20. Phase Contrast

More on phase contrast

The wave number of a high-energy electron in the vacuum can be written

$$k = \sqrt{\frac{2mE_{nr}}{h^2}}$$

which looks like the classical version, because we used a modified kinetic energy

$$E_{nr} = \left(\frac{m+m_0}{2m}\right)\cdot E$$

In the specimen, the kinetic energy will vary with position, so the wave number will, too:

$$K(\mathbf{r}) = \sqrt{\frac{2m[E_{nr} + eV(\mathbf{r})]}{h^2}} = k\sqrt{1+\frac{eV(\mathbf{r})}{E_{nr}}}$$

I have used $V(\mathbf{r})$ to represent the crystal potential in this context. Since $eV(\mathbf{r})$ is a much smaller term than E_{nr}, we can expand to lowest order

$$K(\mathbf{r}) \approx k\left[1+\frac{eV(\mathbf{r})}{2E_{nr}}\right] = k + \frac{\sigma V(\mathbf{r})}{2\pi}$$

The interaction constant is defined as:

$$\sigma \doteq \frac{\pi k e}{E_{nr}}$$

Phase-object approximation

As the electron progresses through an increment dz in specimen thickness, the electron wave function phase advances

$$\psi(x, z+dz) = \psi(x,z)e^{2\pi i K(\mathbf{r})dz} \approx \psi(x,z)e^{2\pi ik\cdot dz}e^{i\sigma V(\mathbf{r})dz}$$

We can find the net phase change by integrating over the entire thickness *T*. First identify that

$$\psi(x, z+dz) \to \psi + d\psi$$

Since dz is very small, we can expand:

$$\psi \cdot e^{2\pi i K(\mathbf{r})dz} \approx \psi \cdot [1 + 2\pi i K(\mathbf{r})dz]$$

Now we can separate variables:

$$\cancel{\psi} + d\psi = \psi \cdot \left[\cancel{1} + 2\pi i K(\mathbf{r})dz\right]$$

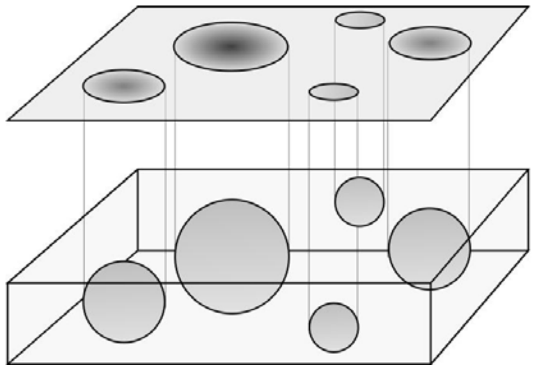
$$\int_{\psi=\psi(x,0)}^{\psi(x,T)} \frac{d\psi}{\psi} = \int_{z=0}^{T} 2\pi i K(\mathbf{r})\cdot dz = \int_{z=0}^{T} 2\pi i\left[k + \frac{\sigma V(\mathbf{r})}{2\pi}\right]\cdot dz$$

$$\ln\left[\frac{\psi(x,T)}{\psi(x,0)}\right] = 2\pi ikT + i\sigma\int_{z=0}^{T} V(\mathbf{r})\cdot dz$$

Projected potential

The integral is the potential integrated over the thickness T. For a real, 3-D sample, this gives us a 2-D function called the projected potential

$$V_t(x,y) \doteq \int_{z=0}^{T} V(x,y,z)\cdot dz$$



Applying the phase-object approximation
Now the wave function at the bottom of the sample is

$$\psi(x,T) = \psi(x,0)\cdot e^{2\pi ik\cdot T}\cdot e^{i\sigma V_t(x)}$$

This is the exit, or final wave function

$$\psi_f = \psi(x,T)$$

The unscattered, or initial, portion of the wave would somehow avoid the specimen potential, so its wave function would be

$$\psi_i = \psi(x,0)\cdot e^{2\pi ik\cdot T}$$

We don't expect it to be a function of *x* if the incident illumination is uniform. We can call the extra phase factor the specimen, or object, function

$$F(x) = e^{i\sigma V_t(x)}$$

So, we could use these new names to write

$$\psi_f(x) = F(x)\cdot\psi_i$$

WPOA
It the specimen is a weak-phase object (i.e., very thin, low density), then $\sigma V_t(x) \ll 1$, so we can expand

$$F(x) = e^{i\sigma V_t(x)} \approx 1 + i\sigma V_t(x)$$

Now the exit wave is

$$\psi_f(x) \approx [1 + i\sigma V_t(x)]\cdot\psi_i = \psi_i + i\psi_{sc}(x)$$

where we have used definitions from previous chapters. Clearly $\psi_{sc}(x) \ll \psi_i$. We usually want $\psi_f(x)$ to be normalized:

$$1 = |\psi_i|^2 + \cancel{|\psi_{sc}(x)|^2}\ , \ \to \psi_i = 1$$

Now we could write

$$F(x) = \psi_i + i\psi_{sc}(x)$$

I need to make a leap ahead to a later concept, which is that the microscope itself, particularly the objective lens, can cause additional phase shifts. If everything is aligned right, the initial wave (the direct beam) is not affected, but the scattered wave is shifted by some phase that depends on the OL defocus. Let's assume that all of the scattered wave experiences the same phase shift $-\chi$ (sticking with the sign convention). Then the wave function in the image plane would be

$$G(x) = \psi_i + i\psi_{sc}(x) \cdot e^{-i\chi}$$

Phase contrast

Let’s find the image intensity, assuming $\psi_i = 1$

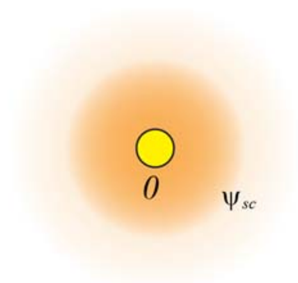
$$\begin{aligned} I(x) &= |G(x)|^2 \\ &= \left|1 + i\psi_{sc}(x) \cdot e^{-i\chi}\right|^2 \\ &= 1 + \cancel{|\psi_{sc}(x)|^2} + i\psi_{sc}(x) \cdot e^{-i\chi} - i[\psi_{sc}(x)]^* \cdot e^{i\chi} \end{aligned}$$

$$I(x) = 1 - 2\,\mathrm{Im}[\psi_{sc}(x) \cdot e^{-i\chi}]$$

For the moment, let’s assume that $\psi_{sc}(x)$ is real. Then

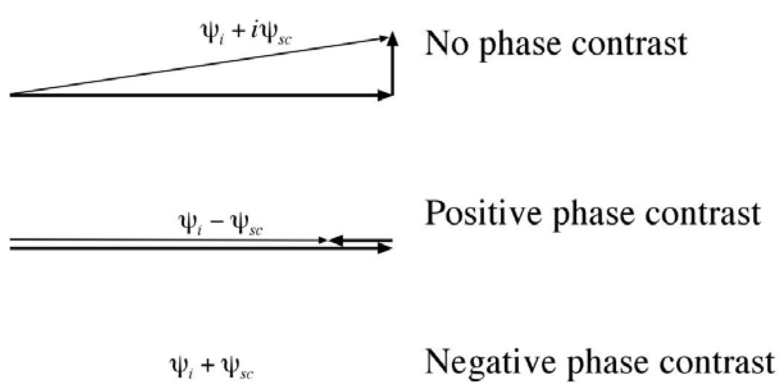
$$I(x) = 1 + 2\psi_{sc}(x)\sin(\chi)$$

So χ controls how the scattered wave combines with the unscattered wave.



Phase contrast explained

The path lengths taken by waves depend on the their trajectories through the objective lens, and these change with the lens defocus. This rotates the scattered wave to rotate in phase in various ways before recombining with the incident beam in the image plane. There are three distinct possibilities:



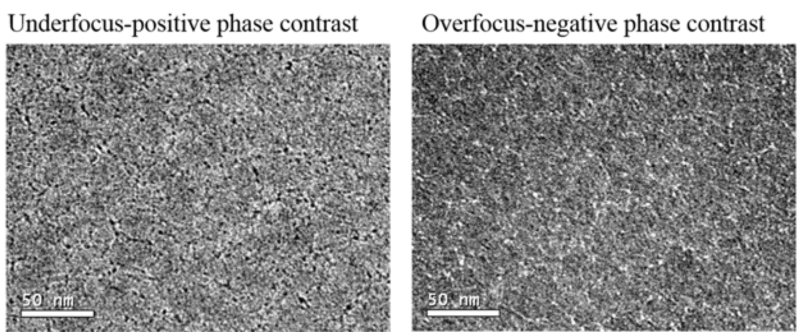
The image intensity is proportional to the magnitude of G. Since $\psi_{sc} \ll \psi_i$, if we don’t induce any additional phase shift ($\chi = 0$), the object has very little effect on the $|G|$, because ψ_{sc}^2 must be very small

$$|G| = \sqrt{\psi_i^2 + \psi_{sc}^2} \approx \psi_i + \cancel{\frac{\psi_{sc}^2}{2\psi_i}}$$

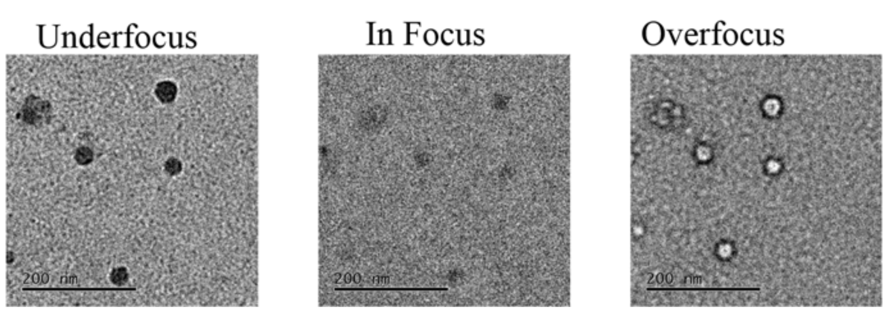
To first-order, there is *no* phase contrast in this case. This is actually the case when a phase object is precisely in focus in the objective lens. So a phase object shows minimum contrast when it is in focus.

If we add an extra 90° phase change to ψ_{sc} ($\chi = -\pi/2$), then $|G| = \psi_i - \psi_{sc}$, and we have *positive* phase contrast, in the sense that the object will appear darker than the background, as we usually expect in a "shadow" image. This makes the image contrast interpretable in the conventional sense, and is approximately what we observe when the objective lens is slightly underfocused . But if we instead rotate back by 90° to remove any phase shift ($\chi = \pi/2$), then $|G| = \psi_i + \psi_{sc}$, and we have *negative* phase contrast. In this case, the object appears brighter than the background., and can be observed with a slight overfocus of the objective lens. We usually prefer positive phase contrast, so we will look at how to accomplish this.

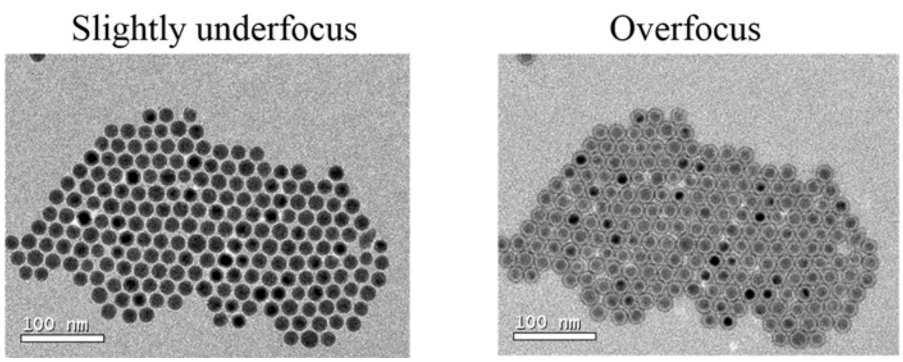
Phase contrast example I: a-C
A common example of phase contrast can be found in many TEM samples: the amorphous carbon support film used for preparation of many types of colloids. And it turns out we do have a way to add or subtract from the phase of the scattered wave. It is done by adjusting the objective-lens defocus, which will examine later. Underfocus of the OL gives positive phase contrast; overfocus gives negative phase contrast. A signature of phase contrast is contrast reversal upon changing the OL excitation through the "true", in-focus setting.



Phase contrast example II: polystyrene spheres
It is hard to tell in the previous example which image shows positive contrast and which shows negative contrast. The example below is a bit more clear, because we can actually see contrast of the polystyrene spheres w.r.t. to the carbon support film in the background. Some interesting things occur at the edges. These are called "Fresnel" fringes. From these fringes, we can see that there is not a perfect contrast reversal between underfocus and overfocus. In fact, the fringes are more accentuated in overfocus, which is another reason underfocus is usually preferred.



Not pure phase objects
A lot of things are not pure phase objects. Specimens with heavier elements, even if they are very thin, may show some phase contrast, along with amplitude contrast, including both diffraction and mass-thickness contrast. The In spheres shown below are such an example. We see evidence of contrast reversal and Fresnel fringes associated with phase contrast. But the spheres are still quite a bit darker than the background in overfocus, so there must be mass-thickness contrast, too. Even though the spheres are all about the same size, some are darker and some are lighter, and we can see bright diffraction replicas offset from the actual particles. These are all indications of diffraction contrast, because the spheres contain some crystallinity.



Lattice fringes (two-beam)
Lattice fringes are what we usually think of when we hear someone say "high-resolution" TEM. Let's apply what we have to a crystal, assuming a two-beam condition. The wave function can be broken down to give an object function

$$F(x) \approx \{\Psi_0(T) + [\Psi_g(T) \cdot e^{2\pi isT}] \cdot e^{2\pi igx}\} \cdot e^{2\pi ikT} \to 1 + i\phi_g \cdot e^{2\pi igx}$$

Here, ϕ_g may be complex, depending on the structure factor, so let's write

$$\phi_g = |\phi_g| \cdot e^{i\delta}$$

Now we can identify the scattered wave as

$$\psi_{sc}(x) = \phi_g \cdot e^{2\pi igx} = |\phi_g| \cdot e^{i(2\pi gx+\delta)}$$

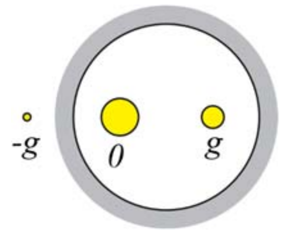
In this case, $\psi_{sc}(x)$ could be complex, so we can't use our previous form for $I(x)$. We find that

$$G(x) = 1 + i|\phi_g| \cdot e^{i(2\pi gx+\delta-\chi)}$$

Which gives

$$I(x) = 1 - 2\,\mathrm{Im}[\psi_{sc}(x) \cdot e^{-i\chi}]$$
$$= 1 - 2\,\mathrm{Im}[|\phi_g| \cdot e^{i(2\pi gx+\delta-\chi)}]$$
$$I(x) = 1 + 2|\phi_g| \cdot \sin(2\pi gx + \delta - \chi)$$

The intensity shows an oscillation that has the periodicity of the lattice planes. In fact, the phase contrast never vanishes in this case. But as χ changes, the planes shift laterally along *x*, so it is not a true representation of the crystal potential.



Lattice fringes (three-beam)
Here is another example. We usually take HR images on a high-symmetry zone axis. Say we let *0*, g and $-g$ all contribute. Then

$$F(x) \rightarrow 1 + i\phi_g \cdot e^{2\pi igx} + i\phi_{-g} \cdot e^{-2\pi igx}$$

If are incident beam is perpendicular to g, then we are sure to have

$$\phi_{-g} = (\phi_g)^* = |\phi_g| \cdot e^{-i\delta}$$

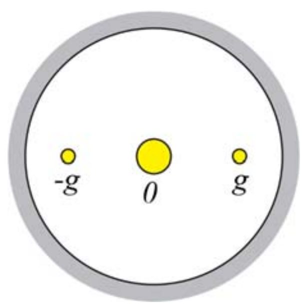
Now our scattered wave is

$$\psi_{sc}(x) = \phi_g \cdot e^{2\pi igx} + \phi_{-g} \cdot e^{-2\pi igx} = 2|\phi_g| \cdot \cos(2\pi gx + \delta)$$

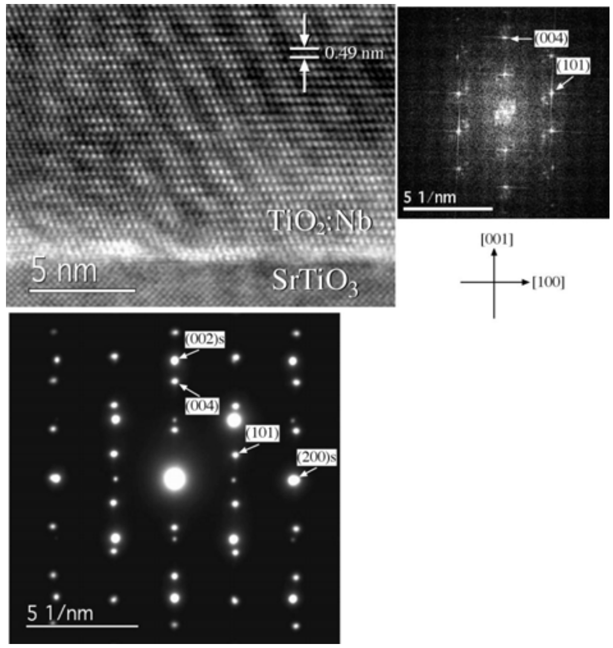
This time, it is real, so we can use our simple form for intensity

$$I(x) = 1 - 2\,\mathrm{Im}[\psi_{sc}(x) \cdot e^{-i\chi}]$$
$$= 1 - 2\,\mathrm{Im}[2|\phi_g| \cdot \cos(2\pi gx + \delta) \cdot e^{-i\chi}]$$
$$I(x) = 1 + 4|\phi_g| \cdot \cos(2\pi gx + \delta) \cdot \sin(\chi)$$

This is more like the phase contrast we usually think of. If $\chi = 0$ (in focus), the fringes disappear. If $\chi < 0$ (underfocus),we have positive phase contrast. If $\chi > 0$ (overfocus), we have negative phase contrast. In no case is there lateral shifting of the fringes.



HR lattice image example
Below is an HR lattice image example, along with its FFT and a diffraction pattern. Nothing special here, but just pointing out that there is a close tie between what we see in a diffraction pattern and the fringes that show up in the lattice image. We should always think of phase contrast lattice images as interference patterns of two or more beams. Even though the bright spots look like atoms, or columns of atoms, the connection is indirect, and what we are seeing depends a lot on defocus. If we want the image to be as directly interpretable as possible, we should take the images with a little bit of underfocus. We will figure out just the right prescription later on.


0.49 nm
TiO2:Nb
5 nm
SrTiO3
(004)
(101)
5 1/nm
[001]
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(002)s
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(200)s
5 1/nm