Keywords: Fresnel reflectivity, Roughness parameter, Debye-Waller factor, Magnetic roughness

In this lecture, we will continue with the techniques of neutron reflectometry. And before we get to any experimental data or examples, it is important that I discuss the way we handle the data, or the way we fit them i.e. the way we evaluate the reflectivity pattern for a given model structure. With this aim, I continue.
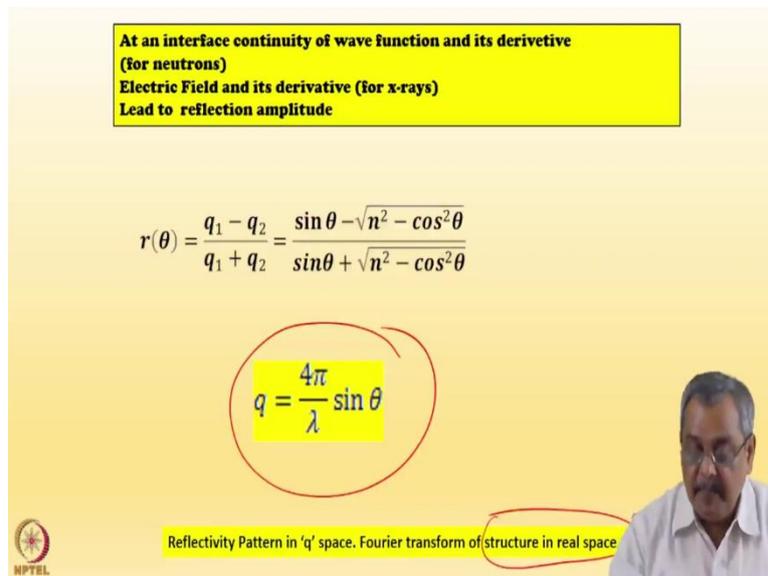
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Earlier I derived the expression for reflectivity at the interface of a single infinite medium while considering the continuity of the wave function and its derivative at the interface. I will just repeat it quickly. So, if the incident beam is of unit amplitude, $e^{iq_1 z}$, and since the reflected beam is going in opposite 'Z' direction then with reflectivity, r, it is, $re^{-iq_1 z}$. Along with this, we also have the transmitted part, $te^{iq_2 z}$, where t is transmission amplitude. Here, q_1 and q_2 are wave vector transfers inside and outside the medium, respectively.

From this, continuity of wavefunction and its derivative across the interface gives us (with z=0),

$$1 + r = t$$

$$q_1(1 - r) = q_2 t$$

From these two equations we can find that the reflection amplitude is,

$$r = \frac{q_1 - q_2}{q_1 + q_2}$$

Using the relation, $q = \frac{4\pi}{\lambda}\sin\theta$ we can get this expression for r in terms of angle.,

$$r = \frac{\sin\theta - \sqrt{n^2 - \cos^2\theta}}{\sin\theta + \sqrt{n^2 - \cos^2\theta}}$$

Like all other diffraction experiments we will be getting the reflectivity pattern in q space. Actually, we plot the reflectivity as a function of either angle or q, momentum transfer, and from there we have to obtain the structure in real space. So, the general part of the task is similar to what you have done for all other diffraction patterns.

We know that $n = \cos \theta_c$. Using this and assuming that θ is small we can write,

$$r(\theta) \cong \frac{\theta - \sqrt{\theta^2 - \theta_c^2}}{\theta + \sqrt{\theta^2 - \theta_c^2}}$$

Here, we have two cases.

i) When $\theta < \theta_c$, $\theta^2 - \theta_c^2$ is negative and so, $\sqrt{\theta^2 - \theta_c^2}$ is imaginary number, say, $i\beta$. In this case, reflection amplitude can be written as,

$$r(\theta) \cong \frac{\theta - i\beta}{\theta + i\beta}$$

and reflected intensity (I) turns out to be,

$$I = r * r^* = 1$$

It signifies the total external reflection from the interface depending on material properties as θ_c is dictated by the density in the medium. So, up to the critical angle reflected intensity is equal to 1.

ii) When $\theta \gg \theta_c$, then I can write

$$\theta - \sqrt{\theta^2 - \theta_c^2} = \theta - \theta\left(1 - \frac{\theta_c^2}{\theta^2}\right)^{1/2} \cong \frac{\theta_c^2}{2\theta}$$
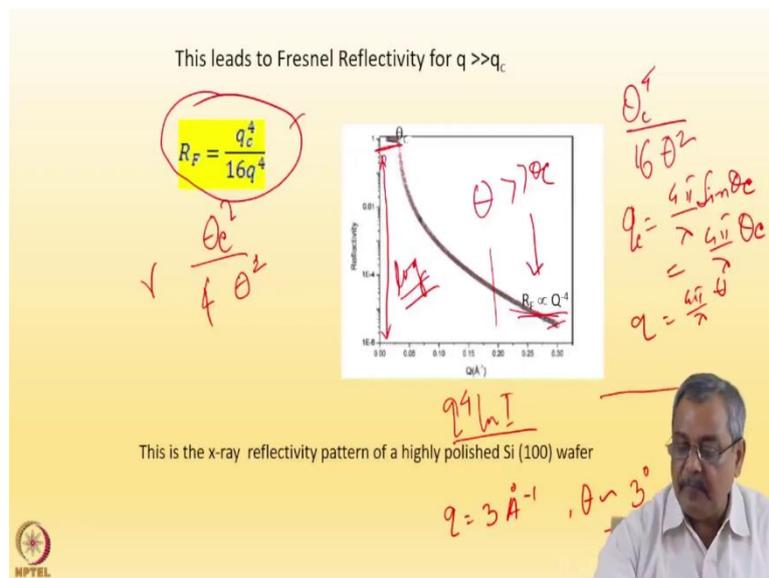
Similarly,

$$\theta + \sqrt{\theta^2 - \theta_c^2} \cong 2\theta$$

So that, reflection coefficient,

$$r(\theta) \cong \frac{\theta_c^2}{4\theta^2}$$

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And reflected intensity in this case is,

$$I = r * r^* = \frac{\theta_c^4}{16\theta^4}$$

Using the relation $q = \frac{4\pi \sin\theta}{\lambda}$, then for larger angles,

$$I = r * r^* = \frac{q_c^4}{16q^4}$$

When $\theta \gg \theta_c$, I have got this region known as Fresnel reflectivity (R_F) where reflectivity falls as q^4 that means if the q is doubled your intensity falls by 16 times.
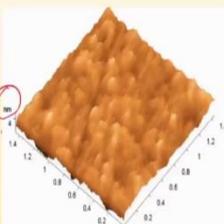
Now, let me point it out to you that because reflectivity falls so rapidly with q and we want to fit the reflectivity data over a reasonably large q range, so, we plot the reflectivity in log scale and our fits are also shown in log scale. Because if I plot it in linear scale, the intensity in this region will be 10^{-4}, compared to 1, and they will get very little weightage when I am trying to fit the data because they are almost in the background. That is why we use log scale for plot of reflectivity.

All the data that I will be showing, you will find mostly it is in log scale or sometimes data will be multiply by q^4 to normalize with respect to Fresnel reflectivity. That is also done often. But in general, we plot reflected intensity in log scale to bring the weightage of the points across the reflectivity profile to similar values.

This is the Fresnel reflectivity for a single layer. But, this calculation has been done assuming that the interface is absolutely flat. Now let us look at a real interface. There is roughness at the interface, and I want to show you this AFM image of a surface.
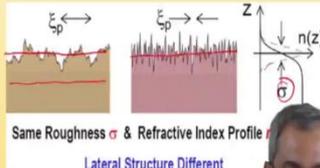
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This is the 3-dimensional AFM picture of a nickel film where height is shown in nanometers. We can see that this is not flat and actually you can see that this height is in a scale of about 4 nanometers. So, the interface is not flat in general, and rather it has got a structure like this which is fluctuating in height. In this fluctuating height, I can assume a mean height and a fluctuation around that value, a Gaussian fluctuation. Pease note that I can talk about this as a Gaussian, with a full width at half maxima, σ, which is a signature of this fluctuation or the roughness.

But this length scale has to be much less of the fluctuation than the linear resolution q or the coherence length of the incident radiation. Because the coherence length dictates up to what distance two rays will interfere, and this length scale of fluctuation has to be smaller than that to invoke this assumption.

$$R = R_F e^{-q^2 \sigma^2}$$

I call it a Debye-Waller like factor. I will remind you that we got a similar parameter when we talked about thermal agitations of atoms in a lattice.

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There is roughness at the interface

AFM of a Ni film
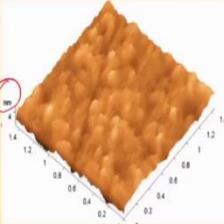
σ = Roughness

σ is the roughness parameter

$$R = R_F e^{-q^2 \sigma^2}$$
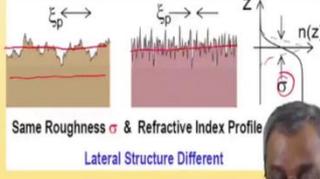
Debye-Waller like

Same Roughness σ & Refractive Index Profile
Lateral Structure Different

'σ' is a parameter, one obtains from reflectometry

If you remember, I talked about thermal agitations of a crystal lattice, they grow larger as the thermal agitation takes place because of temperature, and we wrote down $I = I_0 e^{-Q^2 u^2 / 3}$. $1/3^{rd}$ factor is there because $< \cos^2 \theta \geq 1/3$ for a sphere. This expression we also use it in the Guinier region in SANS where we have used, $I = I_0 e^{-Q^2 R_G^2 / 3}$, where R_G is the radius of gyration of the object. So, whenever we have a fluctuation around a mean we use such relation. Here R_G is basically the length scale over which the object is there. You can find out the distance, and then the R_G gives the radius of gyration. In case of actual dynamic Debye-Waller factor, the time average picture gives me $u^2 / 3$, because the atom is now oscillating in all possible directions and that oscillation has an average amplitude of $u^2 / 3$.

Now in $R = R_F e^{-q^2 \sigma^2}$, σ is roughness parameter. This is a case of static fluctuation where there is an average fluctuation around a mean value. And that is why I called it Debye-Waller like. We are not going to derive it. I have derived the Fresnel reflectivity, and this is the Debye-Waller like factor given there is a roughness of the surface. This roughness parameter also one obtains from the reflectometry data. And I will show you how it is obtained by fitting reflectometry data.

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Let me just quickly tell you that I talked to you about a medium which offers a potential to the neutron beam passing through given by $V(r) = \frac{2\pi\hbar^2}{m} b_{coh}\rho$. This expression has come from the coherent scattering length density.

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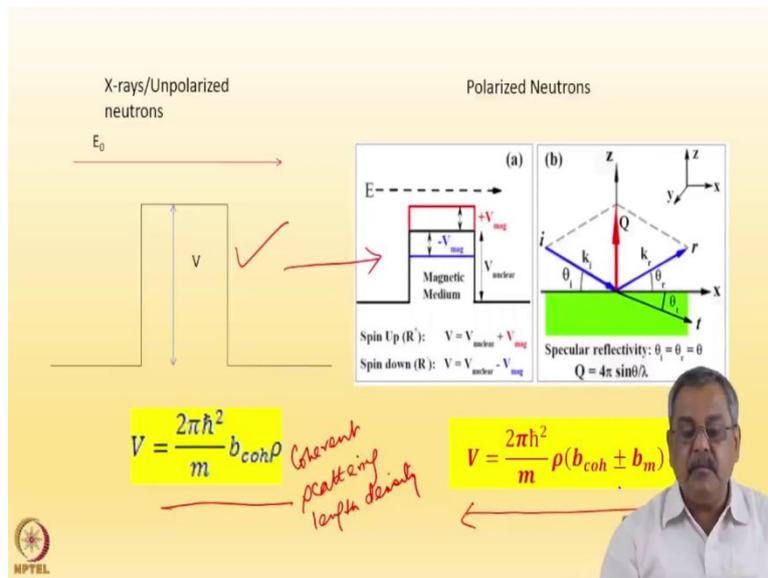
This expression is for a non-magnetic medium. If I have a magnetic medium that is magnetized, then there is a magnetic scattering length density also. We call it b_m. In this case of magnetic medium, the potential will be $V(r) = \frac{2\pi\hbar^2}{m}(b_N \pm b_m)\rho$, where b_N is the coherent scattering length offered by the nuclear potential and 'b_m' is the magnetic scattering length, summed over all the scatterers in a unit volume. That is why ρ, the number density comes into picture and we are talking in terms of scattering length density. We have a magnetic scattering length which is either added or subtracted from the nuclear scattering length depending on the magnetization direction in the medium and the magnetization direction of the neutron, whether they are parallel or anti-parallel. Hence, the magnetized medium offers a potential which is either more or less compared to nuclear component, depending on the polarization of the neutron and polarization of the medium. This is the beauty of neutron scattering. You do not see this in x-rays.

As I stated earlier, this (the left-hand side picture) is from an unpolarized medium, unpolarized neutron reflectometry. But when you have a magnetized medium and a polarized neutron beam (the right-hand side picture), you can see that if this is the nuclear potential, you have either $+V_{mag}$ or $-V_{mag}$ and we have $V = V_N + V_{mag}$ for spin up neutrons, and $V = V_N - V_{mag}$ for the spin down neutrons. The potential energy is given as a sum of the two.

I have also discussed earlier about the specular reflectivity when reflected beam makes the same angle as the incident beam on the surface and follow the Snell's law. Here, we also have a transmitted beam. We also discussed about the critical angle and since the V is different for

up and down neutron, so now for polarized neutron beam, my critical angles for up (+) and down (-) neutrons will be different.

Let us say this is the intensity *vs q* plot for unpolarized neutron then I can say that in case of polarized beam, for up spin we will have one reflected intensity, for down spin we have another reflected intensity, and this difference depends on the magnetic moment density in the medium.
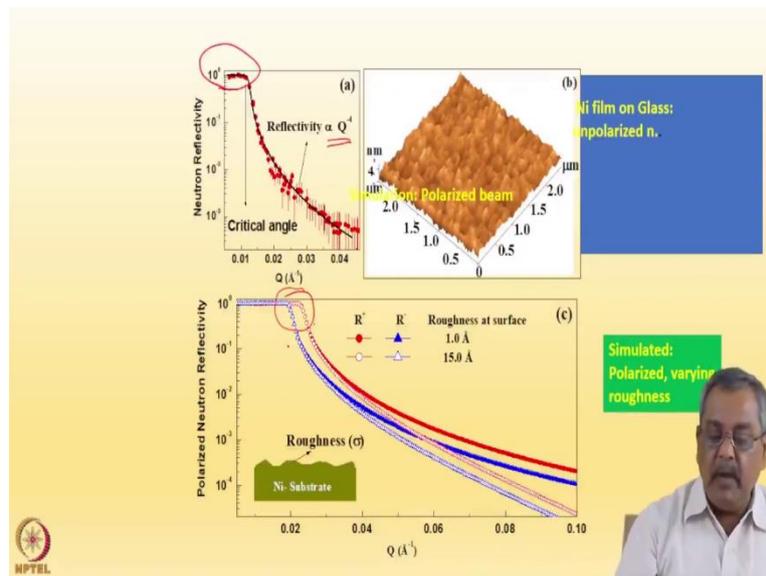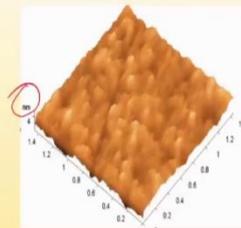
Let me just show you some examples. Now adding roughness and nuclear potential, I have shown you neutron reflectometry a nickel film on glass. It is the data taken in our instrument at Dhruva. You can see that it has got a flat edge, normally for any reflectivity experiment this intensity, below critical edge is taken as 1. Then we have the fall in the reflectivity profile which follows q^{-4} law. This is an experimental data which matches nicely with the Fresnel reflectivity profile.

And here, I have simulated reflectivity profile from the same nickel film. There are two different neutron reflectivity profiles for neutron polarizations up and down, R^+ and R^-. You can clearly see the difference here in the critical angle of the two beams.
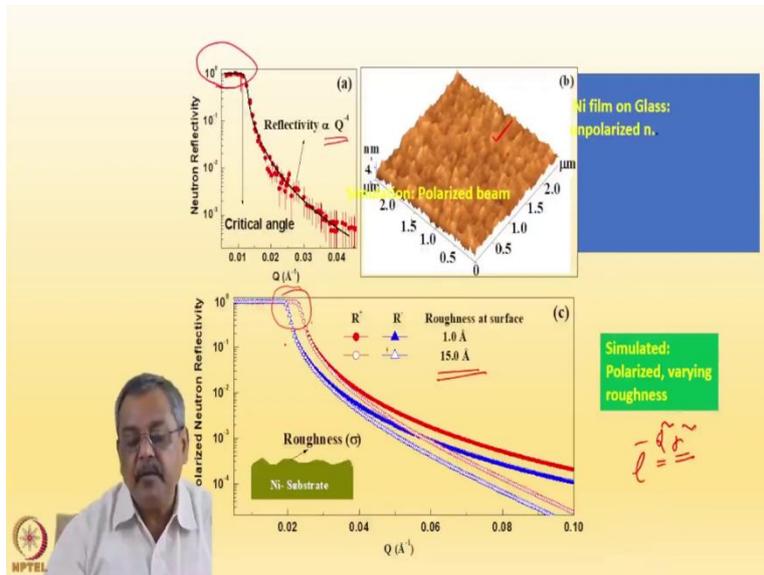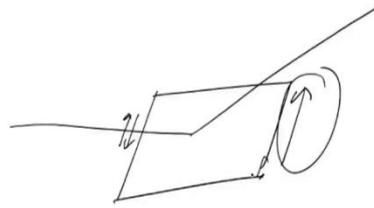
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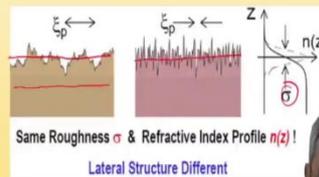

AFM of a Ni film

σ is the roughness parameter

$$R = R_F e^{-q^2\sigma^2}$$

Debye-Waller like

Same Roughness σ & Refractive Index Profile n(z) !

Lateral Structure Different

'σ' is a parameter, one obtains from reflectometry

The way the experiments are done is, as following. The sample surface is magnetized in some direction. You have a neutron beam incident on it and reflected. Now, this neutron, incident neutron beam has a spin, either parallel or antiparallel, with respect to the sample magnetization. And that gives me two reflectivity patterns for different neutron spin polarization directions, R^+ and R^-.

Over and above, the difference in critical angle, this pattern has been simulated for two different interface roughness of 1 angstrom and interface roughness of 15 angstrom. Now, you can see the effect of this Debye-Waller factor here. You can see even for the up polarized neutron, as you go to larger and larger q, the reflectivity falls much faster because of $e^{-q^2\sigma^2}$ factor where σ is the interface roughness. Same is true for the down spin neutrons. Up to some point, they follow the same intensity, but then for the roughness of 1 angstrom the intensity falls much less rapidly than roughness of 15 angstrom.

This shows that from our experiments, I can find out the roughness parameters, from the critical angle I can find out the physical and magnetic moment density of the film.