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# Ultrafast electron microscopy for probing magnetic dynamics

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The spatial features of ultrafast changes in magnetic textures carry detailed information on microscopic couplings and energy transport mechanisms. Electrons excel in imaging such picosecond or shorter processes at nanometer length scales. We review the range of physical interactions that produce ultrafast magnetic contrast with electrons, and specifically highlight the recent emergence of ultrafast Lorentz transmission electron microscopy. From the fundamental processes involved in demagnetization at extremely short timescales to skyrmion-based devices, we show that ultrafast electron imaging will be a vital tool in solving pressing problems in magnetism and magnetic materials where nanoscale inhomogeneity, microscopic field measurement, non-equilibrium behavior or dynamics are involved.

## Introduction

Magnetic materials underlie a wide range of technologies, from data storage<sup>1</sup> and power generation<sup>2</sup> to charged particle optics;<sup>3</sup> in fundamental science, magnetism results from spin or orbital correlations in materials, and is often linked to other ordering phenomena<sup>4</sup> such as superconductivity,<sup>5</sup> ferroelectricity,<sup>6</sup> and charge density waves.<sup>7</sup> Understanding magnetism is crucial for both development of new technological applications and disentangling couplings in strongly correlated materials. Both static and dynamic properties govern the functionality of magnetic materials; this functionality typically relies on a rapid switching or transport of the spin state. Though challenging, a wealth of insight is possible through the direct observation of light- or current-driven magnetization dynamics.

For example, tracking angular momentum flow<sup>8–12</sup> in ultrafast demagnetization is crucial for both understanding the process and for speeding it up; demagnetization at interfaces<sup>13</sup> may show interesting spatiotemporal dynamics.<sup>14</sup> Memory devices based on skyrmions—topologically stable, localized

quasiparticles—involve movement of nanometer-scale magnetic textures on ultrafast timescales.<sup>15–21</sup> As ever-smaller skyrmions are produced,<sup>22–30</sup> it will be increasingly necessary to image their dynamic behavior. It may be possible to better understand the atomic-scale motion of spin and charge involved in coherent ultrafast magnetism.<sup>31–33</sup> Highly nonuniform dynamics could be induced by a localized change in magnetic properties, such as a laser-driven transient modification of magnetic anisotropy.<sup>34</sup> These examples illustrate that understanding transient magnetic properties in space and time necessitates high-temporal-resolution imaging. Several probes of magnetism have been developed for this purpose.

Photons—in the visible,<sup>35,36</sup> extreme ultraviolet,<sup>37,38</sup> or x-ray<sup>39–41</sup> spectral range—have been employed to image magnetic dynamics for many years. Magnetization-dependent differences in absorption or reflectivity produce image contrast, and when the timing is varied between pulses of photons used for excitation, called the pump, and pulses used for imaging, called the probe, high temporal resolution is possible.<sup>42</sup> Ultra-short optical pulses can initiate sudden modifications on the

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magnetic order (see **Figure 1**), such as demagnetization,<sup>36</sup> domain dilation,<sup>43–45</sup> and ferromagnetic-to-paramagnetic phase transitions,<sup>46</sup> all observed using optical pump-probe techniques. These sudden changes can result in magnon generation,<sup>47</sup> all-optical magnetic switching,<sup>48</sup> and spin currents,<sup>49</sup> and can be applied toward technological applications such as terahertz emitters.<sup>50</sup>

However, ultrafast photon-based nanoscale imaging has resolution limits. For example, spatial resolution in magneto-optical Kerr effect (MOKE) microscopy is typically restricted to the micrometer scale without the aid of a near-field probe.<sup>51</sup> X-ray magnetic dichroism (XMCD), responsible for many discoveries of nanoscale magnetic dynamics,<sup>15,43,52–59</sup> can achieve a spatial resolution on the order of ten nanometers, employing imaging beamlines either at synchrotrons with X-ray pulses of tens of picoseconds or longer,<sup>60</sup> or with femtosecond resolution at free-electron lasers<sup>14</sup> and using high-harmonic sources.<sup>61</sup> In contrast, XMCD photoemission electron microscopy takes advantage of the spatial resolution of electron imaging.<sup>54,62–65</sup>

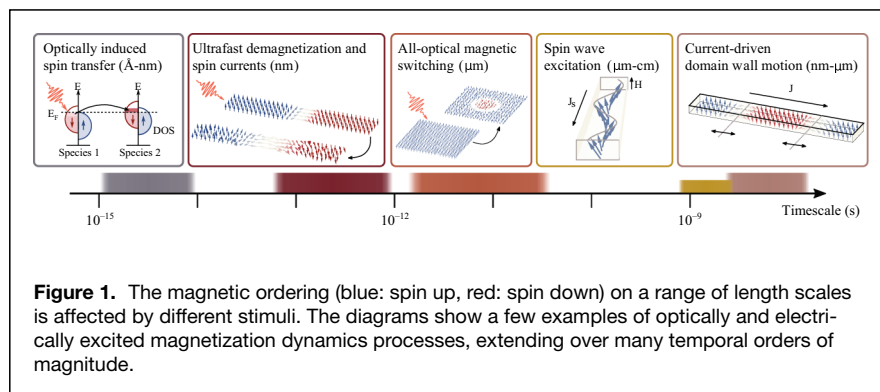
Electrons are a powerful alternative probe of magnetism on the nanoscale. Magnetic image contrast using electrons arises through both inelastic and elastic scattering. With low incident electron energy, scanning electron microscopy with polarization analysis (SEMPA)<sup>66–71</sup> probes the local surface magnetization by sampling the spin polarization of emitted secondary electrons, and spin-polarized low-energy electron microscopy (SPLEEM) offers magnetic contrast in reflection,<sup>72,73</sup> because the inelastic and elastic electron mean free paths depend on the relative orientation of electron spin and local surface magnetization. Higher incident-energy electrons can excite atomic transitions sensitive to material electron spin polarization, analogous to XMCD; this technique, called electron magnetic chiral dichroism (EMCD), employs crystalline diffraction to select outgoing electron orbital angular momentum states corresponding to different transitions.<sup>74</sup> EMCD offers atomic-resolution magnetic contrast,<sup>75–77</sup> but the high dose necessary for a good signal-to-noise ratio may restrict the usability of this technique for short electron pulses. Magnetic dichroism with electron orbital angular momentum post-selection<sup>78–82</sup> relies on similar mechanisms but has the potential

for an adequate signal-to-noise ratio even with femtosecond pulses and kHz repetition rates. Spin-polarized transmission electron microscopy<sup>83,84</sup> may prove capable of both inelastic and elastic magnetic imaging.

### Lorentz transmission electron microscopy

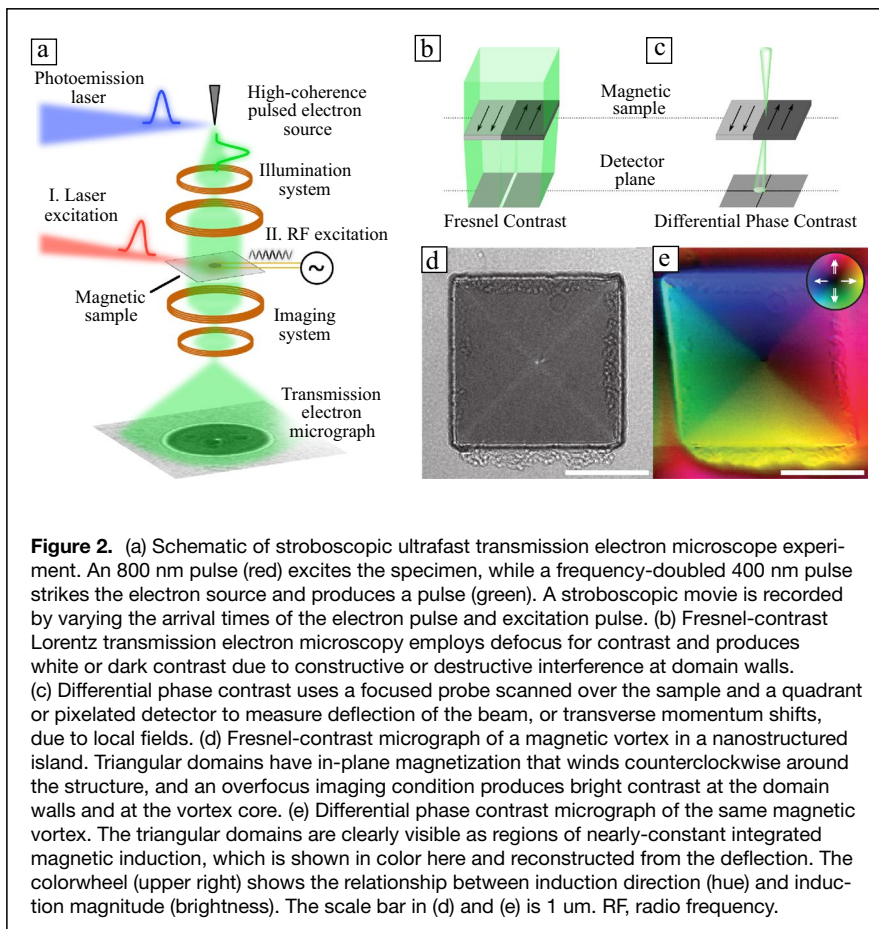
The Lorentz force, an elastic interaction, can be employed for magnetic imaging at a wide range of energies.<sup>85</sup> Lorentz electron microscopy describes modes where the immersive objective lens of a transmission electron microscope is turned off to minimize the external magnetic field applied to a magnetic specimen, and imaging contrast is based on the Lorentz force, as shown in **Figure 2**. Three contrast modes are most commonly used for magnetic imaging in transmission electron microscopy: (1) Fresnel contrast,<sup>86,87</sup> which uses image defocus to transform phase gradients introduced by the Lorentz force onto incident plane wave-like electrons into intensity in an image (Figure 2b and d); (2) differential phase contrast,<sup>88</sup> which uses a focused beam at the specimen and measures the deflection in momentum space due to the Lorentz force with a quadrant or pixelated detector (Figure 2c and e); and (3) holography,<sup>89,90</sup> which uses a biprism to interfere the plane wave passed through the specimen with an off-axis reference wave passed through vacuum. Fresnel contrast is most frequently used for its simplicity, whereas differential phase contrast is more quantitative and increasingly applied as fast pixelated detectors become more common. Holography is the most quantitative of the three contrast modes, but also requires a high transverse coherence,<sup>91</sup> so it is challenging but possible with pulsed electrons.<sup>92,93</sup>

With a pulsed electron source<sup>94</sup> through either photoemission<sup>92,95–100</sup> or a beam chopper,<sup>100–105</sup> and synchronized current, field, or laser excitation of the specimen, sub-picosecond temporal resolution has become possible in stroboscopic transmission electron microscopy.<sup>106–108</sup> This approach has many applications, but some of the first experiments imaged magnetic dynamics with Lorentz microscopy.<sup>101,109</sup> Moreover, using continuous-beam imaging and *in situ* short-pulsed excitation of a sample, insights into the ultrafast generation and stabilization of quenched magnetic states are possible.<sup>110–114</sup>



### Imaging optical pulse-induced magnetic textures

Standard Lorentz microscopy is an excellent tool to investigate magnetic textures induced by femtosecond optical excitation. Imaging the response of magnetic textures to external stimuli has a long tradition. Commonly, variations in temperature<sup>115,116</sup> and temperature gradients,<sup>117</sup> external



magnetic fields<sup>118–120</sup> or electric currents<sup>121,122</sup> are applied on timescales for which the spin system remains close to local thermal equilibrium. In contrast, *in situ* femtosecond optical excitation results in a far-from-equilibrium spin state and enables the creation of novel metastable magnetic textures, which can be characterized by continuous-beam Lorentz microscopy, as shown in **Figure 3**.

As a first example, Eggebrecht et al. recently demonstrated the optically induced generation of a dense vortex-antivortex network in a polycrystalline iron thin film deposited on a silicon nitride membrane.<sup>112</sup> Before excitation, the system shows a magnetic ripple texture, induced by the local fluctuations of the magnetic anisotropy direction (see **Figure 3a**, upper panel). Applying a single intense ultrashort optical pulse raises the temperature of the iron thin film above the Curie temperature. In the center of the optical focal spot, the iron thin film is transiently demagnetized, whereas the underlying silicon nitride membrane remains cold. On a 100 ps timescale, the iron film thermally equilibrates with the membrane and a local ferromagnetic ordering is re-established, but the fast cooling rate (larger than  $10^{12}$  K/s) prevents the system from reaching a homogeneous ordering. Instead, nanoscale ferromagnetic domains are formed, which, at their mutual boundaries, are linked by

topological vortex and anti-vortex defects. The “frozen” magnetic state, as shown in **Figure 3a** (lower panel), is stable at room temperature but can be reset to the initial ripple texture by either applying an external magnetic field or low-fluence optical pulses.

Similar results were obtained for optically excited permalloy ( $\text{Ni}_{80}\text{Fe}_{20}$  high permeability magnetic alloy) microstructures, in which vortex switching and small-scale vortex-antivortex networks can be induced.<sup>113</sup> In this case, the probability of the final magnetic configuration was shown to depend on the strength of the optical excitation and the shape and size of the magnetic structure.

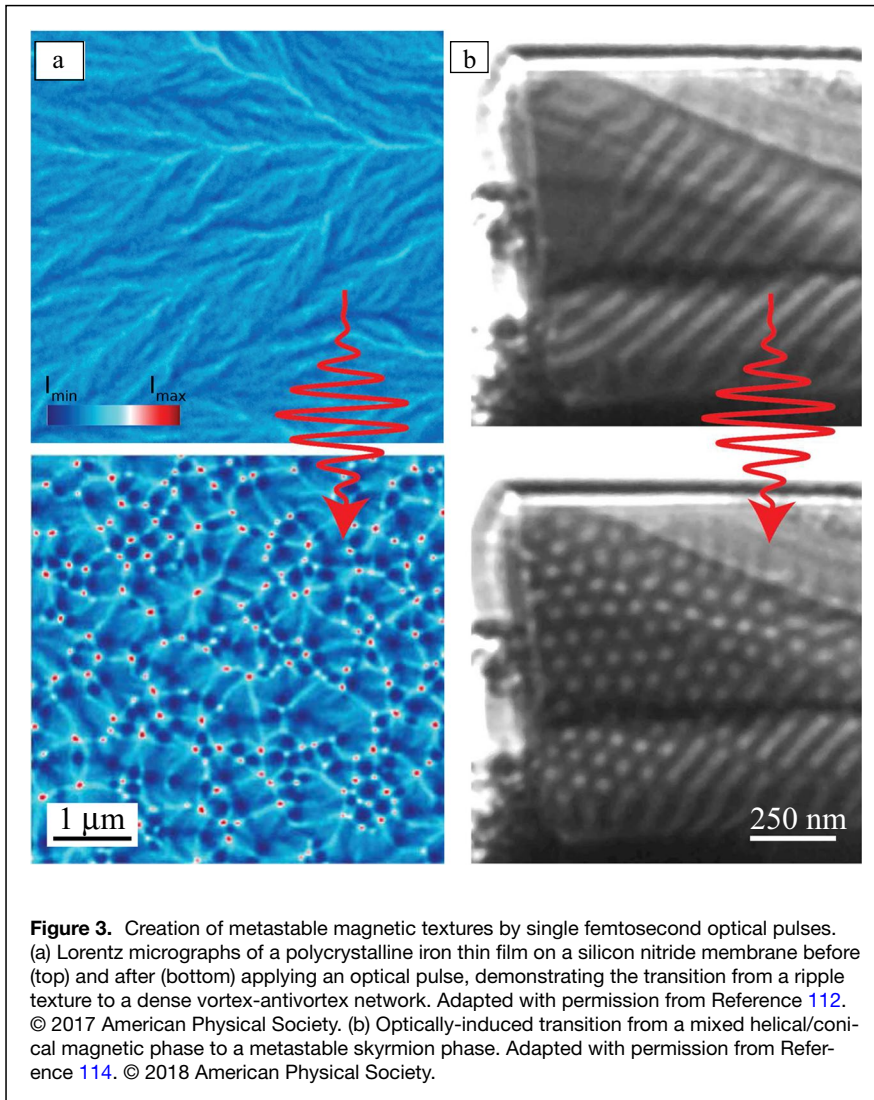
While out-of-plane magnetized samples are generally less suited for Lorentz microscopy, local topological defects such as domain walls or skyrmions can still provide good image contrast with nanoscale resolution.<sup>119</sup> In a recent study,<sup>114</sup> the optical generation of a metastable skyrmion texture in FeGe thin films was probed by *in situ* Lorentz microscopy. In equilibrium, FeGe shows a series of different mag-

netic phases depending on sample temperature and applied magnetic field. A Lorentz micrograph of a mixed phase with helical and conical textures stable at low temperatures and intermediate external magnetic fields is shown in **Figure 3b** (upper panel). Upon laser excitation starting from this phase, the sample temperature rapidly increases, reaching the stability regime of an adjacent skyrmion phase. Fast quenching results in a metastable skyrmion lattice, as shown in **Figure 3b** (lower panel). Such all-optical writing approaches could be a fundamental ingredient for novel skyrmion-based information storage and transport applications. The dynamical evolution upon femtosecond optical excitation in a related sample system was further elucidated using time-resolved x-ray scattering, demonstrating ultrafast switching and the existence of a transient topological fluctuation state.<sup>123</sup>

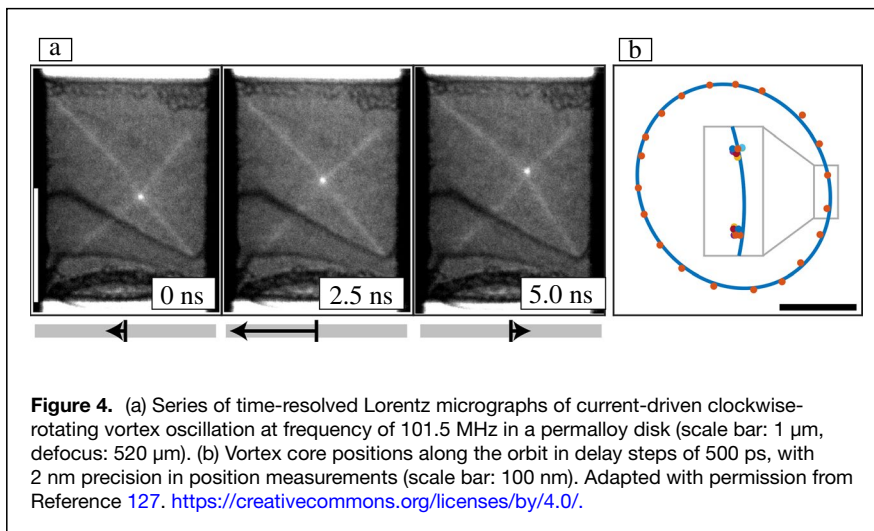
### Ultrafast Lorentz transmission electron microscopy

With stroboscopic electron illumination, ultrafast Lorentz transmission electron microscopy can be applied to image transient changes in magnetic textures and fast dynamics. Experimental challenges in ultrafast Lorentz microscopy include the limited average beam current and the requirement





**Figure 3.** Creation of metastable magnetic textures by single femtosecond optical pulses. (a) Lorentz micrographs of a polycrystalline iron thin film on a silicon nitride membrane before (top) and after (bottom) applying an optical pulse, demonstrating the transition from a ripple texture to a dense vortex-antivortex network. Adapted with permission from Reference 112. © 2017 American Physical Society. (b) Optically-induced transition from a mixed helical/conical magnetic phase to a metastable skyrmion phase. Adapted with permission from Reference 114. © 2018 American Physical Society.



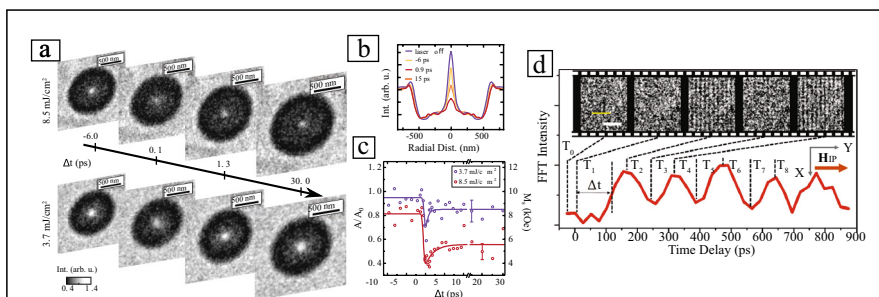
**Figure 4.** (a) Series of time-resolved Lorentz micrographs of current-driven clockwise-rotating vortex oscillation at frequency of 101.5 MHz in a permalloy disk (scale bar: 1  $\mu\text{m}$ , defocus: 520  $\mu\text{m}$ ). (b) Vortex core positions along the orbit in delay steps of 500 ps, with 2 nm precision in position measurements (scale bar: 100 nm). Adapted with permission from Reference 127. <https://creativecommons.org/licenses/by/4.0/>.

for spatiotemporal reversibility of the triggered magnetic dynamics, often necessitating a tailored sample design.

Ultrafast Lorentz microscopy has been employed to study several types of dynamics, including domain wall motion under an oscillating magnetic field<sup>109</sup> or laser-induced thermal gradients,<sup>110,124</sup> laser-induced demagnetization,<sup>100,124–126</sup> and current-driven vortex oscillations.<sup>127</sup>

In two recent examples, Möller et al. investigated the response of a magnetic vortex to an applied high-frequency current.<sup>127,128</sup> The interplay between the current-induced force on the vortex, consisting both of Oersted-field and spin-transfer torque contributions, and the confinement potential within a magnetic nanostructure results in resonant gyration dynamics of the vortex. Driving the vortex gyration by applying 101.5 MHz radio-frequency currents across a 2.1  $\mu\text{m}$  permalloy square, Möller et al. were able to track the vortex position with a 2 nm precision,<sup>127</sup> as shown in **Figure 4**, and observed the frequency dependence of the response in real space.<sup>128</sup> Prior work had mapped the time-averaged motion using continuous-beam Lorentz microscopy.<sup>121</sup> The temporal resolution available in ultrafast Lorentz microscopy now captures the precise phase of the gyration, and can measure the dissipation rate and transient ring-down behavior of the oscillation after the driving current is rapidly switched off.

As a further example, several groups have recently investigated ultrafast demagnetization, shown in **Figure 5**. Ultrafast demagnetization was first observed through optical Kerr spectroscopy in 1996,<sup>36</sup> and its underlying microscopic physical mechanisms are still being debated.<sup>31,129–133</sup> Ultrafast Lorentz microscopy has the potential to



**Figure 5.** Snapshots of laser-induced magnetization dynamics captured by Lorentz microscopy. (a–c) Ultrafast demagnetization of a permalloy disc mapped with 100 nm and 700 fs resolution.<sup>125</sup> (a) Lorentz micrographs recorded for several delay times and pump-laser fluences. (b) Azimuthally averaged intensity profiles, evidencing the delay-dependent variations in the intensity of the central bright spot. (c) Transient magnetization retrieved from image contrast. (a–c) Adapted with permission from Reference 125. <https://creativecommons.org/licenses/by/4.0/>. (d) Magnetization dynamics stimulated by a transient optical grating.<sup>126</sup> Time-resolved micrographs reveal a time-dependent, periodic magnetic contrast, and the evolution of the intensity of the spatial Fourier transform of the image. Reproduced with permission from Reference 126. © 2021 Royal Society of Chemistry. FFT, fast Fourier transform.

provide further experimental insights due to its very high spatial resolution.

Several recent experiments studied demagnetization behavior with ultrafast Lorentz microscopy with picosecond-scale<sup>100,124–126</sup> temporal resolution. Rubiano da Silva et al. recently mapped the partial demagnetization of vortices in nanoscale disks of permalloy with 100 nm spatial resolution and 700 fs temporal resolution,<sup>125</sup> shown in Figure 5a–c. Cao et al. reported on demagnetization of permalloy with a grating pattern caused by interference in the exciting laser, resulting in a coherently precessing magnetic grating,<sup>126</sup> shown in Figure 5d. Zhang et al. observed a peculiar sequence of sub-picosecond demagnetization, recovery over 3 ps, and decay to a paramagnetic state over 12 ps of the spiral spin texture in Mn-Ni-Ga.<sup>100</sup>

Despite this methodical progress, ultrafast Lorentz microscopy still needs to overcome a number of experimental challenges. Since Fresnel contrast is linked to the applied imaging defocus, reaching a sufficient signal-to-noise ratio often requires rather large defoci in the 100 μm range or above, associated with a significant loss in spatial resolution. Therefore, further progress in high-coherence, high-brightness ultrafast electron sources is necessary to reach a spatial resolution on the order of few nanometers.

## Conclusion and outlook

As the experiments presented here show, behavior of magnetic materials at picosecond or shorter timescales may strongly deviate from equilibrium. This far-from-equilibrium behavior is observable with Lorentz electron microscopy both via *in situ* excitation of ultrafast processes that lead to long-lived changes in magnetic textures, and with stroboscopic excitation and imaging of picosecond or faster dynamics. The combination

of a nanometer spatial and femto-second temporal resolution makes ultrafast electron microscopy an excellent tool for imaging magnetic dynamics. Electron microscopes can measure crystal structure, charge states, electric fields and bonding down to the atomic scale, and strain, temperature, phononic and plasmonic band structures on the nanometer scale. The capability to correlate these properties with sub-picosecond movies of magnetic dynamics will enable identification of nanoscale inhomogeneities invisible in bulk measurements, quantification of local fields used to excite dynamics, and direct measurement of coupled behavior like magnetostriction.

Specifically, this tool is well-suited to help answer a number of open questions in magnetism

related to microscopic magnetic energy landscapes,<sup>127,128</sup> angular momentum transfer processes,<sup>134</sup> ultrafast demagnetization at interfaces, skyrmion dynamics, and coherent ultrafast magnetism. The growth of other ultrafast electron techniques, including time-resolved SEMP<sup>67–71</sup> and potentially SPLEEM, opens the door to complementary surface-sensitive imaging of dynamics. New manipulation techniques, such as irradiation by a high-charge pulse of electrons,<sup>135</sup> may grow in use. There are numerous opportunities for ultrafast electron imaging to unravel key problems in magnetism.

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## Conflict of interest

On behalf of all authors, the corresponding author states that there is no conflict of interest.

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