Magnetic properties of nanotextured greigite.

Barbara Lesniak¹, Michalis Charilaou², and Andreas Gehring¹

¹Institute of Geophysics, Department of Earth Sciences. ETH Zurich, 8093 Zurich, Switzerland (barbara.lesniak@erdw.ethz.ch)
²Department of Physics, University of Louisiana at Lafayette, Lafayette, LA 70504, USA

Greigite ($\text{Fe}_3\text{S}_4$) is a ferrimagnetic mineral widespread in sedimentary environments, commonly found in lacustrine and marine sediments that records ancient geomagnetic field variations and environmental processes. However, its magnetic properties are not yet well understood due to the lack of a single crystal greigite suitable for magnetic measurements. In particular, the dependency of its magnetic properties with respect to structural and morphological properties remains uncertain.

In the present study, we analyzed the structural and magnetic properties of synthetic, polycrystalline greigite formed by controlled colloidal synthesis [Rhodes et al. 2017]. X-ray diffractometry and transition electron microscopy reveal that greigite forms flakes of about 100 nm that consist of epitaxial intergrown nanoparticles with a mean coherence length of 19 nm. Therefore, our synthetic greigite can be considered as polycrystalline flakes with a nanotexture. The saturation magnetization ($M_s$) of the nanotextured greigite is $32.7 \text{Am}^2\text{kg}^{-1}$ and the coercivity is $B_c = 41 \text{mT}$. The $M_s$ is about 45% below the value for relatively large, synthetic crystal and this in turn is probably caused by the nanotexture, e.g., interfaces between nanocrystallites. The ratios $M_r/M_s = 0.54$ and $B_{ar}/B_{Sc} = 1.33$ indicate single-domain (SD) particles with pre-dominant uniaxial anisotropy [Roberts 1995]. The FORC diagram at room temperature shows an oval contour plot supporting that the flakes are nanotextured with interacting SD particles. The hysteresis parameters $B_c$ and $M_s$ continuously increase upon cooling to 10 K.

Low-temperature cycling of the magnetization between 300 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 $B \gtrsim 500 \text{mT}$. At weaker fields a slight magnetic induction 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.

Ferromagnetic resonance spectroscopy (FMR) at room temperature shows a resonance field $B_{res} = 213 \text{mT}$ and linewidth $\Delta B = 160 \text{mT}$. Upon cooling the $B_{res}$ decreases continuously down to 50 K followed by a pronounced shift to lower values down to 10 K. The shift goes along with markedly linewidth broadening. The discontinuity of the spectral parameters at $T < 50 \text{K}$ 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.

In summary, the magnetic properties of greigite can be critically affected by the nanotexture. The response of the nanotexture to the magnetization and anisotropy properties can be taken to identify and characterize greigite nanoparticles in natural environments and to critically evaluate their use for paleomagnetic studies.
