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Galaxy-like magnetic domain structures in ferromagnetic nanoparticle arrays observed by electron holography

Kazuo Yamamoto[1*]

[1] Japan Fine Ceramics Center

When ferromagnetic nanoparticles are packed as a face-centered-cubic (fcc) lattice, dipolar ferromagnetic state appears in the lattice film. Magnetic dipoles in each nanoparticle are aligned in one direction with dipole-dipole interaction. This ground state was theoretically predicted by Luttinger and Tisza in 1946 [1]. Magnetic force microscopy (MFM) detected collectively magnetized regions in 12 nm Co nanoparticle assemblies [2]. However, the magnetic domain structures have been obscure because MFM is not sensitive enough to image the in-plane component in the arrays. Electron holography, one of the techniques using a transmission electron microscope (TEM), can detect and image the in-plane component of magnetic flux at higher spatial resolution. Here, we report our use of electron holography to observe the existence of dipolar ferromagnetic domain structures and their behavior.

We prepared 8 nm epsilon-Co nanoparticles by thermally decomposing cobalt carbonyl precursor in high boiling-point solvent. Each nanoparticle was coated with organic surfactants. We dispersed the particles on immiscible liquid and compressed them to obtain close-packed monolayers. We then transferred them to carbon-coated TEM grids [3]. Before loading the sample into the TEM, the film was scratched with a sharp tungsten needle to obtain a vacuum area for electron holography.

Figure 1 shows the TEM image of the particle arrays with partial disordering. The average edge-to-edge separation between particles is 4.2 nm, so exchange effects are minimal. We used Philips CM200 field emission TEM under Lorentz microscopy mode; the standard objective lens was turned off and a Lorentz mini-lens beneath the objective lens was used to focus the sample. To observe virgin domain structures of dipolar ferromagnetism, we cooled the sample to 108 K with a liquid nitrogen cold stage in a field free environment (Zero Field Cooling).

Figure 2(a) shows a 7.5 micron wide region of the array. We took a series of holograms and reference holograms at the same positions, and obtained the magnetic flux distribution shown in Figure 2(b). We observed partially galaxy-like structures in micron scale that were separated by transverse domain walls. A previous simulation predicted vortex structures due to magnetostatic interactions [4]. Our result was similar to the simulation pattern at the sample edges.

Next, we applied a magnetic field to the sample in the TEM. The sample was tilted and the objective lens was turned on. After turning off the lens, the sample was tilted back and imaged in remanence. The center of the galaxy-like structure was shifted to the left, as shown in Figure 2(c). Shifting of a transverse wall by a magnetic field has also been observed in ordinary ferromagnetic permalloy rings [5]. A similar response occurred in our sample without exchange coupling.

We demonstrated the existence of dipolar ferromagnetic domain structures formed in the magnetic nanoparticle arrays, and observed the patterns of the domain structures and their response to the applied field. This observation should clarify the characterization of dipolar ferromagnetism in nanoparticle arrays.

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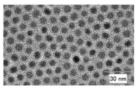

Fig. 1. TEM image of 8 nm epsilon-Co nanoparticle array.

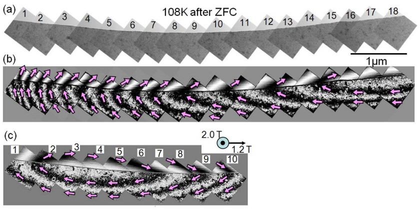

Fig. 2. Dipolar ferromagnetic domain structure after zero field cooling. (a) A series of 18 TEM images taken along the edge of the array film. (b) Magnetic flux distribution around the edge. (c) Magnetic flux distribution after applying magnetic field in the TEM.

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