Off-axis electron holography of self-assembled Co nanoparticle rings

Takeshi Kasama[1], Rafal E. Dunin-Borkowski[2], Michael R. Scheinfein[3], Steven L. Tripp[4], Jie Liu[4] and Alexander Wei[4]

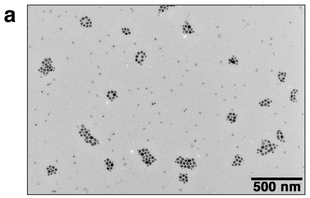
[1] Department of Materials Science and Metallurgy, University of Cambridge, Cambridge, UK, [2] Center for Electron Nanoscopy, Technical University of Denmark, Kongens Lyngby, Denmark,
[3] Department of Physics, Simon Fraser University, Burnaby, Canada, [4] Department of Chemistry, Purdue University, West Lafayette, USA.

Abstract

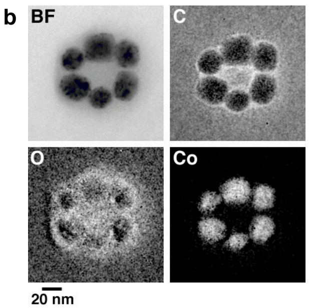
A full understanding of the magnetic properties of closely-spaced ferromagnetic nanocrystals is of great importance for the development of new materials for high-density magnetic information storage and magnetic random access memory applications. Nanoscale ring-shaped elements are of particular interest because they can form magnetic states that exhibit flux closure (FC) and cannot be supported in disk-shaped elements of similar size [Zhu et al. (2000) *J. Appl. Phys.* 87, 6668].

Here, we use off-axis electron holography in the transmission electron microscope (TEM) and micromagnetic simulations to study FC states in self-assembled Co nanoparticle rings that contain between five and eleven 25-nm-diameter crystals. Holograms were obtained at room temperature in zero-field conditions after applying chosen magnetic fields to the samples *in situ* in the TEM by partially exciting the conventional microscope objective lens. Mean inner potential (MIP) contributions to the phase shift were determined by turning the samples over, and subsequently subtracted from each recorded phase image to obtain magnetic induction maps. Our results show that most nanoparticle rings form FC remanent magnetic states, and occasionally onion-like states. Although the chiralities (the directions of magnetization) of the FC states are determined by the shapes, sizes and positions of the constituent nanoparticles, reproducible magnetization reversal of each ring can be achieved by using an out-of-plane (OOP) magnetic field of between 1600 and 2500 Oe.

Sample details and conventional TEM characterization

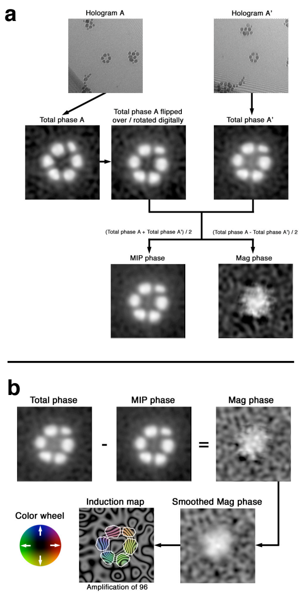


Polycrystalline Co nanoparticles (25-30 nm in diameter) were dispersed in a toluene solution containing C11 resorcinarene and deposited directly onto holey carbon Cu TEM grids (a). A significant fraction of the nanoparticles self-assembled into rings of 5-10 particles. The samples were known from bulk magnetometry techniques to be weakly ferromagnetic at room temperature, but neither the detailed magnetic states of the self-assembled Co particles nor their reversal mechanisms were understood.



A magnified bright-field (BF) image of a six-membered nanoparticle ring and corresponding three-window background-subtracted elemental maps for C, O and Co are shown in (b). There is a thin C-rich shell around each particle. The thicker and less distinct O-rich shell is associated in part with the residue of the surfactant and in part with the presence of ~3 nm of CoO surrounding each crystal. The adjacent Co particles are therefore not in direct contact but are separated from each other by several nanometers.

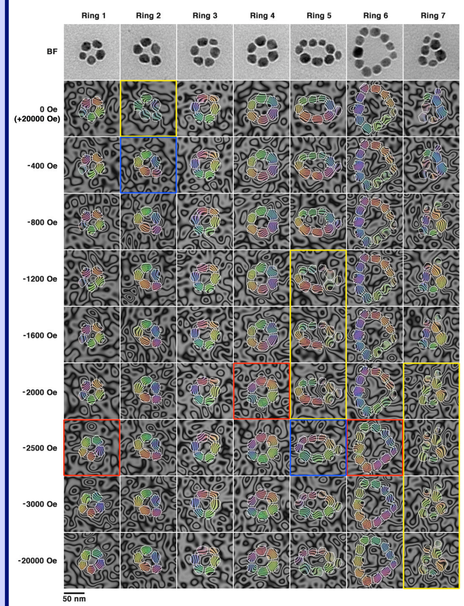
Experimental procedure used to obtain induction maps



In order to investigate the magnetization reversal behavior of the rings, OOP fields were applied *in situ* in the TEM by partially exciting the conventional microscope objective lens. First, an OOP field of +20,000 Oe was applied perpendicular to the plane of the specimen and reduced to zero. Second, the sample was taken out of the TEM, turned over and put back into the microscope. Third, a sequence of chosen OOP fields of up to -20,000 Oe was applied to the specimen in the TEM. The applied field was always reduced to zero before acquiring holograms.

The dominant MIP contribution to the phase shift was calculated from phase images that had been acquired before and after turning the specimen over (a). Their sum and difference were used to determine twice the MIP and twice the magnetic contribution to the phase shift, respectively. Once the MIP contribution had been calculated, it could be subtracted from all subsequent phase images (b). Phase contours and colors were used to form the final induction maps. Smoothing of the phase images was used to remove statistical noise and occasional artifacts resulting from misalignment of the pairs of phase images.

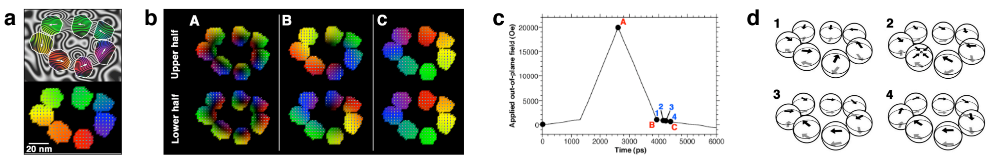
Magnetic induction maps measured using electron holography



Magnetic induction maps were acquired from seven Co nanoparticle rings following the application of OOP fields of different magnitude. The resulting induction maps are shown on the left, with a phase amplification of 96. The red boxes show the lowest applied fields at which the rings were seen to have reversed. The blue boxes show FC states that transformed from onion-like states (marked in yellow).

Although many of the rings exhibit FC states, some of the rings (e.g., 2, 5 and 7) show onion-like states, which were previously thought to be less favored in nanoscale ring geometries. In ring 2, an onion-like state changes to a FC state at a low value of the applied field. Onion-like states are also observed close to the coercivity of remenence (see, e.g., ring 5). Unusually, ring 3 does not reverse even following the application of a -20,000 Oe field. Additional particles close to the rings may stabilize onion-like or non-FC states (see, e.g., ring 7). From the experimental induction maps, the coercivity of remanence is measured to be between 1600 and 2500 Oe.

Micromagnetic simulations of a seven-membered nanoparticle ring



Micromagnetic simulations were used to understand the reversal mechanisms of the Co rings that had been observed using electron holography. The figures show an experimental induction map (upper image in (a)) and simulations (lower image in (a) and the six frames in (b)) of the magnetic states of a seven-membered nanoparticle ring. In the simulation, a 20,000 Oe OOP field was first applied to the ring and reduced to <1000 Oe (c). The letters in (b) correspond to the red capital letters in (c). The simulations show that the reversal mechanism of the rings is highly complicated. During magnetization reversal, the individual Co particles each form magnetic vortices (d-1). On reducing the OOP field, one of the particles switches to a single-domain (SD) state before the others (d-2) and ultimately determines the directions of the moments in the ring (d-3) when it forms a FC state (d-4). The evolution and final direction of the moments in the particles is determined by subtle differences in energy between alternate magnetic states, which are in turn affected by the details of the particle morphologies, orientations and positions.