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Port-Barcarès

Review and News of Quantitative TEM techniques

# Practical: 4D **STEM and DPC**

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**Goal of this practical:** This practical session is about different 4D STEM (*Scanning Transmission Electron microscopy*) imaging techniques. It follows after the HAADF practical with the aim to explore more sophisticated STEM methods.

Note: the microscope needs either a segmented detector with live imaging capabilities, or the equivalent in 4D-STEM. As the different systems have different capabilities, we will give below some basic information of the different setups, and will keep the color coding for variations on the practical on the different setups:

**JEOL F2: 1.4 Å spatial resolution at 200 kV. Two pixelated CMOS detectors before and after the energy filter. Virtual imaging with DPC and Center of Mass (CoM) is possible as well as integrating and differentiating. Strain mapping is also possible.**

**ILLIAD TFS: 0.5 Å spatial resolution at 300 kV. Segmented detector with live DPC iDPC and dDPC and two pixelated detectors and CoM data treatment in the acquisition software. One pixelated detector with CMOS technology (Ceta camera) and one with direct detection: EMPAD.**

**TESCAN TENSOR: 2.8 (3.5) Å spatial resolution BF (HAADF) at 100 kV: pixelated detector and virtual imaging. Virtual DPC or CoM are not yet supported in the software. Precession up to an angle of 3 degrees is available at a mindboggling speed of 72 kHz. Framerate of Dectris is up to 4500 fps at 8 bit. Strain mapping and orientation mapping are possible.**

*If a certain point can't be done on the system, we simply continue and will find other more suitable options later on in the TP.*

STEM is a versatile TEM mode, where the resolution of the image is usually limited by the probe size. Depending on the detector that is used, the contrast formation is different and various aspects of the sample can be studied, from compositional to electrical properties. We are going to study DPC (*Differential Phase Contrast*) that offers the possibility to image electric fields present in the sample, as well as light elements from iDPC (integrated DPC). In DPC, typically a segmented detector is used. It can be seen as a detector with only few pixels, typically 4, 8 or 16 segments (pixels) are used. Finally, we will see how all these techniques can be applied, and much more, at the same time using a fast pixelated detector (4D-STEM) where the diffraction pattern is sampled with many more pixels.

The sample: MBE grown GaN nanowires with AlN insertions, courtesy of Akhil Ajay and Eva Monroy. The sample is prepared by wet dispersion on lacey carbon grid by Martien den Hertog. The sample number is E3812. The GaN nanowires contain10 periods of 5/5 nm GaN/AlN, 7 periods of 5/7 nm, 5 periods of 5/10 nm, on a 700 nm base. 500 nm cap. 100 nm close to the heterostructure undoped. Rest doped with Si $3^{19}$ at $cm^{-3}$.

**System studied:** AlN/GaN heterostructure nanowires containing _polarization-induced electric fields_. What happens in nitrides is that, due to the hexagonal wurtzite structure, the negative charge cloud does not entirely superpose on the positive charge cloud along the c direction of the crystal, resulting in a dipole moment. We will see the effect of this dipole at the interface along this crystal direction. Since the dipoles in the different nitrides (for example AlN, GaN, or InN) are different, we will obtain a fixed charge sheet at the material interfaces, resulting in polarization fields. Strain will also influence the exact value of the polarization field. Since these polarization fields will have a dramatic effect on electro-optical properties (for example a GaN disk between AlN barriers can emit light well below the bandgap of pure GaN due to these fields) we are very interested to measure it precisely. We note that it is not easy to quantify these fields precisely, because of the mixture of material contrast and electrical contrast at hetero interfaces. This practical will therefore merely look at qualitative effects to get some feeling for field mapping.

This practical aims to acquire 4D STEM data, that you will analyze with different methods during the data treatment practical by Knut and Pete: using Center of mass approach and Single Side Band.

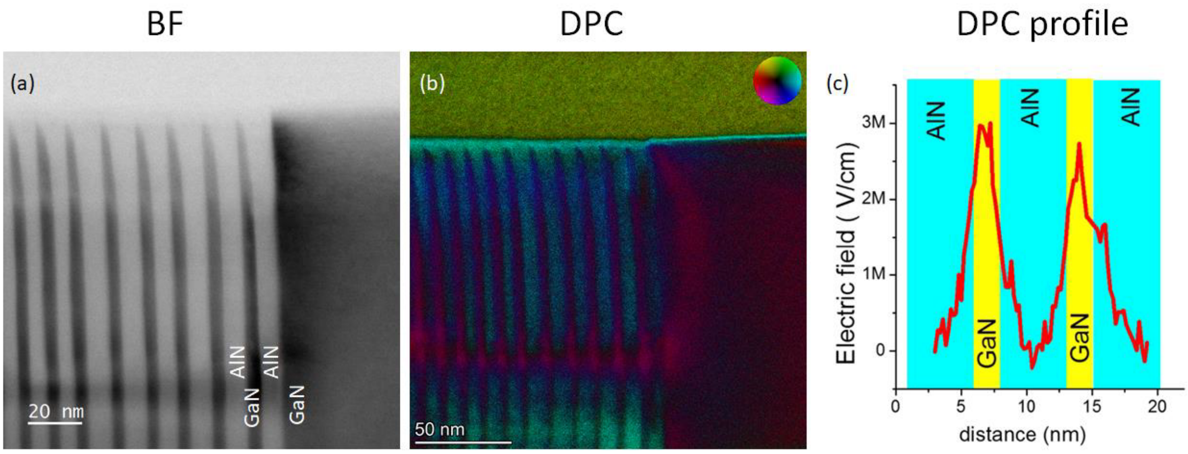


**Figure 1** - BF of the AlN/GaN quantum wells inserted in a nanowire. (b) DPC image of the same region. (c) example of DPC profile in such a sample.

# 1 - DPC *(40 min)*

### *2.1 Principle*

Differential phase contrast (DPC) imaging is a STEM method to visualize an *electromagnetic field* in a specimen by *measuring the deflection of an electron beam* due to the field at each beam-scan point. The beam deflection is measured with a segmented detector. When a segmented detector composed of four segments is used (see Figure 2), the angle and the direction of the beam deflection (beam shift on the detector plane) are measured from the difference between the signal amounts acquired with the two segments opposed to each other. *Dwell times used here are in the other of µs*.

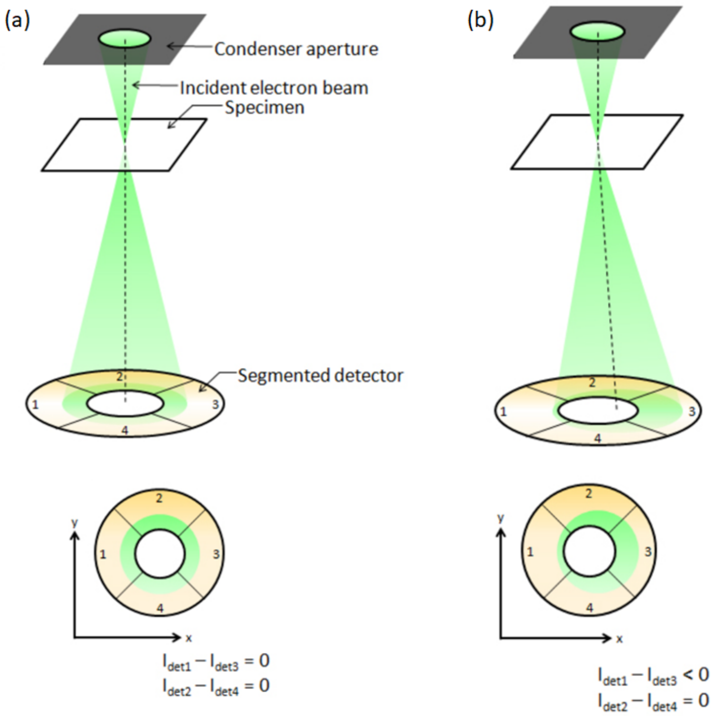


**Figure 2** – Schematic of DPC measurements. (a) probing a region with no field. (b) probing a region where the magnetic or electric field deflects the incoming electron, beam.

The projected electric field can be obtained from the beam shift measured through the Lorentz force as:

$$E_{\perp} = -\frac{\gamma m_e^* v_0^2}{qt} \quad \text{(eq. 1)}$$

Where $q$ is the elementary charge, $\gamma$ is the beam shift measured, $m_e^*$ the relativistic mass of the electron, $v_0$ the relativistic electron velocity and $t$ the sample thickness. Since DPC signal is a measurement of the beam shift, according to equation 1, this is proportional to the electric field, and the DPC image is an image of the electric field present in the sample.

The assumption for this kind of DPC is that the diffracted disk shifts as a whole due to the electric (or magnetic) field, typically referred to as rigid-shift (or hard shift). However, when the field changes over the probe volume, not a rigid-shift but a redistribution of intensity will be seen.

### *2.2 Setting up the microscope*

Alignment at HR conditions, convergence semi angle of about 20 or 30 mrad.
Prior to the practical: find some suitable Nanowires with nice heterostructure, align them on [1-100] and [11-20] and save the positions. The NW cross section is shown below to understand how we will project the structure [10.1021/nl302890f]. When looking along a **[11-20]** like direction we are arriving at a corner of the hexagon, and look along two [1-100] side facets. When looking along a **[-1100]** direction we enter at a side facet, and therefore expect the thinnest region at the edge projecting a corner of the hexagon.

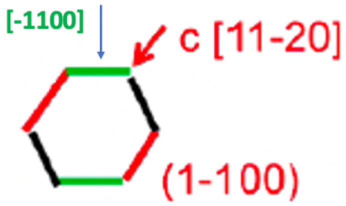


**Figure 3** - GaN nanowire cross section.

### *2.2.1 Rotation* :

Between your STEM image and your diffraction pattern. Before starting any experiment, it is important to understand the rotation. When you are using scan rotation, you are rotating the grid to scan the image. This does not impact the diffraction pattern at each scan point. In order to make a link between an electric (magnetic) field direction at each scan point with the structure we scan, we need to know the rotation between the diffraction pattern and STEM image. Due to different coordinate systems, a y-flip of the diffraction pattern may also be present.

*Verify the rotation between the STEM (HAADF or BF, doesn't matter much) image and your diffraction pattern. Look first at the STEM image, with zero scan rotation, then look at the diffraction pattern with the 0002 diffraction (NW growth plane) clearly visible, then defocus to see a TEM like image of the nanowire: what is the angle between STEM image and diffraction pattern?*

### *2.3 The experiment*

Place your beam at different locations in the NW structure or make a 4D STEM map and look at the diffraction pattern at different locations of the NW. At atomic resolution (**HR** setting in TENSOR, no atomic resolution), do you think we are in the case of 'rigid-shift' or 'redistribution of intensity'?

*Measuring the polarization-induced Electric fields*

*2.3b – Make an DPC image in NB, can you see the Electric field profile similar to the one displayed in Figure 1c? Here we aim to have the probe a bit larger than interatomic spacings, to average out the electric field at atomic length scale. So, the semi convergence angle should be somewhere between 1-3 mrad, See fig.4.*

**On JEOL: both virtual DPC as well as CoM imaging is possible, so we don't need to split in first DPC and then CoM, but could look at both one after the other once a dataset is acquired.**

**On TENSOR: For now, only 1D virtual detectors are available in the standard software. If possible virtual DPC and or CoM will be tried.**

As discussed before, using DPC you can image fields. You can image the electric field present in the sample, thus you can measure electrical properties in a TEM!

*2.3c - What is the order of magnitude of the electric field/magnetic field that I want to study and the respective beam shift?*

Calculate the value of the beam shift for an electric field in the order of 3 $MVcm^{-1}$, using a sample thickness of 200 nm and 200 keV electrons, knowing that the equation 1 in this case is $\gamma = \frac{E_{\perp}t}{3.438 \times 10^5 V}$.

*2.3d – Looking at this particular nanowire heterostructure, with AlN and GaN, what is the minimal spatial resolution needed to study the field gradient? For this sample, what would be a good tradeoff between spatial resolution and field precision? Figure 4 can help you to have an idea. What convergence angle do you need in this case and why?*

Before setting up the convergence angle for performing DPC you should think about which spatial resolution is needed, if you are working with small built-in electric field as in a p-n junction, depletion regions are in the order of 20 nm – 100 nm, while for atomic electric fields, you need better spatial resolution and smaller probe possible, in order to resolve the atomic columns.

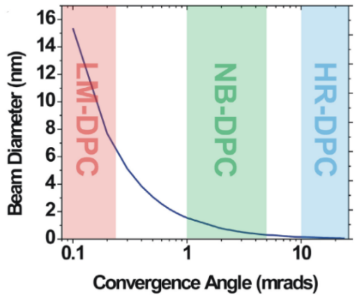


**Figure 4** - Plot of diffraction limited beam diameter as a function of convergence angle, neglecting the influence of source size and aberrations.

*2.3e - How can we get such spatial resolution and field detection sensitivity, should we work in LM, NB or HR? If we perform our experiment in HR, how could we separate the beam shift caused by the atomic fields from the polarization-induced one?*

It depends on your sample needs, if you are studying atomic fields, you need to work in HR (high convergence angles and small probe size). On the other hand, if you are working with p-n junction a bigger probe size can be used, HR is also possible, but you should average the signal measurement in a region bigger than the unit cell of the structure studied, in order to remove the atomic fields. Because of the uncertainty principle, you will always have a tradeoff between spatial resolution and field precision, nicely described in this paper [https://doi.org/10.1016/j.ultramic.2021.113342]

*2.3f – Make one DPC image in NB and another in LM, can you notice any difference?*

By reducing the semi convergence angle, you gain in deflection precision. But you loose spatial resolution. Thus the beam shift sensitivity is increased. In order to fit the transmitted beam in the 4 quadrant detector (or pixelated detector), a longer camera length needs to be used. Thus, you lose some spatial resolution. There is a tradeoff here, between spatial resolution and electric field sensitivity! What is the best for you? Depends on the needs of your sample!

***2.4 Practical issues and Limitations***

*2.4a - Make a profile in your DPC map acquired in NB, can you say how much is the field you have measured? See how much the field profile is changing in the different sized regions.*

Notice that the DPC signal is not calibrated, therefore, you do not know how much the beam shift is, and you cannot use equation 1 to retrieve the electric field. However, you can have a qualitative information about how the electric field varies on the region studied. Therefore, we can conclude that DPC is not directly quantitative! Quantitative information from DPC is possible, but is not an easy task.

*2.4b - Compare two orientations turning about 4 degree around the NW axis, and make a DPC map in NB in each orientation, what do you observe? Orient the sample in condition where the kikuchi lines are avoided, make a DPC map, what do you observe?*

DPC signal is strongly affected by diffraction contrast, depending on the sample, this can hinder the interpretation of the signal obtained.

# 3 - 4D-STEM (40 min)

### *3.1 Principle*

4D-STEM is an extension of DPC, basically adding many more pixels beyond the 4, 8 or 16 of DPC. 4D-STEM cameras capture the *full electron diffraction pattern*, while conventional STEM detectors record a single value per probe position. In 4D STEM, the probe is rastered on the specimen in a 2D array. At each probe position, a 2D diffraction pattern is imaged on a pixelated detector. This generates a 4D data cube that can then be further analyzed. This is possible due to development of *fast pixelated detectors*, dwell times used here are in the other of ms, depending on the camera you use. So-called pixelated 4D STEM gives us more information at the cost of longer pixel times. Since STEM existed, the possibility to do 4D STEM was there. However, with the technology of 10-20 years ago, we were limited to say 10 or 25 images per second, you can easily see that if you want to scan a field of 100 by 100 px, this takes……… a long time… 8 min. So your sample may have gotten serious beam damage or drifted.

If the TEM features more than one camera, try to obtain 4DSTEM maps at the same conditions. How do they compare? Looking at the diffraction patterns do you see a difference?

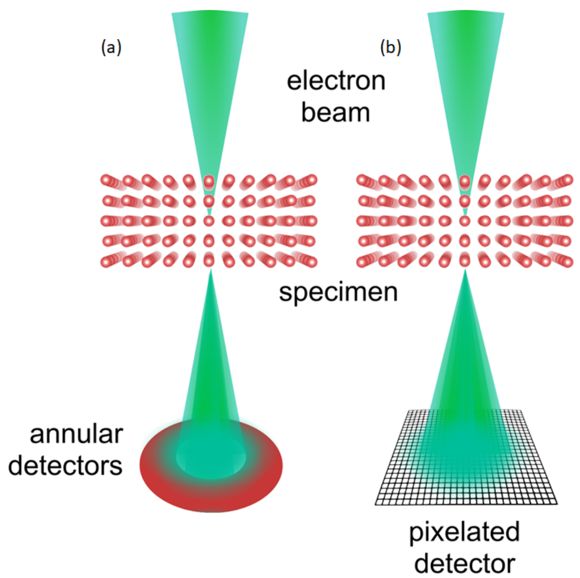


**Figure 5** – (a) Schematic of HAADF method, electrons are collected in the back focal plane by an annular detector. (b) Schematic of a 4D-STEM experiment, electrons are collected in the back focal plane by a fast pixelated detector.

Once you have acquired the 4D STEM map, you can do many things in data treatment. For example add virtual detectors, BF, ABF, ADF, HAADF or DPC!

You can also determine the beam position by calculating the Center of Mass (CoM) in the diffraction pattern recorded on the pixelated detector:

$$\boldsymbol{k}_{CoM} = \frac{\sum_i \boldsymbol{k}_i I_i}{\sum_i I_i} \qquad \text{(eq. 2)}$$

Here the pixel position is defined as $\boldsymbol{k}_i = (k_x^i, k_y^i)$ and the intensity in each pixel as $I(\boldsymbol{k}_i)$. The difference between the results of CoM performed in the unaltered beam (positioned outside the p-n junction), and the deflected beam at the p-n interface: $\gamma_{shift} = \left|\boldsymbol{k}_{CoM}^{shifted} - \boldsymbol{k}_{CoM}^{reference}\right|$, see Figure 6. Then, using equation (1), the electric field can be measured.

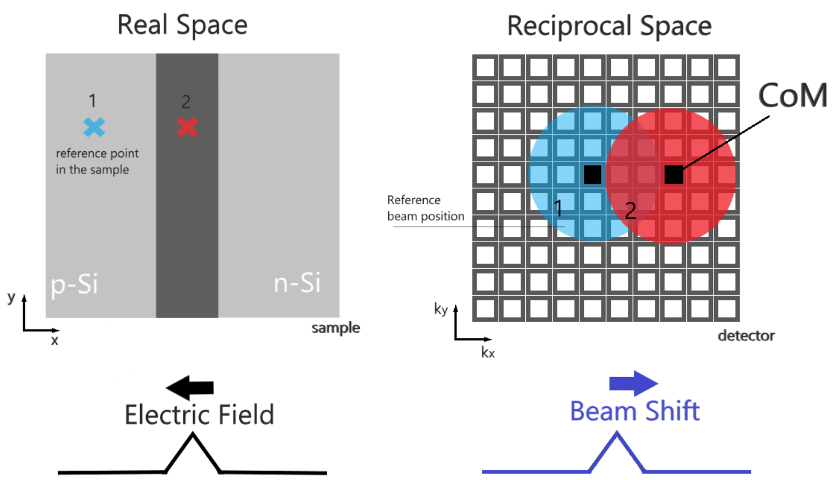


**Figure 6** – Schematic of the Electric field measurement through the detection of the beam shift on a pixelated detector for a p-n junction.

CoM is one method to analyze the beam position. You could also match a template to the diffracted disk using cross correlation, typically referred to as template matching (TM).

### *3.2 Setting up the microscope*

Alignment

### *3.3 The experiment*

*3.3a repeat the same measurements performed in DPC in NB mode, notice that now you have to work with dwell times in the order of ms.*

*If your system is equipped with **Precession**, acquire the same map with and without precession, what do you see? (**TENSOR** and **ILLIAD**?)*

*If your system is equipped with **Energy Filtering**, acquire the same map with and without energy filtering, what do you see? (**JEOL F2** and **ILLIAD**?)*

*3.3b In 4D-STEM quantification is simpler than DPC, because we can easily calibrate the detector and determine the beam shift in rads (usually some µrads for the sample under study). Do you have any idea how this could be done?*

*3.3c In order to perform CoM, you need to analyze the 4D data set posteriorly, this will be done in the practical 4D-STEM on Thursday/Friday with Prof. Knut Muller.*

### *3.4 Limitations*

*3.4a – Acquire a 4D-STEM map in a random crystallographic direction. Look at the diffraction pattern on the camera, do you see anything else rather than the transmitted beam? Move the beam position across the sample, do you see any difference? Do you think this can affect the CoM? Why?*

*3.4b – Depending on the software, you can instantaneously observe the results, however, often in 4D-STEM (depending on your system) you need to post-process the multidimensional data set. What is the size of the data you got?*

4D-STEM can capture a lot of information at the same time, but also generates big data sets, that easily reach a few gigabytes. In addition, 4D-STEM can be done on a pixelated detector where at least a dwell time of ms can be used. Thus, depending on the camera used and the size of the map acquired this imposes some limitations, because the acquisition time may be so long that sample drift will become a problem. However, cameras are becoming faster.

### *3.5* ***Explore more: Strain and orientation mapping*** *(if available)*

*If your system is equipped with* ***Strain Mapping routines****, acquire a 4D STEM map at about 2 mrad semi convergence angle. It is similar to the field mapping conditions used earlier* ***BUT*** *you will use a very different camera length (magnification of the diffraction pattern), as now you are interested to see the diffraction spots and not only the direct beam. You may acquire this map with and without precession. What is the difference? (****TENSOR, JEOL F2*** *and* ***ILLIAD****?)*

Lattice variations are present between the GaN and AlN, and strain relaxation will occur at the NW sidewalls. Can we see it?

*If your system is equipped with* ***Orientation Mapping routines****, acquire a 4D STEM map at about 2 mrad semi convergence angle. It is similar to the field mapping conditions used earlier* ***BUT*** *you will use a very different camera length (magnification of the diffraction pattern, as now you are interested to see the diffraction spots and not only the direct beam. You will need precession. (****TENSOR****)*

Due to strain or nanowire bending (due to growth, strain or wet dispersion) you may see orientation differences along the NW or along the heterostructure. How much is the orientation changing? Think back of seeing orientation changes in the HAADF STEM practical, what is the difference between both experiments (HAADF vs 4D STEM) in terms of ease and getting a more or less exact value for the orientation change?

### *3.6 Electron Ptychography*

Finally, lets obtain 2 4D STEM maps for reconstruction with CoM and SSB:

The first one in HR mode: Scan a field of 100 by 100 positions with a px step of around 0.15 Å.

The second one NB conditions with 2 mrad beam semi convergence angle. Scan a field of 100 by 100 positions with a px step of around 0.3 nm.

You hopefully see this data back in the 4D STEM data treatment practical.

# 4- Conclusions

We looked at various forms of 4D STEM. Using a detector with few pixels (segmented detector for DPC) or with more pixels. If certain sampling conditions are met, 4D STEM data can also be used for iterative electron ptychography (see Pete's talk and practical). Here it is important that the beam positions on the sample have a decent overlap, while this is not so important for other approaches (virtual detectors, CoM or TM).

A different aspect of 4D STEM is also orientation mapping and precession, which is potentially treated in this practical and in the Nanomegas talk and demo.

i DPC can be used to visualize light elements as well as electric or magnetic fields! Quantification of electric fields in 4D-STEM is possible, but you should be careful to minimize diffraction contrast.

Further reading:

[10.1103/PhysRevLett.122.106102] Knut's excellent paper on mapping fields in similar nanowires

[https://doi.org/10.1063/5.0104861] Our paper on detection limits by CoM in a p-n junction