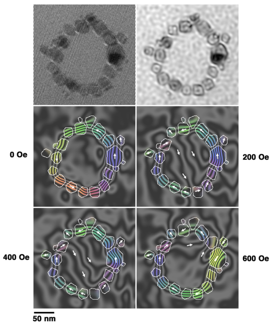
Measuring the magnetic moments of Co nanoparticle rings from their Aharonov-Bohm phase shift

M Beleggia[1], T Kasama[1], R E Dunin-Borkowski[1], J Liu[2] and A Wei[2]

[1]Center for Electron Nanoscopy, Technical University of Denmark, DK-2800 Kgs. Lyngby, Denmark
[2]Department of Chemistry, Purdue University, West Lafayette, IN 47907-2084, USA

1. Introduction

The Aharonov-Bohm effect [1] reveals itself in transmission electron microscopy (TEM) as a phase modulation of the high-energy electron wave induced by magnetic specimens. Here, we use off-axis electron holography [2,3] in the TEM to record electron-optical phase images from rings of closely-spaced 20-nm-diameter single-magnetic-domain cobalt particles, which exhibit a variety of magnetic states at remanence [4,5], including vortex (closure domain) and onion (in-plane-magnetized) states. By means of a newly developed algorithm [6] we measure the magnetic moments of individual rings at room temperature. We show that the number of flux quanta in each ring is not necessarily an integer and may be smaller than unity. We also investigate the hysteretic response of a Co ring by measuring its moment as a function of the out-of-plane magnetic field applied to the sample before hologram acquisition. All measurements are carried out at remanence, producing quantitative "remanent hysteresis curves".



Top-left, an off-axis electron hologram of a ring made of about 20 Co nanoparticles; top right, the reconstructed electrostatic contribution to the phase shift, providing thickness information; bottom, magnetic contribution to the phase shift at various applied reversal fields (holograms are always acquired at remanence) revealing the complex projected magnetic field topography associated to this particular ring. Note in particular the presence of a vortex (0 Oe) and an onion (400 Oe) state at remanence.

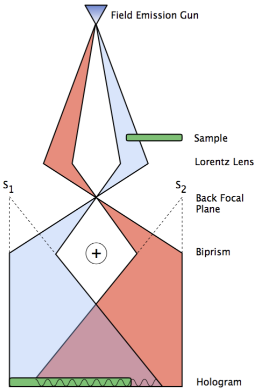
2. Electron holography

Off-axis electron holography relies on the use of an electron biprism to overlap a high energy electron wave that passed through a TEM specimen with another part of the same electron wave that has passed only through vacuum.

The resulting interference fringe pattern can be used to retrieve the phase modulation of the electron wave, described by the "Aharonov-Bohm" expression

$$\varphi(x,y)=C_E\int V(x,y,z)dz-\frac{\pi}{\phi_0}\int A_z(x,y,z)dz$$

which contains information about the electrostatic potential V and the component *A_z* of the magnetic vector potential in the electron beam direction *z* generated by and surrounding the sample. Extraction of quantitative information about the magnetic properties of the sample requires processing of the phase image obtained by reconstructing the hologram.



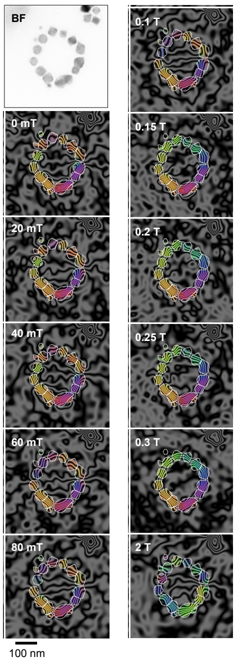
3. Experimental magnetic induction maps

Ferromagnetic Co nanoparticles were deposited at room temperature onto a Cu grid coated with a holey carbon film. A significant fraction of the particles formed ring structures.

Electron holography was used to record phase shifts associated with the rings. The structures were subject to out-of-plane field pulses of varying strengths. All holograms were acquired *after* the pulse, in field-free conditions. The duration of the magnetic pulse was of the order of seconds, produced by manually changing the current in the TEM objective lens.

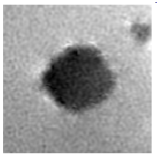
For the ring shown on the right, different values of the applied field result in two transitions in the observed remanent state: at around 0.1 T the number of fringes visible in the inner part of the onion-state ring decreases, signaling the formation of a vortex state; at larger fields, the onion state reappears, with opposite orientation of the moment compared to the low-field state.

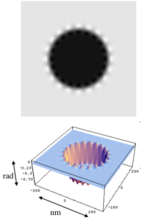
We apply our algorithm to this ring, and measure the variation of the moment, in magnitude and orientation, with applied field.



4. Vortex state and Aharonov-Bohm shift

When the ring is found in a vortex state, the phase shift features a constant plateau in the inner region, surrounded by a constant background: this is a direct visualization of the Aharonov-Bohm phase shift. Here, the phase jump is about 1 rad, corresponding to ~1/3 of a flux quantum.





Comparison between experimental and simulated phase shifts of a Co nanoparticle ring in a vortex state.

5. Algorithm for extraction of magnetic moment

The magnetic moment of a nanoparticle or set of nanostructures

$$\mathbf{m}=\iiint\mathbf{M}(\mathbf{r})d^3\mathbf{r}$$

can be measured from the retrieved phase shift. The phase gradient is proportional to the magnetic induction projected along the electron trajectory

$$\frac{\phi_0}{\pi}\left[\hat{\mathbf{z}}\times\nabla\varphi(\mathbf{r}_\perp)\right]=\int_{-\infty}^{+\infty}\mathbf{B}(\mathbf{r})dz$$

so that, if the phase gradient is integrated over some portion of the field of view

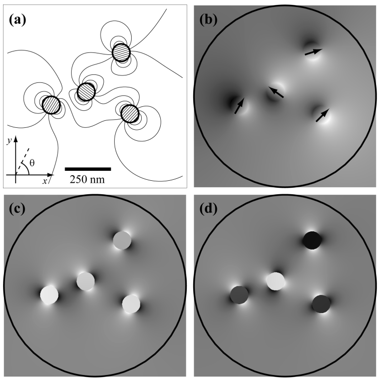
$$\frac{\phi_0}{\pi}\left[\hat{\mathbf{z}}\times\iint\nabla\varphi(\mathbf{r}_\perp)d^2\mathbf{r}_\perp\right]=\iiint\mathbf{B}(\mathbf{r})d^3\mathbf{r}$$

a proportionality can be established between the integrated phase gradient and the volume integral of the induction, a quantity that we refer to as "inductive moment" m_B

$$\mathbf{m_B}=\frac{1}{\mu_0}\iiint\mathbf{B}(\mathbf{r})d^3\mathbf{r}$$

Then, fundamental properties of magnetostatic fields result in a further relationship between the magnetic moment m and the inductive moment m_B. If the region of integration is a circle encompassing all magnetized matter, the relationship between m and m_B is particularly simple (especially for planar moments): $m_B=1/2$ m.

To improve statistics and to reduce the effects of artifacts, the integration is carried out over circles of decreasing radius (until the integration loop touches the particle boundary), and the obtained curve is extrapolated to zero radius with a parabola.

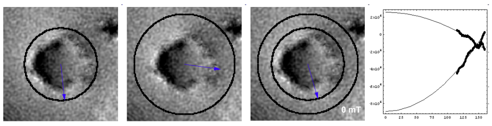


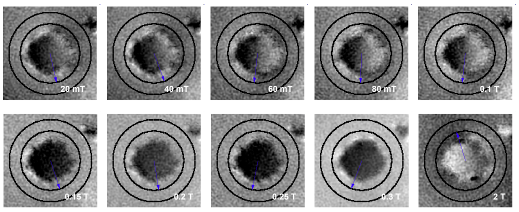
Illustrative example showing simulations of four 50-nm radius disks, each carrying 2☒10^7 Bohr magnetons, magnetized in-plane at angles of 18°, 45°, 60° and 144° measured anti-clockwise from the horizontal axis: (a) magnetic induction map; (b) magnetic phase image, with loop-integral contour and arrows pointing in the direction of each disk's magnetization; (c) and (d) phase gradient components, with the boundary of the circular integration region superimposed The ensemble of disks is representative of a generic magnetic structure with a non-uniform magnetization and a non-trivial shape. Upon application of the algorithm, the vectorial sum of the four moments, i.e. the total moment, is obtained exactly [6].

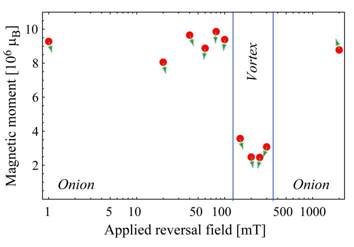
6. Measurements of magnetic moment

For the chosen ring, the acquired phase images allow us to draw circles between 115 nm and 160 nm in radius without overlapping with nearby structures. Integrating over circles of increasing radius (with a 0.5 nm step, i.e. a total of 91 integrations), we obtain a data set for each component of the magnetic moment. We then fit the two data sets with parabolic functions, and take their apexes as values of the moment components. Uncertainties on the measurements are between 3% and 12% for the magnitude and around 2-5° for the orientation of the magnetic moment.

The transition between vortex and onion states, occurring when the reversal field exceeds 100 mT, is clearly visible in the hysteresis loop as a sudden decrease of the total magnetic moment. The onion state reappears, *with reversed polarity*, when the applied field is very large.







Top: Circle of minimal radius (115 nm), circle of maximal radius (160 nm), all circles in between the two, and the fit of the loop-integration results with a parabolic function.

Middle: Data series as a function of applied field.

Bottom: Resulting remanent hysteresis curve, with the moment measured in magnitude and orientation (green arrows).

7. References

[1] A. Aharonov and D. Bohm, Physical Review 115, 485 (1959).
[2] A. Tonomura, *Electron holography*, Springer, Berlin, 1993.
[3] E. Volkl, L. Allard, D.C. Joy (editors), *Introduction to Electron Holography*, Kluwer Academic, 1999.
[4] S. L. Tripp, R. E. Dunin-Borkowski and A. Wei, Angew. Chemie 42, 5591 (2003).
[5] T. Kasama, R. E. Dunin-Borkowski, M. R. Scheinfein, S. L. Tripp, J. Liu and A. Wei, Adv. Mat. 20, 4248 (2008).
[6] M. Beleggia, T. Kasama and R.E. Dunin-Borkowski, Ultramicroscopy (2010) in press.