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Separation of electrostatic and magnetic phase shifts using a modified transport-of-intensity equation

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ABSTRACT

We introduce a new approach for the separation of the electrostatic and magnetic components of the electron wave phase shift, based on the transport-of-intensity equation (TIE) formalism. We derive two separate TIE-like equations, one for each of the phase shift components. We use experimental results on FeCoB and Permalloy patterned islands to illustrate how the magnetic and electrostatic longitudinal derivatives can be computed. The main advantage of this new approach is the fact that the differences in the power spectra of the two phase components (electrostatic phase shifts often have significant power in the higher frequencies) can be accommodated by the selection of two different Tikhonov regularization parameters for the two phase reconstructions. The extra computational demands of the method are more than compensated by the improved phase reconstruction results.

1. Introduction

The ability to image the magnetic domain structure in magnetic materials is critical to understanding the way in which these materials respond to an external field and how this behavior is related to the microstructure of the material. Applications of magnetic materials are wide-ranging and include for example magnetic recording media in which the magnetic domains control bit size, and permanent magnets. There are a number of techniques that can be used to image the magnetic domain structure [1] of which the highest spatial resolution is obtainable using Lorentz transmission electron microscopy (LTEM) [2]. This technique has the additional advantage that the microstructure and composition of the materials can be explored in the same instrument for direct correlation with the domain structure. Recent examples of the use of Lorentz microscopy are the study of nanoscale magnetic interactions in square artificial spin ice lattices, in particular the structure of Dirac strings and “magnetic monopoles” [3,4], *in situ* studies of the motion of domain walls in Ni_2MnGa type ferromagnetic shape memory alloys [5] and the interaction of magnetic domain walls with twin boundaries in FePdCo alloys [6].

In LTEM one measures the phase shift of the incident electron beam due to the sample's magnetic induction, which leads to a deflection of the electron beam. This is achieved by defocusing the imaging lens by a large amount [7], a technique known as Fresnel imaging. Typical deflection angles are in the range of tens to hundreds of microradians, i.e., about two orders of magnitude smaller than typical Bragg angles, so that defocus values of several to hundreds of micrometers are not unusual. The large defocus generates significant defocus blurring, so that the spatial resolution of Lorentz images is usually estimated to be around 10 nm. Along with the defocus blurring, in field emission instruments the Fresnel images also suffer from significant delocalization due to the large spherical aberration of the long focal length Lorentz lens used for this type of observation; C_s values of several meters are not unusual [8].

The combination of a Lorentz pole piece in which the sample can be imaged whilst sitting in a low magnetic field, unlike in conventional TEM objective lenses, with an imaging spherical-aberration corrector provides improved spatial resolution in Lorentz mode, because the correction of C_s down to a few microns lowers the amount of delocalization, and, hence, the amount of defocus needed to render the magnetic domain walls visible. The reduced defocus, in turn, leads to reduced defocus blurring and therefore an improved spatial resolution of the order of 1 nm [9–11]. In addition, the reduced defocus brings the Fresnel image mode into the realm of applicability (the so-called *small defocus limit* [12]) of the transport-of-intensity equation (TIE) formalism for electron wave phase reconstruction [13,14].

The TIE formalism can be used to reconstruct the phase of the electron wave at the detector plane from a through-focus series centered at zero defocus. Due to the small angles involved in Lorentz deflection, one can show that the application of TIE to Lorentz images enables the phase shift of the sample exit wave function to be obtained, provided the small defocus limit is

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observed. The total phase shift consists of two contributions: an electrostatic phase shift due to the mean inner potential and, potentially, any localized electrostatic polarization, and a magnetic phase shift due to the magnetic induction of the sample. In most cases, the magnetic phase shift is rather weak compared to the electrostatic component, so that it can become difficult to extract the magnetic information. It is then common to record a second through-focus series with the sample flipped upside down, since the lack of time-reversal symmetry of the magnetic phase shift will change its sign relative to the electrostatic phase shift. A simple subtraction then produces the magnetic phase shift.

In this contribution, we present an alternative approach to the separation of electrostatic and magnetic phase shifts using the TIE formalism. In Section 2 we describe the transport-of-intensity equation and how it is usually solved using fast Fourier Transforms (FFTs). Then we introduce a new approach for phase shift separation, based on the linearity of the TIE formalism, and illustrate its benefits using a computational model. Finally, in Section 3 we present a series of experimental phase reconstructions on a patterned Permalloy sample and on amorphous rectangular CoFeB islands. We conclude with a brief summary of results.

2. Theory

2.1. The transport-of-intensity equation

Under paraxial conditions, the phase of an electron wave is coupled to the intensity gradient along the electron trajectory. Paganin and Nugent [13] derived the so-called transport-of-intensity equation (TIE) formalism, which expresses this relation mathematically. If the total phase shift of the electron wave is represented by the symbol $\varphi_t = \varphi_e + \varphi_m$, with φ_e the electrostatic and φ_m the magnetic phase shifts, then the TIE reads

$$\nabla \cdot (I_0 \nabla \varphi_t) = -k \partial_z I, \tag{1}$$

where I_0 is the intensity of the in-focus image, $k = 2\pi/\lambda$ is the wave number, and the z-direction is parallel to the electron propagation direction. The ∇ differential operators are two-dimensional and operate in the plane normal to the z-axis, and the symbol ∂_z represents the partial derivative with respect to the variable z. It should be noted that the gradient of the magnetic component of the total phase shift can be related to the integrated magnetic induction by the relation [15]

$$\nabla \varphi_m = -\frac{\pi}{\phi_0} (\mathbf{B} \times \hat{\mathbf{n}}) t, \tag{2}$$

where t is the foil thickness, $\hat{\mathbf{n}}$ a unit vector along the beam direction, and ϕ_0 the flux quantum. Note that this relation is only meaningful for samples with uniform thickness and vanishing fringing fields.

The TIE formalism can be derived in a number of different ways, notably from a paraxial approximation to the free-space Schrödinger equation [16], or from the real-space propagator equation [17]. For Lorentz microscopy conditions, with small scattering angles, the TIE follows directly from a small-angle approximation to the Lorentz lens point-spread function [18]. An elegant numerical solution approach to the TIE formalism was proposed by Paganin and Nugent [13]; their approach uses Fourier transforms to implement the inverse differential operators ∇^{-1} and ∇^{-2}. The total phase shift can be written formally by the following expressions:

$$\phi_t = -k \nabla^{-2} \left[\nabla \cdot \left(\frac{1}{I_0} \nabla (\nabla^{-2} [\partial_z I_0]) \right) \right]; \tag{3}$$

$$\phi_t = -k \nabla^{-1} \cdot \left(\frac{1}{I_0} (\nabla^{-1} [\partial_z I_0]) \right). \tag{4}$$

Comparing this relation with Eq. (2) indicates that the integrated magnetic induction components can be obtained by partially solving the TIE formalism using a single application of the inverse gradient operator ∇^{-1} [19, p. 655], provided that there are no spatial variations in the electrostatic phase shift. However, this is unlikely to be the case for realistic LTEM samples as there will be electrostatic potential variations due to either sample thickness variations or to the lack of materials in between patterned nanostructures. For a derivation of the conditions under which the TIE formalism is applicable to Lorentz images we refer to Ref. [12]. A description of the boundary conditions used to solve the equation can be found in [20].

2.2. Separation of electrostatic and magnetic phase shifts

In this section, we describe a new approach to the separation of electrostatic and magnetic phase components. In the conventional approach, which we will summarize first, one reconstructs the total phase shift, $\varphi_t^+ = \varphi_e + \varphi_m$, using a through-focus series, I_-^+, I_0^+, and I_+^+, for the sample in the upright orientation, indicated by the superscript $+$; the defocus amount is indicated by the subscript. The TIE in this case is given by

$$\nabla \cdot (I_0^+ \nabla \varphi_t^+) = -k \partial_z I^+ \approx -\frac{k}{2\,\Delta f} (I_+^+ - I_-^+). \tag{5}$$

The second equality is valid in the small defocus limit and approximates the longitudinal derivative as the difference between overfocus and underfocus images.

When the sample is flipped upside down, a second through-focus series I_-^-, I_0^-, and I_+^- is used to solve a second TIE for the total phase $\varphi_t^- = \varphi_e - \varphi_m$ (the sign change reflects the fact that the magnetic phase shift is not invariant with respect to time reversal symmetry):

$$\nabla \cdot (I_0^- \nabla \varphi_t^-) = -k \partial_z I^- \approx -\frac{k}{2\Delta f} (I_+^- - I_-^-). \tag{6}$$

The electrostatic and magnetic phase shifts are then obtained by adding and subtracting the two reconstructed phases:

$$\varphi_e = \frac{1}{2} [\varphi_t^+ + \varphi_t^-]; \tag{7}$$

$$\varphi_m = \frac{1}{2} [\varphi_t^+ - \varphi_t^-]. \tag{8}$$

This traditional approach can be used with the TIE formalism as well as with standard off-axis holography phase reconstructions.

The linearity of the transport-of-intensity equation suggests that separation of the phase shifts may be accomplished also by substituting the expressions for φ_t^+ and φ_t^- into the respective TIEs (5) and (6):

$$\nabla \cdot (I_0 \nabla \varphi_e) + \nabla \cdot (I_0 \nabla \varphi_m) = -k \partial_z I^+;$$
$$\nabla \cdot (I_0 \nabla \varphi_e) - \nabla \cdot (I_0 \nabla \varphi_m) = -k \partial_z I^-,$$

where we have made use of the fact that $I_0^+ = I_0^- \equiv I_0$. Adding and subtracting these equations we obtain

$$\nabla \cdot (I_0 \nabla \varphi_e) = -k \partial_z^e I \quad [\text{TIE}-\text{e}] \tag{9}$$

$$\nabla \cdot (I_0 \nabla \varphi_m) = -k \partial_z^m I \quad [\text{TIE}-\text{m}] \tag{10}$$

where we define

$$\partial_z^e I \equiv \frac{1}{2} [\partial_z I^+ + \partial_z I^-]; \tag{11}$$

$$\partial_z^m I \equiv \frac{1}{2} [\partial_z I^+ - \partial_z I^-], \tag{12}$$