Transmission Electron Microscopy
András Kovács and Hongchu Du
Ernst Ruska-Centre for Microscopy and Spectroscopy with Electrons, Peter Grünberg Institute (PGI-5)
Forschungszentrum Jülich

Contents

1. Transmission Electron Microscopy Fundamentals 2
   a. Overview 2
   b. Transmission Electron Microscopy Landmarks 3
   c. General features 4
   d. Simplified structure 5

2. Phase Contrast Imaging and Diffraction modes 7
   a. Bright Field Imaging 7
   b. Thinking in reciprocal space – Diffraction 9
   c. High-resolution TEM imaging 13

3. X-ray Energy-Dispersive Spectroscopy 16

4. TEM experiments 17

5. Submission of the lab report 18

6. Recommended readings 18
1. Transmission Electron Microscopy Fundamentals

a. Overview

Transmission electron microscopy (TEM), together with its associated techniques, is one of the most versatile characterization methods. In short, it can be described as analysis of the interactions between high energy electrons, usually between 60-300 keV, and a thin electron transparent sample. This interaction may generate a number of different signals, depicted in Figure 1, which can be used for composing images, diffraction patterns and spectroscopic analyses. Consequently, TEM techniques can provide information on the sample’s microstructure, chemical composition and electronic properties.

![Figure 1: Most common signals generated on the interaction of a high energy electron beam and a thin sample.](image)

As the TEM instrumentation allows the simultaneous acquisition of the signals generated during the electron beam and sample interaction, a comprehensive sample characterization is possible by the use of TEM.
b. **Transmission Electron Microscopy Landmarks**

The TEM related techniques development occurred as the natural evolution of optical microscopy techniques, especially due to the spatial resolution limitation from the latter due to the diffraction limit. A timeline containing some landmarks regarding the TEM conception includes:

1897 – J. J. Thompson discovers the electron.

1924 – L. de Broglie presents the wave-particle duality theory.

1927 – H. Busch describes the use of non-homogeneous electromagnetic fields to focus electrons.

1931 – E. Ruska presents the electron microscope concept and builds its first prototype, illustrate in Figure 2.


1939 – Siemens company produces the first commercial TEM.

**Figure 2:** First electron microscope prototype, by E. Ruska and M. Knoll.
1940 – W. Glaser and O. Scherzer present fundamental theorems on electron optics.

1943 – J. Hillier develops the Electron Energy-Loss Spectroscopy (EELS).

1951 – R. Castaing develops the X-ray Energy Dispersive Spectroscopy (XEDS).

1970 – The first High Resolution Transmission Electron Microscopy (HRTEM) images are obtained. The spatial resolution, about 0.4 nm, reveals details from the sample’s atomic structure.

1998 – Aberration corrected HRTEM achieves spatial resolution close to 0.1 nm.

2009 – Spherical (Cₜ) and Chromatic (Cₜ) aberration-corrected TEM with resolution down to 50 pm.

c. General features

- High spatial resolution: The main limiting factor on optical microscopy maximum spatial resolution (10⁻⁷ m scale) is related to the diffraction limit according to the Rayleigh criterion (θ ~ 0.61·λ), which limits the spatial resolution to the wavelength. The use of high-energy electrons on TEM, i.e., smaller wavelengths, enhances the achievable spatial resolution up to the 10⁻¹⁰ m scale.

- Simultaneous acquisition of different signals: As mentioned before, TEM instrumentation provides the possibility of simultaneous analysis of different signals generated during the interaction between electron beam and sample. Among the several characterization possibilities are the combination of imaging and electron diffraction for the local crystalline arrangement assessment, and the combination of imaging and spectroscopy techniques for the high spatial resolution chemical analysis of a particular region of interest on the sample.

- Sample preparation: Thin samples, typically below 100 nm thickness, are necessary for most of TEM techniques as the electron beam transmission through the sample is required. Preparing samples at this scale without changing their original features demands extremely controlled and time-consuming procedures.
- **Sampling**: Given its focus on high spatial resolution and the requirement of thin samples, TEM techniques usually deal with the analysis of a reduced sample volume. Consequently, extending the results from the local measurements to describe the whole sample features requires a careful analysis.

\[ \text{(1)} \]

\[ \text{d. Simplified structure} \]

TEM microscopes present a complex instrumentation given the requirement of a precise manipulation of high energy electrons. A simplified diagram is show in Figure 3 and the most important sections are briefly described on the following sections.

![Figure 3: TEM simplified instrumentation diagram.](image)

- **Electron Gun**: This system essentially includes an electron source, from which electrons are extracted by heating (thermoemission guns), by the application of a strong electrical field (field-emission gun) or their combination (Schottky gun), and an assembly of electric dipoles
which supply the acceleration voltage to the extracted electrons. This acceleration voltage typically ranges from 60 to 300kV.

- **Condenser and Scanning lenses:** This system comprises several electromagnetic dipoles and physical apertures, which combination provides the controlled electrons deflection along the column and the generation of an electron beam. The fine tuning of the condenser lens system can generate collimated (parallel) or convergent illumination modes with different spot sizes at the sample. In addition, the electron beam can be shifted and tilted in respect to the optical axis just before the sample by the scanning lenses, allowing then the Scanning Transmission Electron Microscopy (STEM) mode.

- **Sample holder:** The TEM grids are placed into the microscope using the sample holder. This system, together with the goniometer, can provide sample movement along 6 axes (Cartesian $xyz$, tilt $\alpha \beta$, and rotation $R$) inside the TEM column. Advanced experiments, such as *in situ* heating, cooling, and mechanical testing, can be performed during the TEM characterization with the aid of adequate sample holders and external control hardware.

*Figure 4.* A double-tilt TEM holder (Gatan) on a stage (left) and a magnified view of the specimen area (right).

- **Objective lens:** Although the objective lens delivers a magnification of only $\sim$20-50 times, it is the most important lens in the TEM. A high performance is demanded due to the fact that its optical aberration functions determine the resolution of the microscope. It forms the images and diffraction patterns that are magnified by the projector.
- **Signals detection:** The signals generated during the TEM analysis can be detected using several different devices. For the electrons detection, the most important are based on the combination of a scintillator, a photomultiplier and a couple-charge device (CCD). New detector designs allow direct electron detection. Other important detection devices are the X-rays spectrometer and the electron energy-loss spectrometer, which are the fundamental instrumentation for EDXS and EELS techniques, respectively.

2. **Phase Contrast Imaging and Diffraction modes**

   a. **Bright Field Imaging**

   The conventional TEM imaging mode, often referred as bright field imaging (BF), is based on the sample illumination by a collimated and broad electron beam. Figure 5 presents a simplified electron ray path diagram for the most common TEM configurations, BF and electron diffraction. In a simplified way, the adequate focusing combination from the objective and projector lenses determines the projected information at the detection plane, either the magnified image or diffraction pattern. The use of physical apertures at different positions along the TEM column after the sample may provide the image formation with selected diffracted components (dark field imaging - DF) or the selection of a given interaction area for the diffraction pattern (selected area electron diffraction (SAED)).
Figure 5: Simplified electron ray path diagrams for the TEM (a) bright-field mode and (b) selected area electron diffraction mode.

As the image formation on the BF mode is dependent on the electrons transmitted through the sample, the obtained images present mass-thickness contrast. For BF image the transmitted beam is selected by the objective aperture, while the DF image is obtained by positioning the aperture on a diffracted beam. Depending on the configuration of the objective aperture regarding the selection of diffracted components from electron beam, diffraction contrast can be obtained. Figure 6 present examples of BF and DF images of a semiconductor nanowire.
**Figure 6**: BF and DF images from an InAs nanowire. Different crystalline phases and/or orientations can be detected if different diffracted components are selected to generate the image.

(Image courtesy: J.C.G Perez and J. Bettini)

---

**b. Thinking in reciprocal space - Diffraction**

Diffraction patterns are images of the *reciprocal space*. A crystal in reciprocal space can be characterized as an array of points, which is uniquely defined for any given crystal, and each point is associated with a particular set of planes in the crystal. Figure 7 shows the incident wavefront (wavefront normal is $k_i$) being scattered by a plane of atoms to produce a diffracted wavefront (wavefront normal is $k_o$). When $\Theta$ equals to the Bragg angle $\Theta_B$, the Bragg law becomes

$$2 \sin \Theta_B = \frac{\lambda}{|K|}$$

so when we are at the Bragg angle, the magnitude of the vector $K$ has a special value, $K_B$.

---

* The Bragg’s law describe the scattering from a crystal lattice: $n\lambda=2d\sin\Theta$, where $n$ is an integer, $\lambda$ is the wavelength, $\Theta$ is the angle between the incident plane wave and scattered plane wave.
\[ |\mathbf{K}_s| = 1/d \]

where \( d \) is the interplanar distance from a given crystalline plane of the crystal. \( |\mathbf{K}_s| \) has units of nm\(^{-1}\) if \( \lambda \) is measured in nm.

**Figure 7.** (a) Scattering of electron waves on atomic planes, where \( \mathbf{k}_i \) is the incident electron wave normal and \( \mathbf{k}_d \) is the diffracted wave normal. (b) The scattering angle \( \sin \theta \) is defined as \( \mathbf{K}/2\mathbf{k}_i \).

An other important definition. Any vector in reciprocal space can be defined as a combination of the vectors \( \mathbf{a}^* \), \( \mathbf{b}^* \) and \( \mathbf{c}^* \) that are the unit-cell translations in reciprocal space. So the reciprocal vector \( \mathbf{g} \) of an atomic plane from the real space can be defined as

\[ \mathbf{g}_{\text{hkl}} = h\mathbf{a}^* + k\mathbf{b}^* + l\mathbf{c}^* \]

where \( h, k \) and \( l \) are integers and called as Miller indices. The definitions of Miller indices are shown in Figure 8. The definition of the plane \((hkl)\) is that it cuts the \(a\), \(b\) and \(c\) axes at \(1/h\), \(1/k\) and \(1/l\), respectively.
Figure 8. The plane ABC has Miller indices (hkl). The vectors OA, OB and OC have lengths a/h, b/k and c/l.

Crystals have different structures. There are seven lattice systems according to the axial system used to determine their structure such as are body-centered cubic (bcc), face-centered cubic (fcc), hexagonal, tetragonal, rhombohedral, orthorhombic, monoclinic and triclinic. The structure of the crystals imposes certain selection rules that determine which beams are allowed and which reflections will appear in the diffraction pattern. The selection rule is expressed through the structure factor (F) that is the unit-cell equivalent of the atomic scattering amplitude f(\(\mathbf{g}\)). The structure factor is defined as

\[ F_{hkl} = \sum_i f_i e^{-2\pi i (hx_i + ky_i + lz_i)} \]

The structure factor determines whether the reflection of a certain atomic plane appear in the diffraction pattern or not. For example, in the fcc lattice (see Fig. 9), the coordinates of the atoms are (x, y, z)=(0, 0, 0), (\(\frac{1}{2}\), \(\frac{1}{2}\), 0), (\(\frac{1}{2}\), 0, \(\frac{1}{2}\)), (0, \(\frac{1}{2}\), \(\frac{1}{2}\)). We substitute these values of (x,y,z) into equation of the structure factor then

\[ F = f \left\{ 1 + e^{\pi i (h+k)} + e^{\pi i (h+l)} + e^{\pi i (k+l)} \right\} \]

Remember h, k and l are all integers. If all three are either odd or even, then all of the exponential terms are \(e^{2n\pi i}\) and all the phases of the diffracted waves are multiples of 2\(\pi\) and

\(^\dagger\) Fcc and bcc are both cubic.
are in phase. However, if one of \( h, k \) or \( l \) is odd and the others are even (or the opposite), then two of the three phase factors will be odd multiples of \( \pi \) giving two terms of \(-1\) in equation for \( F_{hk} \). Thus the structure factor

\[
F = 4f \text{ if } h, k, l \text{ are all even or all odd,}
\]

\[
F = 0 \text{ if } h, k, l \text{ are mixed even and odd.}
\]

These rules determine the allowed reflections in the electron diffraction pattern of the \( fcc \) structure. In the same way, the allowed reflections for bcc, hexagonal close packed (hcp) or other structures can be determined.

**Figure 9.** (a) Body-centered and (b) face-centered cubic crystal structure. In bcc, the atoms are at \((0,0,0)\) and \((\frac{1}{2}, \frac{1}{2}, \frac{1}{2})\), while in fcc the atoms are at \((0,0,0)\), \((\frac{1}{2}, \frac{1}{2}, 0)\), \((\frac{1}{2}, 0, \frac{1}{2})\) and \((0, \frac{1}{2}, \frac{1}{2})\) positions.

The electron diffraction analysis is carried out after the measurements of both spacing and orientation of the recorded diffracted spots. As the recorded diffraction pattern is taken at the objective lens back-focal plane, the spots spacing refers to the scattering angles and, consequently, to the crystalline planes spacing, according to Bragg’s law. Figure 10 presents an example of electron diffraction pattern from a single crystalline and a poly-crystalline sample. Diffraction from polycrystalline sample can be viewed in the same way as X-ray diffraction from powders. For a completely random polycrystal, the reciprocal lattice is rotated about all axes and produces a set of spotted rings in the diffraction pattern, as shown in Fig. 10 (b).
Figure 10: Selected area diffraction patterns of (a) monocrystalline ZnO and (b) polycrystalline ZnO.

c. High-resolution TEM imaging

High-resolution TEM (HRTEM) images is based on the interference between two or more diffracted beams from the electron beam after the interaction with a thin sample. Crystalline materials present well defined diffraction angles, as described by the Bragg’s law, and their interference generates a periodic pattern corresponding to the sample atomic structure. However, HRTEM images cannot be readily interpreted, especially regarding the correlation of bright spots and the atoms positions as the scattering of the incident electron beam (a plane wave) by the atomic potentials in the sample does not only contain linear contributions (single scattering) but also multiple scattering events besides the instruments influence. Figure 11 presents an experimental HRTEM image, the projected crystallographic structure, and the calculated images for different objective lens excitation (defocus).
Figure 11: HRTEM image (left) of CeO₂ and the projected crystalline structure along the \langle110\rangle direction (right, top). The HRTEM images calculations for two defocus (d) present a contrast reversal, indicating thus that the direct correlation between higher intensity spots on HRTEM images cannot be directly correlated to the atomic positions.

The electron scattering in TEM is described assuming the incident electron beam as a plane wave, which amplitude and phase are altered during the interactions with the sample. Actually, round electromagnetic lenses act on electrons alike optical glass lenses on light. This is illustrated in Fig. 12, where the incoming plane electron wave is diffracted at the specimen. Electrons being diffracted by the same angle, i.e. the Bragg angle for crystalline objects, are focused in the back focal plane of the lens and form a diffraction pattern. The actual image observed in the image plane depends on two components: (i) the interaction of the electrons with the specimen and (ii) the electron optical imaging process by the electromagnetic lenses of the microscope itself. The incoming plane wave

\[ \Psi_0(x) = \exp(2\pi i k_0 \cdot x) \]

is scattered at the atomic potential \( V(x) \). At the bottom of the specimen the exit plane wave \( \Psi_f(x) \) is given by a superposition of plane waves:
\[ \Psi_t(x) = \int \Psi_g \exp \left(2\pi i(k_0 + g) \cdot x\right) dg \]

\( \Psi_g \) is the complex valued Fourier coefficients corresponding to the vector \( g \) in reciprocal space (or Fourier space), which is also called spatial frequency. For an ideal periodic object, the integral can be written as a sum over the reciprocal space vectors and their Bloch wave coefficients. For a non-periodic object, such as a defect in a crystal or an interface, the integral has to be solved numerically by solving the relativistically corrected Schrödinger equation.

**Figure 12:** Schematic illustration of the mode of operation of the objective lens. The specimen scatters the incoming plane wave. Beams scattered by the same angle are focused in the back focal plane of the lens and form an interference pattern in the image plane.

The exit plane wave is modified by the aberrations of the objective lens, which impose an additional phase factor \( \chi(g) \), the so-called aberration function:

\[ \tilde{\Psi}_t(x) = \int \Psi_g \exp \left(2\pi i(k_0 + g) \cdot x\right) \exp \left(-i\chi(g)\right) dg \]

This equation implies that:

(i) the high-resolution image is an interference pattern formed by superposition of plane waves;
(ii) the effect of the microscope can be mathematically expressed by a phase shift, which is a function of the spatial frequency.

Recording electron waves on the CCD camera means measuring intensities. Therefore, as a final result the product of the wave function and its complex conjugate is obtained, i.e. the phase information of the image wave is lost:

\[ I(x) = \Psi_t(x)\Psi_t^*(x) \]

The new generation of transmission electron microscope are able to correct and adjust the aberration functions using aberration correctors that open new ways to obtain directly interpretable images. Figure 13 presents the comparison between the images recorded from the same sample using a conventional TEM and an aberration-corrected TEM.

![Figure 13: Raw HRTEM images from the same CeO₂ nanocrystal from a (a) standard TEM microscope (1.25 Å information limit) and from a (b) spherical aberration-corrected microscope (0.8 Å information limit).](image)

3. **Energy-Dispersive X-ray Spectroscopy (EDXS)**

In Figure 1 we showed that the electron beam generates a characteristic X-ray signal. To do so the electrons must penetrate through the conduction/valence bands and interact with the inner-shell electrons. Above a critical energy, an electron escapes from the inner shell, leave a
hole in the inner shell, but most importantly ionizes the atom. The ionized atom can return to its lowest energy by filling the hole with an electron from the outer shell. This transition is accompanied by the emission of X-ray and an Auger electron. The process is shown in Fig. 14. The characteristic X-rays have a well-defined energy and a line width of few eV that can be used to determine the chemical composition of the sample as shown in Fig. 14 (b).

Figure 14. (a) The ionization process produced by the incoming electron beam that results in characteristic X-ray emission. The image is taken from Williams, Carter: Transmission Electron Microscopy textbook (Springer). (b) A typical EDXS spectrum.

4. TEM experiments

You will learn how to operate the transmission electron microscope working on a focused ion beam prepared electron transparent sample. The sample is a cross-section of a multi-gate substrate consisting of metallic electrodes embedded in SiO$_x$. Such device can be used to measure transport properties of nanowires that is usually positioned on the top. Please determine the structure factor of Si (see in task b) prior to the laboratory work.

Your tasks for today:

a. Record bright-field, dark-field, high-resolution images and selected area electron diffraction patterns during the experiments. What are the important experimental parameters?

b. Tilt the sample to the closest [hkl] zone axis. Record a selected-area electron diffraction pattern of Si. Can you determine the [hkl] zone axis? How the structure factor of Si would look like? A little help: the first group of Si atoms locate on fcc sites and the second bunch on the basis vector $[\frac{1}{4}, \frac{1}{4}, \frac{1}{4}]$. In Si, which reflections are forbidden?
c. Measure the composition of the layers using EDXS.

d. Determine the microscope constant ($\lambda L$) from the following expression

$$R \ d_{\text{hkl}} = \lambda L$$

where $R$ is the distance of the reflection(s) measured from zero beam, $d_{\text{hkl}}$ is lattice plane distance using $d_{\text{111}}$(Si) = 0.313 nm, $d_{\text{110}}$(Si) = 0.192 nm, $d_{\text{100}}$(Si) = 0.163 nm.

e. Using the microscope constant, determine the d spacing of the $d_{\text{100}}$(Si) = ? nm.

5. **Submission of the lab report.** Relevant only for students who do lab course within the “Fortgeschrittenen-Praktikum” of RWTH Aachen.

Please send the report (electronic or written) with the analysis to the following address:

a.kovacs@fz-juelich.de

Dr. András Kovács
Ernst Ruska Centre 1
Forschungszentrum Jülich
52425 Jülich

6. **Recommended readings.**