Magnetic properties of nanotextured greigite

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1 Introduction

Greigite (Fe_3S_4) is a ferrimagnetic mineral widespread in sedimentary environments, where it can record ancient geomagnetic field variations and environmental processes.

At this time, the magnetic properties of greigite are not well understood, because greigite generally forms poly-crystalline particles, i.e., idiomorph single crystal is not avaliable

In particular, the dependency of greigite **magnetic properties** on its **textural** properties, i.e., poly-crystallinity, remains uncertain

2 Structure and Morphology

In the present study, we analyzed the structural and magnetic properties of synthetic, polycrystalline greigite formed by controlled colloidal synthesis[1]. X-ray diffractometry and transition electron microscopy reveal that greigite forms flakes of about 100 nm that consist of epitaxially intergrown nanoparticles with a mean coherence length of 19 ± 2 nm. Therefore, our synthetic greigite can be considered as polycrystalline flakes with a nanotexture.

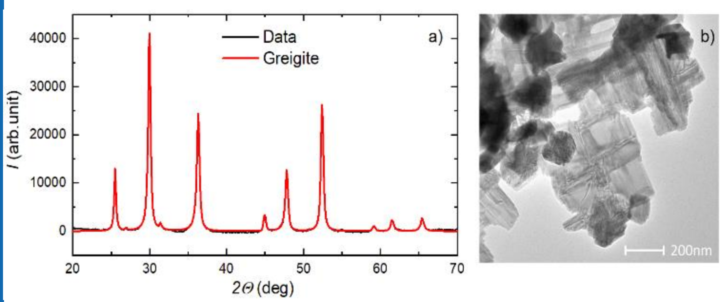

Fig. 1: a) Powder X-ray diffraction patterns measured and calculated of the greigite sample; b) TEM picture showing the flake morphology of the synthesized greigite.

3 Magnetic properties

The saturation magnetization (M_S) of the nanotextured greigite is 32.7 Am2 kg-1 and the coercivity is B_C = 41 mT (fig. 2a). The M_S is about 45% below the value for relatively large, synthetic, crystals and this in turn is probably a textural effect, e.g., interfaces between nanocrystallites. The ratios M_R / M_S = 0.54 and B_{CR} / B_C = 1.33 indicate single-domain (SD) particles with pre-dominant uniaxial anisotropy[2]. The FORC diagram at room temperature shows an oval contour plot indicating that the flakes consist of interacting crystallites in SD magnetic state (fig.2b). Moreover, the hysteresis parameters B_C and M_S continuously increase upon cooling to 10 K (data not shown).

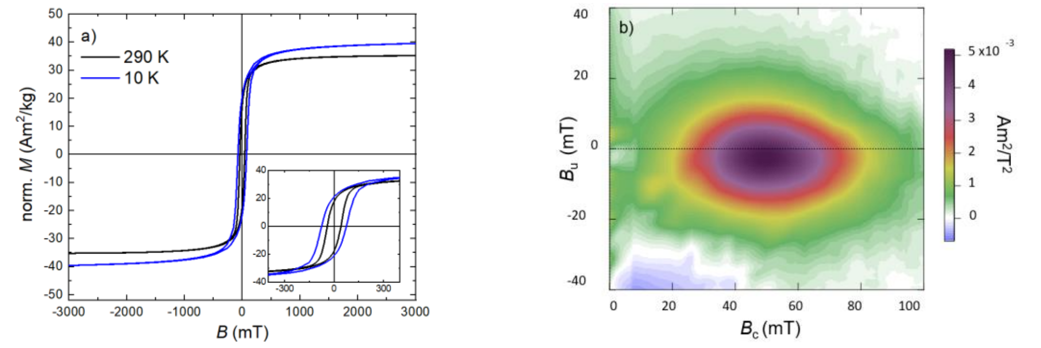

Fig. 2. a) Hysteresis curves at 300 K and 10 K and enlarged range of the hysteresis loop (inset) b) FORC diagram obtained by processing of magnetic data recorded with a VSM in fields up to 1 T.

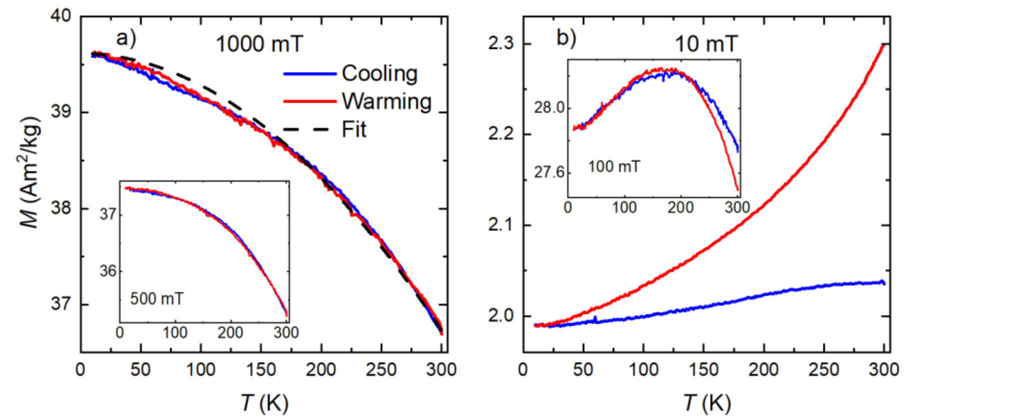

Fig. 3. Low-temperature magnetization cycling (cooling in blue and heating in red); a) curves recorded in 1000 mT field and their fit with the modified Bloch law (dashed line), curves with B = 500 mT (inset); b) cycling at low fields of 10 mT and 100 mT (inset).

Low-temperature cycling of the magnetization between 300 K and 10 K in fields between 10 mT and 1000 mT shows the expected behavior for ferrimagnets with the superposition of the cooling and warming curves at fields B > 500 mT. At weaker fields a slight magnetic increment upon warming is found and the relative increase in magnetization is field dependent. This irreversibility most likely stems from the magnetization of the nanoparticle interfaces and their interactions in the flakes.

4 Ferromagnetic Resonance (FMR) spectroscopy

Ferromagnetic resonance spectroscopy (FMR) at room temperature reveals angular dependence of the absorption intensity which can be subdivided into a low- and a high-field range (fig. 2a,b). Below 200 mT the absorption intensity changes with field orientation and the directional differences tend to zero with increasing B_{ex}. That indicates, that in the low-field range, the randomization of the flakes in the sample is broken. Upon cooling the B_{eff} decreases continuously down to 50 K followed by a pronounced shift to lower values down to 10 K (fig. 5). The shift goes along with markedly linewidth broadening. The strong change of the spectral parameters at T < 50 K points to a change in the effective anisotropy of the flakes most likely due to changes of the magnetocrystalline and the interaction anisotropies in the nanotexture, because the shape anisotropy of the polycrystalline flakes undergoes no significant change.

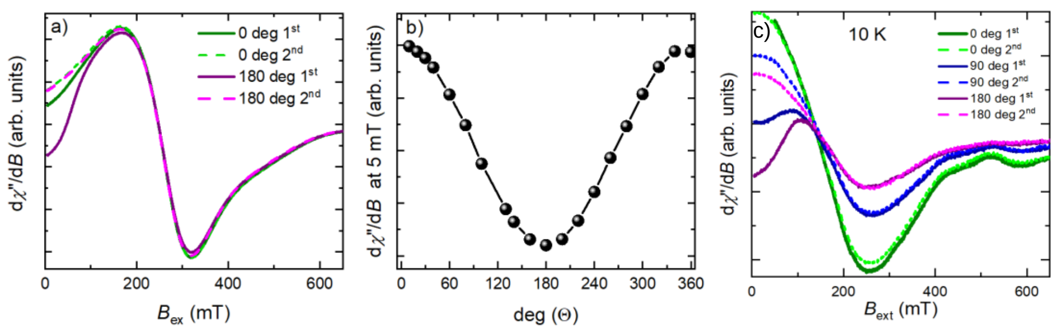

Fig. 4. a) FMR spectra at 300 K, initial one determined as 0° position and after 180° rotation with first scans (solid lines) and second scans (dashed lines), whereat the latter are identical, and the first scans vary in the low-field range at B_{ex} < 200 mT; b) angular dependence of the near zero-field absorption (B_{ex} = 5 mT) obtained from first scans. c) Angular dependence of the FMR spectra at 10 K, the first scans (solid lines) and the second scans (dashed lines) of the initial spectrum (green) and after rotations by 90°(blue) and 180° (purple).

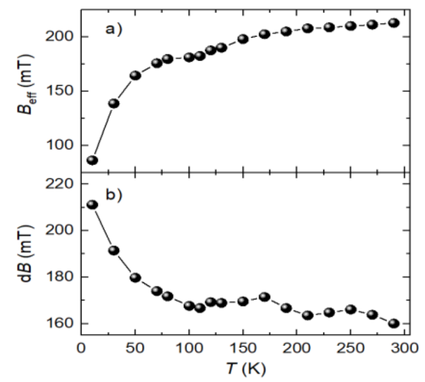

Fig.5. Development of the spectral parameters B_{eff} and ΔB at low temperatures with pronounced changes at $T \approx 50$ K.

5 Conclusion

Greigite reveals magnetic metastability that most likely stems from the **nanotexture**.

The effect of the nanotexture on the magnetization and anisotropy properties may allow to **identify and characterize greigite** nano-particles in natural environments and to critically evaluate their use for paleomagnetic studies.

6 References

1 Rhodes, Jordan M., et al. "Phase-controlled colloidal syntheses of iron sulfide nanocrystals via sulfur precursor reactivity and direct pyrite precipitation." Chemistry of Materials 29.19 (2017): 8521-8530.
2. Roberts, Andrew P. "Magnetic properties of sedimentary greigite (Fe3S4)." Earth and Planetary Science Letters 134.3-4 (1995): 227-236.

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