Development of Aberration Corrected Differential Phase Contrast (DPC) STEM

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In this article we demonstrate that aberration correction for STEM probes has been achieved for field-free Lorentz STEM imaging of magnetic samples, and that, an order of magnitude improvement of spatial resolution has been obtained. We believe, that our achieved <1 nm spatial resolution is currently the best in the world for direct imaging of magnetic structure by electron microscopy.

Introduction
Correctors for spherical aberration (Cs) of electron lenses have resulted in a step-change in performance for both TEM and STEM instruments, making atomic scale imaging and analysis of materials routinely possible. Working in collaboration with JEOL and partners we show that aberration correction and other technologies have enabled an order of magnitude improvement in the capability to image magnetic behaviour in thin nano-scale structures important for current and future information technologies.

The imaging of magnetic structure in the electron microscope has a long history that extends back to the 1950’s. The group of imaging techniques used to generate magnetic contrast are collectively known as “Lorentz microscopy” [1] as they can be understood in terms of the classical Lorentz force ($\mathbf{E} = -e(\mathbf{v} \times \mathbf{B})$) experienced by the beam electrons traversing the specimen. Thin magnetic samples also exhibit a quantum interaction with the beam, via the Aharonov-Bohm effect, whereby the phase of the passing electron waves are altered. Thus, for imaging magnetic samples the Lorentz techniques are a branch of phase contrast microscopy. In TEM mode, Fresnel and Foucault techniques have been extensively used. Both are effective in generating images showing strong magnetic contrast but are limited in spatial resolution or linearity. TEM Holographic techniques have been quite successful and shown to be capable of higher resolution imaging of magnetic induction [2], however, their basis is off-line image reconstruction and they cannot be applied to all sample geometries.

At the University of Glasgow, we have developed the STEM based Lorentz imaging mode of Differential Phase Contrast (DPC) over the last 30 years. In this article we demonstrate that in collaboration with JEOL, CEOS GmbH, Gatan Inc., Deben Ltd and University of Warwick, that aberration corrected DPC STEM has been achieved enabling the study of magnetic structure with world-leading spatial resolution better than 1 nanometre. Furthermore, in contrast to holographic techniques, these images are available in real time at near video frame-rates.

Experimental
On a JEOL JEM-ARM200FCS TEM/STEM equipped with cold field emission gun (C-FEG), a CEOS Cs STEM probe corrector and HR pole-piece, several major developments have been made to successfully realise DPC mode imaging. After describing the general concept of the DPC mode we will deal with each of the required developments in turn.

Figure 1 depicts the setup required for DPC mode imaging, where the focused electron probe is raster-scanned across the specimen with the scattered transmitted cone of electrons being detected in the far-field by a segmented STEM detector. If the specimen is magnetic and contains regions with components of the magnetic induction $\mathbf{B}$ oriented in the specimen plane, then it can be shown that the beam is deflected through an angle:

$$\beta = \frac{e_i}{\hbar} \int \mathbf{B} \times \mathbf{n} \, dz$$

where $e$ is the charge on the electron, $\lambda$ its wavelength, $\mathbf{B}$ the magnetic induction in the specimen and $\mathbf{n}$ the unit vector along the electron trajectory. The classical Lorentz deflection induced by a typical magnetic sample is relatively weak. The deflection angle, $\beta$, is in the range 1-100 micro-radians and is very much smaller than typical diffraction scattering angles which are generally >3 milli-radians. The segmented detector is used to detect such Lorentz deflection
of the beam by measuring difference signals from opposite quadrants. An alternative interpretation of the interaction of a thin magnetic sample on the beam is that the electron beam’s wavefunction after passing through the region containing magnetic induction becomes phase-shifted due to the quantum mechanical Aharonov-Bohm effect [3]. Thinking in these terms, by the action of taking difference signals, the gradient of the phase-change due to the sample is measured and hence the technique produces images showing differential phase contrast.

The process of combining the signals from the detector segments to produce live DPC STEM images is described in more detail later.

**STEM probe formation for magnetic imaging.**

In standard STEM mode, the normally excited objective lens (OL), which produces focused STEM probes with semi-convergence angles, \( \alpha \approx 3-30 \) mrad and has enabled imaging of Si-dumb-bells with information at 0.67 Å. However, the normally excited OL also subjects the sample to a magnetic field of strength \( \sim 2 \) Tesla. A field of such strength would completely saturate the vast majority of magnetic samples, obliterating any magnetic domain structure of interest. Thus, DPC STEM mode imaging must be performed with the OL completely de-excited and the sample residing in field-free or near to field-free conditions. This is easily achieved by switching the microscope into “LOW MAG” mode whereby, the OL goes off and STEM probe formation is controlled by a combination of the variable condenser (CL3) and condenser mini (CM) lenses. In this scenario, and in the absence of an aberration corrector, the diameter of the STEM probe would be dictated by the spherical aberration coefficient of the CM. In collaboration with us, JEOL and CEOS have developed a special optical configuration for the aberration corrector that compensates for the Cs of the CM and have enabled a magnification range that extends up to 2.0 million times. **Figure 2** shows an image for the resulting Ronchigram in this mode that exhibits a flat, aberration corrected, region that extends out to 3.2 milli-radian semi-convergence angle. Using a 70 μm condenser aperture, corresponding to the full diameter of the flat region, STEM imaging of a test sample, Au nano-particles, shown in **Fig. 3(a)**, demonstrated that particles with sizes of the order of 1 nm and smaller could be resolved. In fact, **Fig. 3(b)** demonstrates

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**Fig. 1** Diagram illustrating the concept of DPC STEM imaging. Lorentz deflection of the focused electron probe, through an angle \( \beta_c \), by domains in a thin magnetic sample is detected using a segmented detector.
that the smallest particle that could be resolved had a width of the order of 0.7 nm. Taking the Fast Fourier Transform of Fig. 3(a) (inset of Fig. 3(c)) and forming a radially averaged line profile showed that Fig. 3(a) contains information content up to a maximum spatial frequency of 1.8 nm⁻¹. All of these observations are in agreement with calculations made by CEOS GmbH who expected that the C₃ coefficient of the CM lens should be reduced to the order of several microns and should result in a FWHM probe diameter of 0.8-1.0 nm with spatial resolution defined as being half this value, 0.4-0.5 nm.

While it would be desirable to always operate with the highest spatial resolution, inevitably a trade-off has to be made. Recalling that the Lorentz deflection angle, β, can be as small as a few micro-radians, around 1000× smaller than the optimum probe semi-convergence angle α. Higher sensitivity to small Lorentz deflections can be obtained by reducing α at the expense of spatial resolution. This can easily be done by changing to the smallest 10 µm condenser aperture which reduces α to 450 micro-radians but means that the spatial resolution becomes limited by diffraction. Lorentz sensitivity can be increased by...
a further two times, a further reduced to 215 micro-radians, by the combined adjustment of the CL3 lens and the CEOS corrector’s adapter lens element (ADL). Under these conditions spatial resolution has been measured to be in the range 3-5 nm.

**Nulling and application of in-situ magnetic fields to the specimen**

In "LOW MAG" mode, although the OL is completely de-excited, the sample still resides in a modest magnetic field, ~150 Oe directed perpendicular to its plane. This is the result of remanence from the ferromagnetic pole-pieces. For many thin film in-plane magnetised samples the out-of-plane oriented remanent field has little influence on the static magnetic structure. Its strength is generally very much weaker than the strength of the in-plane magnetic anisotropy. In-situ studies of magnetic reversal behaviour can be accomplished by using either the remanent field, or a stronger field applied by partially exciting the OL. Tilting the sample (generally up to +/- 30 degrees is possible) can then be used to nucleate and grow magnetic domains eventually leading to reversal and saturation of the film. For ultra-soft magnetic samples, where the coercivity is very much less than the remanent field strength, it is desirable to be able to reduce to near zero the strength of the remnant field. Utilising a system developed at University of Warwick we can measure the strength of the remanent field at the sample plane using a Hall-probe TEM rod and apply a reverse current through the OL to null it. In this way, very low field strengths, < 1 Oe can be achieved in the sample region.

**Segmented DPC detector and video chain**

Development of the segmented detector and DPC image acquisition system has required an extensive collaboration involving ourselves, JEOL, Gatan Inc., Deben Ltd and Andrew Armit Designs.

The geometry of the segmented detector employed is depicted in Fig. 4(a). It consists of eight segments arranged into an inner solid quadrant (INT0 to INT3) and an outer annular quadrant (EXT0 to EXT3). DPC STEM imaging, detecting the displacement of the transmitted electron disc, is most simply realised by using a camera length that projects the transmitted electron disc onto only the inner quadrants. However, in previous work [4], we have shown that for polycrystalline magnetic thin films, strong and unwanted electrostatic phase fluctuations arise due to diffraction from the nano-scale crystallites with varying orientations. By utilizing a cameral length that projects the transmitted electron disc across the outer annular (as well as the inner) quadrants, the higher spatial frequency electrostatic fluctuations can be "filtered" from the lower spatial frequency magnetic domain and domain wall features.

Conversion of charge signals from the detector segments into video-level voltage signals has been achieved through the development of the SuperFast 8-channel, 2 MHz bandwidth amplifier by Deben.

![Schematic of the DPC STEM detector and video signal digitisation](image)

**Fig. 4** (a) Schematic of the DPC STEM detector and video signal digitisation, (b) screenshot of the DPC control palette in Digital Micrograph.
& Andrew Armit designs. The SuperFast amplifier is controlled by software and has a wide range of settings enabling selection, for each channel, of input resistance/capacitance for noise reduction/bandwidth selection and gain. "On the fly" arithmetic mixing of channels, is possible and can be used to view live difference signals between segments. However, we prefer to perform such image arithmetic on acquired digital images and utilize the SuperFast amplifier to pass the segment signals unaltered.

Commonly on an advanced STEM instrument such as the JEM-ARM200F, combined image acquisition and point-wise analysis (via Electron Energy Loss Spectroscopy (EELS) or X-ray Energy Dispersive Spectrometry (EDS)) are controlled by Gatan's Model 788 Digiscan II system through Digital Micrograph software. On our JEM-ARM200FCS, the DPC detector adds 8 segment signals to the already lengthy list of signals to be acquired from the common STEM detectors (JEOL ADF1, ADF2, BF, Gatan Model 806 HAADF, Model 807 BF/ADF) and current measurement from the CFEG. Thus, in all, a total of 13 signals were required for acquisition, although not all would be used at any one time. Gatan developed a solution for this by implementing hardware and software that allowed 4 Digiscan II boxes to be operated in parallel. This was achieved in such a way that for recent releases of Digital Micrograph software (from GMS version 2.3.X), this capability is now part of the standard software-base. Live DPC imaging, enabling magnetic contrast to be visualised, has been achieved through a control palette, Fig. 4(b), created in the Digital Micrograph scripting language by the author. By clicking "Start/Stop" or "Grab Frame" buttons, calls are issued to the Digiscan II boxes that start/stop the imaging process. The individual segment images are visible but magnetic contrast can only be seen by displaying live difference images between opposite segments. Two orthogonal direction components are necessary to reconstruct magnetic orientations and these are achieved by viewing the image pairs “INT0 – INT2”, “INT1 – INT3” if using the inner quadrant (“EXT0 – EXT2” & “EXT1 – EXT3” if using the outer quadrant). Based on these image pairs live colour images showing magnetic orientations are also presented. Quantitative determination of Lorentz deflection, and hence, BS×t, the product of the magnetic induction times the thickness of the specimen, is possible by relatively straightforward post-processing of the recorded DPC images. It is the intention to include a real-time capability so that DPC images can be calibrated in terms of quantitative Lorentz deflection.

Investigation of magnetic samples

In this section we present results obtained from applying the DPC system to investigate the properties and behaviour of magnetic specimens in some of our current research.

Iron nanostructures

The fabrication of nano-scale magnetic structures is a lengthy procedure, most commonly achieved by multi-step lithographic techniques in which the shapes to be created are written into a sensitive resist followed by chemical development, metallisation and “lift-off” steps. Alternatively, rapid direct-writing of magnetic nano-structures can be achieved in focused ion beam and scanning electron microscope (SEM) systems where a needle based system is used to inject an organometallic precursor gas into the region of the beam-scanning [5,6]. Using SEM, such electron beam induced deposition (EBID) has been employed to create rectangular iron elements, Fig. 5, and pillars with diameter around 50 nanometres, Fig. 6.

Fig. 5(a) and (b) show greyscale DPC images (obtained using Spot L1 and a 10 micron condenser aperture) that highlight the ground state arrangement of magnetisation in the approximately 600 nm × 400 nm × 40 nm thick rectangular elements which were fabricated on Si3N4 support membranes. As described earlier, Fig. 5 (a) & (b), are produced by subtracting the video signals from opposing segments on the DPC detector and produce a pair of images with orthogonal directions of sensitivity. Inside the elements strong black and white contrast can be seen that corresponds to the magnetic domain structure, while outside of the elements “noisy” phase contrast from the thin carbon coating used for charge dissipation is observed. The arrangement of the magnetic domain orientations in the element is most easily understood by forming the colour map in Fig. 5(c). From this Fig. it can be seen that the element has formed a flux-closing multi-domain Landau type pattern in which the magnetisation tends to be oriented parallel to the element edges and circulates around two vortices within the elements interior. The spatial extent of each of the vortices is dictated by the magnetic properties of the material, specifically the exchange stiffness and the saturation magnetisation. From polycrystalline alloys involving Co, Ni and Fe, vortex widths have been measured to range from 7-15 nanometres wide [7]. In Fig. 5(d), we have utilised the high spatial resolution afforded by aberration correction of the CM lens to measure a DPC intensity profile from the position of the red line in Fig. 5b. Fig. 5d, shows that for the EBID Fe element (with approximate chemical composition 60% iron, 40% carbon) that the measured width of the vortex core is 13.6 nm.

Narrow pillar-like magnetic structures can be formed by allowing the electron beam in the SEM to dwell on a single location. Such pillars have proved highly effective as magnetically switchable trapping sites when directly written on top of out-of-plane magnetised nanostrips (see reference [9] for a fuller explanation). For these pillars, due to their small diameter, ~50 nm, DPC imaging has been used to measure the magnetic field strength needed to switch the pillars magnetisation direction. Figs 6(a) and (b) shows colour DPC maps that depict the nanopillars grown on the edge of a grid support. The colour contrast inside the pillars is not simply interpretable in terms of magnetic structure as it is dominated by electrostatic phase gradients from the changing thickness associated with their circular cross-section. In the free-space region immediately surrounding the tip of the pillar, located inside the dashed ellipse, colour contrast relating to the pillar’s de-magnetising
fields can be observed. Starting in Fig. 6(a), moving in a clockwise direction around the tip of the pillar, the colour contrast changes from blue to red to yellow. Referring to the colour-wheel inset, this indicates the magnetic fields emanating from the tip are divergent and thus it can be inferred that the pillar is magnetised in an upwards direction. A field of strength 1000 Oe was then applied to the pillar in-situ by partially exciting the OL lens and tilting the specimen to near 30 degrees. After de-excitation the OL and returning the specimen to its untitled state, the colour map in Fig. 6(b) was obtained. Again, the direction of the magnetisation in the pillar was inferred by examining the colour contrast associated with de-magnetisation field from the pillar tip. In Fig. 6(b) it can be seen that the colour contrast has altered and changes from yellow to green to blue when moving in a clockwise sense around the tip. This indicates that the de-magnetisation fields are now convergent at the tip and infers that the magnetisation direction has been switched to a downwards orientation by the applied field.

**Multilayered ferromagnetic sample**

The high spatial resolution afforded by aberration correction for the field-free mode makes it possible to investigate the behaviour of multi-layered magnetic films in a cross-section type geometry. For a repeated NiFe ferromagnet / FeMn antiferromagnet multilayer sample we have performed DPC imaging in order to understand aspects of its magnetic reversal behaviour. Fig. 7 shows greyscale DPC images obtained from a

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**Fig. 5** DPC imaging of EBID Fe nano-element. (a) & (b) horizontal and vertical DPC component images, (c) colour map showing magnetic induction directions, (d) Line-trace measurement of vortex core diameter for line indicated in (b).
FIB cross-section of the multilayer (~ 50 nm thick) with the structure NiFe/(FeMn/NiFe)\times10 grown on an oxidised Si substrate with a capping layer of 5 nm of Ta. The NiFe layers had a thickness of 16.5 nm and the FeMn layers 12.8 nm.

Initially the sample was immersed in a large field (around 1000 Oe) to have all the layers in parallel alignment. The DPC image component showing the magnetic induction parallel to the interfaces is shown in Fig. 7 (a) where the NiFe layers appear as bright stripes in the image and the FeMn layers are gray indicating no net induction component in these regions. The variation in contrast within the stripes is a consequence of the granular structure of the film and this gives rise to a diffraction contribution to the phase contrast image. A linetrace from the area indicated by the rectangle in Fig. 7 (a) is shown, this averages the signal over a 74 nm width to reduce the effects of the diffraction contrast from the grain structure. The profile shows the magnetised layer variation very clearly where each magnetic layer is around 16-17 nm wide (i.e. deposited thickness of the film) and the AF layer is 13 nm wide. By tilting the sample in the objective lens field the magnetic state could be altered with individual layers switching and an example of the state part way through the reversal process is shown in Fig. 7 (b) where seven of the eleven magnetic layers have switched their direction of magnetisation, one partially. This can also be seen by comparing the averaged linetraces for Figs 7 (a) & (b) for the two different states. The linetraces show the induction in the ferromagnetic layer very clearly and indeed the interface between the ferromagnetic and antiferromagnetic layers shows the transition which is on the order of 1-2 nm.

**Reduction of magnetization in nano-scale regions by ion irradiation**

Obtaining quantitative measurements regarding the strength of magnetic induction from DPC images is usually straightforward. We have been investigating the use of FIB based ion-irradiation to control the strength of magnetisation in \( \text{Cr}(3 \text{ nm})/\text{Ni}_{80}\text{Fe}_{20}(10 \text{ nm})/\text{Cr}(5 \text{ nm}) \) films deposited on \( \text{Si}_3\text{N}_4 \) electron transparent membranes. The key aim for us was to create and characterise narrow irradiated line defects that could act as trapping sites for domain walls in magnetic nanowires [9,10]. Fig. 8 shows quantitative DPC imaging of a line irradiated at a dose of \( 8 \times 10^5 \) ions cm\(^{-2} \). The components of magnetic induction were mapped parallel, Fig. 8(a), and orthogonal, Fig. 8(b), to the irradiated line. In Fig. 8(a) the irradiated line is observed as a lower intensity feature while it is invisible in Fig. 8(b), the latter is consistent with the component of magnetic induction being continuous across an interface as proved from Maxwell’s equations, even though the magnetisation is discontinuous. The intensity profile from the region indicated in Fig. 8(a) is plotted in Fig. 8(c) where the vertical axis displays quantitative measurement of the Lorentz deflection of the beam. Quantitative determination of the deflection is achieved by dividing the difference images by the “SUM” image (i.e. summing the images from all segments). Since the diameter of the transmitted electron disc incident on the segmented detector relates to the beam convergence semi-angle, \( \alpha \), which is known, then the Lorentz deflection, \( \beta \), can be easily be recovered. In Fig. 8(c), the quantitative profile shows a measured Lorentz deflection angle of \( \beta=4.3 \) mrad for the unirradiated region. This is as expected. For a 10 nm thick \( \text{Ni}_{80}\text{Fe}_{20} \) film with \( B_s = 1 \) Tesla, then the total beam deflection should be \( \beta=6.5 \) mrad. With the DPC

![Fig. 6 (a) & (b) DPC colour maps showing magnetic fields emanating from the tip region (indicated by dashed ellipses) of 50 nm diameter EBID Fe-nanopillars.](image-url)
sensitivity components being oriented at 45 degrees to the direction of the mean magnetisation in the film, then the measured β is reduced by sin(45) = 0.7, yielding β = 4.4 μrad. The irradiation dose of 8×10¹⁵ ions cm⁻² has resulted in a line of width 50 nm, with a measured deflection of 1.3 μrad, corresponding to a reduction of 70% reduction in Mₘ.

Summary

In summary, our collaborative development of an aberration corrected STEM Differential Phase Contrast system has demonstrated quantitative imaging of magnetic structure with spatial resolution in the 1-6 nanometre range. As far as we are aware, apart from ultra-high vacuum based scanning tunnelling microscopy (UHV-STM) of atomic surfaces, we know of no other techniques that currently enables magnetic imaging at this length scale. Excitingly, we envisage that further improvements can be made. All results presented here were obtained with a beam energy of 200 keV. Recently we have begun performing DPC at 80 keV which should lead to around a 4× improvement in magnetic sensitivity and which will be essential for investigating new phenomena in ultrathin, 1-5 atom thick, magnetic layers. Furthermore, DPC imaging is not just limited to magnetic samples. Materials and films containing intrinsic electric fields and polarisation exert similar influence on the electron beam. However, the exciting prospect here is that the OL need not be de-excited for such work. Operating in the more usual aberration corrected OL ON mode, atomic resolution DPC investigations are enabled. We envisage that such powerful imaging may benefit the understanding of charge distributions in bonding, across interfaces and at surfaces and lead to the discovery of new aspects of materials physics.

Acknowledgments

The developments reported and procurement of the microscope were enabled by joint funding from the University of Glasgow and the Scottish Funding Council (through the Scottish Universities Physics Alliance (SUPA)).

The authors would like to take the opportunity to express their gratitude to all staff members of JEOL, Gatan Inc., CEOS GmbH, Deben Ltd., University of Warwick and to Andrew Armit for their invaluable efforts in this collaboration. We are grateful to the following collaborators for samples: EBID Fe nanostructures from the group of H. J. M. Swagten at TU Eindhoven, Netherlands; multilayer ferromagnetic samples from the group of R. M. Bowman at Queens...
University, Belfast, UK; tri-layered \(\text{Cr/Ni}_{80}\text{Fe}_{20}/\text{Cr}\) samples from the group of C. H. Marrows, University of Leeds, UK.

We also acknowledge funding support from the UK EPSRC, grant number EP/I013520/1, which funded one of the authors (M-J.B.) and enabled much of the development work.

References


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Fig. 8 DPC analysis of FIB irradiated lines in a \(\text{Cr/Ni}_{80}\text{Fe}_{20}/\text{Cr}\) multi-layer. (a) & (b) Component images showing the irradiated line, (c) a quantitative plot of beam showing the variation of beam deflection and width of the irradiated line.