The Quantitative Measurement of Magnetic Moments from Phase Images of Nanoparticles and Nanostructures

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The phase shift of the electron wave front in a transmission electron microscope can be used to obtain information about a magnetic sample such as the magnetization or coercivity of a region of interest, as well as about magnetic interactions and transitions between single- and multi-domain magnetic states. A key quantity, whose determination from a phase image has not previously been addressed in depth, is the magnetic moment $\mathbf{m} = \iiint \mathbf{M}(\mathbf{r})\mathrm{d}^3\mathbf{r}$ of a nanoparticle or nanostructure, where $\mathbf{M}(\mathbf{r})$ is the magnetization of the structure and the integral is performed over its volume. The difficulty of measuring $\mathbf{m}$ results from the fact that a phase image does not provide direct information about $\mathbf{M}(\mathbf{r})$; instead, the phase shift is proportional to the projection (along the beam path) of the in-plane components of the magnetic induction $\mathbf{B}(\mathbf{r})$ both within and around the specimen. Approaches for measuring $\mathbf{m}$ based on integration of the phase gradient have previously been suggested [1,2], but neither justified nor derived rigorously.

We have recently shown mathematically that the magnetic moment of a nanostructure can be measured *quantitatively* from a phase image or, equivalently, from its gradient components [2]. The basis of our algorithm is the relationship between the volume integral of the induction, a quantity that we call the "inductive moment" $\mathbf{m_B}$, and the true magnetic moment $\mathbf{m}$. If a circular boundary is chosen when integrating the phase shift in the form

$$\mathbf{m_B} = \frac{\hbar R}{e\mu_0}\int_0^{2\pi} \mathrm{d}\theta[-\sin\theta,\cos\theta,0]\varphi(R\cos\theta,R\sin\theta),$$

then it is always true that $\mathbf{m}=2\mathbf{m_B}$. This relation can be utilized to study particles of arbitrary shape and magnetization state. Furthermore, the two components of $\mathbf{m_B}$ can be extrapolated to a circle of zero radius to yield a measurement free of most artifacts (see Ref. [3]). Here, we illustrate the measurement of magnetic moments from an experimental phase image.

Figure 1 shows an example of an experimental electron hologram and a corresponding magnetic induction map (displayed in the form of contours generated from the magnetic contribution to the phase shift) acquired from three closely spaced ferrimagnetic crystals of magnetite (Fe_3O_4). In order to measure the magnetic moment of the crystal, we integrate the magnetic phase, shown in Figure 2, within circular boundaries of increasing radius, as allowed by the field of view. We then extract the inductive moment from each measurement according to the formula given above. In this case, the minimum radius that lies outside the physical boundary of the three crystals, about 100 nm, yields a moment of 2.29 10^6 μ_B oriented at 136° (see Fig. 2(a)). The maximum radius compatible with the field of view and centered on the same position, ~240 nm, yields a moment of 1.02 10^6 μ_B oriented at 126° (see Fig. 2(b)). After extrapolating the measurements to an integration circle of zero radius, the best-fitting values for the two moment components are $\mathbf{m_B}^x=-(1.99\pm0.14)10^6$ μ_B and $\mathbf{m_B}^y=(1.89\pm0.22)10^6$ μ_B, which, together, result in a vector of modulus $(2.74\pm0.18)10^6$ μ_B oriented at (136±4)°, approximately 20% larger in modulus (but oriented similarly) than that inferred when using the circle of smallest radius.

To compare the results with a predicted value, we estimated the radii of the three magnetic particles to be approximately (20±2) nm; dividing the magnetic moment (twice the inductive moment, or $(5.5\pm0.4)10^6$ μ_B) by the total estimated volume of the particles $(1.0\pm0.2)10^6$ nm^3, we obtain an average magnetization of (0.64±0.12) T. The expected magnetization of magnetite, as reported in the literature, is ~0.6 T, confirming the soundness of our approach.

Future work will include extending the approach to allow measurement of the moment of each particle in a set, as well as true local hysteresis loops of individual nanostructures.

References

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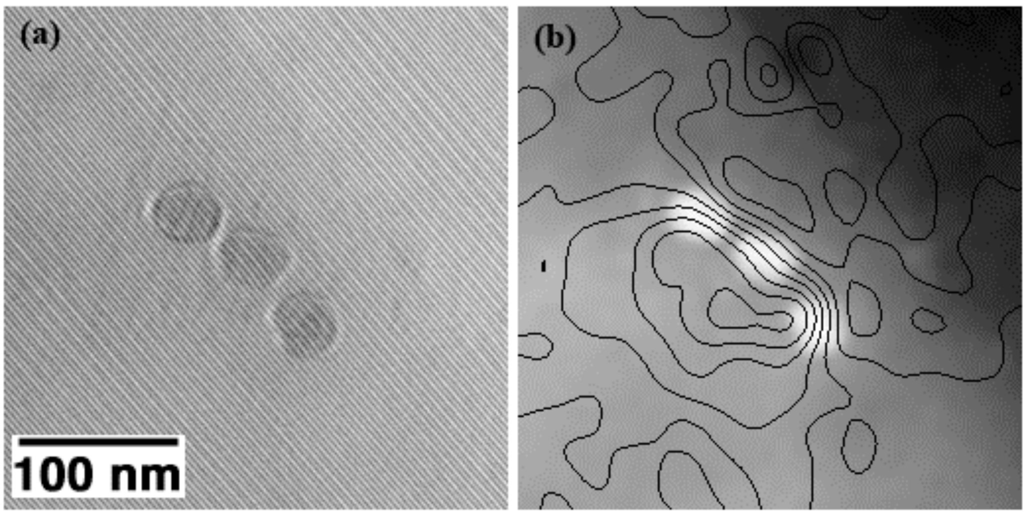

FIG. 1. (a) Off-axis electron hologram recorded at 300 kV in magnetic-field-free conditions from a chain of three approximately equidimensional ferrimagnetic magnetite nanocrystals supported on amorphous carbon. (b) Corresponding magnetic induction map, with 0.0625 rad phase contours superimposed onto the mean inner potential contribution to the recorded phase shift.

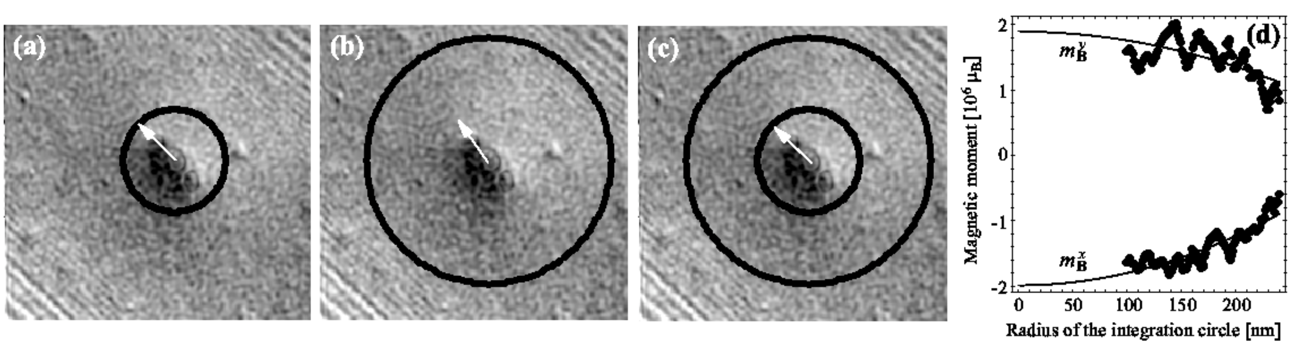

FIG. 2. (a)-(c) Magnetic phase shift corresponding to the hologram and magnetic induction map shown in Fig. 1. The circles and arrows show the integration circle and the direction of the resulting inductive moment for radii of: (a) 100 nm, (b) 240 nm, and c) when using radii between 100 and 240 nm to obtain a value for the magnetic moment extrapolated quadratically to zero radius. (d) Parabolic fit of the two orthogonal components of the measured inductive moment.