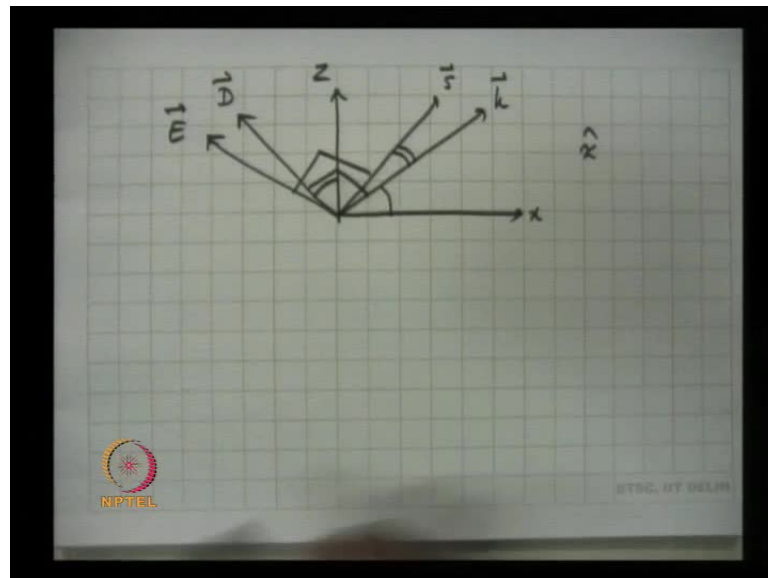


Quantum Electronics
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Module No. # 03
Second Order Effects
Lecture No. # 07
Non - Linear Optics (Contd.)

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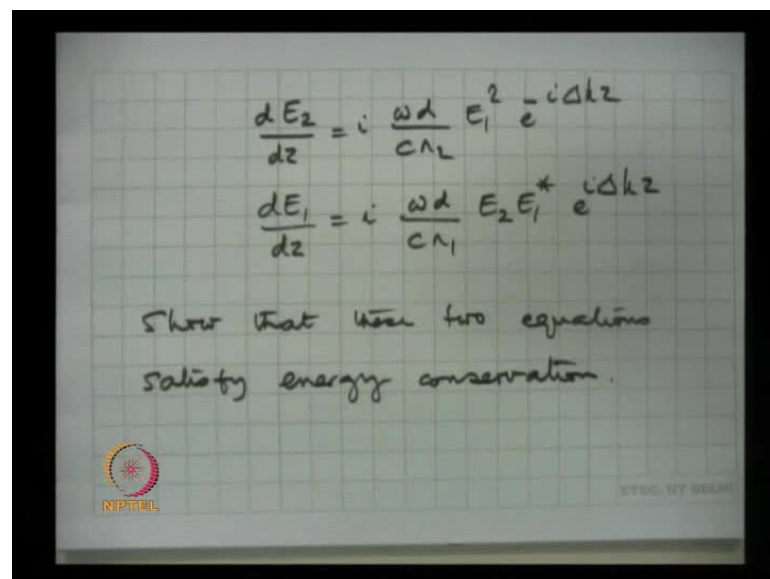
Let me, before we start the lecture, let us look at that the question that we had put last time. Remember, the question was that I have a uniaxial medium and **a wave** around is propagating in some direction; the unit vector to that direction is given. So, remember that the D vector is always perpendicular to the k vector, and if it is an extra ordinary wave, the k vector and the D vector lie in the plane containing k, z and the optic axis, and this D vector. So, this D vector is lying in the plane containing the optic axis and the k vector. And, once I know D, **I can**, this angle, so, this angle is equal to this angle; and once I know D, then I can actually calculate that orientation of the E vector; and once I know the orientation of the E vector, for example, if the E vector happens to be like this,

then the s vector will be like this. So, it is just calculating from the given direction of the k vector, knowing that the D vector lie s in the plane containing optic axis and the k vector, finding out the D vector direction; from the D vector direction, get the E vector direction; and from the E vector direction, get the s vector direction, because this is right angle; E and s are perpendicular, D and k are perpendicular.

So, it is a small angle, up to 2.15 degrees or something like that, the angle between these two. So, this is what I have told you earlier, to check for the angle between the k vector and s vector; this angle is usually very small, a couple of degrees. Do you have any questions?

What is the maximum value of this angle? This is what I asked you to calculate. It is a few degrees, this is **no, no, no, no**... Because, **the** at 0, psi is equal to 0, E and D are parallel; **at psi is equal** to 90, they are again parallel; and, in between, there is a small angle and that angle depends on this n o to any ratio because, remember, the E z by E x was n o square by n e square divided into tan psi. So, the difference, **the** anisotropy is usually not very large, n o and n e are not very far from each other; so, this angular difference is usually very small, it is a couple of degrees; and the number which I gave you in the quiz, that psi was about 60 degrees, and so, the angular difference is about 2 degree or so only; it is a small angle. Anything else? Before we start discussion, **I want**, I will leave a problem to you. Please do this at home.

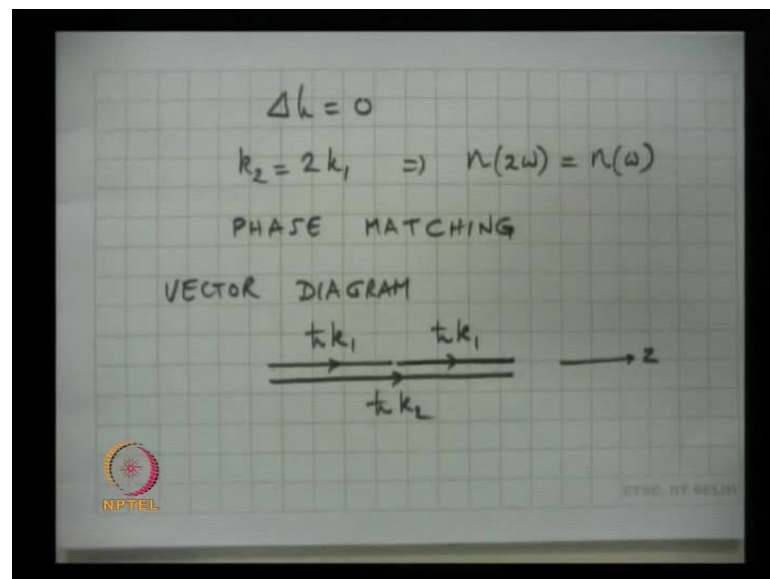
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So, remember these two equations we have written, $\frac{dE_2}{dz}$ is equal to $i\omega d$ by $c n^2 E_1^2$ minus $i\Delta k z$ and $\frac{dE_1}{dz}$ is equal to $i\omega d$ by $c n^1 E_2 E_1^*$ minus $i\Delta k z$. Show that these two equations satisfy energy conservation. E_1 is the electric field of the ω wave, E_2 is the electric field of the 2ω wave, and so E_2 is getting generated from E_1 ; so, there must be energy conservation. So, you need to show between these two equations; this two coupled equation satisfy energy conservation. So, I leave this problem to you.

So, what we have seen is, we have actually solved this first equation under the assumption that E_1 is almost a constant. This is also called a no-pump depletion approximation. The ω frequency, which is the strong wave, which comes into the crystal is called the pump; and, you neglect the depletion of the pump, you neglect the depletion, you neglect the change in the power in the pump for low conversion efficiencies; and, this is usually termed as no-pump depletion approximation; and under this approximation, we solve this equation and we found that, for maximum efficiency, Δk must be 0, which we called as a phase matching condition.

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So, what we found is, for maximum efficiency, Δk is equal to 0; this implies k_2 is equal to $2k_1$, which implies, the refractive index at 2ω must be equal to the refractive index at frequency ω .

The electromagnetic wave at frequency ω and the electromagnetic wave at frequency 2ω must be propagating at the same velocity; but that is not possible normally, because all media are dispersive; so, when you change frequencies, the refractive index will change. So, $n_{2\omega}$ is larger than n_{ω} ; so, how do I satisfy this condition? Also, before we look at this problem, **this is called**, this is called the phase matching and we discussed different interpretations of this.

In the photon picture, as I said, the non-linear medium is helping 2 photons at frequency ω , merge into a single photon at frequency 2ω ; and, phase matching condition is nothing but momentum conservation condition. So, we can draw, what is called as the vector diagram. So, here is the momentum vector of one of the photons, $\hbar \times k_1$ $\hbar \times k_2$, **k, sorry**, another $\hbar \times k_1$ and this is $\hbar \times k_2$. This sum of the two momentas are this length of the vector here, and this must be equal to the length of the $\hbar \times k_2$ vector; and they are all propagating in the same direction, so, **all the vectors are...** So, this is my z axis, z direction.

This is a very nice interpretation in terms of vector diagrams. We will use this vector diagram later, again, in other interaction process; and so, if the sum of these two vectors is different from this length of this vector, then you do not have phase matching, and your efficiency will drop down. So, we will come back to this vector diagram repeatedly when we look at the parametric process also.

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$$\frac{dE_2}{dz} = i \frac{\omega d}{c n_2} E_1^2 e^{-i dkz}$$

$$\frac{dE_1}{dz} = i \frac{\omega d}{c n_1} E_2 E_1^* e^{i dkz}$$

Show that these two equations satisfy energy conservation.

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$$\frac{dE_2}{dz} = i \frac{\omega d}{c n_2} E_1^2 e^{-i\Delta k z}$$

1. $\Delta k = 0$ $k_2 = k_1$
 $n(2\omega) = n(\omega)$
BIREFRINGENCE PHASE MATCHING

2. QUASI PHASE MATCHING (QPM)

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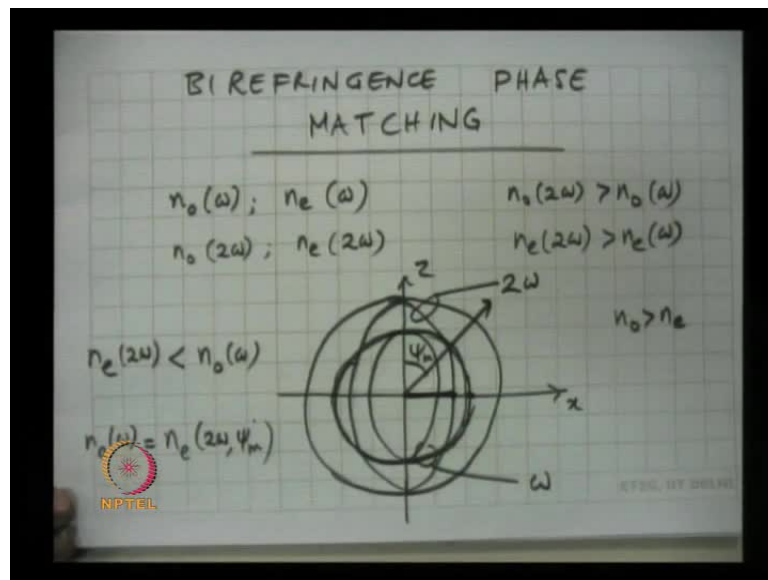
So, this phase matching is an important issue, let me look at this equation - why there is this problem, what is leading to this problem? This problem is coming because of this term - exponential minus $i \Delta k z$, sorry, $i \Delta k z$. So, the equation which we solved, is only this equation for 2ω $i \omega d$ by c into $E_1^2 e^{-i\Delta k z}$; when I integrated this equation, it is this term which is leading to the sinc function. So, **if I make this**, if I make this disappear from this equation, if the equation did not have this term, then I will not have the sinc function, and I will have increase efficiency, that means, the second harmonic will continuously grow as the wave propagates.

So, there are two techniques, there are two primary techniques. In one technique, I made Δk is equal to 0, that means, k_2 is equal to k_1 , which means, refractive index of the medium at 2ω is equal to refractive index of medium at ω . This, I will use the **bi-refrindex** of the crystal, and hence, it is called the Bi-refrindex Phase Matching. Remember, in anisotropic media, there are two refractive indices, there are two waves - the ordinary wave and the extraordinary wave; and the extraordinary wave velocity or the refractive index, as seen by the extraordinary wave, depends on the angular propagation. So, it may be possible in anisotropic media to be able to achieve this condition, by choosing the ω wave to be one polarization and 2ω wave to be another polarization. I will show this; and so, this is called Bi-refrindex Phase Matching, we will discuss this in more detail.

The other technique is introduced as a dependence of this non-linear coefficient d ; d is, in our analysis, is assumed to be constant suppose, I modulate d along the propagation direction periodically; a periodic variation in d will have Fourier components, spatial Fourier components; I can use one of those spatial Fourier components to cancel this term.

Fourier series will have sine and cosine series, sine and cosine or an exponential, is the same; so, I can actually use a periodic variation in the non-linear coefficient to cancel this term, so, **d will...** If I choose a periodic value for periodic function of d , that periodic function will have Fourier components and one of the spatial Fourier components can be used to cancel this effects of this, and that is the second technique, called Quasi Phase Matching or called QPM. This is a very important technique, first proposed by Bloembergen in 1961, and the classic paper on non-linear optics, where he showed that it is possible to overcome this issue of phase mismatch through a periodic variation in the non-linear coefficient.

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So, first what you will do is, we will look at Bi-refringence Phase Matching. So, first we are looking at Bi-refringence Phase Matching. Now, remember, suppose, I take a medium which is anisotropic, so, there are the ordinary refractive index at frequency ω and the extraordinary refractive index at frequency ω ; then, there is also an ordinary refractive index at 2ω and an extraordinary refractive index at 2ω .

So, if I look at second harmonic generation of a 1 micron wave, so, I will have refractive indices **at** of the medium; ordinary and the extraordinary indices at the frequency corresponded to 1 micron wave length, and the ordinary and extraordinary refractive indices corresponding to 0.5 micron wave length.

Now, remember, because of dispersion, n_o of 2ω is greater than n_o of ω and n_e of 2ω is greater than n_e of ω . As you increase the frequency, the refractive index increases. Now, between n_o and n_e at 1 frequency, **depending**, there are kinds of positive and negative uniaxial crystals. So, let me look at a negative uniaxial crystal in which n_o is bigger than n_e . So, let me try to draw a figure; the index surfaces which I introduced in an earlier class; so, let us draw the index surfaces; so, I first draw the index surfaces corresponding to frequency ω . So, I have the ordinary index surface corresponding to the ordinary wave and the extraordinary refractive index surface. So, if n_o is greater than n_e at 1 frequency, the ellipse will lie inside the circle, touching the circle at this; so, this is by z axis, this is x, so, these two are at frequency ω .

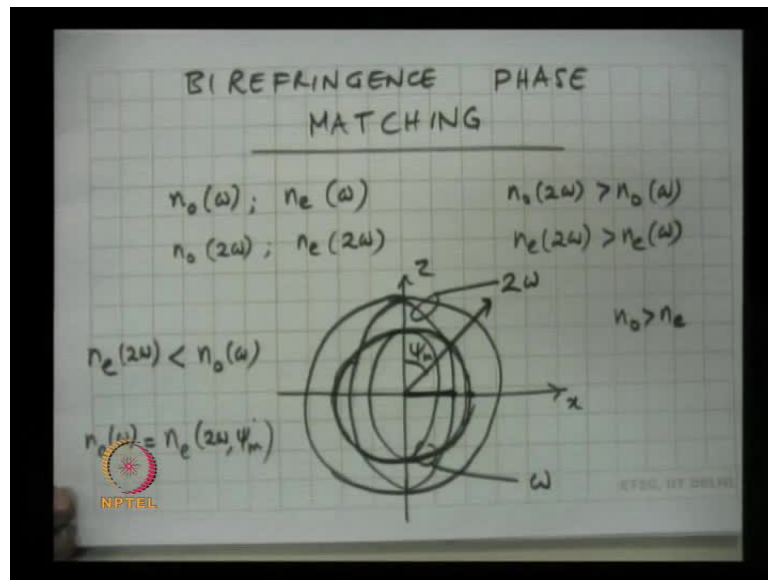
Now, I want to draw in the same figure, the index surfaces corresponding to 2ω frequency. So, because, n_o of 2ω is more than n_o of ω , this circle corresponding to the second harmonic wave, will lie outside the circle. Now, **the ellipse:** the ellipse may intersect this circle, that ellipse cannot intersect this ellipse, because that ellipse has to lie outside this ellipse, because n_e of ω is less than n_e of 2ω . But that ellipse could intersect a circle or may not intersect the circle. So, suppose, I take a situation where the ellipse intersects the circle, so these are for 2ω .

So, under what condition will this intersect take place? Remember, this length is how much? This length is n_e of 2ω , and this length from here to here, the circle is n_o of ω . If this is less than n_o of ω , then the ellipse corresponding to 2ω will intersect the circle corresponding to ω frequency; n_o of the ω is a circle, here; n_e of 2ω is the ellipse; this is n_o of ω , this circle is n_o of ω circle; and this ellipse is n_e of 2ω ellipse. So, this length is the length corresponding to n_e of 2ω ; this length is the length corresponding to n_o of ω ; and, n_e of 2ω should be less than n_o of ω for this ellipse to intersect the circle.

When does the ellipse intersects the circle? Look at this direction. The ordinary refractive index at frequency to ω , sorry, the ordinary index ω becomes equal

to the extraordinary refractive index at 2ω for that direction, some ψ , phase matching, so this some special angle. So, in such a situation, there are directions in which the ordinary refractive index at frequency ω becomes equal to the extraordinary refractive index at 2ω , along that direction.

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As I told you before, please differentiate between n_e of 2ω ψ and n_e of 2ω ; n_e of 2ω is a constant, corresponding to n_e of 2ω 90° , so, n_e of 2ω is the extraordinary refractive index as seen, via wave propagating perpendicular the optic axis. The minimum value of refractive index as seen to the extraordinary wave, for this crystal.

If the extraordinary wave propagates in any other direction, the refractive index seen by the extraordinary wave will be more than, n_e of ω , n_e of 2ω , for this 2ω frequency. So, please differentiate between n_e of 2ω ψ and n_e of 2ω ; this is there is a formula for this refractive index which depends on the angle ψ . So, there are special directions; there will be a special direction, in which, along which the refractive index, as seen by the ordinary wave at frequency 2ω , becomes equal to the refractive index as seen by the extraordinary wave at frequency 2ω ; and that, I can obtain - phase matching - that means.

So, although there is dispersion, I am using the fact that there are ordinary and extraordinary refractive indices; and under this situation, it is possible to achieve what is called as Bi-refrindex Phase Matching because, I am using the bi-refrindex of the crystal to achieve phase matching.

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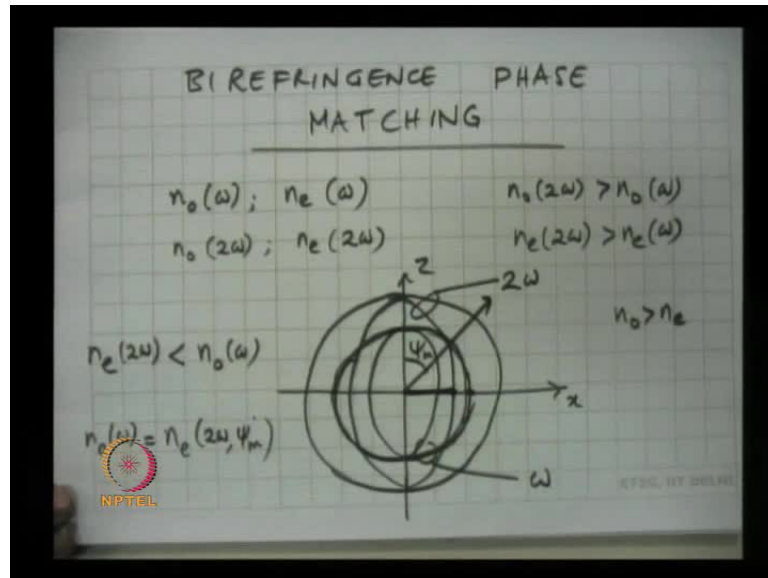
$$\frac{1}{n_e^2(2\omega, \psi_m)} = \frac{\cos^2 \psi_m}{n_o^2(2\omega)} + \frac{\sin^2 \psi_m}{n_e^2(2\omega)} = \frac{1}{n_o^2(\omega)}$$

$$\sin^2 \psi_m = \frac{\left[\frac{1}{n_o^2(\omega)} - \frac{1}{n_o^2(2\omega)} \right]}{\left[\frac{1}{n_e^2(2\omega)} - \frac{1}{n_o^2(2\omega)} \right]}$$

Now, what does this condition imply? Remember, $1/n_e^2(2\omega, \psi_m)$ will be equal to $\cos^2 \psi_m / n_o^2(2\omega) + \sin^2 \psi_m / n_e^2(2\omega)$. Remember, we had obtained the refractive index as seen by the extraordinary wave when it propagates at angle, making an angle ψ with the optic axis. For **phase** matching, this must be equal to $1/n_o^2(\omega)$; because, my phase matching condition is this, and this implies that, $1/n_e^2(2\omega, \psi_m)$ must be equal to $1/n_o^2(\omega)$.

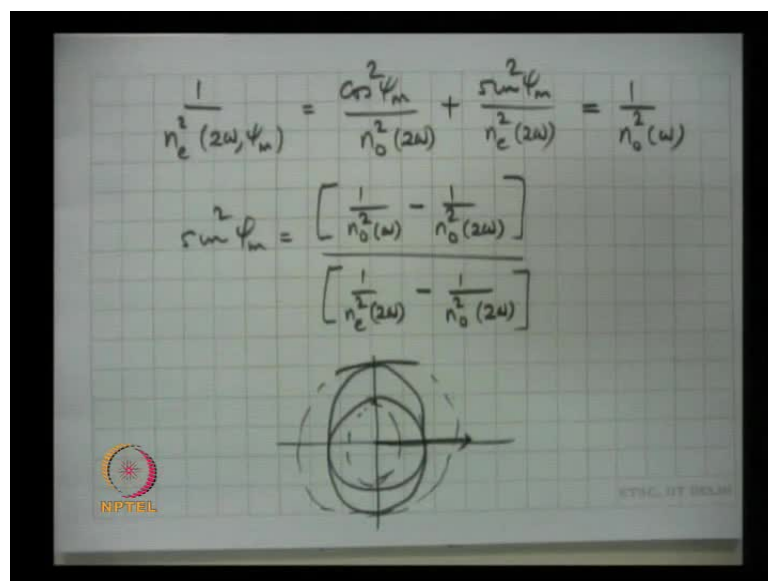
I can simplify this equation and I will get the following equation - $\sin^2 \psi_m$ is equal to $1/n_o^2(\omega) - 1/n_o^2(2\omega)$ divided by $1/n_e^2(2\omega) - 1/n_o^2(2\omega)$. You just write the cosine in terms of sine, simplify this equation; and there is a specific angle, at which, depending on the ordinary and extraordinary refractive indices of the crystal, and depending on the dispersion in the crystal, it is possible to achieve Bi-refrindex Phase Matching.

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If n_e of 2ω , happens to be larger than n_o of ω , then the ellipse will lie outside the circle and there is no Bi-refraction Phase Matching possible. So, Bi-refraction Phase Matching requires anisotropy and certain relationships between dispersion and bi-refraction. Now, please note, that if you deviate from this angle ψ_m slightly, Δk becomes finite; because n_o of ω , although, n_o of ω remains the same, n_e of 2ω will change, as to change in angle.

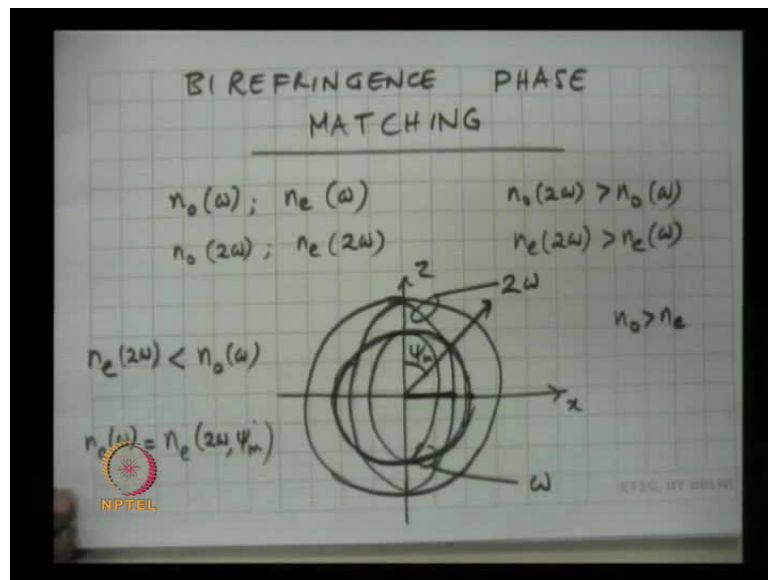
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So, at this point, it is very critical; the angle that you need to propagate the wave in, is very critical, it is called Critical Phase Matching. If this angle happens to be 90 degrees, then the circle, the inner circle, will be tangential to the ellipse. So, it will look like this; so, then, I am just drawing the inner circle, n_o of ω and n_e of 2ω will look like this.

So, I am not drawing the outer circle here and the inner ellipse; so, there is inner ellipse here, corresponding to ω frequency, there is an outer circle corresponding to the 2ω frequency. So, sometimes, it is possible, that this angle, ψ_m become 90 degrees. Sorry, why? No, this coupling is brought about by non-linearity, I will, I will show you.

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Here also, even in this case, the two electric fields are orthogonal; one is ordinary wave and the other is extraordinary wave, so, the polarizations are perpendicular to each other; but there is still coupling, because of the non-linearity. Now, this is a very important issue, I will come to this.

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$$\frac{1}{n_e^2(2\omega, \psi_m)} = \frac{\cos^2 \psi_m}{n_o^2(2\omega)} + \frac{\sin^2 \psi_m}{n_e^2(2\omega)} = \frac{1}{n_o^2(\omega)}$$

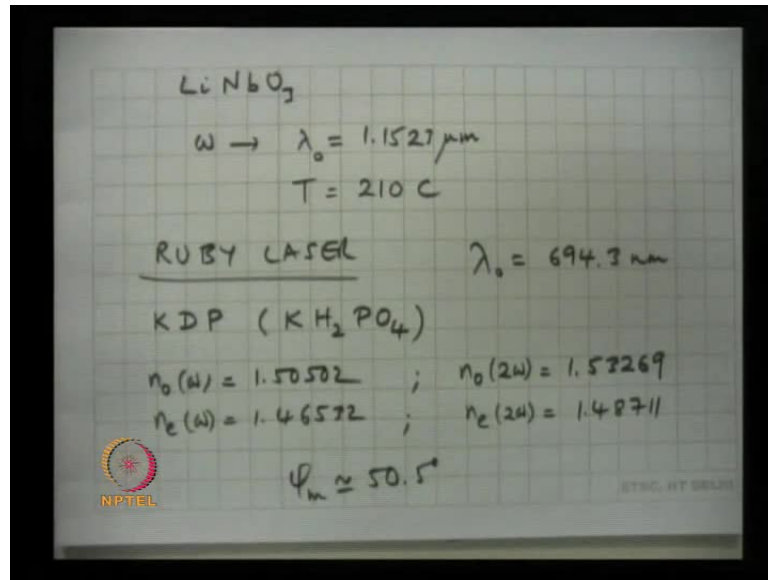
$$\sin^2 \psi_m = \frac{\left[\frac{1}{n_o^2(\omega)} - \frac{1}{n_o^2(2\omega)} \right]}{\left[\frac{1}{n_e^2(2\omega)} - \frac{1}{n_o^2(2\omega)} \right]}$$

$$n_e(2\omega) = n_o(\omega)$$

So, the same situation here, except that, now, it is possible that this angle becomes 90 degrees; in this case, look here, because the two curves are tangential, small deviations are not so critical; even if a small angular mismatch, miss alignment, you will not have a large Δk ; Δk is finite still; but it will not go as fast as at this point, when the two curves are actually intersecting. So, this is called Critical Phase Matching, this is called Non-critical Phase Matching. I am still using bi-refringence, but here, the ω and 2ω waves are both propagating perpendicular to the optic axis; one is an ordinary wave, the other is an extraordinary wave.

So, the ω frequency is an ordinary wave. The 2ω frequency is an extraordinary wave, **but the two refractive indices...** Here, is a situation, where n_e of 2ω becomes equal to n_o of ω . In this case, the phase matching angle is at 90 degree **slope** optic axis, and because of the two curves being tangential to each other, small deviations in angle, will not create a large Δk , and it is called Non-critical Phase Matching. Now, it is possible in some situations, to make this happen, because, remember, these refractive indices are also functions of temperature; so, I can use this equation and find out, if it is possible to vary the temperature, such that, **psi n** becomes 90 degrees.

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So, for example, if you take this, an element called... So, if you take lithium niobate, for example, it so happens, that in lithium niobate, LiNbO_3 , if your ω corresponds to a wavelength of, **lambda is equal to**, λ_0 is equal to 1.1523 micrometers at a temperature of 210 degree Celsius, you can achieve Non-critical Phase Matching. Because, it so happens, that these values are, such that, ψ_m , the solution is 90 degrees; but it is not always possible. So, if you change the wavelength, then you are not able to satisfy this condition with ψ_m and 90 degrees.

So, Bi-refrignce Phase Matching is interesting, because you use bi-refrignce, achieve Phase Matching, but **you could have**, if it is Critical Phase Matching, you have to ensure that your propagation direction is according to this angle. So, let me give you an example; so, the first laser which was built, was what? Ruby laser. What is the wave length? 694.3 **right?** Now, there is a crystal which is very popularly used, KDP, this is KH_2PO_4 , potassium di hydrogen phosphate. It is a uniaxial crystal, it is transparent and visible, and it is used in many second harmonic experiments; you can grow big crystals **of this, of these crystals**, big-size crystals.

So, let me give you the numbers. So, corresponding to this wavelength, n_o of ω is equal to 1.50502, n_e of ω is equal to 1.46532, n_o of 2ω is equal to 1.53269 and n_e of 2ω is equal to 1.48711 Is it possible to achieve Bi-refrignce Phase Matching?

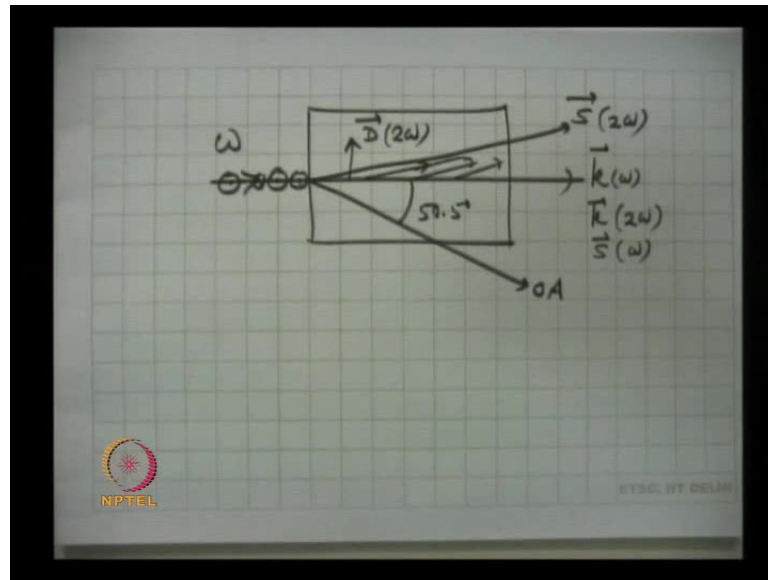
The condition we needed was, any of 2ω must be less than n_o of ω , so, I can achieve Bi-refrignence Phase Matching; I substitute all this values in this equation and get the angle ψ ; so, ψ_m comes to be about 50.5 degrees. So, if I take potassium di hydrogen phosphate crystal, propagate it in an angle of 50.5 degrees with the optic axis and use this laser, I can satisfy the phase matching condition; but please note, in Critical Phase Matching, the s vector of the extraordinary wave is not parallel to the k vector of the extraordinary wave. So, all though the wave front is propagating at 50.5 degrees to the optic axis, the energy of the second harmonic is not propagating at that angle, it is propagating at oblique angle.

So, what will happen is, the ω frequency wave will go along the same direction, but the 2ω wave will deviate; the 2ω gets generated and does not propagate along the z direction; the energy of the 2ω is not propagating along the z direction, because the s vector of the second harmonic, which is an extraordinary wave, is not parallel to the k vector.

So, this effect is called walk-off and this creates a problem in efficiency; because, now, they are no more interacting; if they separate out, they are not interacting any more. So, you need to ensure that, **so**, if you take a beam of a certain cross section, the walk-off, that means, complete beam walk-off will take place over a certain length, depending on this angle; so, you cannot use crystals much longer than that. Because, the ω wave is propagating like this, and the 2ω , we start from here, and then, actually, its continuously feeding off at some other angle. So, **this**, in Critical Phase Matching, there is another problem of beam walk-off; in Non-critical Phase Matching, there is no problem, because, you are propagating perpendicular to the optic axis, s vector of the extraordinary wave, is parallel to the k vector of the extraordinary wave.

So, **it is always**, one always tries to find out the possibility of Non-critical Phase Matching, but otherwise, one has to use the Critical Phase Matching issue and generate the second harmonic.

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So, this is an example of a crystal, **in which...** So, what is expected is, if I take a crystal so, **here is my**, suppose, my KDP crystal, suppose, the optic axis is like this; so, I propagate making an angle of 50.5 degrees with the optic axis; this is the frequency ω ; and tell me, what is the polarization of the ω frequency, with respect to this diagram? Is it in the plane of the diagram or perpendicular plane of the diagram? **Perpendicular plane**. Perpendicular plane, it is an ordinary wave - right?

So, the ω frequency will come like this, this is the polarization state; and the extraordinary wave at 2ω , will get generated in a polarization parallel to this plane. So, this is the direction of k of ω , of k of 2ω and s of ω ; the pointing vector corresponding to ω frequency, the propagation vector of the ω frequency, the propagation vector of the 2ω frequency; but the pointing vector of the 2ω frequency is not parallel to the k vector of the 2ω frequency. So, the energy that is getting generated at 2ω is actually walking-off from the beam, the main beam.

So, here, the beam which is coming from here, and this polarization state, if I want to plot the D vector of the second harmonic; this is D of 2ω ; remember, D vector is perpendicular to the k vector, so, when I say polarization in anisotropic medium, it is the D vector direction. So, I launch a horizontal polarized light in the KDP crystal and I am expecting the generation of 2ω , which is **in the plane of** in this vertical plane.

Now, as I raise this issue, how am I sure that the second harmonic will generate in the perpendicular polarization state? What is there to couple between this polarization and this polarization? Satisfying phase matching is one issue, but there must be a finite coupling. Suppose, I take two pendulums which have an exactly identical time period, and if I make one oscillate, even if the second one is resonant to the first one, there is no energy transfer if there is no coupling. So, if I take two pendulums which are identical, if and if I oscillate one of them, I must induce some coupling. I must put a spring in between or some mechanical object or something, electromagnetic, so that, energy, these two get coupled.

So, first is, this resonance, suppose, I make a coupling, and make them couple in a non-identical, again there is no transfer of energy; if I take two pendulums which are non-identical in length and if there is a coupling, there is no energy transfer; there is coupling but there is no energy transfer. So, two conditions are required - one is resonance and a finite coupling. So, resonance is something like, here, phase matching. The moment I said this is a phase matching condition, the transfer can be constructive all the time; but first of all, there has to be transfer, there has to be generation of the normal perpendicular polarization state. And, to analyze this, I must now go back and look at the complete equation, the vector equation for polarization in terms of the tensor components of D; because, with this analysis, it is not possible for me to explain or find out, whether at all this is possible.

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$$P_i^{(NL)} = 2 \epsilon_0 d_{ijk} E_j E_k$$

$$P_z^{(NL)} = 2 \epsilon_0 \left[d_{zzx} E_x^2 + d_{xxy} E_x E_y + d_{xxz} E_x E_z + d_{xyx} E_y E_x + d_{xyy} E_y^2 + d_{xyz} E_y E_z + d_{zxx} E_x^2 + d_{zxy} E_x + d_{zxz} E_z E_x + d_{\dots} \right]$$

$$d_{xxy} = d_{xyx}$$

So, to analyze this, now, we move to the second aspect of this polarization, and that is, we go back and look at this equation. P_i , remember, P_i is equal to $2\epsilon_0 d_{ijk} E_j E_k$; this is the actual equation connecting the non-linear polarization, this is only the non-linear polarization. This is the i th component of non-linear polarization, depends on the j th and k th components of the electric fields. I am not writing the summation sign, this repeated indices j and k are being summed over.

So, essentially, means, for example P_x non-linear will be equal to $2\epsilon_0$ into $d_{xxx} E_x^2 + d_{xxy} E_x E_y + d_{xxz} E_x E_z + d_{xyx} E_y E_x + d_{xyy} E_y^2 + d_{xyz} E_y E_z$. How many terms will be there? Plus d_{zxy} , sorry xx Plus $d_{zxx} E_x^2 + d_{zxy}$, sorry, $(())$ $xxx, xxy, xxz, xyx, xyy, xyz$, then sorry, sorry plus $d_{xzx} E_x E_z E_x$ plus d etcetera, so many terms? How many? 9 terms. Because these two indices can take three values each, and I get 9 terms.

There is no difference, because they are both coefficients of this one and this one; they are the same, so, these two elements will be equal; that means, the latter two indices can be interchanged without changing the value of the coefficient; so, which means, the d_{xxy} will be equal to d_{xyx} because both are coefficients of $E_x E_y$. Are we adding them right there?

So, I will have two times factor here, so, these two are equal; see when a plus b whole square, I will get $2ab$; so, similar, I will get two times; so, this is... Please remember here also, that these are total electric fields, and this is the total non-linear polarization; this will contain $\omega^2, \omega^3, \omega$ - whatever it is, all kinds of frequencies. So, this equation relates the total polarization to the total electric field. Now, let us apply this to... So, this is the general equation, relating the non-linear polarization to the electric fields because of second order effect; this is the second order non-linearity, because it is the product of two electric fields.

In a third order, I will have $\chi_{ijkl} E_j E_k E_l$, so, this d_{ijk} has 27 elements; and I cannot write this metrics, because they are 3 by 3 by 3 matrix; it is a 3 dimensional matrix. But, as he raised, fortunately there is no difference between $E_j E_k$ and $E_k E_j$, so, d_{ijk} is equal to d_{ikj} ; and that will help me in what is called as contracting indices; and I can transform this into a 2 dimensional matrix, I will show you.

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$$E_j = \frac{1}{2} \left[E_j^{(\omega)} e^{i(k_1 z - \omega t)} + c.c. \right] + \frac{1}{2} \left[E_j^{(2\omega)} e^{i(k_2 z - 2\omega t)} + c.c. \right]$$

$$E_k = \frac{1}{2} \left[E_k^{(\omega)} e^{i(k_1 z - \omega t)} + c.c. \right] + \frac{1}{2} \left[E_k^{(2\omega)} e^{i(k_2 z - 2\omega t)} + c.c. \right]$$

$$P_i^{(2\omega)} = 2 \epsilon_0 d_{ijkl} E_j E_k$$

$$P_i^{(2\omega)} = 2 \epsilon_0 d_{ijkl} \frac{1}{4} \left[E_j^{(\omega)} E_k^{(\omega)} e^{2i(k_1 z - \omega t)} + c.c. \right]$$

So, before that, let us look at what kind of non-linear polarization will I generate at 2 omega frequency. So, for this, now, I must worry about the components of electric vector; so, let me write E_j is a sum of j th component of omega electric field and j th component of 2 omega electric field.

So, I will have half E_j of omega $e^{i(k_1 z - \omega t)}$ plus complex conjugate plus half E_j of 2 omega $e^{i(k_2 z - 2\omega t)}$ plus complex conjugate. Similarly, the k th component of electric field will be simply this with j replaced by k , because, I do not know a **priori**, what are the components of the electric field of the wave at the frequency omega and the frequency 2 omega? - **plus complex conjugate** - So, I need to substitute this into this equation $2 \epsilon_0 d_{ijkl} E_j E_k$; E_j superscript omega is the **electric** j th component of electric field; at frequency omega, this is the j th component of electric field at 2 omega, k th component of electric field at omega, k th component of electric field at 2 omega.

Now, can you tell me **when I so...** I must multiply these two then, multiply by $2 \epsilon_0 d_{ijkl}$ and find out P_i at 2 omega, so what will I get? So, first thing is, I get $2 \epsilon_0 d_{ijkl}$ and half into half is 1 by 4. Now, tell me what terms will I get, when I multiply these two at frequency 2 omega? This into this will give me 2 omega; this complex conjugate and the complex conjugate will also give me 2 omega, that is the complex conjugate, anything else? There is no other term, so, I will have here E_j omega E_k

omega exponential $2 i k 1 z$ minus omega t plus complex conjugate, which I can write as follows, which I can write as follows: P_i of 2ω is equal to half of $\epsilon_0 d_{ijk} E_j \omega E_k$ of omega exponential $2 i k 1 z$ minus omega t plus complex conjugate.

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$$P_i^{(2\omega)} = \frac{1}{2} \left[\epsilon_0 d_{ijk} E_j^{(\omega)} E_k^{(\omega)} e^{2i(k_1 z - \omega t)} + c.c. \right]$$

$$= \frac{1}{2} \left[\tilde{P}_i^{(2\omega)} e^{2i(k_1 z - \omega t)} + c.c. \right]$$

$$\tilde{P}_i^{(2\omega)} = \epsilon_0 d_{ijk} E_j^{(\omega)} E_k^{(\omega)}$$

$$P_i^{(NL)} = 2 \epsilon_0 d_{ijk} E_j E_k$$

$$= 2 \epsilon_0 \left[d_{xxx} E_x^2 + d_{xxy} E_x E_y + d_{xxz} E_x E_z + d_{xyx} E_y E_x + d_{xyy} E_y^2 + d_{xyz} E_y E_z + d_{xzx} E_z E_x + d_{xzy} E_z E_y + d_{zzx} E_z E_x + d_{zzy} E_z E_y + d_{zzz} E_z^2 \right]$$

So, this expression is exactly like the electric field, half of a component plus a complex conjugate; just like I wrote before, P non-linear 2ω as half of something into exponential, this thing plus complex conjugate; remember, we had obtained E square earlier; $\epsilon_0 d E$ squared, we have got. Now, e square is actually $E_j \omega E_k$ of omega, so let me call this as, half of P_i tilde at 2ω exponential $2 i k 1 z$ minus omega t plus complex conjugate, where P_i tilde at 2ω is equal to $\epsilon_0 d_{ijk} E_j$ of omega E_k of omega.

Please differentiate between this equation and this equation; this is the total non-linear polarization, there is $2 \epsilon_0 d_{ijk} E_j E_k$; this is the total electric field; this is the total non-linear polarization; this equation, here, gives me the i th component of non-linear polarization at frequency 2ω , because of the j th and k th components of the electric fields at ω . This electric field contains both ω and 2ω ; this electric fields are only at ω .

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$$P_i^{(2\omega)} = \frac{1}{2} \left[\epsilon_0 d_{ijk} E_j^{(\omega)} E_k^{(\omega)} e^{2i(k_1 z - \omega t)} + c.c. \right]$$

$$= \frac{1}{2} \left[\tilde{P}_i^{(2\omega)} e^{2i(k_1 z - \omega t)} + c.c. \right]$$

$$\tilde{P}_i^{(2\omega)} = \epsilon_0 d_{ijk} E_j^{(\omega)} E_k^{(\omega)}$$

$$P_i^{(NL)} = 2 \epsilon_0 d_{ijk} E_j E_k$$

$$= 2 \epsilon_0 \left[d_{xxx} E_x^2 + d_{xyx} E_y E_x + d_{xzx} E_x E_z + d_{yyz} E_y E_z \right]$$

Now, I leave it to you to find out a similar expression for P_i non-linear ω ; because, when you take a product of these terms, when you multiply those electric fields, when you multiply this $E_j E_k$ here; we substitute and multiply from these products, you will also get terms of the type exponential minus $\psi \omega t$.

So, please pick up those terms and next time, tell me what their equation is. So, P_i non-linear ω , I want to find out; so, I will have to write it in this form - half of something into exponential i , you will get $k_2 - k_1$ into $z - \omega t$ plus complex conjugate, exactly like before, except that, now, this is the component form; earlier, we did not worry about the components of electric vectors and so on, but here, this is now, the actual i th component of non-linear polarization at second harmonic generated by the j th and k th components of electric fields at ω frequency.

So, let me look at an example, **so, okay**, So, first thing is, as I told you, j and k indices can be interchanged, because, there is no difference between $E_j E_k$ and $E_k E_j$; so, I can actually contract these two indices. So, there is a particular standard convectional for contraction, two indices, 11 is represented by 1; 22 is represented by 2; 33 is represented by 3; 23, 32 is represented by 4; 13, 31 is represented by 5 and 12 21 is represented by 6. You can remember very easily, this is 11, 22, 33, 23, 13 and 12 so this is 1, 2, 3, 4, 5, 6.

This is a standard convention used to denote piezo-electric tensors or electro-optic tensors or non-linear optical tensor and so on. Whenever indices are contracted, this is the standard notation, so, when I say d_{14} , it means, i is 1 and the second coefficient is 4, that means, it is either 23 or 32; both are same; so, d_{123} is d_{14} , d_{112} is d_{16} and so on. So, the last two indices are contracted, and so, when you contract this, now, d tensor becomes, what is the dimension of the matrix? 3 cross 6.

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$$d = \begin{pmatrix} d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\ d_{21} & d_{22} & d_{23} & d_{24} & d_{25} & d_{26} \\ d_{31} & d_{32} & d_{33} & d_{34} & d_{35} & d_{36} \end{pmatrix}$$

KDP

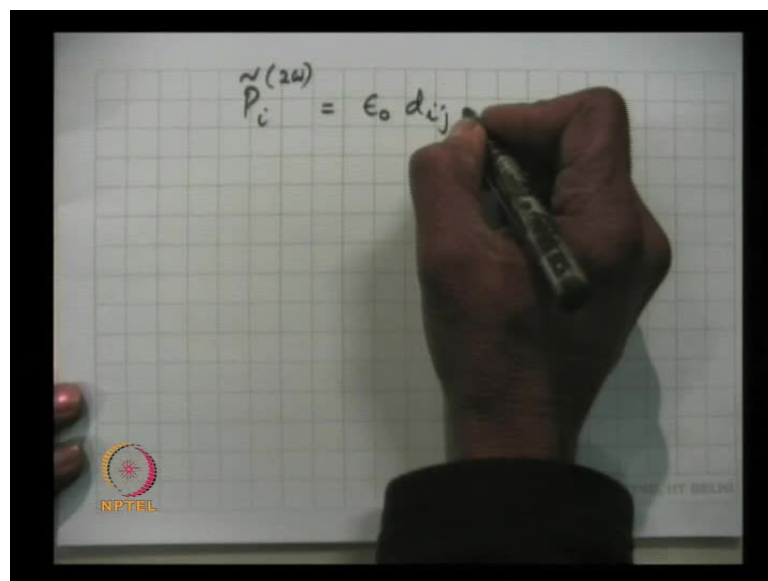
$$d = \begin{pmatrix} 0 & 0 & 0 & d_{14} & 0 & 0 \\ 0 & 0 & 0 & 0 & d_{25} & 0 \\ 0 & 0 & 0 & 0 & 0 & d_{36} \end{pmatrix}$$

So, I will have, I can write the d matrix like this, so, d tensor will be d_{11} , d_{21} , d_{31} , d_{12} , d_{13} , d_{14} , d_{15} , d_{16} , d_{22} , d_{23} , d_{24} , d_{25} , d_{26} , d_{31} , d_{32} , d_{33} , d_{34} , d_{35} and d_{36} ; 18 elements instead of 27.

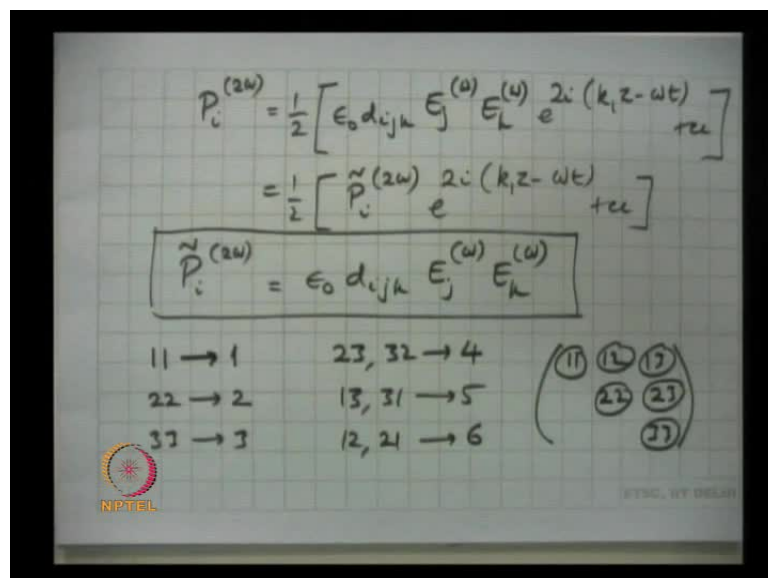
Now, if you know the symmetry properties of the crystal, that symmetry property must be exhibited in the properties of the medium. So, if I can actually use the symmetry properties present in a crystal to find out, which of these elements are 0, which of these elements are equal to each other, which of these elements are negative of each other and so on. **So, I apply symmetry operations on the...** I know, the symmetry properties of my crystal, which is the mirror symmetry, that must be exhibited here; if there is a rotational symmetry by 90 degree, that must be exhibited here. So, any symmetry will be exhibited here; so, actually, **I can** for a medium, I can find out which elements are zero, which elements are non-zero, which elements are equal to each other, and so on.

So, all crystals belonging to one class will have the same d matrix, the values may be different; so, let me give you for KDP, potassium di hydrogen phosphate, **0 0 0...**. There are only three non-zero elements; and similarly, for lithium niobate, there is another matrix; all crystals belonging to the KDP class, ammonium di hydrogen phosphate is another crystal, which is like KDP, it has a same matrix except, values for these coefficients may be different; and as I told you, these are typically 10 to the minus 12 meters per volt, the values.

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$$\begin{aligned}
 \tilde{P}_x(2\omega) &= \epsilon_0 d_{xxx} E_x^{(2\omega)} + \epsilon_0 d_{xyy} E_y^{(\omega)^2} \\
 &\quad + \epsilon_0 d_{zzz} E_z^{(\omega)^2} + 2d_{xyz} E_y^{(\omega)} E_z^{(\omega)} \\
 &\quad + 2d_{xxz} E_x^{(\omega)} E_z^{(\omega)} + 2d_{xxy} E_x^{(\omega)} E_y^{(\omega)}
 \end{aligned}$$

$$\begin{pmatrix} \tilde{P}_x(2\omega) \\ \tilde{P}_y(2\omega) \\ \tilde{P}_z(2\omega) \end{pmatrix} = \begin{pmatrix} d_{11} & d_{12} & & & \\ & & & & \\ & & & & \\ & & & & \\ & & & & d_{36} \end{pmatrix} \begin{pmatrix} E_x^{(\omega)^2} \\ E_y^{(\omega)^2} \\ E_z^{(\omega)^2} \\ 2E_y^{(\omega)} E_z^{(\omega)} \\ 2E_x^{(\omega)} E_z^{(\omega)} \\ 2E_x^{(\omega)} E_y^{(\omega)} \end{pmatrix}$$

So, because of this contraction, my equation is actually, this equation, which I have wrote down earlier, simplifies to \tilde{P}_i of 2ω is equal to $\epsilon_0 d_{ij}$, no, let me write like this, sorry. So, let me write the \tilde{P}_x of 2ω , will be, now, you see, whenever j and k are equal, sorry, whenever j and jk and kj terms, they are equal, so what will I get when I expand? I will have $\epsilon_0 d_{xxx}$, E_x square, E_x at ω square, actually, plus $\epsilon_0 d_{xyy} E_y$ of ω square plus $\epsilon_0 d_{zzz} E_z$ of ω square plus, now, you will have $d_{xxy} d_{xyx}$; and similarly, so, I will have $2 d_{xyz} E_y E_z$ plus $2 d_{xxz} E_x E_z$ of ω plus $2 d_{xxy} E_x E_y$ of ω . There are still 9 terms, actually, these 3 terms contain the twice the number.

Similarly, I can write an equation for \tilde{P}_y of 2ω \tilde{P}_y tilde \tilde{P}_z tilde; so, actually, you can write this entire thing in a matrix equation, \tilde{P}_x tilde of 2ω \tilde{P}_y tilde of 2ω \tilde{P}_z tilde of 2ω is equal to just d matrix; so, $d_{11} d_{12}$ etcetera into E_x of ω square E_y of ω square E_z of ω square $2 E_y$ of ωE_z of ω $2 E_x$ of ωE_z of ω and $2 E_y$ of, sorry, $2 E_x$ of ωE_y of ω .

For example, I can check this; so, \tilde{P}_x tilde ω , so $d_{11} E_x$ square $d_{12} E_y$ square, this is d_{12} plus $d_{13} E_z$ square, this is d_{13} , this is $2 d_{14} E_y E_z$, so, this d_{14} multiply by 2 by $E_y E_z$; then, I have $2 d_{15} E_x E_z$, so $2 d_{15} E_x E_z$ and $2 d_{16} E_x E_y$ $2 d_{16} E_x E_y$.

So, the three component of the non-linear polarization that are generated, are related to the electric fields components at frequency ω , the x and y and z components, through this matrix relationship. So, what I will do in the next class is, look at some examples KDP and lithium niobate; and, as I gave you this example - 50.5 degrees, I need to travel, **at**, making an angle of 50.5 with the optic axis; but 50.5 can be in this cone? Is there any restriction? I will show you, that if you are not careful, you may have no coupling; you will be phase matched, but there is no coupling; because, the non-linear coefficient for that interaction may be 0. So, you need to be careful in not only achieving phase matching, but also, choosing the right direction with respect to the crystal axis, **so**, such that, there is a finite coupling between the ω and 2ω frequency. Are there any questions?

No, that is anisotropic medium; under linear approximation, we were studying the linear properties of anisotropic media because, actually, what is non-linearity doing? It is, non-linearity is generating or coupling different frequencies. So, once an electromagnetic wave gets generated at the new frequency, it will propagate through the medium as per the Maxwell's equation, which we have solved. So, all that the non-linearity is doing is, generating a non-linear polarization which acts as a source to generate electromagnetic waves at the new frequency; and that wave will travel just like a wave in an anisotropic medium which we have analyzed earlier. Is it perfectly all right to use those results here or is it just an approximation? No, **it is perfectly all right**, it is perfectly all right.

It is just like, I can analyze the problem of a pendulum individually, and when I have coupling, I still use the same set of equations, with an extra coupling in between, and say, that the energy transfer etcetera, so, there is no problem at all. Anything else?

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$$P_x^{(2\omega)} = \epsilon_0 d_{xxx} E_x^{(2\omega)} + \epsilon_0 d_{xyy} E_y^{(\omega)^2} + \epsilon_0 d_{zzz} E_z^{(\omega)^2} + 2d_{xyz} E_y^{(\omega)} E_z^{(\omega)} + 2d_{xxz} E_x^{(\omega)} E_z^{(\omega)} + 2d_{xxy} E_x^{(\omega)} E_y^{(\omega)}$$

$$\begin{pmatrix} P_x^{(2\omega)} \\ P_y^{(2\omega)} \\ P_z^{(2\omega)} \end{pmatrix} = \begin{pmatrix} d_{11} & d_{12} \\ & \\ & \end{pmatrix} \begin{pmatrix} E_x^{(\omega)^2} \\ E_y^{(\omega)^2} \\ E_z^{(\omega)^2} \\ 2E_y^{(\omega)} E_z^{(\omega)} \\ 2E_x^{(\omega)} E_z^{(\omega)} \\ 2E_x^{(\omega)} E_y^{(\omega)} \end{pmatrix}$$

This will lead to 4 omega generation also, because, the second harmonic eigen will again while travelling in anisotropic medium, will have 4 omega generation because of non-linearity. Yes.

Now, will 2 omega generation, **will** also lead to 4 omega generation? Yes, in principle, but I need to satisfy the phase matching condition, that, the refractive index at 2 omega must be equal to refractive index at 4 omega; **this will not happen**. So, which in general, will not happen, **so, I will not be...** number 1; number 2, the power generated at 2 omega is already very small, so, at that frequency, there is no non-linearity, **this**, I mean, the non-linearity is very weak corresponding to 2 omega to 4 omega conversion.

So, in general, that does not happen, but it is always possible to have a situation where that is also being satisfied; and then, I have to solve that equation also simultaneously with this.

Thank you, very much.