Limitations and challenges in off-axis electron holography of electromagnetic fields in nanoscale materials

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In contrast to most conventional transmission electron microscopy (TEM) techniques, which only allow the spatial distribution of image intensity to be recorded, off-axis electron holography allows the phase shift of the electron wave that has passed through an electron-transparent specimen to be measured. The phase shift can, in turn, be used to provide information about local variations in magnetic induction and electrostatic potential within and around the specimen. Recent developments in the technique include the reconstruction of electrostatic potentials and magnetic fields in three dimensions, the use of advanced specimen holders with multiple electrical contacts to study nanoscale working devices, improvements in the stability of transmission electron microscopes to optimise phase sensitivity and the development of new approaches for improving temporal resolution using both direct electron detectors and double exposure electron holography. We are currently using the technique to characterize electrostatic potentials and magnetic fields in a wide variety of nanoparticles, nanostructures and thin films that are subjected to electrical biases and externally applied magnetic fields, as well as to elevated and reduced temperatures. Figure 1 shows representative results obtained from a study of the thermomagnetic behaviour of nanoscale grains of magnetite during heating *in situ* in the TEM. The magnetic induction maps show first a horseshoe-like magnetic state and then magnetic phase contours that flow from the bottom to top of the grain at higher temperature.

An important limitation of backprojection-based algorithms for reconstructing magnetic fields in three dimensions is the presence of artefacts resulting from incomplete tilt series of phase images and the inability to include additional constraints and known physical laws. Accordingly, one of our aims is the development of a robust model-based approach that can be used to reconstruct the three-dimensional magnetization distribution in a specimen from phase images recorded as a function of specimen tilt angle using off-axis electron holography. In order to perform each reconstruction, we generate simulated magnetic induction maps by projecting best guesses for the three-dimensional magnetization distribution in the specimen onto two-dimensional Cartesian grids. Our simulations make use of known analytical solutions for the phase shifts of simple geometrical objects, with numerical discretization performed in real space to avoid artefacts generated by discretization in Fourier space, without a significant increase in computation time (Figs 2 and 3). Our forward simulation approach is used within an iterative model-based algorithm to solve the inverse problem of reconstructing the three-dimensional magnetization distribution in the specimen from tilt series of two-dimensional phase images recorded about two independent tilt axes. Results will be presented from studies of magnetite nanocrystals, lithographically patterned magnetic elements and magnetic skyrmions examined as a function of temperature and applied magnetic field. At the same time, we are developing a similar algorithm for the reconstruction of three-dimensional charge density distributions in materials. Preliminary results will be presented from studies of charge distributions in electrically biased needle-shaped specimens, which require the analysis of differences between phase images recorded using two applied voltages, in order to subtract the mean inner potential to the phase shift.

The above studies are part of a wider program of research aimed at recording off-axis electron holograms of nanoscale materials and devices in the presence of multiple external stimuli. Further examples will be presented from studies of electrically biased resistive switching devices and two-dimensional flakes of transition metal dichalcogenides, whose electrical properties can be influenced strongly by the presence of contamination and defects, as well as by their interfaces to metal contacts. We are grateful to J. Ungermann, M. Riese, G. Pozzi, W. Williams, A. R. Muxworthy, M. Farle, M. Beleggia, T. F. Kelly and N. Kiselev for valuable contributions to this work and to the European Research Council for an Advanced Grant.

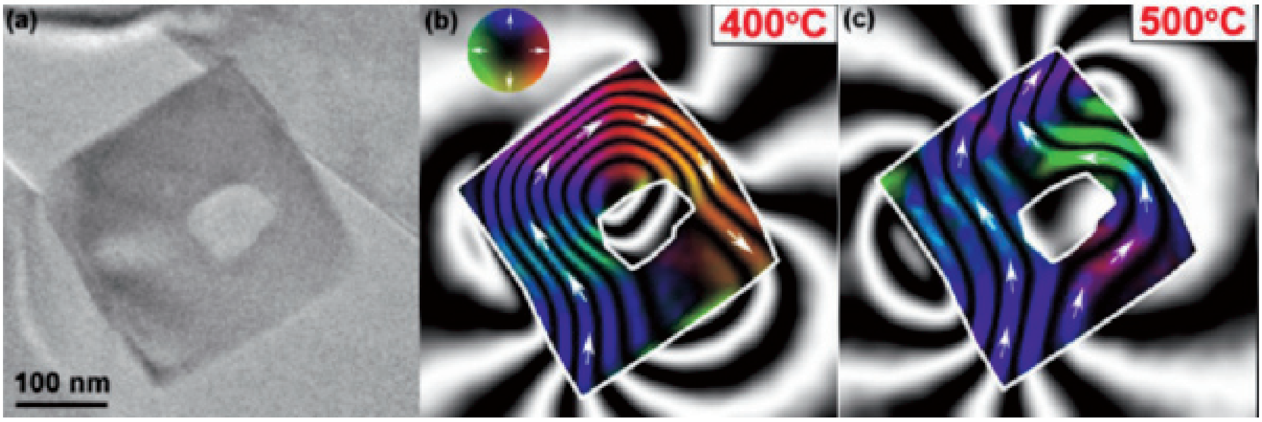

Figure 1. (a) Bright-field TEM image of a 300 nm toroidal magnetite grain. (b, c) Magnetic induction maps recorded using off-axis electron holography during in situ heating to (b) 400 and (c) 500 ˚C. The outline of the grain is marked solid white lines. The contour spacing is 0.39 radians.

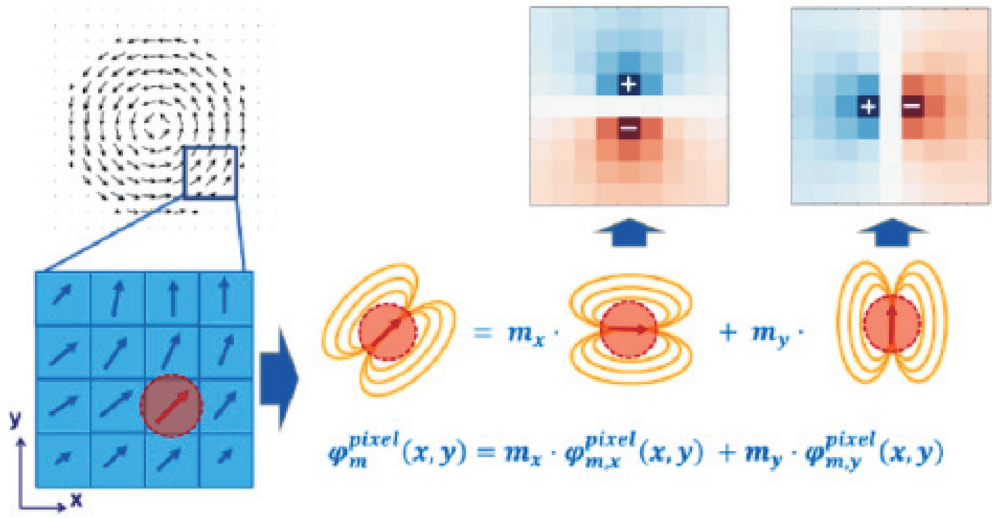

Figure 2. Illustration of the simulation process in our model-based reconstruction algorithm. Each projected two-dimensional magnetization distribution is sub-divided into pixels that are represented by simple geometries (e.g., a disc). The contribution to the phase shift from every pixel is calculated as pre-computed components.

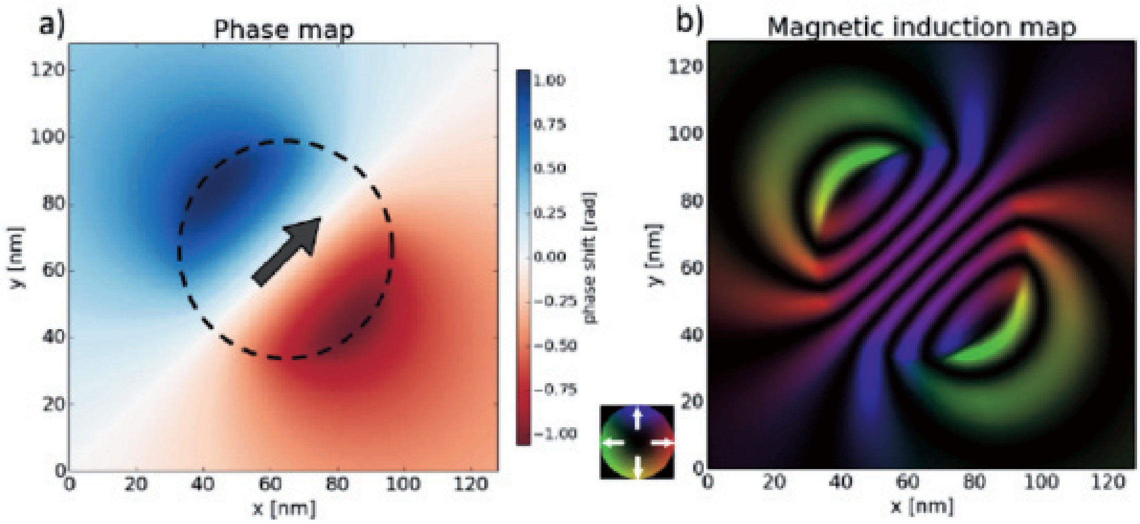

Figure 3. a) Simulated magnetic phase shift of a uniformly magnetized sphere with a radius of 64 nm. The magnetization direction is indicated by the arrow. b) Corresponding magnetic induction map (20 times phase amplified).