Neutron Scattering for Condensed Matter Studies
Professor Saibal Basu
Department of Physics
Homi Bhabha National Institute
Week 9: Lecture 23C

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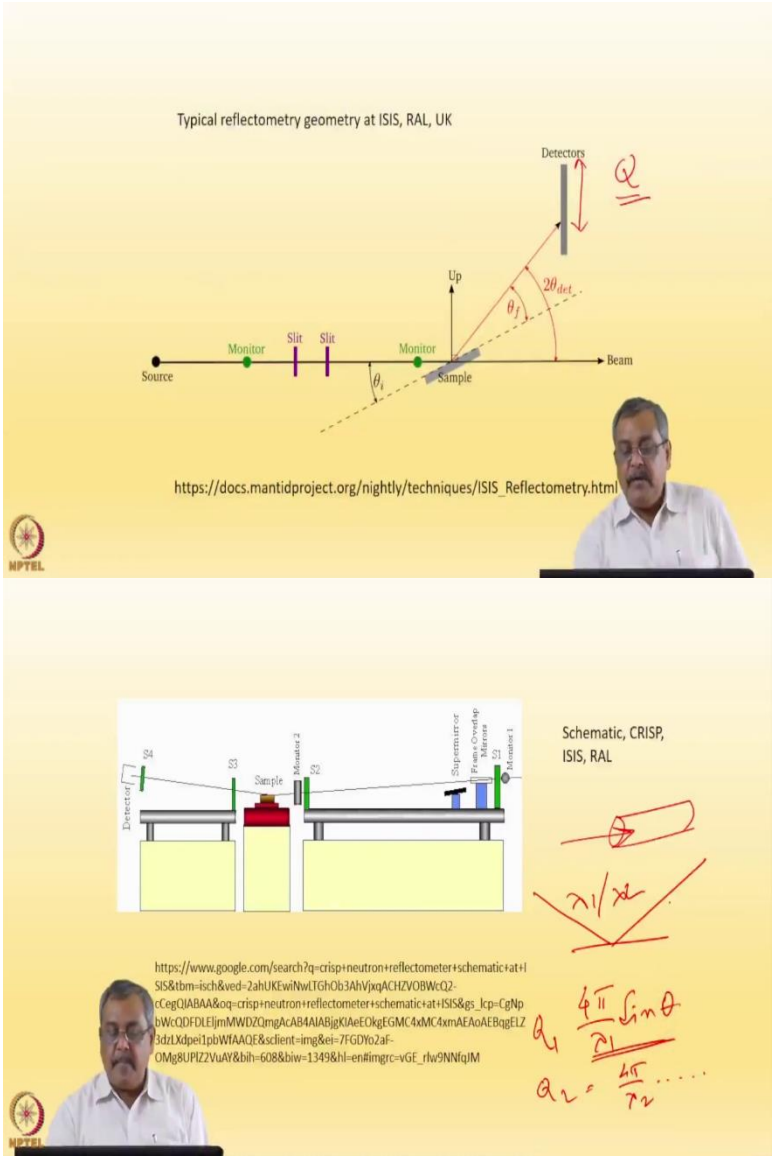

This is a typical design of a reflectometer at ISIS, RAL. This is the CRISP reflectometer. Basically, you tip the beam onto the (horizontal) sample and then the detector catches (the reflected beam) and because it is a polychromatic (incident) beam. Even if it is an end-on detector you can get large *Q* (range) information from the time of flight (ToF). Because this is a polychromatic (incident) beam, the time of flight (ToF) dictates what is the Q value. The

angle remains same for all the wavelengths. As $Q = \frac{4\pi}{\lambda} \sin\theta$, so, for the same angle, based on the time of flight (or λ), you have different Q values.

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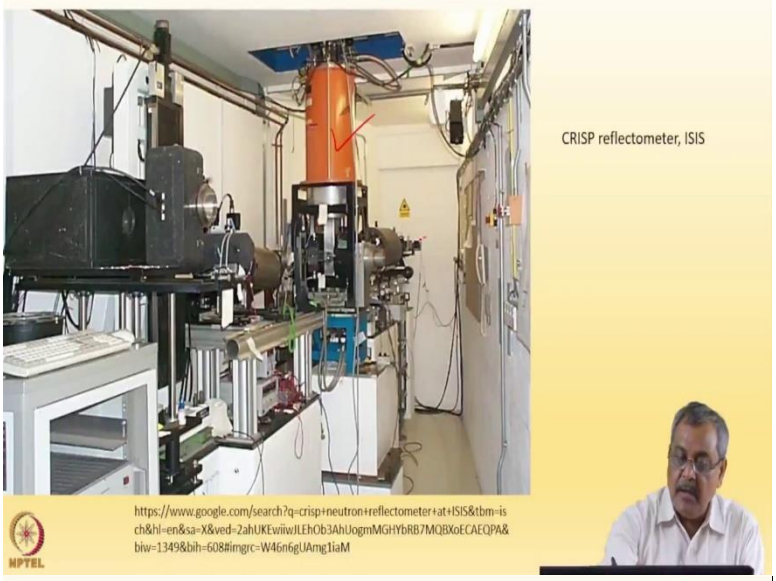

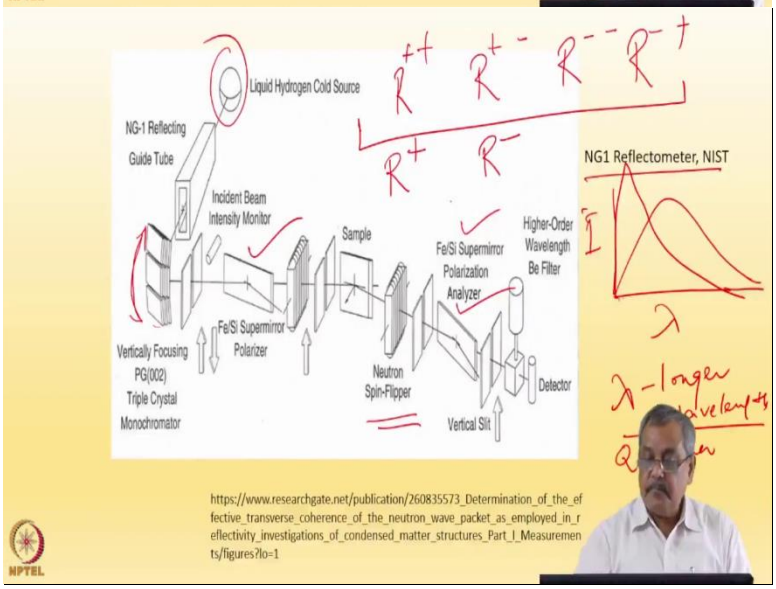

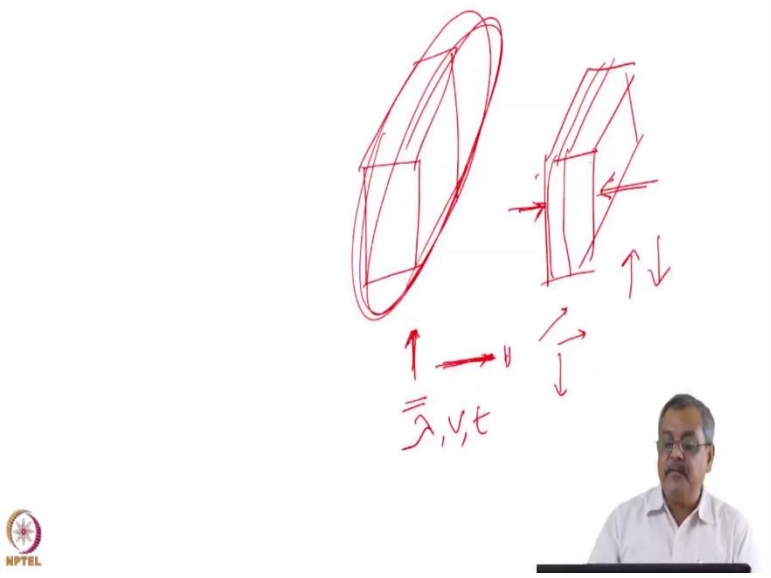

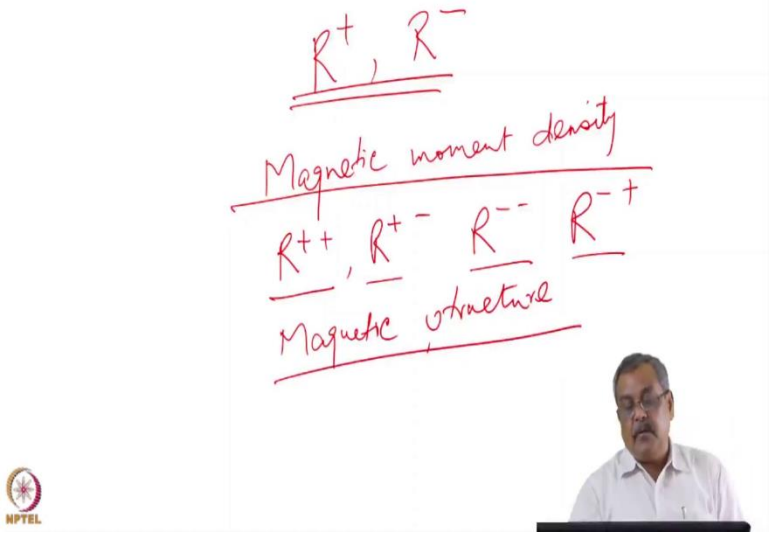

This is the CRISP reflectometer as you can see. Generally, in all the magnetic reflectometers, there will be a large cryostat which is also (seen) over here. The detector is here and the beam comes from the spallation source from this direction. This is just a photograph (of the instrument).

This is the schematic of the NG1 reflectometer at NIST (NCNR reactor). You can see, they have polarizers. Supermirror polarizers can be used for analyzing the reflected beam for up and down neutrons. Neutrons are coming from the Guide and there is a focusing geometry where you focus (the beam) in vertical direction, but obtain the reflectivity pattern in horizonal direction. So, this focusing in the vertical direction helps to get higher intensity without sacrificing the resolution in the horizontal plane. There is also a neutron spin-flipper, which is a DC flipper. It has a structure like this, on which you wind a solenoid so that if this is the neutron spin polarization direction and this is the direction of the solenoid's magnetic field,

then while passing through the solenoid the neutron tends to precess around this field. You choose an appropriate thickness of the solenoid based on the wavelength of the neutron. Depending on the wavelength, the velocity changes, which changes the time spent in the magnetic field and that tells you by what angle the neutron will be precessing (during its passage through the solenoid). Right choice of field and the thickness of the DC flipper can flip, up beam neutrons to down beam. This is about the DC flipper which we use. There are also other kinds of spin flippers, like radiofrequency spin flippers.

(Discussing about NG1 reflectometer schematic) This is a neutron spin flipper. There is the sample (position). There is a polarization analyzer also, which is an Fe/Si supermirror and then you have got a detector. So, the general geometry is very similar to what we have in all other sources.

The other point I would like to mention is that, in this case, it (the reflectometer) is coupled to a liquid hydrogen cold (neutron) source. Reason being, as I discussed earlier also, the neutron intensity versus wavelength, a Maxwellian in the room temperature beam, then in a cold source the spectrum shift to lower energy or longer wavelengths (multiplying intensity at longer wavelengths). Reflectometry as well as small angle instruments use longer wavelength neutrons because we are working in the region of low Q. That is why here the NG1 Reflectometer is coupled with the liquid hydrogen cold source from where the cold neutron beam comes. In this instrument they have got a Pyrolytic Graphite triple crystal monochromator in vertically focusing geometry. Then they have a polarizing supermirror, followed by flipper, sample and again another spin-flipper and Fe/Si supermirror for polarization analysis. So, without flipping and with flipping, you can get the intensity of two (neutron) beams. So, now, in case of (using an) analyzer crystal, you are measuring reflected intensity of up neutron scattered as up neutron or up neutrons scattered as down neutrons, down neutron scattered as down neutron and down neutron scattered as up neutron. So, you measure four set of intensities instead of two. Now, it is clear that when we do not do the spin analysis (of reflected neutron), we have measurement of R^{+} that means, reflectivity of the up neutrons and R^{-} reflectivity of the down neutrons. When you measure R^{+} and R^{-} without any analysis of the reflected polarization, then what we obtain is magnetic moment density. When we do (spin) analysis of the reflected beam, we find out four intensities. These are non-spin flip (R^{++},up-up), non-spin flip (R^{--}, down-down), spin-flip (R^{+-}, up-down), spin-flip (R^{-+}, down-up). We get magnetic structures (from these measurements). It will be clearer when later I discuss this issue with an example.

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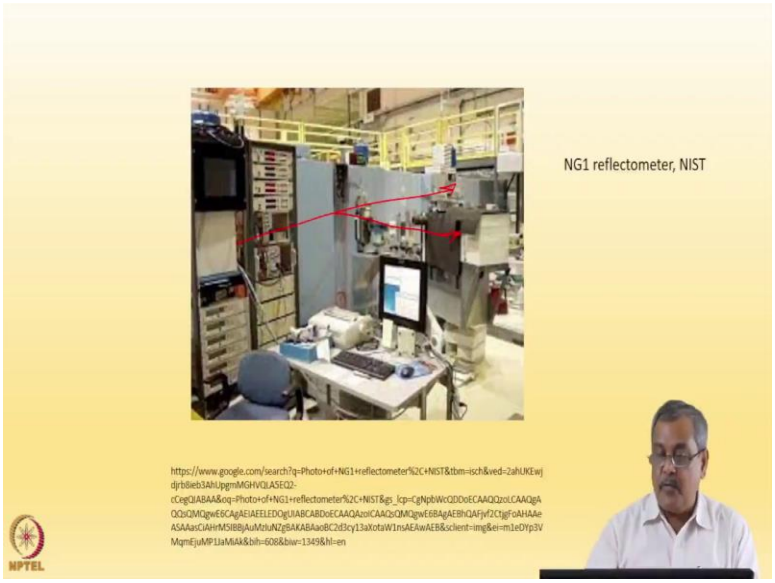

This is the schematic and this is a reflectometer similar to Dhruva. As I told you earlier that, this is a guide from which the beam is reflected into this reflectometer using pyrolytic graphite monochromators.

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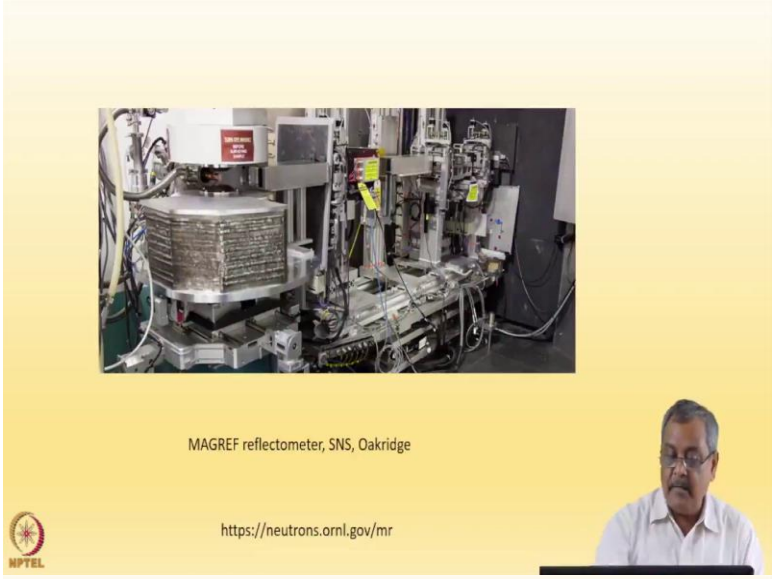

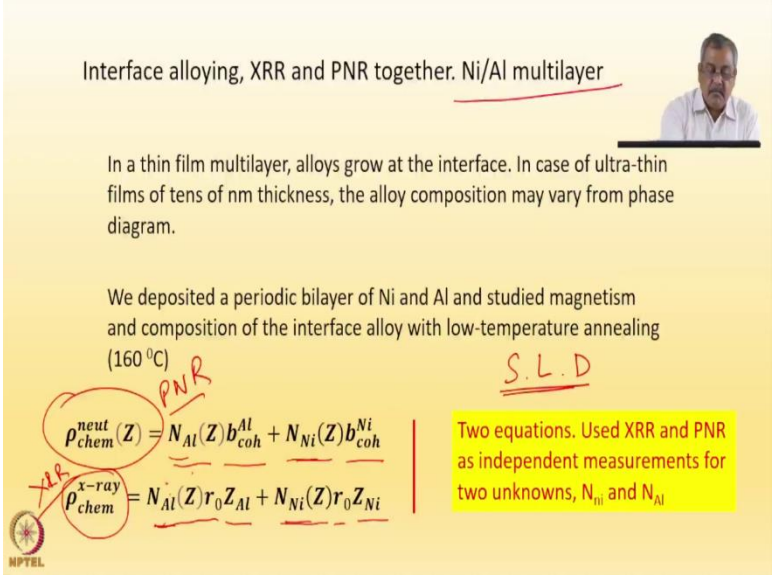

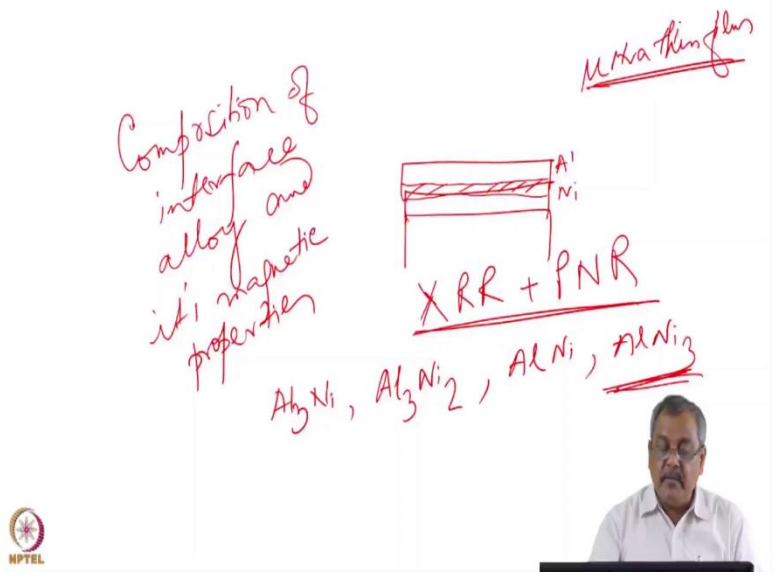

This is the MAGREF reflectometer at SNS, Oakridge. It has a very similar structure to what we showed earlier.

With this, I will now switch over to examples. To start with, I will give an example in which XRR and PNR together were used to understand the interface alloying in a Ni/Al multilayer. In this sample, we deposited periodic bilayers of Ni and Al. When we annealed this multilayer, you will find alloy layers forming at the interfaces and our intention was to determine the composition of interface alloy and its magnetic properties. For this we use XRR and PNR together. I will explain shortly why they were used together.

Ni and Al form a number of alloys and in which the Ni-Al ratio keeps changing, for example, at lower temperature Al_3Ni alloy is formed, then you have Al_3Ni_2 at higher temperature next we have AlNi. you also have $AlNi_3$ forming at higher temperature, which is a very important alloy because it combines the hardness of Ni with the ductility of Al and is used heavily in aeronautical industry. Our interest was, how the alloy forms in ultra-thin films at the interfaces as a function of annealing temperature/time and can one determine its composition using XRR and PNR together.

If we do XRR, we get electron scattering length density which for the Ni-Al alloy is given by,

$$\rho_{chem}^{x-ray} = N_{Al}(Z) r_0 Z_{Al} + N_{Ni}(Z) r_0 Z_{Ni}$$

where r_0 is the classical electron radius 2.818 fm, and *N* and *Z* are the number density and *Z*-value of the corresponding element. So, this equation gives me the chemical density for the given sample and this (density ρ_{chem}^{x-ray})we obtain from (XRR) reflectometry experiment. Similarly, for neutrons it is

$$\rho_{chem}^{neut} = N_{Al}(Z) b_{coh}^{Al} + N_{Ni}(Z) b_{coh}^{Ni}$$

This we get from a PNR experiment as nuclear scattering density (ρ_{chem}^{neut}) and above one we get from an XRR experiment as electron scattering density. In these equations, everything is known except $N_{Al}(Z)$ and $N_{Ni}(Z)$. I can consider XRR and PNR as two independent experiments giving two independent equations, which we should be able to solve algebraically once we obtain the density profile in samples from these two equations. That was our main aim in the XRR and PNR experiments.

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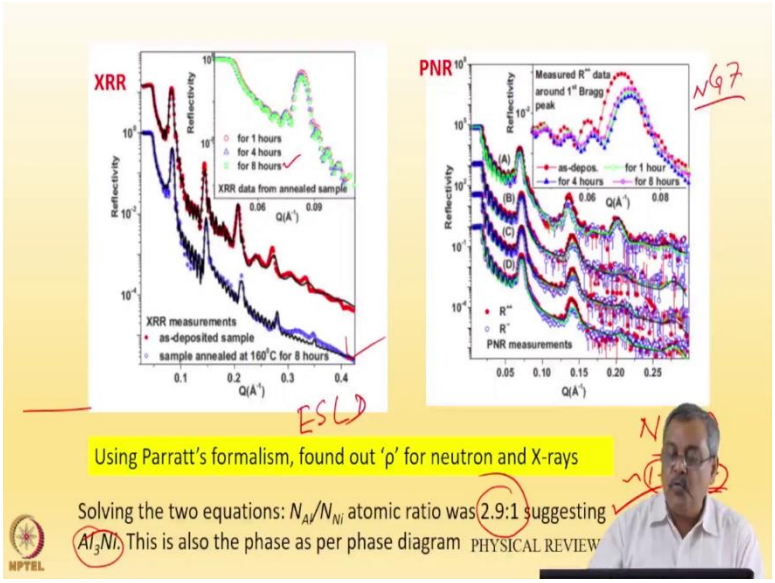

Let me just give you the results here.

This is the XRR result for the Ni/Al sample and this is a PNR result which was obtained at NG7 instrument at NIST. XRR was on a table top machine. We formed the alloy by annealing for 1 hour to 8 hours at a low temperature of around 160 °C.

This is how the polarized neutron reflectometry data vary with respect to (time of) annealing. And this is how the XRR reflectivity pattern changes after 8 hours of annealing. Now, we use Parratt's formalism for both of these data. But independently fitted the nuclear scattering length density from the PNR data and fitted the electron scattering length density from the XRR data. So, we obtained these two values from the experiment and then I attempted to find out what is the Al to Ni ratio?

We found that Ni to Al atomic ratio, that is, $N_{Al}: N_{Ni} = 2.9: 1$. It indicates that at the interface at this low temperature annealing, we obtain the Al_3Ni alloy and it is also commensurate with the fact that at low temperature annealing, this is the first alloy that forms for Ni and Al.

This is an interesting result in which we obtained the physical density of an alloy layer which is typically around 1 to 2 nm or 10 to 20 Å thick in a Ni/Al multilayer and the nature of the alloy. You can see the excellent resolution obtained using this non-destructive reflectometry technique.

This is one example I showed where we can use XRR and PNR together for our samples. Usually, whenever we do PNR, XRR comes as a preamble. That means, usually when we make a sample, we carry out the XRR experiments to obtain its density, thickness, roughness, etc.

with XRR, because XRR data has much higher intensity and we can get better fitted values. Then we go ahead and carry out the PNR. Of course, this will not be true for all the samples. But for most samples, this route is chosen.

With this, I will stop today. I will provide more examples and more interesting results just to show you the kind of experiment that one can do using PNR or polarized neutron reflectometry to understand structure of thin films.