Quantitative Magnetization States of Colloidal Systems via Simplified-LLG and Model-based Fitting of Phase Images

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Off-axis electron holography can be used to record electron-optical phase images of colloidal magnetic systems such as self-assembled nanoparticles deposited onto an electron-transparent substrate (chains, rings or more complicated arrangements). The recorded phase contour maps can be used to provide information about the orientation of the macro-spin (the semi-classical magnetic moment) of each nanoparticle in the assembly. Figure 1(a) shows a bright-field image of Co nanoparticles on a holey carbon film. Self-assembly has resulted in a close-packed arrangement of the particles into a chain that has a width of 3-4 particles. The mean diameter of the particles is ~13 nm. Figure 1(b) shows the magnetic contribution to the reconstructed electron holographic phase image of the same distribution of particles, with phase contours superimposed. A deeper understanding of the effect of magnetic interactions on the self-assembly process requires quantitative information about the magnitude and orientation of the macro-spin in each of the particles in the assembly.

We are therefore developing a Monte Carlo based approach for the quantitative interpretation of such phase images. The approach involves varying the macro-spins in a simulated array of particles that has the geometry of the real sample until convergence with the experimental phase image is achieved. The resulting distribution of moments in the simulation is then used to infer the true magnetization state of the sample.

As for any Monte Carlo based procedure, our approach is facilitated by a good initial guess, which, in general, decreases the chance that the simulation will terminate in a local minimum of configurational space. A suitable initial guess may be obtained by applying a simplified version of the Landau-Lifshitz-Ginzberg (LLG) equation that is suitable for colloidal systems (i.e., without exchange and with high damping). By starting from an initial condition, in which the moments of the particles are either randomly oriented or aligned along a specified direction, the local field acting on each particle can be calculated as a superposition of the dipole fields generated by each of the particles in the assembly. Any local misalignment between the field and the moment results in a torque and in a rotation of the moment towards the direction of the field. A suitable rotation matrix is then applied to each moment to obtain a new distribution of moments. After a few hundred iterations, relaxation is achieved and the system attains equilibrium.

The LLG-relaxed state is initially used to generate a phase image that can be compared with the experimental magnetic induction map, in order to judge the match to the simulated magnetic state. An example of such a procedure is shown in Fig. 2. For this example, the co-ordinates and radii of the nanoparticles in the array were measured from the bright-field image shown in Fig. 1(a). These positions and dimensions were used to generate a moment distribution, which was initiated in the vertical direction and then relaxed according to the simplified LLG dynamics discussed above, to yield the configuration of moments shown in Fig. 2(a). A simulated phase image, which is shown in Fig. 2(b), was then calculated by superimposing the phase shifts of the entire assembly of moments, on the assumption that they are all uniformly magnetized spheres and that the moments all lie in a single plane. The comparison between Fig. 1(b) and Fig. 2(b) is at first sight satisfactory. However, it fails to reproduce a flux-closure structure (a dipolar vortex) near the top of the array. Therefore, a further step is required, in the

form of a Monte Carlo based refinement of the magnetic moment distribution, until a better match to the experimental phase image is obtained. The computational step required to obtain the simulated phase image from the distribution of moments is the "forward projection" needed in the Monte Carlo algorithm to assess whether to accept or reject the outcome of each iteration, by comparison with the experimental image.

Work is in progress to optimize the algorithm and to extract refined distributions of moments from experimental images. Once fully established and tested, similar model-based fitting of phase images may represent a true breakthrough in our ability to quantify physical parameters using electron microscopy.

References

[1] M. Varon, V. Puntes and C. Frandsen are thanked for the sample and for discussions.

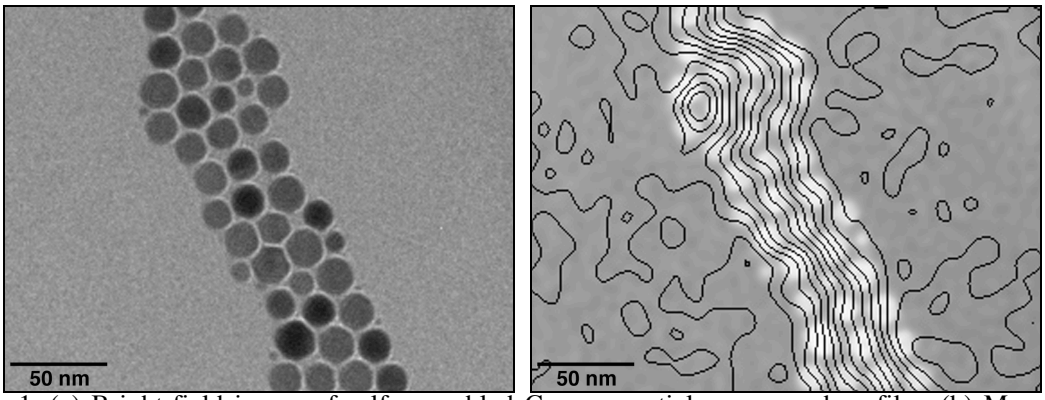

FIG. 1. (a) Bright-field image of self-assembled Co nanoparticles on a carbon film. (b) Magnetic contribution to electron holographic phase shift, with 0.083 radian phase contours superimposed.

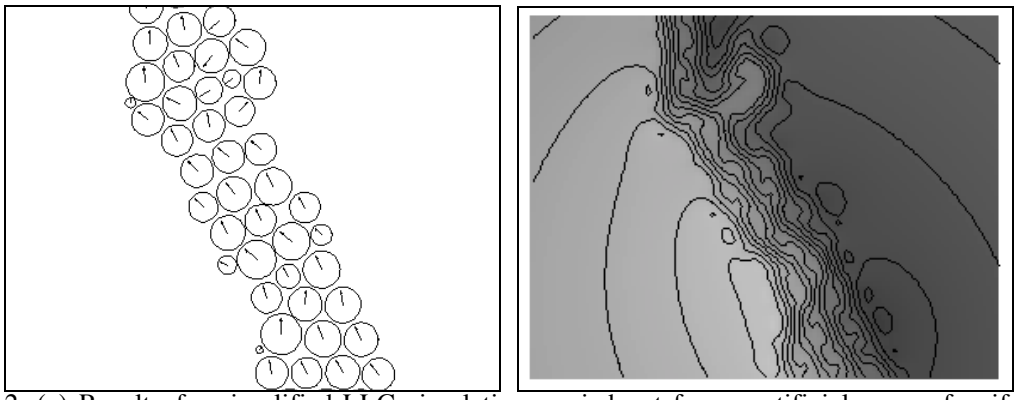

FIG. 2. (a) Result of a simplified-LLG simulation carried out for an artificial array of uniformly magnetized spheres located at the same coordinates as in the real sample, with the sphere diameters measured from the image shown in Fig. 1(a). (b) Magnetic induction map corresponding to Fig. 2(a).