The effect of molecular inhomogeneity on the magnetization of a hematite-ilmenite solid solution with y = 0.83

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The magnetic ordering of hematite-ilmenite solid solutions ((1-*y*) Fe_2O_3 - *y* $FeTiO_3$; $0 < y < 1$) has been a topic of considerable interest for decades in different research fields (e.g. mineralogy, rock magnetism, material science). The end-members have a corundum structure and are antiferromagnetic with Néel temperatures of 948 K for hematite and 54 K for ilmenite. In a first approximation, the magnetic ordering temperature of the solid solutions exhibits a linear increase with increasing mole fraction of Fe_2O_3. Ilmenite-rich solid solutions ($1 > y > 0.5$), referred to as hemo-ilmenite, possess ferrimagnetic ordering; however, spin glass-like properties have also been found in hematite-ilmenite solid solutions for $y > 0.6$ (Ishikawa et al. 1985).

The magnetic properties of the intermediate phases are mainly due to the arrangement of the $Fe^{(III)}$, $Fe^{(II)}$, and $Ti^{(IV)}$ cations, i.e. to the local concentration of $Fe^{(III)}$, which generally replaces $Ti^{(IV)}$ in the lattice. Complex ordering regimes have been reported for synthetic solid solutions with a mole fraction of ilmenite between 0.79 and 0.9. A solid solution with $y = 0.79$ was depicted as re-entrant spin glass whereas one with $y = 0.9$ was considered to be cluster spin glass (Ishikawa et al. 1985). The occurrence of spin glass-like properties was also described for natural hemo-ilmenite particles with $y = 0.86$ (Gehring et al. 2007). The relationship between magnetic ordering and chemical inhomogeneity in the solid solution has not been resolved in detail.

Natural single crystals of hemo-ilmenite forming a solid solution with $y = 0.83$ were investigated in this study. The single crystals have structure which is homogeneous with respect to resolution of the transmission electron microscope. Inhomogeneity on a molecular level is documented by electron paramagnetic resonance (EPR) spectroscopy, which reveals the occurrence of $Ti^{(IV)}$-enriched $Fe^{(III)}$ clusters isolated in the solid solution at $T < 20$ K. The EPR spectral characteristics suggest that the clusters are paramagnetic, i.e. decoupled from the magnetic bulk material. The zero-field and field-cooled magnetizations as well as ac susceptibility measurements indicate magnetic inhomogeneity of the solid solution at low temperature, as manifested by spin glass-like properties at about 40 K. This behavior occurs 60 K below the first indication of magnetic metastability in the solid solution detected by hysteresis experiments. The above finding provides evidence that chemical heterogeneity on a molecular level enhances the dynamics of the magnetization in natural hemo-ilmenite solid solution.

References

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