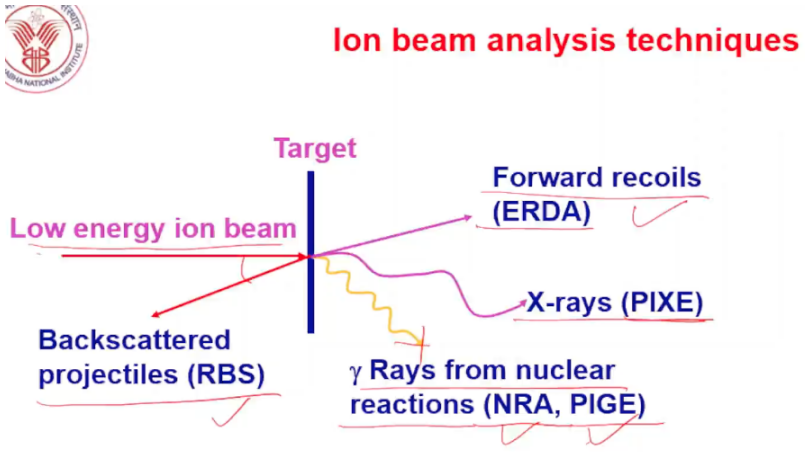
Ion beam analysis: Rutherford backscattering and Elastic recoil detection analysis

B.S.Tomar

Homi Bhabha National Institute

Lecture-18, Module-1

Hello everyone. The previous lecture, I discussed the technique, the nuclear technical technique based on neutrons, that is neutron activation analysis. And also discussed some of the applications of this technique in different areas. Today I will discuss another nuclear technical technique called ion beam analysis technique. As the name suggests, in ion beam analysis technique, we use the charged particle beam. And the charged particle beam may have different types of reactions with the nuclei in the target. And depending upon the type of reaction that is happening, we have different nuclear technical techniques.



So by this schematic, I will first just introduce what are the techniques that I am going to discuss in this sketch. So when the low energy ion beam, it could be proton, alpha or even heavier ions, they bombard the target, then there could be backscattering of the projectile. So the backscattering of the projectile is like Rutherford backscattering.

And there is a, as we will discuss in more details, the specific relationship between the backscattered energy and the mass of the isotopes in the target material. And therefore, we can identify the elements or the isotopes present in the target. And later on, we can even find out their concentrations, their depth profile etc. So this is RBS, Rutherford backscattering spectrometry is a very well-known ion beam analysis technique in material characterization. Now, second type of reaction can be that the projectile hits the nucleus and gives recoil that means that the atoms of the target are taken out of the target material.

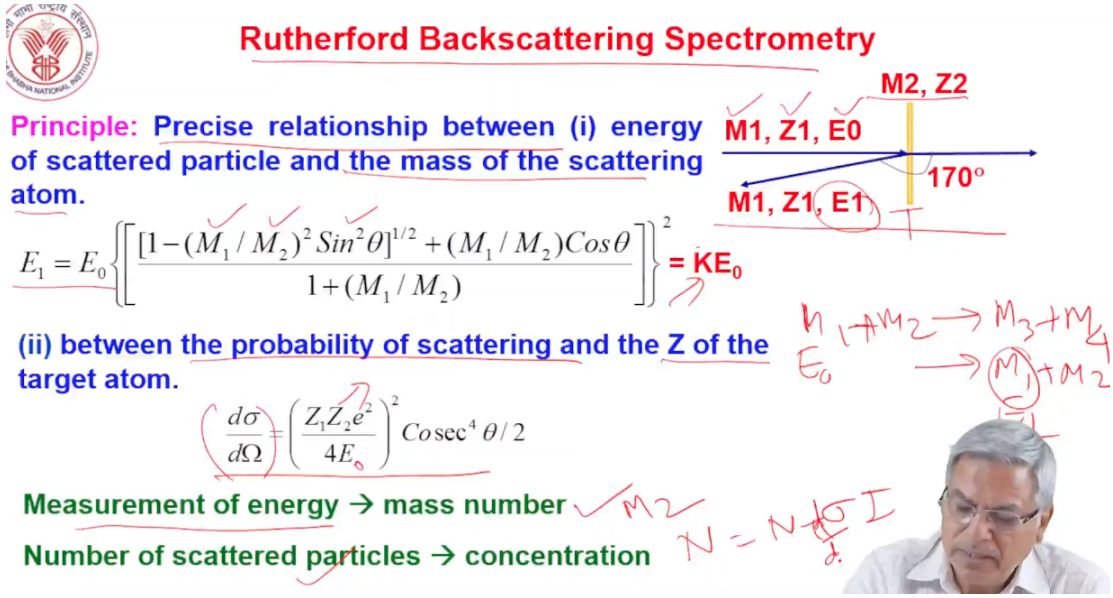
So they come out of the target material and they can be detected by a suitable detector system. So the recoils which are emitted in the forward angle, the forward recoils are detected to tell about the constituent of the target. So this is called the elastic recoil detection analysis. It is purely elastic scattering between the target nuclei and the initial projectile beam. And this also can be used to characterize the different types of materials.

There could be nuclear reactions between the low energy ion beam and the nuclei in the target material. And these nuclear reactions can give rise to the gamma rays. So if you detect the gamma rays from this nuclear reactions, then the analysis based on the nuclear reactions will be called nuclear reaction analysis. And one of the variants of this is particle induced gamma emission, PIGE. Mostly protons or deuterons are used.

So they are actually similar techniques, but the difference will become clear when we discuss each technique in this particular lecture. So in nutshell, these are the four types of nuclear reactions or interactions which we will be utilizing in the different nuclear analytical techniques, the RBS, the ERDA, the NRA and PIGE.

Also, you will see when the projectile beams interact with the target atoms, they can cause ionization in the atomic shells, like for example, K-shell ionization can take place. And this ionization of the K-shell electrons will lead to emission of X-rays. So particle induced X-ray emission, though it involves the atomic orbitals, this also is considered to be an ion beam analysis technique. And mostly people use protons. I will not discuss this particular, basically this gives you the composition of the material, different elements will have characteristic X-rays and those X-rays can be detected by suitable detector and the peak area of the X-ray peaks will tell you the concentration of the particular element in the material. So you may have to have standards and it is similar to X-ray fluorescence, where you use X-rays to bombard the target material or even you can use gamma ray or even you can use beta particles. So the emitted X-rays are characteristic of the elements. So PIXE and XRF, these are other techniques which are used in routine compositional analysis of different types of material.

While the ion beam analysis techniques like RBS, ERDA, NRA and PIGE, they offer much more advantage over simple techniques like PIXE and XRF and it will become clear. So this is not just a routine analysis, but they involve the detailed analysis of the target material, the processes that one is studying and so on.



So let me first discuss the fundamentals of Rutherford backscattering spectrometry. As the name itself implies, it is based on the Rutherford scattering of the projectile by the nuclei present in the target material. And the two important principles behind this are, one is that when a projectile is being backscattered by the target nuclei, there is a precise relationship between the energy of the scattered particle that is the projectile scattered and the mass of the scattering atom.

So basically it is an elastic scattering and as we discussed in the lecture on nuclear reaction, in elastic scattering the kinetic energy is conserved and you can calculate the energy of the backscattered ion exactly if you know the mass of the projectile, target and the initial energy of the projectile as well as the angle theta at which the scattering is going to take place. And so I have just given a schematic of this RBS. We have a projectile of mass M_1, atomic number Z_1 and energy E_0, bombarding the target material, having let us say a particular nucleus with mass and charge M_2 and Z_2 and after the scattering, the projectile is coming out with energy E_1. So θ is equal to let us say 170 or 160, whatever it is. We will discuss why back angles very shortly.

So you can from the conservation of mass and energy and the conservation of linear momentum set up the equation like we did it in that particular lecture to find out the mass of the target nucleus if you recollect, we had solved that equation for energy E_3 of M_3, whereas here we will be determining the mass of M_2 itself. You will recall $M_1 + M_2 \rightarrow M_3 + M_4$ for any nuclear reaction, but here it is elastic scattering so the products are $M_1 + M_2$ only. There the ejectile energy was E_3, here energy is E_1. And so this E_1 is actually nothing but E_3 you can say. And so that equation was solved and the relationship between E_3 and E_1 or here E_1 and E_0 is given by this formula, it contains M_1, M_2 and θ.

$$E_1 = E_0 \frac{\{[[(1-\frac{M_1}{M_2})^2 sin^2\theta]^{\frac{1}{2}} + \left(\frac{M_1}{M_2}\right)cos\theta]]^2}{1+\frac{M_1}{M_2}}$$

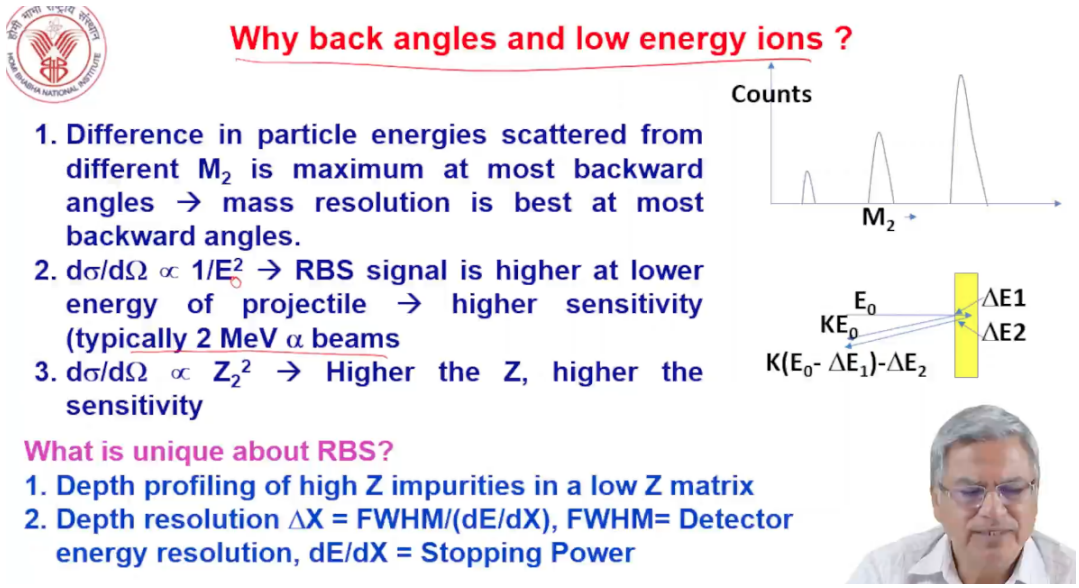
So this term can be clubbed as K. So the backscattered ion energy is K × E_0 and K is called a kinematic factor. It depends simply upon the masses and angles of the projectile and target nuclei. So for a particular value of M_2, there is a particular value of K. So that way you can find out what is the mass of the nucleus which was backscattering the projectile.

The second relationship is between the probability of scattering of the projectile and the Z of the target. And the cross section is given here, probability means essentially the cross section. So the differential cross section, $\frac{d\sigma}{d\Omega}$ per unit solid angle is given by

$$\frac{d\sigma}{d\Omega} = (\frac{Z_1 Z_2 e^2}{4E0})^2 Cosec^4\theta/2$$

where E_0 is the projectile energy. So you can see here, higher the Z of the target nuclei, higher the cross section. So this will be made clearer that for higher Z nuclei, the sensitivity is higher. Cross sections are higher, there will be more events. So you can then from the determination of the number of particles that are scattered or from the counts in the spectrum of the scattered particle, we can find out the concentration of the target nuclei.

So the measurement of the energy of the backscattered particle gives you the mass number M_2 in the target. You can identify. So from the mass number you can of course identify the elements. And from the number of scattered particles, what you will know, scattered particle it will be $N_t\sigma I$ and then you can say, so this σ will be $\frac{d\sigma}{d\Omega}$. So this is the number of particles into t time. And so this tells you the concentration of that particular target.



Now let us ask the question, why back angles? And in fact the RBS is done with the low energy ions, energy of the projectiles is low because we do not want to introduce any nuclear reaction. So it is simply Coulomb scattering. So you need to be below the Coulomb barrier. There are no complications because of the nuclear reaction with the target. So it is a pure Coulomb scattering. So that energy of the projectile will be kept quite low. Not only that, the cross sections for this RBS scattering are, if you see here, cross section for the scattering varies as $1/E^2$. So lower the energy, higher is the cross section for RBS and therefore we go for low energy ions.

So two aspects, why the low energy ions? Because the cross section is inversely proportional to E^2, so that is E_0. So lower the energy of the projectile higher the sensitivity for the RBS for detection of the concentration of the elements. But typically, you know people use 2 MeV alpha particle beams, so you will be below the Coulomb barrier for most of the target nuclei. In addition to this, there would not be any nuclear reaction that would complicate, it would unnecessarily introduce some radioactivity in the sample and so it may become difficult to handle.

So this is the reason for low energy. Second is that why back angles? So back angle is the difference in the particle energy that E_1 backscattered from different masses in the target nuclei is maximum at most backward angles. So if you see here, if you put theta, so essentially you can say $dM_2 / d\theta$, the change in the mass with the angle if you do an exercise, you would find the differences between different masses. So you have got cobalt, iron, nickel, different elements, so their masses are also different. The energy of the different masses will be widely spaced at most backward angles. $dE_1 / d\theta$ will be maximum at θ=180. But you cannot put detector at 180 degrees because that is the beam path, so you keep slightly away from 180, maybe 170, 160 or so. That is the reason for

back angle. So you want to resolve for example here, what I have shown here, M_2, different M_2s in the spectrum. So the projectile spectrum if you record in the back angle, then you will see here, this is low mass, higher mass, higher mass.

So this gap between these different masses will be maximum at most backward angle. So you can say the mass resolution is maximum at most backward angle. Otherwise, you know at lower angle, they will all merge together, the gap will reduce, so you cannot resolve that, they may start overlapping. Secondly, as I mentioned, the cross section are proportional to Z^2, $\frac{d\sigma}{d\Omega}$ proportional to Z^2 of the target elements. And so, higher the atomic number of the nucleus that you are going to investigate, higher the sensitivity.

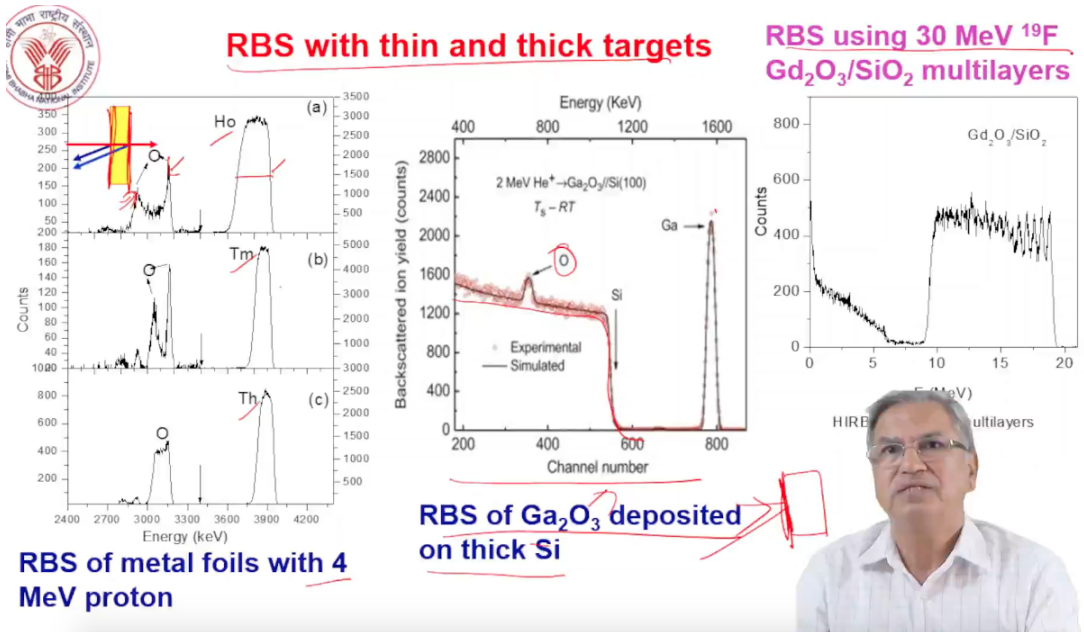
Because more events you will get, you can see here, this is the lower Z, higher Z and still higher Z. So higher the Z, higher the counts you will get, so they are more sensitive. And because of this reason, you know, the RBS is ideal for detecting the higher Z impurity in a low Z matrix. So mostly the applications of RBS involve determination of high Z impurities in a low Z matrix. For example, you have got a plastic that you want to determine substituents, metallic impurities or iodine, bromine, excellent technique for that kind of study.

So there will be several applications. So that is one, determination of depth profile of high Z impurities in a low Z matrix because of this higher Z being more sensitive and they appear at high energy so you can distinguish them from the low Z matrix. And second is the depth profile. You can do depth profiling by RBS technique because the backscattered ion energy will be different depending upon the depth from which the scattering is taking place. I will elaborate this more using this cartoon.

So you have a projectile of energy E_0, bombarding the target and I have shown a thick target. So there will be backscattering at every depth of the target. So from the surface, from the surface backscattered projectile, energy will be $K*E_0$, K is the kinematic factor. So that energy is well known. For a particular M_2, you can find out what will be the energy because K you can calculate exactly, K depends upon M_1, M_2 and θ.

Now, as the projectile is travelling in the target material, it is losing energy and every depth it will get backscattered. So let us say at a particular depth, we will say this particular depth energy lost is ΔE_1. So the energy at this depth will become E_0-ΔE_1. So M_2 at this depth will see a projectile coming at E_0 -ΔE_1 and backscattered from that site. So you are putting a detector here, D. So the backscattered energy at that point will be $K*(E_0$ -$\Delta E_1)$. And again, the backscattered ion will lose energy ΔE_2, so -ΔE_2. You can exactly calculate what will be the energy of the backscattered ion if it is backscattered from a particular depth in the target material. Because the kinematics are very well known, the energy lost is very well known in the target material. You can find out because the stopping powers are known and you can find out the depth.

Suppose you know the dE/dx, you can find out the depth at which the backscattering took place. And so by recording the spectra of the backscattered projectile particles, you can do depth profiling for impurity concentrations and what depth they are, you can see. So depth resolution in RBS, Δx depends upon detector resolution FWHM of the peak upon stopping power, dE/dx. So we can determine the resolution of detector from the normal spectra, stopping powers are known, we can find out the depth resolution. So I will just give you some of the examples of RBS.

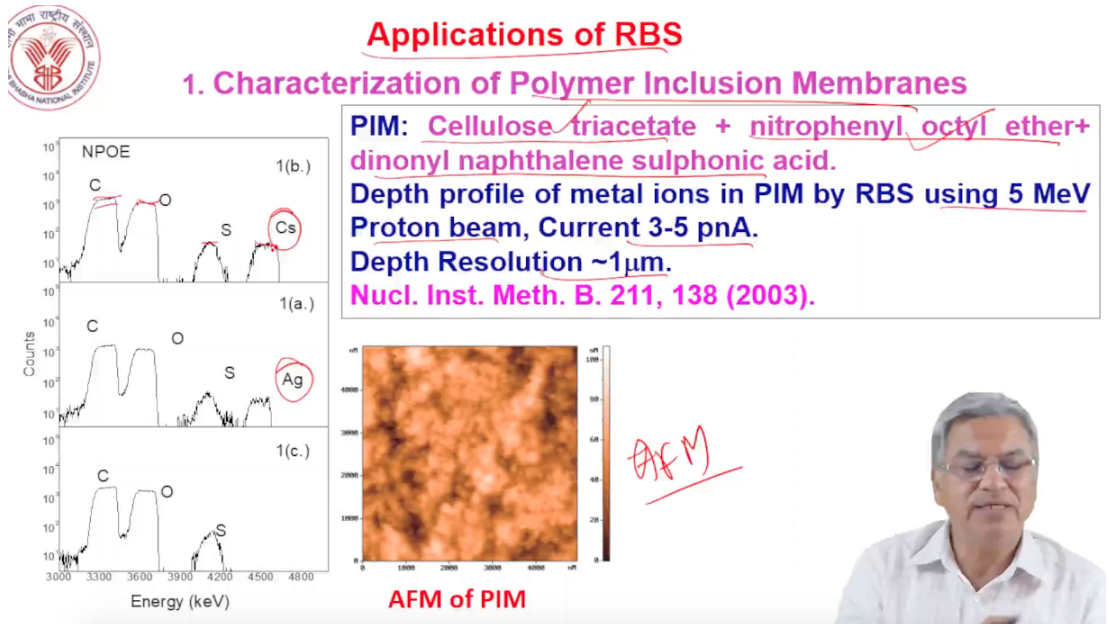


So you can have RBS in thin target or thick target. So thin target means the projectile is passing through the target, it is not stopped by the target. So you can see here, I have shown here some of the RBS spectra using a 4 MeV proton beam at Trombay, we have an accelerator called folded tandem ion accelerator (FOTIA). And so these are the metal foils of holmium, thulium and thorium and their thicknesses are in microns. So when the particle is going, so basically the objective was to see if there is oxidation of this pyrophoric metals on the surface.

If there is oxidation, there will be surface oxygen so when the beam is hitting the surface and so oxygen from both front and back will backscatter. So oxygen at the front, this is the front, this is back, high energy is from the front and low energy is from the back and this is the metal foil. So you can find out the thickness of the foil because you know the dE/dx and you know the energy of the projectile and similarly so for thulium, holmium, thorium, you can find out the oxygen concentration and the thickness of the foil. So this is a typical spectrum with thin foils.

If you have a thick target, the thick target means the projectile will stop somewhere in the target and so the high Z impurities will appear at a much higher energy than the low Z ones and there will be a big hump because of the bulk material.

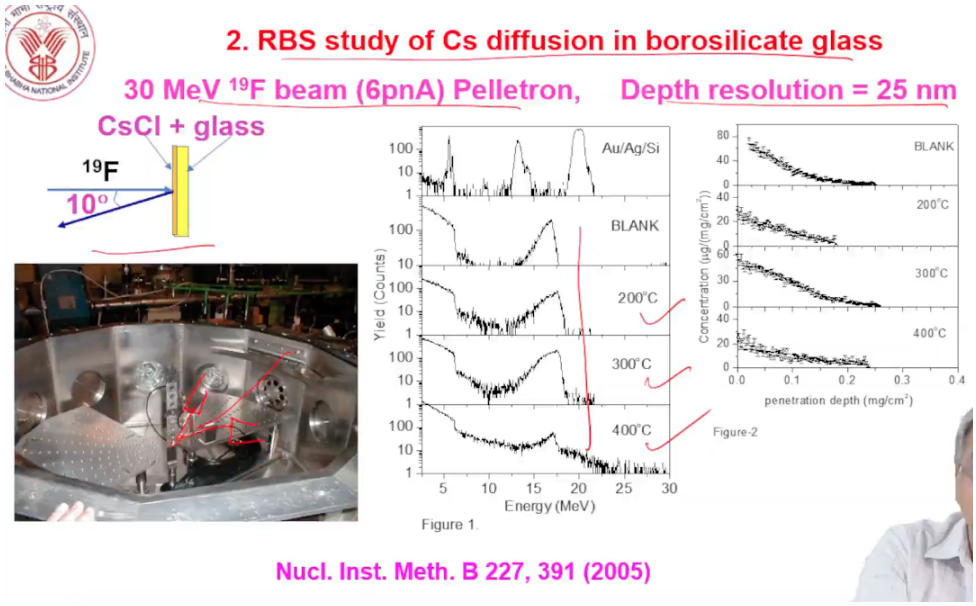
So this is a RBS spectrum of gallium oxide deposited on a thick silicon foil. So on the surface, suppose you have got silicon foil and you have the gallium oxide on the surface and you see the backscattering. So the gallium peak will come at much higher energy but the oxygen of gallium will be at much lower energy because the kinematics are dependent upon M_2 and silicon will be somewhere here and since silicon is very thick, the silicon will not appear as a peak but it will appear as a hump, thick edge type thing. So this is all due to silicon. So this is due to the thickness of the silicon. Since it is very infinitely larger compared to the projectile range, you will see a hump. So thick target will show a hump. And this is a very interesting experiment of multilayers. There were several layers deposited of gadolinium oxide and silica layers, each layer being of nanometers thick and that could be seen in the RBS spectrum. Of course, if you want to have high resolution, you require heavier projectiles like Fluorine-19 because you require heavy ion for better mass resolution. If you want to have high mass resolution then you require heavier ions because the stopping powers are high.



So some of the applications of RBS like this is a polymer inclusion membrane. These membranes are used for separation of metal ions like cesium and silver from the aqueous solution. So the constituents are cellulose triacetate, nitro phenyl octyl ether. This is the bulk material, this is a plasticizer and this is a carrier molecule, di-nonyl naphthalene sulphonic acid, which will take the cesium. And this polymer inclusion membrane like polymer membranes, they can take up cesium. So you want to know whether the cesium is distributed uniformly in the sample or only on the surface. So that was the

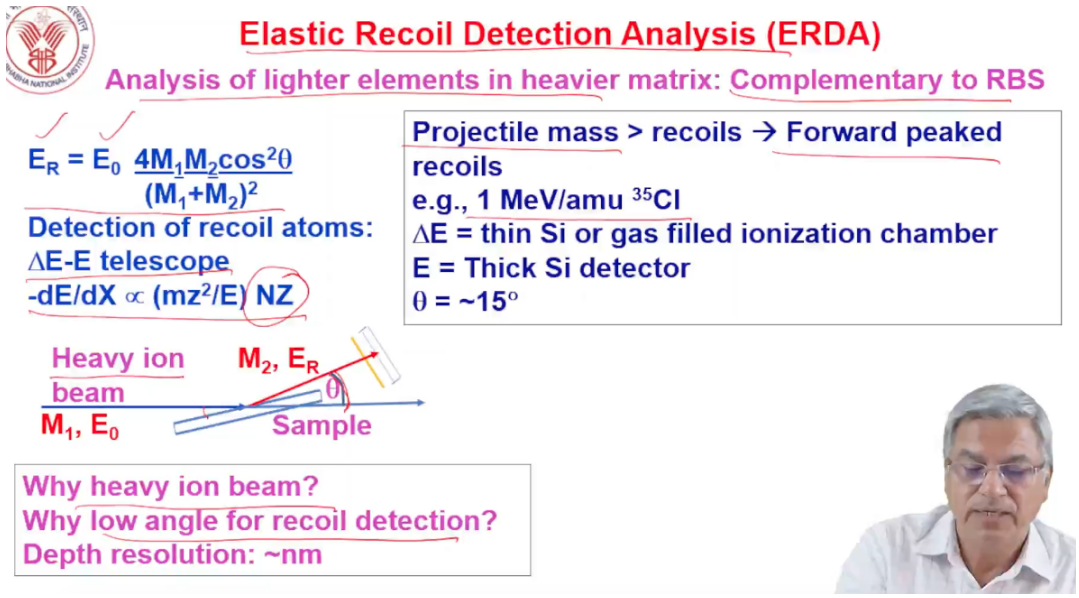
experiment, you can see depth profile of cesium in polymer inclusion membrane was studied by 5 MeV proton beam with a current of 3 to 5 nanoamperes. And now since proton has got very low stopping power, depth resolution is only 1 micron. But then the thickness of these films is of the order of 100 microns, so 1 micron is quite good.

And this is the AFM of the polymer inclusion membrane. And you can see here, carbon, oxygen, sulphur, so these are flat that means the polymer is uniformly distributed and even the RBS spectrum of cesium shows that cesium is uniformly distributed in the entire membrane. So up to 100 micron, the entire thickness of the membrane, the metal ion is distributed. So the idea was to see whether the this carrier molecule, sulphur bearing molecule is distributed uniformly in the membrane.



Similarly, another experiment was Rutherford backscaling spectrometry of cesium diffusion in borosilicate glass. These borosilicate glasses are used for immobilization of the high level waste at our department. And so you want to have higher depth resolution. So we use the fluorine-19 beam, 6 pnA, the depth resolution was 25 nanometer. So this is the typical experiment, the beam comes from here, hit the target and then backscattered Fluorine-19 beams are measured by two detectors. So you have a glass sample and on the surface we have cesium chloride evaporated. Then after evaporation, you anneal this sample at different temperatures. So different temperatures the cesium will go in the depth and you are studying to what depth it has got diffused.

So this depth profiling of your cesium can be done using RBS and then you analyze, so this is the RBS spectrum and this gives the penetration depth versus cesium concentration and the analysis of this gives you the diffusion coefficient of cesium in the glass matrix. These data are useful in subsequent analysis.



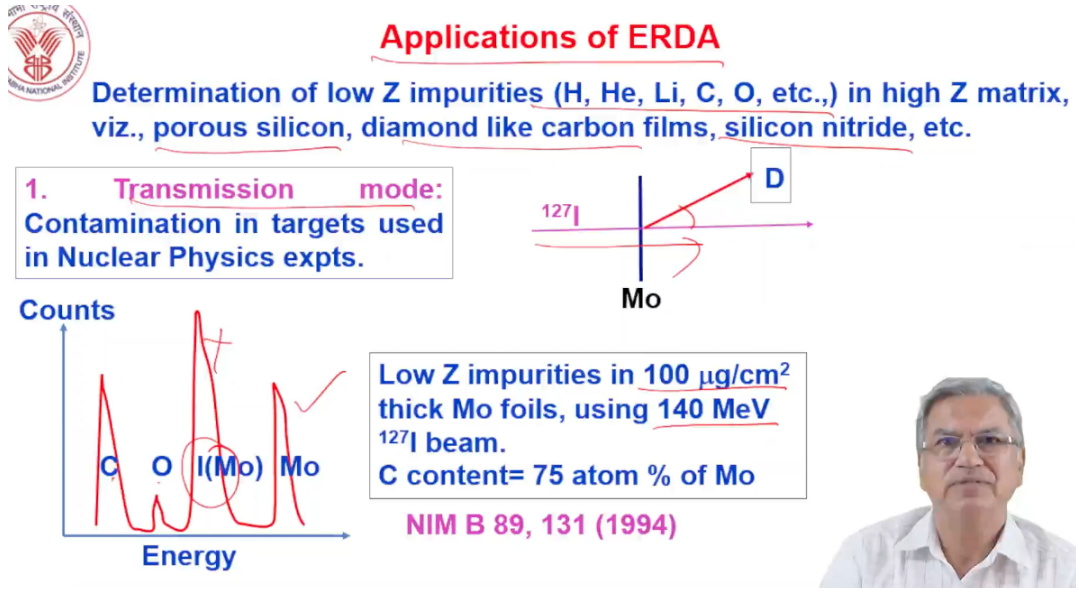
Ok, another technique is elastic recoil detection analysis (ERDA). We have not worked on this but it is also an important technique and which is in fact complementary to RBS because this technique is used for analysis of light elements in a heavier matrix. RBS we use for heavier elements in lighter matrix, this ERDA lighter elements in heavier matrix and why it is so? If you recollect the kinematic equation for elastic scattering, the energy of recoil we can say E_2 is related to projectile energy into $M_1M_2/(M_1 + M_2)^2 \cos\theta$ and so the projectile mass, you are detecting the recoils in forward direction.

$E_2 = E_1 * M_1M_2/(M_1 + M_2)^2 \cos\theta$

So if you have a heavier projectile mass, all the recoils will come in the forward direction. It will give you a bigger kick, the energy of the center of mass will be more and so all the recoils will come in the forward direction. So you put the detector in the forward angle and detect their spectrum by ΔE -E telescope, thin ΔE silicon detector and thick silicon or you can use a ΔE based on gas and the stopping power is given by mZ^2/E into the electron density of the medium. Since you are putting the detector in the forward angle, let us say 15, 20 degree or 30 degree, and the target may be thick , heavy ion beam will not travel much. So what you do, you put the target in a glancing position. So small angle, maybe 15 degree or so and so this is called a glancing position at forward angle you measure the recoils. So, the heavy ion beam is not passing through the target, the recoils may be coming from different depths in the sample.

So why heavy ions? Because we want the higher recoil energies and which will appear at the forward angles. And why low angles? Because with heavy ions as projectiles, the recoils will appear at the forward angles only. The recoils will come in the forward angle only if you have a heavier mass projectile.

So ERDA essentially is used to detect low Z impurities in a high Z matrix. Typically like you have one MeV per nucleon chlorine, so 35 MeV chlorine beam and you have a thin ΔE detector and a thick E detector and keep the telescope at 15 degree or so.



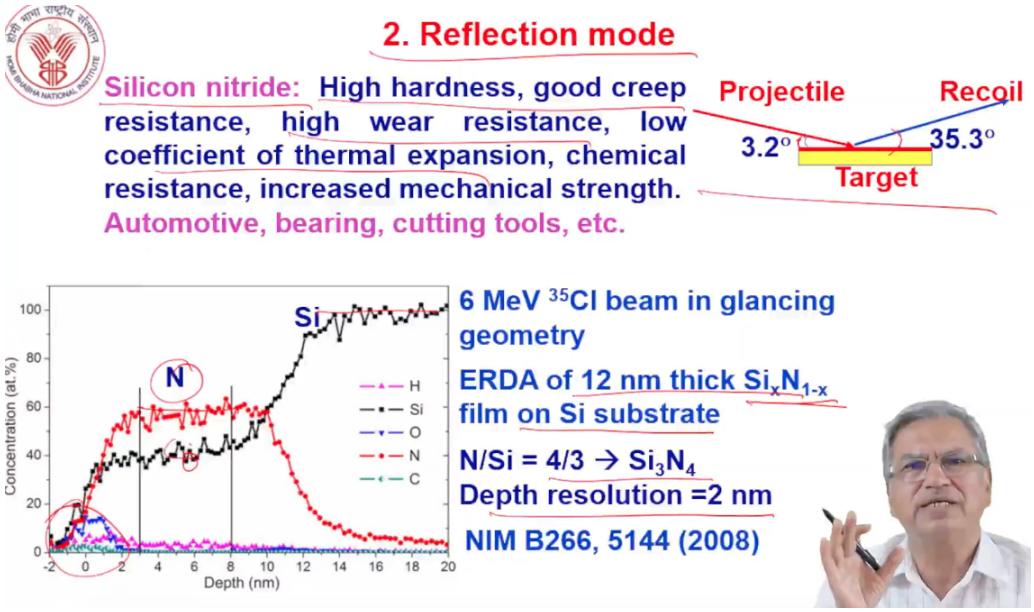
So I'll just quickly give you two applications of ERDA. One of them is, so ERDA is essentially used for low Z impurity detection, hydrogen, helium, lithium, carbon, oxygen in a high Z matrix. And most of the time, you know, these high Z matrices are high technology materials like porous silicon, diamond like carbon films, silicon nitride, etc.

And if you have a very thin target, like micron thick or even less, then you can use it in the transmission mode. This geometry, you don't need to put in the glancing angle beam can pass through this. So we have an iodine-127 beam of 140 MeV. You are determining the impurities in the molybdenum foil of 100 micrograms per centimeter square.

So we can put detector at forward angle. And now you will get the spectra due to different impurities like carbon, oxygen in the form of narrow peaks. And this is the iodine elastically scattered from molybdenum. So this is not of interest. And you have the molybdenum peak, the molybdenum recoils you will get.

And so they essentially wanted to see what the impurity is. So it was basically carbon impurity and the carbon content was found to be 75% of the molybdenum. So they are used in like nuclear physics experiment, we have a metal foil and you want to know what is the impurity in the target. Normally the ERDA is studied in reflection mode because you would have thick target it's like micron thick sample. So you would like to know what are the elements present as a function of depth. So one of the examples is the silicon nitride, it is a very, very technologically important material because of high hardness, good creep resistance, high wear resistance, its a hard material, low coefficient

of thermal expansion, chemically resistant and increased mechanical strength and fine applications in automotive industry, bearing, cutting tools, etc.



So one of the studies was that at glancing angle, they studied the recoils from the target. What is the target? Silicon nitride on a silicon substrate, thick silicon and on which you sputter or evaporate and you make a layer of silicon nitride, you can do even ion sputtering. And so under this position, so glancing angle and forward angle, you see the spectrum. So the thickness of this silicon nitride was actually very thin, it was about 12 nanometers, 12 nanometers thick silicon nitride and the composition is not known because you may be depositing by ion sputtering. But you can see here, you can get a flat concentration of nitrogen and silicon here that is the bulk silicon and you will see some impurities.

These impurities are seen here, low Z impurities and from the ratio of the nitrogen and silicon count, this is silicon, they could find out that it is a Si_3N_4 silicon nitride, Si_3N_4 with the depth resolution of 2 nanometers. So, as I was mentioning, this ERDA is basically used in specific cases where you are looking for a particular information. It is not just routine analytical technique, but it is used whenever you want specific information about the material that you have developed. So that is all I have to say. In the next lecture, I will take the other two techniques nuclear reaction analysis and particle induced gamma emission. Thank you very much.