1. The imaging process

As a first approximation, we consider coherent image, i.e. a plane parallel, monochromatic electron wave falls in the object, and is scattered, the scattered waves interfere and recombined by an electron lens (objective lens) to form an image.

2. The process consists of two parts: (a). The interaction of the incident wave with the object, defining the exit wave from the object, transmission function \( q(x) \); (b) the action of the objective lens in forming the image, transfer function \( t(x) \).

3. The essential question: how is the images intensity distribution related to the object structure?

Phase contrast images

- Contrast in TEM images can arise due to the differences in the electron waves scattered through a thin specimen.
- This phase contrast mechanism can be difficult to interpret because it is very sensitive to many factors such as specimen thickness, scattering factors, and properties of lens, etc.
- Phase contrast imaging can be exploited to image the atomic structure of thin specimens.
- The most obvious distinction between phase contrast imaging and other forms of TEM imaging is the number of beams collected by the objective aperture or an electron detector.
- For BF/DF image only requires a single beam selected by OA for imaging.
- A phase-contrast image requires the selections of more than one beam. In general, the more beams collected, the higher the resolution of the image.

7 spots are used for imaging the lattice
Beam condition of phase contrast image

Beam condition of diffraction contrast image
Only two diffraction spots were used for imaging the GaAS lattice showing one set of fringes from the (111) plane (horizontal). Only (111) diffraction contributed the image. The vertical crystal planes are invisible. The information retracted by image is not complete.
Many beams were selected by an aperture (ring) to form image showing more information about the structure of specimen.

- the question is where are the atoms with respect to the bright and dark contrast dots?
- Do the dark spots or bright spot correspond what atoms (Ga or As), etc.?
- Beautiful image sometimes is difficult to interpret.
Incident beams

2. Abbe theory

- On the BFP of the lens, the Fraunhofer diffraction pattern is formed.
- Rays scattered at angle $\theta$ come together at position $x=f\theta$ (small angle approximation) in BFP

\[ \psi(x) = F[\Psi(u)] = F[Q(u) \cdot T(u)] \]

\[ = q(x) \ast t(x) \]

so image intensity

\[ I(x) = |q(x) \ast t(x)|^2 \]

t(x) is objective lens transfer function

- For easy interpretation, we must have special objects with simple relation of q(x) to structure

1-D illustration of Abbe theory

- Fraunhofer diffraction
- BFP, and aperture
- Objective lens
- q(x)
- specimen
- Image plane
- Q(u) Diffraction pattern
- \(\Psi(x)\) image amplitude
2-D wave functions

\[ Q(uv) = \int q(xy) \cdot e^{2\pi i(ux + vy)} \, dx \, dy \]
\[ = F[q(xy)] \]
\[ \Psi(uv) = Q(uv) \cdot A(uv)e^{i\chi(uv)} = Q(uv) \cdot T(uv) \]

\[ \chi(uv) = \pi \Delta f \lambda (u^2 + v^2) + \frac{1}{2} C_s \lambda^3 (u^2 + v^2)^2 \]

\[ u = \frac{\theta_x}{\lambda}, \quad v = \frac{\theta_y}{\lambda} \]

\[ I(xy) = |q(xy) \cdot t(xy)|^2 \]

*For easy interpretation, we must have special objects with simple relation of q(xy) to structure*
the phase of incident electron wave is altered by specimen and the objective lens.

Phase contrast image is an interference pattern between the forwarded-scattered and diffracted electron waves from specimen.

Interference patterns require close attention of the phases of the wave.

The specimen is approximated as an object that provides phase shifts to the electron wave-front, sometimes in proportion to its scattering potential, since the specimen (crystal) is the packs of atom columns.

The atom columns in crystal act as the phase grating when the planar incident electron waves pass the specimen.

Phase contrast image is the projection of atom columns by combining the exit electron waves already interfering with specimen and objective lens.

A 3D perspective view looking down the (110) direction of Si. Each Si atom is drawn as shaded hard sphere.
Phase contrast image is an interference pattern between the forward-scattered and diffracted electron waves from specimen.

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A HRTEM image showing Si (110) projection
The single atom or atom columns have potential distribution, which interferes with incident electron waves, causing the phase shift of e-beam.
Phase shift by a single atom due to the potential

Phase shift by the atom columns

The crystal acts as a phase grating filter. The contrast produced by this mechanism is called phase contrast
The image is formed based on the amplitude distribution of exit waves, showing the projection of atom columns and locations of atoms.
A surface profile image of CeO2. The image conditions are such that columns of atoms appear as black dots. The direction of viewing is along a [110] direction of the crystal. The specimen is only about 2.5 nm thick.
3. Scattering approximations for validness of this model
- small-angle approximation: scattering angles < \(10^{-2}\) radians.
- Weak scattering approximation: scattered amplitude << incident amplitude
- Fails for heavy atoms, and for 1000 nm carbon film

4. Thin object approximation (phase object approximation) (thickness 100-200 nm and >100 kV -> low accuracy; 30-40 nm and >100 kV -> high accuracy)

the refractive index for the electron wave, \(n\)
\[ n = 1 + V(r)/2E \]

\(V(r)\) is the potential distribution of material.

\(E\) : applied electric field

if \(V(xy)\) is the potential projection along \(z\) direction
so the transmission function is
\[ q(xy) = e^{-i\sigma V(xy)}, \text{ and } \sigma = \frac{\pi}{\lambda E} \]

and then the diffraction amplitude is
\[ Q(u, v) = F[q(xy)] = F[e^{-i\sigma V(xy)}] \]

if \(\sigma V(xy)\) is small
\[ e^{-i\sigma V(xy)} \approx 1 - i\sigma V(xy) \]

this is valid for WPOA (weak phase object approx.)
diffraction pattern : \(Q(u, v) = \delta(u, v) - i\sigma \Phi(u, v)\)

- WPOA shows the crystal acts as phase grating filter. The phase of incident e-beam will be modified by the projected crystal potential.
- The variation of the projected crystal potential causes the changes of e-beam phase.
- The contrast is called phase contrast.
5. Imaging in WPOA

Imaging function

\[ \psi(x) = (1 - i \sigma V(x)) * t(x) \]

where \[ t(x) = F[T(u)] = F[A(u) \cdot e^{i\chi(u)}] \]

\[ = F[A(u) \cdot (\cos \chi(u) + i \sin \chi(u))] \]

Let \[ c(x) = F[A(u) \cdot \cos \chi(u)] \]
and \[ s(x) = F[A(u) \cdot \sin \chi(u)] \]

so \[ \psi(x) = (1 - i \sigma V(x)) * (c(x) + is(x)) \]

\[ I(x) = \psi(x) * \psi(x) &= |\psi(x)|^2 \]

ignore high order term, \( \sigma^2 \)

\[ I(x) = 1 + 2\sigma V(x) * s(x) \ldots \ldots (a) \]

The effect of equation (a) is that multiplying diffraction pattern in BFP by the phase contrast transfer function

\[ S(u) = F[s(x)] = A(u) \sin \chi(u) \]
Phase contrast transfer function

- the phase contrast transfer function (PCTF) is oscillatory, showing maximum transfer of contrast when $\chi(u) = \pm \pi/2$, and zero contrast when $\chi(u) = \pm \pi$
- When $S(u)$ is negative, positive phase contrast results, meaning that atoms would appear dark against a bright background.
- When $S(u)$ is positive, negative phase contrast results, meaning that atoms would appear bright against a dark background.
- When $S(u) = 0$, there is no detail in the image for this value of $U$
\( \Delta f = -58 \text{nm}, C_s = 1.2 \text{ mm}, \) and \( E = 300 \text{ kV} \)

- \( \sin \chi \) starts at 0 and decreases. When \( u \) is small, the \( \Delta f \) term dominates.
- \( \sin \chi \) first crosses the \( U \)-axis at \( u_1 \), and then repeatedly crosses the \( u \)-axis as \( u \) increases.
- \( \sin \chi \) can continue forever, but in practice, it will be modified by other functions (damping effects).
- Once the microscope is selected, the transfer function is fixed independent of specimen.
• $\Delta f = 0$, in the Gaussian defocus. No feature can be observed.
If we fix Cs and E, u increases as ∆f decreases achieving a higher spatial resolution.

Further decreasing ∆f will cause u suddenly jump to cross the u-axis and decrease u.
• If we fix $\Delta f$ and $E$, $u$ increases as $Cs$ decreases achieving a higher spatial resolution.
• Higher spatial frequencies depends on the large diffraction angles and $Cs$ effects become large
• The presence of zeros in the CTF (contrast transfer function) curve means that we have gaps in the output spectrum which do not contribute to the output signal: it is as if these frequencies were filtered out.
• the best CTF is the one with the fewest zeros
Scherzer defocus

- the CTF could be optimized by balancing the effect of spherical aberration against a particular negative of $\Delta f$, called Scherzer defocus.
- At this defocus, all beams will have nearly constant phase out to the first crossover of the zero axis. This crossover point is defined as the instrumental *resolution limit*.
- This is the best performance that can be expected from a microscope unless we use imaging processing method to extract more information.
- The implications of resolution require as many beams as possible being transferred through the optical system with identical phase.
\[ \chi(u) = \pi \Delta f \lambda u^2 + \frac{1}{2} \pi C_s \lambda^3 u^4 \]

\[ \frac{d\chi(u)}{du} = 2\pi \Delta f \lambda u + 2\pi C_s \lambda^3 u^3 \]

let \( 2\pi \Delta f \lambda u + 2\pi C_s \lambda^3 u^3 = 0, \Delta f + C_s \lambda^2 u^2 = 0 \) ....(a)

as shown in figure, when \( \chi = -\frac{2}{3} \pi \), sin \( \chi \) will be near -1

so \( -\frac{2}{3} \pi = \pi \Delta f \lambda u^2 + \frac{1}{2} \pi C_s \lambda^3 u^4 \) ....(b)

combining (a) and (b)

Scherzer defocus

\[ \Delta f_{Sch} = -\left( \frac{4}{3} C_s \lambda \right)^{\frac{1}{2}} \]

Scherzer resolution

\[ r_{Sch} = 0.66 C_s^{\frac{1}{4}} \lambda^{\frac{3}{4}} \]

Based on the electron probe size

\[ d_p^2 = d_g^2 + d_s^2 + d_c^2 + d_d^2 \]

Resolution

\[ \alpha_{optimum} = 0.9 \left( \frac{\lambda}{C_s} \right)^{\frac{1}{4}} \]

\[ d_{min} = 0.8 C_s^{\frac{1}{4}} \lambda^{\frac{3}{4}} \]
• The plots of $\sin \chi(u)$ as a function of $u$ could extend out as far as you want to plot them. In practice, they don’t because of the envelop damping function, i.e. the microscope is unable to image the finest detail due to reasons other than the simple transfer characteristics of a linear system.

• **Envelop damping function** results in the information retrieval limit or instrumental resolution limit. If we set up the Scherzer defocus or damping function, i.e. whichever equals zero first, the phase-contrast images are directly interpretable.

• Beyond the limit, we have to use computer simulation for interpretation.
Envelop Damping Function

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- The effects of envelop damping function could be arisen from chromatic aberration, and energy spread in focal length producing the Gaussian spread in the focus plane, and thereby introducing the Gaussian damping function \( ACc(u,v) \)

- In addition, the partial coherence of beam, sample drift, sample vibration, detector delocalization, etc. introduce the envelop damping effect.
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Imaging intensity in real space

\[ I(x) = 1 + 2\sigma \phi(x) * s(x) \]  

\[ S(u) = F[s(x)] = E(u)A(u) \sin \chi(u) \]

\( E(u) \) is the envelop damping function

Imaging intensity in Fourier space

\[ F[I(x)] = \delta(u) + 2\sigma F(u)E(u)A(u) \sin \chi(u) \]

\( F(u) \) is actually the structure factor
Imaging using passbands

• using minimum contrast defocus as a reference point, we can adjust the defocus to Scherzer defocus.

• As shown in figure, the passband condition settings for CTF allow higher spatial frequencies to contribute to the image, and X is constant or dX/du is small over a range of u which includes the reflection of interest.

The minimum contrast defocus condition
• all contrast is minimized and cannot see anything

in minimum contrast condition:

\[ \sin \chi(u) = 0.3 \]

and

\[ \Delta f_{MC} = -0.44(C_s \lambda)^{1/2} \]
As shown in above figure, we may obtain a pretty picture using high order passbands settings, but it may not give a true structure image of specimen.

at the high-order passbands setting, imaging the specimen is beyond the instrumental resolution limit so we cannot use the intuitive approach for image interpretation.

we should exactly know where the zeros are in the CTF, and use computer simulation to assist the image interpretation.
**Experimental considerations**

- Whenever you are using HRTEM imaging, you must first ask what information you are hoping to obtain.

- Lattice fringes images show lots of straight lines but tell you nothing about the atomic locations. These fringes are giving you information about the crystal orientation on a very fine scale.

- All interpretations and treatments of HRTEM imaging are based on the specimen behaving as weak-phase object. Most specimens of the interest are **too thick**.

- To be really sure that you have correctly interpreted the image, the match between experimental and simulated images should be good over a range of thickness and defocus values.

- To obtain a phase contrast image with atomic resolution, the following procedures are important.

  1. Choose an instrument with low Cs and small $\lambda$.
  2. Good alignment of beam and **comma-free alignment (only for HRTEM)**, and good experimental environment.
  3. Work in thin, flat, and clean regions of the specimen.
  4. Orient the specimen so that beam is aligned along a zone axis.
  5. Correct the astigmatism using optical diffractogram
  6. Find the minimum contrast setting and record a through-focus series.
Experimental considerations

8. Simulated and process the image using available computer program.

9. For HRTEM, it is critical that incident beam is precisely parallel to the optic axis of the microscope. If not, we will see the comma aberration.

10. Comma free alignment involves alternatively applying equal and opposite beam tilts to the incident beam; If there is a residual beam tilt of the incident beam away from the optic axis, then one image will look more distortion than the other. Adjust the beam-tilt controls until both tilted images look equally distorted. Repeat this procedure for the orthogonal direction.

Examples of HRTEM images

HRTEM image of a catalyst surface showing ceria and palladium nano-particles with different morphologies and with different atomic periodicities. Ceria has a large unit cell (58 atomic number) than palladium. The large 0.34 nm fringe spacing is used to identify the ceria crystallites. The ceria nano-crystals are distinctly faceted.
HRTEM image of $\gamma/\alpha_2$ interface in a TiAl alloy. This is a coherent interface between a DO$_{19}$ $\alpha_2$-Ti$_3$Al precipitate plate (bottom) in a L1$_0$ $\gamma$-TiAl matrix (top). There is a matching of the close-packed planes in both crystals, $\{111\}_\gamma \parallel (0001)_{\alpha_2}$, and the interface plane is parallel to these close-packed planes. Burgers circuits are drawn around the partial dislocations at opposite ends of a ledge. The symbols $S_e$, $S_s$ and $F$ indicate the start and finish points of 90° (edge) and 30° (screw) circuits, respectively.
Grain boundary
Phase image showing a ,110, projection of KI incorporated within a 1.6-nm-diameter SWNT, reconstructed from a focal series of 20 images. SCIENCE VOL 289 25 AUGUST 2000
Multiwall nano-tube