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Experimental evaluation of the 'transport-of-intensity' equation for magnetic phase reconstruction in Lorentz transmission electron microscopy

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ABSTRACT

The 'transport-of-intensity' equation (TIE) is a general phase reconstruction methodology that can be applied to Lorentz transmission electron microscopy (TEM) through the use of Fresnel-contrast (defocused) images. We present an experimental study to test the application of the TIE for quantitative magnetic mapping in Lorentz TEM without aberration correction by examining sub-micrometer sized $Ni_{80}Fe_{20}$ (Permalloy) elements. For a *JEOL JEM 2100F* adapted for Lorentz microscopy, we find that quantitative magnetic phase reconstructions are possible for defoci distances ranging between approximately 200 μm and 800 μm. The lower limit originates from competing sources of image intensity variations in Fresnel-contrast images, namely structural defects and diffraction contrast. The upper defocus limit is due to a numerical error in the estimation of the intensity derivative based on three images. For magnetic domains, we show quantitative reconstructions of the product of the magnetic induction vector and thickness in element sizes down to approximately 100 nm in lateral size and 5 nm thick resulting in a minimal detection of 5 T nm. Three types of magnetic structures are tested in terms of phase reconstruction: vortex cores, domain walls, and element edges. We quantify vortex core structures at a diameter of 12 nm while the structures of domain walls and element edges are characterized qualitatively. Finally, we show by image simulations that the conclusions of this experimental study are relevant to other Lorentz TEM in which spherical aberration and defocus are dominant aberrations.

1. Introduction

Imaging the magnetic structure of materials at the nanometer scale is motivated by the scientific study of magnetic phenomena and the technological drive to develop new devices, in particular for information storage devices [1,2]. The magnetic structure can be imaged qualitatively and mapped quantitatively using transmission electron microscopy (TEM) in a variety of contrast modes, so-called 'Lorentz TEM' [3,4,52], for example, Fresnel-contrast defocused images. Magnetic imaging in the TEM is possible because in the presence of a magnetic (and electric) potential, the wave function of the electron undergoes a phase shift [5].

Consequently, quantitative mapping of the magnetic induction vector at nanometer scale spatial resolution can be extracted. However, measuring the phase of an electron wave exiting the sample is challenging because the phase component of the electron wave which reaches the imaging detector is lost due to the quantum mechanical nature of the measurement. Additionally, the phase component is influenced by aberrations of the microscope and the signal collected at the detector further influenced by the modulation transfer function (MTF) of the charge-coupled device [6].

Here we examine experimentally quantitative magnetic imaging using the 'transport of intensity' equation (TIE) in conventional Lorentz TEM as a phase reconstruction methodology. By conventional Lorentz TEM, we refer to a microscope with a significant spherical aberration.

The principle of this methodology, developed by Teague for wave propagation in general [7], is based in the case of Lorentz TEM on Fresnel-contrast images. A focal series enables to correlate the intensity, phase and intensity derivative with respect to the direction of the wave propagation [8]. A detailed description on how this methodology was applied in this study is presented later.

The TIE approach for magnetic imaging in the Lorentz TEM can be a complementary methodology to off-axis electron holography. In terms of phase reconstruction, the TIE is more sensitive at higher spatial frequencies, while off-axis electron holography is better at lower spatial frequencies [9] leading to a suggestion to

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combine both methodologies [10].

For practical application, the TIE methodology has several advantages compared to off-axis electron holography. First, it can be applied in most conventional TEMs, including those with reduced coherency of the electron source, e.g. LaB_6 [8,11]. The method is less demanding in terms of sample requirements. The sample is not required to be prepared so that the region of interest is placed near the vacuum, which may modify the micromagnetic structure. The field of view is larger than typically found in off-axis electron holography. Finally, the TIE approach is more convenient for in situ experiments.

The objective of this research is to test experimentally the quantitative capability of the TIE phase reconstruction approach specifically for magnetic imaging in conventional Lorentz TEM, as manifested in the measurement of the phase gradient, which relates to the product of the magnetic induction, *B*, and thickness of the sample, *t*. The aims of this examination are to determine the limit of detection, and the accuracy of the measured *Bt* product, as well as the spatial detection limit of magnetic structures.

Most reports on quantitative electron phase reconstructions using the TIE methodology are for electrostatic potentials in non-magnetic materials, e.g. Refs. [12–17]. The phase sensitivity of the TIE for the electrostatic potential phase step was estimated theoretically at $\pi/20$ rad [18] and possible phase accuracy of up to $\pi/300$ rad [9]

There are fewer reports on quantitative application of the TIE methodology to magnetic materials, typically applied to micrometer scale structures e.g. Refs. [11,19–22,53]. When applied to the study of magnetic materials, Masseboeuf et al. [23] studied a chemically ordered FePd thin film, reporting a *Bt* detection of several tens of *T* nm using an FEI Titan equipped with a dedicated Lorentz lens. A spatial resolution of 10 nm was estimated based on the detection of domain walls. Phatak et al. [24] applied the TIE to a three-dimensional reconstruction of the magnetic vector potential in a square 1 μm Permalloy element using a JEOL 2100F equipped with a Lorentz objective lens. The vector potential was probed at a spatial resolution of approximately 13 nm.

We tested experimentally the quantitative capability of the TIE by examining cases for phase reconstruction of lithographically patterned $Ni_{80}Fe_{20}$ (Permalloy) elements in terms of varying lateral size and thicknesses. For quantitative analysis, the reconstructions were evaluated in terms of phase gradients along magnetic domains, which represent the *Bt* product. Additionally, the phase reconstruction from magnetic structures in the form of vortex cores, 90° domain walls, and elements edges were evaluated.

Experimentally, the limit of the magnetic detection for phase gradients in magnetic domains was found to be around 10 rad/μm, which in terms of the *Bt* product is 5 T nm in Permalloy elements sized laterally down to approximately 100 *nm.* The accuracy of the magnetic phase gradient reconstruction was under 1 rad/μm. In order to achieve quantitative phase reconstructions, both lower and upper limits for defoci distances were observed of 200 μm and 800 μm, respectively. The smallest magnetic structures that could be quantitatively detected were vortex cores 12 nm in diameter. The magnetic structures of domain walls and element edges were characterized qualitatively by phase reconstructions.

2. Methodology

2.1. Magnetic phase reconstruction using the transport of intensity equation

Magnetic samples imaged at the nanometer scale are described as phase objects according to the Aharonov and Bohm equation [6], which quantifies the influence of electrostatic and magnetic potentials. By assuming a constant contribution from the electrostatic potential to the phase shift, ϕ, of a sample with uniform thickness, *t*, and a constant magnetic induction, *B*, throughout the thickness of the sample, the phase change along one direction in a magnetic domain can be represented by:

$$\phi(x) = \frac{et}{\hbar}\int_{x_0}^{x_0+x} B_y dx \;=\; \frac{e}{\hbar} B_y tx \tag{1}$$

where *x* is the direction in the plane of the sample perpendicular to the optic axis; B_y is the magnetic induction of the sample perpendicular to both *x* and the optic axis; x_0 coordinate represents the location of a domain wall.

Therefore, the in-plane component of the magnetic induction vector, $B_\perp$, is derived by applying a gradient to the reconstructed phase:

$$\nabla\varphi(x, y) = -\frac{e}{\hbar}(B \times \hat{n}_z)t \;=\; -\frac{e}{\hbar}B_\perp t \tag{2}$$

where $\hat{n}_z$ is a vector unit along the *z* direction, namely the optic axis.

As noted, the TIE approach is a general phase reconstruction methodology, which was first applied to visible light microscopy [26], followed by neutrons [27], X-ray microscopy [28], and finally, the topic of this research, for fast electrons.

The TIE shows that the phase can be determined by intensity variation measurements:

$$\nabla_\perp\cdot[I(r_\perp, z)\nabla_\perp\phi(r_\perp, z)] = -k\frac{\partial I(r_\perp, z)}{\partial z} \tag{3}$$

The TIE correlates between image intensity, *I*, phase shift, ϕ, of the electron wave-function and the derivative of the image intensity along the direction of the wave propagation, *z* (optic axis), where $r_\perp$ represents the radial coordinate perpendicular to the optic axis and *k* represents the wavenumber of the fast electrons. Information transfer is attenuated by the square of the spatial frequency, meaning that transmission of low spatial frequencies is suppressed [29,30].

Solving differential Eq. (3) and optimizing the phase reconstruction result can be achieved by several routes. Pagannin and Nugent suggest a Fourier-transfom based approach [31], which was used in this study through the 'QPt' algorithm [32], as represented by Eq. (4):

$$\varphi(r_\perp, z) = -k\nabla_\perp^{-2}\nabla_\perp\left(\frac{1}{I(r_\perp, z)}\nabla_\perp\nabla_\perp^{-2}\frac{\partial I(r_\perp, z)}{\partial z}\right) \tag{4}$$

where $\nabla_\perp^{-2}$ is the inverse Laplacian operator, which in the Fourier space is [33]:

$$\nabla_\perp^{-2} f(x, y) = FT^{-1}\left[\frac{FT[f(x, y)]}{|q_\perp|^2}\right] \tag{5}$$

The symbols *FT* and FT^{-1} represent forward and inverse Fourier transforms, respectively, and $q_\perp$ is the spatial frequency vector normal to the propagation direction.

For solving the TIE equation, the intensity derivative with respect to the optic axis, *z* is estimated [15]. In practice, we acquire two defocused (Fresnel-contrast) images, $I(r_\perp, -\Delta z)$ and $I(r_\perp, +\Delta z)$, and a Gaussian image, $I(r_\perp, 0)$. Due to small magnetic deflection angles, in this work down to several μrad, large defoci distances are required, typically 100–1000 μm, for achieving Fresnel-contrast images with sufficient contrast. Consequently, alignment between defocused images is critical because of the practical large image rotation and change in magnification observed at the microscope. Barty et al. [34] and McVitie et al. [30] show that a misalignment of a single pixel is sufficient to