## **Local Phase Curvature Measurement in STEM With a Pixelated Detector**

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The introduction of fast pixelated detectors to scanning transmission electron microscopy (STEM) has opened new and exciting possibilities for material's characterisation. Pixelated detectors have already been used to enhance imaging with ptychographic techniques, e.g. [1], enhance contrast for magnetic materials in differential phase contrast (DPC) [2], and measure atomic electric fields [3]. In this paper we present a new method of treating phase effects in STEM. We show experimentally, that a small geometric distortion of the central disk in STEM is related to the local curvature of the phase of the detected electron beam. This new method, operating at nanometre resolution in STEM, can be used to image local features in DPC of magnetic materials like domain walls, vortices or magnetic skyrmions; non-atomic electric fields; bend contour contrast; and the differential of the projected thickness of a sample [4].

DPC of magnetic materials is derived from the phase shift induced in the electron beam due to Aharonov-Bohm effect [2]. For the case of a sample with uniformly magnetised domains (along  $\pm y$  direction), Fig 1(a), these regions can be represented by a phase variation in the x direction  $\phi = \pm (2\pi e/h)B_Stx$ , where e is the electron charge, h is Planck's constant,  $B_S$  is the saturation magnetic induction and t is the thickness of the sample. This phase change is related to the deflection of the probe  $\beta_L = (\lambda/2\pi)\partial_x\phi = \pm (e\lambda/h)B_St$ , where  $\lambda$  is the wavelength of the illuminating electrons. It should be noted that a similar derivation can be shown for a wedge-shaped specimen with constant mean inner potential [4]. However, for the general 1D case, the phase can vary over the interaction volume as  $\phi = Ax + Bx^2 + \dots$  (A, B, ... are real) in the x direction - and this variation can cause higher order effects than a simple shift of the probe [5]. This can be seen at the domain wall in magnetic films as illustrated in Fig. 1(a). Experimentally, these effects can be suppressed by choosing a probe size over which the phase variations can be approximated as linear, so that a domain wall profile can be imaged by DPC [2,5].

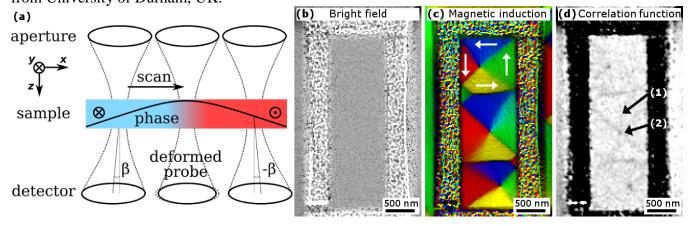
It is important to be able to resolve the higher order phase effects, for this purpose a pixelated detector is used to image a central STEM disk for each point in a scan over a sample. The data is then analysed by a normalised cross-correlation (NCC) algorithm post acquisition by comparison with a chosen mask of the probe. If the shape change of the probe is small, the position of the maximum of the NCC will measure an accurate deflection of the disk as has been demonstrated previously [2]. However a small change of the shape of the disk can be quantified by checking the maximum value of the NCC. As a proof of concept, we have collected a 4D STEM dataset from a thin film magnetic sample of Ni<sub>80</sub>Fe<sub>20</sub> (8 nm thick) with a FIB defined micrometre-sized rectangle that stabilises a multi-domain structure with various phase objects (domain walls and vortex cores). The data was collected on MerlinEM detector [6] with JEOL ARM 200 cFEG STEM at the University of Glasgow in aberration corrected field free mode [7]. The data was analysed by the NCC algorithm which is freely available [8]. The probe semi-angle was 0.5 mrad with the corresponding maximum spatial frequency (5 nm)<sup>-1</sup> and acceleration voltage was 200kV. Reconstructed

images are shown in Fig. 1: (b) is bright field image, (c) is colour combination of two orthogonal components of integrated magnetic induction and (d) is the correlation function sensitive to small shape changes of the central disk.

The correlation contrast produced by the NCC can be derived analytically and it can be shown that in a limit of a small shape change the contrast can be directly related to the quadratic component of the phase of the sample. This derivation will be studied further in a future publication. The new method is closely related to Fresnel imaging in plane wave illumination TEM where the imaging lens is defocused, however, the advantage of this NCC algorithm is that it is a completely focused method. To illustrate the sensitivity to the change of the diameter of the electron probe between points (1) and (2) in Fig 1(d), the analysis shows the difference of the surface area of the disks as 0.3%. The method can be applied to localised electromagnetic fields, sample thickness and mean inner potential analysis and moreover, the high sensitivity to small phase changes could be used to image biological specimens.

## References:

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**Figure 1.** (a) Schematic of a STEM scan over a magnetic domain wall. Linearly varying phase only causes a shift of the probe – blue and red area. Over the domain wall in the middle, the phase change causes a change in the shape of the disk. (b) Bright field reconstruction from 4D STEM data of an image of Ni<sub>80</sub>Fe<sub>20</sub> sample (8 nm thick) with FIB irradiated rectangular area. The probe convergence semi-angle was 0.5 mrad and the imaging was done with the objective lens switched off. (c) Colour combination of x and y components of integrated magnetic induction. Arrows specify the induction orientation. (d) Correlation function reconstruction where the intensity is directly related to the deviation of the shape of the probe due to non-linear phase gradient within the probe-sample interaction volume.