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Lecture - 25 Matrix Form: SHG, SFG, DFG, SHG in KDP Crystal

So, welcome student to the next class of Non-linear Optics and its Applications. So, in the previous class we have studied 2 different principles one is Neumann's principle and another is Kleinman's symmetry. And applying these 2 symmetries we find that it is possible to reduce the elements of d matrix and eventually from 18 different elements, we can go down to 10 different elements and then further we can even go to 2 different elements 2 different distinct elements and making all other elements are 0.

And that one can find by applying Neumann's principle, we suggest that under symmetry operation the structure of the d matrix remain same. So, today we will going to understand the more important thing that how inside this crystal the second harmonic process is generated. So, let us see what we have today.

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So, in order to understand the second harmonic in the crystal first we need to know what is the matrix form for second harmonic and then sum frequency generation and different frequency generation and then we will study about the second harmonic generation in a specific crystal. We use KDP crystal here which is a very well known crystal to generate second harmonic; so let us start with the matrix form.

Second Harmonic Generation (SHG) $P_i^{(NL)}(2\omega) = \omega + \omega; \omega, \omega) = \epsilon_0 \chi_{ijk}^{(2)} E_j^{(\omega)} E_k^{(\omega)} = 2\epsilon_0 d_{ijk} E_j^{(\omega)} E_k^{(\omega)}$ SHG in Matrix form, $\begin{pmatrix} P_x^{(2\omega)} \\ P_y^{(2\omega)} \\ P_z^{(2\omega)} \end{pmatrix}_{3\times 1} = 2\epsilon_0 \begin{pmatrix} d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\ d_{21} & d_{22} & d_{23} & d_{24} & d_{25} & d_{26} \\ d_{31} & d_{32} & d_{33} & d_{34} & d_{35} & d_{36} \end{pmatrix}_{3\times 6} \begin{pmatrix} E_x^{2(\omega)} \\ E_y^{2(\omega)} \\ E_z^{2(\omega)} \\ 2E_y^{(\omega)} E_z^{(\omega)} \\ 2E_x^{(\omega)} E_z^{(\omega)} \\ 2E_x^{(\omega)} E_z^{(\omega)} \end{pmatrix}_{6\times 1}$ = 1 ITELONLINE CENTIFICATION COURSES Dr. Samudra Roy Department of Physic

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So, this is not a very new thing, but still this matrix form is important and one should understand that how this matrix forms are generated. So, in couple of classes ago we have already demonstrated that how the P matrix is represented in terms of ijk and this P non-linear in terms of ijk is written in this way as shown here; so this is the way one can write.

Since we are dealing with second harmonic; so my frequency component will be 2 omega. So, 2 omega is generated as a result of the combination of omega and omega. So, once I add 2 omega; I will generate the frequency 2 omega this is the 2 omega frequency component of the non-linear polarization. In general non-linear polarization there will be many other frequency terms that we have already shown in our earlier classes, but here we are only looking for those term, where the frequency components are 2 omega.

Now the frequency component of the 2 omega can come from this 2 elements E j and E k with omega and omega. So, when they multiplied each other; so, we will have a 2 omega components and that is why here we are saying this is the second harmonic process. So, eventually in terms of d matrix we will have this.

So, once we have this component form now it is necessary to write in a matrix form because at the end of the day what we have this d matrix in our hand. So, we know what is d 11, d 12 most of the cases as I show in the previous classes that it is vanishing most of the components are vanishing. So, we will have a simplified form of the d matrix and then it is easier for us to calculate P x, P y, P z which are the non-linear components of polarization matrix.

So, now if I write these things in matrix form then P x, P y, P z can simply be represented in terms of E x, E y, E z in this way; so one can varily derive this from this.

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So, if I write P x here in the right hand side; in the left hand side it will be 2 epsilon 0 d of x and now ij is the indices that will going to change. So, first indices will be x x; so I will write E x E x. So, that is E square this is the E square term and this is the d 11 index; x x index means d 111 and we know that d 111 means d 11. So, if I multiply the first term is this what about the second term?

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Second term again in the left hand side P x we have 2 epsilon 0 d 1, second term should be d 22. If I write d 22; the second term we will write d here 2 means y y; so, it should be d y y. So, we will have d 2, d 12 E y square; so this is d 12 E y square the second term.

Similarly, for third term we have d 133 and we have d E ZZ; so, its Z square.

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Fourth term we have d for the fourth term which is d 14 here which is d of 1; 4 means 23. 23 means if I write any terms from XY; so, it is XYZ. So, here we have E of Y, E of

Z and this will come twice. Because this is Y this is Z. So, another term will come where we should have this is Z, this is Z and this is Y, but both the cases we know for the permutation symmetry that d of X of YZ is equal to d of X of ZY; so we will should not bother about that.

Since there will be 2 terms; so, 2 multiplied by E y, E z will be here corresponding to d one forth term. In the similar way d 15 and d 16; we have all the terms here. So, this is for P x in the similar way we can write P y and P z, I do hope all you all of you understand how to write this matrix form, which is very important in studying how the second harmonic generating inside the crystal. And the process is quite straight forward once you know what is the component you are talking about then you can readily able to write. And this is the compact form of this if I expand this it will be coming in this way only .

So this is the matrix form of the second harmonic.

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So, we can also able to write the matrix form of sum frequency generation. So, sum frequency generation is nothing, but if we have 2 frequency components if we have 2 frequency components, then what happened that this 2 frequency components can adopt and generate a summation of that; that we know because we have already studied this things. So, omega 3 is a frequency which is generated by omega 1 and omega 2. So, now,

P ith component can be represented term of E j and E k having the component omega 1 and omega 2.

In the similar way, if I start with P x component it would be x and x; x and x, but the frequency components are different if you should not previously it was E x square, but it is now x x, it is now yy, but the frequency components are different and zz frequency components are different. Once we go to the other term then again we have the combination one combination for example, the fourth term.

So, d 14 is essentially d 123; d 123 means d x y z. So, one x y z is there this is x, this is y, this is Z. So, we will have 2 terms one is y with the frequency component and another is Z with the frequency component 2, but other term is still possible which is y with the frequency component 2 and Z is the frequency component 1; both the cases they will go into generate the frequency omega 3. Because when you multiply these 2 things; you will generate omega 1 plus omega 2 if we multiply by this 2, again it will generate omega 3 which is omega 1 plus omega 2 that is why we have 2 terms adding here.

In the similar way we have the other terms exactly the logic is same. So, this is the way of writing of second harmonic for some frequency in matrix form; well if I able to write the second harmonic and sum frequency.



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So why not the difference frequency? So, the difference frequency one can also write exactly in the same form, but one should remember that since the difference frequency generation. So, we have; so P x; so I need to find out omega 3, omega 3 is how I omega 3 is generated? Omega 3 is generated as omega 1 minus omega 2 the difference between these two. So, since it is related to minus of omega 2 instead of having E k omega 2; I should write E of star of E k of star of omega 2.

Because we know that this is E k star of omega 2 is nothing, but E k of minus of omega 2. So, this notation in many books I will found they will follow. So, we are following the same notation here; so now, instead of multiplying this 2 we now have a star here also in the similar way we will have the summation, but in form of star because we are dealing with difference frequency generation. So, these forms are very important. So, I believe you readily you can understand this things once you; once you write this matrix form verily you will find; what is the value of P x, P y, P z that is important because these are the terms that is generating in the second harmonic waves.

So, that is why the direction is important in which direction they are generating the second harmonic wave or the sum frequency wave or the difference frequency wave is important and one can find out the entire things by just writing this matrix form.



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Well after having the knowledge of this matrix form the next important thing that we will going to learn is about the KDP crystal. KDP crystal is the very interesting crystal and in

KDP crystal normally we generate the second harmonic and let us try to find out what properties we have in KDP crystal.

So, KDP crystal is a uniaxial trans, crystal transference in the visible range; obviously, it is transparent. If it is transparent in the visible range, then it is easier for us to apply it in the visible range, normally we walk in the visible range.

And then it is the second harmonic generation is observed in this crystal by illuminating light 1064 nano meter. And it will go into produce the light at 532 which is exactly the half of the wave length or whatever the wave fundamental wave length we have and then we can generate second harmonic. So, here we have a picture here.

So, if you look to the slide there are many information that is dumped here; one is this is the picture of the crystal and the optic axis is also shown. Optic axis is a preferred axis along which we can define many things and along optic axis, the velocity of the ordinary and extraordinary waves are same; that means, the reflective index along this directions are same.

So, this is the very important direction for act for that axis, but here we also show one important thing and that is how the second harmonics are generated. So, this is the experimental figure; this is the experimental figure; if you launch a light around this which is normally invisible because this is in higher region this is. So, gradually what happened that in the output for a KDP crystal; we will get a reddish color of light which we say for the timing is a fundamental. However, fundamental light if I launch with 1064; it is difficult to see because it is not possible to see with your naked eye, you need to put special thing to find to see this higher lights.

But in the experiments we can do that and this is the nature of the fundamental light and important thing is that we can generate from this fundamental; the second harmonics are generated experimentally and this is the second harmonic spot of second harmonic. So I launch one light which is in not in the visible range in the higher sight, but whatever the light is generated is well in our visible range in our blue sight because when the wave length is near around 53-. So, we will have a bluish light which is our second harmonic.

Also we can find out the reflective indexes because the reflective indexes are very important and this reflective index values are given here for the operating wave length say 1064. So, in 1064 if these are the reflective indices; these will be use full to find out the angle theta this angle theta. Because we have a expression of angle theta along which we have a phase matching and this information basically help us to find out what is the value of theta. There is the expression we have already derived this expression in our earlier classes, but important thing is that here we have a crystal which is negative in nature because n of o of omega is greater than n of E of omega.

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So, the velocity of extraordinary wave is the velocity of the extraordinary ray is faster than velocity of ordinary ray. So, that is why it is a negative crystal. So, we know in the negative crystal ne is less than no except this particular direction which is optic axis, where both the values are same. Well after having the knowledge of KDP crystal; next we need to know with this crystal what we can do. (Refer Slide Time: 15:05)

	Figure 14
KDP Crystal	Here $n_o(\omega) > n_e(\omega)$, so the SHG will be under $o + o \to e$ system. Phase matching angle, $\sqrt{\theta} = \sqrt{\sin^{-1}\left(\frac{n_o(\omega)^2 - n_o(2\omega)^2}{n_e(2\omega)^{-2} - n_o(2\omega)^{-2}}\right)} \approx 41^\circ$
$n_o(\omega) = 1.4938$ $n_o(2\omega) = 1.5124$ $n_e(\omega) = 1.4599$	d-matrix of KDP crystal In principal axis system; $d = \begin{pmatrix} 0 & 0 & 0 & d_{14} & 0 & 0 \\ 0 & 0 & 0 & 0 & d_{25} & 0 \\ 0 & 0 & 0 & 0 & 0 & d_{36} \end{pmatrix}$
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So, here as I mentioned that can able to find out what is that angle because in for KDP crystal; we know that there is a certain angle that which the phase matching is achieved. And this is the example this is the value of this theta. And if I now start putting this value in no of omega no 2 omega from here and the operating angle at the operating angle at the operating wave length say 1064 nano meter.

Then we will find that this angle is around 41 degree; so if I launch a electric field around 41 degree; this is a Z axis and this is a x axis; then we should have a phase matching. Also the d matrix of the KDP crystal is shown; so here you can find that d matrix there is only 3 non zero components are there in the d matrix. And this nonzero components are sitting in this way d 14, d 25 and d 36; this threes are nonzero components in the d matrix.

So, we have the d matrix we know along which angle the phase matching is there these 2 very important information of there. So, now what we will do? We will try to find out what is the condition to generate the second harmonics in KDP crystal or now the second harmonics is generated in the KDP crystal.

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So, before that we need to understand few things. So, let us understand what is going on here; so, here we find for KDP crystal 41 degree is the angle preferred angle along which the second harmonic is generated. But we never mentioned that the orientation of the k because k can be in zx plane and k may not be in zx plane, but still the angle might be 41 degree.

So, if I look in more general figure; so one azimuthal angle. So, one can assume which is preferred here as a phi. So, in all cases if I if this vector is rotate into this cone, cone can make this cone. So, in this cone all for all orientation of the k this angle is theta because these angle eventually a dimensional figure. For 3 dimensional figure we have a rotation like this for all cases; this is touching with this point is basically a round wise as shown here.

So, all the points here over this circle the value of the k is same, magnitude of k is same and the direction is changing because my azimuthal angle is changing. But interestingly the phase matching angle which is phi, which is theta here is not changing. That means, all orientation the phase matching condition is valid; so my question is what is there any preferred direction of theta as well as phi for which we have a more value of the second harmonic more or what is the condition to generate the second harmonic; is it that sufficient is the angle is important this phi is important or not? So, this is the special orientation of the k where angle theta is maintained here, but k is in zx plane. So, we start with this condition and try to find out if k is in zx plane; really second harmonic will going to generate or not that is the first thing that we will do; so let us try to understand that.

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	Case 1 : $\vec{k}(\omega)$ is in the $z - x$ plane		
	Here $\vec{k}(\omega)$ is in the $z - x$ plane making the phase matching angle $\theta \approx 41^{\circ}$ with z-axis (optics axis). To achive the PM under $o + o \rightarrow e$ system the fundamental wave must be o-wave. Now o-wave should perpendicular to \hat{z} and $\vec{k}(\omega)$, i.e optic axis and $\vec{k}(\omega)$. That means o-wave in along y-direction.		
	\hat{z} $P_i^{(NL)} = 2\epsilon_0 d_{ijk} E_j E_k$ $E_i = \frac{1}{2} \left[\tilde{E}_i^{(\omega)} e^{i(kz - \omega t)} + c.c \right]$		
	$\overrightarrow{E} = E^{(\omega)} \hat{v} \qquad P_i^{(2\omega)} = \frac{1}{2} \epsilon_0 d_{ijk} \left[\tilde{E}_j^{(\omega)} \tilde{E}_k^{(\omega)} e^{2i(kz-\omega t)} + c.c \right] = \frac{1}{2} \left[\tilde{P}_i^{(2\omega)} e^{2i(kz-\omega t)} + c.c \right]$	$(z-\omega t) + c.c$	
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So, case 1; in case 1 what we have? In case 1 we have k is in z x plane. So, k is in z x plane make an phase angle theta is 41 degree. So, this angle is around 41 degree and z is a optic axis; to achieve the phase matching condition around this o o to e system; that means, 2 ordinary wave that you can merge and generate an extraordinary wave.

So, the fundamental wave must be in ordinary wave that is the first thing that we should look into; that what should be the fundamental wave? And this fundamental wave should be ordinary wave because 2 ordinary wave can generate an extraordinary wave, this is the o plus o equal to e system, since the crystal is negative in nature. So, now the o waves should be perpendicular to the z axis and k that is very important. So, the orientation of the o wave if this is o wave it should be perpendicular to both k as well as the z; the optic axis, the o wave is always perpendicular to k propagation constant, the direction of propagation constant and z.

Here if you see that this is z; k is in this plane. So, if it is perpendicular to both then the E has to be in y direction which is perpendicular to this plane.

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So, now we know my P non-linear is this where e is this, P 2 non-linear is represented by this where half of P this things; I just write the old equation here, but orientation of the k is important. Orientation of the k it is making an phase matching angle, but it is in x z plane; the question is if this is the orientation whether really we will go into get any kind of second harmonic or not.

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So once we have the knowledge of my d matrix; it is readily one can readily find out what is the matrix form of the second harmonic that we have already shown in this class.

But before that I can write the P i 2 omega till the this is the amplitude of the non-linear polarization at 2 omega point is equal to E j omega and E k omega which till the; that means, the amplitude of this. So, now if I try to find out what is the amplitude of the electric field for this system; we can say that electric field around x and electric field around Z is 0 here it is mistake here should be Z; E Z.

So, electric field is only around y direction because if you remember the previous slide k is along this direction, this is our k vector making an angle 41 degree, this is optic axis Z and in order to have the second harmonic; my fundamental wave has to be a has to be an ordinary wave. So, this ordinary wave is perpendicular to Z as well as k so; that means, it is along this perpendicular direction and this perpendicular direction means it is along y direction. Since it is x and Z plane it is along y direction; so that is why E y is not equal to 0 here.

So, now what we will do? We now know what is E x, E Z and E y; E x E Z is 0. So, this is 0, this is 0 all other terms will be 0 d matrix for KDP crystal is given. So, this is my d matrix and P x P y P z is there; so we need to find out what is my P x, P y, P z. Now if I if I calculate this P x, P y, P z we find the interest thing that P x, P y, P z all are 0; that means, even though the k is launched along the direction for which the phase matching angle is satisfied, but still we are not getting any kind of second harmonic because the matrix product. So, what is the conclusion here? Try to understand one by one.

So, first we see that P x, P y, P z can be 0 even though the phase matching condition are there. So; that means, launching of the k is very important the azimuthal angle is important. If I launch the k vector along x z plane, but still the phase matching is there, but will that cannot ensure that the second harmonic will go into generate. So, the matrix product here what the conclusion we have? Let us check the matrix product vanishes for the specific orientation of k vector no non-linear polarization of 2 omega.

Since the k is in as I mentioned is exit plane; so, the matrix product is 0 here. So, the P x P y P z is 0. So, no 2 omega generation can be possible, but you should note that the phase matching is still there. So; that means, only the phase matching condition cannot ensure that they will go into generate the second harmonic. So, only the phase matching condition of the direction of k vector is not only here should have a t.

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So, not only the phase matching condition the direction of the k vector is equally important to excite second harmonic as I mentioned.

So, here we find that phase matching is there I launch the k vector along the direction for which the phase matching in there, but azimuthal angle is very important because I can launch the k vector in infinite orientation for which the phase matching is there, but the direction is different. Based on the k matrix k direction one can know what is the direction of e ordinary and extraordinary which eventually generate the second harmonic.

So, in a next class we find that for KDP crystal which should be the optimum direction for which we can generate the second harmonic. So; that means, inside the crystal the direction is very very important; if I launch the k in direction for which the phase matching is there that cannot insure singly. So, this is not the only condition for generating second harmonic, we need to find out the azimuthal angle that orientation of the k is really very important for which I can generate or maximize our second harmonic.

So, today we will going to conclude our class. So, we just started the generation of second harmonic in KDP crystal; one condition is shown where the k is in exit plane. So, in the next class we show we change the orientation of k and trying to find out how the second harmonic will go into generate. And most importantly if that is the case how the d

matrix will effect and what should be the effective d value which value because there are 3 different d values are there in KDP crystal..

Which value which d value is really responsible for generating the second harmonic that is important. We need to find out that d value for which we are generating the second harmonic. So, with this note let me conclude my class here; so see you in the next class, where we learn more about the second harmonic generation in KDP crystal and about the orientation of the k and optimize the d matrix value so.

Thank you for your attention, see you in the next class.