Chapter 3
Basic Crystallography and Electron Diffraction from Crystals

Lecture 16
Outline

• Kikuchi Line and its indexing
• Double diffraction
• CBED pattern (convergent beam electron diffraction)
Conventional high energy electron diffraction relies on elastic scattering. However, in a thick enough specimen, inelastic scattering will also take place. Inelastically scattered electrons travel in all directions but their distribution peaks in a forward direction, as shown in Figure (a). Note that in reality, the scattering is occurring in 3 dimensions.

More electrons are scattered forward than sideways. This contributes a grey background around the central spot of the diffraction pattern, as shown in Figure (b).
(1). Electrons which have been inelastically scattered can subsequently be diffracted, but only if they are now traveling at the Bragg angle, $\theta_B$ to a set of planes, \{hkl\}.  
(2). Two sets of electrons will be able to do this - those at $(+\theta_B)$ and those at $(-\theta_B)$, as seen in Fig. (a).  
(3). This diffraction results in intensity changes in the background. Because there are more electrons at A than B, **(since electrons passing through A are closer to the incident direction than those through B)** one bright line is developed (the excess line) together with one dark line (the deficit line).  
(4). Because the electrons are inelastically scattered in all directions, the diffracted electrons will form a cone, called Kossel cone, not a beam. Hence we observe Kikuchi lines - not Kikuchi spots, as seen in Fig. (b).  
(5) The spacing of the pair of Kikuchi lines is the same as the spacing of the diffracted spots from the same plane. However, the position of the lines is very sensitively controlled by the orientation of the specimen and Kikuchi lines are often used to set the orientation of a crystal in the TEM to an accuracy of 0.01 degrees.
(6) Kikuchi (1928) described this phenomenon before the development of the TEM; it can occur in any crystalline specimen.

(7) Kikuchi lines are useful for precise determination of specimen in a TEM. When we tilt the specimen, we tilt its reciprocal lattice. Tilts of the reciprocal lattice with respect to a stationary Ewald Sphere do not cause any substantial changes in the positions of the diffraction spots, but the individual spots grow or fade in intensity.

(8) The positions the Kikuchi lines are extremely sensitive to the tilt of the specimen. During a tilt, the Kikuchi lines moves as if they are affixed to the bottom of the crystal. With a long camera length typical for diffraction work, there is significant movement of the Kikuchi lines on the viewing screen.
This simulation shows Kikuchi lines moving in relation to the diffracted spots when the crystal is tilted through small angles.

Tilt angle 20°

Tilt angle 10°
Kikuchi line images
Some Kikuchi line micrographs for silicon are shown.

(a) The Kikuchi lines pass straight through the transmitted and diffracted spots. The diffracting planes are therefore tilted at exactly the Bragg angle to the optic axis.

- The ideal specimen thickness will be such that we can see both the spot pattern and the Kikuchi lines as seen in Fig. (a). This is one of the few situations when thinner is not necessarily better. In most cases, the specimen is the thinner and the better.
(b) The crystal has now been titled \textit{slightly away} from the Bragg angle, so that the Kikuchi lines no longer pass through the transmitted and diffracted spots.
(c). The crystal is tilted so that **more than one set of planes** are diffracting. Each set of diffracting planes has its own pair of Kikuchi lines.
Indexing Kikuchi Lines

The separation between the two Kikuchi lines is the same as the separation between the (hkl) diffraction spot.

(a) Only Diffraction Spot Pattern

(b) Diffraction Spot Pattern with Kikuchi Lines
Indexing Kikuchi Lines

The separation between the two Kikuchi lines is the same as the separation between the (hkl) diffraction spot and the (000) spot.

We can index the Kikuchi lines by measuring their separations in much the same ways as we index diffraction spots. Consider two different pairs of Kikuchi lines from the planes \((h1k1l1)\) and \((h2k2l2)\). The separations between their pairs of excess and deficit lines, \(p1\) and \(p2\), are in the ratio:

\[
\frac{p_1}{p_2} = \frac{\sqrt{h_1^2 + k_1^2 + l_1^2}}{\sqrt{h_2^2 + k_2^2 + l_2^2}}
\]

Figure shows ratios of \(\sqrt{32}\) and \(\sqrt{8}\) for indexed (440) and (220)
Indexing Kikuchi Lines

The angles between intersecting Kikuchi line pairs are the same as the angles between their corresponding diffraction spots, at least so long as the Kikuchi line are not too far from the center of the view screen. These angles are helpful for indexing Kikuchi lines in the same way that the angles between pairs of diffraction spots are useful for indexing diffraction patterns. For example the angle, $\Phi$, between the (220) and (400) Kikuchi line in left figure is:

$$\Phi = \arccos \left[ \frac{1}{\sqrt{8}} [220] \cdot \frac{1}{\sqrt{16}} [400] \right]$$

$$= 45^\circ$$
Indexing Kikuchi Lines and constructing Kikuchi lines

For a crystal oriented precisely on a zone axis, we can generate an indexed Kikuchi line pattern from its indexed diffraction. Each (hkl) Kikuchi line is drawn perpendicularly to the line between the (000) and (hkl) diffraction spots, bisecting this line. The procedure is shown in figure.

Kikuchi line, (-400), bisects the line between (000) and (-400)

Kikuchi line, (400), bisects the line between (000) and (400)

Diffraction Spot Pattern with Kikuchi Lines
Specimen Orientation and Deviation parameter (s)
The positions the Kikuchi lines are extremely sensitive to the tilt of the specimen. During a tilt, the Kikuchi lines move as if they are affixed to the bottom of the crystal. With a long camera length typical for diffraction work, there is significant movement of the Kikuchi lines on the viewing screen. The Kikuchi lines can be used to determine the sign and magnitude of the deviation parameter, s, which quantifies how accurately the Laue condition is satisfied.

Deviation Parameter, s:
- $x$ is the distance between the diffraction spots and its corresponding bright Kikuchi line (E-line)
- $r$ is the distance between the (000) and (hkl) diffraction spots.
- $\lambda$ is the wavelength
- The unit of s is Å⁻¹ or nm⁻¹

\[
S = \frac{x}{k/r} = \frac{x}{g^2 \lambda}
\]
The sign of $s$

- $s$ points from the Ewald sphere to the reciprocal lattice point.
- For Kikuchi line, $s=0$, when the Kikuchi line runs exactly through its corresponding diffraction spots.
- $s<0$ if the excess line lies inside its corresponding diffraction spot $g$. In this case the reciprocal lattice point lies outside the Ewald sphere.
- $s>0$ if the excess line lies outside its corresponding diffraction spot $g$. In this case the reciprocal lattice point lies inside the Ewald sphere.
\[ \Delta k = g + s \]

Diagram showing the relationship between \( k, g, \) and \( s \) with angles \( \theta \) and \( \alpha \). The diagrams illustrate different configurations for \( s_g = 0, s_g < 0 \), and \( s_g > 0 \).
Kikuchi Map

- As we tilt the sample, the diffraction spots fade or grow in intensity, always at the same positions.
- The Kikuchi lines move with the tilt, as if they were fixed to the crystal planes of sample.
- If we know which Kikuchi lines are on the screen, we can use the Kikuchi lines as “roads” to “drive” the tilt from one spot pattern (crystal zone axis) to another.
- This is extremely useful for orienting specimens along particular crystallographic axes.
- Based on Kikuchi lines, we can easily find the zone axis (How?).

As seen above figure, we can extend the Kikuchi line to create a second pattern from one Kikuchi pattern.

- E.g. knowing the [001] pattern we can construct the [101] pattern since a pair of lines is common to both, i.e. ±(020) lines have zone axes [101] and [001] pole.
- So we draw ±(020) lines from the [001] pole 45 ° to the [101] pole. Although the angle between the [001] and [101] pole is 45 °, we draw the ±(020) lines as parallel and straight because we are always looking at a small segment of the Kikuchi pattern.
- To find [101] pole pattern in operation, we just need to keep ±(020) lines visible while tilting specimen 45 ° about [001]
The Kikuchi lines move with the tilt, as if they were fixed to the crystal planes of sample.
Based on this principal and procedure, we can add [112] pattern, [111] pattern, and [011] pattern as seen in Fig.

- (a) Construction of the [112] (tilt 35°) pattern from the [001] and [101] pattern by extending lines common to each pair of pattern. The [11-1] pair is common to the [101] and [112] pattern the (-220) pair is common to [001] and [112] pattern.

- (b) Other poles can be added such as [011] and [111].

- All constructions are based on the zone law, e.g. [011] pole and ±(200) satisfy: 0x2+1x0+1x0=0.
Experimental Kikuchi Map for fcc crystal and index Kikuchi lines in the schematic map.

Maps for other cubic materials are available in handbook, or refer to J. W. Edington "Practical Electron Microscopy in Materials Science", 1976, Van Nostrand Reinhold Company.
If we know which Kikuchi lines are on the screen, we can use the Kikuchi lines as “roads” to “drive” the tilt from one spot pattern (crystal zone axis) to another.

• Figure shows when keeping a low order [200] Kikuchi band in view while tilting a specimen, the specimen passes through a series of low order zone axes.

Example 1: Use Kikuchi Map to find new pattern

Tilt around [200] band of 18.5 ° from [001] to [013]
Example 2: a [001] diffraction pattern of an fcc crystal was tilted 45° along (020) Kikuchi band to a 110-type pole. 1. Sketch the pattern, label the zone axis and index the low order diffractions. 2. If the crystal was now tilted 40°54” along the (111) Kikuchi band, what pole would be present?
Example: a [001] diffraction pattern of an fcc crystal was tilted 45° along (020) Kikuchi band to a 110-type pole. 1. Sketch the pattern, label the zone axis and index the low order diffractions. 2. If the crystal was now tilted 40°54” along the (111) kikuchi band, what pole would be present?

Solution: on (020) band, there are two types of [110] pole, [-101] and [10-1].

Kikuchi band direction: $b = p_1 \times p_2$

$$b = p_1 \times p_2 = \begin{vmatrix} i & j & k \\ 0 & 0 & 1 \\ -1 & 0 & 1 \end{vmatrix} = 0 + 1j + 0 = (010)$$

parallel to (020)

Pole [10-1] results in (0-20)
So pick [-101], and the angle between [001] and [-101] is

$$\theta = \arccos\left\{ \frac{[001]}{\sqrt{0+0+1}} \cdot \frac{[-101]}{\sqrt{1+0+1}} \right\} = 45^\circ$$

This confirms the tilt is 45° tilt
Zone axis is [-101], and low order diffractions are \{020\}, \{111\}, and \{202\}

The distance between spots, $r$, 

So we can obtain the [-101] pattern:
Question 2: If the crystal was now tilted 40°54’ along the (111) kikuchi band, what pole would be present?

Assume new pole is [hkl], and (111) band is common to [-101] and [hkl] pole.

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After calculation, we know that entire sample tilt procedure is first following (020) band and tilt 45° to put (-101) onto zone axis; and then tilt tilted 40°54’ along the (111) kikuchi band to put (-1-23) onto zone axis, as shown in next page.
The sample tilt procedure is first following (020) band and tilt 45° to put (-101) onto zone axis; and then tilt tilted 40°54” along the (111) kikuchi band to put (-1-23) onto zone axis, as shown in figure.
HW# 16 (a) Sketch and label a [112] diffraction pattern including Kikuchi band for an fcc material.

(b) which Kikuchi band should be used and how many degrees should the sample be rotated to get the [011] diffraction.

(c) Sketch and label the [011] diffraction pattern including Kikuchi band consistent with (b).

Due day: 11/3/08
Double Diffraction

• An electron is diffracted twice before leaving the specimen.

• The beam from the first diffraction serves as the incident beam for a second diffraction, i.e. a diffracted electron acts as an incident beam for a second diffraction.

• Forbidden diffractions in low symmetry crystals are often observed when the specimens are of modest thickness.

• Strong diffractions can act as primary beams for a second diffraction, exciting a diffraction around them. With the second diffraction, intensity may appear at positions of forbidden diffractions.

• Sometimes it is possible to perform tilting experiment to test if double diffraction is occurring. Through tilting, some forbidden diffraction become weaker and weaker until vanished, while the major allowed diffractions keep intensity.
(a) [110] zone axis pattern for a fcc crystal. The arrowed diffractions, which are forbidden in fcc such as (001), are caused by double diffraction; (b) and (c) same pattern but progressively tilted around the g(001) axis. The intensity in the (001) diffraction is completely absent in (c).
Conventional electron diffraction techniques use a parallel incident beam. These techniques are called Selected Area Diffraction (SAD). When the SAD patterns from area less than ~ 0.5 µm, the SAD pattern cannot be used to precisely describe the material structure. The minimum size of SAD aperture usually is 0.5 µm.

In contrast, Convergent Beam Electron Diffraction (CBED) uses a convergent beam of electrons to limit the area of the specimen which contributes to the diffraction pattern. Each spot by CBED then becomes a disc within which variations in intensity can usually be seen. Such patterns initially seem more difficult to interpret but they contain a wealth of information about the symmetry and thickness of the crystal and are widely used in TEM.

The big advantage of CBED over SAD techniques is that most of the information is generated from small regions beyond the reach of other diffraction techniques. The CBED technique is also called micro-diffraction technique. Actually more useful than SAD pattern, especially for nano-scaled materials. For thickness measurement and crystal space group determination. See textbook for more application.
Small convergent angle, $2\alpha$

Ray diagram showing how to form CBED pattern
large convergent angle, $2\alpha$
Example CBED Pattern showing the diffraction discs instead of spots

[111] zone axis CBED pattern for Cu-15 at% Al, recorded at 80 kV. Both the whole pattern (a) and bright field (b) symmetries are equal to $3\overline{m}$.

Small convergent angle, $2\alpha$
CBED whole pattern with Kikuchi and HOLZ line, large convergent angle, 2\(\alpha\)

CHEM 793, 2008 Fall
CBED whole pattern with Kikuchi and HOLZ line, very large convergent angle, \(2\alpha\), and low camera length (200 mm), showing a wide area of reciprocal space. This beam condition is used to form STEM image.