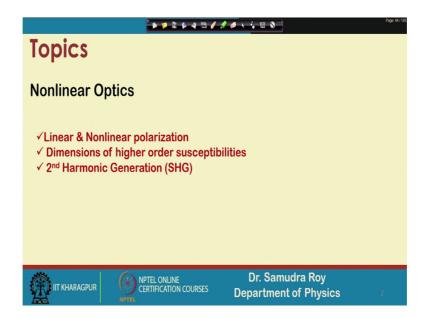
## Introduction to Non-Linear Optics and Its Applications Prof. Samudra Roy Department of Physics Indian Institute of Technology, Kharagpur

# Lecture - 13 Second Harmonic Generation (SHG)

Welcome student to the next class of introduction to non-linear optics and its application. So, in the previous classes we have find a very important concept about the susceptibility. And specially how it relates with the frequency and then find out one important law, which is called the millers rule, where we can find the susceptibility of different frequency and can correlate them with a particular frequency.

So now today let us see what we have in our class.

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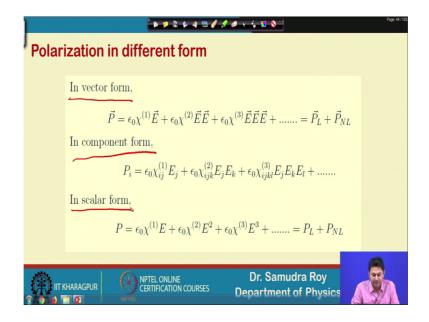
So, today we will have 3 very important concept in non-linear optics one is linear and non-linear polarisation, the first one we will going to learn what is the meaning of linear and non-linear polarisation. However, these term has already been introduced to you that is nothing new, but we will going to learn in detail.

Then the dimensions of higher order susceptibility. So, susceptibility in general is a quantity which has some kind of dimension. So, we will find out to what will be the dimension of the susceptibility when it is higher order

and then very important concept second harmonic generation. So, second harmonic generation is one of the important concept as we learned from the previous courses of previous classes. That when we launch particular frequency and if it is passed through a non-linear medium, then what happened? That this frequency gets double. So, we will get a frequency having twice than the input frequency and this is generally called the second harmonic generation.

So, today we will going to learn, how the second harmonic generation is there from the very first principle calculation or very simple calculation ok. So, first polarisation is different form so if I look this slide.

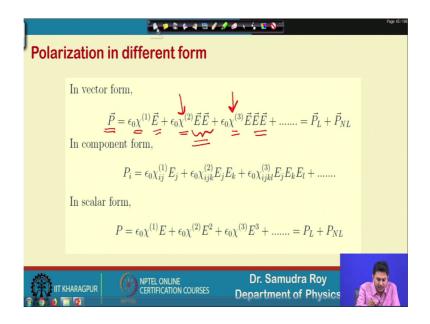
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Then you will find there are 3 forms written here one is in vector form that is the first one this is in the vector form. And then one is a component form and then in a scalar form. In the books if you find any non-linear optics book you will find that the polarisation is always defined by these 3 forms, either it is in vector form that is the first one or it is in component form that is the second one. And or it is in the third form like the scalar form we called as a scalar form.

So, every form has their own significance. So, let us try to understand 3 different form in a more detail way. So, in the first form you can see that the left hand side is a vector quantity.

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So, P is essentially a vector which is related to the susceptibility tensors where these quantities are first order second order ah tensor and so on. And then it is related to the electric field E. The first term is a well-known term P is equal to epsilon 0 chi 1 E, but the second third fourth fifth etcetera terms are which generally different way. You can see that one term is written is E, E this is nothing a like a multiplication of 2 vector rather this is a notation this is a tensor kind of notation this product is non dot or cross product I just write EE to ensure that the interacting become a vector, but this product may be related to some tensor property because these quantity are tensor.

So, in order to understand the polarisation is a vector quantity, we need to use this

particular form in more realistic representation. So, if I say polarisation; that means, that the left hand side polarisation P is a vector quantity. If it is a vector quantity the right hand side should also be vector, but here this terms are normally tensor if these are the tensor quantity then we need to write the E square E cube term in such a way that the entire component should form a vector. So, that is why this is some sort of a form where we can say that the polarisation is indeed a vector quantity. So, in order to have the sense that the polarisation has some vector property we normally use this form.

So, second from the component form are very important, because we are basically representing in the first form, but in a more general way or rather more simple way. So, that we can appreciate that this quantity chi is essentially a vector a tensor.

Polarization in different form In vector form,  $\vec{P} = \epsilon_0 \chi^{(1)} \vec{E} + \epsilon_0 \chi^{(2)} \vec{E} \vec{E} + \epsilon_0 \chi^{(3)} \vec{E} \vec{E} \vec{E} + \dots = \vec{P}_L + \vec{P}_{NL}$ In component form,  $P_i = \epsilon_0 \chi^{(1)}_{ij} E_j + \epsilon_0 \chi^{(2)}_{ijk} E_j E_k + \epsilon_0 \chi^{(3)}_{ijkl} E_j E_k E_l + \dots$ In scalar form,  $P = \epsilon_0 \chi^{(1)} E + \epsilon_0 \chi^{(2)} E^2 + \epsilon_0 \chi^{(3)} E^3 + \dots = P_L + P_{NL}$ 

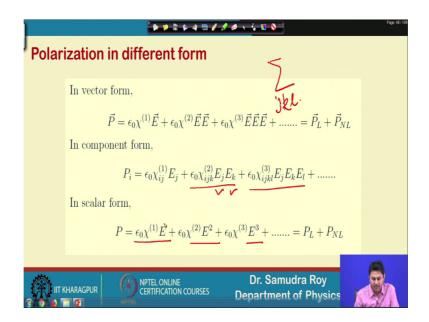
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So, first here in component wise I write. So, Pi is essential the ith component of the vector P and if it is ith component it should be related to the different component of the E field because polarisation is not only depending on the one particular field a component E, but there may be a combination since it is E square E cube these dependents are there see if I go to the first term you can see Pi is related to E j and chi ij. So, chi ij is nothing but a tensor quantity having few components and then i is there and j is there. So, there are one j here for this chi and another j is here for E.

So, these 2 j are there. So, we know that this is called the Einstein notation we know that if the induces are repetitive then; that means, essentially we are summing it over so; that means, if I write Pi in a more general way I should write it is a j epsilon 0 ij and then Ej this i is ith component, but here this j is summation.

So now if there are 2 components are there jj, which is sitting side by side or repetitive indices then we should not use this summation sign. So, just reduce the complexity we just simplify the equation and this is called the Einstein notation. In the future classes also we will not going to use the summation sign all the time whenever you have this repetitive indices then you will find you should understand that it is summed over that in indices ok. So, this is the first term.

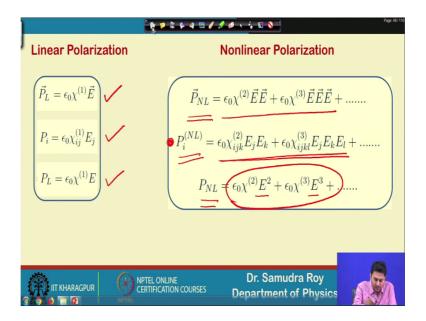
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In the second term we have chi ijk when it is ijk they are should be related to j and k because it is a square term. So, 2 components should be involved when 2 components are there then there should be ijk. Then the third order effect where we should have ijk and l also because jkl all are there. When jkl all are there then it should be some over all this indices. So, if I write in terms of summation this summation is essentially i ith component. So, it is j and then k and then l. So, all these things is summed over this.

And finally, we have the scalar form. So, scalar from is a most simple form, but it is very useful to understand the fundamental effects for example, second order effect third order effect second harmonic generation, third harmonic generation, sum and different frequency generation. All these important effect can be understood with this these terms, where nothing is there nothing vector component or any vector sign or any component is there only I am using the amplitude of E. Since we are using the amplitude of E only the E component is only the amplitude is important we called it the scalar form, which is very simple and very useful you can see that the vector sign as well as the ij ij term is removed here ok.

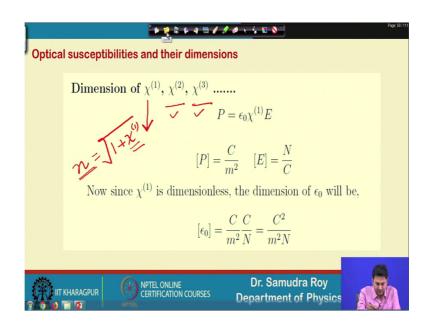
After understanding this particular form that there are 3 forms.



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Now we will go back to our second slide that we represent we just separate out this polarisation in 2 major parts. One is the linear polarisation where E is P is proportional to eventually proportional to the electric field E.

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In vector form, in vector form we should have like this the first term. In vector form in component form we have the second that Pi is epsilon 0 kappa ij ej this is the normal representation when the polarisation is simply ah linear in nature. And PL stands for the linear polarisation where polarisation and electric field are proportional to each other and P and E are related with this form. So, this is the most standard form we always use for linear case.

However for non-linear case we just change the notation because in our case the nonlinear polarisation is rather important. So, that is why what we do that is separate out the terms which is related to E square E cube and so on. So, in vector from again I should write EE EEE and so on. This is a vector notation and I write this is the P non-linear polarisation also the ith component of the non-linear part of polarisation can be represented with the this terms, which is the same terms that we have already explained in the previous slide. And finally, PNL, PNL is the non-linear polarisation and this nonlinear provisions related to electric fields this is the scalar form. So, that is why E square E cube are related to that.

So, we have 3 different forms here and these 3 different forms are now divided to linear part and non-linear part and in the future classes we will going to use the non-linear part.

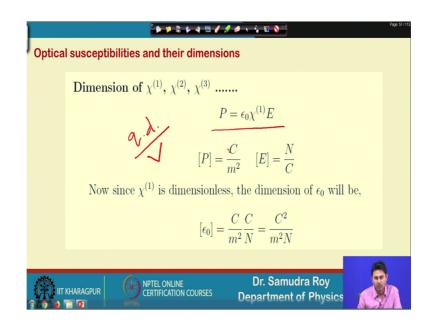
And normally we use this component form which is very useful and it should one should understand how things going on, but today on maybe in the next class we will be more give you more emphasis, we will give more emphasis on in to this particular terms or the linear representation which basically leads to very important outcomes in terms of physics.

So, let us now go back to our go to our next slide ok. In the next slide what we basically represent is the dimensions of the susceptibility terms. So, optical susceptibility should have some kind of dimension because the susceptibility is not now first order in non-linear optics we have second and third order susceptibility. So, when the second and third order susceptibility that it should have some kind of dimension.

However, we know that the first order susceptibility should not have any kind of dimension, why? Because we know that the refractive index is a dimensionless quantity and refractive index is represented by this term. So, when the refractive index is represented by this term; that means, it is one is added with some quantity which is the first order susceptibility. And this first order susceptibility should be dimensionless. So, that the total ah N; that means, the refractive index should be dimensionless.

But what happen when chi 2 and chi 3 are there, one has to investigate ok. So, let us do that this is not very very critical thing to understand. So, let us start with the first-order ah polarisation of the linear polarisation. So, in linear polarisation what happened?

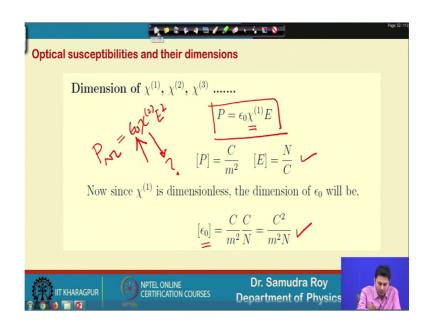
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That it is related to E and there is a proportional to relation. And then P is the polarisation term and it should have some dimension. And we know that polarisation is a dipole moment per unit volume dipole moment is nothing but charge in to distance, charge into distance divided by the volume. So, this volume should have 3 distance; that means, this centimeter centimeter centimeter and this distance is centimetre. So, if I cut it should be centimetre square and this charge is coulomb. So, coulomb divided by centimeter square or in this unit it is meter square. So, coulomb divided by meter square is essentially the dimension of the polarisation, we know the electric field dimension is newton per coulomb.

So now if I combine this 2 into this equation then can have the dimension of epsilon 0. So, most of you may be aware of the epsilon 0 dimension, but for me to be very honest I never remember all this term very clearly. So, that is why I just cross verify and try to of find out this dimension in a different way.

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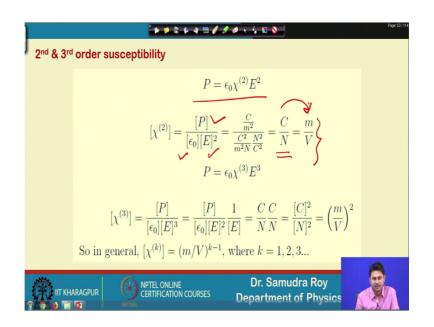


So, here if we know that this is the dimensionless quantities. So, epsilon 0 dimension can be easily figure out from this equation, from the first equation given in here this is dimensionless we know that I know what is the dimension of E I know what is the dimension of P. So, when once we know the dimension of P and E if I put that then the dimension of epsilon will be C 0s coulomb square divided by meter square per newton as simple as that.

Why the dimension of epsilon 0 is important because whenever we try to find out for example, if I try to find out the non-linear the second order susceptibility then the P non-linear is epsilon 0, then chi 2 and then E square. So, epsilon 0 is common both the cases. So, epsilon 0 dimension is important I try to find out the dimension of this quantity. So, once we know the dimension of epsilon 0 E and P dimension is already written here, then I can readily find out what is the dimension of chi 2 in the same way I can find out what is the dimension of epsilon 0 is on. So, that is why the dimension of epsilon 0 is importance.

So, epsilon 0 dimension is now known it is coulomb square divided by meter square newton. Once it is known then what we do we just use that to find of the dimension of chi 2 and higher orders.

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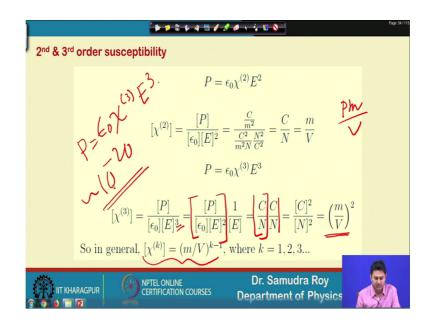


So, so here we try to find out as I mention in the previous slide. I like to mention I like to find out the dimension of chi 2. So, here is my equation the polarisation is epsilon 0 chi 2 E square. This polarisation is essentially the non-linear polarisation, but even if it is a non-linear polarisation the dimension of the polarisation, linear polarisation and non-linear polarisation should be same there should not be any difference.

So, if I try to find out what is the dimension of chi 2 or the second order susceptibility, then it should be the dimension of polarisation divided by the dimension of epsilon 0 which we have already figure out in the previous slide and then E square of that. So, if I now put term by term. So, the dimension of P is coulomb per meter square that we have already figure out in the previous slide.

Epsilon 0 again we you are going to use this is coulomb square divided by meter square and then newton. And it will be multiplied by the dimension of electric field which is newton by coulomb and then make if I make the square of that it should be newton square by coulomb square. And after combining all these things finally, we have an dimension like coulomb per newton. So, coulomb per newton can easily be represented by meter per volt, this is the more useful unit and normally we use this unit meter per volt whenever we define the second order susceptibility. But I for the students I give you a simple problem try to verify that how this coulomb per newton becomes meter per volt. So, this is some sort of classwork or homework for you to figure it out. So, very easy you can I believe all of you can do that ok. So, after figure out after figuring out the dimension of the second order susceptibility, now we go to the next susceptibility next order susceptibility; that means, the third order susceptibility. The procedure is exactly same. So, if I now put third order susceptibility.

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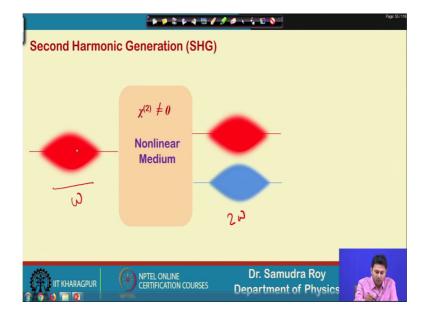


So, my equation is simply P is equal to epsilon 0 chi 3 and then E cube, one we have E cube the equation is exactly the same only thing is that in the previous case it was a square now it should be E cube other things are almost same.

So now this E cube term I just divided square E because the dimension of this quantity we have already figure out, and it comes out to be coulomb per newton and then 1 by E is again coulomb by newton. So, when we have coulomb coulomb per newton and coulomb per newton it should be coulomb square newton square and essentially it will be meter square per volt square. So, if I go on doing this things for higher order susceptibility the kth order susceptibility simply become meter per volt to the power k minus 1. And then where k is 1 2 3 this is general form.

So, this is the way one can easily derive the susceptibility the unit of the susceptibility and normally the value of the second order susceptibility is of the order of same 10 to the power minus 20. So, this is a very, very small value. So, since this order is 10 to the power 20 in terms of meter per volt normally this meter is not used normally it is used picometer divided by volt because the order is so small. So, in case of the chi 3 and all these things also we are going to measure this things or try to find out, what is the value and you will find that this value is very small since the value is very small normally we use picometer or this kind of units ok.

Now, the next thing is if I go back to the slides our next slides is the second harmonic generation.



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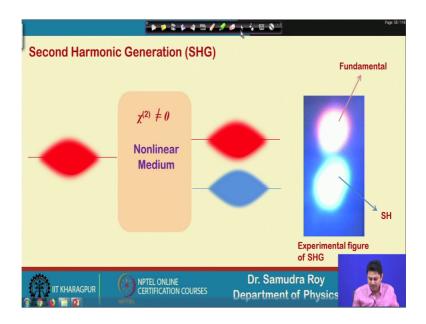
So, this is a very, very important concept today we will going to start that, the second harmonic generation is something where if I launch an electric field of a frequency omega as this animation suggests that I am launching a light which is, I am launching a light which is red here.

And then I it is passing through the non-linear medium where chi 2 is not equal to 0 I am getting 2 light in the output one is red and another is blue here blue representing the

frequency a higher frequency if this frequency say omega. So, this frequency is say 2 omega if this is red and if I increase the frequency with a multiplication of 2 then the colour of light should change and eventually we will have a blue light, where the the frequency of we know that frequency of the blue light is much higher.

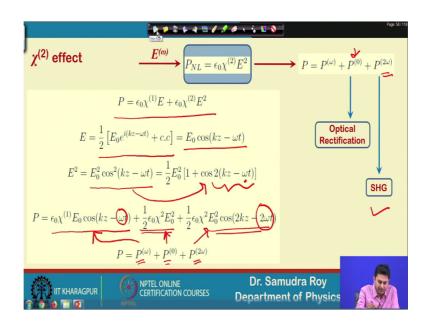
So, this is a this is a just a schematic representation of how the second harmonic generation process is occur a very simplification or simplified diagram we have, but also we have, also we have a experimental figure. So, let me show that the experimental figure.

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So, yeah this is the experimental figure. So, in this experimental figure you can appreciate that we have a spot a ready spot which is the fundamental wave or the wave which is launched to the system. And we are getting something which is blue is in nature.

So, if this is the fundamental wave we write is a fundamental then the other thing is the second harmonic; that means, the frequency is twice than whatever we have. So now, this is the second harmonic generation process.



We now need to understand, how this second harmonic second harmonic generation process can be appreciated through our mathematical form. So, the treatment is very straightforward.

So, let me let me give you the idea how one can do so. Electric filed E is launched when the electric filed E is launched to the system because of the non-linear polarisation we have a representation is square here. So, if the E square term is there. So, what will happen let us see. So, my total polarisation P is epsilon 0 chi E and epsilon 0 chi 2 E square. Total electric field I write this electric field as E 0 E to the power ikz minus omega k plane wave plus the complex conjugate of this plane wave. So, that the total electric field become a real one. So, my total electric field E 0 cos kz minus omega t is the real electric field.

Now, what we will do that I will put this electric field the value of this electric field into the equation. So, when I put this equation I need to put the value of E square. So, E square I calculate if my E is E 0 cos kz minus omega t my E square is E 0 square cos square kz minus omega t. Now this cos square term we can write in different way. So, we know that cos square theta 2 cos square theta is nothing but 1 plus cos 2 theta. So, 1 plus cos 2 theta divided by half is cos square theta. So, I just replace this things here. So, when I replace I will I then have 2 terms one is half  $E \ 0$  square and another is half  $E \ 0$  square multiplied by  $\cos 2$  of kz minus omega omega t.

Now, you can appreciate that this 2 term is here. So, this is the frequency omega and in front of that one 2 term is appearing because of the E square term. So, this 2 term basically gives you the second harmonics. So now, when I put this E square to my final mother equation which is P is equal to epsilon 0 chi 1 E plus epsilon 0 chi 2 E square then it simply gives you the first term which is my E. So, epsilon 0 chi one multiplied by E 0 cos theta and the second term is whatever we have derived here.

So, if I separate out these term and these term you will find in P there are 3 components are there one component is having frequency omega. Another components where the frequency component omega is not there at also this is a 0 frequency component and another components where the frequency component is 2 omega. So, these 3 components are containing 3 different frequencies. So now, if I write my polarisation P so this P p can be represented in 3 part P omega P 0 and P 2 omega.

So, when P omega is there; that means, I am talking about this term having the frequency component omega 0 P 0 is here which gives me no frequency and another component this is a this is a constant kind of term, we are going to understand what is the meaning of constant kind of term and another term which is very, very important inside the polarisation that this polarisation part is vibrating is the frequency 2 omega.

So now here in the right hand side I have listed that which term is giving what the first term P omega is nothing but the representation of the fundamental wave. The fundamental will wave will be represented by this second term which does not contain any kind of frequency that is why it is written P 0 if you look carefully, it is the optical rectification term.

So, this is the average term basically that gives some sort of a rectification it is called the optical rectification and finally, 2 omega. So, this P 2 omega basically generate the second harmonic. So, this is the complete mathematics which is very simple to understand just make E square E is  $\cos E 0 \cos theta$  if I make a square of that and then

divide into 2 part you will readily find there is a 2 omega term that is appearing inside the P.

So, once we have the P where we have 3 different components. So, see this 2 omega component is represented as a second harmonic. It basically leads to the wave that is now vibrating with the frequency double than the fundamental frequency.

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Now, let us understand what is the physics behind it this is a very, very important thing and I should emphasise that we should understand carefully.

So, what happen let us try to understand step by step I am launching an electric field E with a frequency omega. So, if this is my representation of the frequency. So, once we have the representation omega so; that means, this is my frequency. So, this E is launched with the system this system has a non-linear frequency non-linear polarisation. So, when we have a non-linear polarisation the dipole here this is basically representation of the dipole this Euler part this is the dipole this is the dipole. So, when the dipoles are now going to vibrate inside the medium with different frequencies.

So, the vibration of the dipole is basically represented by P. If you remember that P now

have 3 components P omega P 0 and P 2 omega. So, total P is represented by then this box. So, the first term what happen that the dipole will vibrate with the frequency omega because here we have a omega component. When the dipole is vibrating with the frequency omega it will start when the polarisation is vibrating with the frequency omega; that means, the charge particle is vibrating it will start generating a frequency same as the fundamental one that is 2 omega.

The second part it should not have any kind of frequency parts. So, it will not going to vibrate, but it will going to do something that we will going to discuss in maybe in the next class, but let us go to the last term. The last term we have a frequency component 2 omega; that means, the dipole is now going to vibrate with the frequency of 2 omega. When the dipole is vibrating with the frequency 2 omega it now start generating a corresponding electric field that is having a frequency 2 omega. So, here is the new frequency that will going to generate because of the vibration of the electric vibration of the polarisation of the 2 omega.

So now the now if I now summarise the picture so these are the total picture. In this total picture I find that if I launch an electric field it is passing through the a non-linear material. So now, we have a fundamental wave having a frequency omega this is fine, but apart from that we have another wave E omega which is now vibrating of vibrating with a frequency 2 omega. And this E 2 omega is basically the second harmonic. So, this is basically the second harmonic wave. So, this second harmonic wave is generated because the polarisation is now vibrating with a frequency 2 omega.

So, this is the physics of second harmonic very simply I tried an make you understand in a very simple way putting the some diagram so, that you can readily understand exactly what is what is going on. I am launching an electric field because of this non-linear term we have a square term here we called a non-linear polarisation, which is depending on E square and when you put this E square term into the system we find that there are a few terms appearing one is in independent of any frequency we called as optical rectification term.

And most importantly another frequency term is there which is twice of the fundamental

frequency and that gives rise to second harmonic generation. So, with this note let me conclude here in the next class we will learn more about optical rectification and other effects.

So thank you for your attention, see you in the next class.