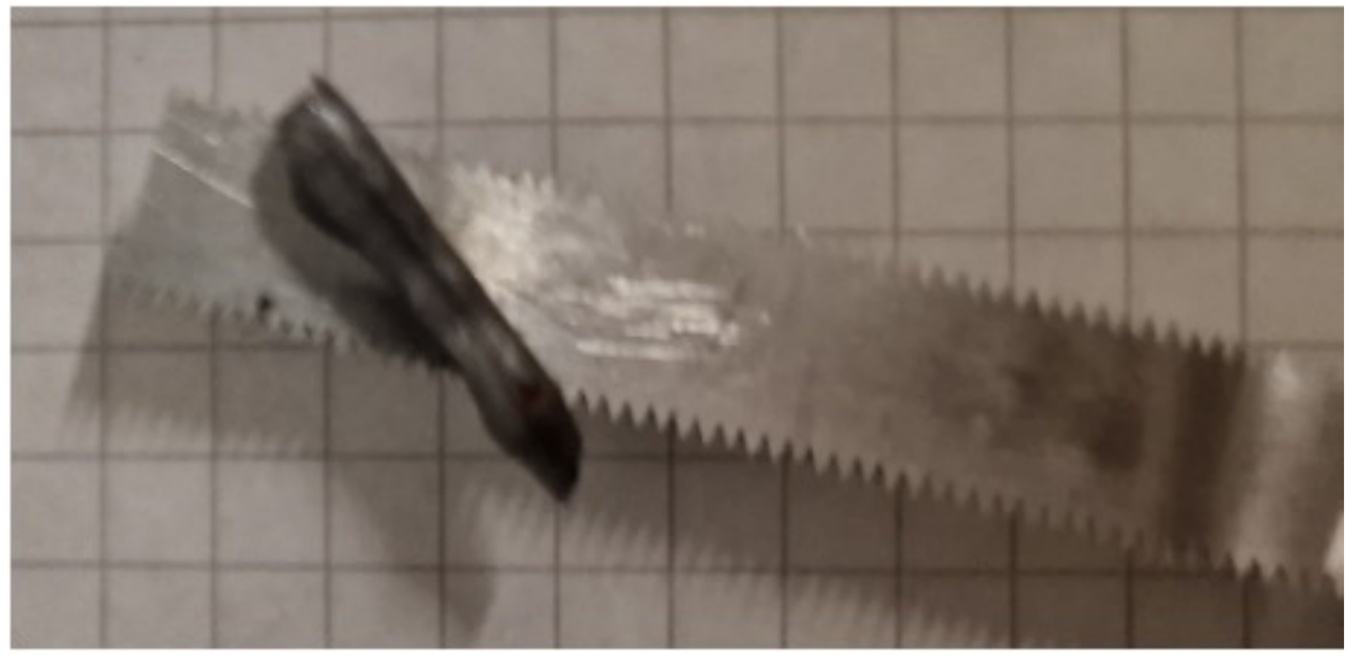
$^{160}GdRu_2Si_2$ Crystal for neutron measurements





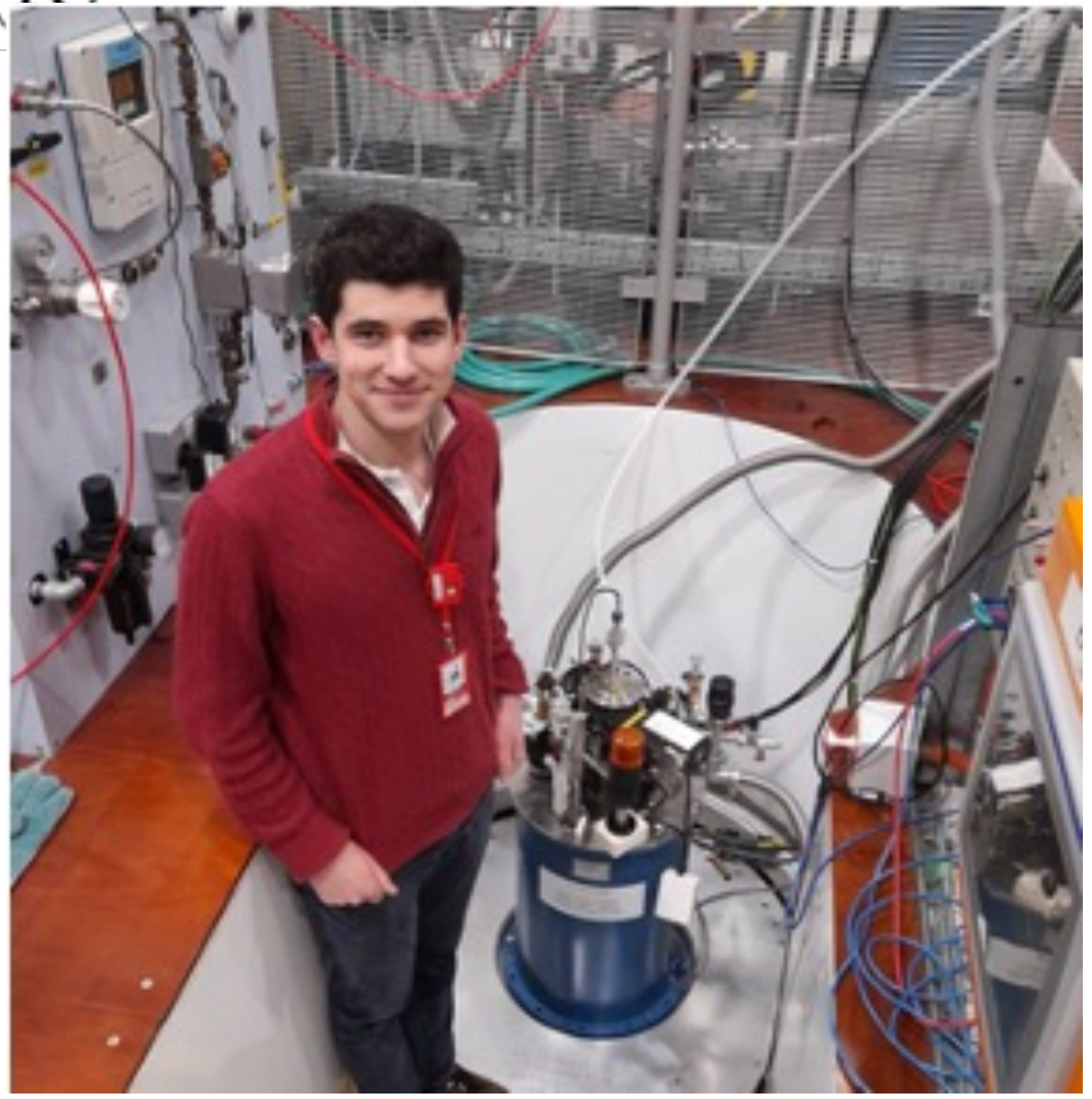






G.A. Wood PhD Thesis, Warwick 2024

WISH diffractometer,
ISIS Neutron Source, UK



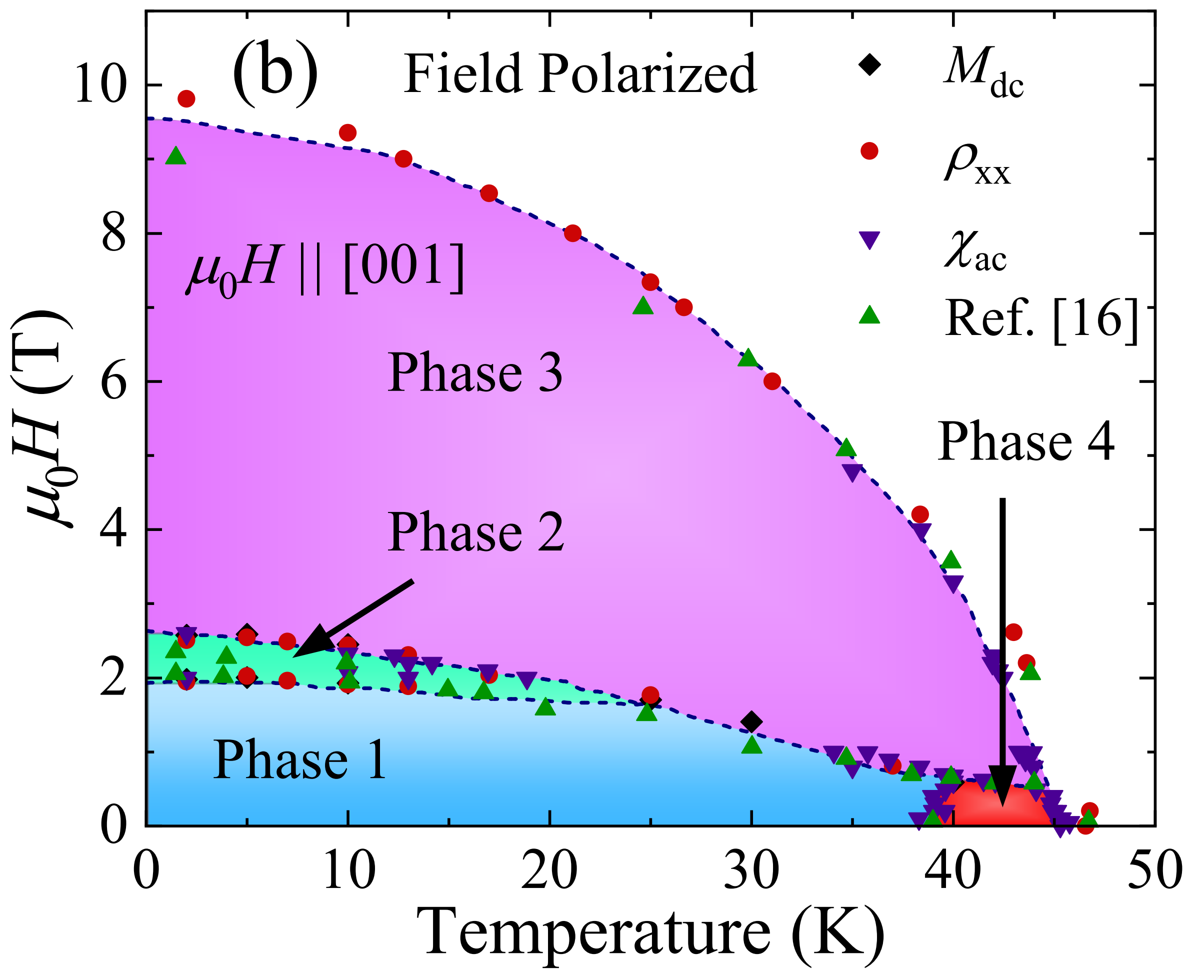
PHYSICAL REVIEW B **107**, L180402 (2023)

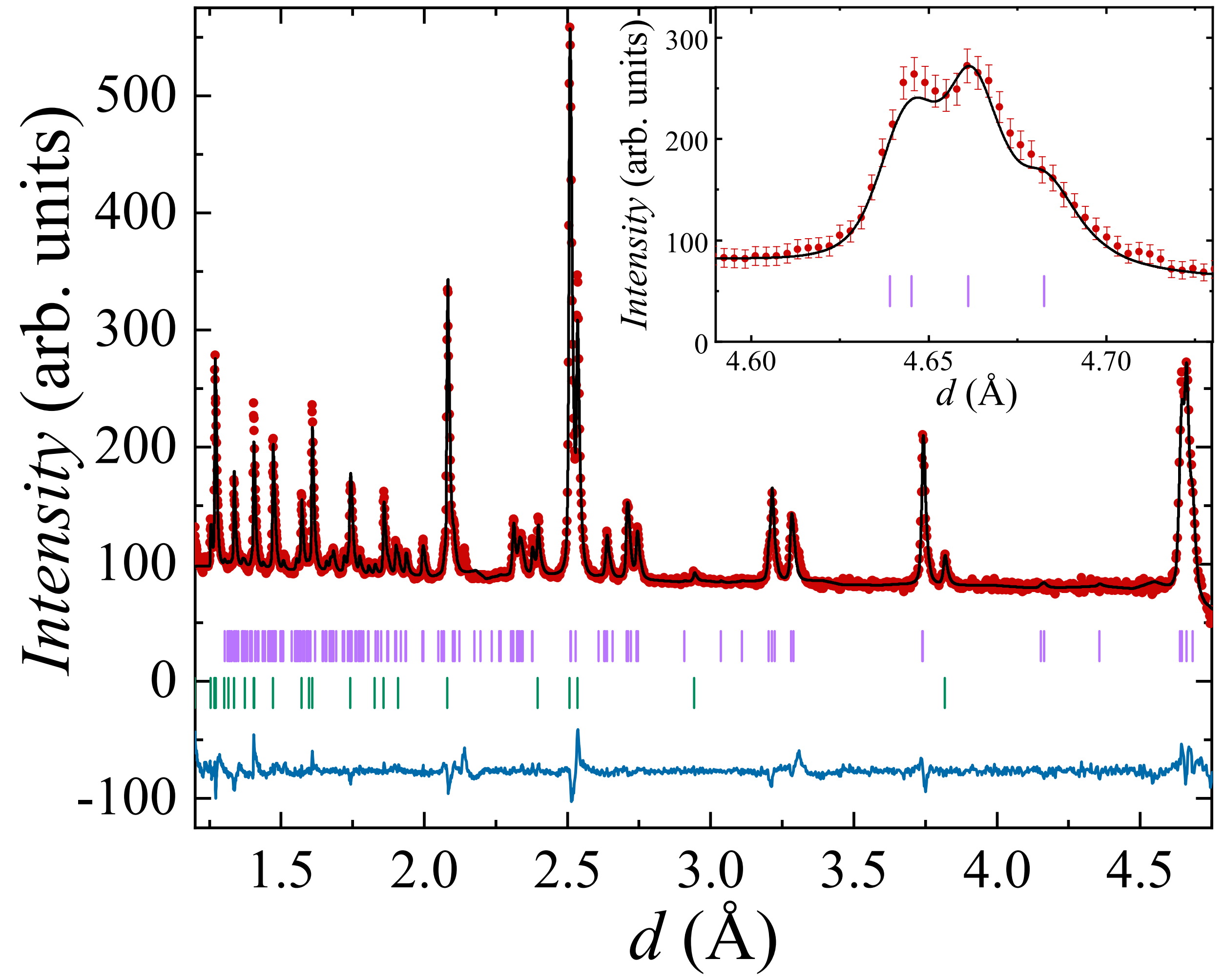
Letter

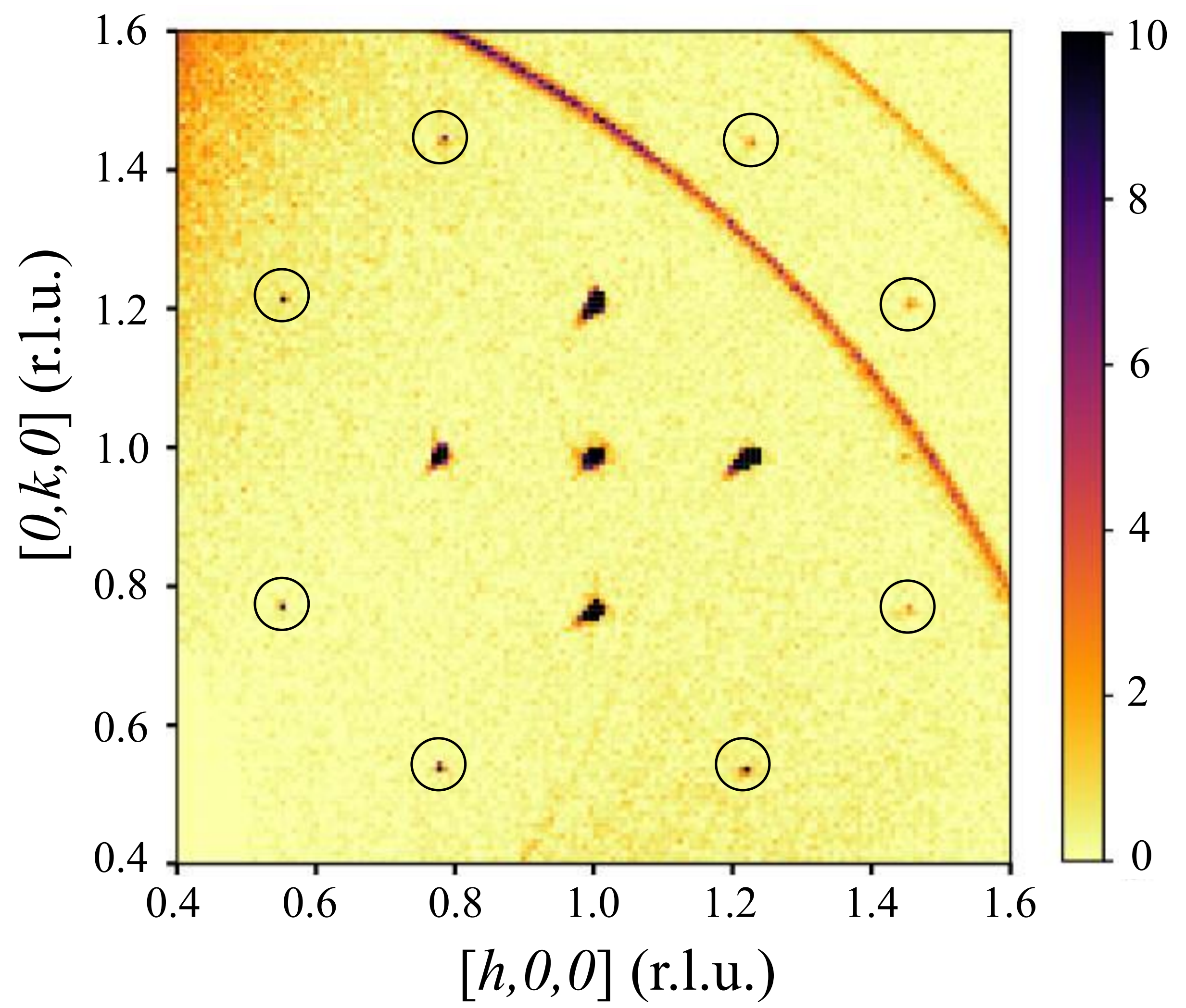
Double-Q ground state with topological charge stripes in the centrosymmetric skyrmion candidate $GdRu_2Si_2$

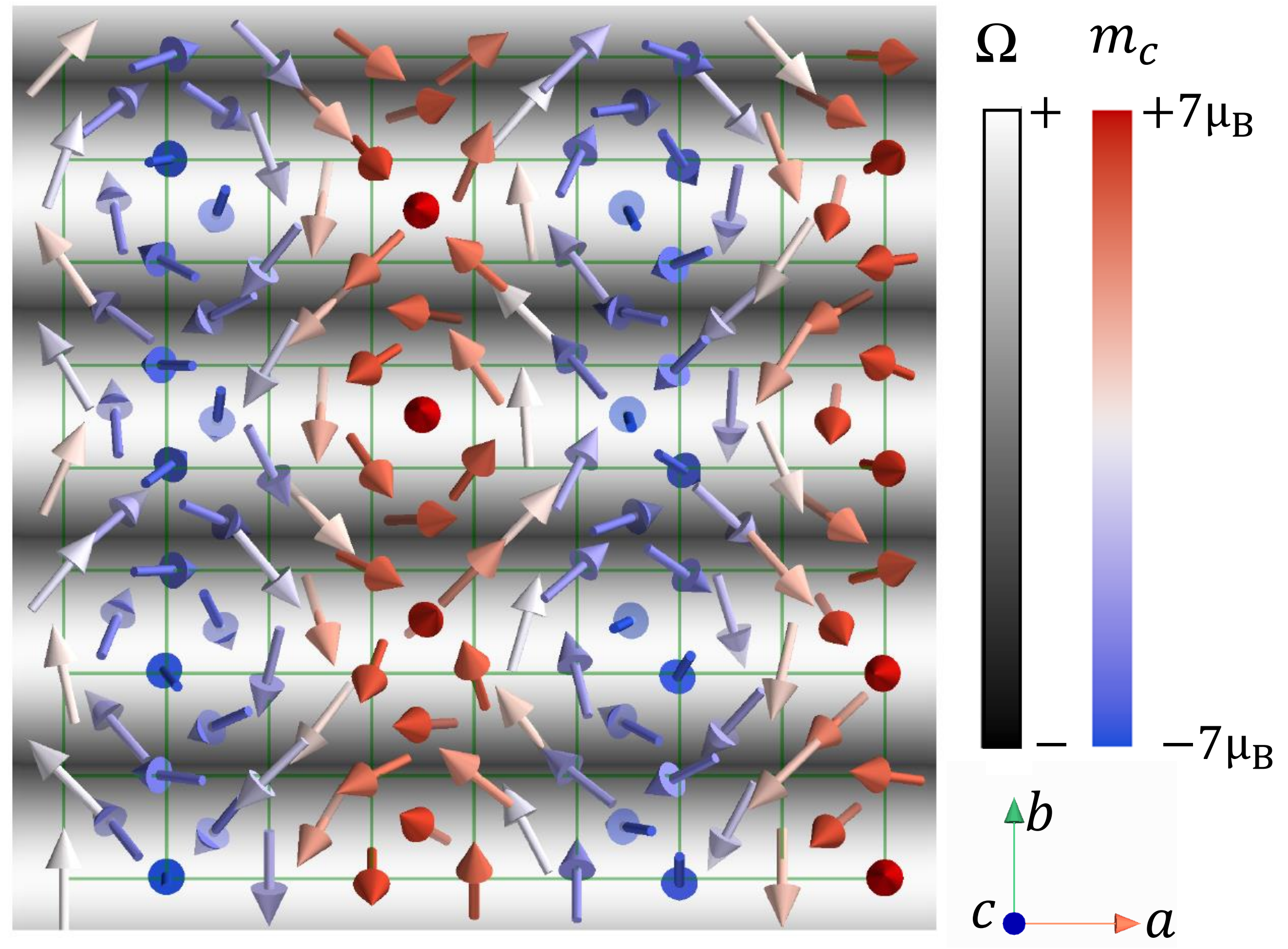
G. D. A. Wood,[1,*] D. D. Khalyavin,[2] D. A. Mayoh,[1] J. Bouaziz,[3] A. E. Hall,[1] S. J. R. Holt,[4,5] F. Orlandi,[2] P. Manuel,[2] S. Blügel,[3] J. B. Staunton,[1] O. A. Petrenko,[1] M. R. Lees,[1] and G. Balakrishnan[1,†]

- Determination of the double-Q magnetic ground state (Phase 1) of the centrosymmetric skyrmion candidate GdRu2Si2 via neutron diffraction.
- Ground state has a 1D topological charge density.
- Establishes $GdRu_2Si_2$ as a magnetically diverse system in which phase transitions between distinct topological spin textures can be probed.



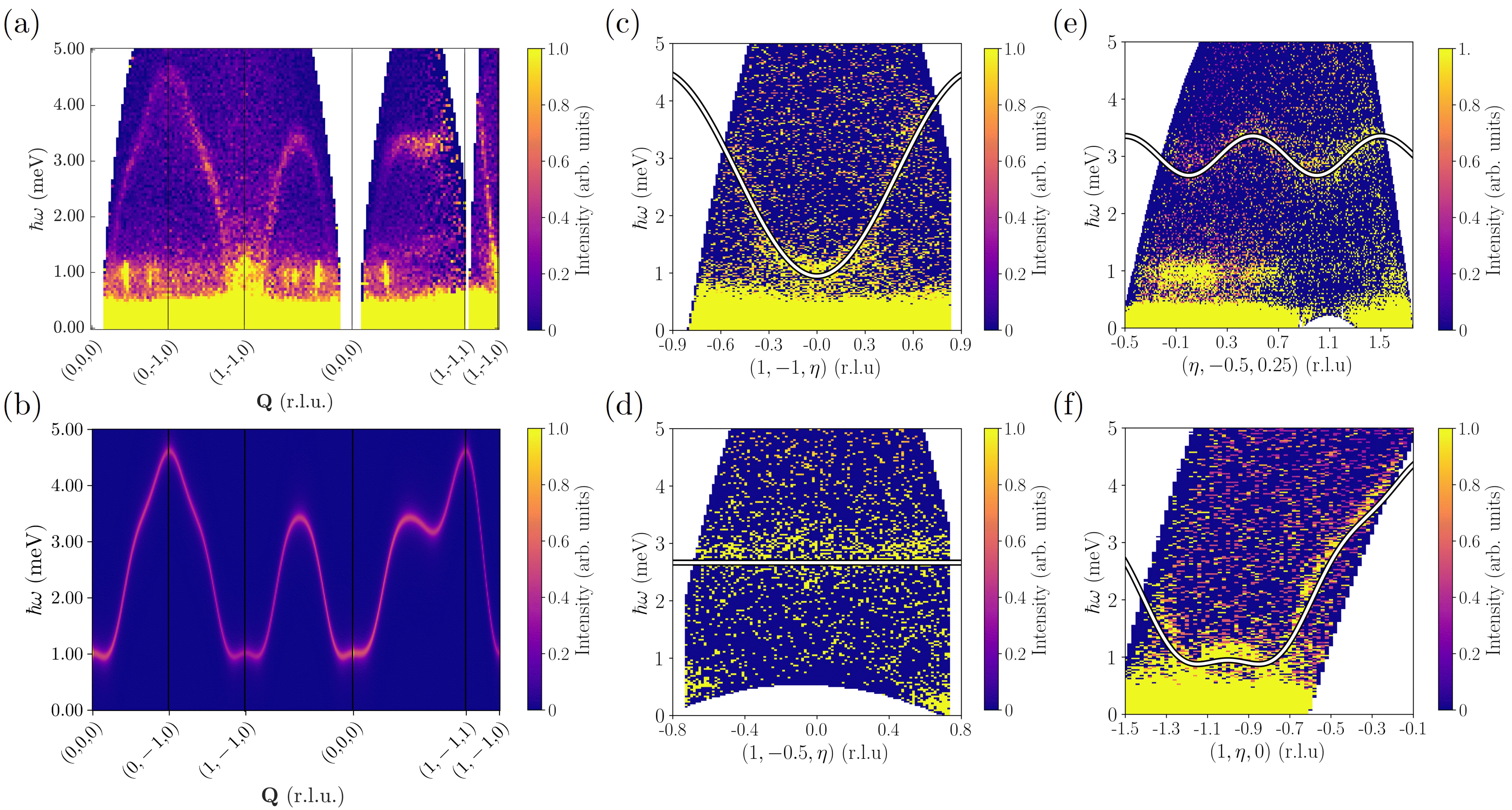


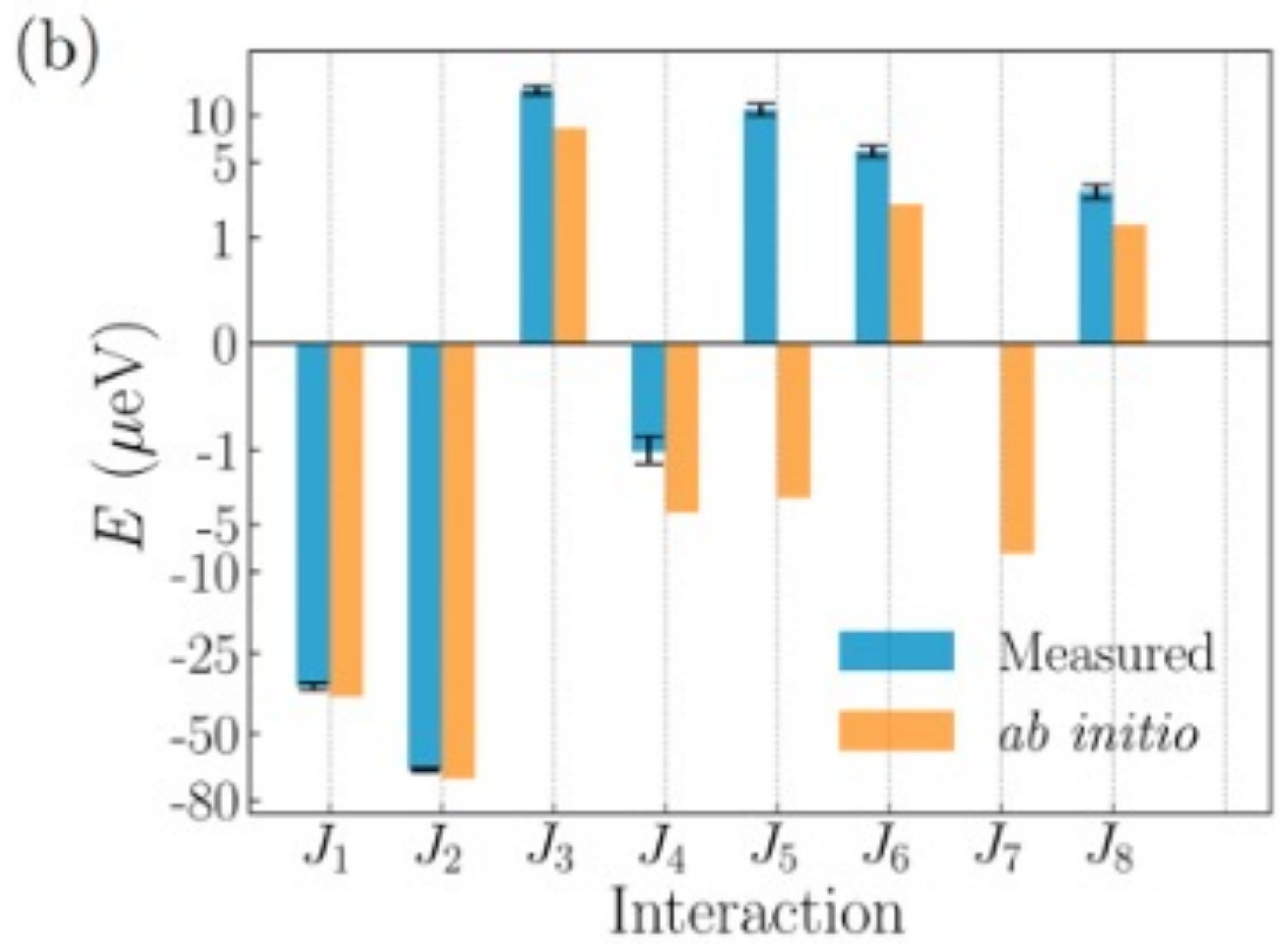




A Magnon Band Analysis of $GdRu_2Si_2$ in the Field-Polarized State

G A. Wood et al npj Quantum Materials 10, 39 (2025)





Spin excitations of $GdRu_2Si_2$ are measured and modelled in the field polarized phase using inelastic neutron scattering on the LET spectrometer. A magnetic Hamiltonian for the system is derived.

M. Gomilšek et al *Anisotropic* ***Skyrmion and Multi-q Spin Dynamics in Centrosymmetric*** ***Gd_2PdSi_3***, **Physical Review Letters 134, 046702 (2025).**

L. Gries et al, ***Uniaxial pressure effects, phase diagram, and tricritical point in the centrosymmetric skyrmion lattice magnet*** ***$GdRu_2Si_2$***, **Physical Review B 111, 064419 (2025)**

B. M. Huddart et al, ***Field-orientation-dependent magnetic phases in*** ***$GdRu_2Si_2$*** ***probed with muon-spin spectroscopy***, **Physical Review B 111, 054440 (2025)**

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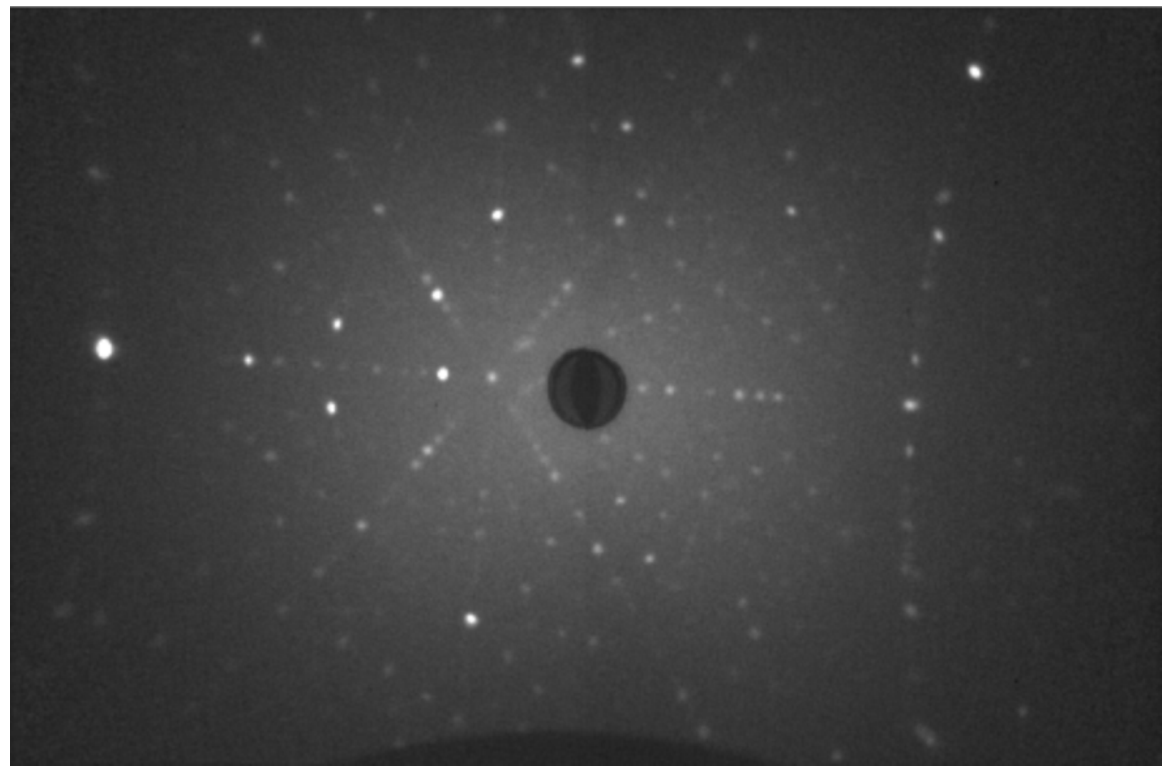
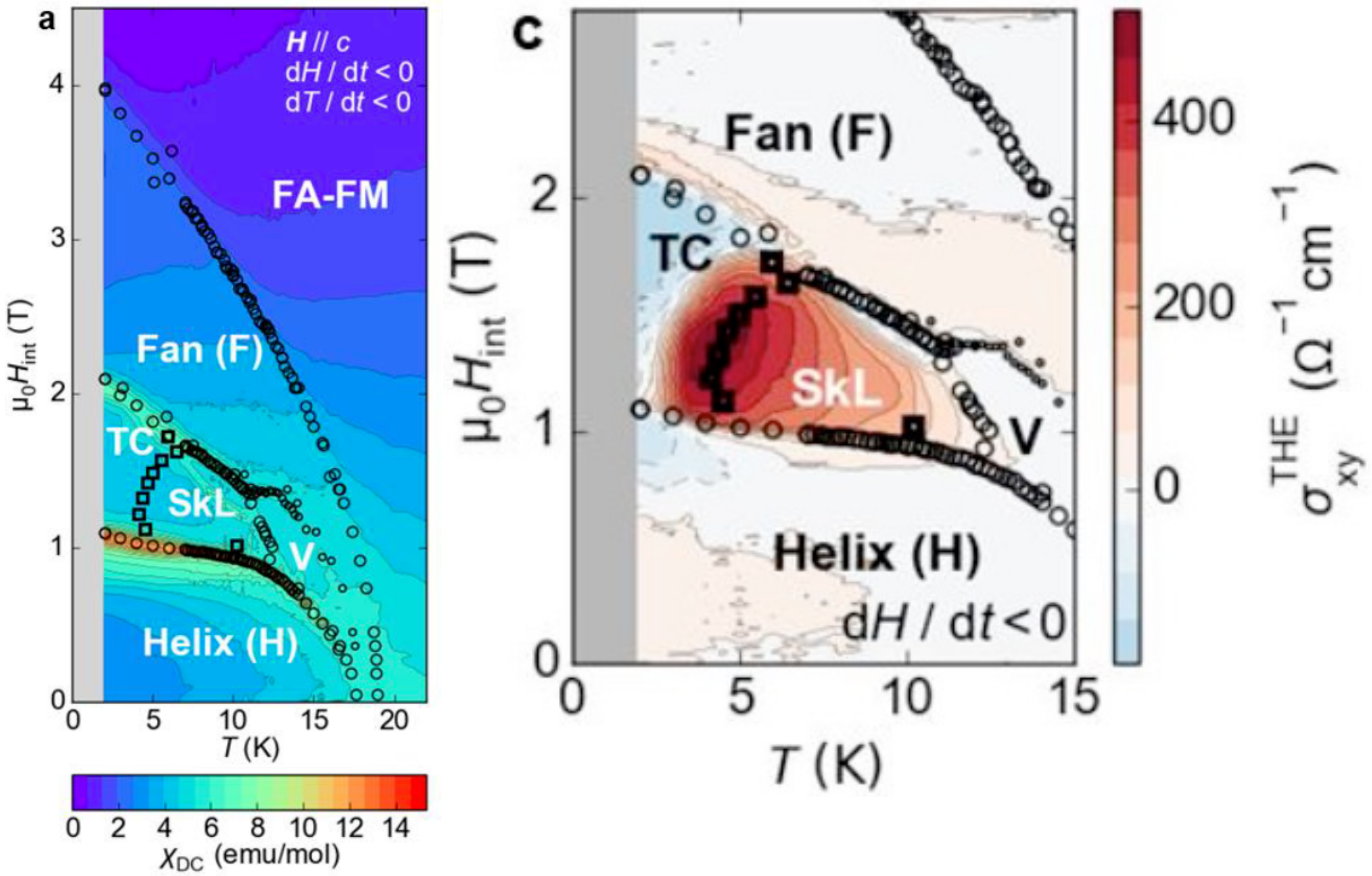
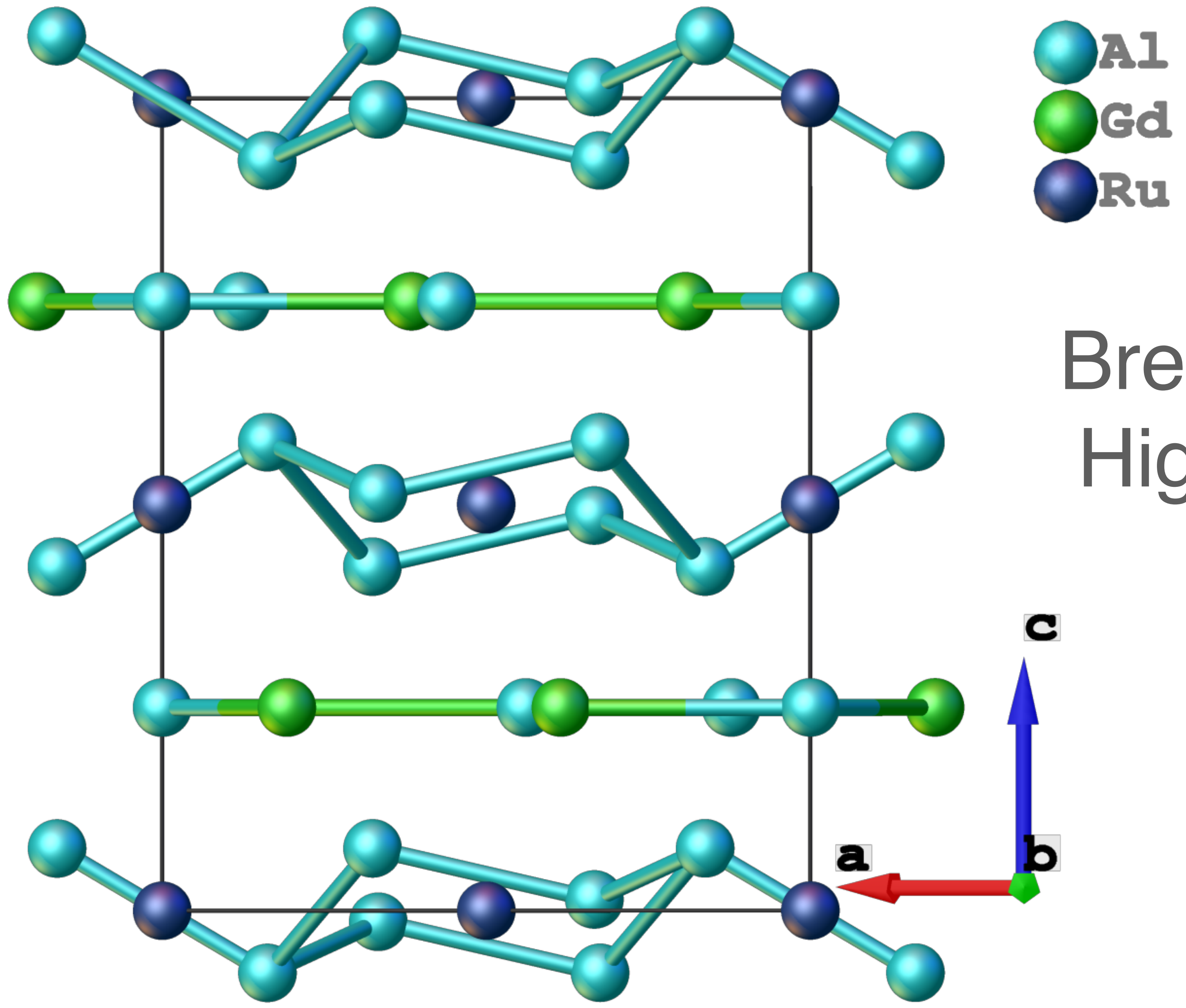
$RE_3Ru_4Al_{12}$ (RE= Gd)

$Gd_3Ru_4Al_{12}$



Large high quality single crystals of $Gd_3Ru_4Al_{12}$ grown using the optical floating zone technique





Breathing Kagome lattice
Highly frustrated system

$Gd_3Ru_4Al_{12}$: $P6_3/mmc$

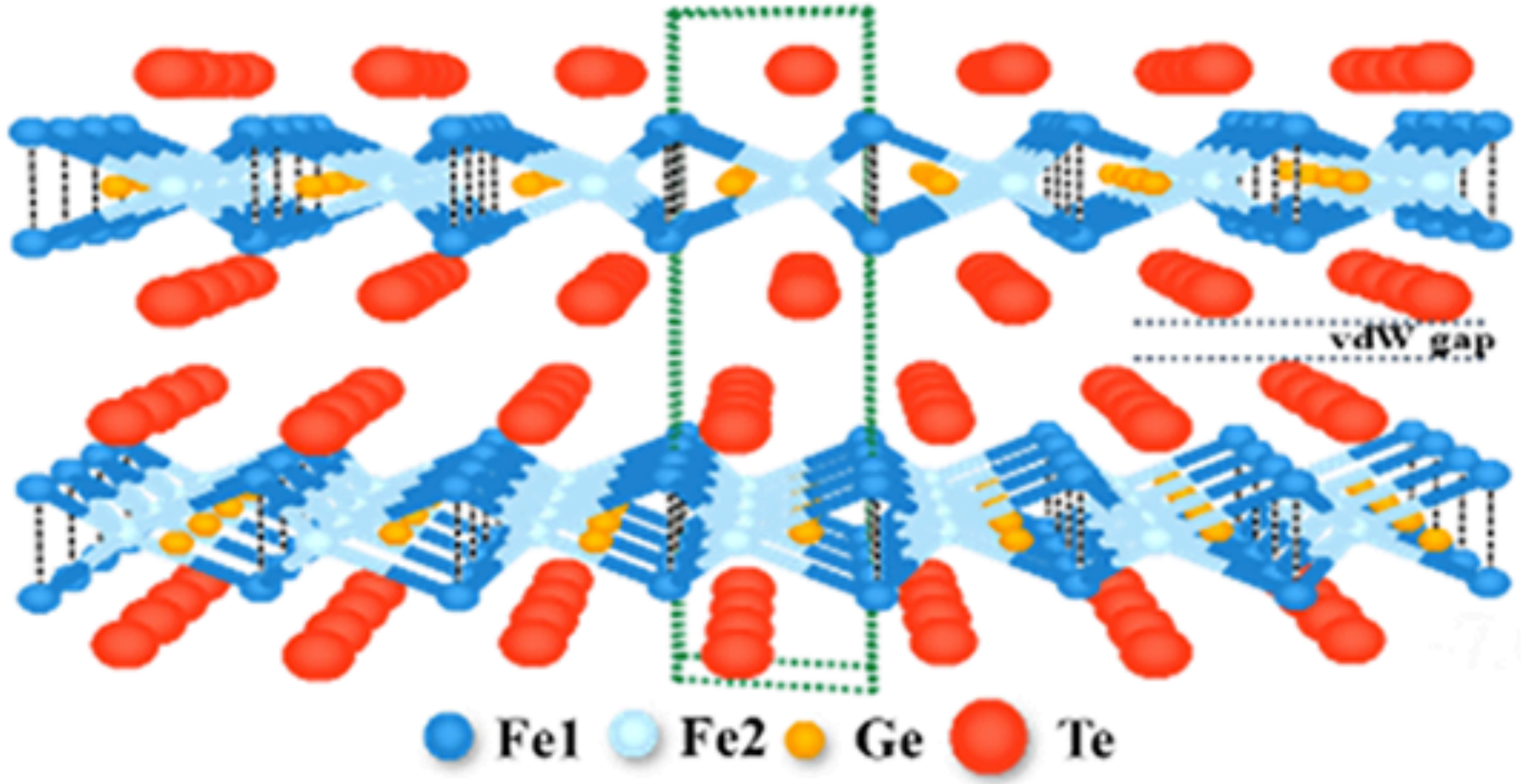
M. Hirschberger *et al. arXiv:1812.02553*

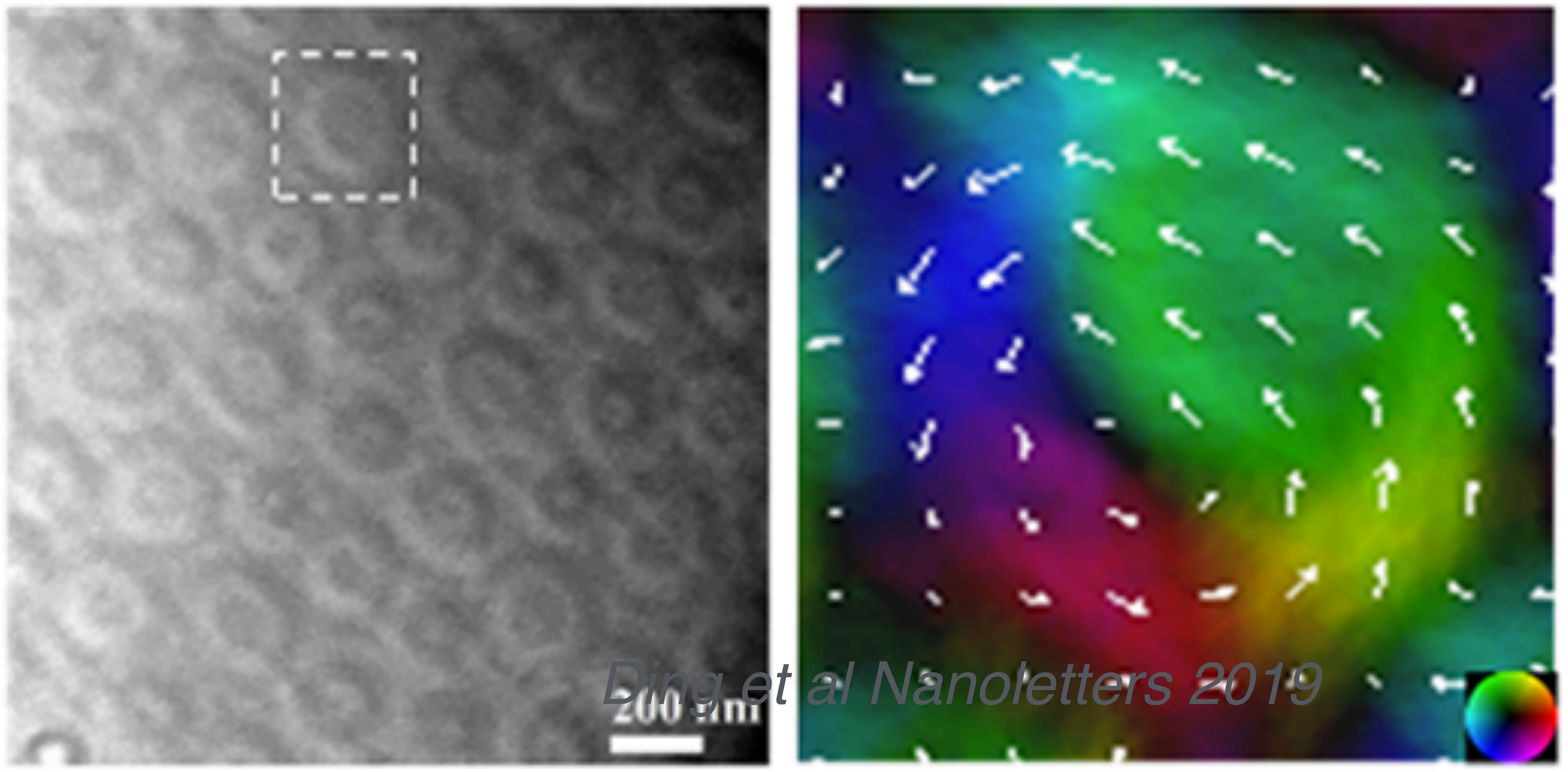
Daniel Mayoh

Layered Materials : FGT

Fe_3GeTe_2 , Fe_3GaTe_2, Fe_5GeTe_2

Fe_3GeTe_2





Ding et al Nano Lett. 20(2), 868-873 (2020)

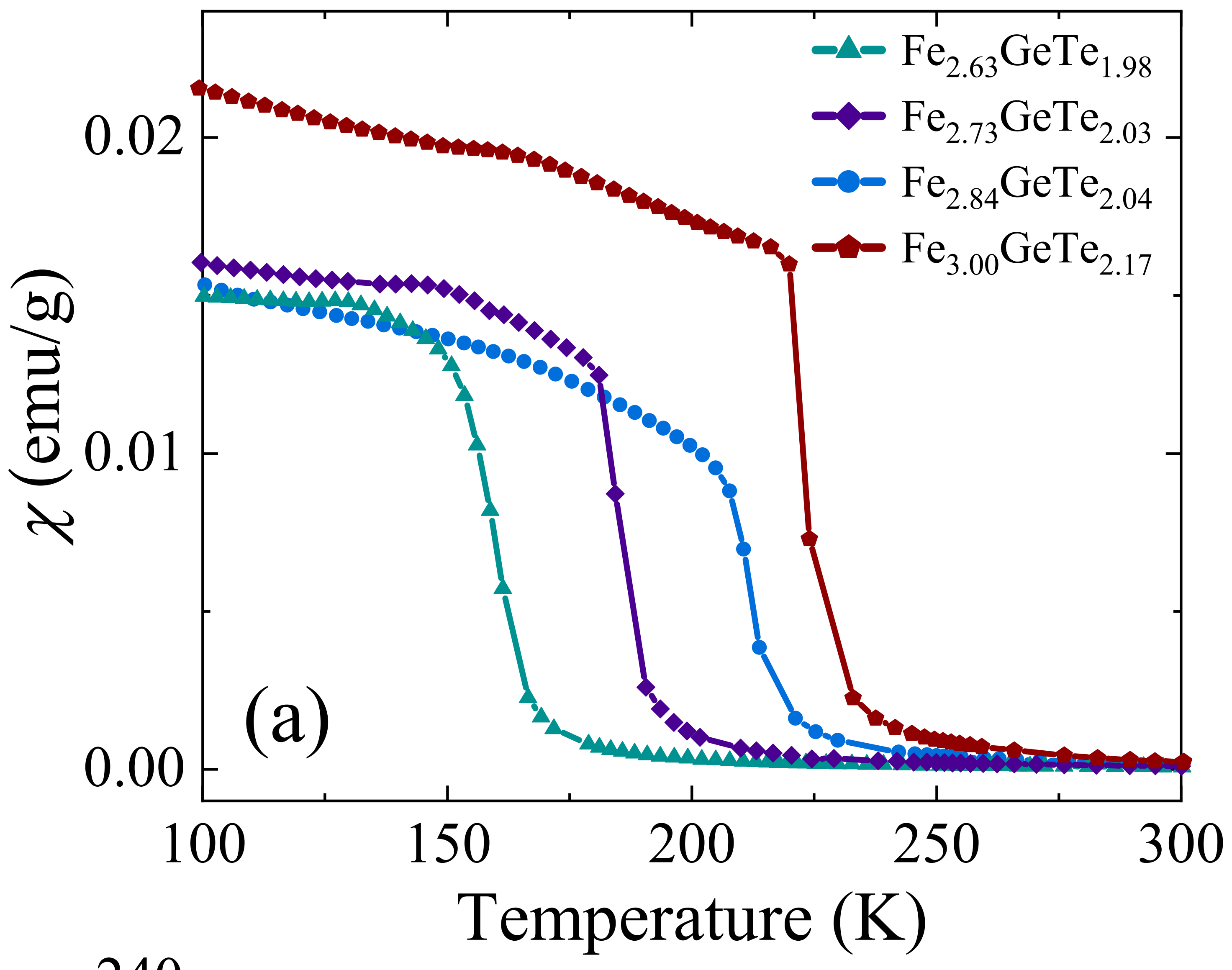
- Fe_3GeTe_2 is a 2D van der Waals magnet.
- centrosymmetric hexagonal, *$P6_3/mmc$*
- It is a ferromagnetic metal with Tc ~ 220K.
- Evidence of observation of Skyrmion bubbles.
- Tc dependent on the actual Fe content.

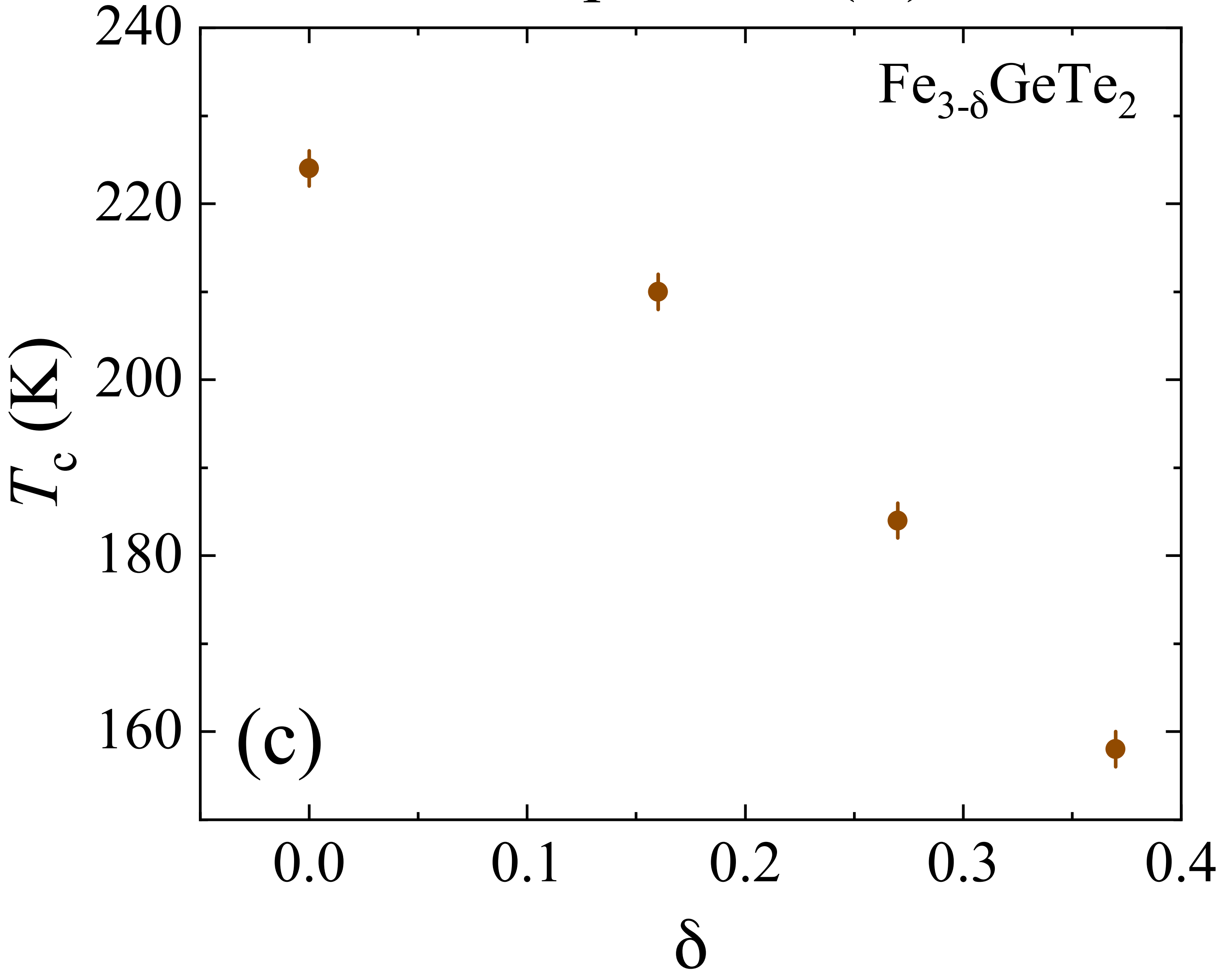
LTEM image of a skyrmion 'bubble' lattice observed at ~93K, in 600 Oe.

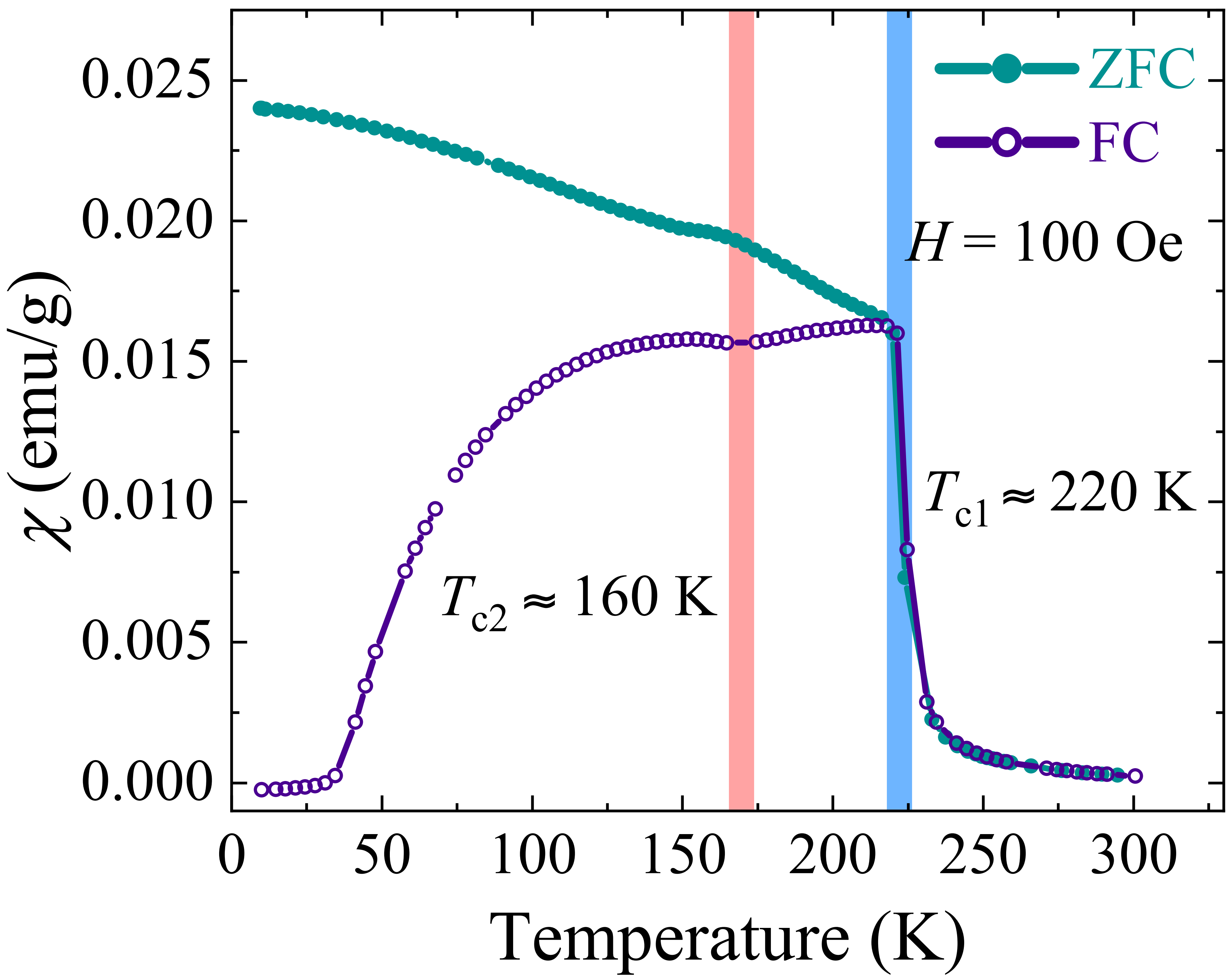
Spin textures: Competition between the uniaxial magnetic anisotropy and magnetic dipole–dipole interaction

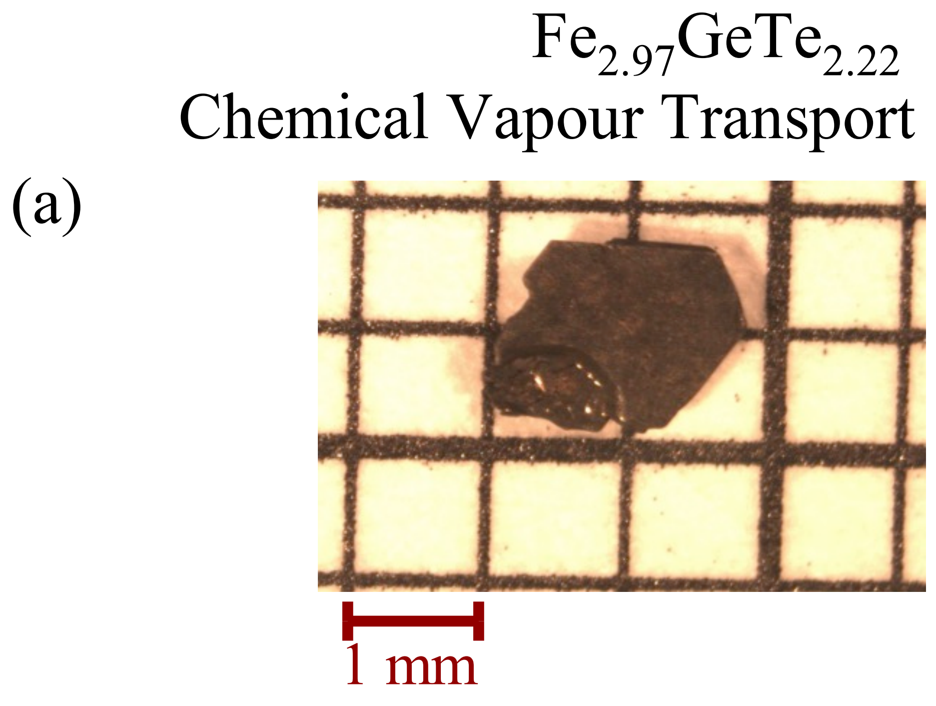
Fe_3GeTe_2 Magnetism

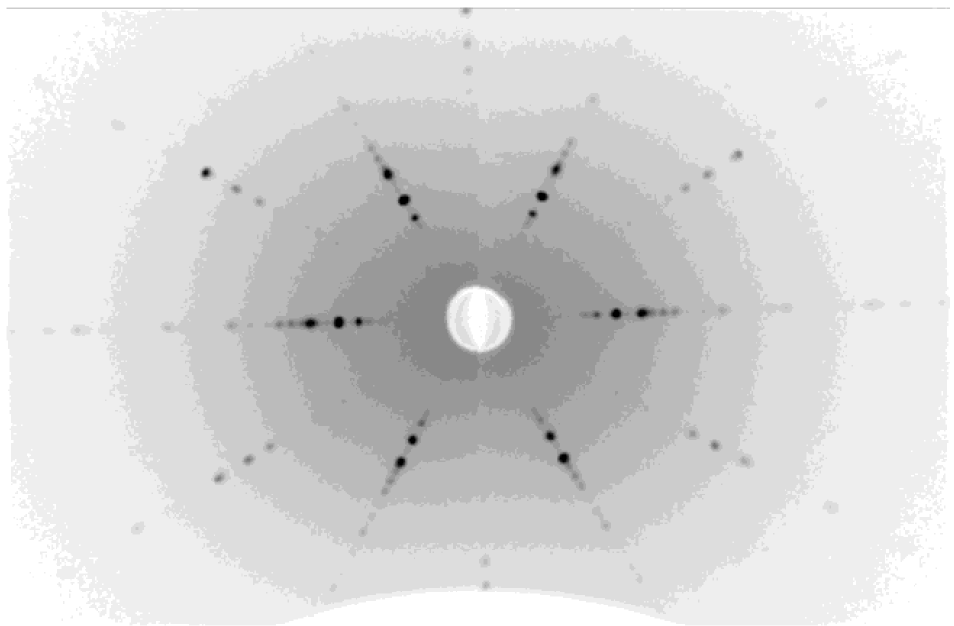
T_C variation with Fe content in the single crystals









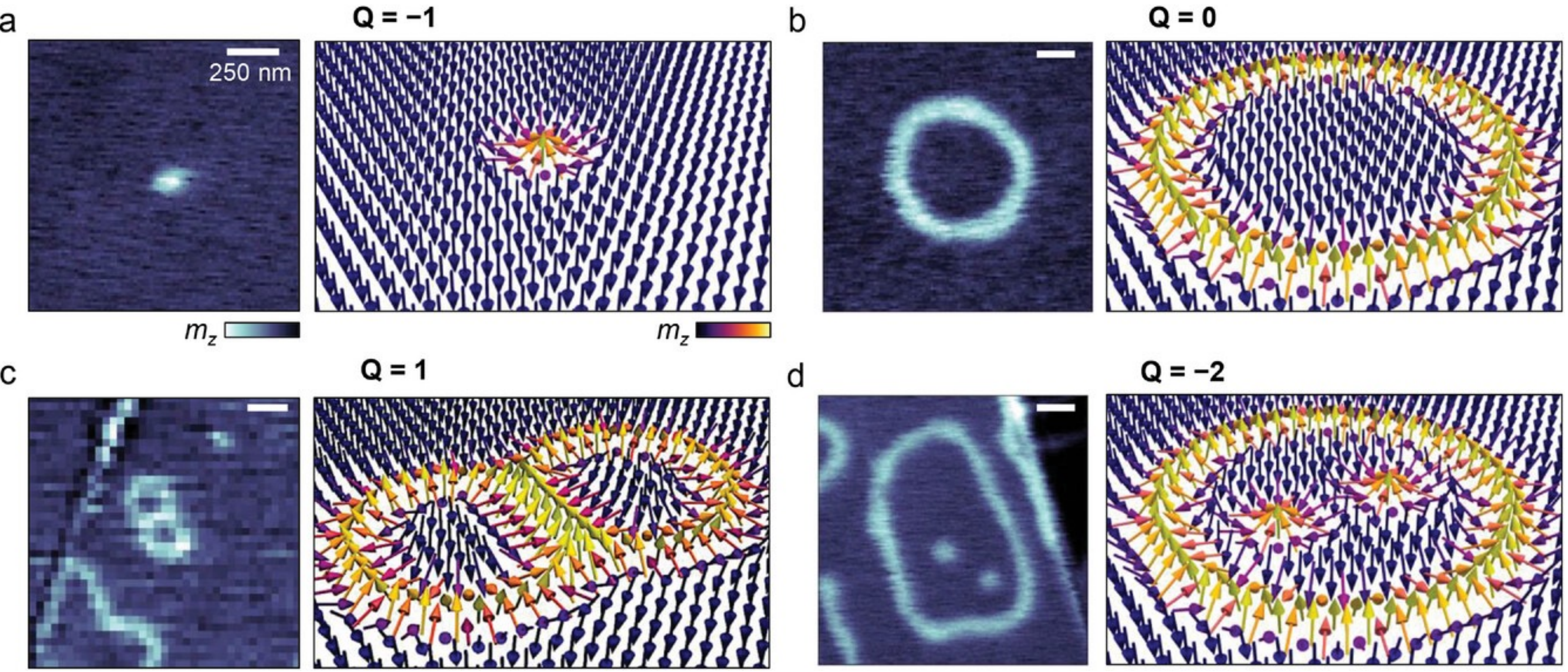


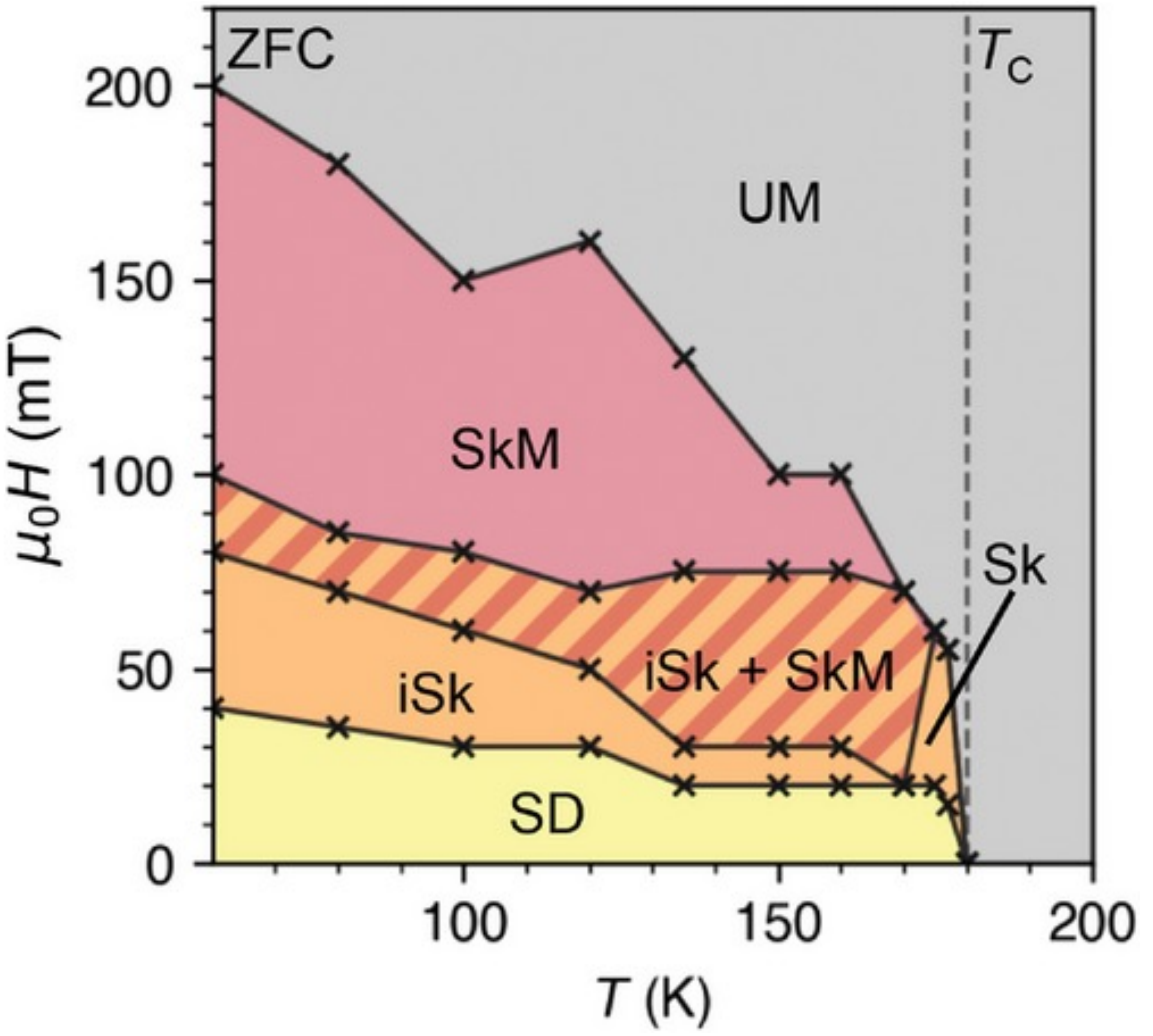
The kink in the magnetisation curves at ~160K is the region where the skyrmions are observed.

D. A. Mayoh et al ACS Crystal Growth & Design **21**, 6786-6792 (2021)

Skyrmions, Skyrmionium, Skyrmion bag and skyrmion sacks

Scanning transmission x-ray microscopy on $Fe_{0.27}GeTe_2$.





a) Skyrmion **b)** Skyrmionium **c)** Skyrmion bag **d)** Skyrmion sack

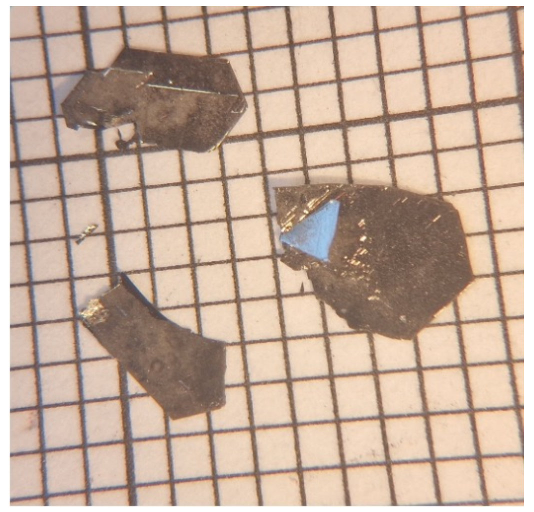
L. Powalla *et al.*, Advance Materials **35**, 2208930 (2023).

D Backes *et al Valence-state mixing and reduced magnetic moment in*
$FeGe_2Te_3$ single crystals with varying Fe content probed by x-ray spectroscopy
Nanotechnology **35** 395709 (2024)

D Backes et al *Strain-Modulated Ferromagnetism at an Intrinsic van der Waals Heterojunction*, Advanced Functional Materials 2400552 (2024)

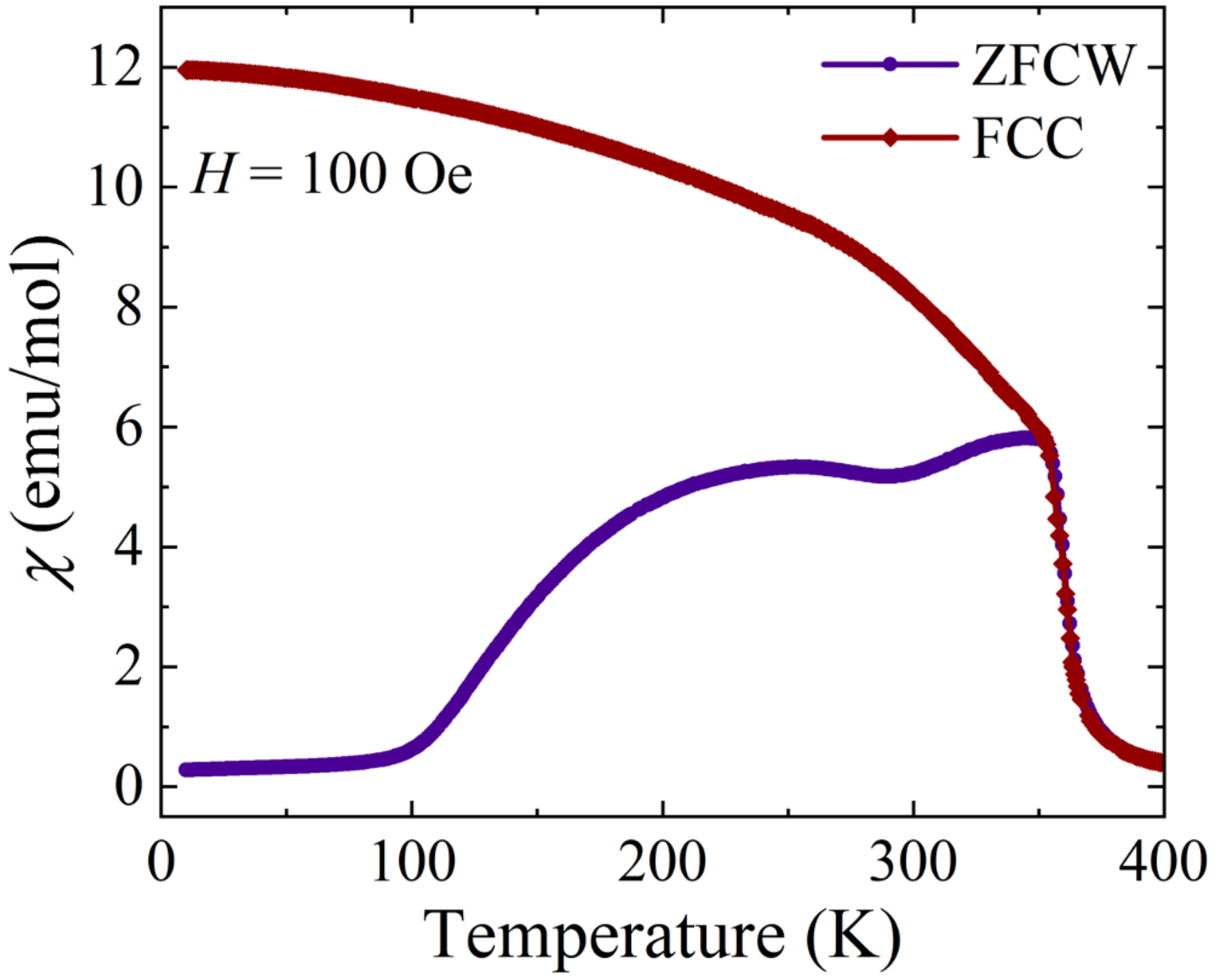
S Vaidya et al *Direct evidence from high-field magnetotransport for a dramatic change of quasiparticle character in van der Waals ferromagnet*
Phys. Rev. Research **6** L032008 (2024)

Fe_3GaTe_2



Large, very shiny hexagonal crystals

1 mm



T_c range ~350-380 K

G. Zhang Nat Comms. 13 (2022) 5067
G. Hu, Adv. Mater 2024 (2024) 2403154

Fe_5GeTe_2

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- Bulk single crystals exhibit a **T_c of 310 K**
- Nanoflakes exfoliated from single crystals exhibit **T_c** ranging from **270 to 300 K**

A. F. May 2D Mater. 9 (2022) 015013
A. F. May, ACS nano 13 (2019) 4436
A.F. May Phys. Rev. Materials 2 (2019) 10440

Fe_3GaTe_2 and Fe_5GeTe_2

Cu_2OSeO_3 Nanoparticles

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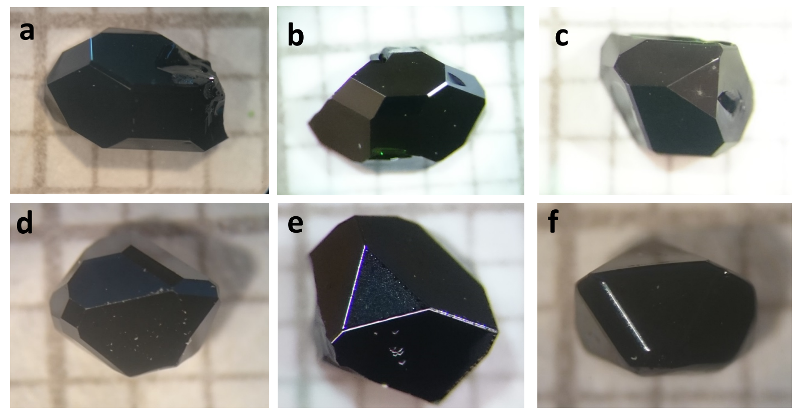
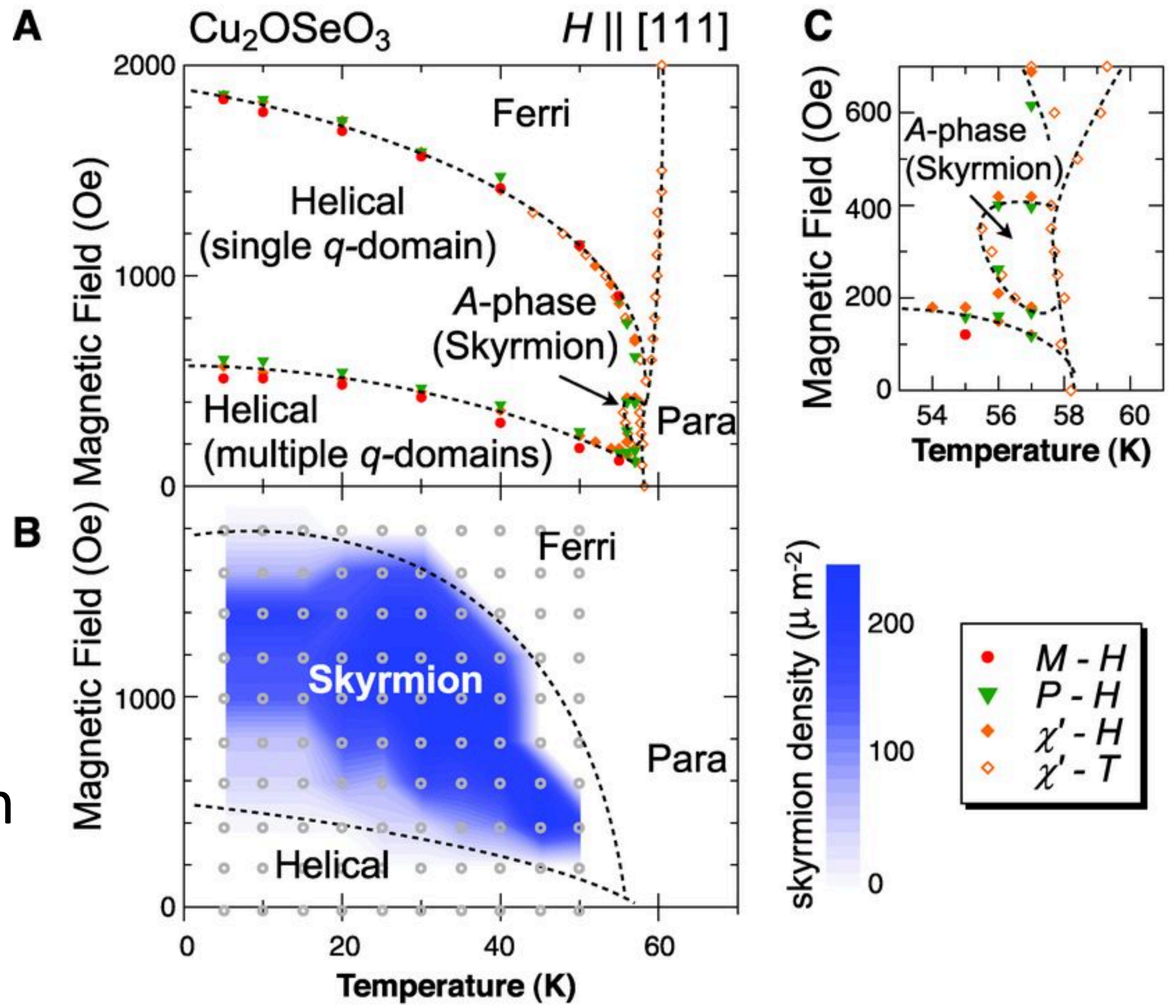
Size effects

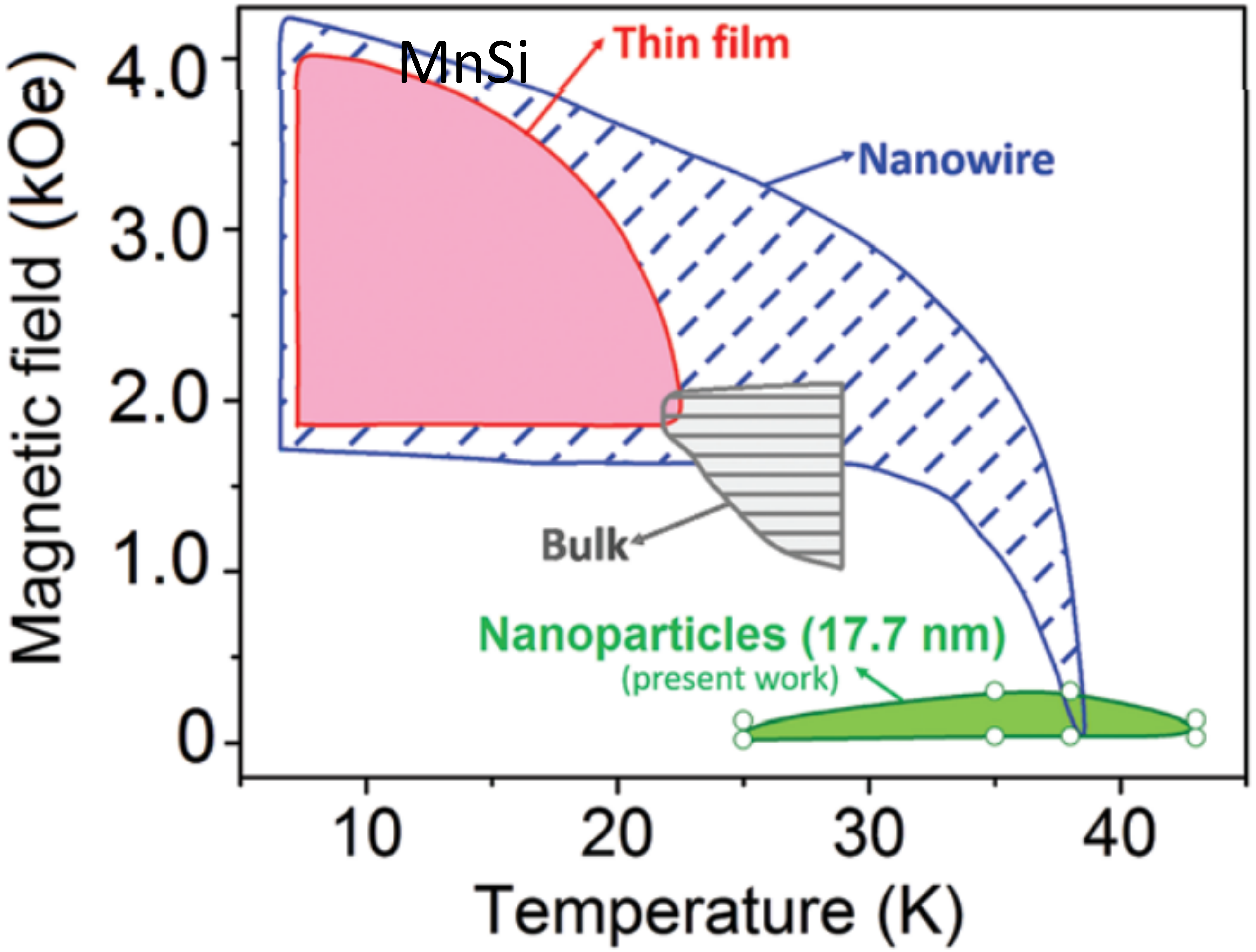
Crystals



~100 nm Thin film



Seki, Shinichiro, *et al*. "Observation of skyrmions in a multiferroic material." Science 336.6078 (2012): 198-201.



Das, Bhaskar, et al. "Effect of size confinement on skyrmionic properties of MnSi nanomagnets." Nanoscale 10.20 (2018): 9504-9508.

Material	T_c (K)	λ_m (nm)
MnSi	30	18
$Fe_{1-x}Co_xSi$	<36	40 ~ 230
MnGe	170	3
FeGe	278	70
Cu_2OSeO_3	59	62

- In B20 materials the SkX is stable over a wide T and H rang (Thin films)
- The Skyrmion pocket is smaller in 3D (crystal)
- Different results for different geometries for MnSi
- Motivation to investigate nanoparticles of Cu_2OSeO_3

Nanoparticles synthesis route

The $CuCl_2$ solution was slowly added to the Na_2SeO_3 solution producing a precipitate which gradually changed from milky green to blue upon adding additional $CuCl_2$. This precipitate, which forms via the reaction

$$Na_2SeO_{3(aq)} + CuCl_{2(aq)} \longrightarrow 2NaCl_{(aq)} + CuSeO_3.2H_2O_{(s)} \quad (1)$$

Cu_2OSeO_3 nanoparticles were synthesised by heating $CuSeO_3.2H_2O$ in an alumina crucible under a flow of O_2 in a tube furnace. At the initial heating stage, a dehydration process takes place in multiple steps leading to the overall reaction [42]

$$CuSeO_3.2H_2O \xrightarrow{\Delta T} CuSeO_3 + 2H_2O \uparrow. \quad (2)$$

A thermal decomposition then takes place to convert the $CuSeO_3$ into Cu_2OSeO_3 nanoparticles via the following reactions:

$$4CuSeO_3 \xrightarrow{\Delta T} Cu_4O(SeO_3)3 + SeO_2\uparrow, \quad (3)$$

$$Cu_4O(SeO_3)_3 \xrightarrow{\Delta T} 2Cu_2OSeO_3 + SeO_2 \uparrow. \quad (4)$$

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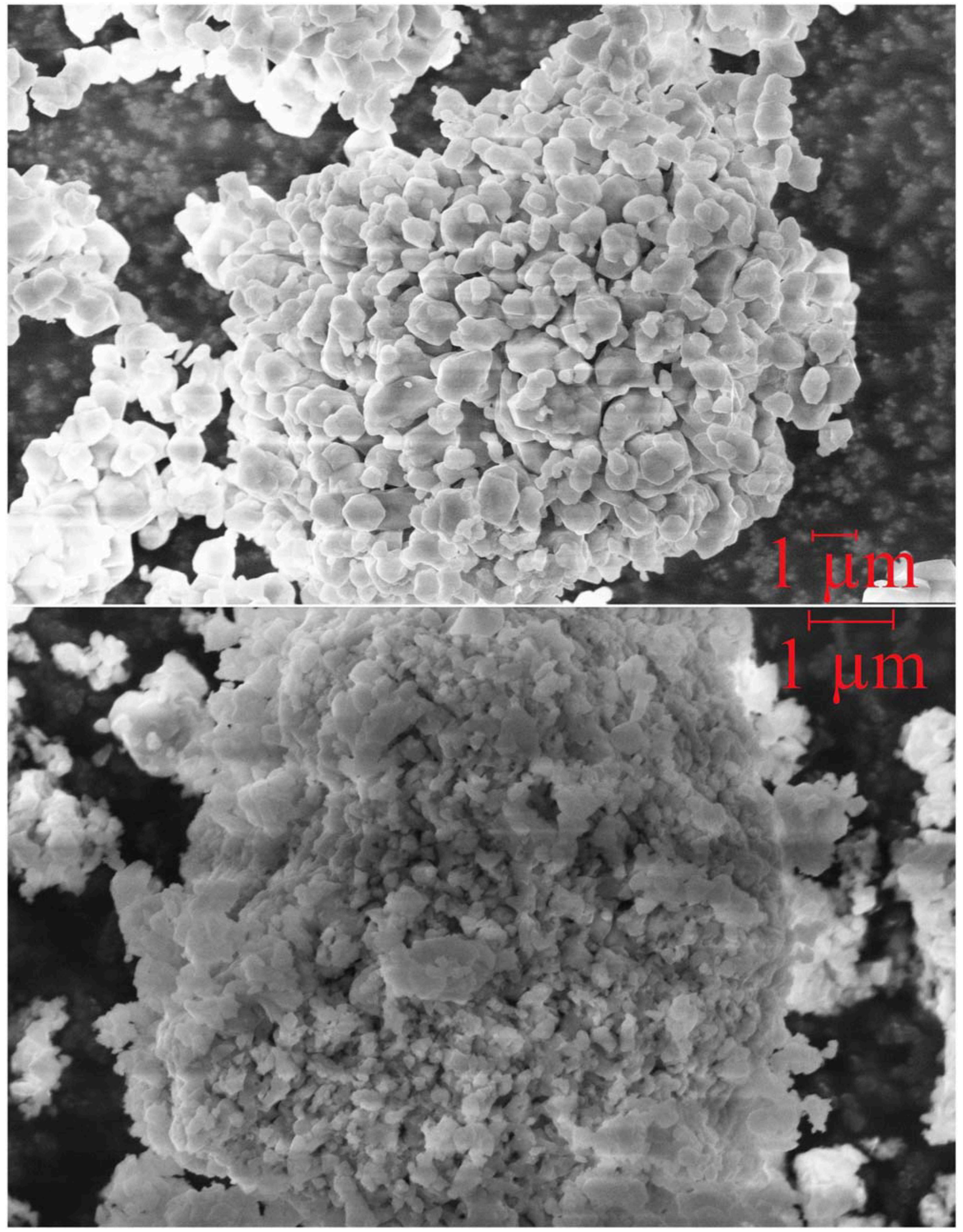
Nanoparticles synthesis

Table 1. Synthesis conditions of samples and sizes of Cu_2OSeO_3 particles. Samples A—D were prepared by precipitation and thermal treatment. Sample SS was prepared via a solid state reaction.

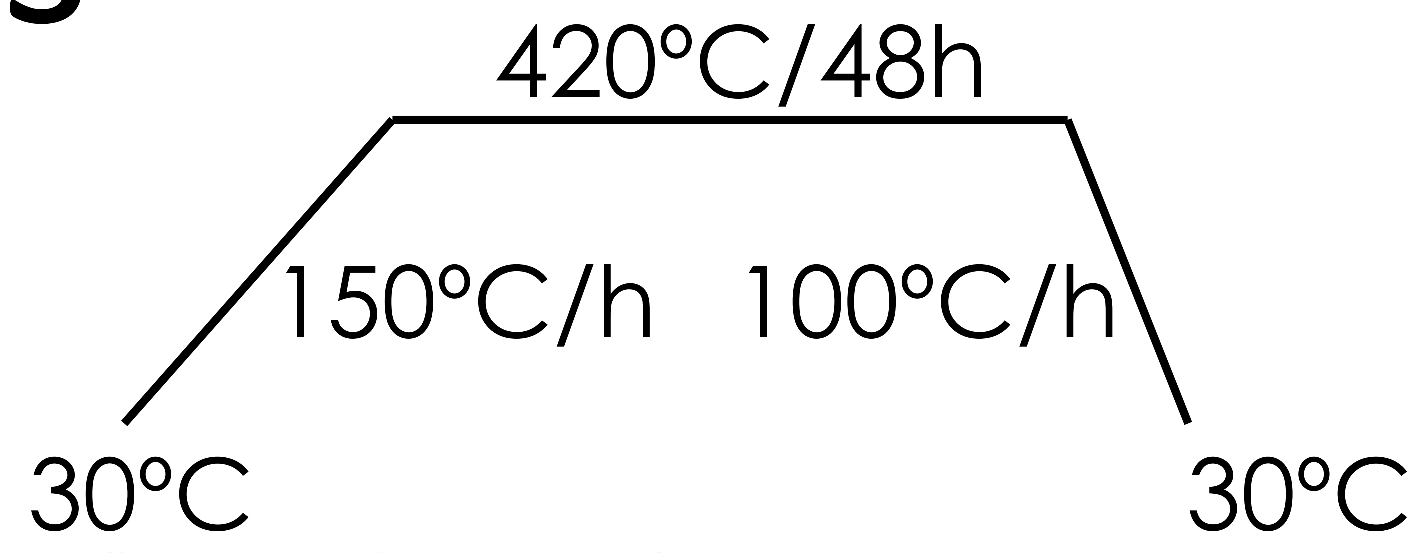
Sample	Temperature (°C)	Dwell time (h)	Particle diameter	
			PXRD	SEM/TEM
A	400 then 420	288 then 48	~3.5 μm	1–8 μm
B	420	72	~3.2 μm	0.8-2 μm
C	450	24	~112 nm	15-250 nm
D	470	24	~126 nm	—
SS	650	96	—	10-100 μm

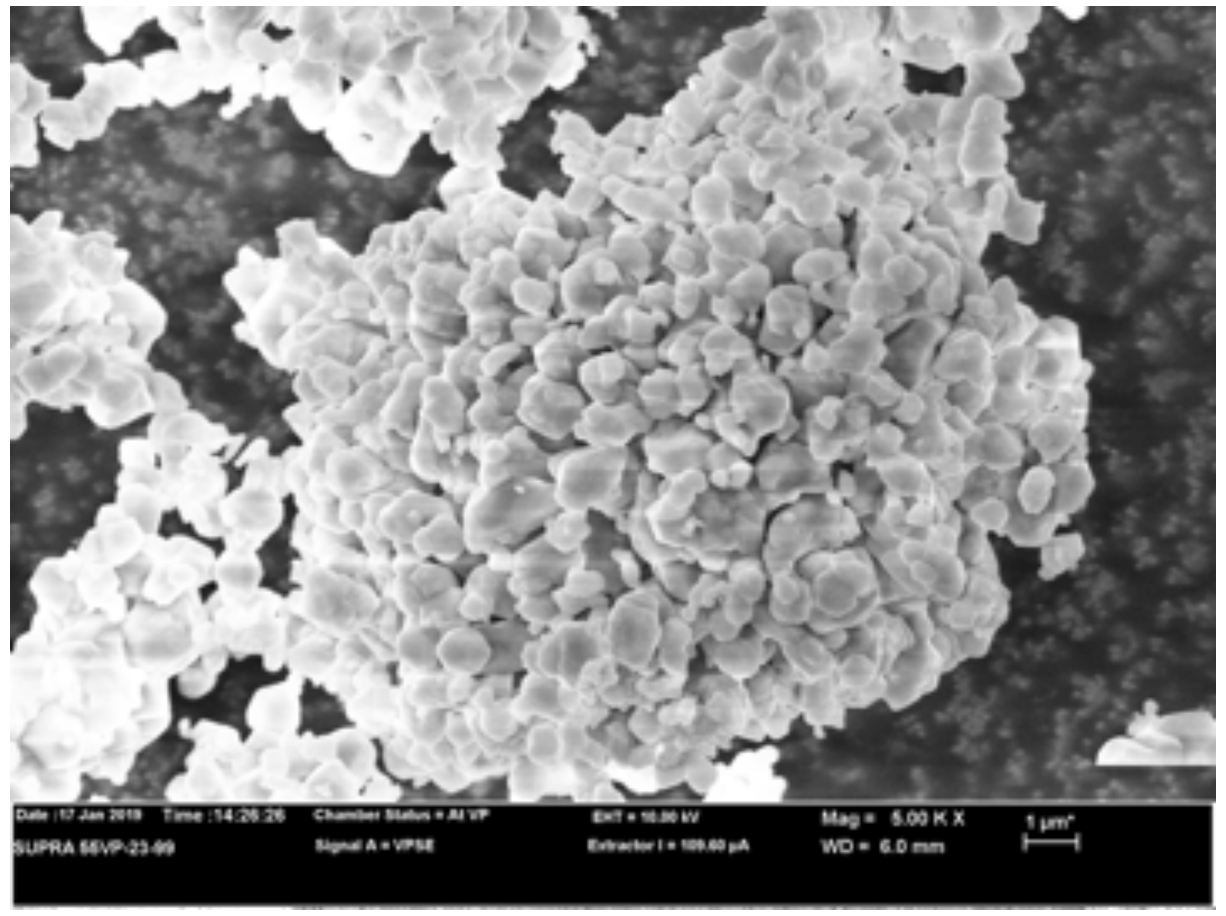
SEM images of Cu2OSeO3 nanoparticles from (a) Sample B and (b) Sample C. The sizes of the individual particles and aggregates can be clearly seen.



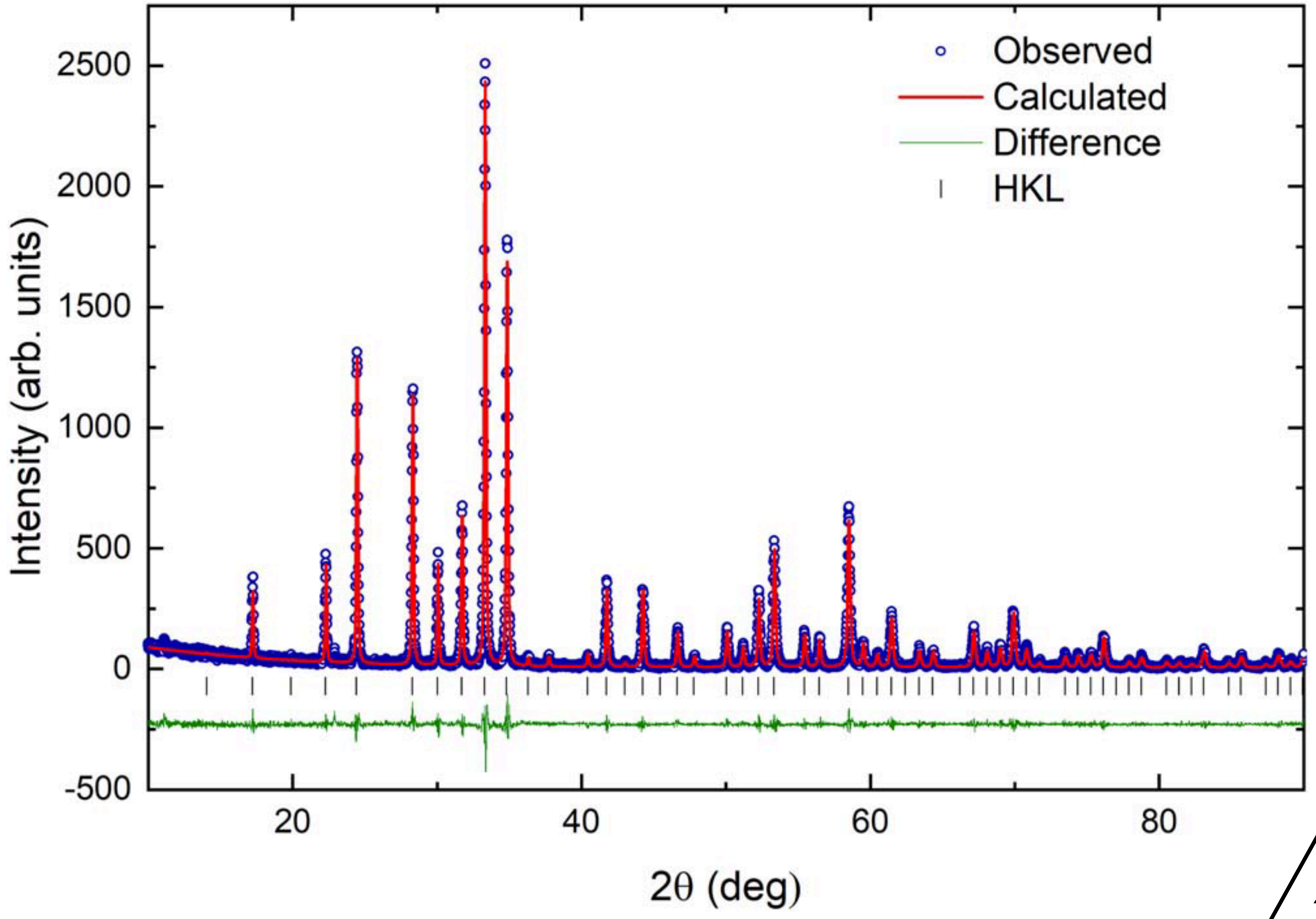
S J R Holt et al Materials Research Express 8 116101 (2021)

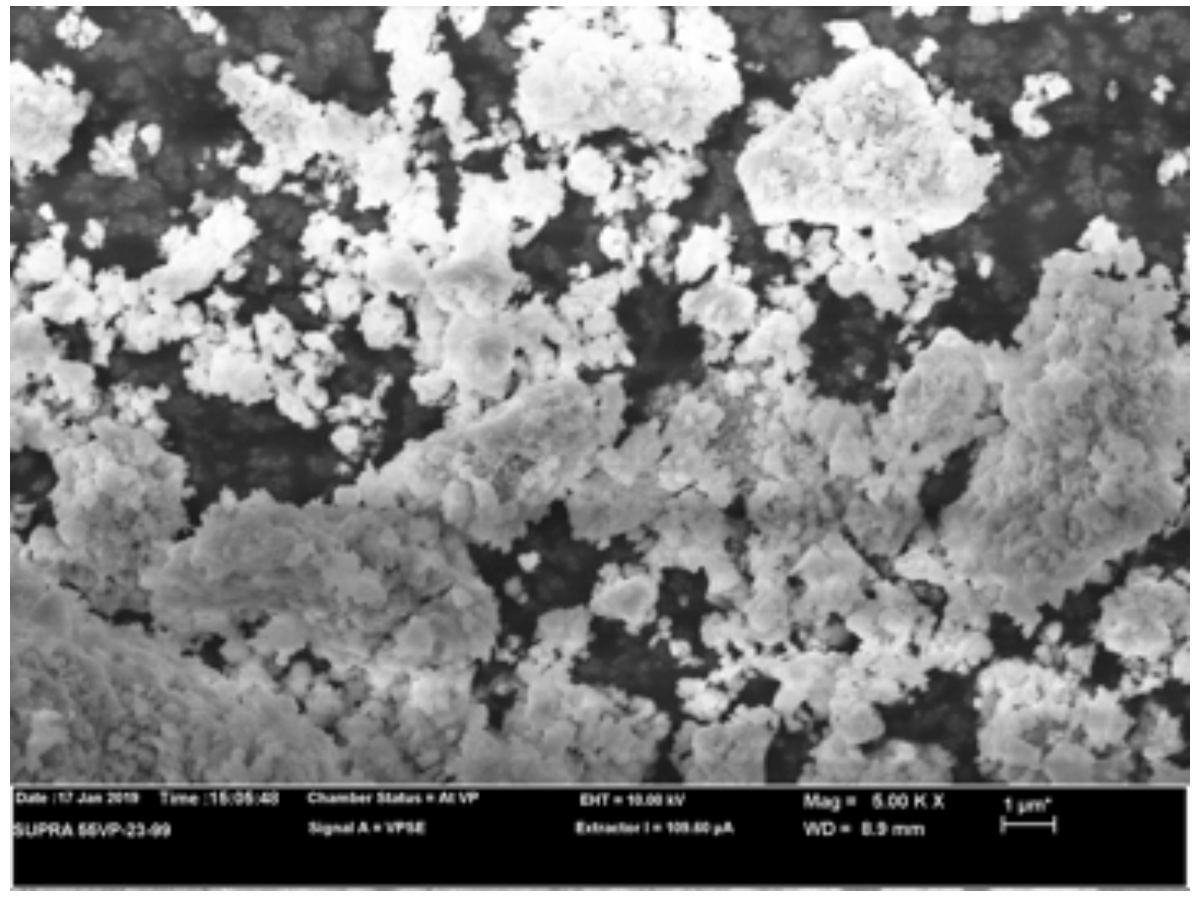
Nanoparticle sizes

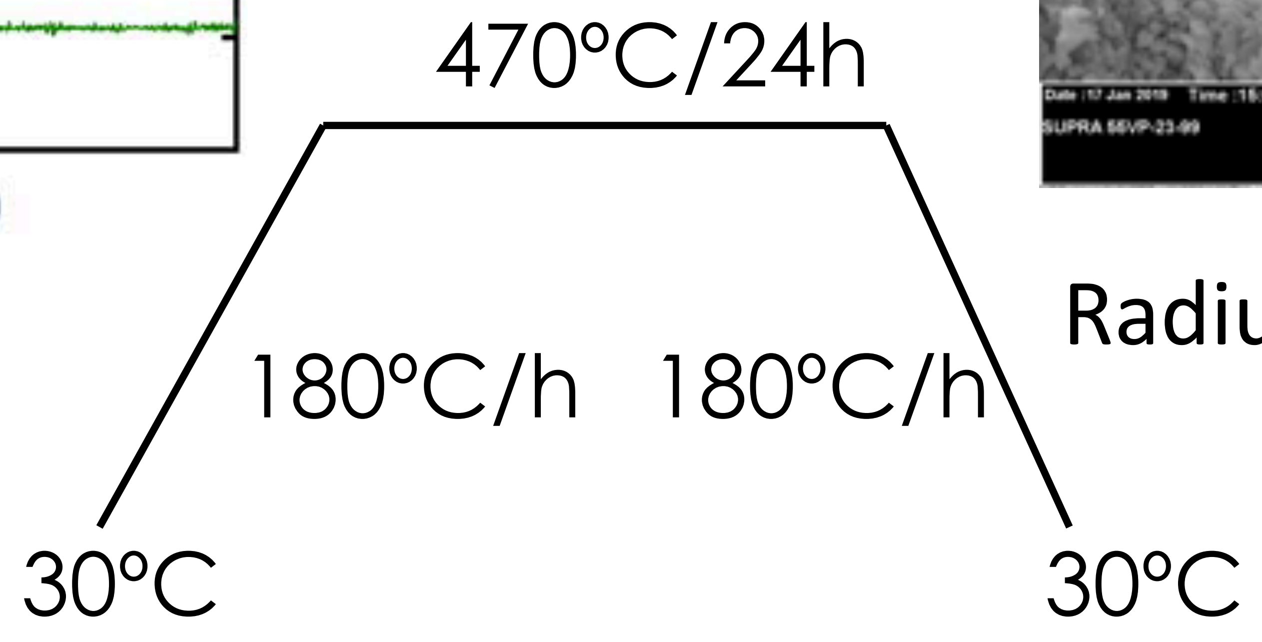




Radius of nanoparticles ~ 1 μm







Radius of nanoparticles (35±5) nm

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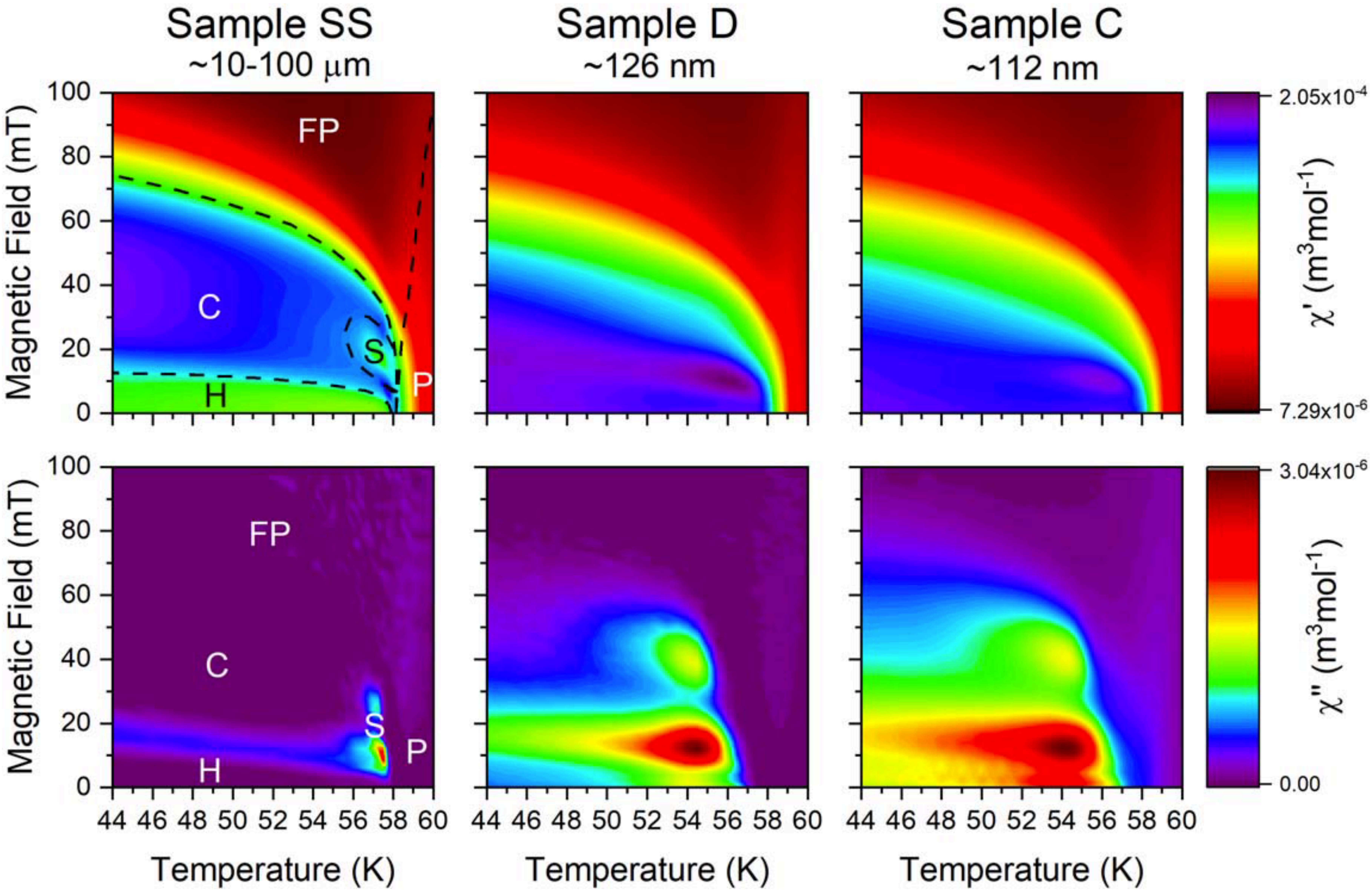

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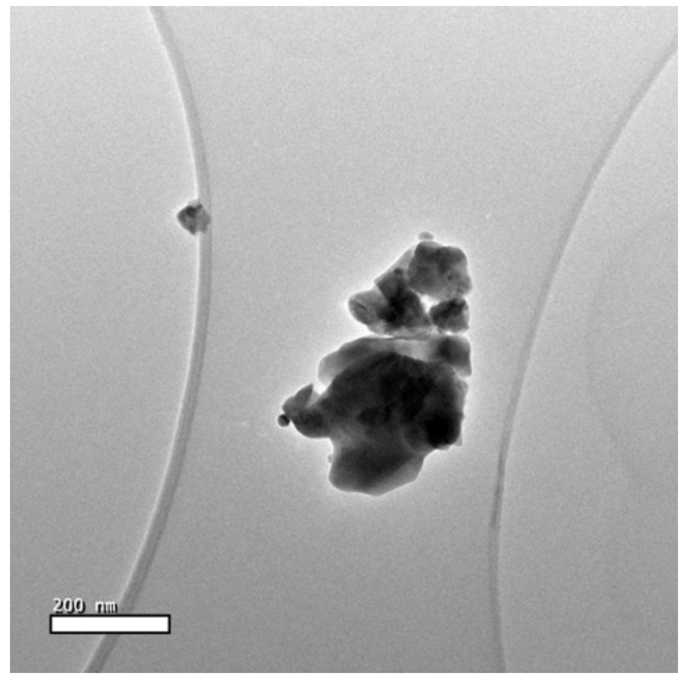
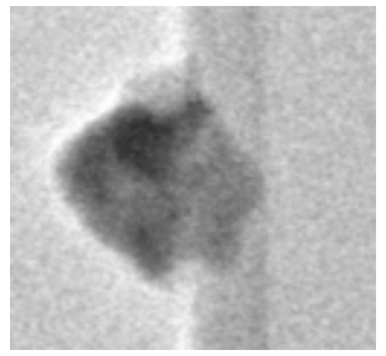
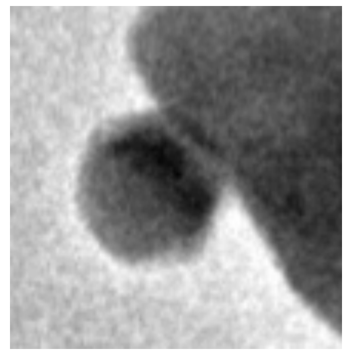
TEM of the nanoparticles

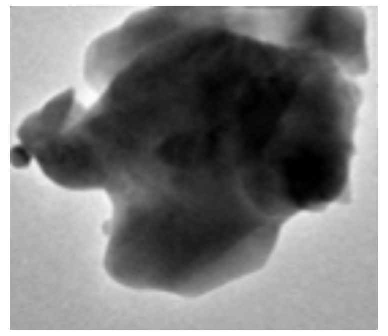




~53 nm diameter



~19 nm diameter



~250 nm diameter

- Inconclusive results, no skyrmions observed
- Search in small nanoparticles by holography
- Larger nanoparticles prove too thick for clear signal
- Roughness of the nanoparticles poses problems for resolving magnetic phase

Tuning topological spin textures in size-tailored chiral magnet insulator particles
PR Baral et al The Journal of Physical Chemistry C,126 (28), 11855-11866 (2022)

S J R Holt et al Materials Research Express 8 116101 (2021)

Summary and Conclusions

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- Crystals exhibit skyrmion lattices, and other spin structures. (Chiral Soliton lattices)

- Skyrmions in centrosymmetric materials.

- Possibilities of observing skyrmion phases in several layered/2D materials.

- Skyrmions in nanoparticles?

- Skyrmions close to RT? Search for new materials goes on….

https://go.warwick.ac.uk/supermag

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X-ray Diffraction Group RTP

Clemens Ritter

THE SKYRMION PROJECT

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THANK YOU

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