Introduction to Non-Linear Optics and Its Applications
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Lecture - 24
Neumann's Principle (Contd.)
Centrosymmetric System

Welcome students to the new class of this Introduction to this Non-linear Optics and its Application course. Today lecture number 24. In the previous class we have started a very important concept called Neumann's principle. The Neumann's principle suggests that if I put some kind of symmetry operation on a crystal in terms of a susceptibility, which is a tensor that will going to change under the symmetry operation. But since the operation is symmetry in nature whatever the d matrix or susceptibility matrix you will going to find after putting this kind of operation, they are same that of the before.

So, before symmetry operation whatever the d matrix we will have, after applying this operation we will going to have the same d matrix if my operation is symmetry in nature; symmetry related to that crystal.

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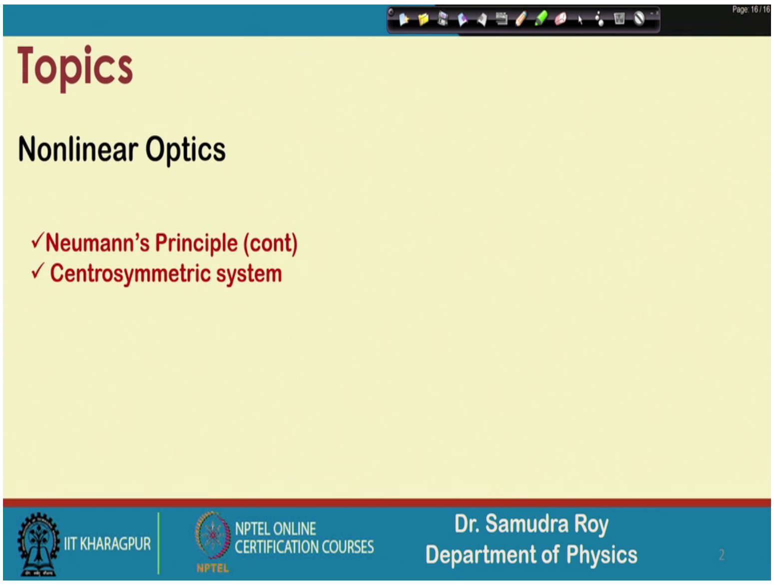

So, let us again try to find out because the Neumann's principle, we just state the things, but we never show any kind of application of that. So, today we will going to show some application on that. So, today we will going to show some application on that and then we will find a very interesting application of Neumann's principle and find that centrosymmetric system, where we have a center of the symmetry how the susceptibility tensor will going to change for that. And eventually we find that the second order tensor or the second order susceptibility or the second order nonlinearity will not be there; that means, it will going to vanish in case of centrosymmetric this is the classical example of Neumann's principle. So, let us continue with the Neumann's principle what we have.

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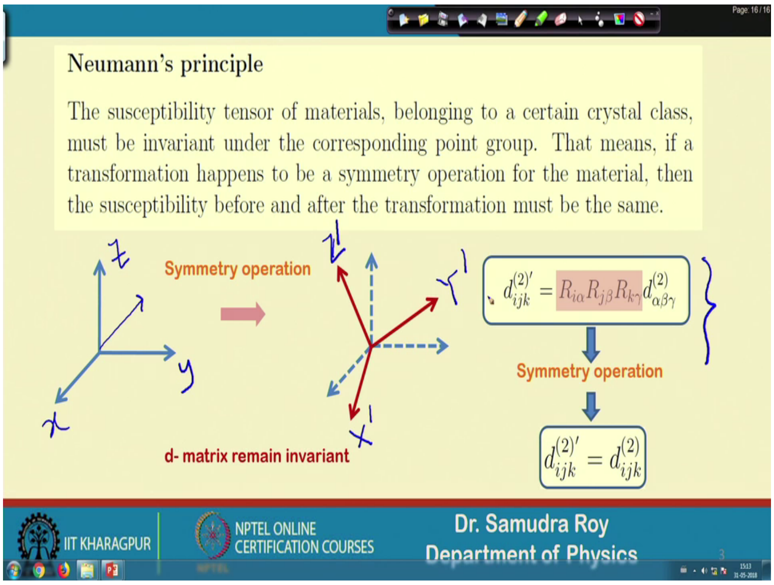

So, as a slide suggest that the Neumann's principle susceptibility tense tensor of the material belonging to a certain crystal class must be invariant under corresponding point group. That means, if I rotate or if I change this put some kind of symmetry operation, the d matrix will remain in variant. So, now, here once again let us understand this equation which is written here, very looking very simple equation that d ijk is the ij kt-h component of d matrix under symmetry operation and if I put some kind of symmetry operation like changing the coordinate system.

So, this coordinate new coordinate new coordinate system the d matrix has a prime on over that. So, this is under prime coordinate system; say this is a non-prime coordinate system and this is a prime coordinate system if this is x y z this is a principle axis say suppose, and this new one is big X big Y and big Z which is a prime coordinate system or if I put some kind of prime it will be easier you to understand. So, in this coordinate system the red one the d matrix the form of the d matrix will change; like the component of the vector change in here if I have a vector here the component will going to change if I rotate my coordinate system, in terms of vector also in terms of vec I mean if I in place of vector if I use say some sort of matrix or tensor then the tensor components also going to change under this rotation or this kind of symmetry operation.

So, if I put this operation over a matrix, then the relationship between the old and new matrix will be represented by this particular form as it is shown. That ijk component is represented by alpha beta gamma; and this alpha beta gamma is a repeated one; that means, if I put summation sign over that it should be over alpha beta gamma.

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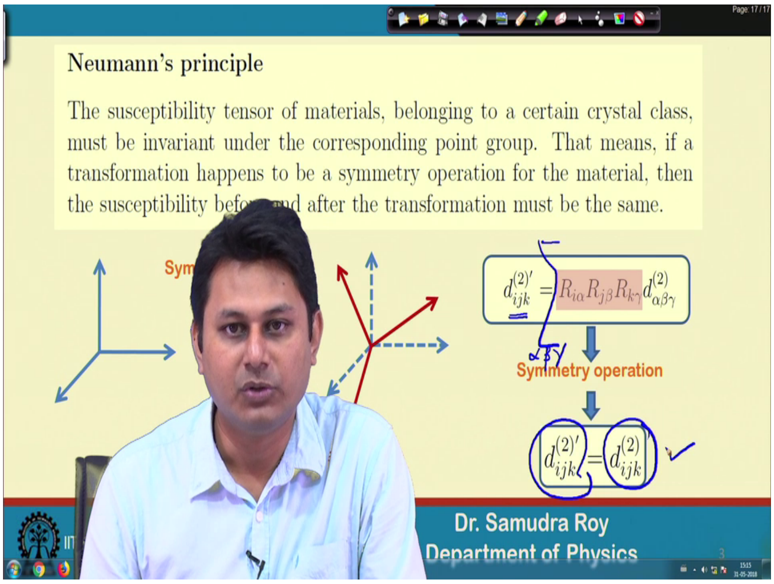

So, alpha beta gamma will going to change in order to have one specific component in d ijk we will have to make a some over alpha beta gamma and all the cases we need to find out what is my Rj R 1 R i alpha R j beta and R k gamma and then only we can able to find out what is my ijk

Now, is my operation is symmetric in nature, then after doing everything whatever we will get as d ijk prime, it should be the same value that we start with that is it should be equal to d ijk; because eventually after applying the symmetry operation this things will not going to change. So, you will eventually have the same value of d matrix. So, this basically gives us additional information, through which we can find out what should be over d matrix components.

So, let us now go to our next slide, ok.

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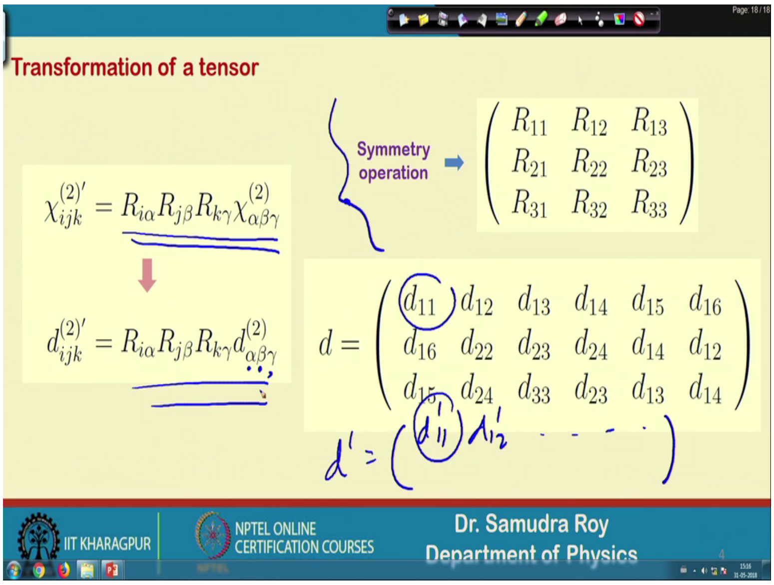

So, this is the old again this is the old slide where a we will now say that my whatever that matrix we are going to use as a transformation matrix, rotation matrix or in general operation matrix, this is symmetric in nature. That means, if I use this matrix components to find out what is my new susceptibility components. So, this operation is symmetric, but the operation rule is again the same thing. So, these are the operation rule and this is my total matrix. So, this is the structure I have given R, I have given d I will going to apply over that. So, my goal here is to find out the prime. So, if I want to find out it should be d 11 prime d 12 prime and so on.

So, what is the relationship between d 11 and d 11 prime I can figure out with this R. So, one component there will be several number of terms are appearing because it will be alpha beta gamma. So, alpha beta gamma is a running indices or a damming indices. So, it will going to rum from 1 to 3. So, there will be in principle many terms there and it depends on how the symmetry operation or the symmetry matrix will look like. So, let us take a example so, that you can understand exactly what I am trying to say.

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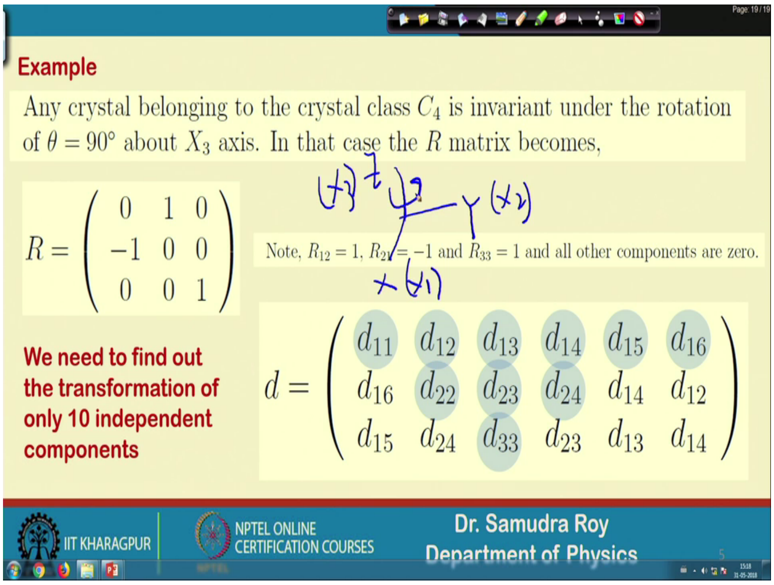

So, suppose we have a crystal which belonging to the crystal class C 4 and; that means, it is invariant under the rotation of 9 degree about X 3 axis. So, we have a axis and if I rotate this axis.

So, we have a coordinate system. So, suppose we have a XYZ coordinate system X Y and Z and if XYZ is represented here X 1 X 2 and X 3this is basically X 3 this is basically X 2 and X 3. So, if I rotate these things to 90 degree or pi by 2, then whatever the crystal we have since it is the symmetry operation, we will have the same crystal again. So, first we need to find out once the operation is given to us the first we need to find out what is my R here. That means, we need to first find out the symmetry operation matrix.

So, here we can see since we are rotating 90 degree.

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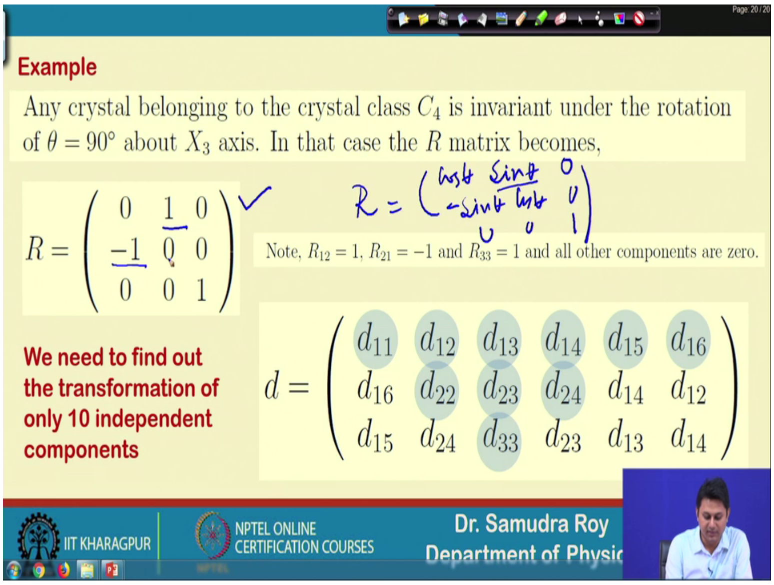

So, my R matrix will be simply 0 1 0 minus 1 0 0, 0 0 1 because rotation matrix you know it was cos theta, sin theta 0 minus of sin theta cos theta 0, and 0 0 1 this js the rotation matrix around z. So, sin theta means sin 90 degree it should be one. So, this negative sign give me minus 1 here, sin theta here also we have 11 and cos 90 degree cos 90 degree is 0 and this one is there. So, eventually we will have a matrix R here. One thing you should note that most of this elements are 0 here, only 3 elements are there in R matrix which are non-zero R 12 R 21 and R 33. So, these are the terms that will go into effect on this rotation thing or this operation thing, but the others things are not going to since it is 0, it is not going to play any role.

So, the m here is to if a d matrix is given like this, we need to find out the transformation of these coefficients. So, now, you can see that there are only 10 different coefficients are there; so if you able to find out after transformation what should be my coefficient. So, we just need to find out this 10 independent coefficient, because others are the same values. So, once we know this 10 independent coefficient I can make after the rotation operation whatever the new matrix we have, we know that these and old matrix is same then we will try to find out a one to one correspondence between them, and eventually independent values of this. So, let us see how we can do that.

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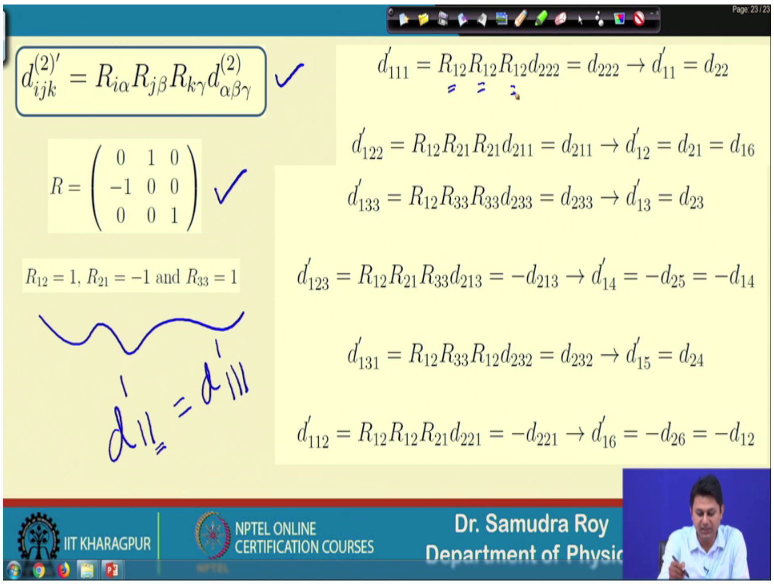

So, R is given to us. So, R is given to us and d matrix also given to us. So, let us start the operation. So, one by one we will going to do. So, this is mine my main equation for transformation in the left hand side, R is given where these are the non-zero elements that we have written explicitly it should be R 11 R 21 and all this things now what we will do? We will try to find out first we will try to find out what is my d prime 11 this is the first element.

D prime 11 is nothing but d prime 111 because I need to use the extended form not the contracted form, because 3 coefficients are there. So, once we write d 111. So, I need to write R 1 here, R 1 here R 1 here. So, let me give you the idea how to do that.

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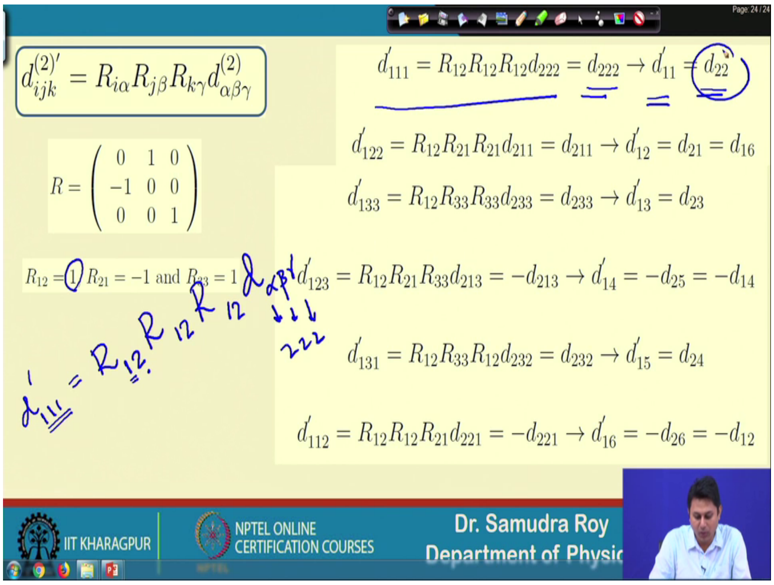

So, I need to find out this is my rule. So, I need to find out 111 prime. So, this is my rule. So, rule suggests ijk is 111. So, R 1 it should be here R 1 should be here, R 1 should be here and d alpha beta gamma will be there, I do not know will be the alpha beta gamma.

But if you look very carefully whatever the value you put except 2 you will get 0 always because only R 1 2 is 1. So, if I write one here then this value has to be 2. If you 2 if you put one then R 1 1 is 0. So, entire multiplication you go to 0; for non-zero elements I am just writing what should be the non-zero elements here. So, you have to put R 2 here in the same way since it is 1 you have to put R 2 here and 2 here. So, that you have non zero elements.

All the other elements will going to vanish once you find d 1 11. So, if I put 2 2 2 here. So, automatically my choice of alpha beta and gamma will be 2 2 2. So, this is essentially the equation that we have. So, we find that d 2 2 2 if nothing but d 1 1 prime or d 1 1 prime is equal to d 2 2; that is my first coefficient and this coefficient d 1 1 prime can be represented in terms of d 2 2 which is a non-prime coefficient ok.

Now next what should be the value of d 2 2 d 1 2 2 the next one next one is d 1 2.

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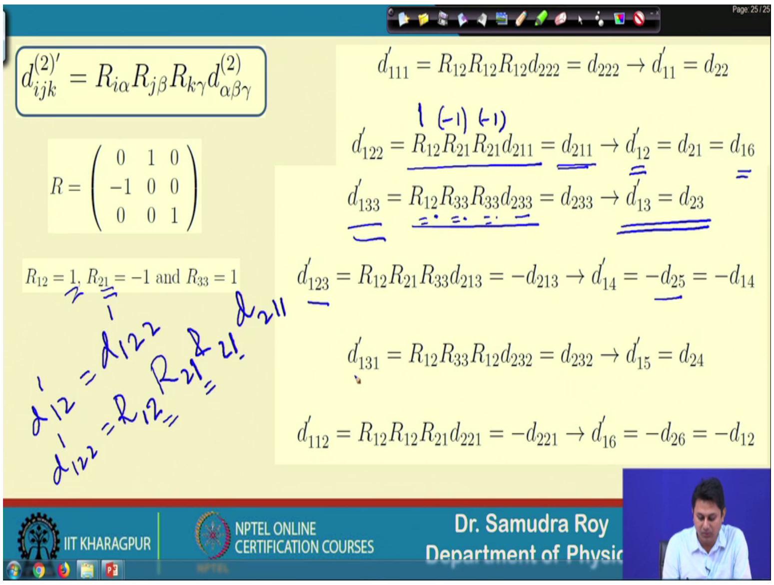

So, d 1 2 prime is d 1 2 2 prime, now we know that d prime 1 2 2 means I have to put R 1 here, R 2 here and R 2 here because ijk here is 1 2 2. So, I is equal to 1, j is equal to 2 k is equal to 2. Again we find that if as soon as I put R 1 here, only for non-zero components if I want to find I need to put R 2; because R 1 2 is only one other terms are 0. When I put R 2 here, then we find that 2 one is a component which is non-zeros I have to put 1 1 here and here again I want to put we need to put R 2 1 here, so that here this 2 1 1 I put like this.

So, see this expression in the expression, I am essentially writing the same thing. Now R 1 1, R 1 2, R 2 1, R 2 1 this is 1, this is minus 1 and this is minus 1 if I am multiplied it should be 1. So, eventually we have d 2 1 1 is equal to eventually we have from this we have d 1 2 prime is d 1 6. So, you can see that even after finding 10 independent components, this 10 independent component again there are equal to each other; so d 2 1 and d 1 6 that we know that they should be equal and we are getting the same thing once again.

Now what happened to due 133 the next term? So, again in the similar way we have to put R 13 and 3 here. So, R 3 3 is non-zero. So, I need to put 3 here, 3 here and once I put R 1 I need to put two. So, that we have a non-zero elements here and all the other elements if you calculate meticulously see you will find that all the other elements will be 0 because of this R matrix components.

So, now d 2 3 3 you need to put. So, from here we can find that d 1 3 prime is essentially d 2 3 in non-prime system. So, in the similar way we find d 1 2 3 which is this d 1 3 which is this and so on. So, how many components we figure out here independent components we know that in the previous slides; we as we have already mentioned that how many components we need to calculate. So, we need to calculate d 1 1, d 1 2, d 1 3, d 1 4, d 1 5, d 1 6, then d 2 2, then d 2 3, d 2 4. So, 10 independent components we need to find out and we start with d 1 1 d 2 2 d 3 3. So, let us see in the next slide how many components we figure out in non-prime and what is the relationship between this 2 from prime prime to non-prime prime

So, how many components we here find? This is one component this is second component this is third component and this is forth component this is fifth component this is sixth. So, 6 different components we figure out again there are 4 different components still remaining, but we will going to use the similar prose due to figure out of this 6. So, let us see whether we can find out the 6 or not. These are the next 4 components and again we will use the same thing. So, let me do that once again for example, any arbitrary component if I want to find out the d 2 2 3.

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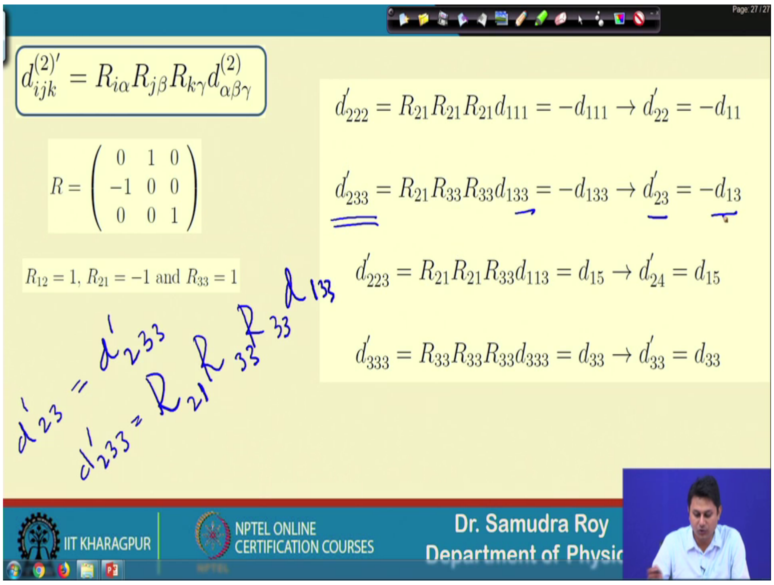

So, d prime 2 3 it is components if I like to find it will be do d 2 3 3. So, d prime 2 3 3 if I put the R value I need to put R 2 here, R 3 here R 3 here and d and here R 2 3 3 once I put then I have to put R 1 here, R 3 here, and R 3 here. Once I put we will find that here I need to put 133. So, d 2 3 is essentially minus h of d 1 3.

So, this from this one we one thing we can find that, they are related to because after making the transformation whatever the d matrix you will get, that should be equal to the previous one so; that means, this component if this component is minus of this component, then there should be some relationship. So, we will find what is the relationship. So, let me do that

So, eh we figure out in this 2 slide we figured all 10 independent components in prime-prime or after operating the symmetry operation on to the system what should be the form of the d matrix we figure out well. So, let us now try to find out what is going on. So, these are the entire list of whatever the values we figure out and that is important; so after figure out all the components.

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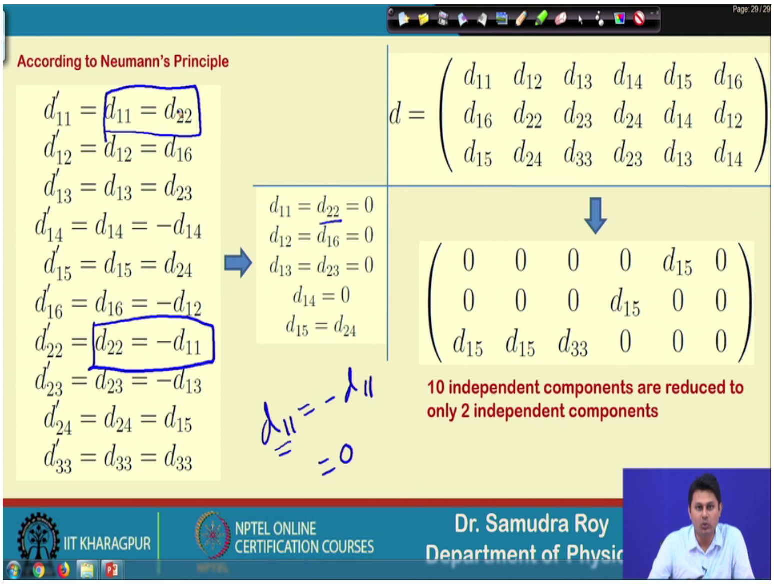

So, d 1 1 is related to non-prime. So, d 2 2, d 1 2 is related to 1 6 and so on. But here one thing you should note that d 1 1 prime and d 1 1 they has to be in the same value because I am applying the symmetry operation once I am applying the symmetry operation my Neumann's principle is valid, and Neumann's principle suggests that if you apply the symmetry operation then the new component which is prime component should be equal to the old one. So, d 1 d prime 1 1 should be equal to d 1 1; d prime 1 2 should be equal to d 1 2, d prime 1 3 should be equal to 1 3 and so on so; that means, we will have a relationship between that this 2 d 1 1 and d 2 2 should be equal, d 1 2 and d 1 4 1 6 should be equal, d 1 3 d 2 3 should be equal. So, we have these conditions.

So, now from here we find some kind of interesting expressions that is really interesting. So, here d 1 1 we find one equation its d 2 2 in the similar way we find d 2 2 is equal to d minus 1 1. See if I compare this 2 equation we will verily find if I now compare this 2 things we find d 1 1 is equal to minus d 1 d this has to be this now it is never possible that d 1 1 equal to d minus of 11; that means, d 1 1 has to be 0. Once we know d 1 1 only solution of this equation is d 1 1 is 0. So, d 1 1 is 0 means d 2 2 is 0 because d 1 1 and d 2 2 is 0.

So, you can see that applying Neumann's principle, we can find out exactly what is the value of d matrix what is the value of d matrix and find there are many zeros in d matrix. So, d 1 1, d 2 2 is 0 that we find what else? Let us see this equation d 1 6 and d 1 2 are same here we find another equation d 1 6 and d 1 2, but they are related with a negative sign. Now from this equation these and this equation we can verily say that d 1 2 and d 1 6 has go be 0, otherwise these 2 equations cannot be satisfied simultaneously.

What else? We can find that d 1 3 and d 2 3 and again here d 2 3 and minus of d 1 3. If I again compare this 2 things like the previous cases, we find that this has to be 0 d 1 3 equal to 0 or d 2 3 equal to 0 and that is the only solution for this 2 given equation. So, again find this twos are 0 what about the 1 4? We have a self we have a equation, where it suggest that d 1 4 is equal to minus of d 1 4, again this equation cannot be possible unless the value of d 1 4 is 0.

So; that means, done 4 value is 0 anything else? D 1 4 we have one equation, d 1 5 is equal to d 2 4, but we do not have any other equation so; that means, d 1 5 and d 2 4 for this symmetry operations are same or the d elements the matrix elements on d is same for this 2 for 1 5 and d 1 4 and 2. Apart from that there are other equation which has consistent that d 3 3 is d 3 3 that is fine no problem in that. So, we have almost all the equation in our hand, here also we have d 2 4 and d 2 5. So, this 2 equations suggest cannot find any any explicit value of d 1 5, we just have the relationship and this relationship suggests that d 1 5 and d 2 4 is same.

So, once we have all this information. So, all the values of d 1 1 and d 1 2 is known to us we started with these values these are our starting point. So, once we have this one in our hand. So, next thing we apply the symmetry operation and when I apply the symmetry operation we find this values are zero there is a relationship this. So, from this we will have finally, this expression of d matrix and this expression suggest that d 1 1 is 0 this is 0, this is 0, this is 0, the 1 5 which is not equal to 0, I put this 1 5 1 6 again 0 from here we can write all this others things are 0. So, d 1 5 I should write here because d 2 4 and d 1 5 are same. So, this 2 are the same elements then 0 0, d 1 5 is here d 1 5 is here this is already because in this operation it is already there.

And then d 3 3 is one independent coefficient that is sitting here. So, eventually how many independent coefficient are there in d matrix, we started with 27 then reduce it to 18 then we reduce it to 10 and ap now after applying Neumann's principle, we reduce it to 2 that is I think a significant reductions of the coefficient now we find that we have only 2 components which are independent to 2 each other and this symmetry operation basically gives us the idea that how one can find the d matrix different d matrix well.

Let me finally; tell the centrosymmetric system what is going on. So, in crystallography we have some kind of system, which is centrosymmetric in nature in centrosymmetric what happen? The crystal have a center of symmetry so; that means, if I change the point x y z to minus x minus y minus z. So, the material should be distinguishable; that means, there is a center of symmetry somewhere. So, we have this centre of symmetry around this center of symmetry we have this some kind of indistinguishable point here

So, the once I go from minus 6 x y z to minus x minus y minus z my matrix here will going to change let me go back to previous slide. So, this is the matrix the operation matrix form. So, R it is the very important that first we form first we know what is my R matrix. So, R matrix should have this kind of form that minus 1 0 0 0 minus 1 0 and 0 0 minus 1. So, this is the form of the R matrix. So, once we have the R matrix in our hand, then the next thing is to find out for this operation what should be the new elements. We will going to us the exactly the same expression that we are going to use for last few classes or last few slides and that is this.

So, susceptibility second order susceptibility will be represented in this way, but here one important thing we should know that this operation matrix diagonal in nature.

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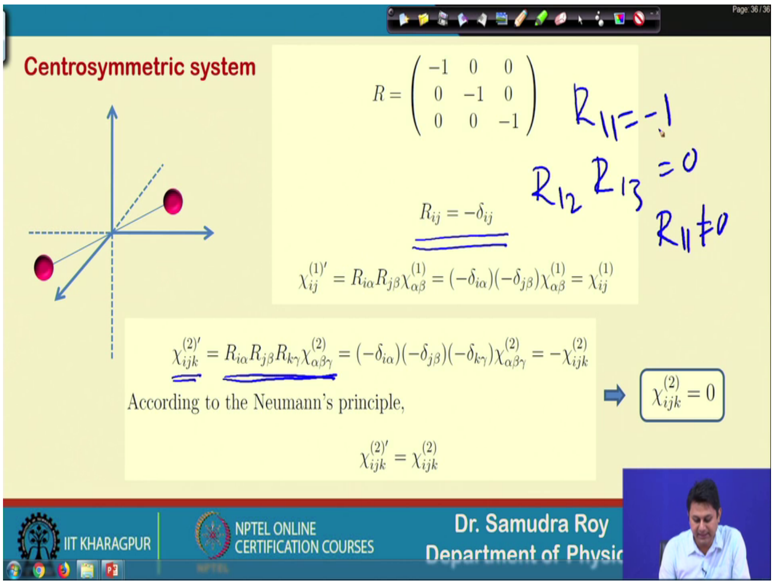

So, R ij is essentially delta ij with negative sign; that means, R 1 2, R 1 3 it will be 0 only R 1 1 is not equal to 0 and this R 1 1 value is minus 1. In the similar way R 2 1, R 2 3 are 0 and R 2 2 is minus 1. See if I write this thing we will find important information. So, one important information this is the chi in kai ij is a, this is the second order susceptibility chi ij is the second order susceptibility oh or the first order susceptibility rather.

. So, first order susceptibility if I apply this R operator on this first order susceptibility, we find that it should be ij there are 2 components it should be R i alpha Rj beta and this alpha beta. So, once we have this relationship in our hand, then we put this delta operator then we find that it will be minus 1 minus one and it is one. So, i j and ij are same so; that means, there is no change for centrosymmetric system there is no change of I apply this symmetry operation, there is no change of first order susceptibility. The first order susceptibility remain conserved, but what about the second order susceptibility second order susceptibility is more important here. So, ijk component will be there because it is a second order susceptibility and they are related to alpha beta gamma and alpha beta gamma; so summation over alpha beta gamma.

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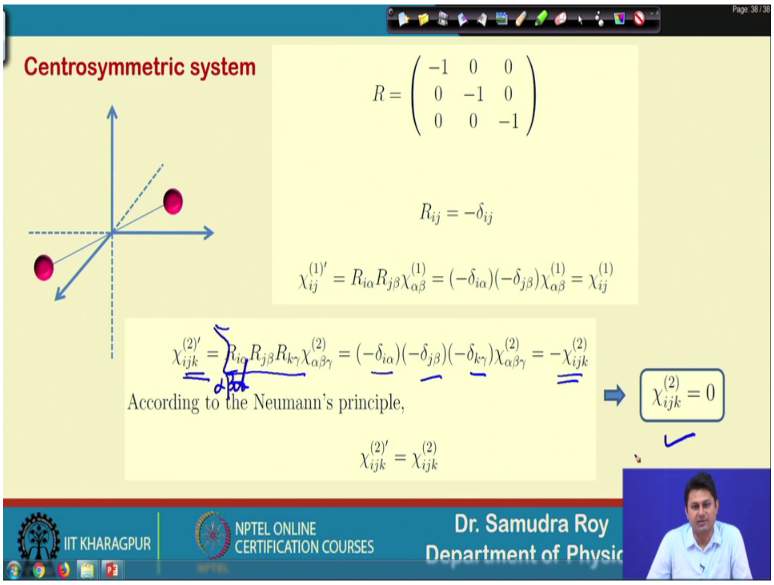

But this alpha beta gamma if I go on to have different values, all the values will be 0 only this delta function values suggests that only the delta ii delta jj and delta kk will give you something otherwise all the contribution is 0.

So, if we do these things we will find that susceptibility ijk is equal to susceptibility jijk with an negative sign. And this is 2 for all the ijks and these suggest a very important expression that susceptibility ijk for all the ijks is 0. So, this susceptibility value ijk was second order susceptibility 0 under this centrosymmetric system suggest that for centrosymmetric system, there is no second order susceptibility. So, second order susceptibility will be 0 or second order susceptibility will vanish for centrosymmetric system.

Well, this is the thing we like to we like to discuss in today's class. So, we will going to end here. So, in the next class we will start from more important applications, and today we find a very important thing that under symmetry operation the structure of the d matrix will not remained change it will be same thing that a before the symmetry operation. And from that we can reduce the different components of the d matrix and only first centrosymmetric system if I apply the Neumann's principle, we will find that the second order susceptibility going to vanish. And that is the very important and classical example of the symmetry operation, and in the next classes we will find out more on these things.

And with, this note let me conclude here. So, see you in the next class.

Thank you for your attention.