High Resolution TEM

<u>Purpose of the practicum</u>: acquiring a hands-on experience in TEM operation, understanding of crucial optical and experimental parameters for imaging, understanding the relation of experiment and calculation in HRTEM.

<u>Results to be achieved</u>: experimental estimation of instrument parameters (magnification, spherical aberration and defocus); experimental recording of HR images, estimation of parameters and theoretical calculation of the images for these parameters.

<u>Description of experiment</u>:
- sample – Si [110] with flat amorphous areas;
- basic alignment of TEM and sample – height, orientation, rotation center, defocus, astigmatism;
- acquisition of lattice images from a thin crystalline region at ~x300K;
- acquisition of the image time series of amorphous at ~x300K magnification and underfocus ~-500nm for Cs-Df-astigmatism determination.

<u>Description of calculations</u>:
- calibration of magnification using images of crystalline areas (DM);
- determination of optical parameters of the microscope using diffractograms of amorphous areas (DM, Polar transformation script, Gauss filter script, Excel or Origin);
- determination of defocus values of defocus series and calculation of images for estimated microscope parameters (Df, Cs) and different thicknesses; estimation of the thickness (DM, EMS).

Theoretical background

Under ideal imaging and sample conditions a HRTEM image is an interference pattern of unscattered electrons and those scattered by the sample. The interference pattern is affected by phase modulation introduced by the aberration of the objective lens, i.e. the image is a convolution of the point transfer function of the objective lens $T(r)$ with the object exit wave function $\psi_o(r)$ [1, 2, 3, 4].

$$\psi(r) = \psi_o(r) \otimes T(r) \qquad (1)$$

The phase modulation by the objective lens is better understood in the Fourier space at the back focal plane of the objective lens (Fourier space) where the Fourier transform of the image intensity is given by

$$\psi(q) = \psi_o(q)T(q) \qquad (2)$$

With $\psi(q)$ and $T(q)$ being the Fourier transforms of the object-exit wave function $\psi_o(r)$ and $T(r)$ respectively, q is the spatial frequency, and the phase contrast transfer function $T(q)$ is defined as

$$T(q) = A(q)Sin\chi(q) \qquad (3)$$

where A(q) is an aperture function and $\chi(q)$ is a phase modulation function due to lens aberrations.

Considering only defocus and Spherical aberration, $\chi(q)$ then takes the form:

$$\chi(q) = 0.5\pi\lambda^3 C_s q^4 + \lambda\pi\varepsilon q^2 \tag{4}$$

where λ is the wavelength of the electrons, ε is the defocus, C_s is the spherical aberration constant. If we assume the aperture function $A(q)$ to be = 1, then the function $\chi(q)$ describes the phase modulation of the exit wave function as it propagates through the imaging system. $\chi(q)$ can be evaluated by analysis of the Fourier power spectrum of high resolution images. Power spectrum of the image is expressed as

$$P(q) = |\psi(q)|^2 \tag{5}$$

Combining equations (2), (3), (5) we get

$$P(q) = \psi_o{}^2 A \sin^2 \chi(q) \tag{6}$$

For an amorphous specimen the probability of scattering can be assumed to be constant for all spatial frequencies therefore the intensity of the power spectrum is defined mostly by the contrast transfer function

$$P(q) \sim \sin^2 \chi(q) \tag{8}$$

$\sin \chi$ takes particular values -1, 0 and +1, when the following condition is met

$$\chi(q) = 0.5\pi\lambda^3 C_s q^4 + \pi\varepsilon\lambda q^2 = n\frac{\pi}{2} \tag{9}$$

Thus a power spectrum (radial distribution of intensity of FFT pattern of the image) of the amorphous specimen, which is proportional to $\sin^2 \chi$, will contain minima corresponding to zeros of $\sin \chi$ and maxima corresponding to ±1. Each maximum in power spectrum may be associated with odd and minima with even integer **n**.

Rearranging and dividing equation (7) by πq^2 gives the linear relationship

$$C_s \lambda^3 q^2 + 2\lambda\varepsilon = \frac{n}{q^2} \tag{10}$$

If the image is taken at strong under focus conditions ($\varepsilon << 0$) the integer **n** takes only negative values and thus Cs and ε can be unambiguously determined as a slope and intersect of linear regression in $q^2 <-> \frac{n}{q^2}$ coordinates.

Content of the practicum and consequent report: determination of spherical aberration coefficient C$_s$ and defocus value

Calculations will be conducted on Digital Micrograph, demo version of the program will be provided for installation on personal computers.

Experimental part

Alignment of the column.

Acquisition of Si[110] images for magnification calibration. Calibration should be performed at exactly the same imaging conditions as series acquisition afterwards: the same current of objective

lens and the same magnification ~x300K. Precise calibration of magnification is essential for this task as far as eq.(10) is written in the units of spatial frequencies, i.e. the scale of q should be known and the accuracy of default camera calibration is not enough.

Acquisition of the images of amorphous region. Due to the moderate dynamical range of CCD camera of the microscope the transfer of high spatial frequencies is suppressed by noise. In order to enhance oscillations of transfer function for high q (get more data points for better linear fit) it is advised to acquire multiple images at the same place and imaging conditions and average them afterwards. This will increase signal-to-noise ratio significantly. Images should be acquired at significant underfocus (~x3 Scherzer focus) in order to produce reasonable number of experimental points (minima and maxima of the radial intensity distribution).

Report should contain detailed description of experiment with explanation of the meaning of performed operations.

Calculation part

Calibration of magnification.

- Start Digital micrograph.
- Under the menu **File/Open** open the image showing the HRTEM image of Si[110].
- To calibrate the image go to Microscope/Calibrate image from diffractogram.
- Calibrate your image by following the instructions given by the Digital Micrograph software.
- Use the known values for Si reflections in the [110] orientation (d_{111}=0.314 nm, d_{200}=0.272 nm) to calibrate your image and remember to divide the value by two if you select a pair of reflections.
- After calibration of the image note down calibration value of the image in pixel in nm by right clicking on the image image display/calibration.
- Close the image.

Estimation of Cs and defocus.

- Open the experimental amorphous image stack
- Open the following Digital Micrograph scripts:
  - Polar projection.s
  - Stack align by correlation to average to accuracy and sum.s
  - Gaussian high-low pass filter.s
  - Hanning window.s
- Choose Rectangular ROI tool from a toolbox and select ROI dragging the mouse over the stack image with Alt pressed. Then run "Stack align by correlation to average to accuracy and sum.s" script (Ctrl+Enter) and wait until you get a drift corrected average image of amorphous region with reduced noise, this can take some minutes.
- Select 1Kx1K ROI and copy it to separate window (Ctrl+C, Ctrl+Alt+V). Run "Hanning

Window.s" script and apply 25% hanning in order to hide drift artifacts at the edges. Perform background subtraction by running Gaussian high-low pass filter.s with parameter equal to -100.

- Calibrate the image by entering the values you noted from the calibration part of the exercise (right click, image display/calibration)
- Calculate the FFT of this image.
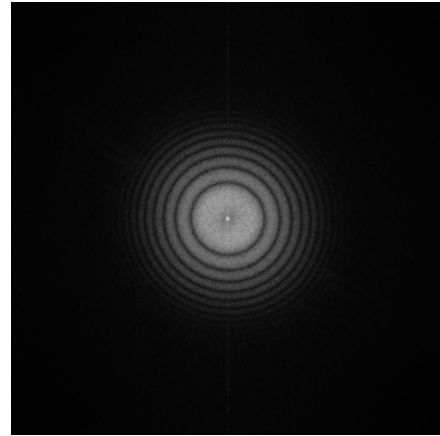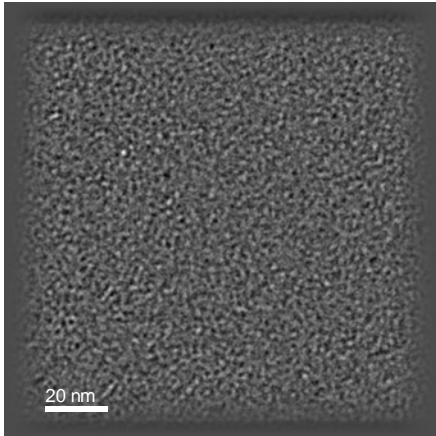
Hanned and calibrated amorphous image   Diffractogram of amorphous image

- Calculate power spectrum (diffractogram) by changing the FFT image from complex to real (Edit/Change data type/real/modulus). Perform background subtraction by running Gaussian high-low pass filter.s with parameter equal to -100.
- Open the script "Polar projection.s" and run it on the diffractogram, you should get a projection of the modumus of CTF function.
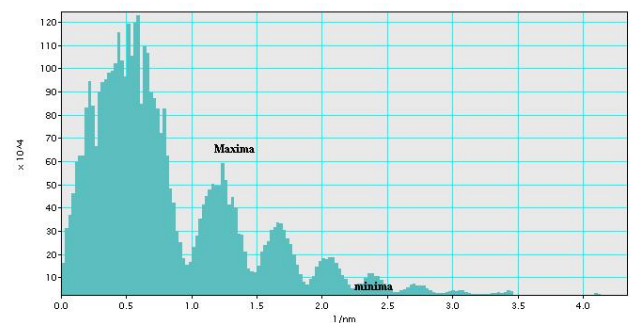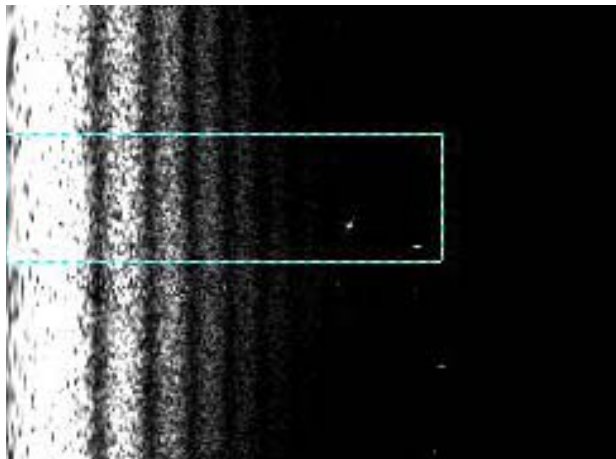
Example of polar projection of diffractogram.   Intensity profile of radial distribution.

- Calculate an intensity profile and integrate to obtain the maxima and minima values for the contrast transfer function CTF.
- Correctly assign the n values to each extremum, the wrong assignment will lead to non-linear plot.

- Draw a plot (in Excel or Origin) in accordance to eq.(10) and determine the values of defocus and Cs assuming a wavelength for 200kV electrons 0.0025 nm. Optionally estimate the value of astigmatism.

Report should contain step by step description of calculations illustrated by figures and resultion df and Cs values.

Relevant references:

[1] O.L. Krivanek, Optik 45 (1976) 97.

[2] W.M.J. Coene and T.J.J. Denteneer, Ultramicroscopy 38 (1991) 225.

[3] P.Buseck, J.Cowley, L.Eyring, High Resolution Electron Microscopy and Associated Techniques, pp. 546-548

[4] L.J.Allen, W.McBride, N.L. O' Leary, M.P Oxley, Ultramicroscopy 100 (1991) 91

[5] M.Kuzuya, M. Hibino, J.Electron Microsc. 30,2, (1981) 114-220

[6] K. Wong, E.Kirkland, P. Xu, R. Loane, J.Silcox, Ultramicroscopy 40 (1992) 139-150