

Introduction to Non-Linear Optics and its Applications
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Lecture – 27
SHG in LiNbO₃

So, welcome student, to the next class of Introduction to Non-Linear Optics and its Application. So, in the previous class, we studied that how one can generate second harmonic in the crystal like KDP crystal. So, one important thing there we find is that the launching angle of k . So, k is in such a direction that it matches that the phase matching is there, but also we need to find out a suitable azimuthal angle.

So, this suitable azimuthal angle eventually increase the d values and that is why instead of writing d we find d effect and also we find out that these d values in a d matrix there are several d values are there, but finally, when second harmonics are generated only one or two d values are there which are basically responsible for that. So, not all d value are used only one or two d values may be used to generate the second harmonic.

So, in the previous case we find that for generating second harmonic the orientation of k is important and also the direction of P_z is important. So, P_z is in z direction, but that will not going to give the second harm desired second harmonic because our condition is two ordinary wave will generate extraordinary wave. So, z direction was not the direction of extraordinary wave. So, we need to divide this component in such a way the p_z component such a way one direction of the P_z component can generate the extraordinary wave which is at 2ω frequency then only we can generate the second harmonic properly.

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Topics

Nonlinear Optics

SHG in LiNbO₃ Crystal

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So, we will extend this concept. So, let us see what we have in today's lecture. So, today we will going to learn the second harmonic, but not in KDP crystal in a different crystal it is called the lithium niobate crystal. So, this crystal has different properties different d matrix. So, obviously, the condition of phase matching will going to change and also the launching angle or other issues will be there. So, how do will we take care of that issues. So, we will going to discuss in this particular lecture, ok.

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SHG in LiNbO₃ Crystal

$$d = \begin{pmatrix} 0 & 0 & 0 & 0 & d_{31} & -d_{22} \\ -d_{22} & d_{22} & 0 & d_{31} & 0 & 0 \\ d_{31} & d_{31} & d_{33} & 0 & 0 & 0 \end{pmatrix}$$

$d_{22} = 2.10 \text{ pm/V}$
 $d_{31} = -4.35 \text{ pm/V}$
 $d_{33} = 27.20 \text{ pm/V}$

$d_{33} \sim 30 \text{ pm/V}$

Non-critical phase-matching

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So, go back to our slides. So, this is the d matrix structure of lithium niobate. So, we can see that only three distinct elements are there in the d matrix. So, in previous lecture from the previous lecture we already find that this d matrix is start with 27 independent components and then by applying the symmetry it can reduce as to 10 components. And then further if I introduce some symmetry like Kleinman symmetry or using this Neumann's principle, where we can put some kind of operation over this d matrix and as a result we will find that few of the d matrix components are 0 and there are few nonzero elements and this non-zero elements are important.

Here we find there are few d matrix few elements of the d matrix, three elements are there three different elements are there, but most importantly this elements we should look d_{33} this is the element which has the maximum value. So, this is really. So, what is the value of d_{33} . So, d_{33} is of the order of 30 picometer per volt which is really a large value compared to the other values. This is almost ten times bigger than the d_{22} and more than ten times rather and 1.5 times or so, bigger than d_{31} . So, obviously, this two things five times bigger than this rather. So, this 2 if I compare this components we can find that d_{33} really having a large amount of d values.

So, we know that the efficiency is directly proportional to this d d components or this d values which are involved in the second harmonic. So, from the beginning our aim should be how to make our system. So, that d_{33} should involve in generating second harmonic. So, that is our aim that how that second harmonic will be involving through d_{33} that we need to think of.

Well, also one thing I should mention here, that this particular structure in noncritical phase matching system the k vector is almost in the direction of x vector why I am writing or showing this figure is important because for lithium niobate system the refractive index is temperature dependent. So, ordinary and extraordinary refractive index is depending on temperature.

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SHG in LiNbO₃ Crystal

$$d = \begin{pmatrix} 0 & 0 & 0 & 0 & d_{31} & -d_{22} \\ -d_{22} & d_{22} & 0 & d_{31} & 0 & 0 \\ d_{31} & d_{31} & d_{33} & 0 & 0 & 0 \end{pmatrix}$$

$d_{22} = 2.10 \text{ pm/V}$
 $d_{31} = -4.35 \text{ pm/V}$
 $d_{33} = 27.20 \text{ pm/V}$

n_o(ω) = n_e(2ω)

Non-critical phase-matching

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So, by varying temperature there is a possibility that we can make the condition we can make really make the condition $n_o(\omega) = n_e(2\omega)$, but not only theta involvement. So, that means, this angle should be $\pi/2$ and that one can achieve in lithium niobate system, ok.

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SHG in LiNbO₃ Crystal

n_e(λ) = 0.6
n_o(λ) = 1.2

n_o(ω) = n_e(2ω)

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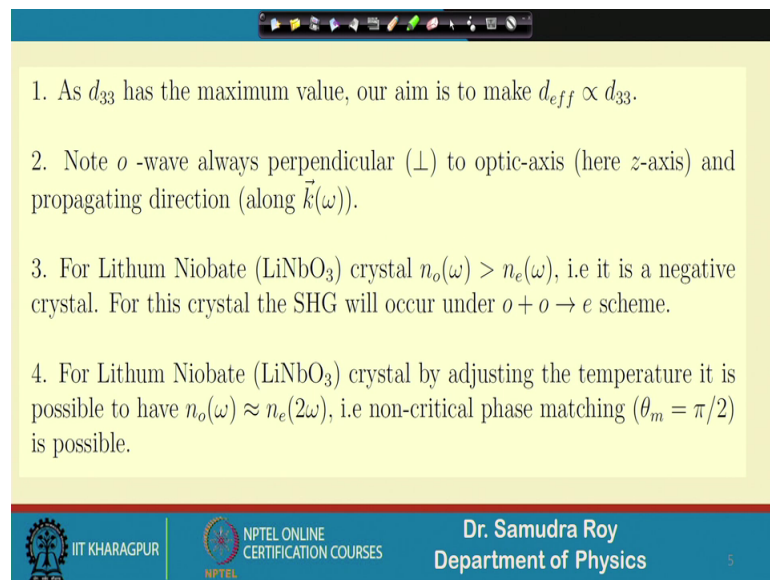
So, let us see what we have in the next slide. So, in the next slide basically we plot refractive index for this lithium niobate structure and this refractive index plot suggest that at particular wavelength say if I look very carefully here we have some wavelength

say around 1.2 or something and here we have this is the value of n lambda. So, this is n of ω which or n let me write once again. So, this say n of lambda equal to say around 1.2 and if I go back here in this point so, this value is the same value of whatever the value we have here. This is n , this is ordinary so, I should write o . So, this is n_e because this is over extraordinary line, n of e of lambda equal to say around 0.6 which is half of that.

So, that means, if I write in terms of frequency n_o at ω is equal to n_e at 2ω which is which is a condition that one can achieve when the phase matching is critical in nature; that means, k vector is moving in x direction, where this two points are coinciding. So, this two points are coinciding so, there is no θ dependency here; so, that that is the direction where we should launch our light.

So, if I do that then this noncritical phase match has some facilities. Since is a noncritical k vector and S vector both will follow the same. So, S vector is also going to the same direction. So, these are additional advantages that we have. So, if we have this kind of phase matching issues then we need to find out few other things.

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1. As d_{33} has the maximum value, our aim is to make $d_{eff} \propto d_{33}$.

2. Note o -wave always perpendicular (\perp) to optic-axis (here z -axis) and propagating direction (along $\vec{k}(\omega)$).

3. For Lithium Niobate (LiNbO_3) crystal $n_o(\omega) > n_e(\omega)$, i.e it is a negative crystal. For this crystal the SHG will occur under $o + o \rightarrow e$ scheme.

4. For Lithium Niobate (LiNbO_3) crystal by adjusting the temperature it is possible to have $n_o(\omega) \approx n_e(2\omega)$, i.e non-critical phase matching ($\theta_m = \pi/2$) is possible.

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So, here we whatever we have discussed it is written in a compact form in a note form. So, what we have let me read it once again that d_{33} has a maximum value. So, our aim should be $d_{\text{effective}}$ proportional to d_{33} as I mentioned that the second harmonic should be in such a way that it should be proportional to d_{33} .

Now, o-wave always perpendicular to the optic axis and the propagation direction vector \vec{k} that you should always know that whenever we calculate the second harmonic and if it is o o plus e system then we should be careful that the fundamental wave should be ordinary wave and the generated wave that is 2ω wave, the generated wave which is having the frequency 2ω should be extraordinary. So, that that thing we should remember and we should do these things very carefully that how this waves are generated.

For lithium niobate crystal n_o is greater than n_e as I mentioned and it is a negative kind of crystal and since it is a negative kind of crystal the phase matching condition that one should have is o o plus o o tends to e system. So, this is a scheme through which the second harmonics will generate. And, as I mentioned for lithium niobate crystal by adjusting the temperature there is a possibility that we can have $n_o(\omega) = n_{2\omega}$ that we have already shown and that means, my phase matching angle is around $\pi/2$; that means, \vec{k} is making $\pi/2$ or \vec{k} is perpendicular to the optic axis and that is the direction along which we have phase matching.

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Case - I

Diagram showing wave propagation along the x -axis (\vec{k}) in a crystal with the optic axis along the z -axis. The fundamental wave $\vec{E}^{(\omega)}$ is along the z -axis. The generated wave $\vec{P}^{(2\omega)}$ is also along the z -axis. Handwritten notes indicate $E_x^{(\omega)} = 0$, $E_y^{(\omega)} = 0$, and $E_z^{(\omega)} = E_1$. The generated wave is identified as an Ex-O wave.

$$\vec{P}^{(2\omega)} = 2\epsilon_0 \begin{pmatrix} 0 & 0 & 0 & 0 & d_{31} & -d_{22} \\ -d_{22} & d_{22} & 0 & d_{31} & 0 & 0 \\ d_{31} & d_{31} & d_{33} & 0 & 0 & 0 \end{pmatrix}_{3 \times 6} \begin{pmatrix} 0 \\ 0 \\ E_1^2 \\ 0 \\ 0 \\ 0 \end{pmatrix}_{6 \times 1}$$

Note: The polarization component $\tilde{P}_z^{(2\omega)}$ will generate a EM wave having frequency component 2ω with polarization along z direction (along optic axis). That means the generated wave is actually an Ex-O wave. The generated 2ω wave and the fundamental ω wave both are Ex-O waves. Hence the phase matching is not consistent here as, $n_e(\omega) \neq n_e(2\omega)$.

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So, next we will try to understand the second harmonic generation in two different cases like we have done for KDP crystal. Here the two different cases are; in first case the electric field is now z direction. If the electric field around z direction it is perpendicular to the \vec{k} , because \vec{k} is along x direction. So, the this one possibility because \vec{k} has to be in

x direction, the phase matching is in this direction only. So, electric field that we can launch this is a trivial case that I launch in z direction.

Now, if the electric field is launch along z direction. So, readily you can understand that this fundamental electric field is not a ordinary wave this is an extraordinary wave, but anyway if this is the case let me figure out what should be the value of P_x , P_y , P_z but from the beginning we know that this wave is extraordinary wave, but still we need to figure out what is going on here to understand the system.

So, E_x is 0, E_y is 0 and E_z is 1, this is our this is our field the field components x, y, z components of the electric field. Now, if this electric field components are this and d matrix which is already given in the previous slide so, P_x , P_y , P_z for second harmonic we can write this in the matrix form. And if I write this in a matrix form only the y component will going to survive, all the other component will be vanished because E_x and E_z are there sorry only z component will be survived the other two will going to vanish.

Now, here unlike the KDP crystal we can see there are many nonzero d elements are there. In that case only three here only three terms were there which was nonzero. Most of the cases with 0 coefficients were there. So, that is why most of the terms was 0, but here there is a possibility that we have some kind of nonzero term and indeed we find that P_x is equal to 0, P_y is equal to 0 and P_z that is generating which is not 0.

And also proportional to d_{33} that was our initial aim that my polarization at second harmonic or polarization at 2ω should be involved with d_{33} coefficient. So, d_{33} coefficient should be involved in generating the second harmonic and here that is the case. So, that means, my polarization would generate a second harmonic along this direction, propagation will generate the second harmonic along this direction because P_z is along this direction. So, it will going to generate a 2ω along this direction.

Now, the important question that we have already mentioned before starting this discussion that what is the what kind of wave $E_{2\omega}$ is? $E_{2\omega}$ is perpendicular to k and it is along the z direction, it is along the z direction. If it is perpendicular to the k and if it is along z direction so, z and k are in the same plane. So, what should be the direction of ordinary wave? Ordinary wave is along y direction that we first to need to

figure out, this is the direction of ordinary wave. Ordinary wave is along y direction, that is for sure.

If ordinary wave is along y direction, k is along z direction, what should be the direction of extraordinary wave? We know that extraordinary wave is perpendicular to ordinary wave and k that is the rule. So, in this particular case extraordinary wave will be along the z direction. So, that means, $E \propto \omega$ which is along z direction which is our second harmonic field that is generated by $P \propto \omega^2$, the z component of the polarization is eventually an extraordinary wave. So, I should put the E here. Now, we can easily find that the fundamental wave which is generating this extraordinary and the second harmonic that is generating is also an extraordinary wave.

If two waves are extraordinary the phase matching condition. So, we will never going to have two extraordinary wave to merge an extraordinary wave. This is a frequency ω , this is a frequency ω , this is a frequency of 2ω , then my phase matching condition would be n of extraordinary of ω will be n of extraordinary of 2ω which never possible. So, that means, the phase matching angle we make in this way, but there is a possibility that I can generate e along z direction I can launch e on z direction also P_z is not equal to 0; that means, P_z is still generating. P_z is generating means it will going to generate a second harmonic wave along z direction.

So, second harmonic wave can also generate along z direction there is a possibility, but that will going to contradict the fact that my phase matching things where n of ω should be equal to n of 2ω which basically not possible.

So, even though all the terms are nonzero we will not going to find any kind of second harmonic. So, that is the very important example even though it looks like trivial from the beginning, but we should understand the fact that if the term is nonzero that does not mean every time it will going to generate second harmonic. So, there are few condition that should follow and this condition when followed strictly then only the second harmonic generation is possible under this birefringence phase matching. So, that means, second harmonic generation is a very critical phenomena and you have to be very careful in generating the waves and second harmonic and you should know the physics behind that and every time you should keep that in mind.

So, in the note we mention that the polarization component P_z will generate electromagnetic wave having the frequency component 2ω with polarization along y direction along optic axis; that means, the generated wave is actually an electromagnetic extraordinary wave as we mentioned. The generated 2ω wave and the fundamental wave ω wave both are extraordinary and that will be the reason why the phase matching should not be. So, there should be 2ω here, it is again a typing mistake. So, n of ω will be never equal to n of 2ω . So, that means, this orientation of e is not a valid orientation. So, we need to do something new.

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Case-II

Diagram showing the coordinate system and the orientation of the electric field \vec{E} and wave vector \vec{k} .

Electric field components:

$$E_x^{(\omega)} = 0$$

$$E_y^{(\omega)} = E_1 \sin \psi = E_o^{(\omega)}$$

$$E_z^{(\omega)} = E_1 \cos \psi = E_e^{(\omega)}$$

Polarization vector $\vec{P}^{(2\omega)}$ components:

$$\vec{P}^{(2\omega)} = 2\epsilon_0 \begin{pmatrix} 0 & 0 & 0 & 0 & d_{31} & -d_{22} \\ -d_{22} & d_{22} & 0 & d_{31} & 0 & 0 \\ d_{31} & d_{31} & d_{33} & 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} 0 \\ E_1^2 \sin^2 \theta \\ E_1^2 \cos^2 \theta \\ 2E_1^2 \sin \theta \cos \theta \\ 0 \\ 0 \end{pmatrix}$$

Handwritten notes:

- $\vec{P}_x^{(2\omega)} = 0$
- $\vec{P}_y^{(2\omega)} = 2\epsilon_0 d_{22} E_1^2 \sin^2 \theta |_{(o+o)} + 4\epsilon_0 d_{15} E_1^2 \sin \theta \cos \theta |_{(o+e)}$
- $\vec{P}_z^{(2\omega)} = 2\epsilon_0 d_{31} E_1^2 \sin^2 \theta |_{(o+o)} + d_{33} E_1^2 \cos^2 \theta |_{(e+e)}$
- Phase matching: $o+o \rightarrow e$, $e+e \rightarrow e$

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So, now what we were doing that we are launching the electric field making an angle ψ with the z axis. So, previously E was in z direction now E is in arbitrary direction with an angle ψ . Thus figure is shown here E is k is along this direction this is the direction along which the phase matching is there, but the orientation of E basically is important. Even though the phase matching is there P_z is not equal to 0, still we find that there is no electric field second harmonic generation. So, here we need to find out what is going on in terms of this E . So, E is launched with an angle ψ .

So, that means, we can figure out what are the components. So, first thing we need to find out what are the components. So, E_x should be 0, because E is eventually in y - z field or y - z plane, E_y is the component along the y direction. So, it should be $E_1 \sin \psi$ and z is $E_1 \cos \psi$. Now, you need to be careful enough to write these component

in terms of ordinary and extraordinary because along y direction we mention it should be a ordinary wave. So, I write this $E \sin \phi$ is a component that is generating ordinary wave and the other component which is along z direction should be the extraordinary wave that issue we have already deal with in the we have already discussed in our previous slides, that if the electric field along the z direction the electric field is an extraordinary wave.

So, here that is the z component in extraordinary wave. So, once we have the E_x , E_y , E_z , the next thing is to find out my polarization. Now, you can see that there are many nonzero terms sitting here. So, E_x was 0 E_y was not 0. So, I put this E_z was not 0, 2 of E_x , E_y 2 of E_y is z is not 0, but other two terms is 0 because both the cases it is involved with E_x term.

So, now if I calculate term by term so, P_x turns out to be 0, there is no P_x ; P_y there is a combination of term if I calculate P_y how to calculate these multiplied by this 0, d 22 this which is nonzero, this 0 multiplied by this is a 0, d 31 multiplied by this is again nonzero. So, this term and this term, two terms are there and also we find out what is my P_z . P_z is 3 11 and another is d 33 $E \cos^2 \theta$.

So, there are many terms are there are many terms that is in this in this matrix. So, as a result we have different term in P_x , P_y and P_z . So, P_z is containing two terms, P_y is containing two terms and P_x is containing no terms it is 0. So, now, you should be careful enough to find out that what is the combination. So, in both the cases if I find it is $E \sin^2 \theta$, so, that means, $E \sin \theta$, ok, here is a notational problem actually here if I write it psi. So, it has to be psi, but I have written is theta. So, let us consider this angle is theta. So, this angle should be theta, this angle should be theta.

So, if I consider this thing here that $\sin^2 \theta E \sin E \sin^2 E \sin^2 \theta$ is basically the components of ordinary wave. So, two ordinary waves are merging to generate the polarization along y direction. Also, here we have $E \sin^2 \theta \cos \theta$; that means, here we eventually have one ordinary component of field one extraordinary component of field. So, ordinary plus extraordinary component can generate polarization along y direction and in P_z also two ordinary waves are generating and as a result I am getting some kind of wave a some kind of P_z . Here also in the similar way two extraordinary wave are combined to generate the P_z term.

So, here one by one if I consider so, you can understand that these two ordinary. So, what should be the electric field associated with P_y . P_y will going to generate an electric field of 2ω along this direction. This is electric field along 2ω and the direction is y . So, that means, this is this has to be an ordinary wave. So, two ordinary wave will going to generate something ordinary which is again violating the phase matching. So, this term the contribution of the these term will not be there. But, here we have a situation where one ordinary and one extraordinary wave can generate an ordinary wave of 2ω , this is ω of ordinary wave, this is ω of ordinary wave. It is going to generate a ordinary wave up to ω this is a type II kind of phase matching.

So, type II kind of phase matching is here. What about P_z ? Two ordinary wave P_z will generate an electric field along this direction. This is the direction of extraordinary wave which is generating 2ω . So, once we have electric field 2ω along z direction; that means, this contribution this contribution will going to generate an electric field which is extraordinary in nature. So, what should be the scheme here? The scheme is ordinary plus ordinary which is generating extraordinary. So, this is the frequency component ω , this is the frequency component ω , this is the frequency component 2ω . So, this is also valid. So, this term is also valid.

What about the last term? Last term, again two extraordinary wave will going to generate P_z . P_z will going to generate an extraordinary wave. So, extraordinary two wave will leads to one extraordinary wave of 2ω . So, ω , ω it will going to generate 2 of ω . So, again it is violating. So, this is 2 of ω again this is violating because the condition is not valid.

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$\tilde{P}_x^{(2\omega)} = 0$

$\tilde{P}_y^{(2\omega)} = 2\epsilon_0 d_{22} E_1^2 \sin^2 \theta|_{(o+o)} + 4\epsilon_0 d_{15} E_1^2 \sin \theta \cos \theta|_{(o+e)}$

$\tilde{P}_z^{(2\omega)} = 2\epsilon_0 d_{31} E_1^2 \sin^2 \theta|_{(o+o)} + d_{33} E_1^2 \cos^2 \theta|_{(e+e)}$

Type II Phase matching

Handwritten notes:
 $E_1 \cos \theta = E_0^{(\omega)}$
 $E_1 \sin \theta = E_0^{(\omega)}$

Note: $\tilde{P}_y^{(2\omega)}$ leads to the generation of a SH field $E_o^{(2\omega)}$ which is a *o*-wave. Now $E_o^{(2\omega)}$ is generated by combining two *o*-wave of frequency ω (1st term) as well combining one *o*-wave and one *e*-wave (2nd term).

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So, once we know these components carefully then we can readily find out that what is going on. So, now, as I mentioned this term this shaded term is a type II kind of phase matching, where P_y leads to generation of second harmonic along y direction we call it E_{2y} . So, which is a which is a ordinary wave and $E_{2\omega}$ is generated with a combination of one ordinary wave of frequency ω , that is the first term here and another combination of 2ω , which is the second term here. So, first term was E_1 , first term was again this notation was which it should be either it should be θ or it should be ψ . So, there is a notational mismatch, but anyway.

So, this angle is θ ; if this angle is θ so, $E_1 \cos \theta$ is along z direction. So, this is an extraordinary component and $E_1 \sin \theta$ is ordinary wave because it is along y direction. So, both the cases it is ω component. So, these two combination basically one ordinary and one extraordinary wave generate this term and this term basically gives us a type II kind of phase matching.

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Type II Phase matching

$$\vec{k}_o^{(\omega)} + \vec{k}_e^{(\omega)} = \vec{k}_o^{(2\omega)}$$

$$n_o^{(\omega)} + n_e^{(\omega)} = 2n_o^{(2\omega)}$$

Vector Diagram

$o+l \rightarrow o$

$o+l \rightarrow l$

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Well, we are almost at the end of this class. So, type II phase matching if I look carefully what is going on so, in this picture so, in type II kind of phase matching the wave vector k_ω of ordinary wave though vector k_ω of extraordinary wave they will go to merge and generate k_ω of an ordinary wave. So, that is one combination, but you should remember that another combination is still possible. So, we have already learnt that.

So, there are two possibilities ordinary waves plus extraordinary wave can this two and ordinary wave or ordinary wave extraordinary wave can these two one extraordinary wave both the cases the phase matching can be possible and this kind of phase matching is called type II, kind of phase matching which we just learnt in the example that is the second harmonic generation one part of type II phase matching can be possible. And the refractive indexing term of refractive index we have the relation between the refractive index like this is a phase matching condition in terms of refractive index.

If we plot the vector diagram we can see that one ordinary one extraordinary wave, they can merge to generate one ordinary wave and in the figure also we can see that if I launch an electric field in this direction when it pass through the system, if it generate the electric field which is ordinary in nature then it will vibrate along this direction. So, we will start from this electric field lounge electric field, but the second harmonic field will change its polarization state and ordinary wave that will vibrate in y direction, it will be

in y direction. So, this will be a schematic figure to show how the second harmonic will be generating in lithium niobate crystal where type I and type II phase matchings are there together.

So, with this note let me conclude here. So, we have learned a very important in the last two classes we have learned very important problem; one is second harmonic generation in KDP crystal and in second harmonic generation in lithium niobate crystal and in lithium niobate crystal you find the orientation of the k is fixed, but if I change the orientation of E , then there is a possibility to generate type II kind of phase matching.

So, with this note let me conclude here. So, see you in the next class and thank you for your attention.