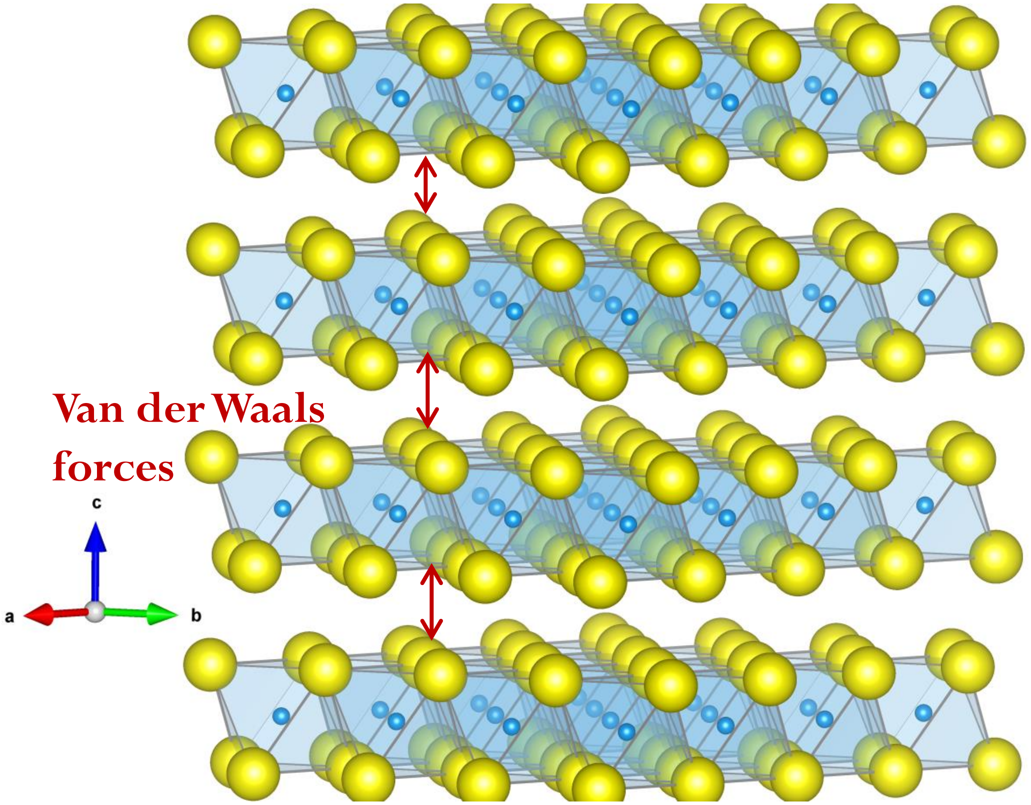
Sino-Russia meeting on frontiers of neutron scattering (SRNS-2024)
October 8–11, 2024, Ekaterinburg

Magnetic states of iron-containing transition metal chalcogenides

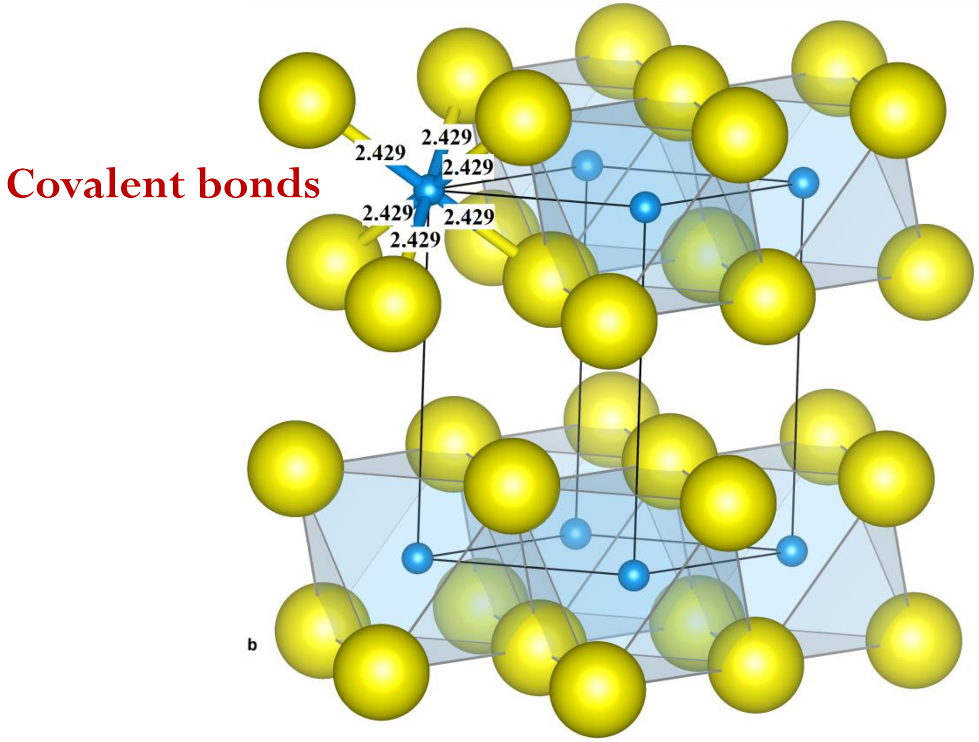
Nosova N.M., Selezneva N.V., Gubkin A.F., Baranov N.V.

Transition metal chalcogenides MX_2



CdI_2- type

1. The possibility of delamination and obtaining graphene-like monolayers

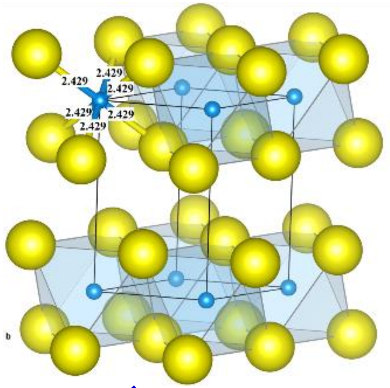


2. Ability to insert (intercalate) other atoms or structural fragments

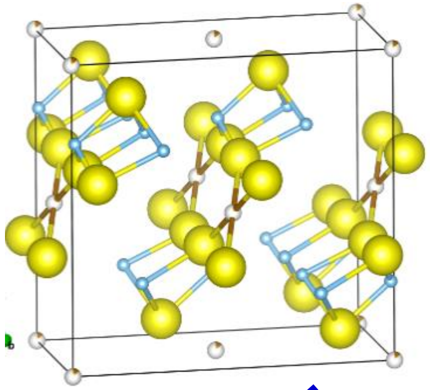
Structure types

Ordering of Fe atoms in layers leads to the formation of different superstructures

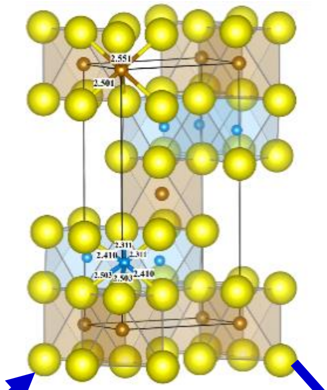
CdI_2



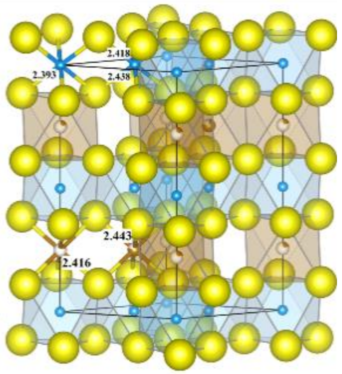
M_5X_8



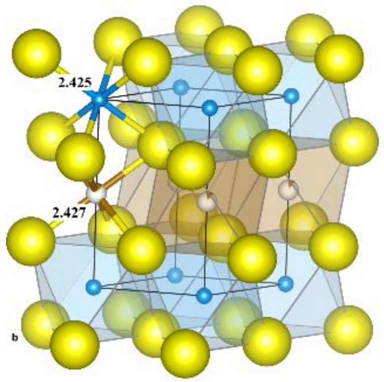
M_3X_4



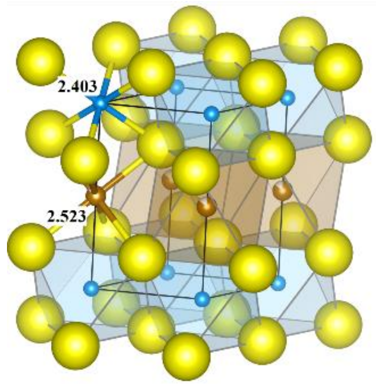
CdI_2



M_2X_3

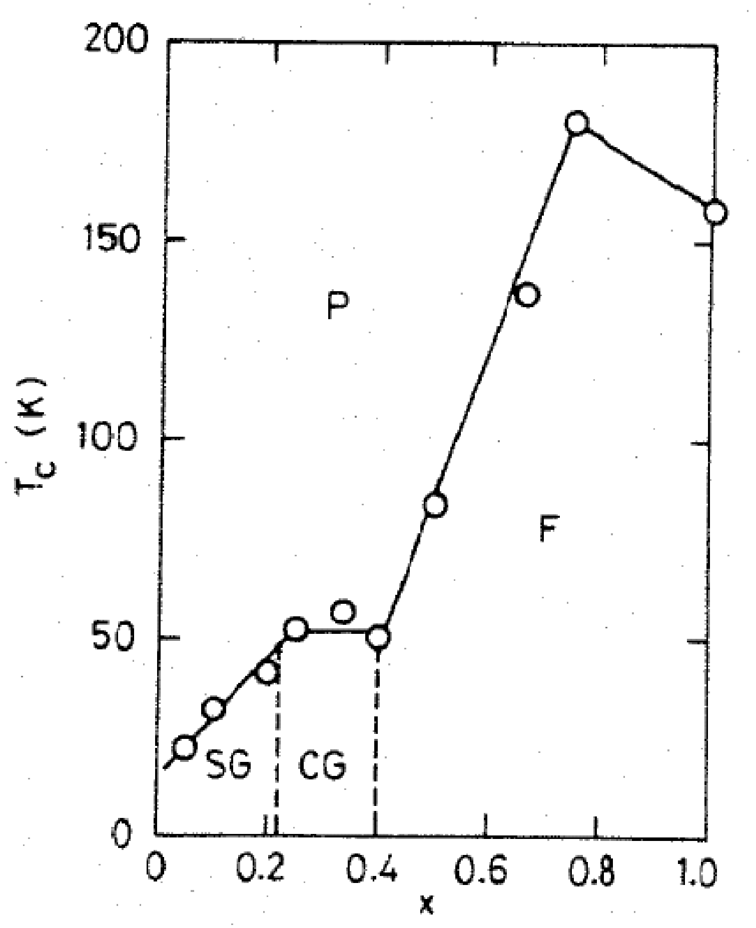


NiAs

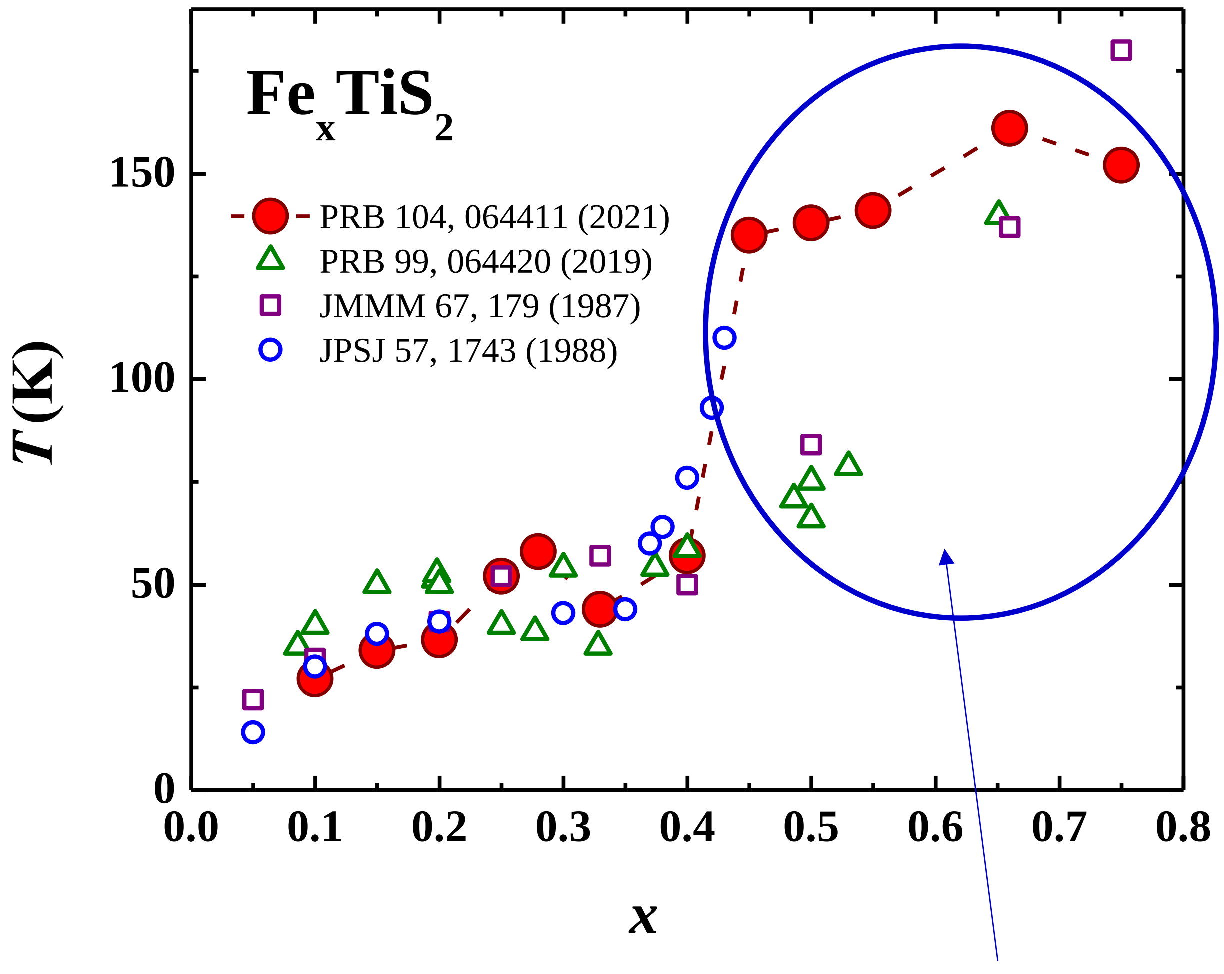


Puzzle #1

Concentration dependence of critical temperatures of magnetic transformations in the system Fe_xTiS_2

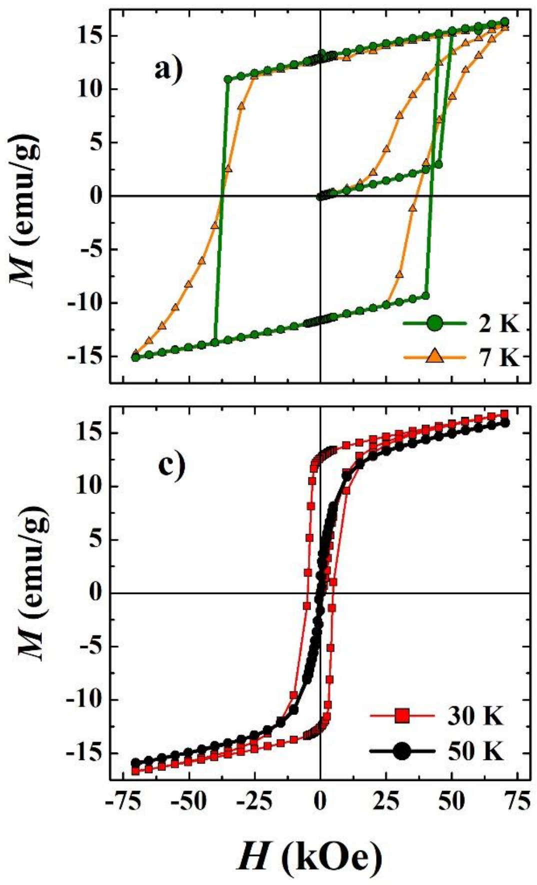


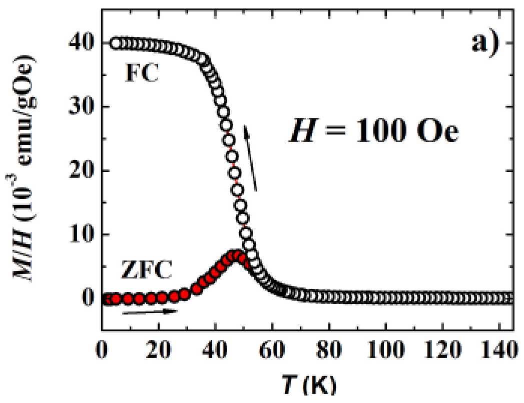
M. Inoue et al., Advances in Physics, 38 (1989) 565

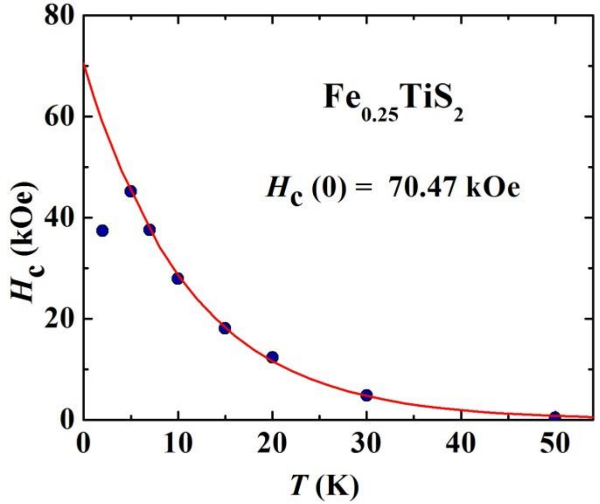


a discrepancy in the data

$Fe_{0.25}TiS_2$: magnetization behavior





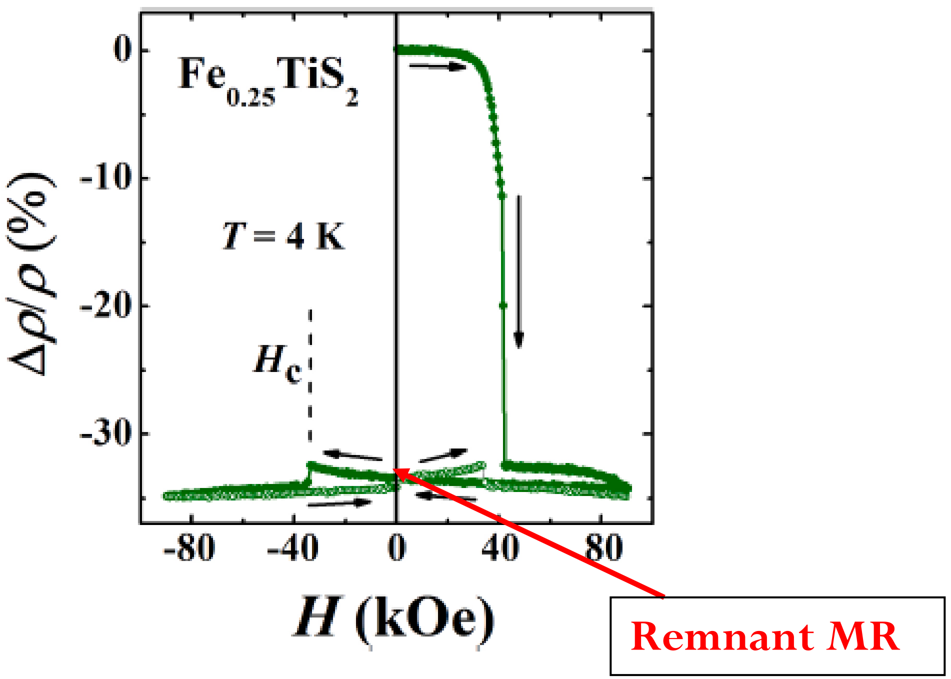


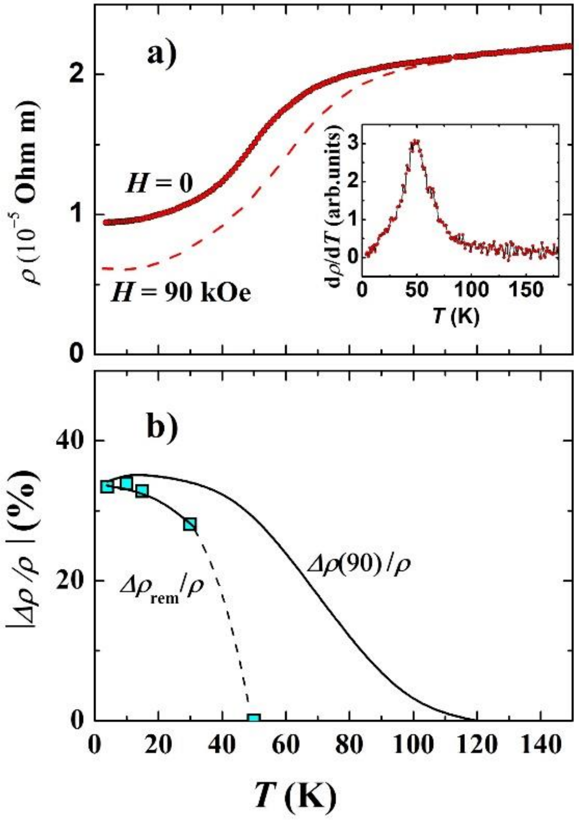
$$H_c(T) = H_c(0) \cdot \exp(-\alpha T)$$

It looks like it's a ferromagnet !

Puzzle #1

$Fe_{0.25}TiS_2$: magnetoresistance





Giant MR implies it's an antiferromagnet !

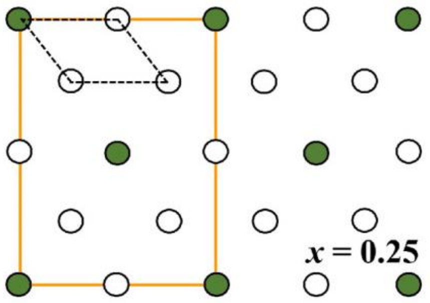
Field-induced phase transition to a metastable high coercive ferromagnetic state!

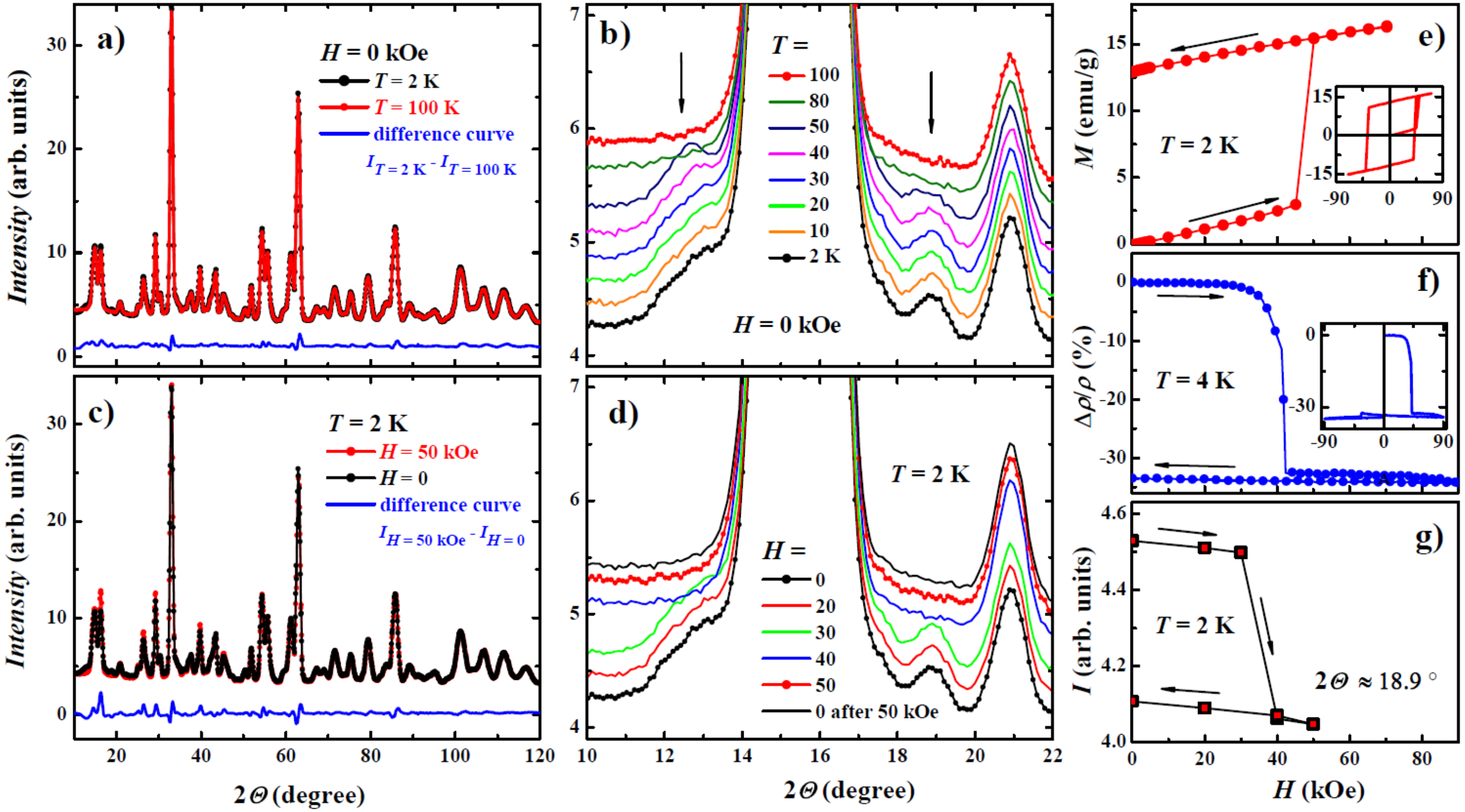
XMCD experiments: orbital magnetic moment in M_{orb} ~ 0.25 – 0.6 μ_B/Fe in Fe_xTiS_2 (Shibata et al., J.Phys. Chem. C 125 (23), (2021)12929)

$Fe_{0.25}TiS_2$: neutron diffraction

superstructure

$$2\sqrt{3} \times 2 \times 2$$





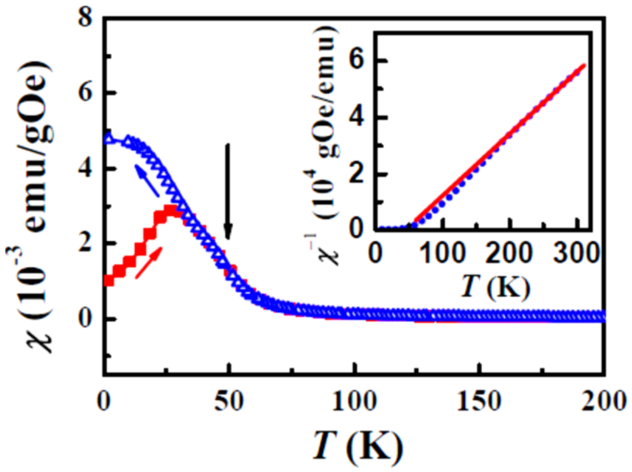
The appearance of new magnetic reflections upon cooling indicates the formation of an AFM order which is transformed to the FM state under application of a magnetic field.

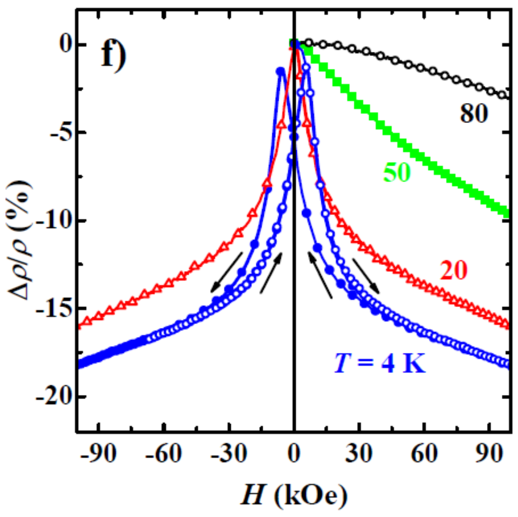
A. Podlesnyak, M. Frontzek
Neutron Scattering Division, Oak Ridge National Laboratory, Oak Ridge, USA

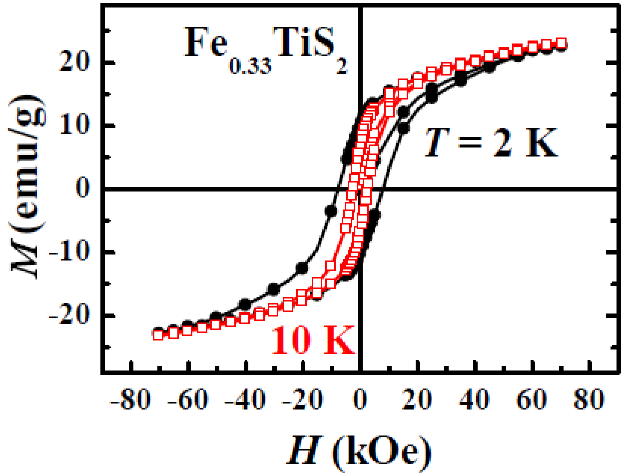
What happens if we increase the Fe content?

Puzzle #1

$Fe_{0.33}TiS_2$: absence of a long-range magnetic order

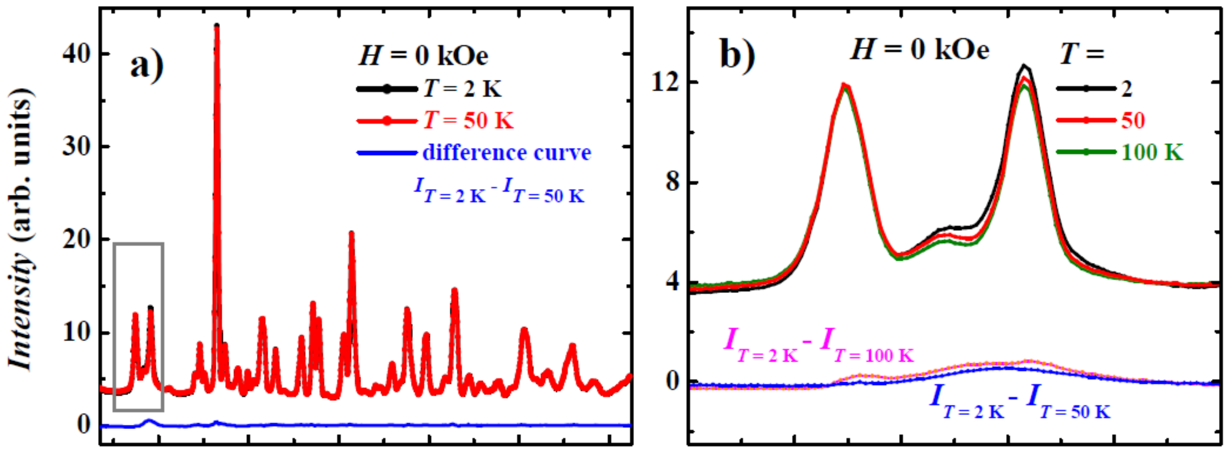






superstructure

$$\sqrt{3} \times \sqrt{3} \times 2$$



Cluster glass behavior! **No long-range magnetic order!**

Broad diffuse magnetic maximum indicates the appearance of short-range magnetic correlations with cooling below 50 K.

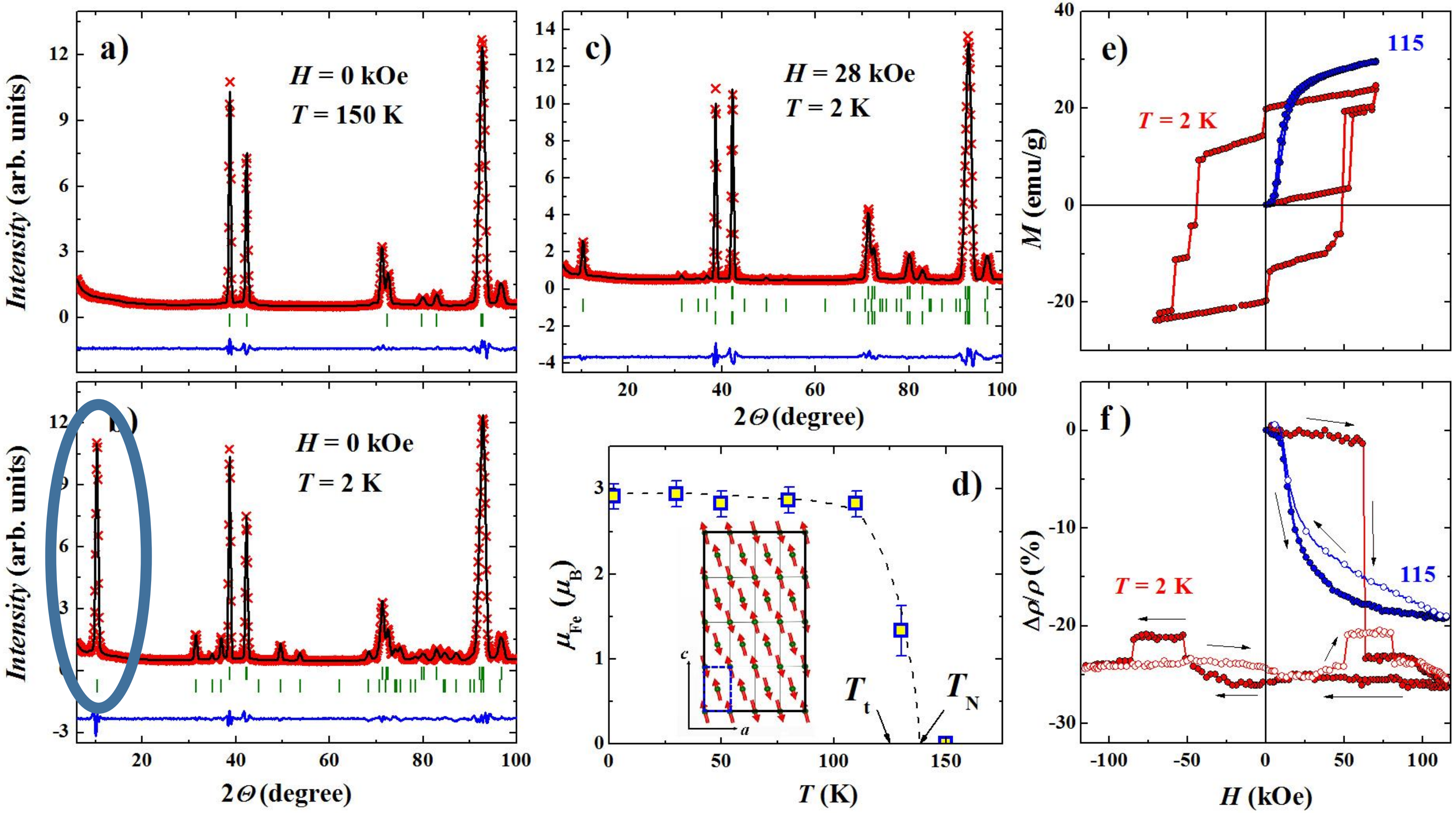
Absence of a long-range magnetic order may result from frustrations of exchange interactions due to the formation of a triangular network of Fe atoms in the *ab* plane

Puzzle #1

$Fe_{0.5}TiS_2$: reappearance of a long-range magnetic

superstructure

$$\sqrt{3} \times 1 \times 2$$

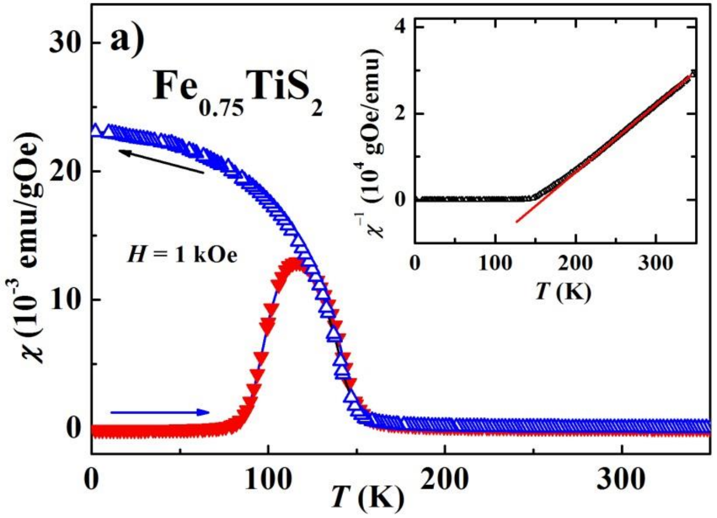


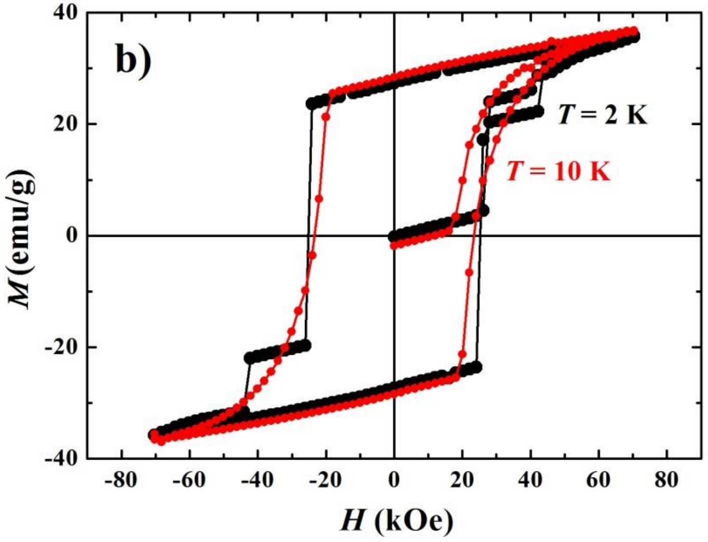
AFM order transforms into metastable FM state in applied magnetic fields.

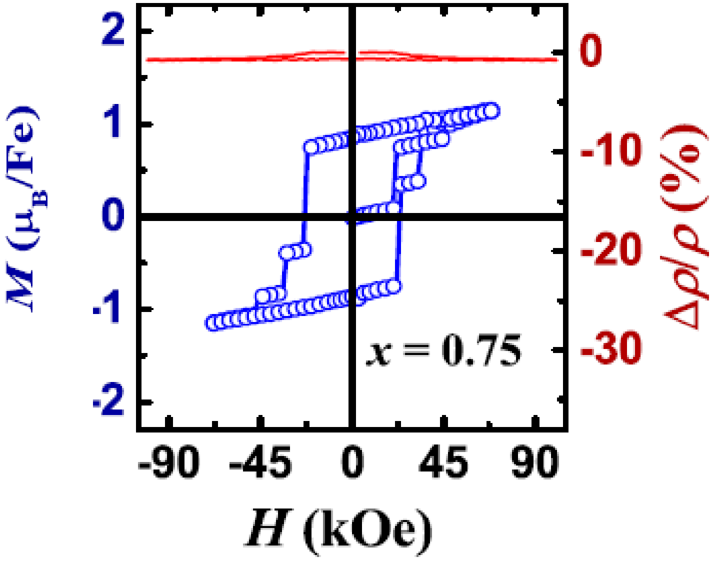
The field-induced FM state is stabilized by magnetoelastic interactions.

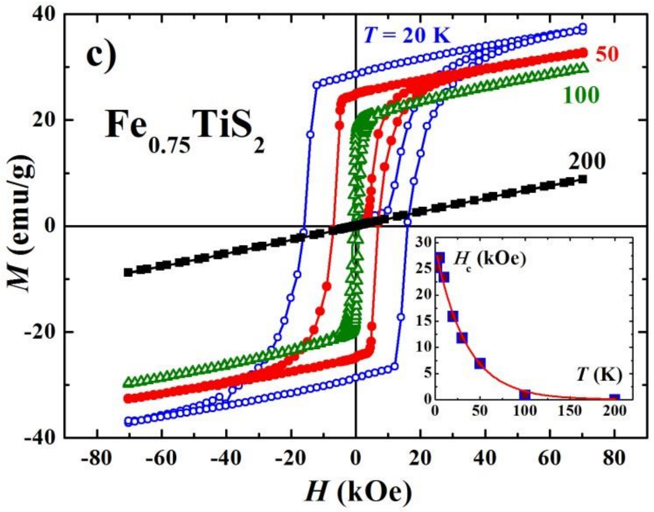
Puzzle #1

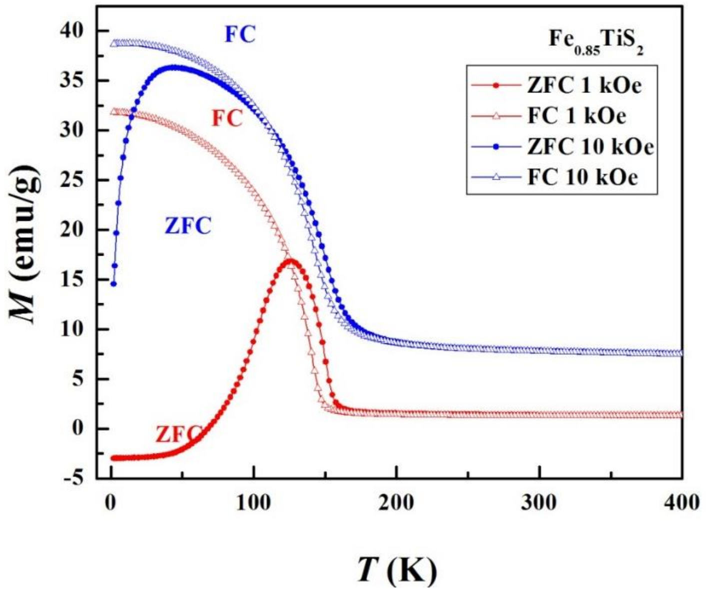
$Fe_{0.75}TiS_2$ and $Fe_{0.85}TiS_2$: ferrimagnetic ordering ?

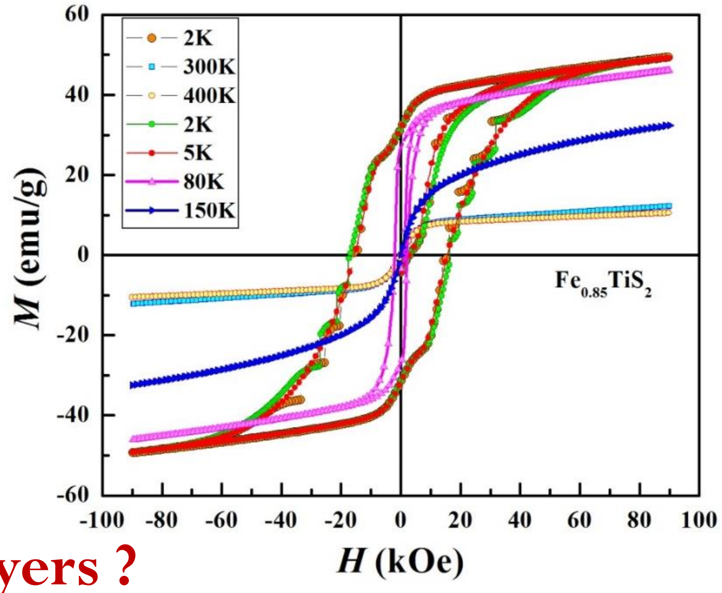










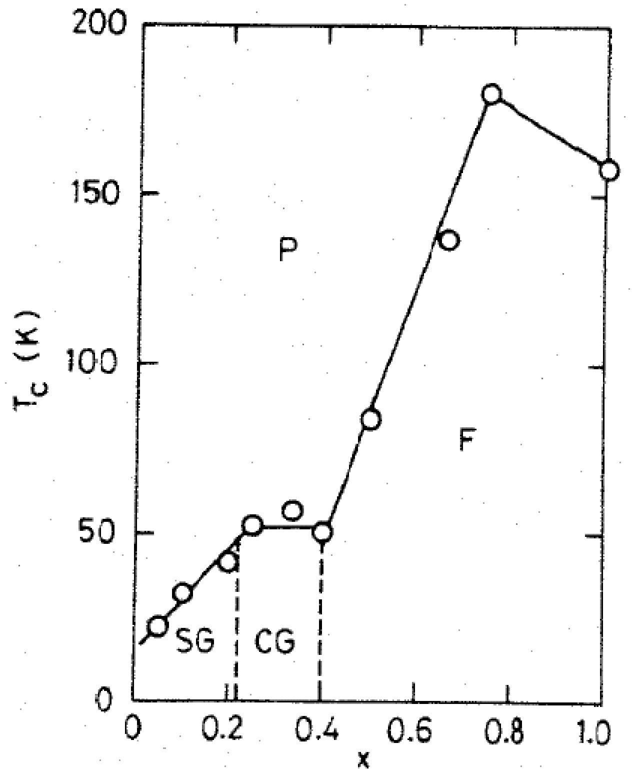


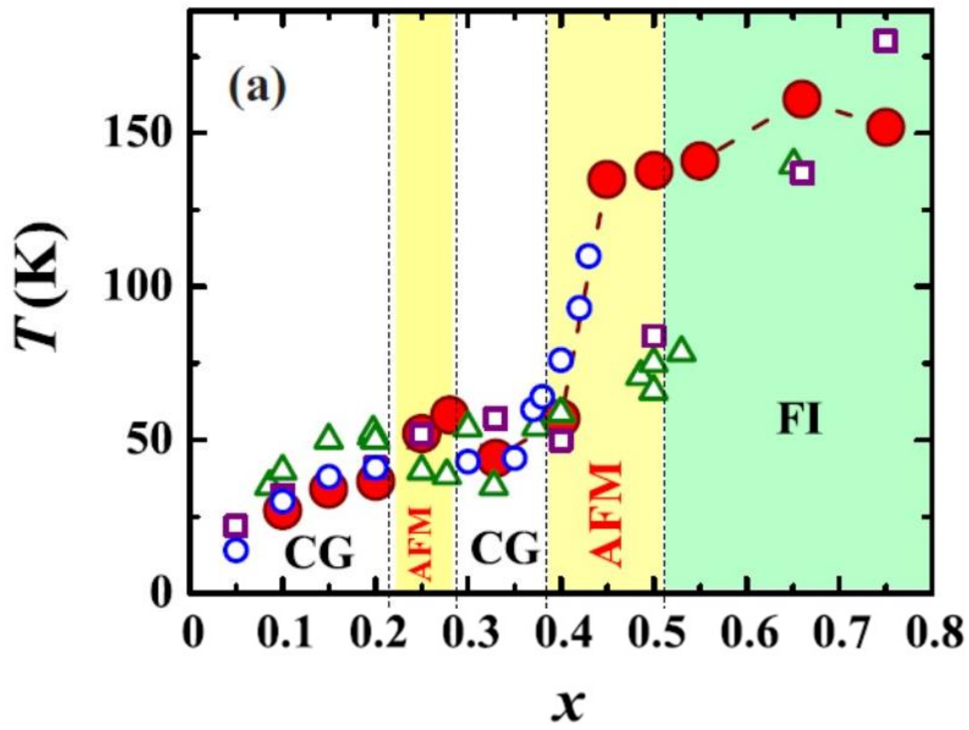
Mixing of Ti and Fe atoms in cationic layers ?

Puzzle #1

Fe_xTiS_2: changes in magnetic state and coercivity

CG → AFM → CG → AFM → FI *Iron content ↑*





Magnetic states and magnetic properties of Fe_xTiS_2 closely relate with the concentration of Fe atoms and their distribution over the lattice

Puzzle #1

PHYSICAL REVIEW MATERIALS **7**, 014401 (2023)

Relationship between magnetoresistance behavior and magnetic states in intercalated compounds Fe_xTiS_2

N. V. Selezneva,[1] E. M. Sherokalova,[1] A. Podlesnyak,[2] M. Frontzek,[2] and N. V. Baranov[1,3,*]
[1]*Institute of Natural Science and Mathematics, Ural Federal University, 620083 Ekaterinburg, Russia*
[2]*Neutron Scattering Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831, USA*
[3]*M.N. Miheev Institute of Metal Physics, Ural Branch of the Russian Academy of Science, 620108 Ekaterinburg, Russia*

PHYSICAL REVIEW B **104**, 064411 (2021)

Multiple magnetic states and irreversibilities in the Fe_xTiS_2 system

N. V. Selezneva,[1] N. V. Baranov,[1,2] E. M. Sherokalova,[1] A. S. Volegov,[1,2] and A. A. Sherstobitov[1,2]
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[2]*M.N. Miheev Institute of Metal Physics, Ural Branch of the Russian Academy of Science, Ekaterinburg 620108, Russia*

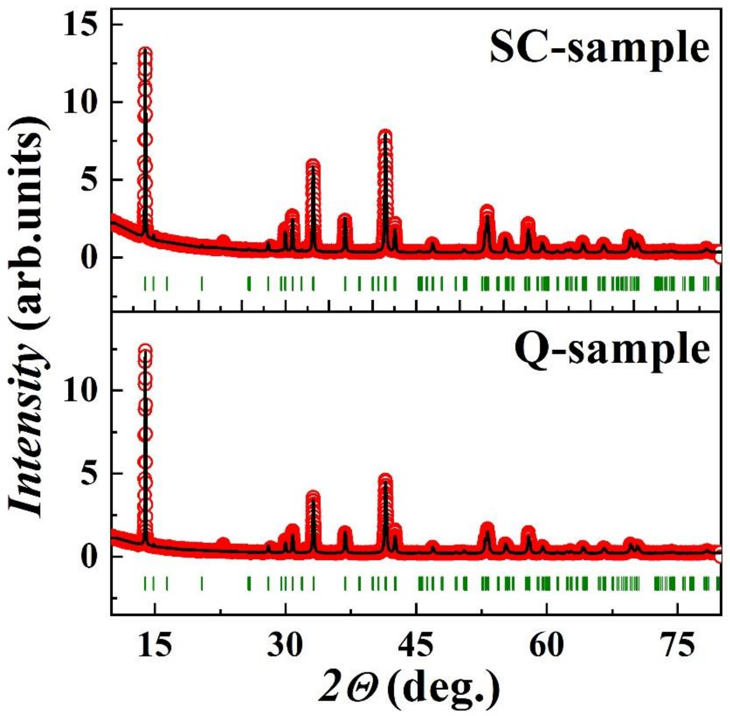
PHYSICAL REVIEW B **100**, 024430 (2019)

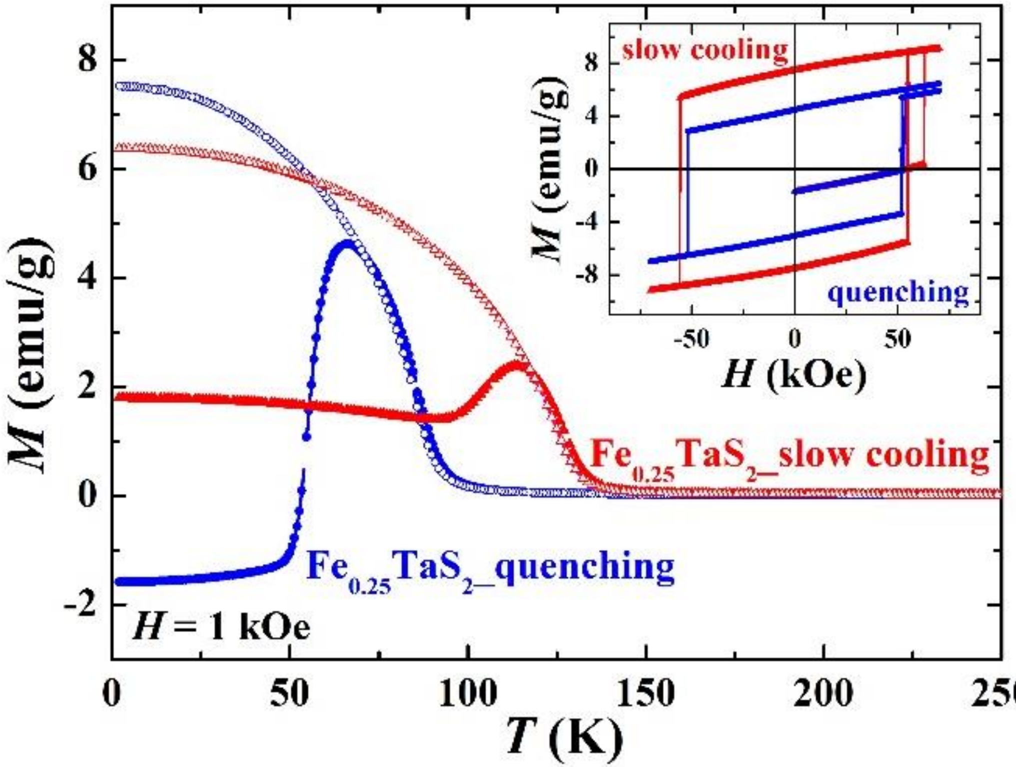
Magnetic phase transitions, metastable states, and magnetic hysteresis in the antiferromagnetic compounds $Fe_{0.5}TiS_{2-y}Se_y$

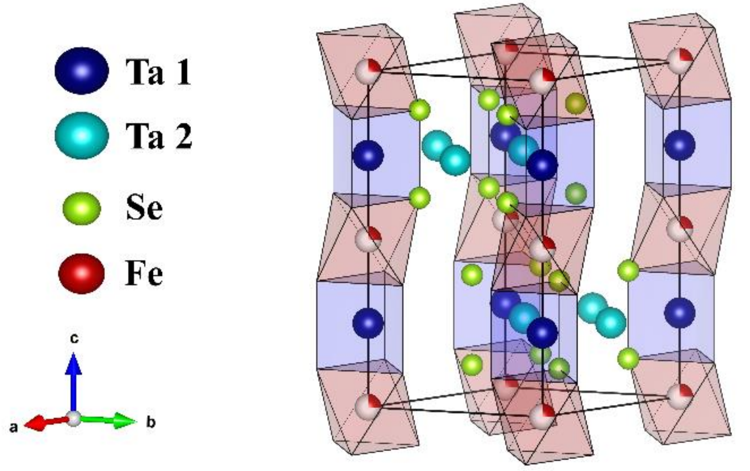
N. V. Baranov,[1,2] N. V. Selezneva,[2] E. M. Sherokalova,[2] Y. A. Baglaeva,[2] A. S. Ovchinnikov,[1,2] A. A. Tereshchenko,[2] D. I. Gorbunov,[3] A. S. Volegov,[1,2] and A. A. Sherstobitov[1,2]

Puzzle #2

Spin state of iron in intercalated and substituted layered compounds Fe_xTaCh_2 and $Fe_xTa_{1-x}Ch_2$ (Ch = S, Se)



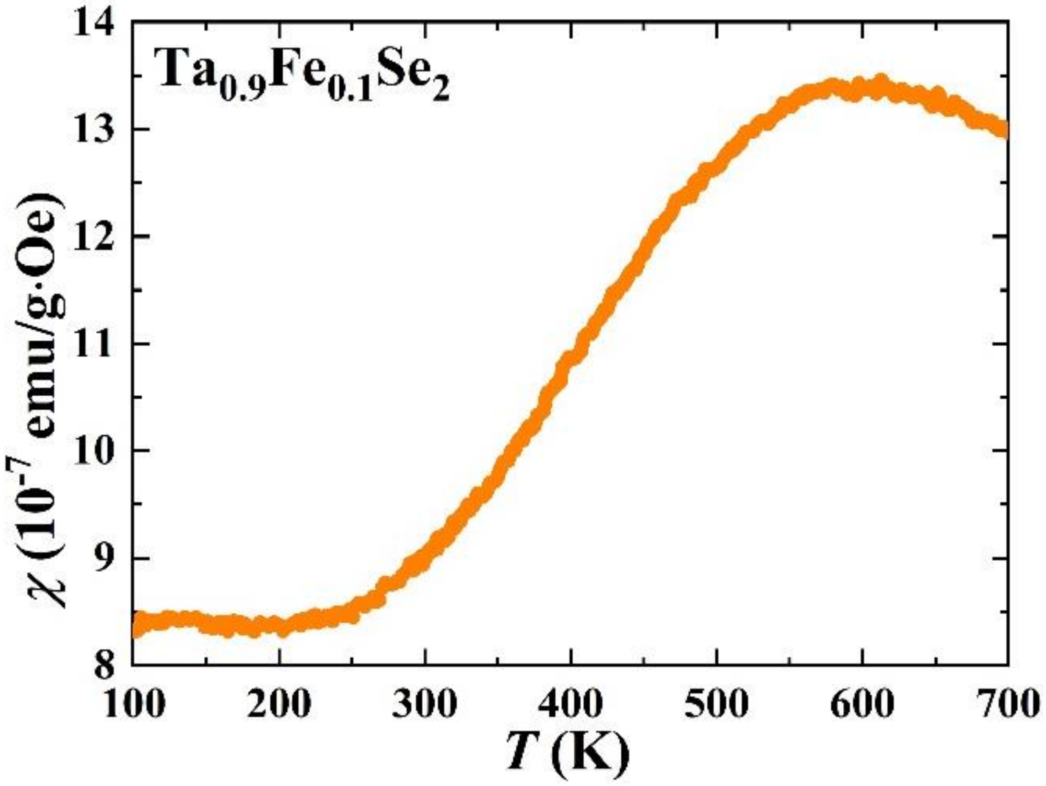




Various methods of preparation and heat treatment do not have a strong effect on the lattice parameters of the main phase in the samples, but they significantly affect the magnetic critical temperature and magnetic hysteresis of the samples

Puzzle #2

Spin state of iron in intercalated and substituted layered compounds Fe_xTaCh_2 and $Fe_xTa_{1-x}Ch_2$ (Ch = S, Se)

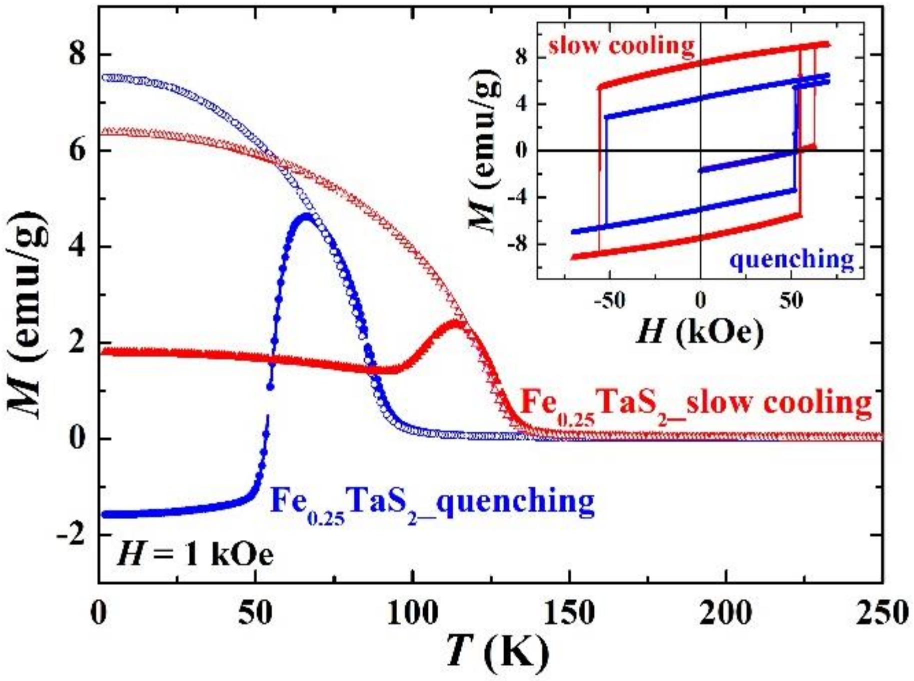


Fe atoms:

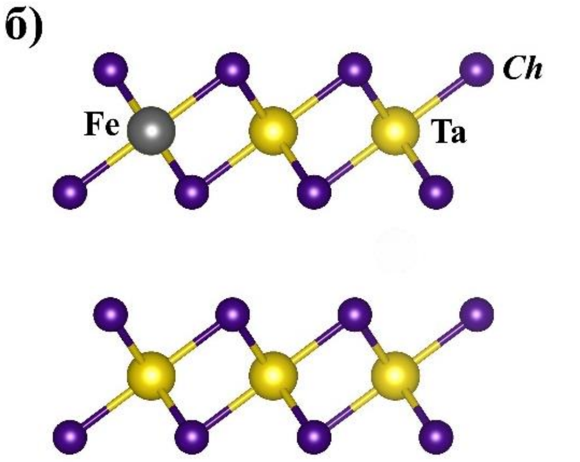
the transition from low-spin (LS) to high-spin (HS) state with increasing temperature in the range 200 – 400 K*

[*] K-T. Ko et al., Phys. Rev. Lett. **107**, 247201 (2011)

Puzzle #2

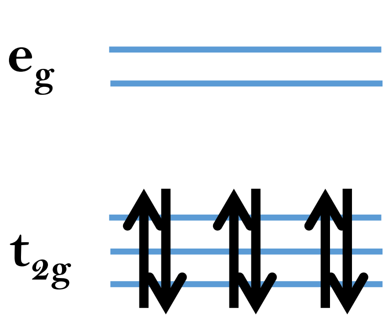


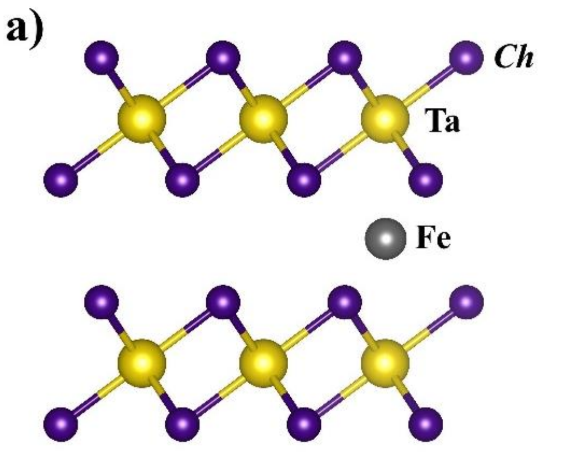
The observed distinctions can be ascribed to the difference in the distribution of Fe atoms over the crystal lattice



Low-spin (LS) state Fe^{2+} ($3d^6$)

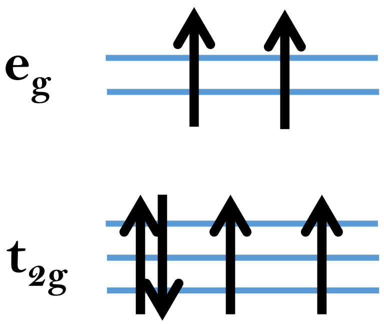
$S = 0$





High-spin (HS) state Fe^{2+} ($3d^6$)

$S = 2$



Depends on the splitting between t_{2g} and e_g states in the crystal field

Puzzle #2

Physica B 673 (2024) 415492

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ELSEVIER

Crystal structure, magnetic and transport properties of $Fe_{0.25}TaSe_2$

N.M. Nosova [a,*], N.V. Selezneva [a], D.A. Shishkin [a,b], N.V. Baranov [a,b]

[a] Institute of Natural Science and Mathematics, Ural Federal University, 620083, Ekaterinburg, Russia
[b] M.N. Miheev Institute of Metal Physics, Ural Branch, Russian Academy of Science, 620108, Ekaterinburg, Russia

It is necessary to reveal the influence of the crystal-field effects on the spin state of iron in the intercalated and substituted compound based on Ta*Ch*$_2$

Iron-containing transition metal chalcogenides

1. N.V. Baranov, E.M. Sherokalova, N.V. Selezneva, A.V. Proshkin, A.F. Gubkin, L. Keller, A.S. Volegov, E.P. Proskurina Magnetic order, field-induced phase transitions and magnetoresistance in the intercalated compound **$Fe_{0.5}TiS_2$** // Journal Of Physics-Condensed Matter. 2013. V. 25, P. 066004 (9).

2. A.F. Gubkin, E.M. Sherokalova, L. Keller, N.V. Selezneva, A.V. Proshkin, E.P. Proskurina, N.V. Baranov Effects of S-Se substitution and magnetic field on magnetic order in **$Fe_{0.5}Ti(S,Se)_2$** layered compounds // Journal Of Alloys And Compounds. 2014. V. 616, P. 148-154.

3. N.V. Selezneva, E.M. Sherokalova, A.S. Volegov, D.A. Shishkin, N.V. Baranov Crystal structure, magnetic state and electrical resistivity of **$Fe_{2/3}Ti(S,Se)_2$** as affected by anionic substitutions // Materials Research Express. 2017. V. 4, P. 106102 (17).

4. N.V. Baranov, N.V. Selezneva, E.M. Sherokalova, Y.A. Baglaeva, A.S. Ovchinnikov, A.A. Tereshchenko, D.I. Gorbunov, A.S. Volegov, A.A. Sherstobitov Magnetic phase transitions, metastable states, and magnetic hysteresis in the antiferromagnetic compounds **$Fe_{0.5}TiS_{2-y}Se_y$** // Physical Review B. 2019. V. 100, P. 024430 (14).

5. N.V. Selezneva, N.V. Baranov, E.M. Sherokalova, A.S. Volegov, A.A Sherstobitov Remnant magnetoresistance and virgin magnetic state in **$Fe_{0.25}TiS_2$** // Journal Of Magnetism And Magnetic Materials. 2021. V. 519, P. 167480 (8).

6. N.V. Selezneva, N.V. Baranov, E.M. Sherokalova, A.S. Volegov, A.A Sherstobitov Multiple magnetic states and irreversibilities in the **Fe_xTiS_2** system // Physical Review B. 2021. V. 104, P. 064411 (14).

7. N.V. Selezneva, V.S. Nosovets, E.M. Sherokalova, D.A. Shishkin, N.V. Baranov Crystal structure and properties of layered compounds **$Fe_{0.75}TiS_{2-y}Se_y$** // Solid State Sciences. 2022. V. 134. P. 107049 (8).

8. N.V. Selezneva, E.M. Sherokalova, A. Podlesnyak, M. Frontzek, N.V. Baranov Relationship between magnetoresistance behavior and magnetic states in intercalated compounds **Fe_xTiS_2** // Physical Review Materials. 2023. V. 7, P. 014401 (11)

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Associate Professor

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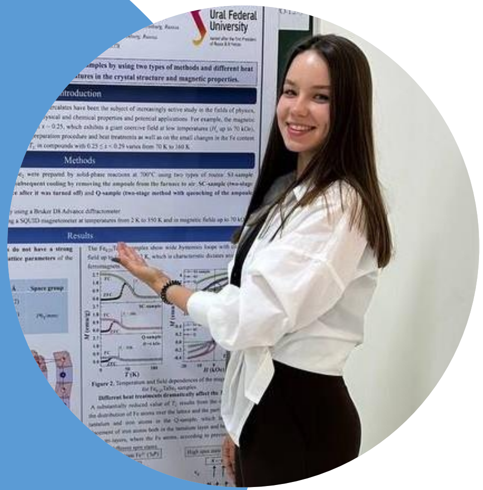

Junior Researcher

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Thank you for your attention!

Supported by the RSF (Grant No. 22-13-00158)

Ural Federal University
named after the first President of Russia B.N.Yeltsin

Sample preparation and research methods :

- **Polycrystalline samples were synthesized by solid-phase reaction method. Single crystals by using a modified Bridgman method.**
- **X-Ray examination: Bruker D8 Advance x-ray diffractometer.**
- **Neutron powder diffraction measurements in magnetic fields up to 50 kOe (WAND diffractometer, High Flux Isotope Reactor, ORNL, USA), and DMS diffractometer H = 28 kOe, Spallation Source SINQ, PSI, Switzerland).**
- **DC magnetization measurements: SQUID MPMS (70 kOe), MPMS (90 kOe), Quantun Design, USA**
- **The transversal magnetoresistance in magnetic fields up to 100 kOe, DMS-1000 system, Dryogenic Ltd, UK.**