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The quantitative measurement of magnetic moments from phase images of nanoparticles and nanostructures—I. Fundamentals

Marco Beleggia *, Takeshi Kasama, Rafal E. Dunin-Borkowski

Center for Electron Nanoscopy, Technical University of Denmark, DK-2800 Kgs. Lyngby, Denmark

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ABSTRACT

An approach that can be used to measure the magnetic moment of a magnetized nanoparticle or nanostructure from an electron-optical phase image is introduced. The measurement scheme is based on integration of the gradient of the measured phase image within a circular boundary that contains the structure of interest. The quantity obtained is found to be directly proportional to the magnetic moment of the particle, with a constant of proportionality that does not depend on the particle's shape or magnetization state. The measurement of magnetic moments from both simulated and experimental phase images is demonstrated, and strategies are presented that can be utilized to overcome sources of error associated with, for example, the presence of neighboring magnetic particles and the perturbation of the holographic reference wave.

1. Introduction

Measurements of electron-optical phase shifts in transmission electron microscope (TEM) images, using techniques that include off-axis electron holography [1,2] and approaches based on the transport of intensity equation [3,4], are increasingly used to record maps of the projected in-plane magnetic induction of magnetized nanocrystals [5,6], nanostructures [7–9], thin films and devices [10,11], often with sub-10-nm spatial resolution [12]. Such phase images can be used to obtain quantitative information about parameters 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 parameter, whose determination from a phase image has not previously been addressed in depth, is the magnetic moment of a nanoparticle or nanostructure

$$\mathbf{m} = \iiint \mathbf{M}(\mathbf{r})\,\mathrm{d}^3\mathbf{r}, \tag{1}$$

where $\mathbf{M}(\mathbf{r})$ is the position-dependent magnetization of the structure and $\mathbf{r}$ is a three-dimensional position vector. The conceptual and mathematical difficulty of measuring $\mathbf{m}$ stems from the fact that a phase image does not provide direct information about the magnetization field $\mathbf{M}(\mathbf{r})$; instead, the phase shift is proportional to a projection of the in-plane components of the three-dimensional magnetic induction vector field $\mathbf{B}(\mathbf{r})$, both within and around the specimen. Approaches for measuring $\mathbf{m}$ based on integration of the phase gradient have previously been suggested [13,14], but neither justified nor derived rigorously.

Here, we show mathematically that the magnetic moment of a nanoparticle or nanostructure can be measured quantitatively from either a phase image or its gradient components. We show that the approach can be employed to study particles with an arbitrary shape and magnetization state, and that no assumptions are necessary to extract the information. We establish strategies that can be used to identify and overcome sources of error, and we demonstrate the measurement of magnetic moments from both simulated and experimental phase images.

Fig. 1 shows an example of a representative experimental electron hologram and a corresponding projected magnetic induction map (comprising contours generated from the magnetic contribution to the recorded phase shift) acquired from three closely spaced ferrimagnetic crystals of magnetite (Fe_3O_4). The induction map shown in Fig. 1 is discussed and analyzed in detail below.

2. Basic considerations

The approach that we describe in this paper is based on the fundamental expression for the magnetic contribution to the electron-optical phase shift (referred to as "magnetic phase") recorded from a magnetized specimen, which, in the reference frame of the microscope, can be written in the form [12]

$$\varphi(\mathbf{r}_\perp) = -\frac{e}{\hbar}\int_{-\infty}^{+\infty} A_z(\mathbf{r}_\perp, z)\,\mathrm{d}z, \tag{2}$$

* Corresponding author.
E-mail address: mb@cen.dtu.dk (M. Beleggia).

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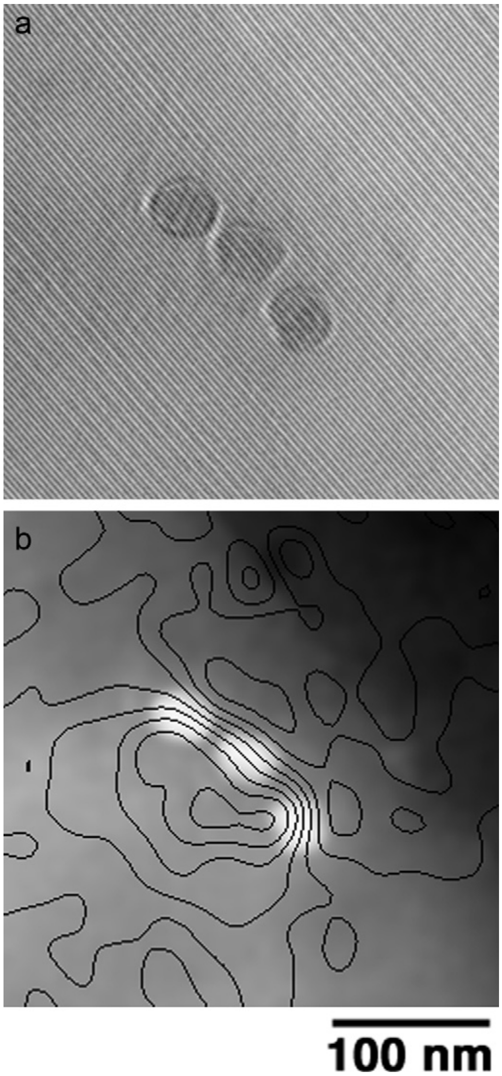

Fig. 1. (a) Off-axis electron hologram recorded at 300 kV in magnetic-field-free conditions from a chain of three approximately equidimensional ferrimagnetic magnetite nanocrystals supported on amorphous carbon. The crystals were extracted from a magnetotactic bacterium and deposited onto a holey carbon support film. The hologram was acquired using a Philips CM300 field emission gun TEM equipped with a Lorentz lens and an electrostatic biprism. (b) Corresponding magnetic induction map, formed by generating contours of spacing 0.0625 radians from the magnetic contribution to the recorded phase shift and superimposing them onto the mean inner potential contribution. The magnetic and mean inner potential contributions were determined by calculating half of the difference and half of the sum of phase images, between which the magnetization direction in the specimen had been reversed by applying a magnetic field to the specimen using the field of the conventional microscope objective lens.

where $\mathbf{A}$ is the magnetic vector potential, the electron beam direction is along $-z$, and $\mathbf{r}_\perp$ is a two-dimensional position vector in the object plane. The two orthogonal components of the magnetic phase gradient are

$$\partial_x \varphi(\mathbf{r}_\perp) = -\frac{e}{\hbar}\int_{-\infty}^{+\infty} \partial_x A_z(\mathbf{r}_\perp, z)\,\mathrm{d}z \tag{3}$$

and

$$\partial_y \varphi(\mathbf{r}_\perp) = -\frac{e}{\hbar}\int_{-\infty}^{+\infty} \partial_y A_z(\mathbf{r}_\perp, z)\,\mathrm{d}z. \tag{4}$$

As the relationship between $\mathbf{B}$ and $\mathbf{A}$

$$\mathbf{B} = \nabla \times \mathbf{A}, \tag{5}$$

can be written explicitly as

$$B_x = \partial_y A_z - \partial_z A_y \tag{6}$$

and

$$B_y = -\partial_x A_z + \partial_z A_x, \tag{7}$$

Eqs. (3) and (4) can be rewritten in the forms

$$\begin{aligned}\frac{\hbar}{e}\partial_x \varphi(\mathbf{r}_\perp) &= \int_{-\infty}^{+\infty} B_y(\mathbf{r}_\perp, z)\,\mathrm{d}z - \int_{-\infty}^{+\infty} \partial_z A_x(\mathbf{r}_\perp, z)\,\mathrm{d}z \\ &= \int_{-\infty}^{+\infty} B_y(\mathbf{r}_\perp, z)\,\mathrm{d}z - A_x(\mathbf{r}_\perp, z)|_{z=-\infty}^{z=+\infty} \\ &= \int_{-\infty}^{+\infty} B_y(\mathbf{r}_\perp, z)\,\mathrm{d}z\end{aligned} \tag{8}$$

and, similarly,

$$-\frac{\hbar}{e}\partial_y \varphi(\mathbf{r}_\perp) = \int_{-\infty}^{+\infty} B_x(\mathbf{r}_\perp, z)\,\mathrm{d}z \tag{9}$$

due to the fact that any vector potential generated by finite magnetic sources must decay to zero at infinity. Eqs. (8) and (9) highlight the fact that the phase gradient component along a given direction is directly proportional to the magnetic induction component in the perpendicular direction, projected along the beam path. This relationship between the components of the phase gradient and the projected induction can be written in a single expression

$$\begin{aligned}\mathbf{B}_\mathrm{p}(\mathbf{r}_\perp) &= \int_{-\infty}^{+\infty} \mathbf{B}(\mathbf{r})\,\mathrm{d}z = \frac{\hbar}{e}[-\partial_y \varphi(\mathbf{r}_\perp), \partial_x \varphi(\mathbf{r}_\perp)] \\ &= \frac{\hbar}{e}[\hat{\mathbf{z}} \times \nabla\varphi(\mathbf{r}_\perp)]\end{aligned} \tag{10}$$

and also forms the basis of proposals for magnetic vector field electron tomography [15,16].

Integration of the phase gradient would result in the expressions

$$\frac{\hbar}{e}\iint \partial_x \varphi(\mathbf{r}_\perp)\,\mathrm{d}^2\mathbf{r}_\perp = \iiint B_y(\mathbf{r}_\perp, z)\,\mathrm{d}^2\mathbf{r}_\perp\,\mathrm{d}z = \iiint B_y(\mathbf{r})\,\mathrm{d}^3\mathbf{r} \tag{11}$$

and

$$\begin{aligned}\frac{\hbar}{e}\iint \partial_y \varphi(\mathbf{r}_\perp)\,\mathrm{d}^2\mathbf{r}_\perp &= -\iiint B_x(\mathbf{r}_\perp, z)\,\mathrm{d}^2\mathbf{r}_\perp\,\mathrm{d}z \\ &= -\iiint B_x(\mathbf{r})\,\mathrm{d}^3\mathbf{r}\end{aligned} \tag{12}$$

in which the integrals are performed over some domains to be specified. Although the integrals in Eqs. (11) and (12) do not provide the "magnetic moment" as defined in Eq. (1) directly, their forms suggest that a relationship may exist between the quantities described in each of the expressions and the components of the desired moment $\mathbf{m}$. In this paper we show that such a relationship does exist, and we establish an approach for measuring $\mathbf{m}$ experimentally. For convenience, the quantity

$$\mathbf{m_B} = \frac{1}{\mu_0}\iiint \mathbf{B}(\mathbf{r})\,\mathrm{d}^3\mathbf{r} \tag{13}$$

will be referred to below as "inductive moment". The inductive and magnetic moments have the same units (A m^2 or J/T in SI), and we choose to measure both quantities in multiples of Bohr magnetons $\mu_B = 9.274 \times 10^{-24}$ J/T.