Magnetization Reversal in Self-Assembled Cobalt Nanoparticle Rings

PURDUE UNIVERSITY

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

UNIVERSITY OF CAMBRIDGE

[1] Department of Materials Science and Metallurgy, University of Cambridge, CB2 3QZ, UK; [2] Center for Electron Nanoscopy, Technical University of Denmark, D-2800 Kongens Lyngby, Denmark; [3] Department of Physics, Simon Fraser University, Burnaby V5A 1S6, British Columbia, Canada; [4] Department of Chemistry, Purdue University, West Lafayette, IN, 47907-2084 USA

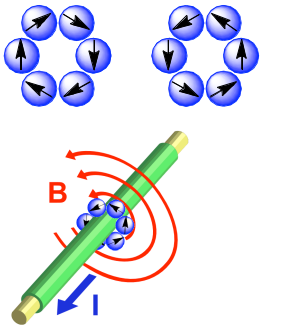
Abstract: Ferromagnetic cobalt nanoparticles (dispersed in toluene by C11 resorcinarene) can spontaneously assemble into nanorings of discrete particle count. The Co nanoparticle rings support bistable flux closure (FC) states at room temperature, as characterized by off-axis electron holography. In many cases, the FC polarization can be reversibly switched by exposure to a coaxial magnetic field. Magnetodynamic simulations provide some insights into the complex mechanism of field-induced FC reversal.

Introduction. In-plane flux closure (FC) is a bistable, low-energy magnetic state commonly observed in thin plates and discs of soft magnetic materials. FC states in such microstructures are often destabilized by a central singularity, referred to as a magnetic vortex. This destabilization is not present in ring-like structures, making them the preferred geometries for supporting binary FC states.

Magnetic rings have been suggested as nonvolatile elements in novel device architectures for magnetoresistive random-access memory (MRAM).[1] Furthermore, FC states can be polarized (switched) by transaxial currents in accordance with Ampère's law, with a fundamental limit for FC switching in the low picosecond range.[2] These provide a motivation to produce magnetic rings on the smallest possible scale.

Limits for magnetic nanorings? Most methods for fabricating magnetic nanorings involve top-down lithography, and are so far limited to dimensions on the order of 100 nm. However, the minimum size for stable FC states depends on the remanence of the supporting magnetic material, and can be expected to be well below 100 nm.

Here we show that Co nanoparticle rings can be produced below 100 nm by dipole-directed self-assembly, and support stable FC states at 298 K. We also observe a remarkable reversal in FC polarization by applying an out-of-plane magnetic pulse, suggesting a novel magnetodynamic mechanism for FC switching. To our knowledge, there is no macroscopic analogy to the field-induced FC switching of magnetic nanoparticle rings.

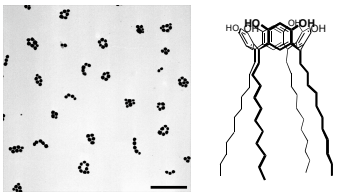


FC states in magnetic nanoparticle rings (CW and CCW)

FC polarization induced by current

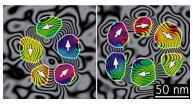
FC switching by exposure to coaxial magnetic field

Self-assembly. Co nanoparticles (d_{av}=27 nm) with a thin oxide shell were dispersed in toluene using C11 resorcinarene, a macrocyclic surfactant.[3] These particles exhibited dipolar character and were weakly ferromagnetic at 298 K (M_R/M_S=0.13; H_C=175 Oe). The Co nanoparticles were deposited onto holey carbon TEM grids, a significant fraction of which assembled into rings comprised of 5–10 particles. Nanoparticle ring formation could be optimized as a function of particle and resorcinarene concentration (ca. 10^{12} particles/mL and 1–10 _M, respectively).[4]



TEM image of Co nanoparticle assemblies deposited from toluene solutions containing C11 resorcinarene (right). Bar = 250 nm.

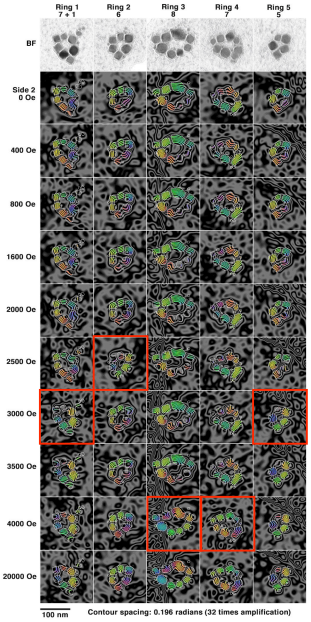
Electron holography (EH). This TEM technique enables in-plane magnetic flux to be visualized with nanometer spatial resolution, and is a powerful method for studying magnetism in isolated nanostructures.[5] FC states in Co nanoparticle rings were recorded by EH at 298 K under zero-field conditions, with continuous magnetic flux observed within each ring ensemble despite irregularities in ring symmetry or particle shape.[4] These observations are consistent with a magnetostatic description of FC states in cyclic ensembles of single-domain particles.



EH images of Co nanoparticle rings taken at 298 K using a Philips CM-300 equipped with a Lorentz lens (300 keV). The polarization of the FC states are depicted using arrows and colors (red = right, yellow = down, green = left, blue = up). The magnetic flux enclosed between adjacent contours is $h/128e$, and correlates inversely with the contour linewidths.

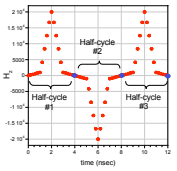
EH also records phase information related to FC polarization. A statistical sampling of Co nanoparticle rings revealed a 50:50 (racemic) mixture of CW and CCW states, indicating the absence of polarization bias despite prior exposure to a magnetic field (H_z) during electron beam focusing. The FC states were stable at 298 K, and remained constant upon additional exposure to H_z in the same direction, up to +2 T.

Field-induced reversals. FC polarizations could be switched by exposing the Co nanoparticle rings to H_z in the opposite sense. Threshold $(-)H_z$ values for FC reversal range from 2500–4000 Oe for rings comprised of single-crystal Co particles (at right), and 1500–2500 Oe if the particles contained twinning defects (not shown). Reliable FC reversal was observed in *ca*. 80% of the rings examined, regardless of their initial polarization. In most cases, subsequent exposure to higher H_z did not result in FC scrambling, clearly indicating a memory effect.



EH images of Co nanoparticle rings after exposure to H_z of varying strength. All images were acquired under zero-field conditions (H_z=0 Oe). Images with red borders indicate threshold of FC reversal.

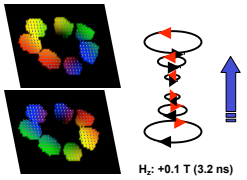
Magnetodynamic simulations. The H_z-induced FC reversals could be reproduced using a 3D simulation tool (LLG Micromagnetics). Constructs based on the Co nanoparticle rings were used to model changes in magnetic induction as a function of H_z, administered as a series of half-cycles (see below). Initial exposure during electron beam focusing was simulated by using a H_z pulse to +2 T in the first half-cycle, to yield a CW-polarized FC state. Pulsing H_z to –2 T in the second half-cycle induced a FC reversal to CCW; a third H_z pulse to +2 T switched the FC state back to CW. The simulations are matched by experiments involving multiple H_z-induced FC reversals in Co nanoparticle rings.



after _-cycle #1 (t=4 ns): CW | after _-cycle #2 (t=8 ns): CCW | after _-cycle #3 (t=12 ns): CW

Simulation of FC switching in a Co nanoparticle ring. Magnetization dynamics were calculated with a time step of 0.2 ps, using a gyromagnetic frequency of 17.6 MHz/Oe and a damping constant of 1.0. Local magnetic moments were calculated as discretized cubes approximately 3.5 nm per side.

The magnetodynamic simulations also reveal intermediate states, prior to FC reformation at zero field (H_z=0). A reduction in H_z near the end of a half-cycle produces a double vortex with a center of inversion, with the in-plane magnetization at the top and bottom of the nanoparticle ring polarized in opposite directions (see below). Further reduction in H_z ($\leq$ 0.08 T) results in the annihilation of the double vortex and a restoration of the FC state.



Magnetodynamic simulation of an intermediate double-vortex state, obtained by reducing H_z from +2 T (*t*=2 ns) to +0.1 T (*t*=3.2 ns). *Left*, upper and lower images represent top and bottom slices of a Co nanoparticle ring, viewed from the top. Right, schematic of double vortex (stacked FC states) with polarizations relative to M_z (blue arrow).

The relation between H_z and FC polarization is not yet fully resolved. However, we note the presence of a residual out-of-plane magnetization (M_z) at zero field, whose direction is defined by prior exposure to H_z and may play a role in FC switching. A magnetodynamic correlation between remanent M_z and the in-plane FC states would be distinct from the recent description of vortex core reversals by AC magnetic pulses, which occur independently of FC polarization.[6]

Acknowledgments

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