Laser: Fundamentals and Applications
Prof. Manabendra Chandra
Department of Chemistry
Indian Institute of Technology, Kanpur

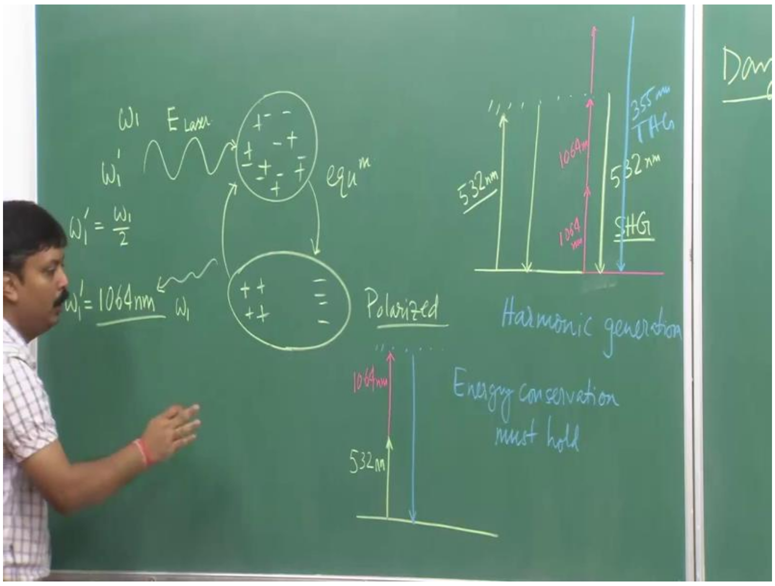
Lecture – 31
Non-linear optical processes

Hello and welcome back. We have been discussing about second order optical nonlinearities particularly second harmonic generation. And in the last class we derived a phenomenological expression for this second order polarization and from there we said you know how we can get second harmonic generation.

We also said that there is necessity to synchronize the phase velocities of fundamental and second harmonic wave in order to have you know decent amount of second harmonic output. We will talk about this you know what is importance or in other word why this phase matching is important in Non-linear optics we will do that in a bit before that we would like to talk about the process s h g.

In the last class we when we derived that expression phenomenological expression for second order polarization, we saw that there is a deceive part and then there is that frequency dependent part and this frequency corresponds to the frequency twice that of the fundamental frequency. Now, what actually happens there physically? So, when intense laser light you know acts on a matter particularly if I take a Non-linear optical material then under equilibrium condition.

(Refer Slide Time: 01:50)



You have a very symmetric Charles distribution. So, this is random Charles distribution everywhere. So, this is very much isotropic.

When a strong laser field acts on this one, this is coming from this one it you know creates a non equilibrium state of this kind of you know Charles distributed state. So, I can just show it by a cartoon where I can distort this Charles distribution like this. So, this is random and equilibriums situation, and this is a non equilibrium distort it is you know condition of the Charles distribution, and this is Polarized right. So, you create an asymmetry in the Charles distribution and when this is very much you know very large then what will happen this is non equilibrium state it will not remain as it is forever.

It will rather quickly would like to this is the state when the photon is acting on this one, and this would like to go back to this state. When it goes back then what will happen it will release the energy in the form of light. Now if I you know if I sent in light of a frequency say omega 1, and if the distortion is not too much that is the movement of the electrons from it is equilibrium position is not too far, then I am going to have omega 1 back in terms of scattering, and this is a linear scattering or Rayleigh scattering that we are aware of very much.

If the distortion is really large when the field is extremely large then it can. So, happen that you know energy that is coming back is much higher than my input. I can show that using an energy diagram, this is my ground state and now if I just give a large amount of photon material is forced to take up that photon, and it go somewhere we normally we topic that by you know dashed line we call it a virtual state.

And because it will not stay in this non equilibrium state for longer time it will revert back to it isotropic condition which is the stable equilibrium condition and the result is the getting back the same frequency of light this is linear scattering. Now if my intensities very high what we can have suppose I come with a frequency omega 1 prime. Such that omega 1 prime is equal to omega 1 by 2 just for simplicity I could take like omega 2 omega 3 does not matter.

Now, this is my omega 1 prime and I will give you certain number suppose this one was say 532 nanometer light good choice it is a green color it is a 532 nanometer light, and I get back my 532 nanometer light in terms of linear scattering or Rayleigh scattering. Now if I have another wavelength which is having half of the frequency; that means twice that of the wavelength.

Essentially omega 1 prime is 1064 nanometer that is very easy to calculate if you know the conversion of frequency and wavelength. Now, what I am going to do I am going to allow the you know material to interact with 1064 nanometer light that is my omega 1 prime and having very you know large electric field, coming from a strong laser beam.

So, if I write somewhere here, this guy will take it here this is my 1064 nanometer write. Now the if electric field is very strong, while one photon takes this molecule up here, but there is possibly no real state, there are too many photons there, which are you know ready to interact with the material even at that condition. Because of large density of photon that can force a system from here to take up a photon further obtain 64 nanometer and go up here.

Once it is goes here it has rest such a condition. Again it will it would like to come back to this condition which refers to the ground state. So, from here it will come back to the ground state directly here now this corresponds to this amount of energy which refers to 532 nanometer.

I have used 21064 nanometer light from in intense laser light, and force a molecule to take 2 of these photons to go to some polarized state, and then scatter back the total energy that it consumed which is 2 photons of 1064 nanometer essentially which is equal to 532 nanometer.

This particular process that I just showed you is known as second harmonic generation Now I could and it is done that if I have sufficient you know photon density from a high intensity laser it will it cannot may not stop here. In some cases it can take another one photon here and it can come back, there it will come back with a photon which is higher than even 532 nanometer.

This energy if it comes from this place this will corresponds to 355 nanometer approximately and this is known as third harmonic generation, you can imagine if would have taken 4 photon successively and jump back to this ground state it would emit you know photon whose frequency would be 4 omega 1 prime, and that would equate to 266 nanometer and that is known as fourth harmonic for this particular 1064 nanometer fundamental light.

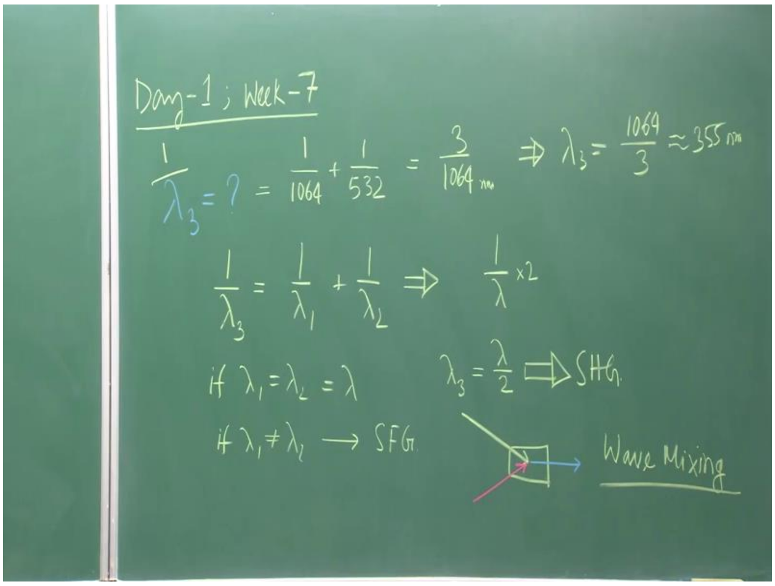
This is the pictorial representation of this second harmonic generation, or third harmonic generation, or fourth harmonic generation so, on. So, in general this is the picture for harmonic generation process. This is specific process in second order amount the second order non-linear optical processes. Now if I have a situation where I come up with a 2 photons 2 incident photon having 2 different frequencies, what we may have we may have the following situation. So, far we have shown like all the photons are of equal frequency.

Now if I have say one photon of 532 nanometer, and then another photon of 1064 nanometer the lengths are not to be scaled. I still have my laser source; I can force my material to take up the photons. So, if we take one 532 nanometer photon and then another 1064 nanometer photon from there. So, it will go up here and then it will release a photon of the energy which is equal to the sum of both of this photons right.

Because the Energy must be conserved right, Energy Conservation must hold. So, I can say that if suppose this is my omega 1 this is my omega 2, do not confuse with this terminology anymore. If this is my omega 1, and this is omega 2, and this one is something omega 3, then my omega 3 must be equal to omega 1, plus omega 2.

I must have the energy conservation for this particular process. So, at this point if I ask you I have used 2 photons having frequencies 532 nanometer and 1064 nanometer, they have someway mixed in the material and generated a third photon having omega 3 frequency. So, corresponding wavelength, this is I should correct this one this is actually lambda 1, and lambda 2. I can have the corresponding omega 2 and omega 1 of course, and this have lambda 3. So, I have this lambda 1 equals to 532 lambda 2 equals to 1064 what is my lambda 3.

(Refer Slide Time: 14:00)



So, my question is lambda 3 is what how do I find out that is very easy to do. I know this equation from energy conservation. So, this is my unknown, how to do that it is 1 by lambda 3 equals, to 1 by lambda 1, plus 1 by lambda 2, lambda 1, and lambda 2 my incident photons lambda 3 is the photon that I am getting out of this non-linear process.

Which is again a second order non-linear optical process because I am using 2 different photons to get a third single photon; now, you can immediately check this I am talking about 2 different frequencies. If my lambda 1 and lambda 2, if lambda 1 equals to lambda 2 equals to say some lambda then what will be the process then this process will lead to 1 by lambda into 2 right.

Essentially my lambda 3 is equal to lambda by 2. So, lambda by 2 means essentially twice the frequency so; that means, that is a second harmonic process. This will give me second harmonic generation when lambda 1 equals to lambda 2.

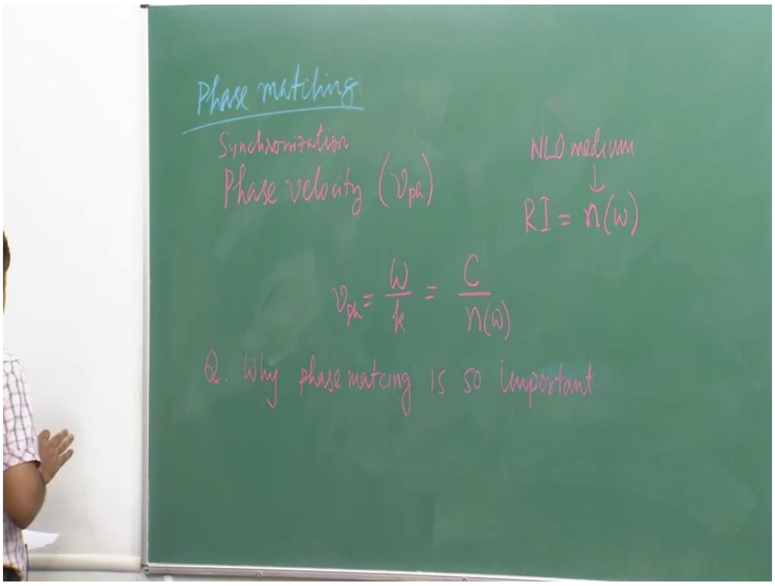
When lambda 1 and lambda 2 are different this particular process is known as sum frequency generation. So, in general if lambda 1 is not equal to lambda 2 this one will give me sum frequency generation. And in this way we can calculate what is the frequency of this particular photon that is what is lambda 3. So, lambda 3 will be equal to one by 1064 plus one by 532 right.

So, essentially that will be 3 by 1064 nanometer right that is 1 by lambda 3 equal to therefore, lambda 3 is equal to 1064 by 3 equals to roughly equals to 355 nanometer. And this process is also known as wave mixing why because you are having total 3 different photons here we are taking the second order process only. So, 3 different frequencies they are as if being mix. So, I can show it pictorially that this is suppose a non-linear optical crystal and one light is coming here, another light is coming here, and the third light is coming out.

They are being mixed here and this is why this is also known as wave mixing, wave mixing has tremendous importance in various different fields starting from physics, chemistry, spectroscopy, material science, and you know in electrical engineering everywhere you will find applications of this process of wave mixing which is a non-linear optical phenomena all right.

Let us now start our discussion on phase matching. So, we said that we need a synchronization of the phase velocities.

(Refer Slide Time: 18:33)

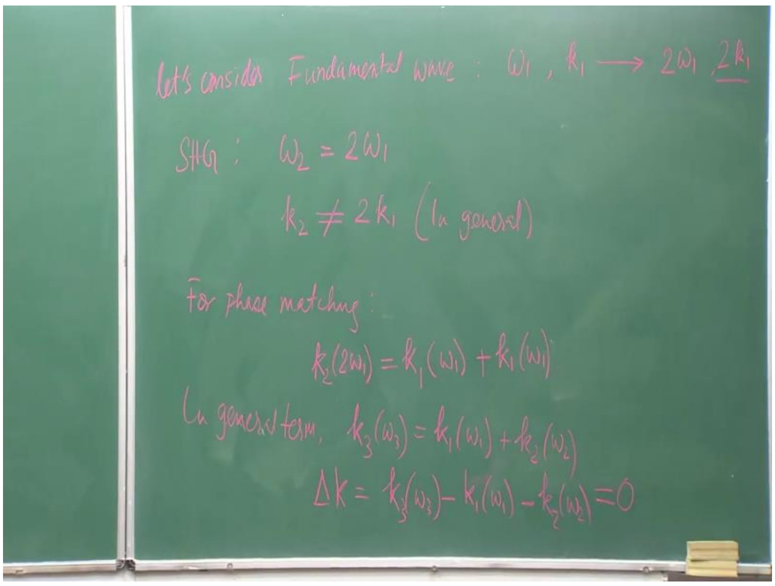


So, we said that we need the synchronization of the phase velocities corresponding to the fundamental and the harmonic wave. Now what is Phase Velocity; Phase Matching is essentially the synchronization of. So, we have to match their phase velocities phase velocity is the velocity of an of a light wave, which is essentially a plane wave when it propagates through a medium correct. And how and this medium generally we characterize, characterized by the refractive index.

So, if I take an NLO medium through which the various propagating and we are considering a velocity as phase velocity. This now NLO medium is characterized by reflective index RI equals to n. And this reflective index in general is frequency dependence, and how is this phase velocity related to this refractive index. So, the thing that we know is this where omega is the frequency, and k is the wave vector of the light that we are considering of a plane wave and this we can write in terms of the velocity of light and the refractive index n omega.

At this point the important question to ask is the why phase matching is, important we have been asking we need to have phase velocity phase velocity phase matching. So, why is it? Important to get this phase matching. So, at this point we will consider a fundamental wave having a frequency omega 1 and a wave vector k 1.

(Refer Slide Time: 21:48)



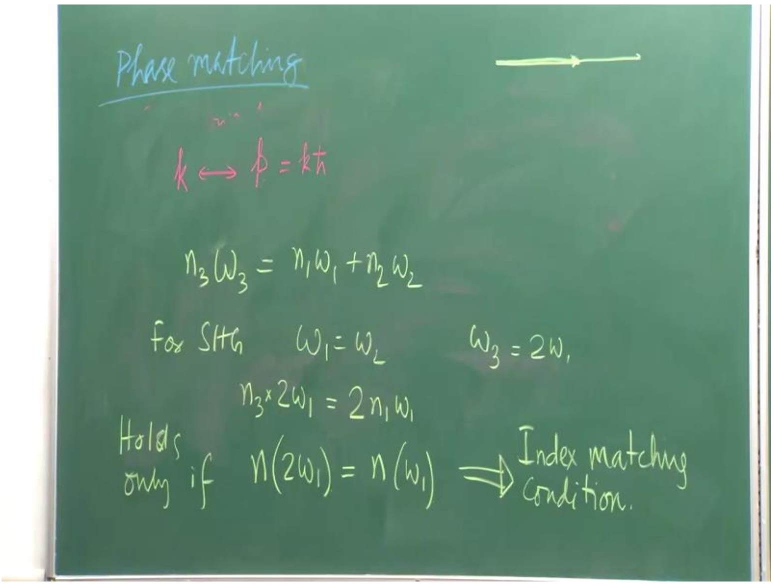
So, the answer that let us consider fundamental wave. So, many cases in books we will also see this is called a polarizing wave. So, they are same , the particular frequency that is used to induce non-linear polarization is known as polarizing wave or fundamental wave. And this fundamental wave is characterized by frequency omega 1 and wave vector k 1 what it will do this fundamental wave will induced polarization at the second harmonic frequency which is equal to 2 omega 1 right. So, this will lead to you know polarization created at frequency 2 omega 1 and corresponding phase velocity will be 2 k 1 right.

Now, the SHG 23.19 that will be created that will be characterized by it is frequency omega 2 and wave vector k 2. So, for SHG process it has to be that omega 2 equals to 2 omega 1 is bound to happen. And this wave vector k 2 it may not be equal to this 2k1.

Generally actually this not equal to 2k1 this is in general. Now, if that is true then how do I achieve the phase matching? So, the phase matching will take place what do we need is I can write as k 2, which is function of frequency 2 omega 1 is a sum of k 1 omega 1 and another photon having the same value right.

This is what is needed to be fulfilled if I really want to have a phase matching you know if I want to have the SHG to occur, and if I write it in general term in general term this I can write k 3 omega 3 equals to k 1 omega 1 plus k 2 omega 2. When I am not certain if it is you know omega 1 equals to omega 2. This is most general form and I can rewrite this one in terms of in term of delta k which is equal to k 3 omega 3 minus this 2 quantities that is k 1 omega 1 minus k 2 omega 2 and this delta k must be 0. So, this is the condition to achieve phase matching in a non-linear optical medium upon interacting with the intense laser light.

(Refer Slide Time: 26:45)



Now, interestingly this wave vector is related to the momentum right because k is related to momentum p which is k h cut correct. So, p is equals to k h cut, this relation you know simply tells me that the energy exchange here essentially what I am having is an energy exchange between different photons right you know coming from fundamental omega 1 2 the second harmonic 2 omega 1 there is an energy exchange.

This energy exchange is between these waves are possible only when I have this fulfilled or in other word my law of conservation of momentum is intact. So, law of conservation of momentum holds. If I consider this process of wave mixing happening in a collinear geometric that is my wavelength 1 is coming in the same direction this 1 is also happening in this 1 and the outgoing light is going this direction. So, in that case what we can write in terms of the refractive index and the frequency following this general relation and we know how this omega k and the refractive index is related. So, knowing that I can write sorry; omega 3 equals to n 1 omega 1 plus n 2 omega 2 right.

Now for SHG I have to have omega 1 equals to omega 2, and not only that I should also have omega 3 equals to 2 omega 1 then. If I put that it is in 3 into 2 omega 1 equals to n 1 omega 1 plus n 1 omega n 2 omega 1 right. So, n 2 also here will be n 1 because I am considering it was same frequency. So, for same frequency it will phase the same refractive index. Essentially it will be 2 n 1 omega 1.

Now this is true only when the n I can write in terms of this one this is a, n 3 is to start with n 3 is a function of omega 3 omega 1 is a function of n 1 is function of omega 1 n 2 is the function of omega 2. This is only possible when I have this condition fulfilled and this holds only if n function of 2 omega 1 is equal to n omega 1, is known as index matching condition and also known as wave vector matching condition.

Now in general this condition is not fulfilled because I have 2 different frequencies and that due to dispersion these 2 frequencies will have different refractive indexes. So, there are certain ways to fulfill that we will look at that in the following class.

Thank you very much.