Introduction to Non-Linear Optics and Its Applications
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Lecture - 41
Third Order Nonlinear Effect

So, welcome student to the next class of Introduction to Non-Linear Optics and its Application. So, in the previous class we have discussed something related to chi 2 effect. In fact, entire classes for so for whatever the classes we have taken, so we mainly cover the chi 2 effect. So, today we will going to start the next higher order effect which is called the chi 3 effect. So let us see what we have in today's class.

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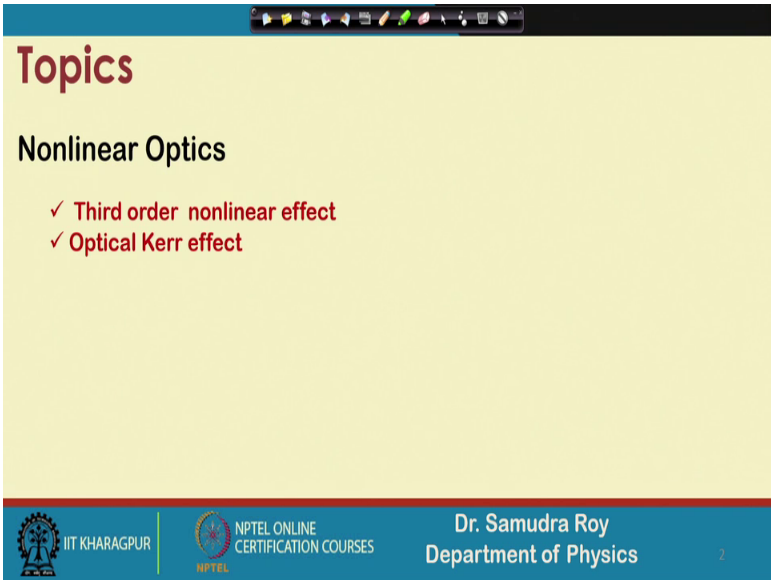

So, today we have third order non-linear effect. So far we have we have been discussing of second order non-linear effect. So, today we will going to start another important thing which is third order non-linear effect. And then we like to discuss something about Kerr effect if the time permit we will discuss in detail otherwise we will do that in the next class. So let us go back to the slides.

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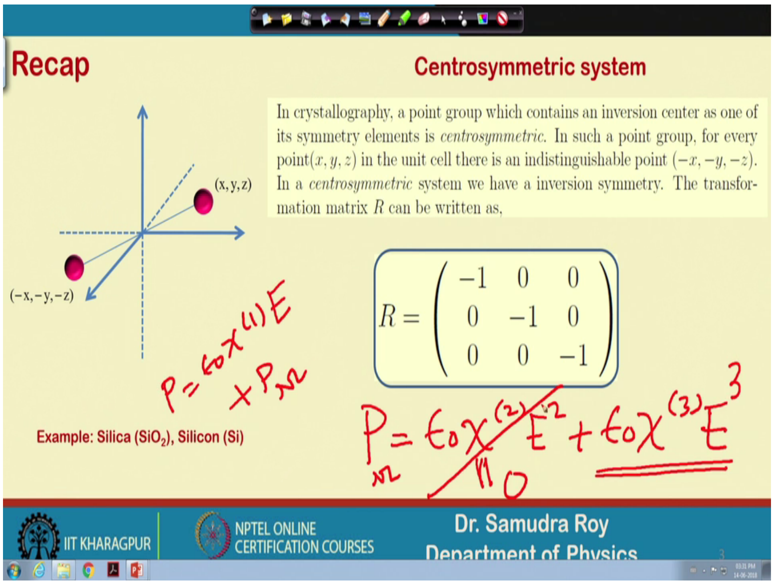

So, this is something we have already done that the centrosymmetric system. Why we are discussing the centrosymmetric system in this particular class that is important, because centrosymmetric system is such a system where we have the effect of chi 2 0. That means, if I write a relation related to polarization and electric field we know that the relation is P is epsilon 0 P non-linear other chi 2 E square and epsilon 0 chi 3 E cube.

This is the non-linear part of the polarization that we always, always use. If we write the total polarization it will be as usual P equal to epsilon 0 chi 1 E plus P NL. And this P NL is now represented in terms of this that we are doing in several time. So, in centrosymmetric system what happened that we already this is shown to you that this chi 2 is 0 this term is 0; that means, in the P non-linear term we never will going to have this affects.

So, in that case what should be the expression of the non-linearity of non-linear polarization? So, this is the expression of non-linear polarization and the in this expression we find that chi 2 is 0 so it will start with the effect of chi 3. So chi 3 is the first higher order effect that one should consider if the system is centrosymmetric in nature.

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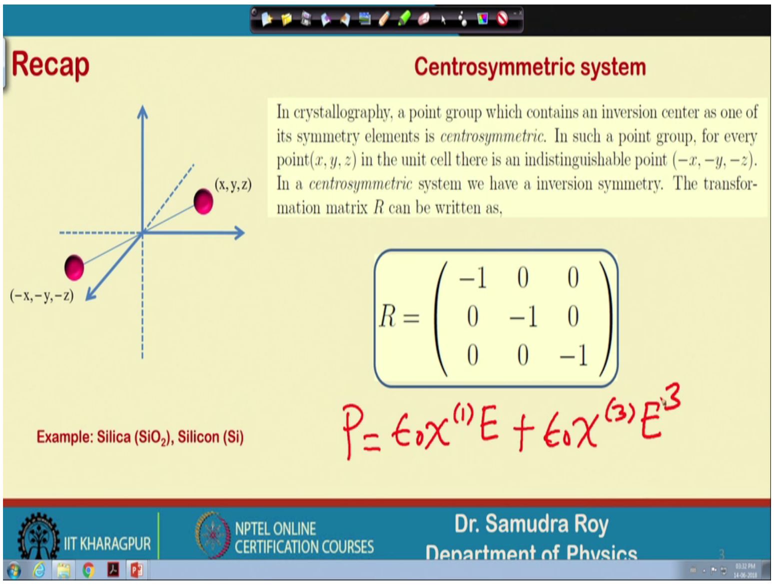

so if I write the polarization in centrosymmetric system the total polarization will be the linear part, the linear polarization which is epsilon 0 E chi 1 E is proportional to E plus the second term will start from chi 3 not chi 2 because chi 2 is 0. Well what is centrosymmetric system; that again let me describe briefly. That is a system where we have a centre of symmetry, if I change the co-ordinate xyz for a point.

Then we will have another point in the exactly minus x minus y minus z. So; that means, this quantity should have or this material which is centrosymmetric in nature they have a symmetry against this transformation. So, this transformation matrix R can be represented in this. So you can readily see that if R is written in this particular form then if I use operate over some xyz.

So, as a result we will get minus x minus y minus z. So if I go minus x minus y minus z. So, there should be a symmetry operation and it is a indistinguishable point in minus x minus y minus z. Silica silicon these are the few materials which are the example of centrosymmetric system. Once we have a centrosymmetric system now the next thing to find out how the non-linear.

Term optical nonlinearity will be affecting here; and as I mention the first higher order effect will be the chi 2 effect. So, first higher order effect will be the chi 3 effect, because chi 2 is 0 here. Why chi 2 is 0 and why chi 3 is the first effect first higher order effect.

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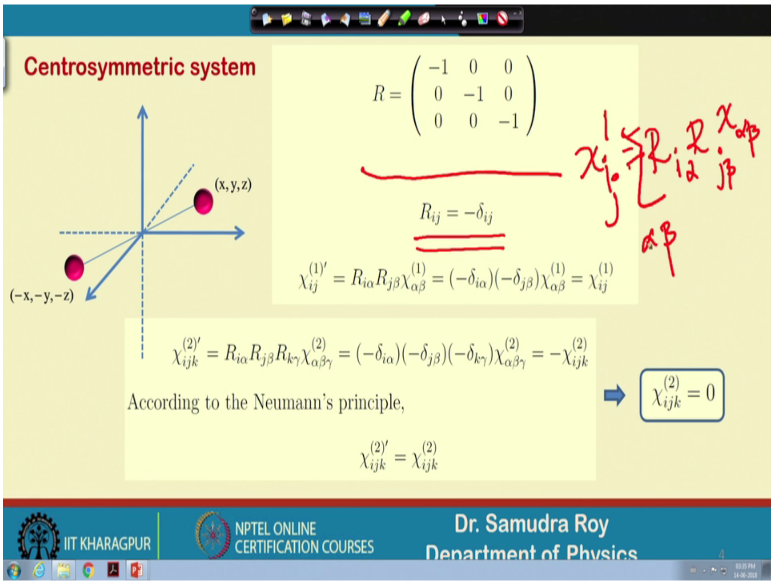

We can understand this simple form where, if a transformation matrix is giving to us. Then we can readily fine with this transformation, what should be the coefficient of the original matrix after transformation what is the coefficient of that particular component of the matrix we can readily know with this transformation rule. So, here R let me remind you once again how this rule works. That if R is given like this, then every point R ij is now nothing, but a delta function because only the diagonal elements are there with a negative sign.

So, it should be minus of delta ij and we know a general transformation rule. That is if I want to find out say chi ij prime; that means, chi ij is a quantity of the tensor element at prime frame. It should be related to R like R i R j alpha beta chi alpha beta; that means, alpha beta is repeated index. So that means, it should be sum over alpha beta like this. And that means, if I sum over alpha beta then whatever, the value we have in the right hand side should be the value of chi ij at prime frame.

So, just using this rule this is the rule that we have already discussed in our previous classes. So, the exactly the same thing is written here, and if I now going to use this rule you can readily see that for ij chi is the first order susceptibility, it will not going to change, whatever the value we have after having the symmetry operation this value will be same.

So, there is no change of the susceptibility which is of first order. What about the second order susceptibility, second order susceptibility here it should be ijk, because the number of elements as soon as I go to second order susceptibility, we know the number of elements ijk will now change from 1 to 3 in each cases. So, we will have 27 elements.

And now, if I write this ijk, in terms of the ijk prime in terms of R and their previous components namely called alpha beta gamma, where alpha beta gamma is a over sum. Then this relation suggest that it should be minus delta i alpha minus delta j alpha minus delta k alpha, because R i alpha R j alpha j beta and R k gamma is nothing, but the delta function with the negative sign.

And when I put all this term together I will find the susceptibility ijk with the negative sign. So, what is the meaning of that? Now it is a symmetry operation, so whatever the value we have that should be equal to the same value after the transformation. So, after the transformation I have ijk prime before the transformation it was ijk.

So, ijk and ijk prime should be same. If I do these things so we will find that it is possible. So, this is negative of ijk alpha ijk chi ijk 2 prime is negative of chi ijk 2, but this has to be same. So, this has to be same, but they are related with the negative sign; that means this quantity is 0. So, second order susceptibility has to be 0 to satisfy this particular symmetry operation according to the Neumann's principle. So, what is Neumann's principle also discuss in this course in some previous classes.

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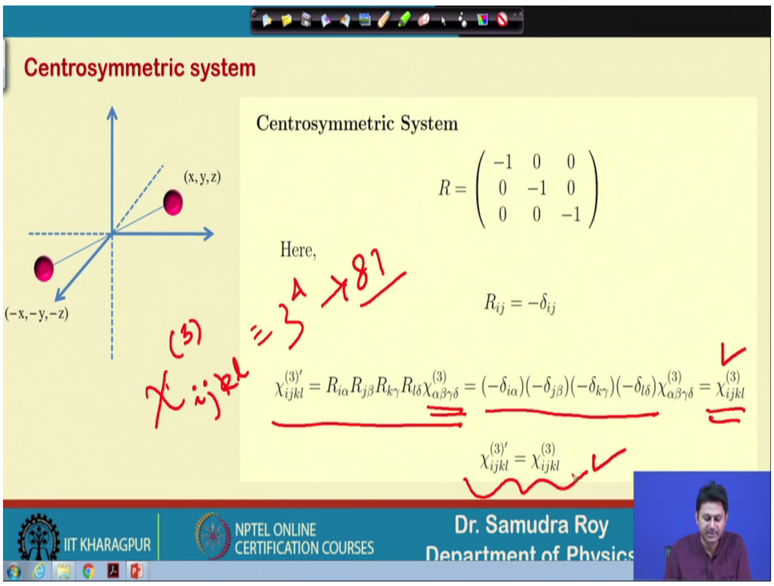

So, now you we will extend these things for third order susceptibility. So, once we say it is a third order susceptibility then I basically, I am talking about chi 3. So, chi 3 should have 4 indices here i j k and l. So this matrix in principal should have 3 into 10 to the power 3 into 3 to the power 4 elements. So, 81 elements it should have, but when relate with these symmetry operation R. The rule of this operation is alpha chi ijkl prime is R i alpha R j beta R k gamma R l delta and alpha beta gamma delta.

So, this will be over from and exactly in the previous way the way we have done we can do the same treatment here. And then i alpha i alpha R i alpha R j beta and R k gamma, I just replace with this matrix form which is minus 1 minus 1 minus 1 and minus 1 in all the cases, so there will be 4 time minus 1. And if I multiply we will have the same element chi i delta kj, because here is a delta function when alpha beta gamma delta are same for example, here alpha will be this quantity will be non 0 only when alpha equal to i. And that means, I am talking about delta ii delta ii nothing, but minus 1.

So, it should be minus 1 minus 1 minus 1 minus 1 and if I multiply all this 4 minus ones I will have a plus 1. So, I will have the same quantity that we should do we should have after having the symmetry operation. That means, the second order susceptibility even if it is 0, but the third order susceptibility is a non-vanishing.

That means, the first higher order effect one should expect the higher order non-linear effect rather, one should expect from this third order effects. So, third order effect basically the first higher order effect here in centrosymmetric system. So, that is why it should be very, very important it should be very, very important to understand; what is the effect of these chi 3.

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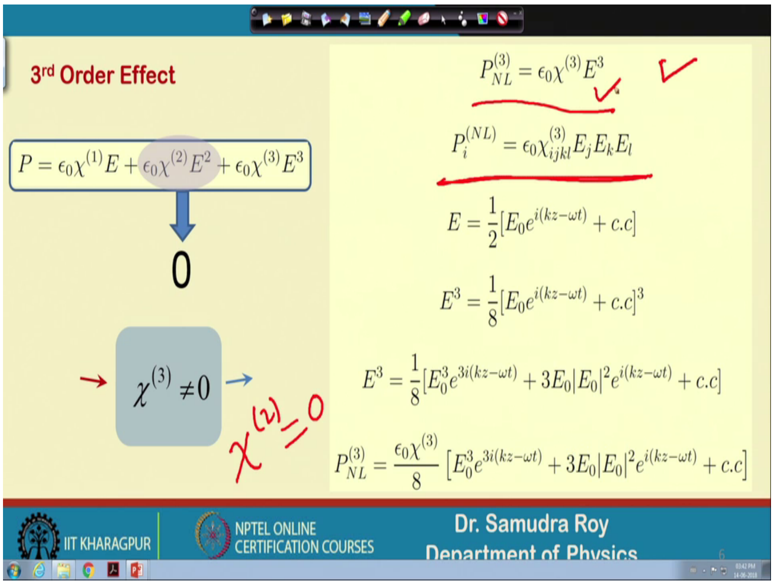

So, chi 2 effect of the chi 2 we elaborately discuss in our previous class, and now in our previous all our classes rather. So, now, we will do the same thing that this is the material where chi 2 is equal to 0, but chi 3 is not equal to 0. I should write it here, I should write it here chi 2 is equal to 0, because I am dealing with some material, which is centrosymmetric in nature then chi 3 is a dominating term.

If I launch something here something will come out from this element, which is non-linear in nature in terms of chi 3 where chi 3 is not equal to 0. So, the question is what we are launching if it is a electric field. So, what kind of phenomena it will be there in the presence of chi 3 and what we will have in the output that is our goal. So, our P is now as I mentioned is modified this term will not be there anymore this is a scalar form of P.

So, P non-linear here is related to chi 3 and it is now proportional to Eq. So, P non-linear is now proportional to E cube, if I write in component form I should write pi ith component of the non-linear polarization is kappa ijkl ejekel is the component form standard component form we used in many cases. So, if we start with this expression, which is a scalar expression. Then we can find out what should be the value of PNL, and how different kind of frequency can mix because of this E cube term.

So now, let us take our total electric field is this is a pill this is a standard plane wave form half E 0 E to the power i kz minus omega t plus complex conjugate of that quantity.

So that means I am launching an electric field having a frequency component omega, because here we have omega components sitting.

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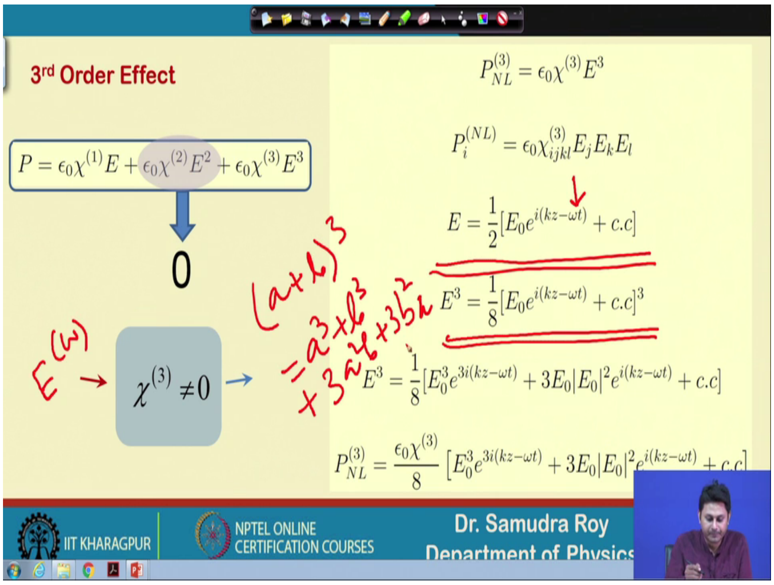

So, an electric field with frequency component omega which is a plane wave I am launching this here. In this particular system, what should be the value of a E cube. So, E cube is a cube of this quantity so when I make a cube of this quantity we will have 1 by 8 E E 0 E to the power ikz minus omega t plus complex conjugate whole cube of that thing.

When I have a whole cube of that thing then the next thing is that we can expand this in form of a plus, it will be expanded like a plus b whole cube. So, I will have a cube term, I will have b cube term and I will have a 3 a square b plus 3 b b square a this kind of terms it will be there. So, now, if I use this E cube and I will get these things as I mentioned.

So, if I write this in the proper way so the first term 1 by 8 will be there the first term is a cube of these things. So, when I make cube it will be E 0 cube E to the power of 3 i kz minus omega t, because I am making a cube of that. And also the complex conjugate will also be cube because there is a b cube term. So, I will enter this complex inside this complex conjugate I can write that there should be 1 term which is a complex conjugate of that first term. Also there will be a couple term we called it 3 E square b. So, this 3 a square b term 3 is here E square is this square of that and then complex conjugate complex, conjugate and this square term can be written in this particular form.

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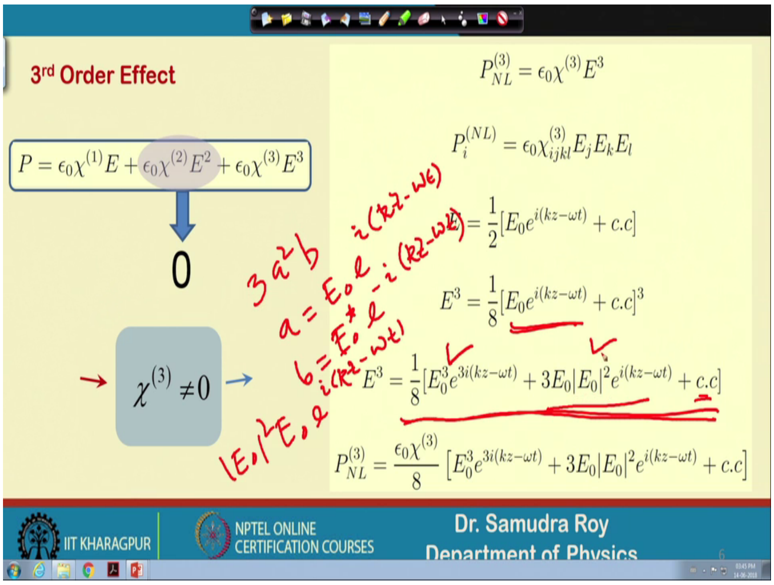

So, if I write a square 3 a square b this is my term. If my a is E 0 E to the power of i kz minus omega t and if my b is E 0 star E to the power of minus i kz minus omega t. Then if I multiply this to making square and b then I will have 1 term, which is mod of E square because complex conjugate is multiplied with this and another term I will have simply E 0 E to the power of i kz minus omega t.

So, exactly the same thing is done here, so I will get this term. So, please note that 1 term I will have a frequency component containing 3 omega and another term I have a frequency component that is having omega. So, two different frequency components are coming when I make E cube term. So, this E cube now I will put here in this expression, because E cube is known I now put this here. Once I put this here, I will have the same expression just multiplied by epsilon chi 3. So, my P non-linear which is important P non-linear 3 this 3 means I am talking about the third order effect only chi 3 term is involved.

It is epsilon 0 chi 3 divided by 8 multiplied by E 0 E to the power of 3 i kz minus omega 3, then 3 E 0 mod of E 0 square E to the power i kz minus omega t plus complex conjugate. So, this entire system entire thing now having two different frequency components.

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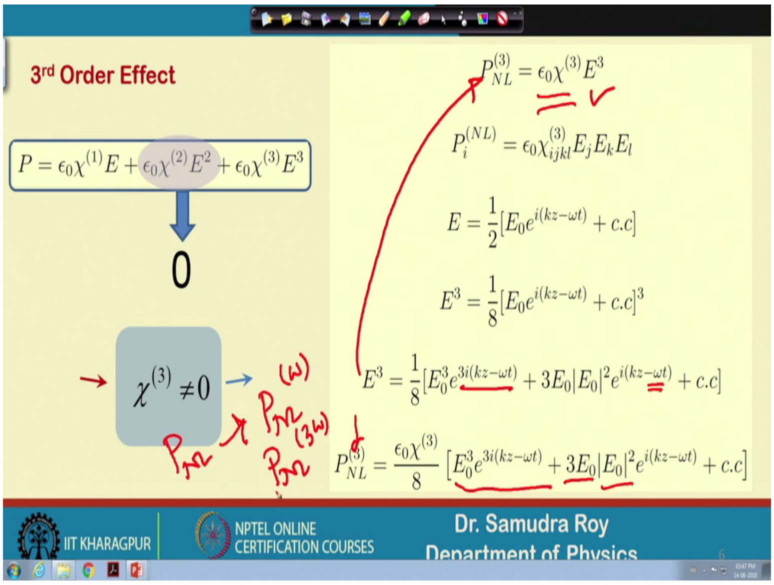

So now, the polarization here nonlinear polarization here; that means, the dipoles inside this system will going to vibrate two different frequencies one is P non-linear omega and another is P non-linear 3 omega. So, 2 components are there inside the P non-linear having two different frequency components.

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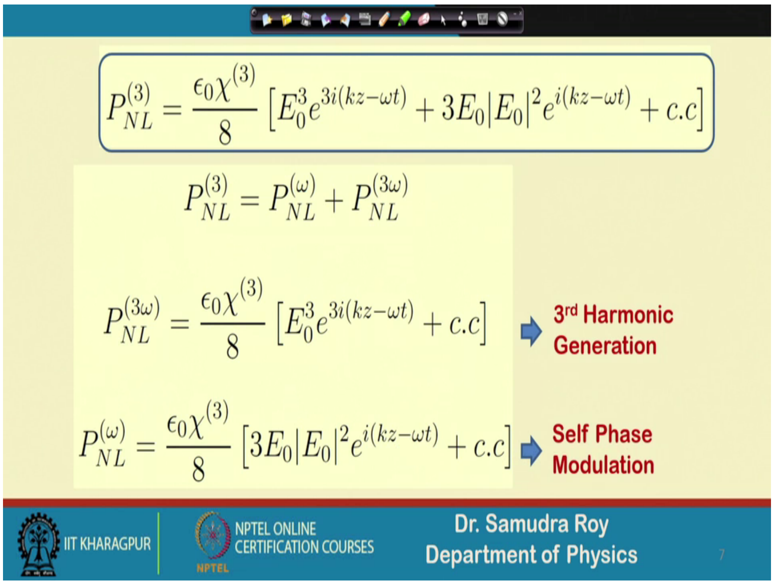

So, two different frequency components; that means two different kinds of vibrations; so let us see here, so P non-linear we already have here. So, P non-linear is epsilon 0 chi 3 8 multiplied by E 0 E to the power 3 kz minus omega t 3 this quantity that we have already shown. Now I write this P non-linear into 2 part P non-linear omega and P non-linear 3 omega. Writing omega means I am talking about this component writing 3 omega means I am talking about this component.

So, these two components basically now separate out so I can write P non-linear 3 omega is now this things. P non-linear omega is now these quantities this amplitude is important that is why I am writing several time this. This quantity epsilon 0 chi 3 divide by 8 will be same for both the cases, but this amplitude is now slightly different for 2 cases, but the most interesting thing is that it will vibrate.

So, to this polarization is now going to vibrate with the frequency 3 omega and another polarization component will be there, which will be vibrating with the fundamental frequency that it should have and that is omega. So, the polarization component that is now vibrating with the frequency 3 omega will going to generate third harmonic. That we can say because in the second harmonic generation case exactly the similar phenomena, phenomena similar phenomena was there.

And in that case what happened that it was proportional to polarization non-linear polarization was proportional to E square term. And as a result we will we have we got a frequency component 2 omega. So, now, here we are having a frequency component 3 omega, because the electric field will be generated by the polarization term that is vibrating inside the system the dipoles will be vibrating in this frequency. If this frequency is now 3 omega, so it will go into generate electric field of 3 omega. On the other hand another component is still sitting here which is very important, which will going to vibrate with the same fundamental frequency omega. So, it will basically generated electric field of frequency omega.

So, it basically leads to another very, very important process and other very, very important phenomena which is called self-phase modulation. We will discuss third harmonic generation and self-phase modulation which are the 2 key important phenomena in third order effect we will discuss in detail in this course.

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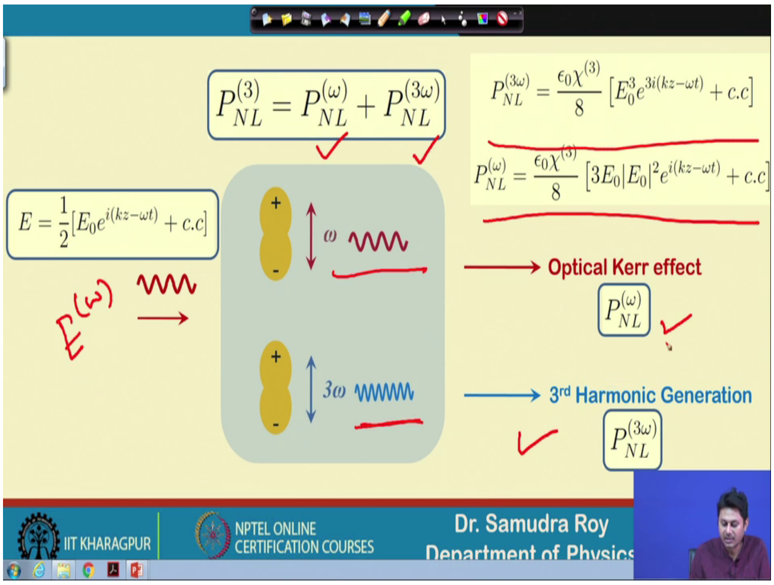

Well, let us try to understand once again as I mentioned in the previous slide. We have 2 polarization components, when we write the polarization in terms of frequency. So, this polarization should have 2 components 1 is P non-linear omega and P non-linear 3 omega these are the 2 frequency components of the polarization, I wrote this components here P 3 omega is this quantity P omega is this quantity. So, electric field having a frequency component omega is launched.

So, I have here in the input E omega something like this. So dipoles are now vibrating at two different frequencies, because there are two polarizations are involved. The polarization having frequency omega will leads to a vibration of dipole. And this dipole will now going to vibrate with the frequency omega, which is the fundamental frequency. And dipole can also vibrate because of the non-linear polarization term containing 3 omega frequency and it will give rise to a field of 3 omega frequency.

So, this is third harmonic generation which is responsible for this free non-linear polarization. And this is called optical Kerr effect which is responsible for the polarization which is now vibrating as a fundamental frequency omega?

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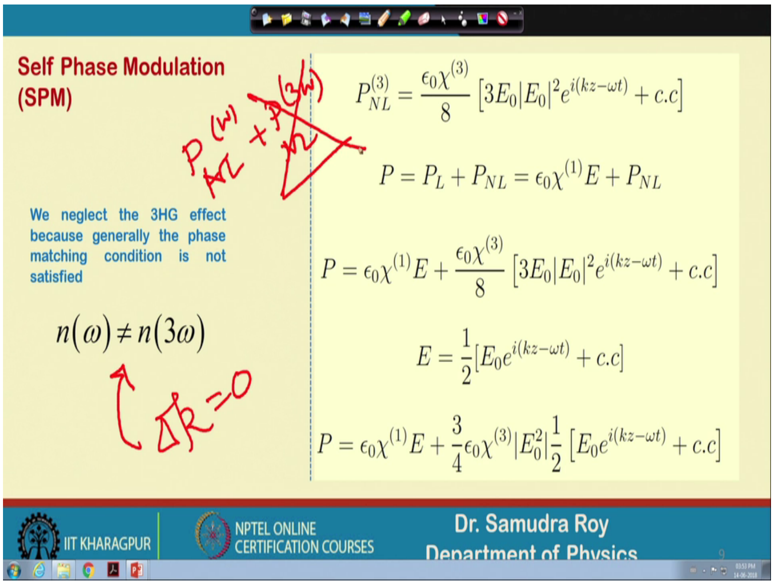

So, now let us try to understand what is self-phase modulation rather Kerr effect. So, Kerr effect is something which will be just try to understand here. So, in the polarization we have 2 term in our hand one is P linear P non-linear omega, and another is P non-linear 3 omega. So, we know that this 3 omega term will leads to third harmonic generation which we will discuss in detail by the way. But normally in order to excite a third harmonic wave, we need a phase matching condition our old phase matching condition delta k equal to 0.

In this case the delta k equal to 0 condition can leads to in terms of frequency it leads to an expression n omega is equal to n of 3 omega, but normally we know that due to dispersion n omega can never been n of 3 omega. So, we will not going to get third harmonic normally. So, it is very difficult to get the third harmonic.

We need to need to have a proper kind of crystal where we can make this kind of arrangement. So, that the dispersion profile is matching properly and we will get a third harmonic effect, but normally we will not going to get this. So, in this P non-linear term we will not bother about for the timing this term. This term will not be there in the P non-linear, which containing a frequency 3 omega.

Because this 3 omega frequency can leads to third harmonic, and we will not going to discuss the third harmonic right now, because the phase matching is normally satisfied here. So, what is the term then we have in the P non-linear. The term that we have is the frequency having the component omega component that means, the fundamental frequency we will have in this P non-linear term. So, P non-linear is now this term only, so in if I write my total polarization only containing this omega term in P non-linear then we will have epsilon 0 chi 1 E this is the linear term plus P non-linear.

If I now explicitly write these things then this is the first term and this is the second term that will appear because of this third order effect. So, you can see that the coefficient of third order effect is sitting here. Now if the electric field is represented in terms of half epsilon 0 E to the power i kz minus omega t and plus complex conjugate.

So, here I can have a epsilon 0 E to the power i kz omega t this terms are there with the complex conjugate is also there. So now, if I write these things if I take this 3 outside and write 3 by 4 and put this half inside, then I will have 1 term half of E 0 E to the power this quantity plus complex conjugate, by taking E 0 outside to this bracket. So, if I do that the next term the third the second term can be written in this particular form, P is equal to epsilon 0 chi E. This second term if I modify slightly it will be 3 by 4 epsilon 0 chi 3 mode of E 0 square then half of this quantity.

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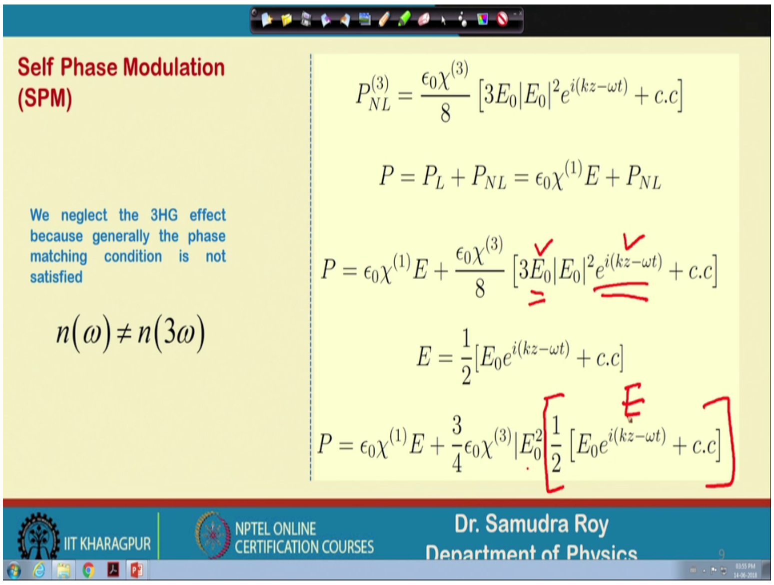

I write it separately to show that this is nothing, but my total E. Mind it total E is a total electric field, this total electric field. Now have a amplitude term and the phase term. I just write amplitude and phase and complex conjugate together with a half then this term become E.

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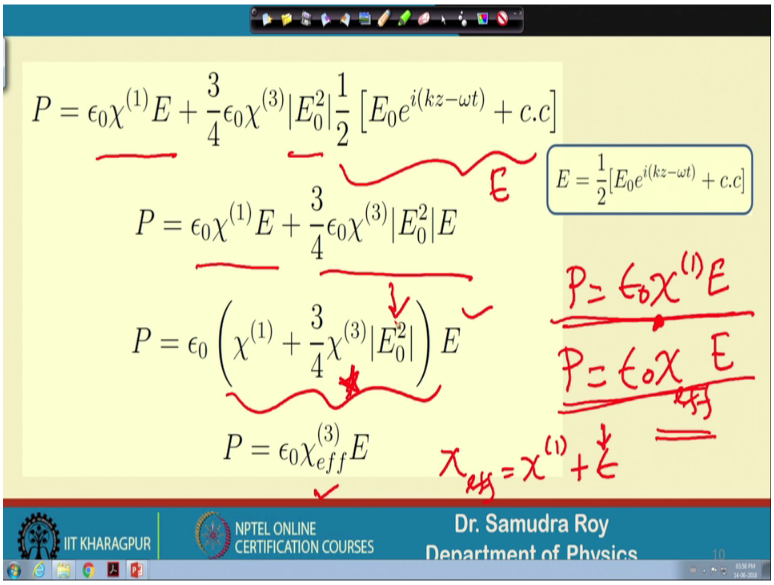

When this term becomes E, then I can now write my polarization in a different way. So, polarization in now epsilon 0 chi 1 E 3 by 4 epsilon 0 this mod of E square have these things which we have E is now half of epsilon E 0 E to the power ikz omega t plus cc i just replace these to key this term to E. So, I will have epsilon 0 chi 1 and 3 by 4 mod of E 0 E. So, now, if I take E common, then I will write this and inside I will have chi 1 plus an additional term here.

So, I write this chi 1 plus 3 chi 3 mod of E 0 as another term. So these two term together I write chi effective. So, now, here we have so normally we have polarization is equal to epsilon 0 chi 1 E, but after doing this process now we have introducing this non-linear polarization I have chi effective E. The expression looks pretty much same, both the cases it is proportional to the electric field, but here inside this expression just we need to replace this chi 1 to chi effective. Where effective is related to chi 1 as chi effective is equal to chi 1 plus something plus I write it as epsilon.

So, this epsilon is now related to the third order non-linear effect. So, the final thing is now I write polarization exactly the way I have written in the polarization the linear form. I can also write in a linear form, but this is looks linear, but this is not a linear form because in chi effective we have mod of E 0 square term that is important.

So, now, this chi effective can leads to one very important phenomena which is called the Kerr effect, because, chi effective is something which can change the refractive index because refractive index is related to that. So obviously, the chi effective is now going to change with the refractive index and this refractive index will be changed due to the presence of the third order nonlinearity. So, in the next class we will start from here and try to find out what will be the effect of chi 3 on refractive index. And because of the change of refractive index which is now a function of intensity.

Because I am launching some light from outside and it will going to mix up with the system and change the corresponding refractive index. So that means, the refractive index is now function of intensity. So, when the refractive index is functional intensity some fascinating things will come out. So, we will discuss in the next class. With this note let me conclude here.

So, we will start in the next class the Kerr effect. So we just started what is the meaning of Kerr effect, but the detail calculation we will do in the next class .With this note let me conclude here, and see you in the next class.

Thank you for your attention.