Layer-Dependent Magnetic Ordering in Natural 2D Magnetic Insulators

Muhammad Zubair Khan[1,*], Oleg E.Peil[2], Sebastian Wintz [3], Markus Weigand[3], Sergio Valencia[3], Florian Kronast[3], Maik Zimmermann[4], Muhammad Awais Aslam[1], Johann G. Raith[4], Christian Teichert[1], Aleksandar Matković[1,*]

[1]Chair of Physics, Department Physics, Mechanics and Electrical Engineering, Montanuniversitat Leoben, Austria.
[2]Group of Computational Material Design, Materials Center Leoben Forschung GmbH, Leoben, Austria.
[3]Department of Spin and Topology in Quantum Materials, Helmholtz-Zentrum Berlin, Germany.
[4]Chair of Resource Minerology, Montanuniversitat Leoben,Austria.

Motivation

Magnetic monolayers show great promise for future applications in nanoelectronics, data storage and sensing. Especially for the applications in magentoresistive tunneling junctions, **2D magnetic insulators** are very attractive material candidates. They enable atomically sharp interface and preservation of long range magnetic ordering down to monolayer thickness. While the research mainly focus on synthetic iodide and telluride based compounds, naturally occuring layered materials are vastly overlooked. Here we explore **magnetic ordering in iron-rich phyllosilicates**. These layered systems are inherently magnetic, **ambient stable** and can be thin down to monolayers. [1,2]

Structure of phyllosilicates

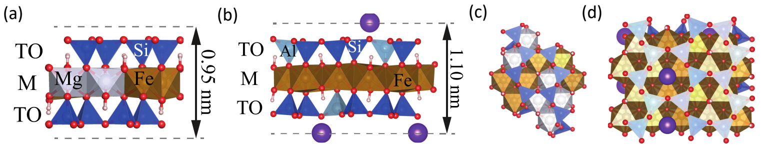


Figure 1: (a,b) side and (c,d) top views of (a,c) Fe-substituted talc- minnesotaite and (b,d) Fe-substituted mica- annite. Presented are relaxed structures obtained from ab-initio calculations. The octahedral units with substituted Fe are indicated by orange colour, and Mg containing octahedrals are shown in grey. The interlayer species (K) are represented as purple spheres, Si and Al tetrahedral groups are shown in dark and light blue respectively. The H and O atoms are denoted as white and red spheres.

Local magnetic moment

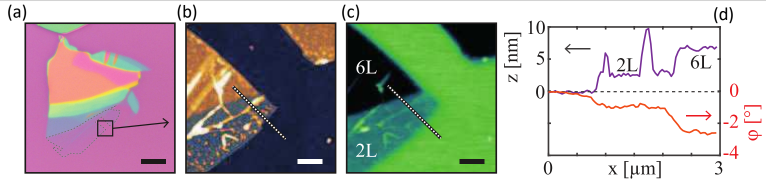
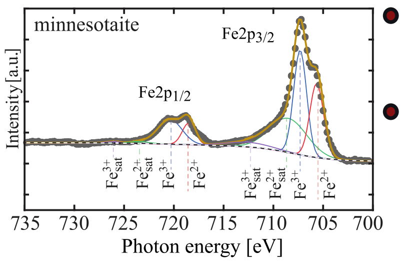


Figure 2: (a) optical micrographs (60 x 60 μm^2 scale bar 10 μm) of exfoliated annite. Dashed lines indicate the thin thin flake region and the solid square mark the area from which the AFM micrographs are presented. (b) AFM topography image of exfoliated flakes (6x6 μm^2, scale bar 1μm, z-scale 15 nm), (c) second-pass phase lag in 180 mT out-of-plane external field,(d) flake height (left y scale) and MFM phase lag (right y scale) cross-sections.

Fe incorporation in phyllosilicates

X-ray absorption Spectroscopy (XAS)



- XAS spectra indicates the partial occupation of Fe^{2+} in high spin configuration, which is evident from satellite peak.
- Two-fold effect of Fe^{3+}:
  - Enhancing ferromagnetic interactions.
  - Reducing the crystalline anisotropy.

Figure 3: XAS spectrum of minnesotaite recorded by photoemission electron microscope (PEEM) across Fe $L_{3,2}$-edge. XAS shows that Fe is present in both Fe^{2+} and Fe^{3+} oxidation states.

Raman spectral features of phyllosilicates

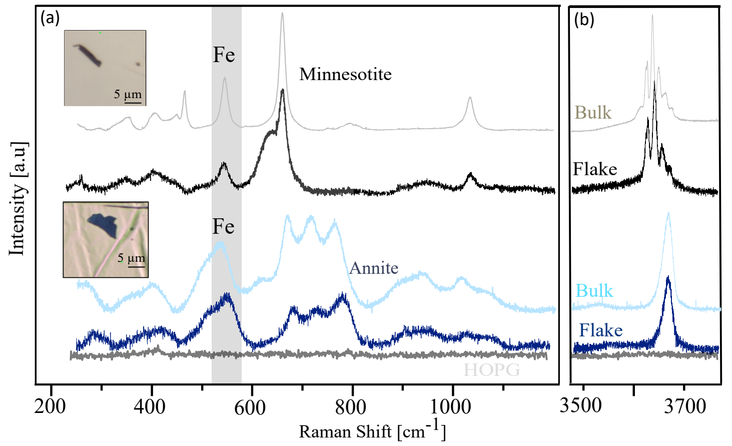


Figure 4: Raman Spectra of thin Fe-rich phyllosilicates showing the (a) fundamental vibrations (250-1200cm^{-1}),and (b) OH vibrations (3500-3700 cm^{-1}). Inset present optical micrographs of the measured flakes on HOPG. Above each each spectrum of exfoliated flakes, reference spectra of bulk mineral specimen are shown.

Layer dependent magnetic ordering

X-ray magnetic circular diochroism (XMCD)

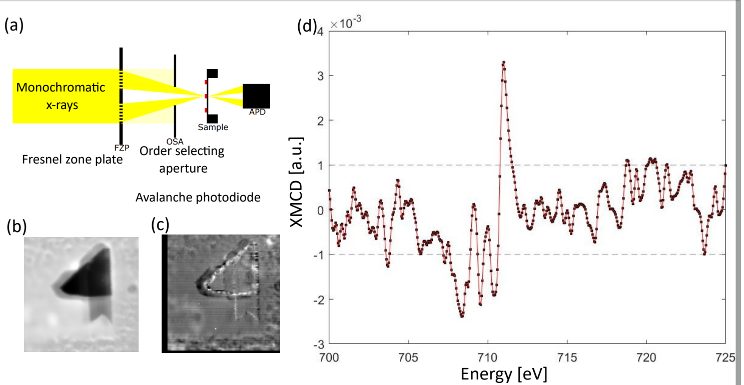


Figure 5: (a) Schematic diagram of Scanning transmission X-ray microscope (STXM), (b) STXM image of flake, (c) XMCD map corresponding flake at 709 eV, (d) XMCD spectra obtained from pre-selcted area.

- Very weak XMCD contrast was observed (from selected ROIs) with intensity of about 10^{-3} of the total signals.
- The actual sample temperature is very likely somewhat higher than the indicated sample holder temperature, having the sample just at the limit of reaching magnetic ordering.
- Due to a minute differences in the positive and negative helicity of X-ray spectra, mapping at several pre-selected energies was carried out to obtain XMCD contrast.

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Contact

aleksandar.matkovic@unileoben.ac.at
muhammad.khan@unileoben.ac.at
unileoben.ac.at/matkovics-lab/
@2D_Mat_Lab

References

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2. Khan, MZ, et al. Advanced Physics Research, (2023) DOI.org/10.1002/apxr.202300070