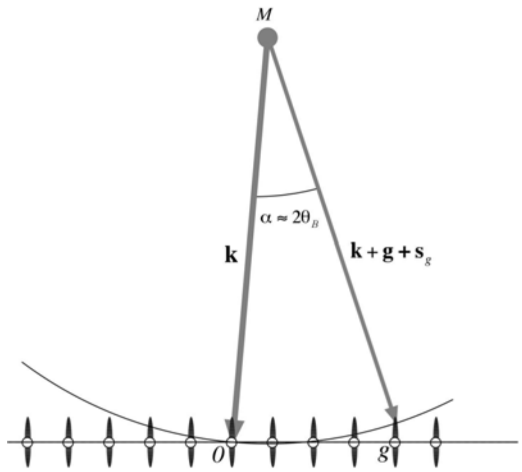
16. Kikuchi Diffraction

Changes in diffraction angle

We often see diffraction peaks from reflections whose planes are not at the precise Bragg condition. So it seems like the scattering angle for those peaks may be a little off from what we expect using Bragg's law. Let's call the actual diffraction angle α . At the Bragg condition, it should be $\alpha = 2\theta_B$.



Even if we are not at the Bragg condition, the scattering is elastic, so we find that:

$$k = |\mathbf{k} + \mathbf{g} + \mathbf{s}_g|$$
$$\cancel{k^2} = \cancel{k^2} + 2\mathbf{k}\cdot(\mathbf{g}+\mathbf{s}_g) + (\mathbf{g}+\mathbf{s}_g)^2$$
$$2\mathbf{k}\cdot(\mathbf{g}+\mathbf{s}_g) = -(\mathbf{g}+\mathbf{s}_g)^2$$

We can find specify the angle α between the incident and scattered beams as follows:

$$\mathbf{k}\cdot(\mathbf{k}+\mathbf{g}+\mathbf{s}_g) = k^2\cos\alpha = k^2 + \mathbf{k}\cdot(\mathbf{g}+\mathbf{s}_\mathbf{g})$$

Now solve for α :

$$k^2\cdot(1-\cos\alpha) = -\mathbf{k}\cdot(\mathbf{g}+\mathbf{s}_\mathbf{g})$$
$$k^2\cdot\sin^2(\alpha/2) = \frac{1}{2}(\mathbf{g}+\mathbf{s}_\mathbf{g})^2$$

We are usually concerned with ZOLZ reflections, with $\mathbf{g}\perp\mathbf{s}_\mathbf{g}$:

$$\sin(\alpha/2) = \frac{1}{2k}\sqrt{g^2 + 2\,\cancel{\mathbf{g}\cdot\mathbf{s}_\mathbf{g}} + s_\mathbf{g}^2}$$
$$\alpha = 2\cdot\sin^{-1}\left[\frac{g}{2k}\sqrt{1+\left(\frac{s_\mathbf{g}}{g}\right)^2}\right]$$

When $s_\mathbf{g} = 0$, we get Bragg's law

$$\alpha = 2\cdot\sin^{-1}\left(\frac{g}{2k}\right) = 2\theta_B$$

Away from the Bragg condition, we have

$$\alpha = 2 \cdot \sin^{-1}\left[\sin\theta_B \sqrt{1+\left(\frac{s_g}{g}\right)^2}\right]$$

In TEM, the scattering angles are quite small, so we just keep the lowest-order correction:

$$\alpha \approx 2\theta_B \cdot \left[1+\frac{1}{2}\left(\frac{s_g}{g}\right)^2\right]$$

This is a very small correction. Say we have a small beam tilt of ϕ. A previous result gives

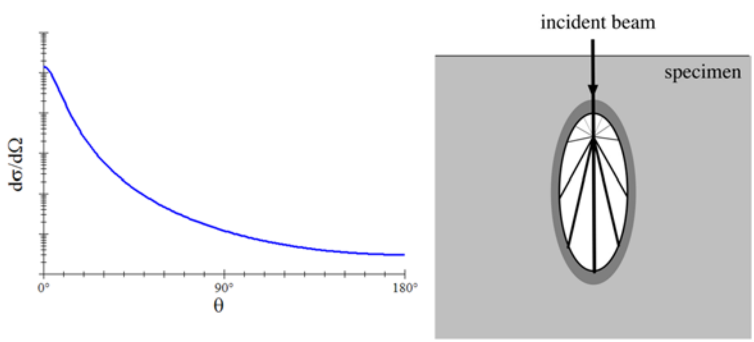
$$s_g \approx g \cdot \phi - \frac{g^2}{2k}$$

So the correction is about

$$\alpha - 2\theta_B \approx \theta_B \cdot (\phi - \theta_B)^2$$

Incoherent, diffuse scattering

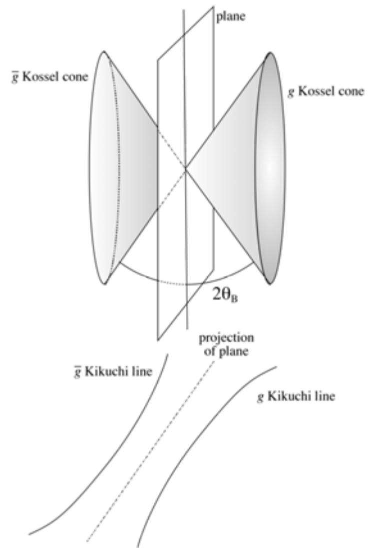
Elastic scattering can occur in any direction, though most of the scattering (of any type) is forward. We can see this from a simple calculation of the differential scattering cross-section of an atom using the Rutherford model:



But even if the scattering is elastic, the coherence of an electron w.r.t. the incident beam decreases with every scattering event, because some energy gets transferred to the specimen. After multiple scattering events, our incident beam is accompanied by a diffuse "plume" of incoherent electrons. These still have close to the same energy as the incident beam, but they have lost their coherence. We can imaging this source of incoherent electrons radiating outward from every point in the specimen, but concentrated in the forward direction.

Kossel cones

Since the diffusely scattered electrons are propagating in every direction (but mostly forward), some of them will be at an angle θ_B w.r.t. a particular set of diffracting planes. The diffraction off these planes will also be at angle θ_B from the planes. But there is not just one pair of incident/scattered wave rays at θ_B from the plane, but rather a whole cone of such rays. We can picture the cone with one of the planes (it doesn't matter which one if we are far from the specimen) at its apex, oriented with the normal to the plane along its axis. In fact, there is another cone on the opposite side of the plane that also describes all possible pairs of incident/scattered wave vectors that are possible paths for Bragg scattering. These two cones are called Kossel cones. We might call the one extending in one direction the g Kossel cone and the opposite one the $\bar{g}$ Kossel cone. The angle between them is $2\theta_B$.



Kossel cone construction

Say the diffraction planes has $x = y = 0$, and the cone apex is at $z = -L$ .The cones make an angle θ_B w.r.t. the Bragg plane, so they satisfy

$$\frac{x}{\tan\theta_B} = \pm\sqrt{y^2 + (z+L)^2}$$

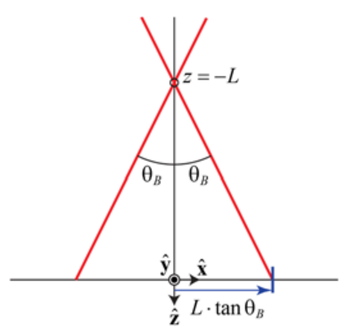
Consider the intersection of the cones with a horizontal plane below the specimen at $z = 0$. The trace is

$$x = \pm\tan\theta_B\sqrt{y^2 + L^2}$$

This described hyperbolas in the + and - directions. If we are very close to the origin (small angles), expansion to lowest order gives:

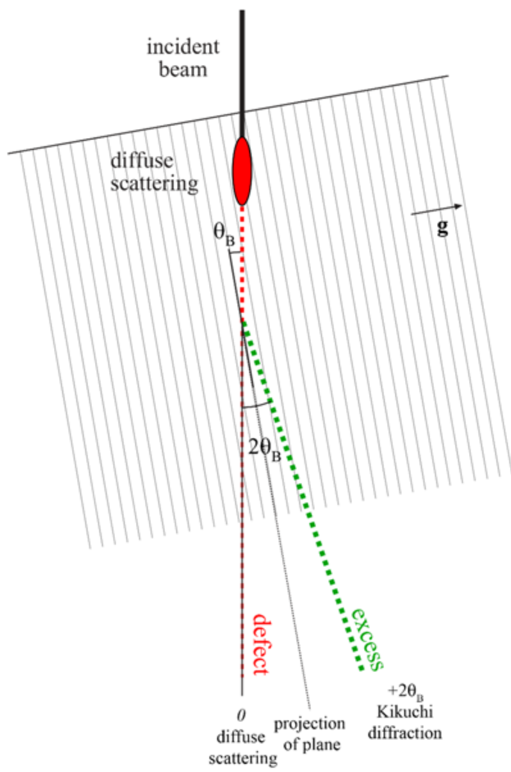
$$x \approx \pm L\cdot\tan\theta_B\cdot\left(1+\frac{y^2}{2L^2}\right)$$

This describes parabolas extending in opposite directions. If y is small, they are just lines at $x = \pm L\cdot\tan\theta_B$. We can think of L as the camera length. This describes where the traces of the cones in the diffraction pattern.



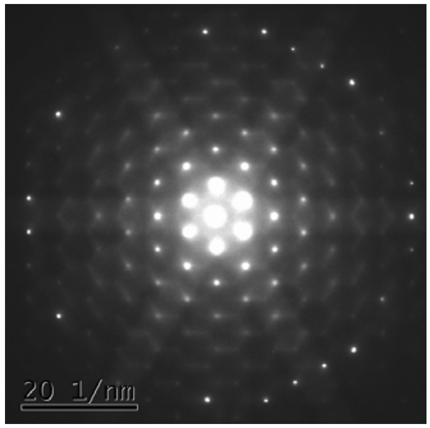
One thing to notice is that the Kossel cones tilt with the sample. We are not very concerned with the incident beam direction at this point, except to say that the diffuse scattering is strongest in the forward direction.

Kikuchi diffraction
The diffusely scattered electrons can diffract off of Bragg planes and contribute to diffraction patterns. This requires more than one scattering event: 1) incoherent, diffuse, elastic scattering (which may involve multiple scattering events, and is strongest in the forward direction), followed by 2) coherent, elastic scattering (only one scattering event needed).



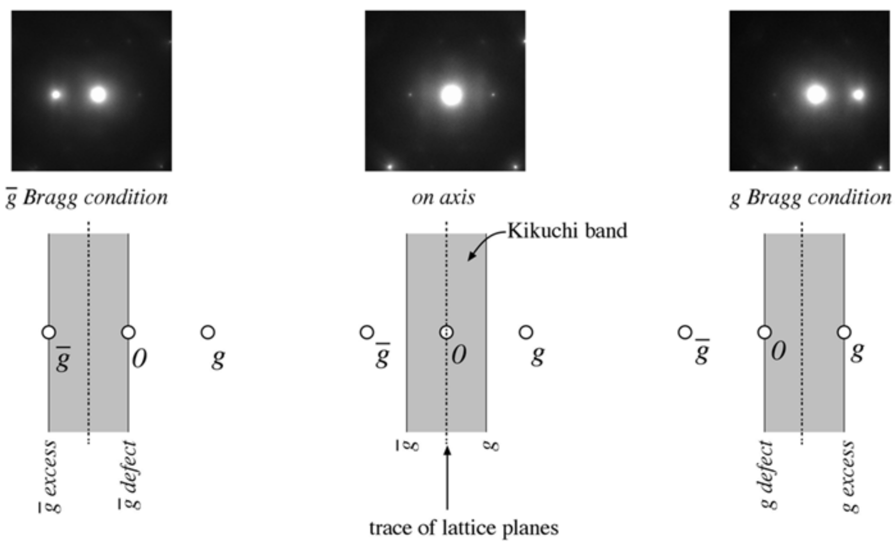
Say a set of planes is oriented at the Bragg condition w.r.t. the incident beam. In addition to the diffraction spot from these planes, we also expect the diffuse electrons propagating in the forward direction to Bragg diffraction from these planes. Since these are distributed on the Kossel cone, the scattered intensity should form a bright line segment passing through the diffracted spot. This is called the "excess" Kikuchi line. The original, forward direction will have lost the contribution from those diffuse electrons, so there will be a dark "defect" (or "deficient") Kikuchi line passing through *0*. The region in between these lines is called the Kikuchi "band". We can always think of the Kikuchi band is rotating with the sample. The projection of the Bragg planes is in the middle of the band, halfway between the excess and defect Kikuchi lines.

Example: Si<111>
Kikuchi bands can be seen in the selected-area diffraction pattern below taken from a Si crystal on a <111> zone axis. The most prominent are the {220} Kikuchi bands.



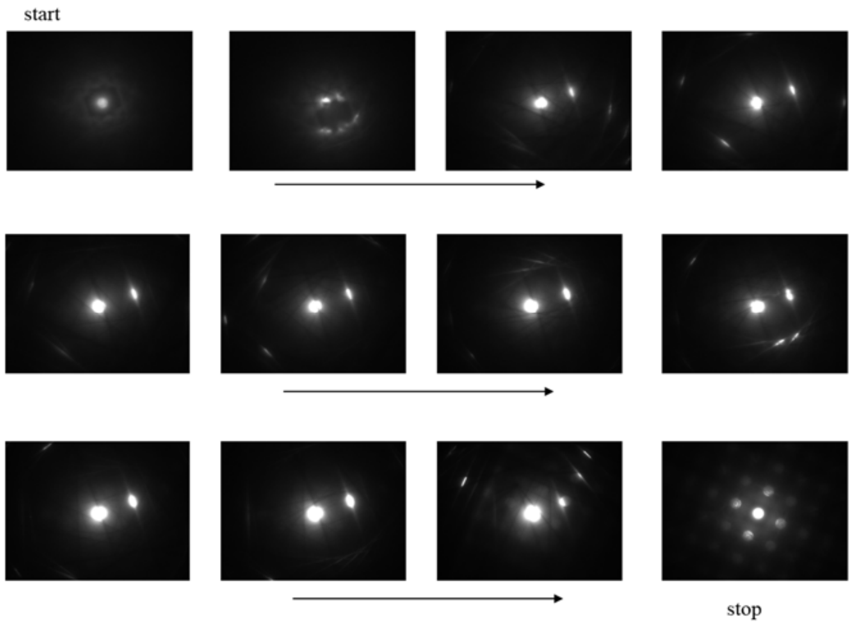
Kikuchi bands

We saw that diffraction spots move with the direct beam, whereas Kikuchi bands move with the sample. (Even though the diffraction spots don't move with sample tilt, they do change intensity.) When we are at the Bragg condition for reflection *g*, there *g* Kossel cone traces out the excess Kikuchi line, and the $\bar{g}$ Kossel cone traces out the defect Kikuchi line. But the excess/defect assignment is fungible; when we rotate the sample to the Bragg condition for reflection $\bar{g}$, the $\bar{g}$ Kossel cone traces out the excess Kikuchi line and the *g* Kossel cone traces out the defect Kikuchi line. On axis, the excess/defect distinction is lost. We still see a Kikuchi band of width $2\theta_B$ that is symmetrical.



Tracking a Kikuchi band

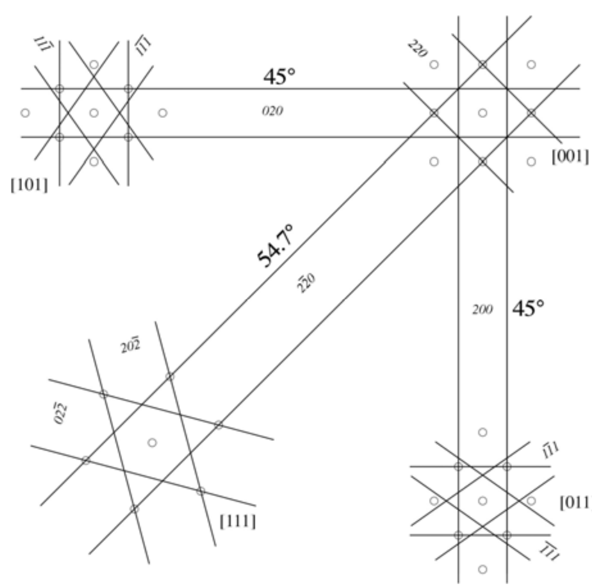
The Kikuchi bands are especially useful when we need to orient a specimen precisely on a particular zone axis, or reorient from one zone to another. The Kikuchi bands always run perpendicular to the $\mathbf{g}$ vector they are associated with. If we find a $\mathbf{g}$ vector that is shared by the two zones, we can follow its Kikuchi band like a highway leading between the zones. If we lose track if where the band is, we can just tilt the sample slightly to reach the Bragg condition for *g*. Then, the excess and defect lines will be very clearly visibly and the search can be resumed.



Note that, whereas we can see Kikuchi bands in selected-area diffraction patterns, they are even stronger in convergent-beam diffraction patterns, which we will discuss shortly. Some examples here use a convergent beam, rather than a parallel beam.

Kikuchi map: fcc

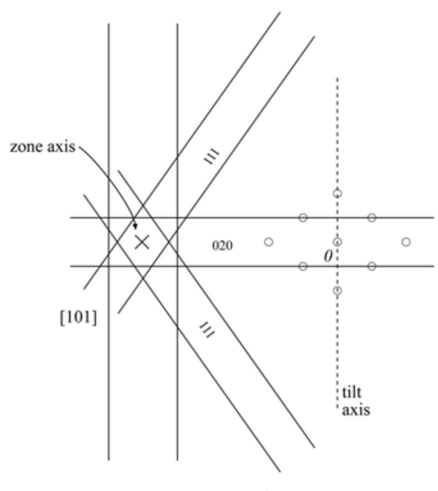
The Kikuchi bands connect various directions, like straight highways looping around the surface of a globe, similar to lines of longitude, although not all pass through the poles. We could plot them in various projections. Usually we pick one reference zone axis and show the bands connecting that axis to other major zone axes. A Kikuchi map for an fcc crystal is shown below. The [001] zone is the starting point. We can connect from there to the [011] zone that is 45° away by following the 200 (or 400) Kikuchi band. (Notice that, according to the Weiss zone law, 200 resides on both zones.). Or we could find the [101] zone, instead, by tilting 45° about an axis 90° from the first one, following the 020 (or 040) band to the [101] zone axis. The [111] zone is 54.7° from the [001] zone, following the $2\bar{2}0$ band.



If we look at this map carefully, we see that the $02\overline{2}$ band must connect the [111] zone to the [011] zone. Also, the $20\overline{2}$ band connects [111] to the [101] zone. But these bands don't seem to point in the right directions on this map, because we had to distort some of the angles on the directional sphere to plot them on a single map.

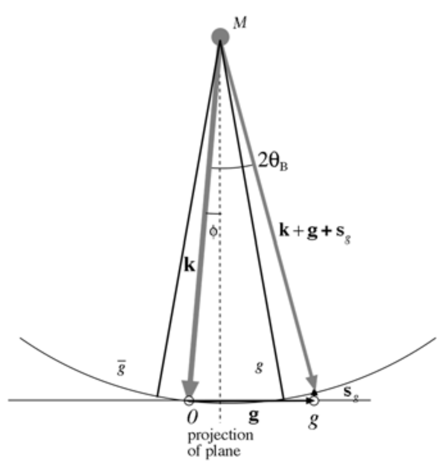
Orienting on the zone axis with Kikuchi lines

Say we are near a low-index zone axis and want to tilt the specimen so that the beam is parallel to the zone axis. We will see diffraction spots, including the direct beam, at roughly fixed positions, regardless of the sample tilt. But the Kikuchi lines will move with the sample tilt. The actual location of the zone axis is where the Kikuchi bands intersect, not where the direct beam is. As we tilt the sample, the Kikuchi bands will move with it, so we can observe both the zone axis location and the beam direction, bringing them into alignment.

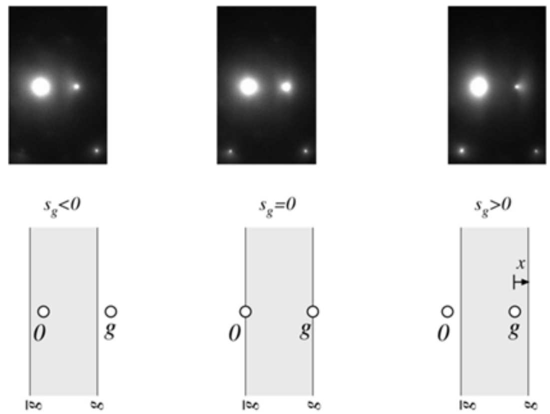


Measuring s_g

We said the excess Kikuchi line passes through the reflection at the Bragg condition ($s_g = 0$). On the zone axis, the excess line is just the *g* line, passing halfway between *g* and *0*, and the reciprocal-lattice point *g* is outside the Ewald sphere ($s_g < 0$).



Apparently, if the *g* line passes on the other side of the g reflection, $s_g > 0$. In other words, the position of the *g* line relative to the *g* reflection provides a way to measure s_g.



For small tilt angles, the relationship is linear. If the *g* line is offset from the reflection by a reciprocal-space distance x, the tilt from the zone axis is:

$$\phi = \theta_B \cdot \left(\frac{2x}{g} + 1 \right) = \frac{g}{2k} \cdot \left(\frac{2x}{g} + 1 \right) = \frac{x}{k} + \frac{g}{2k}$$

For small angles, the excitation error varies as

$$s \approx g\phi - \frac{g^2}{2k}$$

Using the value of ϕ above, we get:

$$s = g \cdot \left(\frac{x}{k} + \cancel{\frac{g}{2k}} \right) - \cancel{\frac{g^2}{2k}} = \frac{gx}{k}$$

Not surprisingly, the excitation error is proportional to x. We could write it as:

$$s \approx 2\theta_B \cdot x$$