MerlinEM
4D-SPED Application Note

Scanning Precession Electron Diffraction with the MerlinEM Direct Electron Counting Detector

NanoMEGAS Precession Electron Diffraction System

In this application note, we will present results acquired with the NanoMEGAS Scanning Precession Electron Diffraction (4D-SPED) toolkit and the MerlinEM Direct Electron Counting Detector. The MerlinEM detector is integrated into the NanoMEGAS TopSpin application and synchronised with the DigiSTAR precession hardware.

Fig. 1: Diffraction patterns from a similar sample area of single crystal Si-SiGe. The MerlinEM detector was used with 10 ms dwell time and conventional optically coupled camera with 20 ms dwell time (both used 10 ms/pixel or 100 Hz precession frequency).1 Contrast calculated as a log between the minimum and maximum data value. Line Profile represented in Fig. 2 (Data courtesy Dr Joaquim Portillo, NanoMEGAS).

1 Additional experimental information is given in the Appendix.
Why Direct (Electron Counting) Detection?

In Fig. 1, we show a single frame from a 4D-SPED experiment conducted with a MerlinEM detector and a conventional camera imaging the same sample. Inelastic scattering (signal between the diffraction spots in the centre of the patterns) and single-electron events are clearly visible in the MerlinEM signal. The conventional camera sufficiently detects high intensity diffraction spots, but the poor signal to noise results in a loss of higher order diffraction information. The comparison of diffraction patterns demonstrates the clear improvement of the background noise with Direct Electron Detection, even though the imaging was performed with half the dwell time.

To further feature the improvements of Direct Electron Detection, we focus on the inelastically scattered signal profiles between the diffraction spots (as noted with lines in Fig. 1, plotted in Fig. 2). A clear profile of the electron counts with a decreasing minimum the further away from the centre of the pattern is achieved with MerlinEM. The conventional camera only achieves imaging of inelastic electrons for the highest intensity area close to the central diffraction spot and the signal is otherwise dominated by fluctuating noise of the CCD chip.

Getting more information from samples

In the previous section, we demonstrated that a direct electron counting detector collects data with a better signal to noise ratio for each probe position/precession compared to the conventional camera. In this section, we will compare the difference this makes for the 4D-SPED image reconstructions. We will show that Direct Electron Detection expands what is achievable in precession electron diffraction.
Improved Mapping Results

MerlinEM and conventional camera 4D-SPED datasets from the same sample were collected with NanoMEGAS DigiSTAR hardware. Datasets were exported to NanoMEGAS (.blo) format and analysed by Astar software. The sample contained different sized grains on the order of 10’s of nm to μm and as such, it is a good test of the sensitivity of reconstruction methods and data collection quality.

**Fig. 3:** A 4D-SPED reconstruction comparison - bright field, phase and phase reliability maps from Timetal 575 ® Ti alloy sample collected by a conventional camera and the MerlinEM Direct Electron Detector. α phase is in red and β is in green on the centre phase maps. White indicates 100% phase reliability on the bottom reliability maps. The data was collected with 10 ms precession frequency and 1° precession angle at 200kV. (Data courtesy Dr Ian MacLaren, University of Glasgow).
When compared, the conventional camera and MerlinEM maps (Fig. 3) demonstrate an increased clarity of diffraction information from the MerlinEM system. Not only is finer detail in the phase map achieved but greater confidence in phase reliability is also observed. Additional maps, full experimental details and discussion of results can be accessed in: MacLaren, I. et al, Microscopy and Microanalysis, 26(6), 1110-1116.

In general, the MerlinEM detector offers additional advantages and versatility due to its design. Each electron gets counted in hardware by analog to digital circuitry at pixel level - the signal is digitised straight away. This means that dose fractionation does not result in reduced signal to noise due to the readout of the camera. If for now we ignore the multitude of applications in 4D-STEM and EELS, the MerlinEM detector offers great potential for diffraction-based experiments - diffraction tomography, multipass data collection, ptychography, time-resolved imaging and others.

We would like to acknowledge NanoMEGAS for the integration of the MerlinEM detector to their 4D-SPED acquisition toolkit. We would like to thank Dr Ian MacLaren from the University of Glasgow for the example datasets in Fig. 3 and Dr Joaquim Portillio from NanoMEGAS for datasets in Figs 1 and 2.

Appendix (Experimental details):

**Fig 1 and 2:**
Data collected with TFS Spectra 300 probe corrected, monochromated X-FEG microscope at The University of Cambridge. The MerlinEM 1R (256x256 pixel detector) was used with 10 ms (at single precession) dwell time using 1º precession angle. Additional parameters: spot size 8, CL2 aperture 30 µm. The conventional camera (Stingray) was used with the same settings but with double (20 ms) dwell time (two precessions per pixel). The sample was a single crystal Si-SiGe.

**Fig 3:**
Data collected with JEOL ARM200cF probe corrected microscope at The University of Glasgow. The microscope was operated at 200kV in a TEM mode. A small aperture (10 µm) was used to achieve low beam current and 0.3 mrad convergence semi-angle. Precession was performed with 1º angle and 10 ms exposure per pixel. The sample was a dualphase Ti alloy, Timetal ® 575. Additional details and further information about the experiment can be found in publication: MacLaren, I. et al, Microscopy and Microanalysis, 26(6), 1110-1116.

MerlinEM key specifications:

Noiseless readout, zero dark counts; no dead time with 1-bit (18000 fps), 6-bit (3200 fps) or 12-bit (1600 fps) imaging; 30 keV - 300 keV operation; pixel size 55 x 55 µm; active area: 14 x 14 mm (256 x 256 pixels) or 28 x 28 mm (512 x 512 pixels); DQE at 60 keV: 1 at Zero frequency, 0.45 at Nyquist; MTF at 60 keV: >0.62 at Nyquist.
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4D-STEM and virtual STEM detection

Key methods:

- 4D-STEM
- HAADF
- ADF
- ABF
- BF
- CBED

MerlinEM key specifications: noiseless readout, zero dark counts; no dead time with 1-bit (18800 fps), 6-bit (3200 fps) or 12-bit (1800 fps) imaging; 30 keV - 300 keV operation; pixel size 55 x 55 μm; active area: 14 x 14 mm (256 x 256 pixels), 28 x 28 mm (512 x 512 pixels), 56 x 14 mm (1024 x 256 pixels); DQE at 60 keV: 1 at Zero frequency, 0.45 at Nyquist; MTF at 60 keV: >0.62 at Nyquist.

Using a pixelated detector in scanning transmission electron microscopy (STEM) opens a plethora of opportunities. Techniques like long range and atomic resolution electromagnetic field imaging, ptychography, fluctuation electron microscopy and others would not be as efficient or outright impossible without resolving a full diffraction pattern for each point of the scan. Using a pixelated detector in STEM results in a 4D dataset (2D scan and 2D diffraction dimensions) - hence 4D-STEM. It will eventually become a goto technique in STEM.

In this note, we will show examples of using a MerlinEM detector to generate established signals in STEM. We will use LiberTEM\(^2\) software to reconstruct the data and specifically its GUI web interface. We will show examples of virtual STEM images and convergent beam electron diffraction (CBED) from a few different samples. The figure below shows some examples of data which will be shown in this application note.

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2 https://libertem.github.io/LiberTEM/
Virtual detection in STEM

By acquiring a full diffraction pattern for each point of a scan, we can recreate standard STEM signals after the experiment by summation of the information within the respectful area of diffraction patterns. We essentially replace multiple STEM detectors (this could be annular detectors, a bright field detector and/or DPC segmented detector) with a single pixelated detector. Apart from the versatility of this approach, an additional advantage is that all the virtual detectors have a calibrated response and therefore data can be readily used in techniques like atom counting or others.

MerlinEM with its high dynamic range and versatile read-out system is ideally suited for 4D-STEM data acquisition. Hardware based electron counting offers exceptionally low noise by removal of thermal background. After the electron is detected, every process is digitised therefore no additional noise is added by the read-out system. Fig. 2 shows an example of a single probe position CBED diffraction pattern from a 4D-STEM dataset. In the example the contrast within the central probe can be clearly seen together with single electrons further away from the optical axis. The frame time was 0.5 ms and dynamic range was set to 6-bit. With these settings, a 256x256 probe position dataset can be taken in 32 seconds, which will minimise sample drift issues. The maximum speed of MerlinEM can be achieved with 1-bit dynamic range (18.8k fps) and it allows it to image the same area within ~3.5 seconds.

Fig. 2: A single probe position from a 4D-STEM dataset together with a line profile demonstrating single electron sensitivity and zero thermal background noise. The frame was taken with 0.5 ms frame time and 6-bit detector dynamic range.

3 If an annular detector is available above the position of the pixelated detector, the annular dark field images can be acquired together with the pixelated detector to collect additional information from larger scattering angles.
Atomically resolved interface in TiN/HfO2 sample

Fig. 3 shows a set of virtual STEM images generated from atomically resolved imaging of TiN/HfO2 interface. High angle annular dark field (HAADF), annular dark field (ADF), annular bright field (ABF) and bright field (BF) images were generated from corresponding areas of the detector which are shown in insets to the right of each image. The images follow the basic characteristics of each detector type: HAADF resolves high atomic number species as the brightest in the bottom half of the image following the adsorptive-contrast characteristics; ADF and ABF show a varying strength mixture of adsorptive- and phase-contrasts and BF shows mostly phase-contrast. The advantage of 4D-STEM here is that we can redefine each detector’s area to achieve ideal imaging for a given material type and even explore this after the experiment. This offers a maximum versatility together with options to explore the data further with modalities like DPC, Centre of Mass, electromagnetic field imaging and others.

Fig. 3: Demonstration of atomic resolution virtual STEM imaging in 4D-STEM of TiN/HfO2 interface. The summed parts of the detector are shown in the insets next to the figures. Data and sample courtesy of AEM group at TU Darmstadt by Alexander Zintler and Leopoldo Molina-Luna.
Complex sample with depth and material changes

Another desirable feature of 4D-STEM is the ability to characterise complex samples. This is demonstrated in Fig. 4 where multiple position averaged convergent beam diffraction patterns (PACBED) are shown with the respective area they were taken from. The sample is conventionally prepared lead free antiferroelectric. The overview on the left in Fig. 4 is a bright field image generated by summing the central disk area. Numbered PACBED patterns show:

- high order Laue zone lines in thicker parts of the sample in 1, 2 and partially over interface in 6,
- first order Laue zone ring in 3 and 4,
- a thin part of the sample showing multiple diffracted disks in 5.

Interestingly, other effects like shifts of the whole pattern (between 3 and 4) are visible which makes the classic STEM imaging difficult - the probe cannot be positioned in the centre of the detector for the whole sample. This kind of effect might be present due to the local differential of the thickness, crystal bending or possibly strain.

Fig. 4: Example of 4D-STEM data in a conventionally prepared lead free antiferroelectric sample. Diffraction patterns from each numbered areas were summed and their logarithm was plotted to show differences in local crystal structure. The size of the white circle corresponds to the size of the area summed. Data and sample courtesy: Alexander Zintler, Hui Ding, Leopoldo Molina-Luna, AEM group, TU Darmstadt.
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Ptychography using a scanning transmission electron microscope

Key methods:

- Ptychography
- 4D-STEM
- Atomic resolution imaging

MerlinEM is a fast, electron counting, pixelated detector for the transmission electron microscope. Its high dynamic range, radiation hardness and versatile readout system make it an exciting tool for scientific research.

MerlinEM key specifications: noiseless readout, zero dark counts; no dead time with 1-bit (18800 fps), 6-bit (3200 fps) or 12-bit (1600 fps) imaging; 30 keV - 300 keV operation; pixel size 55 x 55 μm; active area: 14 x 14 mm (256 x 256 pixels) or 28 x 28 mm (512 x 512 pixels); DQE at 60 keV: 1 at Zero frequency, 0.45 at Nyquist; MTF at 60 keV: >0.62 at Nyquist.

Electron ptychography

Electron ptychography is a computational imaging technique. It is used to generate an image of a sample from a large dataset containing interference patterns. Ptychography can recover a complex phase related to the sample, which is not directly accessible due to the nature of the physical measurements.

Ptychography in scanning transmission electron microscopy (STEM) is applied to 4D-STEM data (where a 2D diffraction pattern is acquired for each point in 2D scan) and can be used with a focused or defocused probe. A complex phase of the sample is generated post experiment by reconstruction techniques. A simplified schematic of STEM ptychography is shown in Fig. 1.

Fig 1: Schematic of 4D-STEM ptychography. 4D data is acquired by pixelated detector and resulting images are reconstructed in a computer. Reconstructed image from graphene bilayer (courtesy of Christopher S. Allen, ePSIC, Diamond Light Source, Harwell Oxford, UK)
Ptychography in STEM

Ptychography in STEM can be separated into two distinct methods, depending on whether a focused or a defocused probe is used:

- **Focused probe data** can be reconstructed with a single-step method with a single-side band (SSB) algorithm based on a weak phase object approximation\(^1\) and Wigner-distribution deconvolution (WDD)\(^2\) which is applicable to a wider selection of samples. The main advantage of focused probe ptychography is that the additional STEM signals can be collected at the same time (i.e. HAADF, EM fields). An example image of MoS\(_2\) sample are shown in Fig. 2.

- **Defocused probe data** can be reconstructed by iterative algorithms\(^3\). It can be effectively used to reduce the dose in STEM to image beam sensitive materials and cover a larger field of view of the sample. An example from a graphene sample is shown in Fig. 3.

Electron ptychography is a very active research area. It is applicable to any probe imaging and can be used to correct lens imperfections, scanning inconsistencies and/or access higher resolution than that defined by the probe forming aperture.

MerlinEM, an electron counting detector, can be used to acquire 4D-STEM data useful for various forms of ptychography. Its single electron efficiency, zero read-out noise and high dynamic range make it a highly applicable and versatile detector technology.

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Examples of ptychography publications featuring MerlinEM detector in STEM ptychography

  Very fast momentum resolved STEM imaging (>10,000 fps) was used to image convergent beam electron diffraction (CBED) with only tens of electrons per probe position. An example of beam sensitive imaging was given for a ZSM-5 Zeolite sample with a dose as small as ~ 200 e / Å².
  **Note:** The limit of 1-bit imaging of MerlinEM is ~18,800 fps which is getting closer to standard STEM speeds with an annular detector (53 μs pixel dwell time).

- **Wen, Y et al. Nano letters 19. 9(2019): 6482–6491.**
  2D materials were studied by 4D-STEM. A combination of electron ptychography, electric field imaging and virtual annular detection was used to identify low and high atomic number atoms simultaneously in a single experiment.

- **Song, B et al. Physical review letters 121. 14(2018): 146101.**
  This publication demonstrates a possibility to combine EELS and Ptychography in the same experiment. By omitting bright field CBED disk, the image of the sample can be still reconstructed in ptychography. If a detector with a hole in the centre is used this would allow a free path of the bright field signal to spectrometer.

  Study of defocused low dose imaging for 2D materials science. MerlinEM was used as an electron counting, sensitive detector to collect a defocused STEM CBED probe.

  Atomic electrostatic maps of 1D channels in 2D semiconductors using 4D scanning transmission electron microscopy. A comprehensive study of 2D materials using 4D-STEM and MerlinEM detector. A simultaneous electric field and ptychography imaging were applied to 1D interface in low dimensional materials.
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Application Note

Using 4D-STEM to measure electromagnetic fields in the transmission electron microscope

Key methods:

- Differential Phase Contrast (DPC)
- Centre of Mass (CoM)
- Cross-correlation (CC)

MerlinEM is a fast electron counting pixelated detector for the transmission electron microscope. Its dynamic range, radiation hardness and versatile readout system make it an exciting tool for scientific research.

MerlinEM can be used as a 4D-STEM camera where a full diffraction pattern is acquired at each point in a scan. Applications of 4D-STEM imaging are wide ranging; established methods including bright field imaging (BF), high angle annular dark field imaging (HAADF), annular bright field imaging (ABF) and differential phase contrast (DPC) are easily accessible through post processing. Exciting emerging techniques like ptychography, momentum transfer imaging and pattern matching would not be possible without access to the full diffraction information enabled by pixelated detectors like MerlinEM. A selection of open source software is available for analysis of 4D-STEM and MerlinEM data¹.

Key specifications: noiseless readout; no dead time with 1-bit (14400 fps), 6-bit (2400 fps) or 12-bit (1200 fps) imaging; 30 keV - 300 keV operation; pixel size 55 x 55 μm; active area: 14 x 14 mm (256 x 256 pixels) or 28 x 28 mm (512 x 512 pixels); DQE at 60 keV: 1 at Zero frequency, 0.45 at Nyquist; MTF at 60 keV: >0.62 at Nyquist.

¹ LiberTEM https://libertem.github.io/LiberTEM/index.html
HyperSpy https://hyperspy.org/
pixelatedDPC https://github.com/matkraj/pixelatedDPC
pixSTEM https://pixstem.org/index.html
py4DSTEM https://github.com/py4dstem/py4DSTEM
fpd https://gitlab.com/fpdpy/fpd

4D-Stem Schematic

Scanning Directions

Electron Beam

Sample

Pixelated Detector
This note will focus on quantitative imaging of electromagnetic fields in the transmission electron microscope in scanning mode. Electromagnetic fields can be separated into two length scales:

\[ F = -e \mathbf{E} - e (\mathbf{v} \times \mathbf{B}), \]

where \( e \) is the charge of an electron, \( \mathbf{E} \) is the electric field, \( \mathbf{v} \) is the speed of the electron and \( \mathbf{B} \) is the magnetic induction. Transmitted electrons acquire momentum which is integrated over the thickness of the sample.

In this part we focus on imaging of magnetic fields, however, essentially the same methodology can be used to analyse long-range electric fields in e.g. biased sample with a PN junction.

In the case of magnetic fields, the measurement is only sensitive to the component of the field perpendicular to the electron trajectory. It is also important to mention that the vast majority of magnetic samples need field free conditions, otherwise the magnetisation of the sample is saturated in the high field created by the objective lens of the TEM. This additional momentum results in a shift of the central probe (disk) on a detector, which can be measured quantitatively.

Quadrant and annular quadrant detectors are the established detection methods for measurement of the disk shifts. In the case of MerlinEM, multiple opportunities for signal analysis are available. We can process the data with virtual DPC and annular DPC detectors, allowing quick analysis but sacrificing most advantages of having the full diffraction information available at each point in a scan. The convenience of this approach is that we can define a suitable detector geometry and its centre post experiment.

Fast acquisition, electron counting and noise-less readout make MerlinEM an ideal tool for dynamic TEM imaging. Timescales below 100 ns are reported in pump probe experiments. Imaging of in-situ dynamic processes with up to 2400 fps is possible with 6-bit dynamic range. In the case of EM fields, TEM can be used in fresnel mode (defocused illumination) to image fast field dynamics. MerlinEM can be also used in a binary mode with up to 14400 fps with individual frames acquired in 70 μs.

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2 Ni\(_8\)Fe\(_{20}\) sample was made by Azzawi Sinan and Del Atkinson from University of Durham, UK.
4D-STEM data can be also used to calculate the Centre of Mass (CoM) of the pattern on the detector. This method has an advantage in that the shift of the probe is measured linearly - contrary to the quadrant method, where the selection of the centre of the pattern is important and the beam shifts have to be within a certain range.

**Fig 2:** Centre of mass generated image from the same dataset as the image above. The small discrepancies in signal between FIG 1 and FIG 2, calculated with different methods, are artefacts related to imperfect placing of the virtual detector in the quadrant method.

A lot of industrially relevant research is applied to polycrystalline materials, for which the CoM method might not be the ideal approach. This can happen if the size of the features (crystalline grains) is comparable to the size of the beam. Under these conditions, the intensity in the central diffraction disk is not flat but has variations due to crystallites meeting different diffraction conditions. Therefore, with CoM and quadrant based approaches the final image also contains diffraction contrast which is not related to the deflection of the probe. This is overcome by a method based on computer vision, where the probe position is analysed by a template matching algorithm. Each diffraction pattern in 4D-STEM dataset can be cross-correlated with an idealised mask pattern. To achieve even greater signal to noise, a 2D gradient is applied to the data before cross-correlation. This way we can achieve a superior imaging mode, which isolates the magnetic contrast from the short-range electrostatic crystallite contrast.

**Fig 3:** Cross-correlation (CC) based analysis of the same 4D-STEM dataset. The superior contrast of the CC method is evident when compared with Figs 1 and 2. The analysis code is based on GPU acceleration and is freely available. The details about processing and methodology can be found in Krajnak M. et al, Ultramicroscopy 2016.

Even very complicated samples (e.g. perpendicularly magnetised multilayer polycrystalline films) can be quantitatively imaged with the cross-correlation method. An example of such an image is shown below.

**Fig. 4:** A comparison of magnetic field images from segmented (left) and cross-correlation (right) methods. The sample consists of 15 repeated layers of [Co(1.6)|Ru(1.4)|Pt(0.6)] capped with Pt(2.4) on the top and Ta(10)|Pt(8) on the bottom (number in brackets is the thickness of the given layer in nanometres). In such a complicated structure DPC detection shows no magnetic contrast at all and only cross-correlation can be used. In this case the sample is magnetised out of plane so it needed to be tilted to show any contrast. It is remarkable that even if the effective magnetic thickness is only 17% of the thickness of the sample, the contrast can be still recovered in a multilayered polycrystalline structure. Image courtesy Kayla Fallon, details can be found in Fallon K. et al, preprint https://arxiv.org/abs/1901.03652.
2. Atomic scale electric fields

Electric fields can also be measured on atomic scales with MerlinEM. It was shown that an average momentum transfer from a transverse electric field can be measured by a CoM algorithm in thin samples. Momentum transfer can be described by:

$$\langle p_\perp \rangle = \int \int p_\perp \cdot I(p_x, p_y) \, dp_x dp_y$$

where is $p_\perp$ the average transverse momentum transferred to the beam by the sample, $I(p_x, p_y)$ is the recorded diffraction pattern (normalised) with the components representing the coordinate system in the momentum space. This equation essentially denotes a CoM calculation of the measured diffraction pattern. In this scenario, the probe size is smaller than the atomic spacing. Even if there was a probe shift due to the long range EM fields, this shift would be overcome by effects of atomic electric fields. Probe shifts in nanodiffraction are usually in the range of tens of microradians or smaller, whereas the expected average shift for atomic resolution imaging can be close to the size of the probe defining aperture e.g. >5/10 milliradians (about 3 orders of magnitude larger).

In recent publications, MerlinEM was successfully used to measure atomic electric fields - a quickly developing field of research. Interestingly, once acquired, the 4D-STEM data can be used for multiple types of analysis; focused probe ptychography and atomic electric field imaging essentially require the same type of data. In the image below, 4D-STEM data of a 2D material, MoS$_2$, was used to generate a phase image (by ptychography), a transverse electric field image (by CoM) and a charge distribution image (by divergence of CoM). Thus illustrating the richness of information available from a single dataset.

![Fig 5](image)

*Fig 5:* Phase (left), the magnitude of the electric field (middle) and the charge distribution (right) images generated from the same 4D-STEM dataset acquired by MerlinEM detector in MoS$_2$ with a horizontal defect present in the middle of the image. The image was obtained under CC BY 4.0 licence from Fang S. et al, Nature Communications volume 10, 1127 (2019).