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Recent Developments in Lorentz Microscopy

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Abstract

Lorentz transmission electron microscopy (LTEM) has gradually evolved into a quantitative technique for analyzing the magnetization state of samples. This review presents latest advances of imaging techniques, including using spherical aberration correction to enhance spatial resolution, the application of Fresnel-variant 4D LTEM to improve temporal resolution, and the development of novel in situ sample holders. Discussion on vector field electron tomography (VFET), which enables three-dimensional reconstruction of magnetization information is also included. In addition, it provides a brief overview of how these advancements have been applied in recent material studies.

Keywords

LTEM, VFET, MBIR, In situ sample holder, Spherical aberration correction, AI.

1. INTRODUCTION

The advent of electron microscopy has been long established, offering a powerful capability for observing microstructures that far surpasses the limitations of optical microscopy. Lorentz transmission electron microscopy, in particular, is highly suitable for the quantitative analysis of magnetic domain structures of magnetic materials at sub-50 nm scales [1]. Simultaneously imaging both the microstructure and the magnetic domain structure of materials enables direct investigation into how often non-uniform microstructures influence magnetic behavior.

The early research on Lorentz transmission electron microscopy (LTEM) was conducted by Hale and Boersch and Raith [2,3]. High-energy electrons passing through magnetic materials (often in the form of two-dimensional thin films) are deflected by the Lorentz force generated by the magnetic field within the thin film. From a quantum mechanical perspective, the electron wave passing through the sample experiences a phase shift influenced by the magnetic vector potential associated with the thin material, which depends on the sample's magnetization. Therefore, LTEM imaging offers direct insights into the magnetization state of the sample.

The Fresnel imaging mode in LTEM leverages the deflection of high-energy electrons caused by the Lorentz force as they pass through magnetic domains in the sample. This results in bright and dark contrast at domain boundaries, providing a visualization of the magnetic domain structure. Commonly used for observing the overall magnetic domain configuration and dynamics, this mode offers qualitative insights into magnetic structures but does not provide direct quantitative magnetization data. The Foucault mode uses an objective aperture to select electrons deflected in specific directions by magnetic domains, creating contrast images that reveal detailed magnetic domain structures and interactions.

LTEM has evolved from a qualitative technique for observing magnetic domains into a quantitative analytical tool for determining the magnetization state of samples. The field has seen steady, albeit slow, improvements in spatial resolution and image quality. Despite its

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powerful capabilities in the study of magnetic materials, there are still some limitations that need to be addressed. In recent years, there have been several technological advancements related to LTEM.

Conventional high-voltage electron microscopes are not suitable for observing irradiation-sensitive magnetic materials. As a result, LTEM typically exhibits relatively high spherical aberration (from 50 to 8000 mm) and chromatic aberration (approximately 20–40 mm) [4]. Moreover, since Fresnel-mode LTEM often requires imaging at high defocus values, its spatial resolution is limited. In recent years, spherical aberration correction of the objective lens in LTEM has led to significant improvements in its spatial resolution.

The microstructural evolution of materials under specific conditions is a key research focus, as it reveals performance during processes [5]. In LTEM, applying in situ magnetic fields is challenging due to space constraints and thin samples, and external fields can deflect the electron beam, reducing imaging accuracy and resolution. Recent developments in in situ sample holders have avoided modifications to the electron microscope itself while ensuring compatibility and performance, offering significant practicality and flexibility [6].

The time resolution achieved in LTEM based on conventional Foucault or Fresnel imaging modes is approximately 40 milliseconds, allowing visualization of quasi-static magnetization behaviors, such as gradual changes under weak magnetic fields [7]. Fresnel-variant 4D LTEM is a new technique based on 4D scanning transmission electron microscopy (4D-STEM), which uses pump-probe methods with electron beams to observe the dynamic behavior of magnetic structures with high spatial and temporal resolution. In recent years, it has already seen further applications. The focus of investigation has expanded beyond simple magnetic domains to encompass more practically relevant magnetic structures with greater complexity, such as magnetic skyrmions.

VFET is an emerging advanced TEM technique based on phase analysis in TEM and electron holography (EH). When applied to LTEM, it enables the characterization of the three-dimensional magnetization of magnetic materials. Since the 2010s, VFET has been used to reconstruct all three components of the magnetic vector potential with high spatial resolution. The method for forming three-dimensional magnetic vector potential can also be based on image iterative reconstruction techniques, which accurately describe the magnetic field distribution in magnetic structures. By combining the forward model of image formation from LTEM experiments with a prior magnetic structure model, researchers can transform the tomography problem into a Maximum A Posteriori (MAP) estimation problem and use model-based iterative reconstruction (MBIR) algorithms to achieve high-precision reconstruction of the magnetic vector potential.

In recent years, artificial intelligence (AI) technology has made significant progress in the analysis of LTEM images. Through deep learning models, the accuracy of magnetic phase reconstruction from single defocused images has been greatly improved, especially demonstrating stronger robustness in noisy environments. These methods, utilizing deep generative priors and convolutional neural networks, effectively overcome the limitations of traditional phase retrieval techniques, enhancing both the precision of imaging and processing efficiency. Furthermore, machine learning models trained on simulated data enable the automatic identification and quantification of complex magnetic structures such as magnetic skyrmions, greatly advancing efficient and automated analysis of magnetic materials.

This paper aims to review recent advancements that have contributed to the development of LTEM, while also providing examples that demonstrate the latest applications of the technique.

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2. DEVELOPMENTS OF IMAGING TECHNICS

2.1. Spherical aberration correction

In LTEM, the objective lens is typically weakened or even turned off to minimize interference, allowing better observation of magnetic domains. However, this weak-lens design reduces the focusing power of the electron beam, and the deflection differences between electrons of varying energies become more pronounced compared to conventional TEM. At high accelerating voltages, the shorter electron wavelength can partially compensate for the effects of spherical aberration on image resolution—one of the motivations behind the development of ultra-high-voltage electron microscopes (up to megavolt levels) in the 1960s and 70s. However, from the particle nature of electrons, higher voltages imply higher-energy beams, which can damage irradiation-sensitive magnetic materials. Additionally, Fresnel-mode LTEM often requires imaging at high defocus values, further limiting spatial resolution to around 5–10 nm [4].

Spherical aberration correction in LTEM has been implemented specifically on the objective lens, leading to significant improvements in spatial resolution. Phatak et al. demonstrated the application of a corrector on a JEOL 2100F microscope, which is equipped with a dedicated Lorentz lens, to achieve high spatial and phase resolution images of magnetic monopole defects at the vertices of artificial spin ice—an accomplishment that was previously not possible [8]. The experimental results demonstrated remarkable effectiveness: the corrector reduced the spherical aberration coefficient from 120 mm to 0.01 mm, lowering the information limit of the microscope from 0.73 nm to 0.43 nm, while also decreasing the defocus required for phase reconstruction from 36 μm to 19 μm.

2.2. In situ sample holder design

To gain a comprehensive understanding of a material's magnetic behavior, it is crucial to image the sample in its pristine state, remanent state, under the influence of externally applied magnetic fields or currents, and as a function of temperature. These observations yield essential micromagnetic data and provide insights into features such as domain wall characteristics, the existence of nucleation and pinning centers, as well as the characteristics of magnetic phase transitions.

In reported studies, LTEM sample holders often utilize sample stages equipped with miniature electromagnets. However, existing implementations still face challenges such as complex modifications to the electron microscope, image distortion caused by external fields, and difficulties in maintaining continuous observation. In recent years, new sample holder designs have emerged, such as the double-tilt in situ magnetic field holder developed by Yang et al., which has addressed these issues to varying degrees [9].

Building on the Philips/FEI double-tilt TEM sample holder, a magnetic-field-applying double-tilt holder was developed by incorporating a "U"-shaped magnetic component at the holder tip and modifying the sample cup. This design retains the dual-axis tilt capability of the original holder while addressing the issue of lateral deflection of the electron beam in the applied magnetic field. Using the Philips CM200-FEG transmission electron microscope at 200 kV and 16,500× magnification, continuous voltages ranging from 0.2 to 1.6 V (with a step size of 0.2 V) were applied to the U-shaped magnetic component. Under these conditions, the measured beam drift ranged between 10 and 200 nm.

As shown by Arita et al., they used LTEM to image the magnetic domain walls injected into permalloy wires, which were subjected to a magnetic field through a sample holder they developed [4,10]. One issue with applying the magnetic field in situ is the deflection of the electron beam. Although the magnetic field applied by the sample holder developed by Arita et al. is limited to ±200 Oe, the advantage of their design lies in the fact that a second set of

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magnetizing coils mounted on the holder compensates for the deflection of the electron beam caused by the external applied magnetic field.

2.3. High temporal resolution imaging

4D electron microscopy has been developed to investigate structural dynamics with high temporal and spatial resolution, and it has been widely applied to a variety of specimens—including gold, graphite, silicon, iron, and carbon nanotubes—as well as a range of phenomena such as transient and oscillatory lattice distortions, morphological changes, and phase transitions. However, none of these prior studies have incorporated magnetic imaging. In study, the application of a Fresnel-mode variant of 4D Lorentz electron microscopy was reported for the first time, enabling the direct visualization of magnetization dynamics—such as domain wall nucleation, oscillation, and wave propagation—induced by pulsed heating in a soft magnetic Ni thin film [11]. Here, we achieved 4D imaging of magnetic domain walls using the defocused Fresnel method in Lorentz ultrafast electron microscopy (UEM), offering both spatial and temporal resolution in situ. The time-dependent changes in magnetization, revealed by variations in image contrast, were measured by inducing structural deformation in the specimen via pulsed optical fields, which modulate the in-plane magnetic field components. The experiment captured, in real time, the processes of domain wall (DW) nucleation, annihilation, and their repeated oscillatory reappearance—showing periodicities of 32 ns and 45 ns—in nickel (Ni) thin films deposited on two types of substrates. In particular, for Ni films on Ti/Si_3N_4 substrates and under minimal residual magnetic fields, the oscillatory behavior corresponded to a distinctive traveling wave train featuring periodic magnetization reversals. The domain walls within this wave propagated at a speed of 172 m/s and had a wavelength of 7.8 μm. This temporal resolution is exceptionally high and matches that of other advanced time-resolved imaging methods such as time-resolved magneto-optical Kerr effect (TR-MOKE) microscopy, various forms of photoemission electron microscopy (PEEM), and scanning transmission X-ray microscopy (STXM).

Following this, ultrafast studies on more complex magnetic structures have gradually emerged, such as magnetic skyrmions. These are topologically stable magnetic configurations that have broad applications in spintronics, including next-generation magnetic storage devices. However, their dynamic behavior on the nanosecond or faster timescale remains a technical challenge. A study in 2018 developed a system that combines femtosecond laser excitation with LTEM, using magnetic multilayer film systems (such as FeGe) [12]. By precisely irradiating the sample with femtosecond laser pulses, the skyrmion structures can be written or erased in a non-contact and controllable manner; LTEM is then used to capture the dynamic evolution of the magnetic structure. Under cryogenic conditions, stable skyrmion states are demonstrated, and for the first time, the real-time imaging of their creation and annihilation within the nanosecond timescale is achieved.

3. DETERMINATION OF THE THREE-DIMENSIONAL MAGNETIZATION CONFIGURATION

The retrieval of three-dimensional magnetization information can be achieved using LTEM observations. By collecting a series of phase shift images at various tilt angles—known as a tomographic tilt series—both electrostatic and magnetic vector potentials can be reconstructed in three dimensions, provided that data is acquired from multiple tilt axes that are mutually orthogonal. Typically, this involves four tilt series: two acquired while the sample is upright and tilted about the x- and y-axes, and two more obtained with the sample inverted. These orthogonal tilt datasets enable the reconstruction of two components of the vector field, with the third inferred by applying a divergence condition (e.g., for magnetic induction or vector potential, under the Coulomb gauge). In 2019, Daniel Wolf et al. demonstrated how to use

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holographic VFET to reconstruct all three components of the magnetic induction and electrostatic potential of Co/Cu nanowires with sub-10-nanometer spatial resolution [6]. Compared to traditional electron holography combined with tomography methods, VFET offers higher accuracy.

In 2022, Zhang et al. proposed a deep learning-enhanced three-dimensional magnetic reconstruction method called MagNet [13]. This method improves the reconstruction quality of traditional VFET in the presence of missing wedge regions by constructing a micro-magnetic simulation magnetic texture library and training a U-shaped convolutional neural network. The study shows that MagNet outperforms the traditional VFET method in handling the missing wedge problem, significantly improving the quality of the reconstructed magnetic induction field. When high image quality is required, there is significant noise, or the data acquisition conditions are poor, model-based iterative reconstruction (MBIR) methods also offer higher accuracy and resolution. Prabhat et al. proposed a MBIR algorithm to reconstruct the magnetic vector potential of magnetic nanoparticles [14]. By integrating a forward model based on TEM image formation with a prior model, they formulated the tomography task as a Maximum A Posteriori (MAP) estimation problem. The vector potential is then obtained by iteratively minimizing the MAP cost function, leading to improved reconstruction performance compared to VFET. More recently, Aurys Silinga et al. demonstrated the use of iterative reconstruction methods to reconstruct the three-dimensional magnetization distribution in nanowire structures from holographic vector field electron tomography measurements [15]. By associating electron phase measurements with the magnetic configuration of the nanostructure, they observed multiple magnetic domains in an L-shaped ferromagnetic cobalt nanowire.

4. ARTIFICIAL INTELLIGENCE IN LTEM OBSERVATION

Artificial intelligence (AI) technology has shown great potential in enhancing the spatial resolution and image analysis efficiency of Lorentz transmission electron microscopy (LTEM). McCray et al. (2023) proposed a deep generative prior-based single-image phase retrieval method, which combines convolutional neural networks with the physical forward model of LTEM imaging to achieve high-quality magnetic phase reconstruction from a single defocused image [16]. Compared to traditional multi-image methods based on the Transport-of-Intensity Equation (TIE), this model demonstrates higher robustness and accuracy in the presence of noise and defocus offsets, with phase reconstruction errors decreasing by over 35% on simulated data. The study also showed that this method, while maintaining structural detail resolution, effectively mitigates sample damage during imaging, providing a new tool for high-throughput dynamic magnetic structure research.

Further expanding on its applications, McCray et al. (2024) developed a machine learning model trained on simulated data to automatically identify and quantify magnetic skyrmions in LTEM images [17]. The model not only accurately detects the positions of skyrmions but also quantifies their size, boundary thickness, and ellipticity. When tested on experimental images, the model achieved a position detection accuracy of 97.6% and an error of less than 5% in radius estimation, significantly improving the efficiency and consistency of magnetic structure analysis. By processing large image datasets in batches, this method lays the technical foundation for statistical analysis of skyrmion lattices and collective behavior modeling.

These two studies showcase the forward-looking application of AI methods in LTEM image processing. They not only optimize the image reconstruction process but also advance the automation of complex magnetic structure analysis, providing technical support for future research on magnetic structures at the sub-nanometer and sub-millisecond timescales.

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5. CONCLUSION

As electron microscopy technology continues to advance, LTEM has evolved into a high-precision technique capable of quantitative magnetization analysis. This paper reviews key technological breakthroughs in terms of spatial resolution, temporal resolution, and the acquisition of three-dimensional magnetization information. Aberration correction techniques have significantly improved imaging accuracy, enabling researchers to observe finer magnetic structures. Meanwhile, the development of new in situ sample holders has enhanced experimental flexibility and reproducibility without requiring modifications to the main instrument. The emergence of Fresnel-variant 4D-LTEM has, for the first time, allowed researchers to track magnetic domain dynamics in real time with nanosecond-scale temporal resolution. Additionally, VFET based on electron holography and deep learning-enhanced reconstruction methods have greatly improved the accuracy of 3D magnetization distribution reconstructions, extending its applicability in the study of complex magnetic structures.

The introduction of artificial intelligence has further accelerated LTEM's evolution into an "analytical tool", improving both image processing efficiency and accuracy. Phase reconstruction and magnetic domain structure recognition methods based on deep neural networks not only demonstrate strong robustness in noisy environments but also enable large-scale statistical analysis of magnetic structures. Overall, the continued development of these technologies has elevated it to a new level, broadening its application prospects in spintronics, magnetic storage materials, and fundamental magnetism research, and providing strong technical support for the study of magnetic materials. Achieving even higher temporal resolution (femtosecond level), improved observation resolution (such as further aberration correction), more convenient and low-error 3D vector reconstruction methods, as well as broader material applicability and compatibility with external conditions, remain important research goals.

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