Magnetic minerals in rocks record the direction and intensity of the ambient magnetic field during formation, providing, for example, varied information of the geomagnetic field and past tectonic plate motions. In order to reliably interpret paleomagnetic measurements, the mechanism of chemical remanent magnetisation (CRM) which can induce and subsequently alter magnetic remanence must be fully understood. Currently, models of CRM processes only exist for the smallest, uniformly magnetised grains, termed single domain (SD). However, the magnetic signal from rocks is often dominated by slightly larger grains containing non-uniform magnetisation states and these are termed pseudo-SD (PSD) grains.

Magnetite (Fe$_3$O$_4$) is the most magnetic naturally occurring mineral on Earth, carrying the dominant magnetic signature in rocks and providing a critical tool in paleomagnetism. The oxidation of Fe$_3$O$_4$ to other iron oxides, such as maghemite ($\gamma$-Fe$_2$O$_3$) and hematite ($\alpha$-Fe$_2$O$_3$), is of particular interest as it influences the preservation of remanence of the Earth’s magnetic field by Fe$_3$O$_4$. During oxidation, the inverse spinel ferrite Fe$_3$O$_4$ reacts with oxygen to form the Fe$^{2+}$ cation deficient phase $\gamma$-Fe$_2$O$_3$, which can then further oxidise to form hexagonally-close-packed $\alpha$-Fe$_2$O$_3$.

Environmental transmission electron microscopy (ETEM) enables the detailed investigation of localised chemical reactions under gas atmospheres. Off-axis electron holography permits nanometre-scale imaging of magnetic induction within and around materials as a function of applied field and temperature. The complementary use of these advanced TEM techniques can be used to reveal local changes in magnetisation in minerals as they alter during in situ heating in a controlled atmosphere.

In the present study, synthetic Fe$_3$O$_4$ particles in the PSD size range (< 200 nm) were heated in situ in an ETEM to a temperature of 700 °C in 8 mbar of O$_2$. Oxidation of the Fe$_3$O$_4$ particles was investigated using bright/dark-field imaging and electron energy-loss spectroscopy (EELS). Figs. 1a & 2a show native smooth-surfaced Fe$_3$O$_4$ grains and complementary EELS analysis of the Fe 2p $L_3$ edge (Figs. 1c & 2c) is in good agreement with that of pure Fe$_3$O$_4$. Close examination of Fe$_3$O$_4$ particles after in situ heating revealed surface degradation in the form of nanoparticles (Figs. 1d & 2d) and EEL spectra showed pre-peaks close to the Fe 2p $L_3$ edge (Figs. 1f & 2f) that are indicative of oxidation. The associated effect of CRM was investigated using off-axis electron holography, in the form of reconstructed magnetic induction maps, where the oxidised grains exhibited a loss of overall remanence (Figs. 1b & 1e) and degradation of magnetic vortices (Figs. 2b & 2e).

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Fig. 1: (a) Bright-field TEM image of two Fe₃O₄ particles, with SAED pattern (inset); and (b) their magnetic induction map. (c) Fe 2p L₃ edge of the EEL spectrum acquired from the particles in (a). (d) Dark-field image of the Fe₃O₄ particles after in situ heating. (e) Magnetic induction map of the particles in (d); and (f) associated EEL spectrum.

Fig. 2: (a) Bright-field TEM image of attached Fe₃O₄ particles, with SAED pattern (inset); and (b) magnetic induction map. (c) Fe 2p L₃ edge of the EEL spectrum acquired from the particles in (a). (d) Dark-field image of the Fe₃O₄ particles after in situ heating. (e) Magnetic induction map of the particles in (d); and (f) associated EEL spectrum.