NANO 703-Notes

Chapter 17-Diffraction from small volumes

Two-beam intensity

Remember the effective excitation error for beam g?

$$s_{eff} = \sqrt{s^2 + \frac{1}{\xi^2}}$$

The general two-beam result for g was:

$$\Psi_g(T) = i \frac{\sin(\pi s_{eff} T)}{s_{eff} \xi} e^{-\pi i s T}$$

Next we find the intensity for the g beam:

$$I_g = |\psi_g(T)|^2 = \frac{\sin^2(\pi s_{eff} T)}{(s_{eff}\xi)^2} = \frac{\sin^2[\pi \cdot (T/\xi) \cdot \sqrt{1+w^2}]}{1+w^2}$$

where $w = s\xi$.

Alternative form

Been there, done that, you say. Well, an alternative form is

$$\psi_g(T) = i\left(\frac{\pi T}{\xi}\right)\mathrm{sinc}(\pi s_{eff} T) e^{-\pi i s T}$$

Now the two-beam intensity is

$$I_g = \left(\frac{\pi T}{\xi}\right)^2 \mathrm{sinc}^2(\pi s_{eff} T) = \left(\frac{\pi T}{\xi}\right)^2 \mathrm{sinc}^2\left(\frac{\pi\sqrt{1+w^2}\,T}{\xi}\right)$$

Variation with thickness/excitation error

We should expect to see oscillations in intensity with both thickness and excitation error. We could plot I_g vs. T/ξ at carefully selected values of w. Or, we could plot I_g vs. w for nice values of T/ξ:
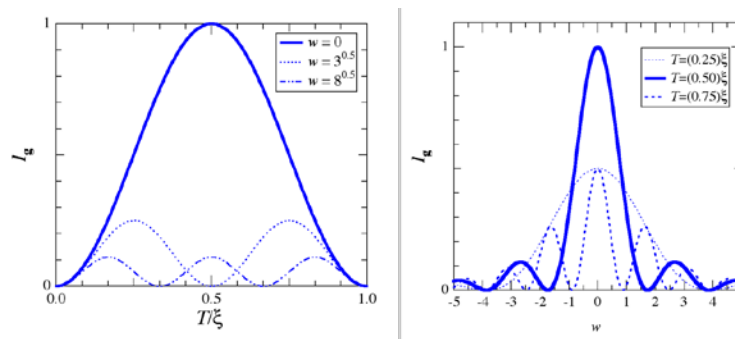
In the first case, the oscillation is sinusoidal, but the amplitude and period decrease as we move away from the Bragg condition. In the second case, we see the sinc-squared function, with the period of

oscillation in w getting shorter as the sample gets thicker. These intensity variations are called "pendellosung" oscillations. The show up in X-ray diffraction, too, from very thin films.

Kinematical approximation

A simpler picture starts to emerge if we are away from the Bragg condition, in which case $w^2 \gg 1$. Let's assume the diffraction is so weak that the direct-beam intensity is undiminished from its incident value, so $\Psi_0 = 1$. This is called the "kinematical" approximation. Starting with the good old Howie-Whelan equations, we can say:

$$\frac{d\Psi_{\mathbf{g}}}{dz} \approx \left(\frac{\pi i}{\xi_{\mathbf{g}}} \right) \cdot (1) \cdot e^{-2\pi i s_{\mathbf{g}} z}$$

To find $\Psi_{\mathbf{g}}$, we can integrate over the foil thickness T:

$$\Psi_{\mathbf{g}} = \left(\frac{\pi i}{\xi} \right) \cdot \int_{z=0}^{T} e^{-2\pi i s z} dz = \left(\frac{e^{\pi i s T} - e^{-\pi i s T}}{2 s \xi} \right) \cdot e^{-\pi i s T} = i \left(\frac{\pi T}{\xi} \right) \operatorname{sinc}(\pi s T) e^{-\pi i s T}$$

Then

$$I_{\mathbf{g}} \approx \left(\frac{\pi T}{\xi} \right)^2 \cdot \operatorname{sinc}^2 \left(\frac{\pi w T}{\xi} \right)$$

Basically, we have assumed $|w| \gg 1$, meaning that $s_{eff} \approx |s|$ and $|s| \gg 1/\xi$.

Interpretation

Let's take another look at the integral we just did. Forgetting the factor in front, we can say:

$$\int_{z=0}^{T} e^{-2\pi i s z} dz = \int_{z=0}^{T} (1) \cdot e^{-2\pi i s z} dz = \Im\{L(z)\}$$

The integral is a Fourier transform over the square envelope, or "shape" function

$$L(z) = \begin{cases} 1, & 0 \le z \le T \\ 0, & \text{otherwise} \end{cases}$$

This function basically tells us where to evaluate the crystal potential. It describes the shape of a thin foil. Its Fourier transform (in terms of s) is:

$$L(s) = T \cdot \operatorname{sinc}(\pi s T) \cdot e^{-\pi i s T}$$

So here is another way to write the diffracted amplitude:

$$\Psi_{\mathbf{g}} = \frac{i\pi}{\xi} \cdot L(s) = \frac{i\lambda \cdot F_{\mathbf{g}}}{v} \cdot L(s)$$

The diffracted amplitude is distributed in reciprocal space as a function of s about the reciprocal-lattice point g. The distribution is just the Fourier transform of the envelope function describing the shape of our

specimen. This is a general result for small crystals (i.e., nanomaterials), not just thin foils. The diffracted intensity is proportional the FT of the shape function of the specimen.

$$ I_g = \left(\frac{\lambda}{v}\right)^2 |F_g|^2 \cdot |L(s)|^2 $$

Dynamical vs. kinematical

Let's see how far we are off using this approximation. Near the Bragg condition, kinematical theory only works for very thin (small samples), but as we move farther from $s = 0$, the approximation gets better.
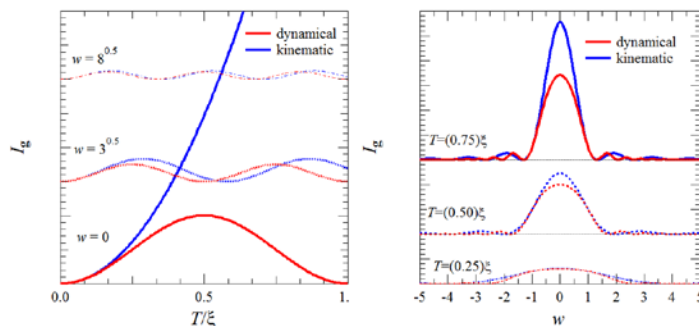

Limitations of kinematic theory

Kinematic theory breaks down when we are near the Bragg condition and the sample is thicker. Say we are at $s = 0$. Kinematic theory says, in this case:

$$ I_g(s=0) = \left(\frac{\pi T}{\xi}\right)^2 $$

But the intensity has an upper limit of $I_g = 1$ (the same as our incident beam). So we definitely don't want to use kinematic theory if $T > \xi/\pi$. It works best to describe the intensity when: 1) the sample is thin and 2) we are far from the Bragg condition.

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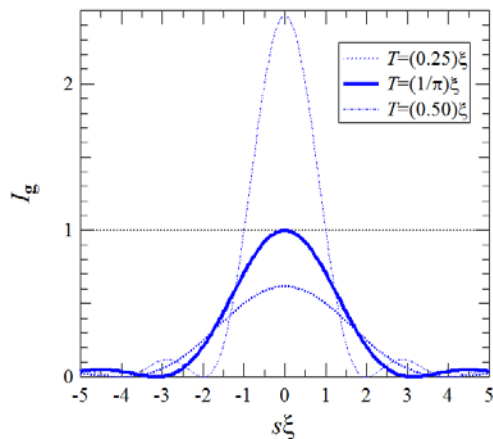
Relrods

There is a geometric interpretation of this. Imagine that, for a small crystal, each reciprocal-lattice point is now distributed into a halo around the point we would associate with an infinite crystal. For a think foil, this is a rod extending normal to the foil direction, called a "relrod". Actually, the rod oscillates in strength around the point where $s = 0$. But the rod moves with the crystal; it doesn't really matter what direction the beam comes from. So kinematical theory lets us think of diffraction geometrically.
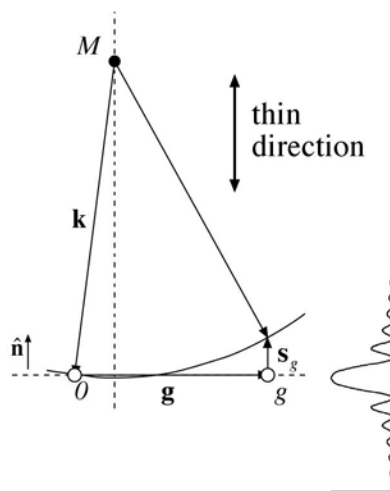

Diffraction from thin crystals

It doesn't matter which reciprocal lattice point we were talking about before. The intensity scales overall as $\left| F_{\mathbf{g}} \right|^2$, but the geometric factor $\left| L(s) \right|^2$ is the same for all reciprocal lattice points. Thus, we can imagine a relrod attached to *every* reciprocal lattice point in our thin crystal. The intensity of each diffraction spot depends on where its relrod intersects the Ewald sphere. Now we know why so many spots appear in a selected-area diffraction pattern from a thin foil. We don't have to be right at the Bragg condition for any particular reflection to show up, because the reciprocal-lattice points are all stretched out in the thin direction.
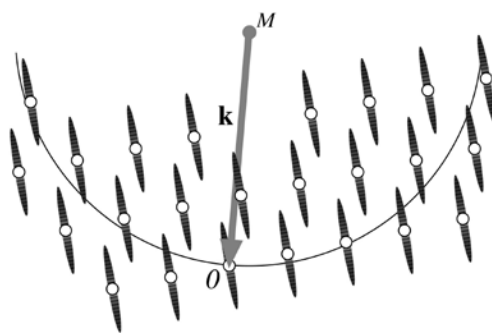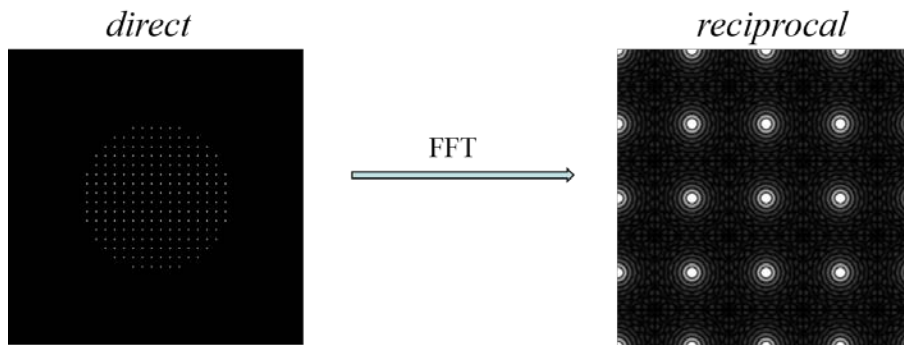

Shape function: circle

Since we think the FT of the shape function determines the distribution of intensity in reciprocal space, we can calculate a few 2-D examples. Say we have a circle that is uniformly filled. Let's find its FFT:
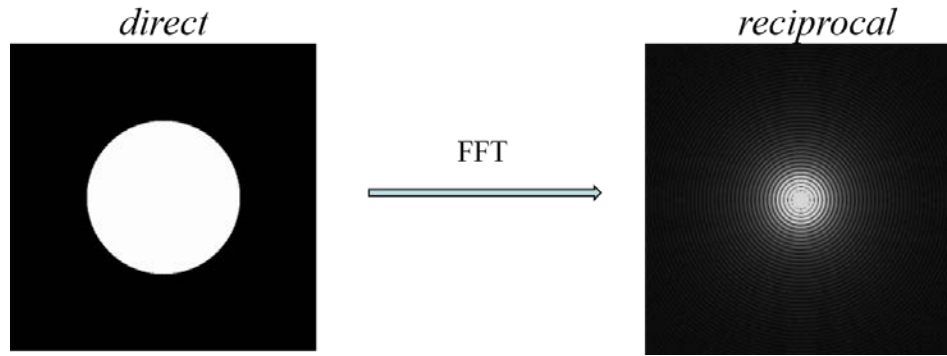
Now within the image the circle is the shape function for a square lattice of points.

The lattice gives rise to an array of reciprocal-lattice spots. The intensity around each spot is spread out by the shape function of the circle.

Shape function: thin foil
Here is another example. Say we have a thin slab of material that is uniformly filled. Its FFT gives a central spot with a streak in the thin direction.
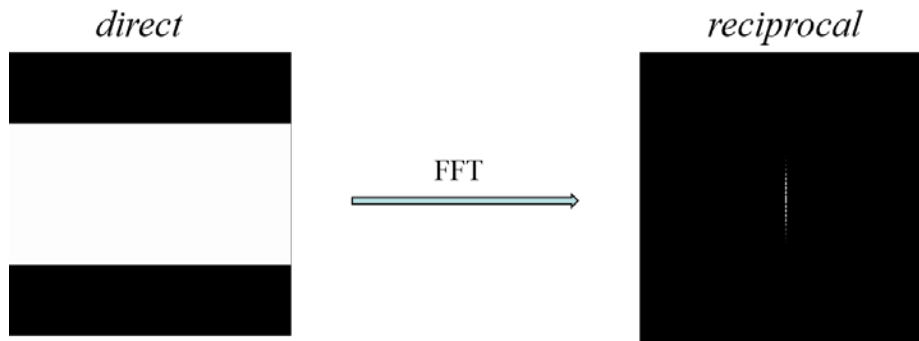

Now if we use that slab to be the shape function of a lattice of points, we see streaks about every reciprocal-lattice point extending in the thin direction.
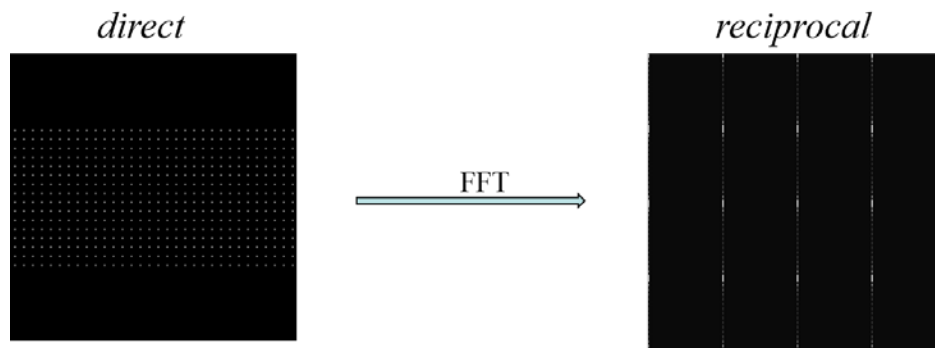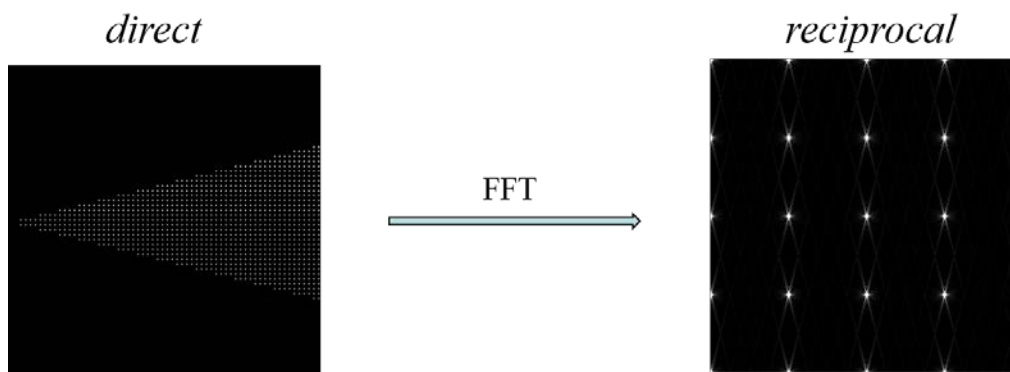
Shape function: wedge

This example is not so simple. In TEM, we often have a thin foil of non-uniform thickness. Let's assume a uniform slope. The FFT gives one streak perpendicular to the top surface and another perpendicular to the bottom surface.
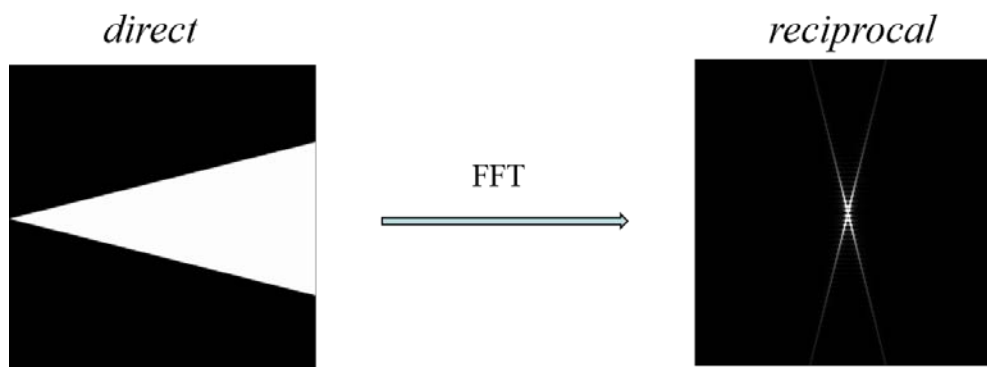
If the wedge-shaped object contains a lattice, we expect a pair of streaks for each of the lattice points.

We are pushing the limits of kinematical theory here a little bit. A full dynamical treatment, with the beam passing down through the two wedge interfaces, reveals that the two streaks do not actually cross. Instead, they veer away from each other. Even at the Bragg condition, then, we actually expect a diffraction spot from a wedge-shaped foil to split into two closely spaced spots. Assuming a two-beam condition for reflection g, the splitting in reciprocal space about the original location of g is given by $\sin\phi/\xi$, where ϕ is the wedge angle and ξ is the extinction distance. The splitting is most pronounced for stronger reflections, which have small ξ, and large dynamical effects.

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