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Recent advances in Lorentz microscopy

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ABSTRACT

Lorentz transmission electron microscopy (LTEM) has evolved from a qualitative magnetic domain observation technique to a quantitative technique for the determination of the magnetization state of a sample. In this review article, we describe recent developments in techniques and imaging modes, including the use of spherical aberration correction to improve the spatial resolution of LTEM into the single nanometer range, and novel *in situ* observation modes. We review recent advances in the modeling of the wave optical magnetic phase shift as well as in the area of phase reconstruction by means of the Transport of Intensity Equation (TIE) approach, and discuss vector field electron tomography, which has emerged as a powerful tool for the 3D reconstruction of magnetization configurations. We conclude this review with a brief overview of recent LTEM applications.

1. Introduction

Lorentz transmission electron microscopy (LTEM) is ideally suited to the quantitative analysis of magnetic domain structures at the sub-50 nm length scale [1]. The ability to image both the microstructure and the magnetic domain structure of an engineering material in the same instrument allows for a direct study of how the often inhomogeneous microstructure plays an important role in influencing the magnetic behavior.

After the early developments by Hale [2] and Boersch and Raith [3], LTEM has seen a steady but slow stream of improvements in both spatial resolution and image quality. In classical terms, high energy electrons traversing a magnetized foil are deflected by the Lorentz force generated by the magnetic induction of the foil. In a quantum mechanical approach, the electron wave passing through the sample is imparted a phase shift which is dependent on the magnetic vector potential of the thin foil. The magnetic vector potential in turn relies on the magnetization of the sample and not on the magnetic induction, and as such LTEM images give direct information about the magnetization state of the sample. It should be noted that the effect of stray fields is also included in these images and has to be treated correctly in interpreting them. For many thin foils the stray fields are minimum and we obtain direct information about the magnetization of the sample; however in magnetic nanostructures, the LTEM images contain integrated information from the magnetization as well as the stray fields. This results in split spots in the diffraction pattern oriented according to the directions of magnetization in the sample, and in magnetic contrast visible in the images. The out-of-focus (Fresnel) mode gives rise to images in which the positions of the magnetic domain walls appear as alternate bright and dark lines, and these can be used to obtain qualitative information. The Fresnel mode is also useful for real-time studies of magnetization reversal (see Section 2.3 below) as it is relatively easy to implement, and additionally, a through-focal series of Fresnel mode images forms the input data set from which phase reconstruction of quantitative magnetic induction maps is carried out, as discussed in Section 3.3 below. For the Foucault mode the objective lens is kept in-focus and one of the split spots in the diffraction pattern is blocked by displacing an aperture located coplanar with the diffraction pattern. Only the domains in which the magnetization orientation is such that the electrons are deflected through the aperture, appear bright. The Foucault mode is also qualitative, and is more difficult to implement than the Fresnel mode, because the image contrast depends critically on the lateral position of the blocking aperture and on how close the aperture plane is to the back-focal plane of the imaging lens.

For realistic sample thicknesses, the Lorentz deflection angle is typically in the range of tens of micro-radians down to sub micro-radian, i.e., several orders of magnitude smaller than Bragg scattering angles, making it easy to distinguish the two. Direct observa-

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tion of the structure in the split central diffraction spot using low-angle diffraction (LAD) (or small angle electron scattering (SAES)) provides semi-quantitative information about the magnetic domain structure and can also be used to follow magnetization processes. It should be noted that LAD provides global information from the whole of the illuminated specimen area rather than local information.

Differential phase contrast (DPC) microscopy is a scanning TEM (STEM)-based technique. The local Lorentz deflection at the position of the electron probe is determined using an annular quadrant detector sited in the far-field, and the difference signals from opposite segments of the detector provide a direct measure of the two components of the Lorentz deflection, β_L at each position of the probe across the specimen. The DPC technique requires an increase in instrumental complexity compared with the Foucault and Fresnel imaging modes. In addition, a relatively long time is needed to record each image, meaning that recording images that show changes in magnetization as a function of applied stimuli such as short current pulses in the millisecond time range is not possible unless time-dependent effects do not contribute significantly.

In order to obtain quantitative images, one needs to consider the effect of the magnetic material on the phase of the incident electron waves. This wave-optical approach (see Section 3) allows for reconstruction of the phase shift across the image, from which quantitative maps of the magnetic induction can be reconstructed, as is discussed in Section 3.3. Recent advances in aberration correction have lead to improvements in both spatial resolution (down to around 2 nm) and in minimum magnetic induction that can be imaged, as will be discussed in more detail below.

A number of review papers and books that include chapters on the theory and practical application of LTEM have been written in the last few years, and these give a good introduction to the classical and wave-optical theories that describe the technique, to the various modes that can be employed, to methods by which magnetic samples can be maintained in a magnetic field-free environment within the microscope, and to the use of *in situ* experiments to follow magnetization reversal and other processes such as the change in magnetic structure across phase transitions. Examples include chapters in *Magnetic Microscopy and its Applications to Magnetic Materials* edited by De Graef and Zhu [4], a chapter by Petford-Long and Chapman in *Magnetic Microscopy of Nanostructures* [5] and a recent review article by Petford-Long and De Graef [1].

The focus of the current paper is to review the new advances that have contributed to the development of LTEM, in addition to presenting examples that illustrate recent applications of the technique.

2. Developments in techniques and imaging modes

In this section we focus *only* on developments that extend the techniques beyond those discussed in the review papers cited above.

2.1. Imaging modes

Taniguchi et al. [6] have recently presented a method to obtain Foucault images in a non-dedicated LTEM instrument: the objective lens is switched off to provide a field-free region at the sample, and the condenser lens is then used to bring the electron beam to a cross-over in the plane of the selected area aperture, which is used as the blocking aperture. Taniguchi et al. also note that the same configuration can be used to image low-angle diffraction (LAD) patterns at high magnification.

2.2. Spherical aberration-correction

In the last few years, the resolution of LTEM images has been greatly increased by the use of spherical-aberration correction [7]. Objective lenses for Lorentz TEM have much higher aberrations than standard TEM lenses (spherical aberration, C_s, values of 50–8000 mm, and chromatic aberration, C_c, of 20–40 mm versus values of 1–2 mm for C_s and C_c for a high resolution lens). These high values, combined with the fact that LTEM images for Fresnel microscopy and for phase reconstruction are recorded with very high defocus values, substantially limits the spatial resolution that can be achieved for most applications to 5–10 nm. Phatak et al. reported the use of a C_s corrector on a JEOL 2100F microscope fitted with a dedicated Lorentz lens, to obtain high spatial and phase resolution images of magnetic monopole defects at the vertices within an artifial spin ice, which had previously not been obtainable. The corrector enabled the C_s coefficient to be reduced from 120 mm to 0.01 mm, resulting in a reduction in the information limit of the microscope from 0.73 nm to 0.43 nm, together with the ability to reduce the defocus needed for phase reconstructions from 36 μm to 19 μm (see Section 3.3 below).

More recently, McVitie et al. [8] reported results from a spherical aberration-corrected STEM, which has been further improved with the addition of an eight-segment detector, to enable collection of DPC images with a spatial resolution of around 1 nm. The authors comment on the challenges associated with the small Lorentz deflection angles that are subtended by thin TEM samples with low magnetic induction, and on the possibility of using a pixelated detector, after the method reported by Pennycook et al. in which an image of the diffraction disk is recorded at each position of the probe as it is scanned across the sample, and the image is then processed to extract the magnetic contrast [9].

Chromatic aberration correction has not yet been applied to Lorentz TEM, however the main advantage is that it would allow analysis of thicker specimens. This is extremely important when imaging bulk magnetic materials, because of the change in magnetostatic energy terms associated with making a thin TEM sample from the bulk. In addition, correction of C_c would enable a lens with a larger pole-piece gap to be used, thus increasing the space available for *in situ* stages.

2.3. Imaging magnetization reversal behavior

Obtaining a full understanding of the magnetic behavior of a material relies on being able to image specimens in their as-grown state, in remanent states, in the presence of applied fields or currents, and as a function of temperature. From these can be derived basic micromagnetic information, together with features such as the domain wall nature, the presence of domain wall nucleation and pinning sites, and the nature of magnetic phase transitions. In addition, the effects of materials processing (for example, patterning into small elements) on magnetic domain structure and magnetization reversal mechanisms can be analyzed.

In situ magnetic fields can be applied, which enables the local magnetization reversal of a sample to be followed in real-time. A number of approaches have been proposed, including tilting the sample into the field of the objective lens or using a sample holder to which small electromagnets are mounted. Further sophistication can be achieved by combining application of a magnetic field with application of current, as shown by Arita et al. [10] who showed the use of LTEM to image domain walls injected into Permalloy wires using a holder that they had developed. One of the issues associated with *in situ* applied fields is deflection of the incident electron beam, and although the holder developed by Arita et al. is limited to an applied field of ±200 Oe, an advantage of its design is that the deflection of the electron beam by the applied