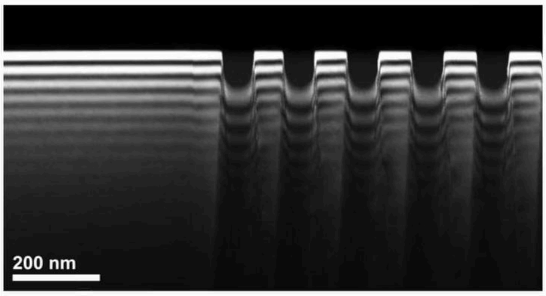
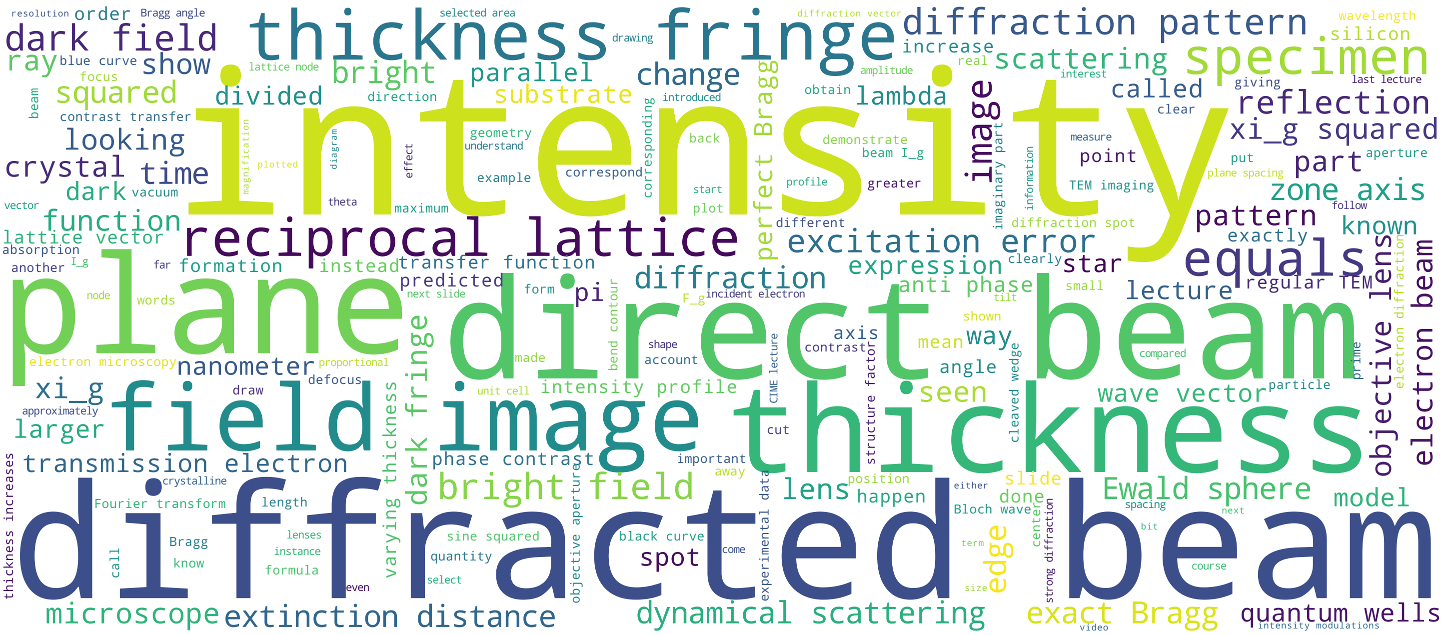


Dynamical effects – thickness fringes

Transmission Electron Microscopy

Prof. C. Hébert & Dr D. Alexander



EPFL

Welcome to CIME's lectures on transmission electron microscopy for materials science. In the last lecture I introduced dynamical scattering. That is the multiple elastic scattering that can occur as a beam propagates across the thickness of a TEM sample. I further showed how this effect leads to a strong interaction between the direct beam and different diffracted beams, that, in the two-beam case, can be conceived of as the diffraction and re-diffraction of a propagating beam. In this lecture I'm going to demonstrate one of the most evident effects of dynamical scattering in TEM imaging: namely the formation of thickness fringes. That is, patterns of bright and dark fringes that can be seen in images of crystalline objects of varying thickness.

Notes

Summary

0m 06s

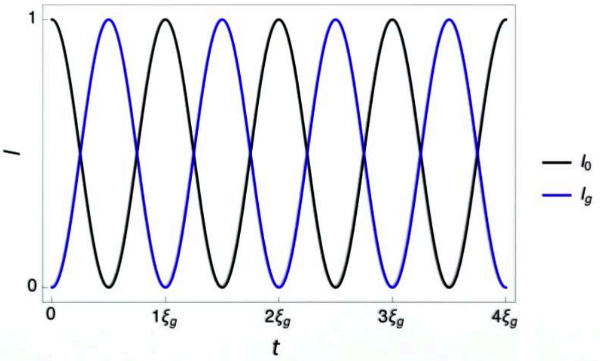
2-beam dynamical scattering intensity

$$I_g = \frac{1}{(1+\xi_g^2 s^2)} \sin^2\left(\pi t \underbrace{\sqrt{\frac{1}{\xi_g^2} + s^2}}_{s'}\right) \quad \text{where: } \xi_g = \frac{\pi V_0 \cos\theta_B}{\lambda |F_g|}$$

$$I_0 = 1 - I_g$$

Exact Bragg condition: $s = 0$

$$I_g = \sin^2\left(\frac{\pi t}{\xi_g}\right)$$



Transmission Electron Microscopy

We are going to consider the formation of these thickness fringes in the case of a two-beam scattering condition. As shown in the last lecture, the intensity in the diffracted beam, I_g, is given by this expression here, in the two-beam case. Where s is the excitation error – that is the deviation from the perfect Bragg condition – t is the sample thickness, and xi_g is the extinction distance, given by this formula here, where V_0 is the volume of the unit cell, theta b is the Bragg angle for scattering, lambda is the wavelength, and F_g is the structure factor for the diffracted beam of interest. Further, sometimes this part of the expression – the root of one over xi_g squared plus s squared – is referred to as s-prime, the effective excitation error. This expression refers to an incident intensity of one. So since we are in a two-beam condition, and all the intensity must either be in the direct beam I_0 or the diffracted beam I_g, clearly the intensity in the direct beam is given by one minus I_g. Now if we consider the case where we are at the exact Bragg condition, then the excitation error s equals zero. For this s equals zero case, you can easily see that I_g simplifies to: sine squared of pi t divided by xi_g.

Notes

Summary

0m 59s

2-beam dynamical scattering intensity

$$I_g = \frac{1}{(1+\xi_g^2 s^2)} \sin^2\left(\pi t \underbrace{\sqrt{\frac{1}{\xi_g^2}+s^2}}_{s'}\right) \qquad \text{where: } \xi_g = \frac{\pi V_0 \cos\theta_B}{\lambda |F_g|}$$

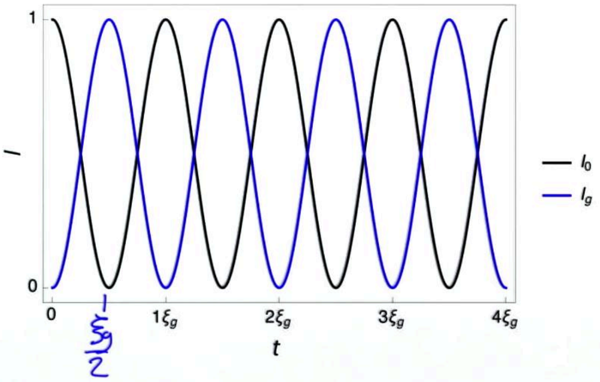
$$I_0 = 1 - I_g$$

Exact Bragg condition: $s = 0$

$$I_g = \sin^2\left(\frac{\pi t}{\xi_g}\right)$$

$$I_o = \cos^2\left(\frac{\pi t}{\xi_g}\right)$$

$$t = n\,\xi_g \Rightarrow I_g = 0$$



Transmission Electron Microscopy

Further, since the intensity in the direct beam equals one minus I_g, then that intensity is given by: I_0 equals cosine squared of pi t divided by xi_g. If we plot these two intensities against thickness, it is clear that both of them modulate sinusoidally with thickness, as shown on these curves here, where the black curve refers to the intensity in the direct beam and the blue curve, the intensity in the diffracted beam. Further, we can see that their modulations are in anti-phase with each other. Looking first at the black curve I_0, the intensity in the direct beam, we can see that, when the sample has no thickness, obviously this intensity is at a maximum, because there can be no scattering. However, as sample thickness increases, this intensity decreases, until when the sample thickness equals xi_g divided by two, the intensity goes to zero. Equally, at that thickness, the intensity in the diffracted beam is at a maximum. Looking now at this blue curve for the intensity in the diffracted beam, this in turn decreases to zero, where the thickness equals one xi_g and then two xi_g and so on. Thus, when thickness t equals an integer number of extinction distances, then the intensity in the diffracted beam equals zero, for this exact Bragg condition situation.

Notes

Summary

2m 29s

2-beam dynamical scattering intensity

$$I_g = \frac{1}{(1+\xi_g^2 s^2)} \sin^2\left(\pi t \underbrace{\sqrt{\frac{1}{\xi_g^2} + s^2}}_{s'}\right) \quad \text{where: } \xi_g = \frac{\pi V_0 \cos\theta_B}{\lambda |F_g|}$$

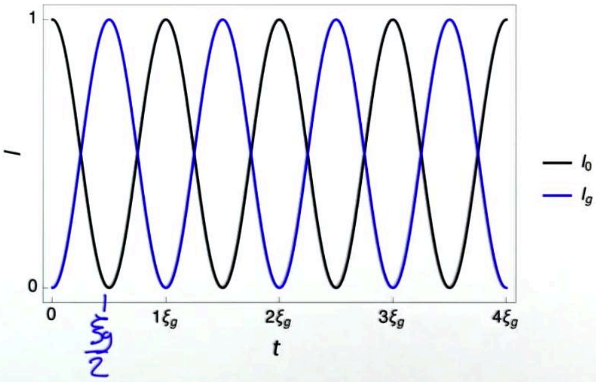
$$I_0 = 1 - I_g$$

Exact Bragg condition: $s = 0$

$$I_g = \sin^2\left(\frac{\pi t}{\xi_g}\right)$$

$$I_0 = \cos^2\left(\frac{\pi t}{\xi_g}\right)$$

$t = n\,\xi_g \Rightarrow I_g = 0$: "extinguished"



Transmission Electron Microscopy

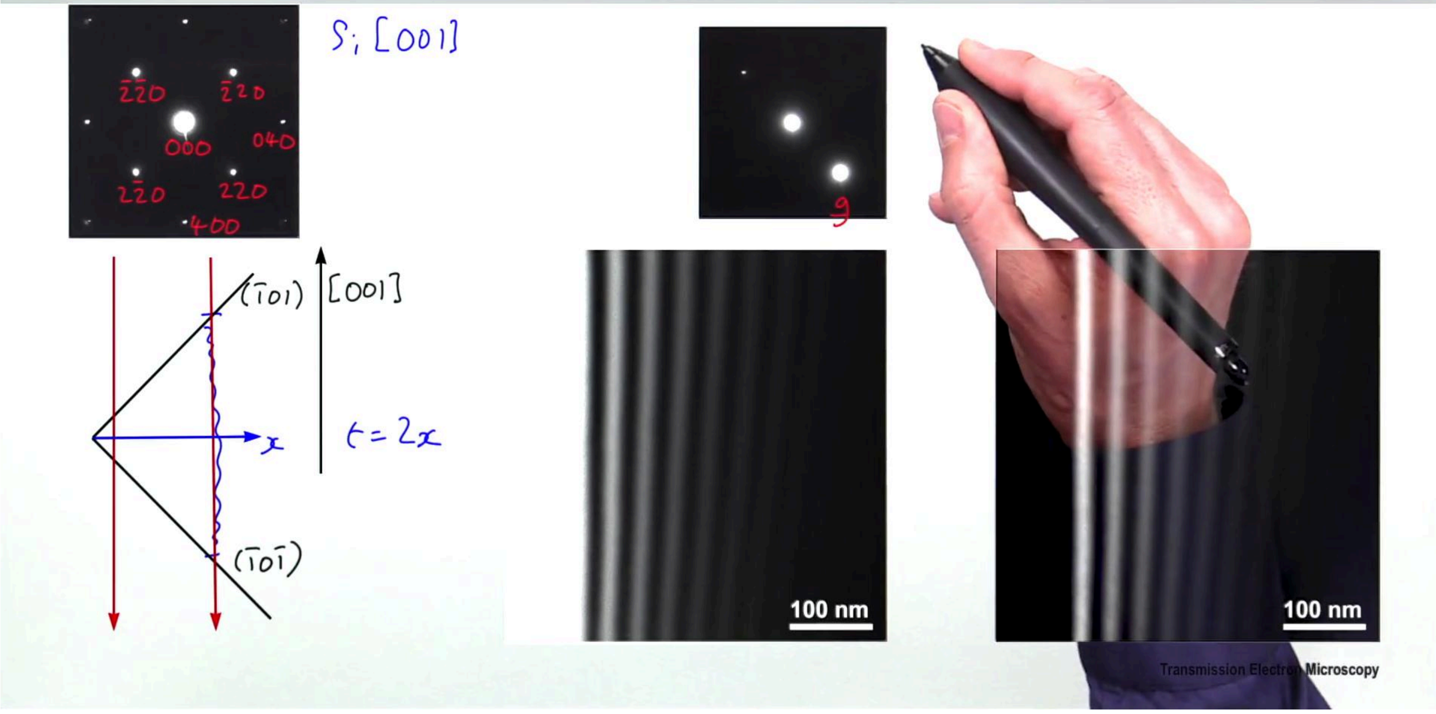
And that is why xi_g is called the extinction distance, because, when t equals xi_g, the intensity in this reflection g is extinguished. What would be nice is if we could measure these intensity modulations experimentally, and in the next slide, I will show how that can be done with simple bright field and dark field imaging.

Notes

Summary

4m 03s

2-beam imaging of thickness fringes

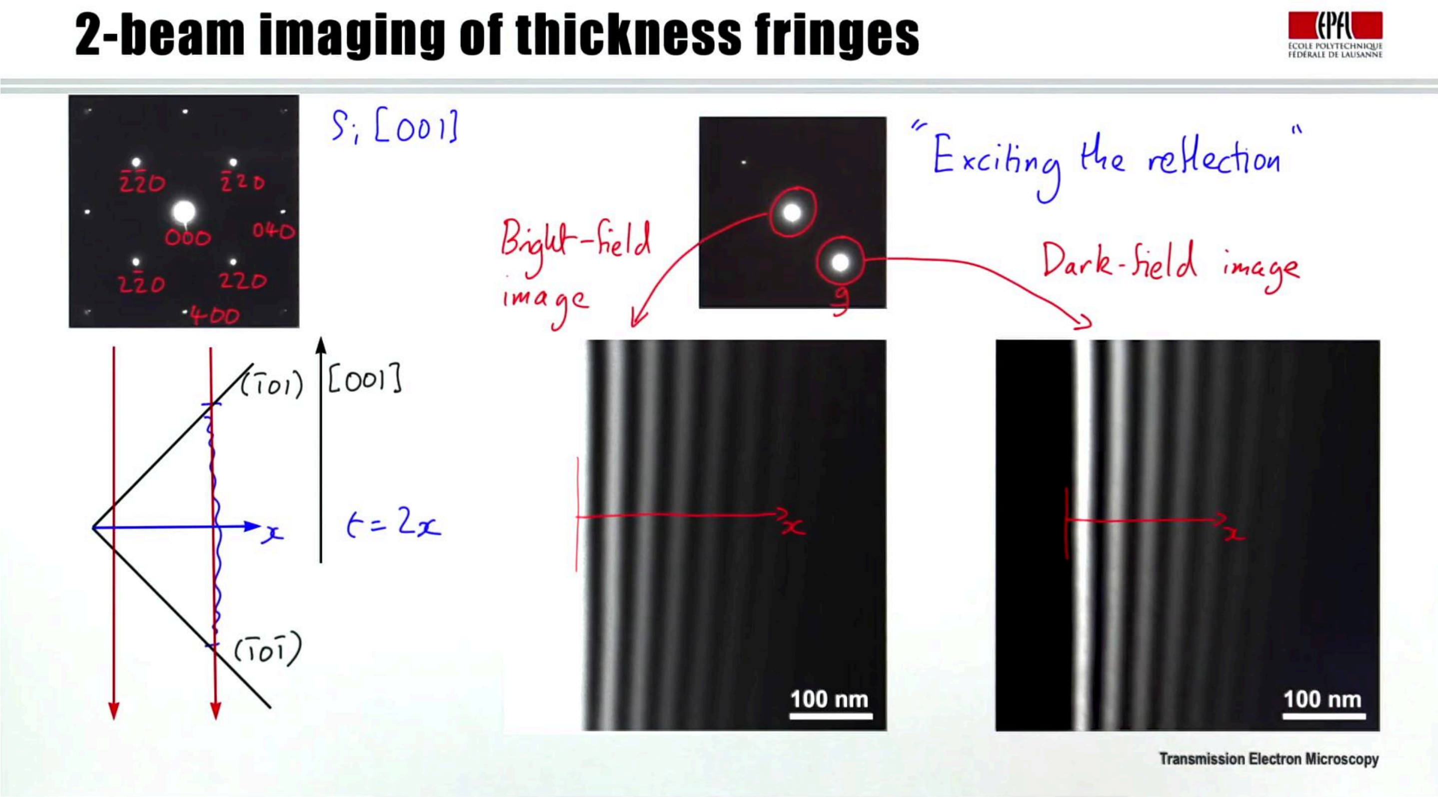


To demonstrate these intensity modulations with thickness, I use a sample of silicon with a basic zone axis of [0 0 1]. Here is the diffraction pattern on that zone axis, which can be indexed as follows. In the center: the direct beam 0 0 0; surrounded by some 2 2 0 type reflections, with further out 4 0 0 type reflections. The sample is a so-called “cleaved wedge”. That is, it has been made by cleaving the silicon on its {1 0 1} type planes. Thus, if we look in cross-section, the sample has a form like this: here we have the [0 0 1] axis, and this is solid material bound by these {101} type planes. We look at this sample in projection, with the electron beam traversing it as follows. Clearly, as we move away from the vacuum edge of the sample, the thickness in projection increases. Further, if we call the distance from the edge x, then, with this geometry, the thickness is given by the simple formula: t equals two times x. I have tilted the sample such that this 2 2 0 reflection is at the exact Bragg condition, giving this two-beam diffraction pattern here, where here is the direct beam, and this is g_220. Tilting in this way is known as “exciting the reflection”.

Notes

Summary

4m 26s

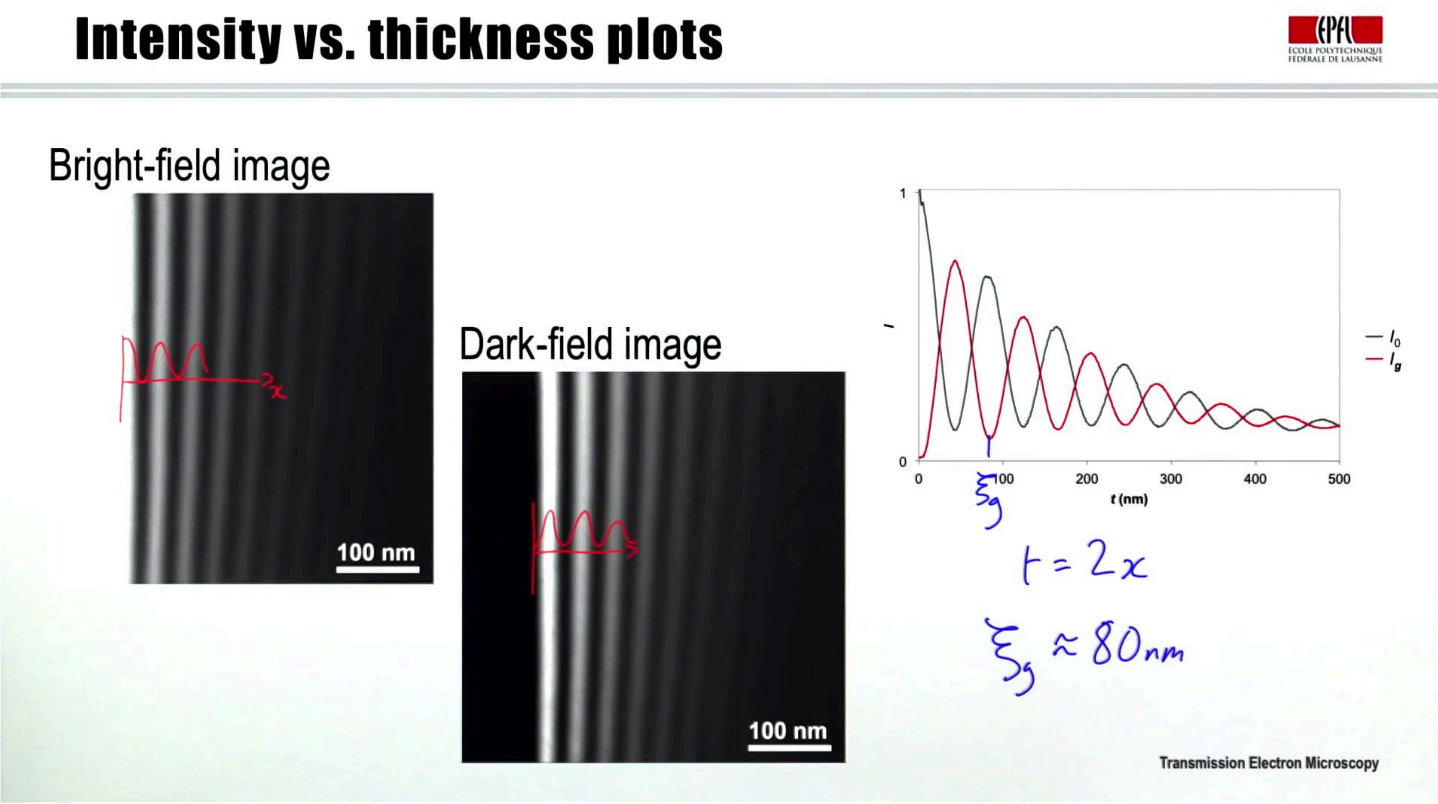


I have then inserted an objective aperture, in order to select just the direct beam and cut out the diffracted beam here. When we do this, effectively we spatially map the intensity in this direct beam, making this bright field image here. In this image, here is the vacuum and then here is the edge of the sample. So x measures this way here. And as we have seen, as x increases, so does the thickness of the sample. And clearly, with increasing thickness, we see a pattern of bright and dark fringes, as predicted by the formula for the intensity in the direct beam. I have then displaced the objective aperture, in order to select the diffracted beam. So now we make a dark field image, where we spatially map the intensity I_g in that diffracted beam. Similarly to the bright field image, we again see a pattern of bright and dark fringes but, as predicted, these fringes are an anti-phase with those in the bright field image. So this is the edge of the sample, this is x, and we can see that, in the vacuum of course it is dark, because there is no diffraction, and then at a certain thickness, when the bright field image is dark, the dark field image is bright.

Notes

Summary

6m 10s

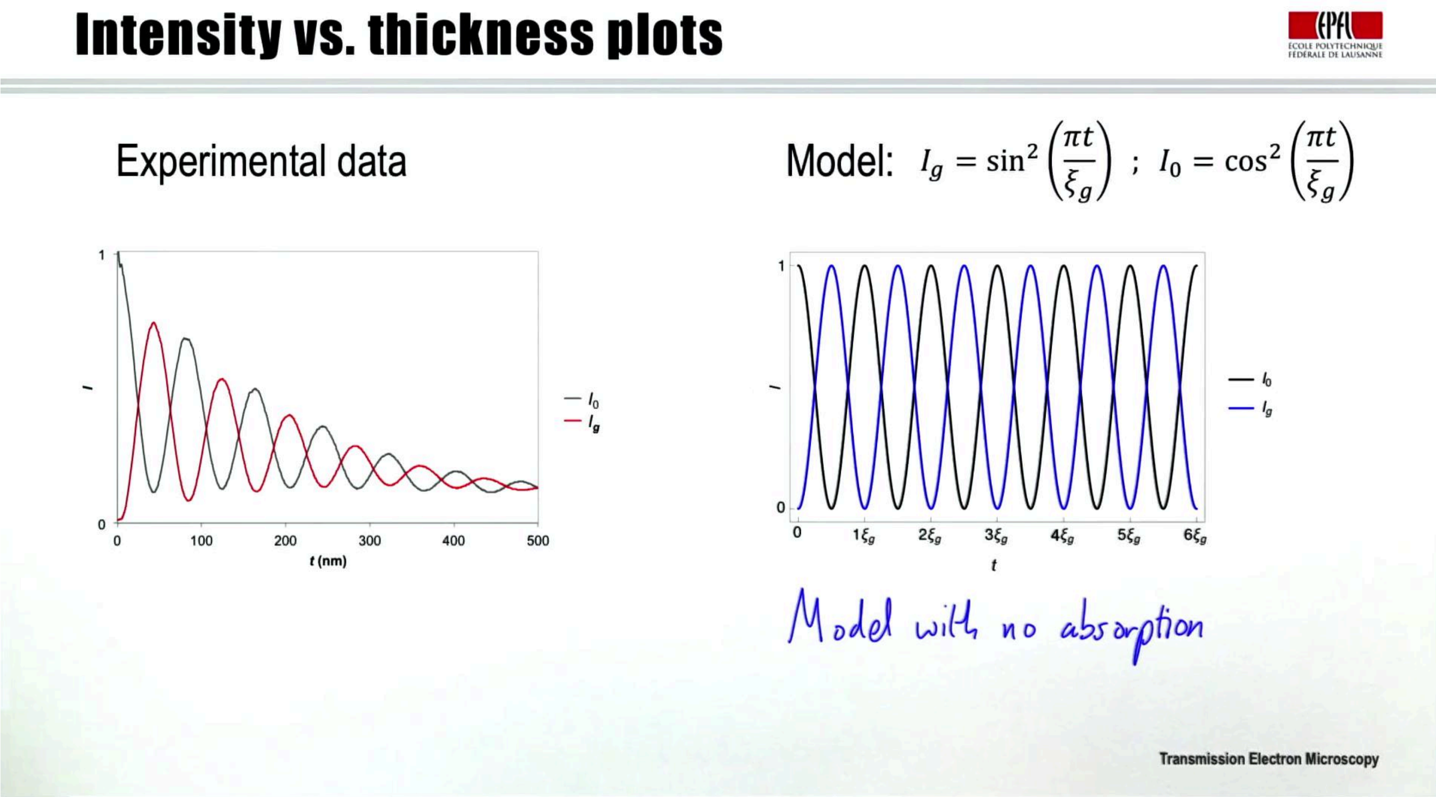


On this slide, I take this bright field and dark field image pair, and pull out the intensity profiles with respect to thickness. So looking at the bright field image, we can see that the intensity profile is bright here, descends to being dark and then it is bright and dark, and so on. I have taken this profile, and plotted it against thickness t, where I have simply said that t equals 2 x, as previously discussed, with x being that distance from the edge of the sample. I have done the same thing for the dark field image, taking its intensity profile, and plotted these two profiles on this curve here. Where, on the grey curve, I show the intensity in the direct beam, and in the red curve curve, I show the intensity in the diffracted beam. The intensity of both these curves has been normalized to the intensity in the vacuum in the bright field image. As predicted, these curves show sinusoidal modulations in anti-phase with each other. And if we look, we can see that the extinction distance xi_g is approximately equal to 80 nanometers. We now take this plot and compare it to the theoretical model from before.

Notes

Summary

7m 41s



That is what we do on this slide where here I show that experimental data with the intensity in the direct beam and the intensity in the diffracted beam, in grey and red respectively. Compared to that simple model for the intensities discussed earlier, where in the black curve we have the intensity in the direct beam and the blue curve the intensity in the diffracted beam. As predicted by the model, we have those sinusoidal modulations in anti-phase with each other. However, in the experimental data, the maximum intensity decays with thickness. Further, the minimum intensity increases a bit with thickness. The reason this is not seen in the model is because this simple model did not account for absorption effects. However, as sample thickness increases, as well as there being more dynamical scattering, there is also absorption of the transmitted electrons. And it is because of this that we see these changes in maximum and minimum intensity as thickness increases.

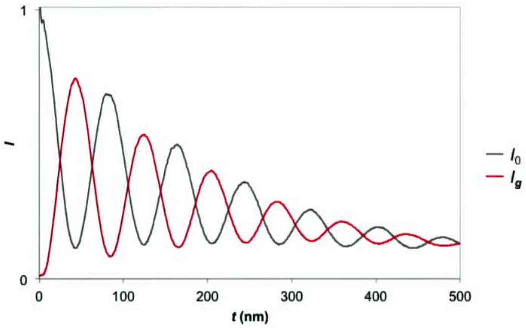
Notes

Summary

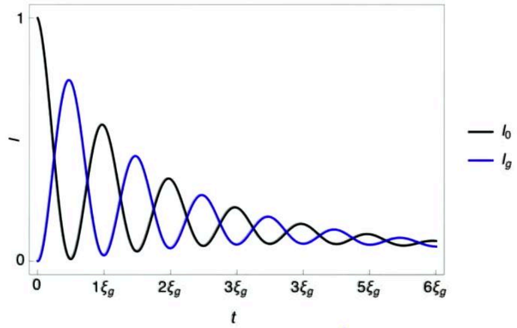
9m 02s

Intensity vs. thickness plots

Experimental data



Model (with absorption)



$\xi_0' = 10\xi_g \ , \ \xi_g' = 15\xi_g$

Transmission Electron Microscopy

To demonstrate that, here I have changed the model to account for absorption in a rather simple way. Specifically, I have introduced imaginary parts of the extinction coefficient, both for the direct beam and the diffracted beam. Where for the direct beam I have introduced an imaginary part of extinction distance xi_0-prime equal to ten times xi_g and I have given the diffracted beam an imaginary coefficient, xi_g-prime equal to 15 times xi_g. Once I have accounted for absorption in this rather simple way, we can see that the model is strikingly similar to the real experimental data.

Notes

Summary

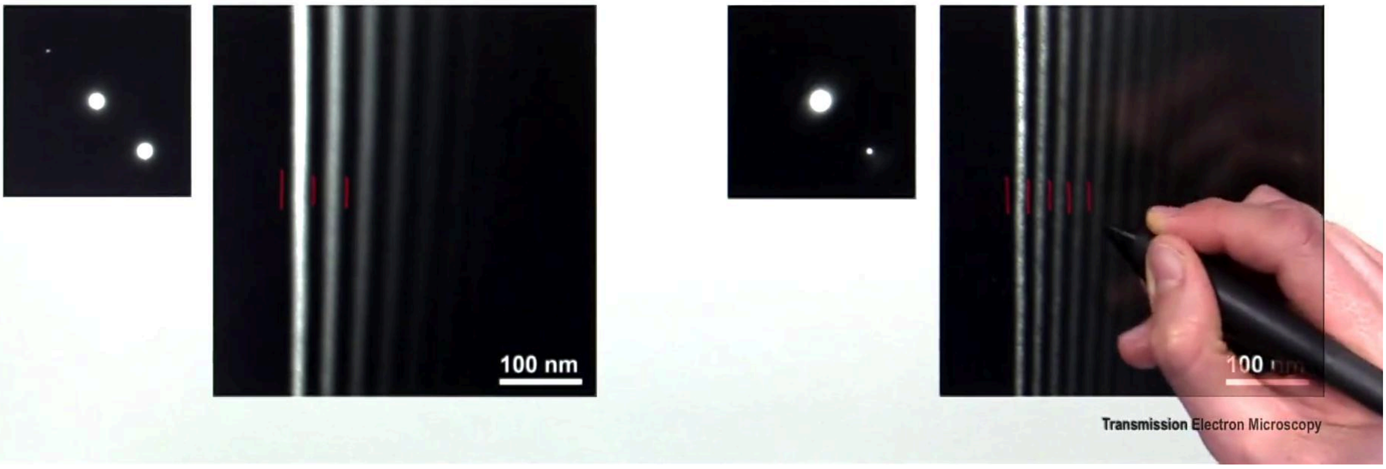
10m 09s

Thickness fringes as function of *s*

$$I_g = \frac{1}{(1+\xi_g^2 s^2)}\sin^2\left(\pi t \underbrace{\sqrt{\frac{1}{\xi_g^2}+s^2}}_{s'}\right)$$

$s > 0$: s' increases

$s = 0$



These bright and dark fringes are typically known as “thickness fringes”, although they are also sometimes called “extinction contours”. So far, we have looked at the case of the exact Bragg condition when s equals zero and the diffracted beam is strongly excited. However, we can ask: what happens when we change the value of s, in this case where I make s greater than zero? Looking at the diffraction pattern from the general region of interest, we can see that the intensity in that diffracted beam has decreased compared to the s equals zero case, because we are no longer at the exact Bragg condition. If we consider this quantity here, sometimes known as s prime or the effective excitation error, then clearly when s is greater than zero, this quantity is larger than when s equals zero. Thus, the intensity in the diffracted beam will be extinguished at smaller thickness variations than the s equals zero case. And that is exactly what we see here, in this dark field image made from g with s greater than zero. Compared to the s equals zero case, it is clear that the separation between these thickness fringes has decreased.

Notes

Summary

11m 00s

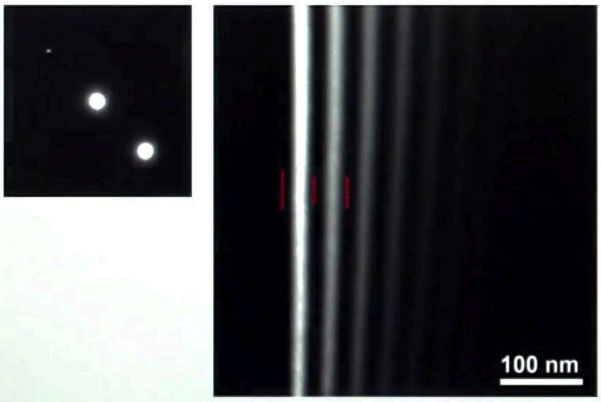
Thickness fringes as function of *s*

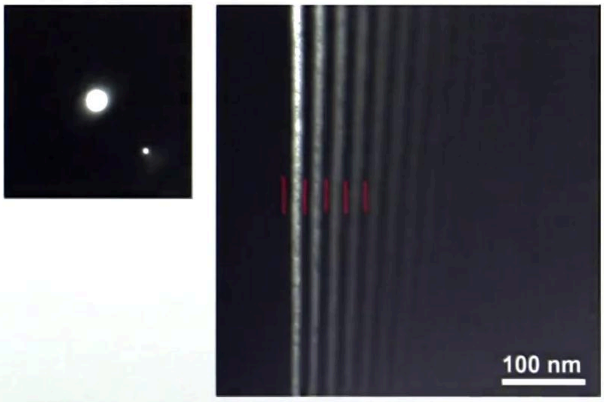
$$I_g = \frac{1}{(1+\xi_g^2 s^2)} \sin^2\left(\pi t \underbrace{\sqrt{\frac{1}{\xi_g^2} + s^2}}_{s'}\right)$$

$s > 0$: s' increases

$I_g = 0$ for $t \approx 45\,nm$

$s = 0$





Transmission Electron Microscopy

Indeed, whereas before we had extinction of the diffracted beam for a thickness of about 80 nanometers, now we have extinction for a thickness of just 45 nanometers. In this case I have increased the excitation error s such that s squared is about the same order of magnitude as one over xi_g squared. But we can ask what happens if I make s squared much larger than xi_g squared? That is what we discuss on the next slide.

Notes

Summary

12m 28s

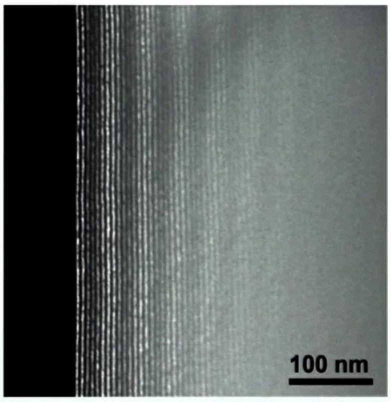
Thickness fringes: weak beam imaging

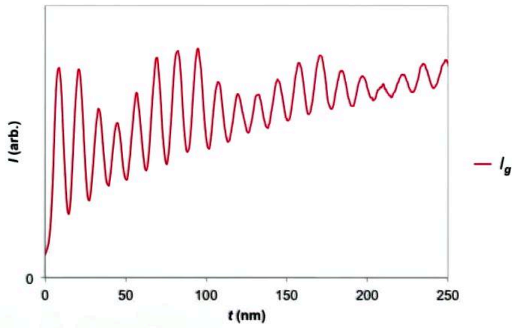
$$I_g = \frac{1}{(1+\xi_g^2 s^2)} \sin^2\left(\pi t \sqrt{\frac{1}{\xi_g^2} + s^2}\right)$$

If $s^2 \gg \frac{1}{\xi_g^2} \Rightarrow I_g \propto \sin^2(\pi t s)$

$s \approx 0.075\,\mathrm{nm}^{-1}$

"g-3g"





Transmission Electron Microscopy

We can see that, if we can deviate sufficiently from the perfect Bragg condition, we can make s squared much larger than one over xi_g squared. In this case, one over xi_g squared plus s squared is simply approximately equal to s squared. Thus, the intensity in the diffracted beam becomes proportional to: sine squared of pi t s. This condition is known as a weak beam imaging condition, one that is often used for analysis of defects such as dislocations. On this slide, I have set up such a weak beam imaging condition, indeed in this case, what is known as a g - 3 g condition. On the diffraction pattern, here is my reflection g. However, what I have done is, instead of putting g at the exact Bragg condition, I have put 3 g here at the exact Bragg condition. If we consider the Ewald sphere construction with the reciprocal lattice – here is the direct beam and here is 3 g – the Ewald sphere intersects that 3 g exactly. Now if I mark in the g and also the 2_g, we can see that reflection g has a large and positive excitation error s here, making this week beam condition, where s squared is much larger than one over xi_g squared. Indeed in this experimental case, s is approximately equal to: 0.075 nanometers to the minus one.

Notes

Summary

13m 00s

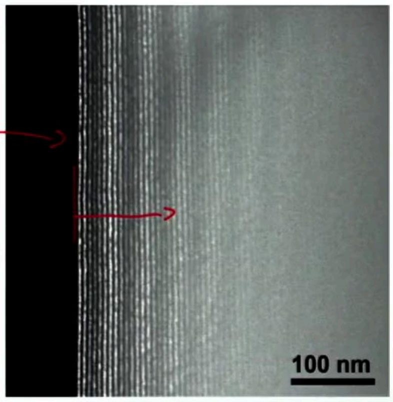
Thickness fringes: weak beam imaging

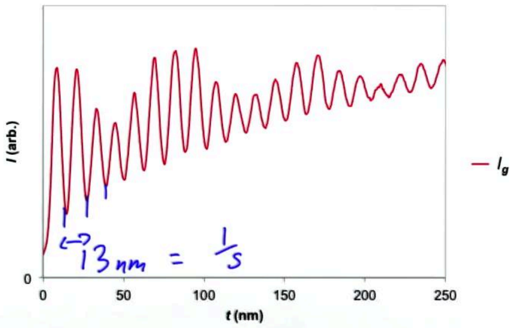
$$I_g = \frac{1}{(1+\xi_g^2 s^2)} \sin^2\left(\pi t \sqrt{\frac{1}{\xi_g^2} + s^2}\right)$$

If $s^2 >> \frac{1}{\xi_g^2}$ => $I_g \propto \sin^2(\pi t s)$

$s \approx 0.075\,nm^{-1}$

"g-3g"





Transmission Electron Microscopy

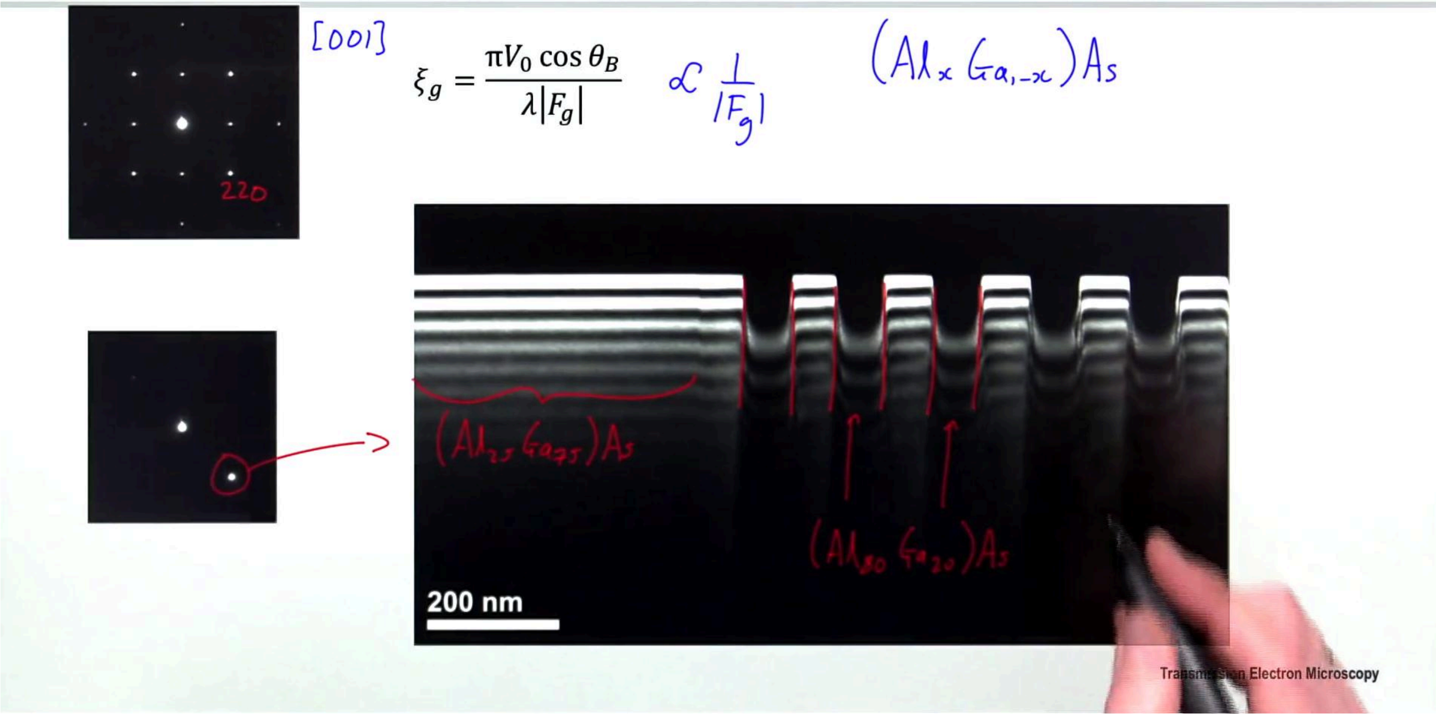
Now looking at the dark field image made from that reflection g, we can see that, compared to before, there is an extremely dense frequency of bright and dark thickness fringes. Pulling out an intensity profile, with respect to the thickness, we can see that here we have very frequent dark fringes at spacings of about one every 13 nanometers, where that distance corresponds to one divided by s, this intentionally large excitation error.

Notes

Summary

14m 48s

Thickness fringes as function of F_g



As well as being a function of the excitation error s, if we consider the formula for the extinction distance here, we can see that thickness fringe frequency will also be a function of F_g, the structure factor for the reflection, such that xi_g will be proportional to one over F_g. This is a case that I illustrate on this slide using a sample of a cleaved wedge not now of silicon but of aluminum gallium arsenide, or AlGaAs as it is sometimes known, where the aluminum and gallium ions can substitute for each other on the same atomic sites. The sample again has a basic zone axis of [0 0 1], and I have excited this 2 2 0 reflection here, giving this two-beam pattern here. And then made a dark field image from that excited reflection. On the left of this dark field image, we have thickness fringes in the substrate, where the substrate has a composition of about 25 percent aluminum and 75 percent gallium on these shared atomic sites. On this substrate, quantum wells have been deposited which are perfectly epitaxial. However, they have a different composition. In these quantum wells, we instead have a composition of about 80 percent aluminum and 20 percent gallium on the shared sites.

Notes

Summary

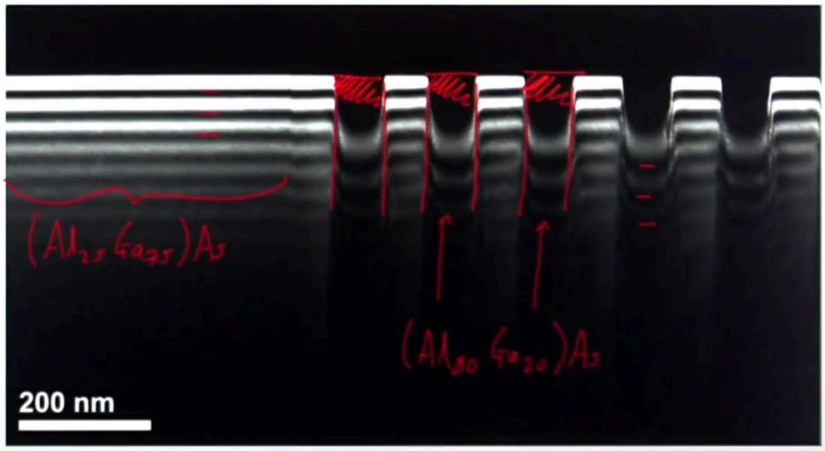
15m 24s

Thickness fringes as function of F_g

[001]

$$\xi_g = \frac{\pi V_0 \cos\theta_B}{\lambda|F_g|} \propto \frac{1}{|F_g|}$$

$(Al_x Ga_{1-x})As$



Transmission Electron Microscopy

If we look closely at the thickness fringes in the quantum wells we can see the fringe separation is larger than the fringe separation in the substrate. In other words, the quantum wells have a larger extinction distance than the substrate. This is reasonable because aluminum scatters less than gallium. So it has a smaller atomic scattering factor than gallium. Thus, as the percentage of aluminum increases, the atomic scattering factor from this shared site decreases, decreasing the structure factor for that reflection and increasing the extinction distance. Another thing you might notice is that the onset for the thickness fringes is further away from the edge of the sample in the quantum wells than in the substrate. That is due to another effect. These quantum wells oxidized badly, thus the first part of the thickness is amorphous oxidized material and does not count to the crystalline dynamical diffraction scattering effect needed to make the thickness fringes.

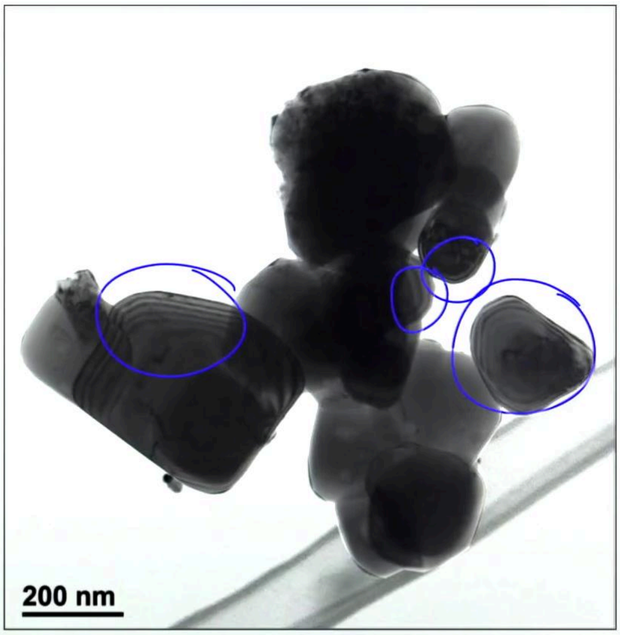
Notes

Summary

17m 08s

Thickness fringes in nanocrystals

$BaTiO_3$ powder



Thickness fringes

Transmission Electron Microscopy

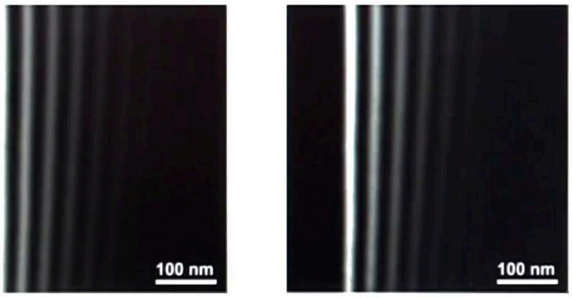
This cleaved wedge sample geometry which I have used to illustrate thickness fringes is little used in regular TEM analyses. Thus, in a way, these illustrations have been rather academic. However, thickness fringes will regularly be observed in regular TEM imaging. Here, I show a bright field image of barium titanate nanocrystals. The crystals are randomly orientated with respect to the electron beam. We can see that some of the crystals have rather low contrast. These crystals must be in a weak scattering condition, far away from a low index zone axis. However, we have other particles which are darker, showing strong diffraction contrast. The particles have varying thickness and, being in a strong diffraction condition, they have increased dynamical scattering, and we can see that as the thickness changes away from the edge, we see thickness fringes here, in this particle here, on this edge here and this edge here. This illustrates how you may well see thickness fringes from dynamical scattering in regular TEM data of, for instance, such randomly oriented nanoparticles, or maybe nanorods, or other crystalline objects of strongly varying thickness.

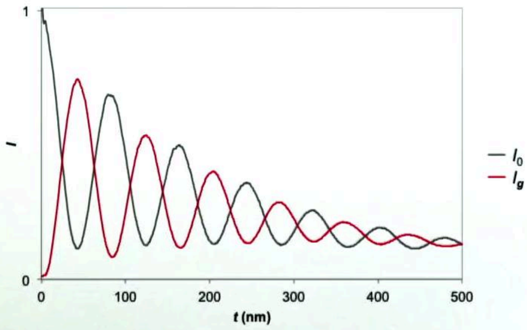
Notes

Summary

18m 18s

Dynamical effects – thickness fringes summary





- I_0 & I_g measured by 2-beam bright-field & dark-field imaging
- At $s = 0$, I_0 & I_g modulate & are in anti-phase with t, giving thickness fringes/ extinction contours on wedge-shaped sample
- Thickness fringe frequency with t also function of s, F_g

Transmission Electron Microscopy

To summarize, in a two-beam condition, I have measured the intensity in the direct beam I_0, and the intensity in the diffracted beam I_g, using bright field and dark field imaging. At the perfect Bragg condition, with s equals zero, we see that I_0 and I_g modulate sinusoidally, and are in anti-phase with each other with increasing thickness, giving bright field and dark field images such as these, where these patterns of bright and dark fringes are known as thickness fringes, or extinction contours. We further see that, when we pull out the intensity profiles of I_0 and I_g on these wedge shaped samples, that the profile matches very nicely a model which accounts for some absorption effects. I have gone on to show that, if we increase s, then the frequency of these thickness fringes increases, in keeping with theory. Finally, with an example of barium titanate nanocrystals, I have shown how such thickness fringes are often observed in regular TEM data, when your image samples of strongly varying thickness.

Notes

Summary

19m 49s

Dynamical effects – thickness fringes summary

In the next lecture, I'm going to continue on this theme of dynamical effects and TEM imaging, this time looking at what happens when our crystalline sample is deformed or bent, giving rise to the appearance of bend contours in bright-field and dark-field images.

Notes

Summary

21m 02s