Neutron Scattering for Condensed Matter Studies
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Week 9: Lecture 23A

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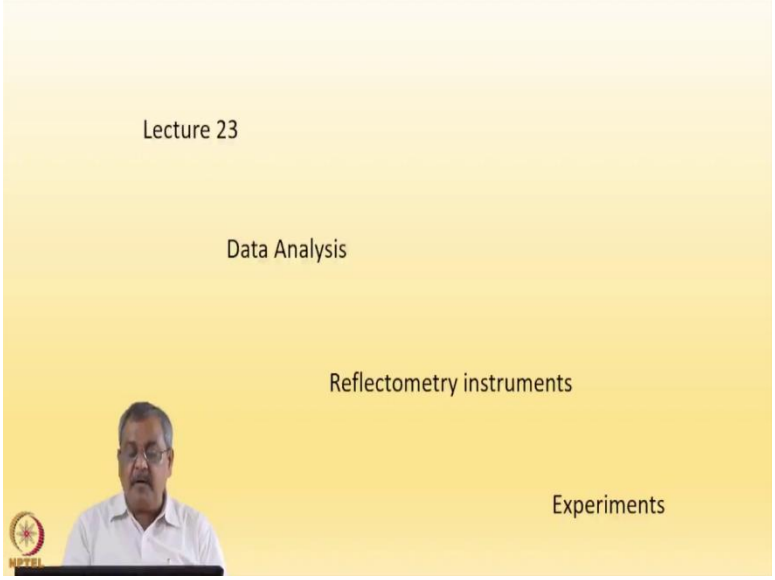

We are on Lecture 23 in the series. For past few lectures I have been discussing Neutron Reflectometry, specifically Polarized Neutron Reflectometry for thin film characterization. In this lecture, I will discuss the data analysis techniques for neutron reflectometry. I will familiarize you with reflectometry instruments which is there at Dhurva and few more from the best sources, to give a general idea about the kinds of instruments that are available and then I will deal with examples or studies that have been done using these instruments.

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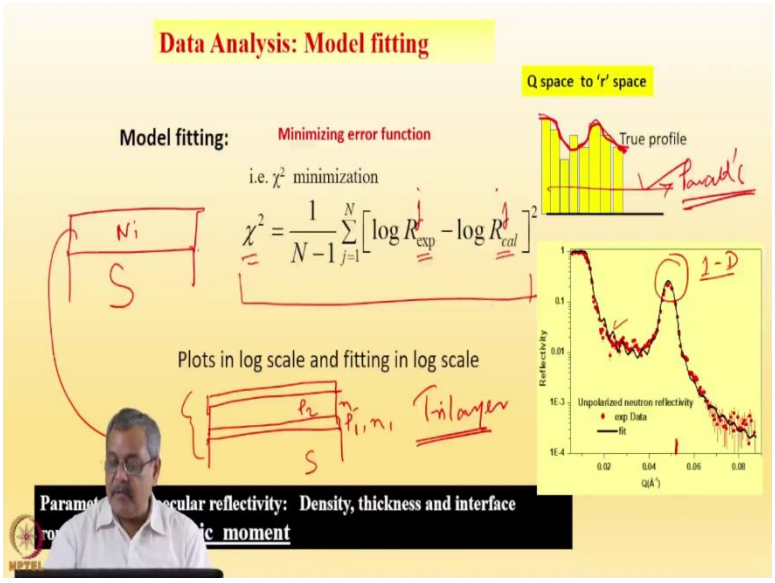

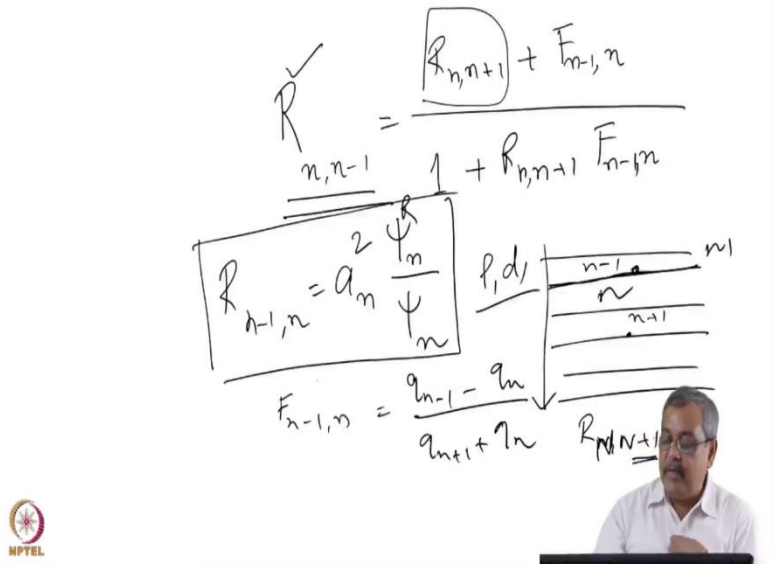

First, we will discuss the data analysis part. I have introduced you to Parratt formalism which can give you the model reflectivity for a model structure in real space. If you remember it, I wrote the following expression.

$$R_{n-1,n} = a_{n-1}^4 \left| \frac{R_{n,n+1} + F_{n-1,n}}{1 + R_{n,n+1} F_{n-1,n}} \right|$$

where layers of known densities, thicknesses all are embedded in this model. And for this model, if I want to calculate the reflectivity index at the boundary of, say, n-1th and nth layer then I need the reflectivity at the next boundary which is nth and n+1th layer and then this becomes the recursion formula. So, to know $R_{n-1,n}$ I need $R_{n,n+1}$ so on and so forth. Here, $F_{n,n+1} = \frac{q_{n-1} - q_n}{q_{n-1} + q_n}$ which is a reflected amplitude between n-1 and nth layer.

So, here to find out $R_{n-1,n}$ I need $R_{n,n+1}$ and this series was truncated. I mentioned to you earlier that $R_{N,N+1} = 0$ where N was the final layer (N+1) layer is the vacuum below the substrate. Whatever goes inside the substrate, nothing comes back and starting from this given structure I can calculate the reflectivity at the air-film interface. This is the handle for our model fitting. Now, once I have a defined structure, I can find out the reflectivity using Parratt formalism.

Let us consider an example on a single layer, say, a single nickel layer. I am using this example because I will show you the actual data collected for the single nickel layer on the substrate and the corresponding fit. This is basically a nickel film which is also used as a mirror in our experiment. Now, in this case, I can represent this nickel layer consisting of a (virtual) thin layer at the substrate-film interface, which will have a different density then that of bulk of the Ni layer and then at the air-film interface also there is a (virtual) thin layer. So, the single layer, for the purpose of fitting the data I might break it up into a trilayer. And when I make it into a trilayer, then for this trilayer I start with some assumed values of densities, thicknesses and interface roughnesses. That means, I represent these layers as a histogram of densities and the refractive index.

Once I have this histogram, I have shown here a schematic that if this is the true density pattern it is possible to represent it as a distinct histogram like this, and for this histogram I can use Parratt's formalism to calculate the reflectivity index. Now, in this case, I have created a trilayer. Now, I keep playing the game of chi-square minimization, which is given by

$$\chi^2 = \frac{1}{N-1}\sum_{j=1}^{N}\left[\log R_{exp}^{j} - \log R_{cal}^{j}\right]^2$$

where R_{exp}^{j} are the experimental values and R_{cal}^{j} are the calculated values from the used model for each data point. This is the chi-square value and I have to minimize it to fit this model. Please remember, I started with an assumed model, which gives me a histogram for the given sample, here it is a single Ni layer and I broke it up into 3 virtual layers. For all 3-layers I have an assumed density, thickness and then I fit the model through χ^2 minimization.

In the picture here, unpolarized neutron reflectivity data is shown for a Ni/Ti multilayer film, you can see the experimental data and the fits. The experimental data has been fitted using this chi square minimization technique.

Let me repeat, Parratt formalism is the starting point of model building. Using Parratt formalism, I can find out the reflectivity of a model structure and I keep changing it till I get a good fit with the experimental data. And that will be my result or the fitted values in the real space given this reflectivity pattern. In real space, I will get the result in terms of density, thickness, interface roughness etc. These chi-square minimization processes are also varied. Nowadays, for reflectometry data, each major source possibly will have its own optimization software available to you. For example, NIST (National Institute for Standards and Technology, USA) does provide you such a program and if you go to their online site, you will get a program to fit your reflectometry data.

In this fit shown here for an unpolarized reflectometry data of a multilayer film with a periodic bilayer. We observe the Kiessing oscillations due to the total thickness of the film. Along with this we have also got a Bragg peak arising due to the periodic bilayer acting as a one-dimensional crystal. For this artificial crystal I had this Bragg peak just like an actual crystallographic Bragg peak, only the number of layers is small. So, there are a finite number of planes, it is a one-dimensional crystal. Because this (periodicity) is in a mesoscopic length scale, the Bragg peak appears at a much lower q-value, here at 0.05 $Å^{-1}$.

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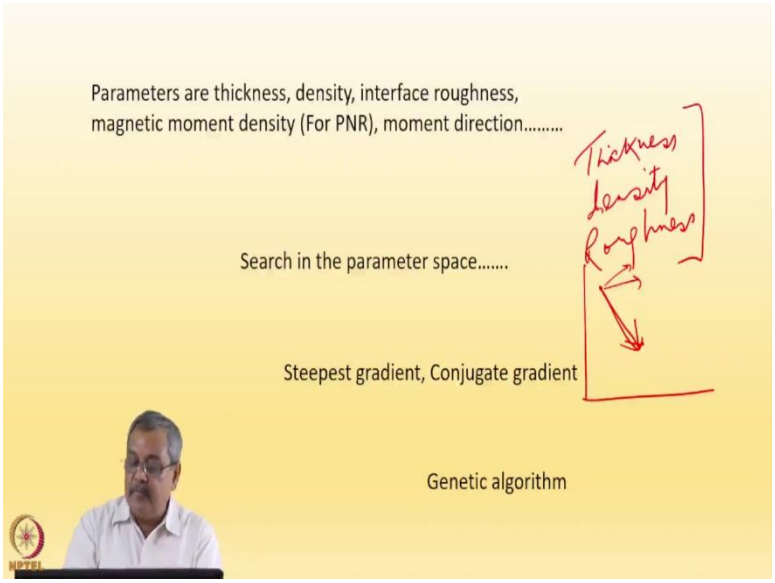

Now, let me repeat that parameters are thickness, density, interface roughness, and magnetic moment density and direction in case of polarized neutron reflectometry. We have to search in the parameter space, we know that it is important that we get the global minimum in the parameter space which is actually the true solution for the given problem, true solution means the parameters are the true parameters of the sample which you do not know actually.

So, you are looking for a global minimum in parameter space starting from some assumed structure. There are several well-known techniques in this chi- square called minimization process. One is steepest gradient technique and another is conjugate gradient. In case of steepest gradient at one point in parameter space (it is a multi-dimensional parameter space) with parameters like thickness, density, roughness etc. If we assume that they are independent of each other then we can have a 3-dimensional parameter space and one point means a particular thickness, a particular density and a particular roughness. You start from there, calculate the gradient in χ^2 at this point by slightly changing the parameters and then move in the direction where the fall is the fastest. I have used genetic algorithm which is slightly different from these (above techniques) and I would like to discuss briefly with you: what is genetic algorithm (GA) technique? I have found that one can reach physically reasonable solutions in a much faster time using GA.

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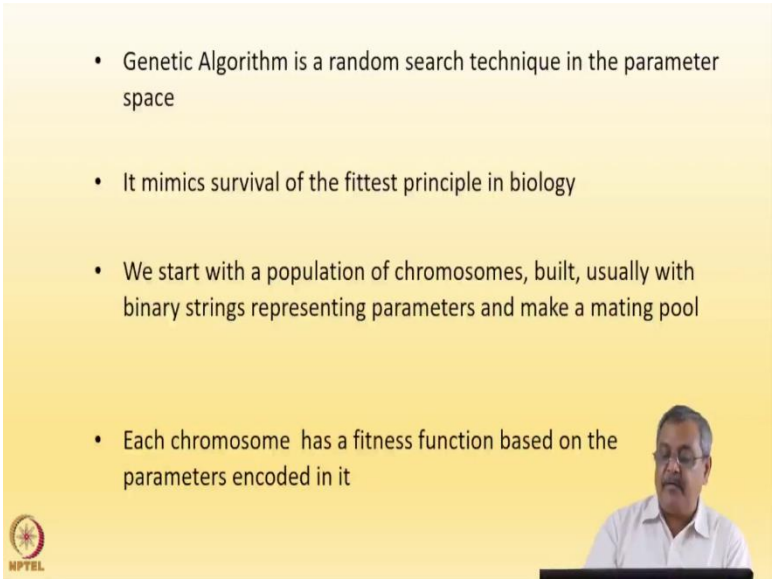

It is a random search technique in the parameter space and it mimics Darwin's survival of the fittest principle. We start with a population of chromosomes! The chromosomes as I will show you, are built usually with binary strings made with parameters (expressed in binary) and we made a mating pool of these chromosomes, where we mate the chromosomes, get new chromosomes. Each chromosome has a fitness function (signifying quality of fit) and from this we get a new pool of chromosomes, which will have their own fitness functions. Let me explain to you.

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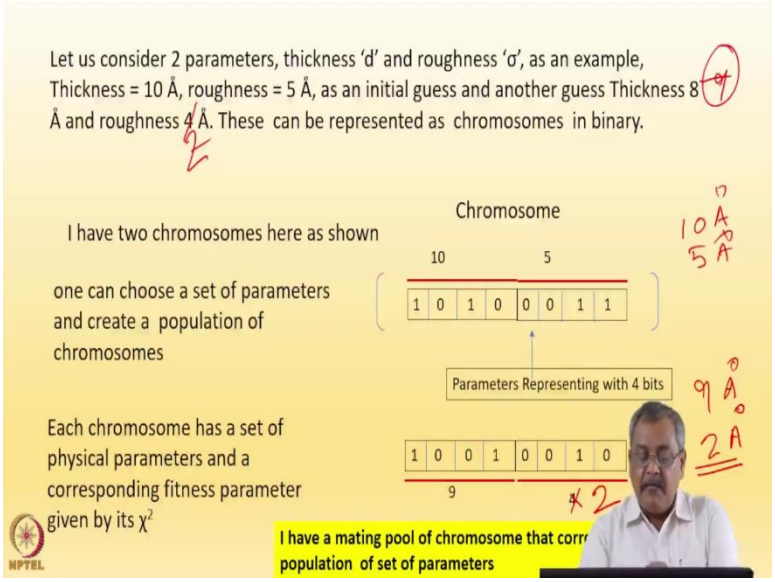

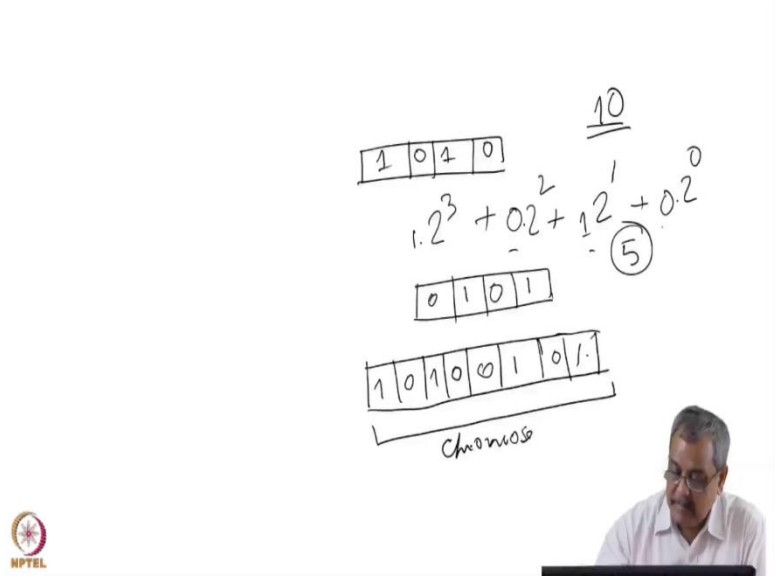

Let me take just 2 parameters of a film, let us say thickness *d* and roughness σ as an example, let us consider the thickness is 10 Å and roughness is 5 Å as an initial case and another case let us take thickness is 9 Å and roughness is 2 Å. Now, if I use a 4-bit byte, the chromosomes look like as shown.

For 10 Å thickness, it will be $1 \times 2^3 + 0 \times 2^2 + 1 \times 2^1 + 0 \times 2^0$, so in binary 4-bit scheme it will be represented as 1010. Similarly, the roughness of 5 Å will be represented as 0101 in 4-bit scheme. Here, I have used 4-bit representation, we can increase the number of bits to get more resolution. Now, with these 2, I can build a chromosome, which is 8-bit long and for the used thickness and roughness it will be 10100101. Now, this thickness and roughness together forms a chromosome for my genetic algorithm simulation.

I can also take another set of parameters for example, say, thickness 9 Å and roughness 2 Å. For this combination, chromosome will be 10010010. So, one chromosome I created for 10 Å thickness and 5 Å interface roughness and another one I have created for 9 Å thickness and 2 Å interface roughness. Now, I will be mating these two chromosomes just like real life chromosomes. And how do you do that.

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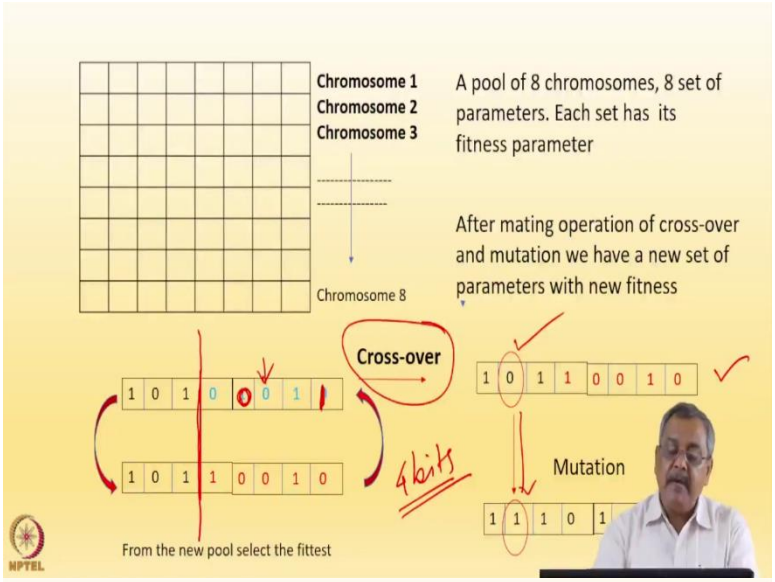

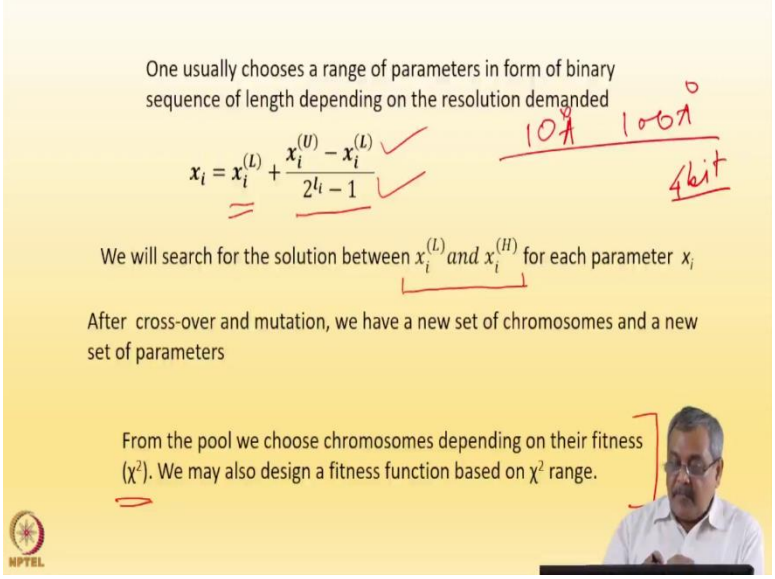

First, I have got a pool of such chromosomes. I showed you two such chromosomes but you can make a pool of these chromosomes. Let us have two chromosomes 10100011 and 10110010 in the pool and here I randomly choose a particular site. Then I cut this part, bring it here and cut this part and take it there. This is known as cross-over operation. So, now with this cross-over operation from this point, the sequence 101 remains and the next part it will be 10010. This is a crossover. Similarly, this part goes on added there. So, after cross-over, there will be two new chromosomes. Now, I choose any one of them. And at a random site, I switch the bit in this binary representation. Switch means if it was 0, I have made it 1. So now, I get after crossover and mutation, a new value of parameters given by 11101010.

So, I have used 4 bits to represent my parameters. And I have explained you the genetic algorithm operations that I will carry out and I will get this chromosome. This gives me a new chromosome after cross-over and mutation.

So, this pool now will have new 8 chromosomes from the previous generation. We usually choose a range of parameters in form of binary sequence. It depends what is the upper limit of parameters, what is the lower limit of parameters.

Suppose, I have thickness in the range of let us say 10Å to 100Å and I want to write them as 4 bit chromosomes, then lower value, $x_i^{(L)}$ will be 10 and $x_i^{(U)} - x_i^{(L)}$ will be 90, and then gives you $2^{l_i} - 1$ means 2^4-1intervals. There are 15 gaps. So, basically, starting from the lowest to the highest l have 16 values of thickness.

Similarly, I can generate the chromosome for the roughness, depending on the length I choose and the lower and upper values. Now, when I am looking for a solution in parameter space, say for thickness in case of which is x_i, let us say, I will be looking for solution in range $x_i^{(L)}$ and $x_i^{(U)}$ for each parameter.

Here, in the previous case I described you the parameters thickness and roughness. There can be more parameters. If it is a multilayer sample, there are many, many, thicknesses, so word length will be long, and depending on that chromosome lengths will be large. And for each parameter depending on how much resolution you want the number of bits in the byte will increase. And, after crossover and mutation, we will have a new set of chromosomes giving a new set of parameters. So, again from this pool, we choose chromosomes, depending on their fitness.

Now, Darwinian fitness is biological fitness. In our case, the fitness is the χ^2. We evaluate $\chi^2$for the new parameters and then from them we choose the more fit ones, that means, with lower value of chi-squares and this choice we can design in our way. Here we chose an exponential way of choosing this chi-square values from the range of parameters. But there can also be several other techniques, I do not want to give you any specific technique.

The fact is that from the new pool, we choose chi-square values depending on the fitness. This process goes on, once we have got a new pool, we choose the chromosomes which have got better fitness. Then between them, we carry on again mating; mating means cross-over and mutation. Again, we get a new set. So, we can start with a large number of parameters or chromosomes and keep on coming down to smaller and smaller values for χ^2. And hopefully, we will hit the global minima or the actual solution in the parameter space. The advantage of genetic algorithm is the following.

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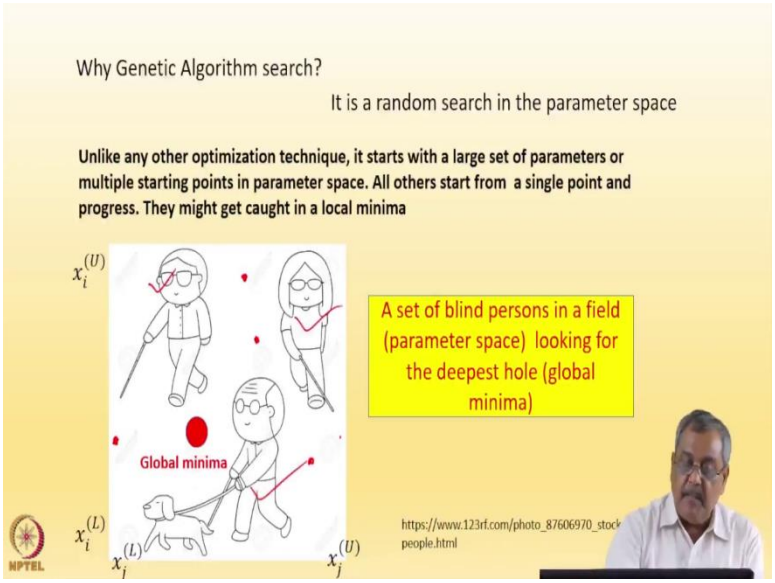

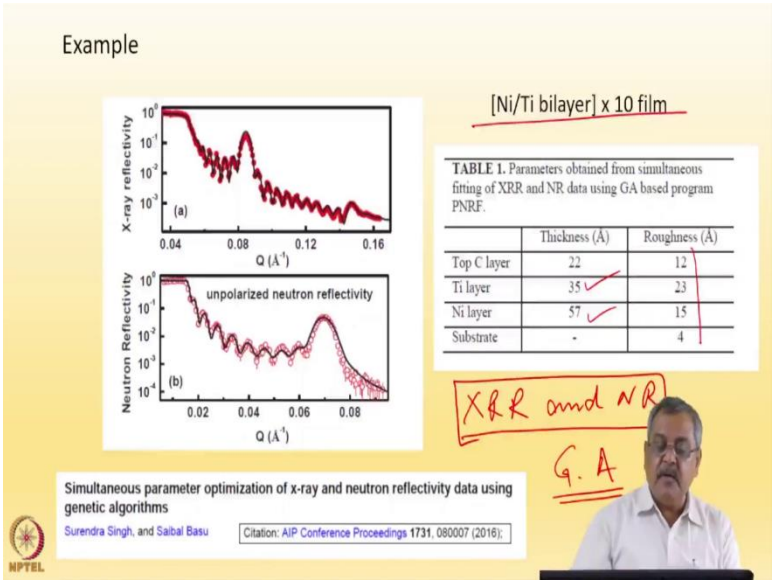

The biggest advantage is that unlike any other optimization technique, the genetic algorithm process starts with a large set of parameters or multiple starting points in parameter space. As I told you earlier, steepest gradient or conjugate gradient techniques, start with only one point and there is a good chance that it will get stuck in a local-minima.

But here in this case in this particular GA search technique, because we start with many points. I have this sketch as an example. It is like a set of blind persons in a field, which is a parameter space as shown here in a square parameter space with lower limit and upper limits on the parameters. And they are looking for the global minima in a blind search. They are blind people. But every time they reach a new parameter set, we choose the lower chi-square values. And that is how they are approaching this global minima. And because I am starting from

multiple points, multiple number of say blind people, I have a better chance of missing this local minima and reaching the global minima.

So, this is about genetic algorithm. I just use one example where we have fitted data using genetic algorithm. It was again that nickel titanium bilayer which I showed you sometime back and you can see that I had minimized the chi-square and the fit quality using genetic algorithm is evident from the plot for neutron reflectivity and polarized neutron reflectivity as well as X-ray reflectivity.

Here actually, interestingly, as I told you, that XRR and unpolarized neutron reflectometry, both of them give structure of the film. So, in this case, these two experimental data are fitted together with relative weightage decided by us depending on their experimental errors and the fit. You can that see we have got average roughness and thickness of the nickel layer and titanium layer using genetic algorithms.

Usually, the data that I will be showing for our experiments, we use the genetic algorithm technique for fitting. But there is no restriction on techniques and other techniques are also equally good. Biggest advantage of genetic algorithm is that you start from multiple points in the parameter space and have a better chance of reaching the global minima unlike other techniques.