Phase Contrast
Lecture 6

• Scattering/Amplitude Contrast
  – BF/DF: thickness, tilt
  – Low-medium magnification
  – Applications
    • Crystal defects: dislocations, stacking faults, phase boundaries, precipitates, defect clusters
    • Contrast: \( g \), type of the fault, its depth in the crystal
    • Quantitative determination of \( b \)
    • Resolution: strongly excited: 10 nm; weak beam: 1 nm

• Phase Contrast:
  – High magnification
  – Highly coherent beam
  – Large defocusing
  – Resolution: 0.1-0.2 nm
  – Reliable interpretation: Simulation

What is contrast?

\[
C = \frac{I_2 - I_1}{I_1} = \frac{\Delta I}{I_1}
\]

• Eyes:
  < 5% - can’t detect
  <10% - difficult
• Enhance digital image electronically
Interference Effects

Fresnel fringes around a hole in a carbon foil

The discovery of carbon nanotubes

Single atoms and clusters of atoms of tungsten on the surface of a few atomic layers of graphite
Effect of two beams reaching the image

Aperture: two beams (the incident and one diffracted)

\[
\Psi = \exp(ik \cdot r) + \phi_g \exp(i\vec{k}' \cdot \vec{r}) = \exp(ik \cdot r) \left[ 1 + \phi_g \exp(i\Delta k \cdot \vec{r}) \right] = \exp(ik \cdot r) \left[ 1 + \phi_g \exp(ig \cdot \vec{r}) \right]
\]

Unit amplitude

\[
\phi_g = R \exp(i\delta)
\]

\[
R = \frac{\pi \sin(ts/2)}{\xi_g}
\]

\[
\delta = \frac{\pi}{2} - \pi s
\]

The total intensity distribution: \[
I = 1 + R^2 + 2R \cos(\vec{g} \cdot \vec{r} + \delta) = 1 + R^2 + 2R \sin \left( \frac{2\pi}{d} - \pi st \right)
\]

- Lattice resolved (the resolving power of the microscope?)
- At least one order + the incident
- More beams: sharpen the details
- The visibility: orientation and thickness

\[
st = 2n\pi \quad R = \frac{\pi}{\xi_g} \frac{\sin(ts/2)}{ts/2} = \frac{\pi}{\xi_g} \frac{\sin(n\pi)}{n\pi} = 0
\]

The fringe: not observable

- The position of the fringes: no simple relation to the position of atomic planes (Next page)
Lattice Fringes: Thickness and Orientation

Thickness variation changes the contrast: no simple relation to the atomic planes

(111) Lattice fringes
Sodium faujasite, d = 1.44 nm

Similar effects, when s changes
Summary of Theory

Phase shift arising:
• at the specimen ~ scattering
• the spherical aberration
• the defocus of the objective lens

Two or more beam recombined to produce intensity difference from point to point

Instrumental factors, phase plates

• High gun brightness
• Medium acceleration V, low Cs
• Parallel beam illumination: the illumination aperture < 0.1 mrad
• Highly stable
  - Accelerating Voltage ~ 1ppm: low energy spreading
  - Vibration-free mechanical stability
• Coma-free on-axis alignment
• Sample height and orientation
• Simulate through-thickness-through-focus

A coherent source of illumination

Very thin < 3nm
Phase Shift: Specimen

- Path difference relative to the wave front in vacuum

\[ \phi_s(r) = \frac{2\pi}{\lambda} \int_{-\infty}^{\infty} [n(r) - 1] dz \approx \frac{2\pi}{\lambda E} \frac{E + E_0}{E + 2E_0} \int V(r) dz \]

Variation in \( n \) from point to point across the specimen \( n(r) \propto \) atomic potential

- The exit wave amplitude:

\[ \psi(r) = \alpha_s(r) \exp[i\phi_s(r)] \]

Ignore the local amplitude modulation: \( \alpha_s \approx 1 \)

\[ \psi(r) \approx 1 - i\phi_s(r) = 1 - i\sigma \phi_p(r) \]

\[ \sigma = \frac{2\pi me}{h^2} \]

\[ \phi_p(r) = \int_{-\infty}^{\infty} V(r) dz \] The projected specimen potential
Lack of Contrast at exact focus

Empirical methods to enhance the contrast of images

- Reducing the size of the objective aperture – limited resolution
- Introducing a focusing error (defocus) – Usual method in HRTEM
- Simple interventions in the BFP (Schlieren contrast) – Appeared from time to time
- Use of BFP phase plates – Optical...

\[
I = |\psi_i + i\psi_{sc}| \approx |\psi_i|^2 - 2\psi_i\psi_{sc} < I_0
\]

For thin sample, \( \psi_{sc} \ll \psi_i \)

The phase object is invisible

Knife edge blocking half of the transverse spatial frequencies

A phase shift of 90° between the direct and scattered waves (thin sample)
Phase shift: Instrumental Factor

Spherical aberration:

- Wave fronts: equal phase
- Rays: trajectories orthogonal to the wave fronts

Two rays: \( R, R + dR \)

- Optical path difference due to the angular deviation
  \[ ds = \varepsilon dR \]
- Total path difference or the phase shift relative to the optical axis

\[
W(\theta) = \frac{2\pi}{\lambda} \Delta s = \frac{2\pi}{\lambda} \int_0^R ds = \frac{2\pi}{\lambda} \int_0^R \varepsilon dR
\]

Part of the outer zone of a lens at a distance \( R \) from the optical axis

Angular deviation!!
Phase shift: Instrumental Factors

Spherical aberration

Change of specimen position

Change of focal length

Scherzer formula

Total angular deviation: \( \varepsilon = \varepsilon_s + \varepsilon_a + \varepsilon_f \)

The phase shift: \( w(\theta) = \frac{2\pi}{\lambda} \int_0^\theta \varepsilon d\theta = \frac{2\pi}{\lambda} \left[ \frac{1}{4} C_s R^3 - \frac{1}{2} \frac{(\Delta f - \Delta a) R^2}{f^2} \right] \approx \frac{2\pi}{\lambda} \left[ \frac{1}{4} C_s \theta^3 - \frac{1}{2} \Delta z \theta^2 \right] \)

\( \theta = \frac{R}{f} \)

\( \Delta z = \Delta f - \Delta a > 0, \text{ underfocus} \)
Phase shift: Instrumental Factor

The wave aberration: $C_s$ and $\lambda$

Introducing reduced coordinates

$$\theta^* = \theta \left( \frac{C_s}{\lambda} \right)^{1/4} ; \quad \Delta z^* = \Delta z (C_s \lambda)^{-1/2}$$

The reduced wave aberration:

$$\frac{W(\theta^*)}{2\pi} = \frac{\theta^{*4}}{4} - \frac{\theta^{*2}}{2} \Delta z^*$$

Scattered waves are shifted with a phase of 90°.

Scherzer focus: $\Delta z^* = 1$

$W$ has the value 90° over a relatively broad range of scattering angles
Plane wave: \( \psi = \psi_0 \exp(ik \cdot \vec{r}) \)

Exit wave: \( \psi_e(\vec{r}) \sim \psi_0 a_s(\vec{r}) \exp[i\varphi(\vec{r})] \exp(ik \cdot \vec{r}) \)

- \( r \): radius vector in the specimen plane

Fourier transform

\[
F(\vec{q}) = \int_{S} \psi_e(\vec{r}) \exp(-i\vec{q} \cdot \vec{r}) dS
\]

- \( q \): radius vector in the diffraction plane

Inverse Fourier transform

\[
\psi_m(\vec{r}) = \frac{1}{M} \int_{S} F(\vec{q}) H(\vec{q}) \exp(i\vec{q} \cdot \vec{r}) d^2q = \frac{1}{M} \psi_s(\vec{r})
\]

- \( H(\vec{q}) = \exp[-iW(\vec{q})]M(\vec{q}) \)

Pupil function: aberration/aperture/defocus
Contrast-Transfer Function

CTF: The imaging properties of an objective lens
  • Independent of any particular specimen structure

A specimen with a single spatial frequency \( q = 1/d \)

The exit wave: \( \psi_s(x) = 1 - \varepsilon_q \cos(2\pi qx) + i\varphi_q \cos(2\pi qx) + ... \)

The Fourier transform: \( F(\pm q) = \frac{1}{2}(-\varepsilon_q + i\varphi_q) \)

The image: \( \psi_m(x') = 1 + \sum_{\pm q} \frac{1}{2}(-\varepsilon_q + i\varphi_q) \exp[-iW(q)] \exp(2\pi i qx') = 1 + (-\varepsilon_q + i\varphi_q) \exp[-iW(q)] \cos(2\pi qx') \)

The intensity: \( I(x') = |\psi_m(x')|^2 \)
  \[ = 1 - 2 \cos W(q) \varepsilon_q \cos(2\pi i qx') + 2 \sin W(q) \varphi_q \cos(2\pi i qx') + ... \]
  \[ = 1 - D(q) \varepsilon_q \cos(2\pi i qx') - B(q) \varphi_q \cos(2\pi i qx') \]

  • \( D(q) \): the CTF of the amplitude structure of the specimen
  • \( B(q) \): the CTF of the phase structure

\[ B(\theta^*) = -2 \sin W(\theta^*) = -2 \sin \left[ \frac{\pi}{2} \left( \theta^{*4} - 2\theta^{*2} \Delta^* \right) \right] \]

Ideal CTF: \( B = 2 \) for all \( \theta^* \)
• Scherzer resolution/point resolution
  The first zero: The maximum k for which phase coherency is preserved at Scherzer defocus
  \[ d_p = \sqrt[3]{\frac{C_s \lambda^2}{6}} \]
Influence of Energy Spread and Illumination Aperture

Energy spreading: $\Delta E = 1-2$ eV (thermionic); 0.3-0.5 (Field Emission)
Finite size of the gun: illumination aperture $\alpha_i$. ($\alpha_i < \alpha_o$, partially spatially coherent)

$\Delta E$ results in $\Delta f$: electrons with different $\Delta f$ superposed incoherently at the image

$$I(x') = 1 - B(q)K_c(q)\varphi_q \cos(2\pi qx')$$

Envelope function: $K_c(q) = \exp\left[-\left(\frac{\pi\lambda q^2 H}{4\sqrt{2}}\right)^2\right]$

$$H = C_c \frac{\Delta E}{E} \frac{1+ E/E_0}{1+ E/2E_0}$$

Damps the CTF for increasing q

Information limit: the damped CTF goes to 0
- Chromatic aberration
- Energy spread
- Fluctuation in high voltage
- Beam convergence
- Spatial incoherence
Recap: CTF

- **Contrast Transfer Function (PCTF)**
  - **Universal curve**
    - Independent of specimen and microscope
    - Oscillatory nature
    - Depending on:
      - Envelope function: Coherent/partially coherent illumination
        - Temporal: Focal spread
        - Spatial: finite divergence
        - Not affect the positions of the zeroes
  - **Focus**
  - **Scaling factor used for specific objective lens/beam energy**
Resolution Limit

- Interpretable image resolution: structural/point res
  - At optimum (Scherzer) defocus: PCTF largest band of spatial frequencies without any phase reversal — the first zero

- Instrumental resolution: information limit
  - Envelop functions: cut-off 15% for image processing
  - Fine details present, not directly interpretable
  - Focal Series Reconstruction

- Lattice-fringe resolution: finest spacing's that can be obtained ...... obsolete
  - Fine structures may not contain atomic information
Lattice and atomic imaging

• Comparatively very few lattice images are directly interpretable in terms of atomic arrangement
  – Phase reversal: difficult to achieve optimum image defocus
  – Best focus can be achieved by using:
    • edge for Fresnel/a defect
    • Knowledge of focal step sizes

• Small-unit-cell materials
  – A few diffracted beams contributing
  – Fourier/Self-images: identical images occur periodically in defocus \( 2d^2/\lambda \)
  – Contrast reversal by a half period

• Thick samples
  – Dynamical complexity
  – In the vicinity of the thickness extinction contour
    • Low direct beam
    • Strong diffraction between diffracted beam: dominating the image contrast
    • Dumbbells... different from the projected atomic separations ... except for highly specific thickness and defocus... cannot be interpreted as atomic positions
Application: an example

Accommodation of lattice misfit:
Stress relieve results in: interfacial misfit dislocations

Cross section of
Ge/Si(001)
heterointerface

Apendix
Outline

• High Resolution TEM
  – Elementary principles of Phase Contrast
  – Weak-phase Object Approximation
  – Multi-slice Method
  – Pseudo-weak-phase Object Approximation
  – Resolving Power

• Applications of HRTEM
  – Identify crystal structures
  – Microstructure and defects

• Examples of HRTEM

• Recent Progress of HRTEM
Brief History of HRTEM

• 1949 Scherzer: Fundamental HRTEM Contrast Theory
• 1956 Menter: Experimental observation
• TEMs: Mechanical, thermal and electrical stabilities
  – 1930s, exceeded the resolution of VLM
  – 1970s, the first ‘structure images’
  – 1980s, 0.2 nm, interpretable resolution limits
  – Mid-1990s, FEG – information limit 0.1 nm