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Direct observation of the interaction between elastic and magnetic domain walls below the Verwey transition in magnetite using electron holography

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The Verwey transition has an enormous impact on the magnetic properties of magnetite at low temperatures – the magnetocrystalline anisotropy increases by an order of magnitude and the magnetic easy axis switches from the $<111>$ directions of the cubic phase to the [001] direction of the monoclinic phase. On cooling through the transition, the [001] easy axis of the monoclinic phase may be chosen to lie along any one of three $<100>$ directions of the parent cubic phase, resulting in the development of transformation twinning. Numerous studies have proposed that a strong interaction exists between the ferroelastic twin walls and the ferrimagnetic domain walls in magnetite. Nevertheless, the nature of this interaction remains highly controversial. Key questions include: i) what kinds of magnetic domains are common?, ii) are magnetic domain walls strongly pinned by the twin walls or can they be moved independently? and iii) how does the twin microstructure that develops on cooling through the transition depend on the magnetic microstructure that exists above the transition, and vice versa?

Here, we use off-axis electron holography in the transmission electron microscope to study the magnetic domain structures in synthetic multi-domain magnetite, both below and above the Verway transition. Electron holography allows magnetic structures to be imaged quantitatively at a nanometer scale. Lorentz electron microscopy is employed to make dynamical observations of the nucleation and translation of transformation twins and magnetic domain walls as the sample was repeatedly cycled through the phase transition. All TEM observations were performed at 300 kV using a Philips CM300ST field emission gun TEM.

The distribution of magnetic domains above and below the transition was generally very different. The cubic phase is characterised by a low density of magnetic domain walls, whereas the monoclinic phase contains a higher density of closely-spaced lamellar domains. In contrast to previous studies, magnetic closure domains within the monoclinic phase were also observed to be relatively common.

Regions showing a clear interaction between magnetic domain walls and twin domain walls were observed. A typical feature is the presence of irregular twin boundaries formed by the impingement of monoclinic domains that nucleated in different parts of the crystal. The monoclinic domains on either side of the boundary contain much finer scale needle-like twins, with the orientation of needles switching through 90˚ across the boundary. The magnetisation in each monoclinic domain is perpendicular to the needle twins, and, except in rare cases, does not appear to be greatly affected by the needle twins themselves. In one case, however, a zigzag magnetic domain structure coincident with the needle twinning was observed, suggesting that different classes of needle twins are present: some which leave the easy axis untouched and other that cause the easy axis to change direection. The irregular boundaries between mononcinic domains are shown to be 90˚ magnetic domain walls. Both conventional and ‘divergent’ 90˚ walls are observed using Lorentz microscopy. Micromagnetic models show that a conventional 90˚ wall is converted to a divergent wall by passage of a 180˚ Bloch wall through the adjacent monoclinic domain. The irregular boundaries are imobile, except at temperatures very close to the Verwey transition, where lateral migration of the boundary prior to the transition was observed