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Direct Observation of Field-Driven Magnetic Reconfiguration in Self-Assembled Nanoparticle Chains

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Magnetic fluids exhibit self-assembly behaviors that alter the macroscopic magnetic properties of concentrated samples. However, direct experimental insight into these nanoscale processes in liquid phases is limited, primarily due to the difficulty of probing structure and magnetic interactions simultaneously at the nanoscale. In this study, we use transmission electron microscopy (TEM) to examine the connection between self-assembly and magnetic properties in magnetic fluids of iron oxide nanoparticles.

Our approach combines complementary TEM techniques to probe this relationship across scales and environments. Off-axis electron holography1 images magnetic interactions within assemblies, while precession-assisted 4D-STEM maps the crystallographic orientation of individual particles. Liquid-cell TEM[2] enables real-time observation of self-assembly dynamics in liquid. In situ magnetization curves measured on individual and interacting nanoparticles reveal the impact of dipolar coupling on macroscopic properties such as coercivity and remanence. Together, these methods link nanoscale organization, magnetic configuration, and collective behavior.

Our results demonstrate the successful characterization of self-assembly processes using cryo-TEM[3], providing a structural basis for magnetic behavior. Building on this, we investigated the magnetic configuration of chains of magnetite nanocubes using electron holography under external magnetic fields. By varying the field, we examine the interplay of dipolar interactions, magnetic anisotropy, and Zeeman energy. Below 100 mT, the magnetization remains aligned along the chain axis, reflecting dominant dipolar interactions. Above this threshold, reorientation occurs, with the magnetization of individual particles aligning with the field and exhibiting antiferromagnetic coupling between neighboring particles. Additionally, applying fields along different crystallographic directions ⟨100⟩ and ⟨110⟩ yields orientation-dependent magnetization curves reflecting cubic anisotropy of magnetite at the single-particle level. Ongoing measurements aim to quantify this anisotropy and extend these observations to dynamically assembling systems in liquid environments.

Overall, these results provide experimental insight into how nanoscale structural organization governs magnetic interactions in magnetic fluids.

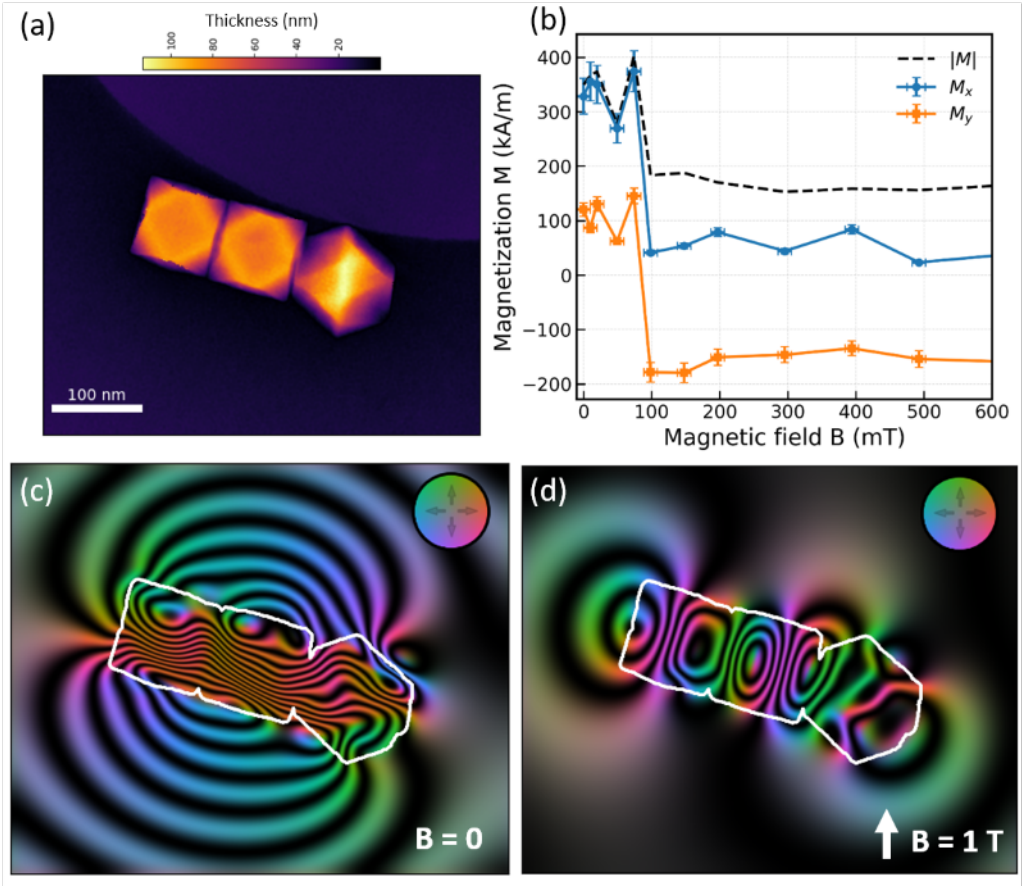

Figure 5: Caption figure: (a) Chain of magnetite nanocubes imaged by electron holography. The magnetic induction lines are shown in (c) without external field and (d) with a field of 1 T applied along the y-axis. (b) Magnetization curve measured on the chain.

1 X. Ye et al., "Quantification of magnetization of nanostructures via three complementary methods,"Appl. Phys. Lett., vol. 126, no. 24, p. 242403, Jun. 2025, doi: 10.1063/5.0266886.
[2] G. Ummethala et al., "Real-time visualisation of fast nanoscale processes during liquid reagent mixing by liquid cell transmission electron microscopy,"Commun Chem, vol. 8, no. 1, p. 8, Jan. 2025, doi: 10.1038/s42004-025-01407-3.
[3] M. Khelfallah et al., "Structural and Magnetic Properties of Ferrofluids Composed of Self-Assembled Cobalt Ferrite Nanoflowers: A Multiscale Investigation,"J. Phys. Chem. C, Jul. 2024, doi: 10.1021/acs.jpcc.4c00790.

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