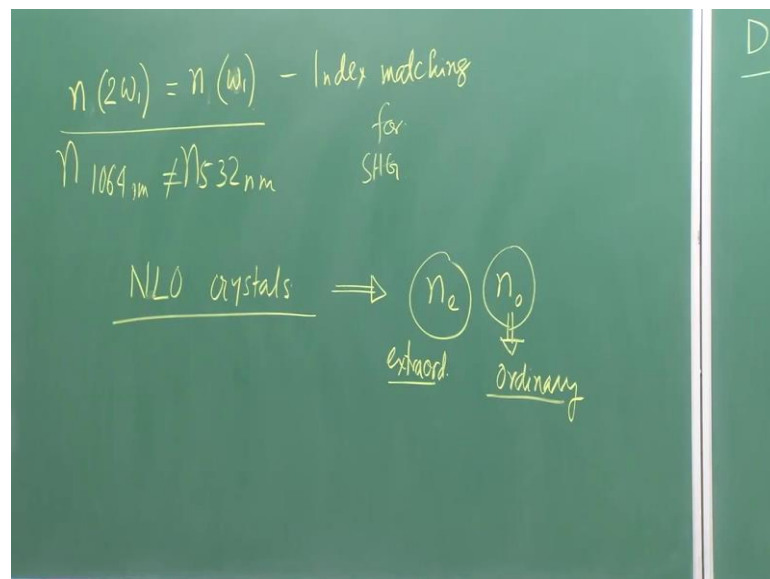


Laser: Fundamentals and Application
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Lecture – 32
Aspects of SHG and Application of non-linear optics

Hello and welcome back, so in the last class we will looking at the phase matching conditions, and we found the related index matching condition. So, the index matching condition for SHG was like this.

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So, specifically we can also remove this n_2 n_1 n_3 . So, essentially this means that the refractive index of the harmonic wave and the fundamental light should be same, in order to have phase matching. So, this is this was my index matching rate condition for SHG.

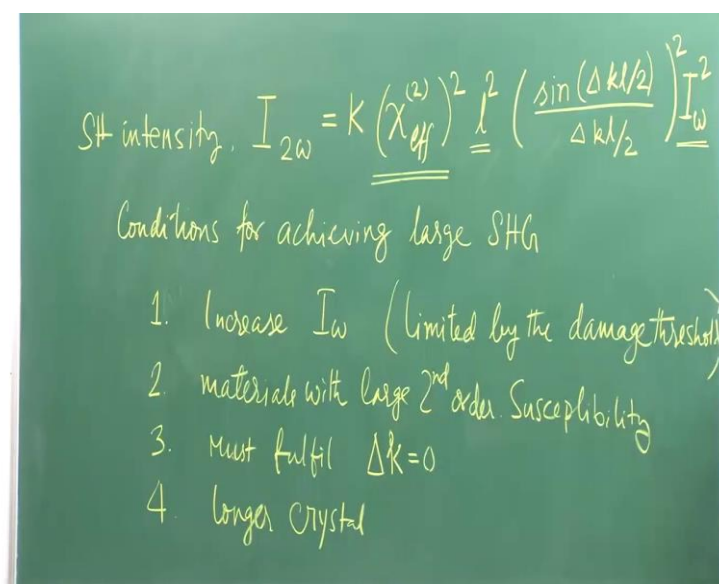
Now, in generally you can understand that this is not possible right, normally because the suppose I have an incident wave of frequency 1064 nanometer, than the double the frequency we will have an wavelength of 532 nanometer. So, there is wide gap in the frequency. So, there is something called dispersion right. So, which is strongly wavelength dependent property? So, in that case the refractive index of say 1064 nanometer and that of 536 nanometer they are not going to be same.

So, if you want to have SHG you need to have this concern fulfilled and you know that this is difficult so there must be some way out. So, the way out is to use a birefringent crystal. So, you need non-linear optic crystals, particularly we will which will be birefringent meaning that they will have two different refractive indices. So, these birefringent crystals they will have two diffractive indexes as n you know they are described as n o and n e, this n o stands for ordinary and this stands for extra ordinary ok.

So, this extra ordinary light we will pass the fundamental light, sorry ordinary refractive index we will take care of the fundamental wave while the other refractive index the you know extra ordinary refractive index we will take care of the harmonic wave . So, actually what you have to do, you have to first identify the optical axis through which the light will pass through without any kind of change. So, you have to identify the optic axis of the crystal that by refringent crystal or analo crystal that we are talking about, and you have to send the incident field at a particular angle with respect to the optic axis of the crystal and this particular angle is dependent on the material that we are using.

So, in that case you can ultimately achieve this condition and you can have SHG process, with lot of you know power, lot of intensity now we will look into one particular or we will look at the ways to enhance the second harmonic intensity. So, I will just write down one expression for second harmonic intensity, so second harmonic intensity.

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SH intensity, $I_{2\omega} = K (\chi_{eff}^{(2)})^2 L^2 \left(\frac{\sin(\Delta k L/2)}{\Delta k L/2} \right)^2 I_{\omega}^2$

Conditions for achieving large SHG

1. Increase I_{ω} (limited by the damage threshold)
2. materials with large 2nd order Susceptibility
3. must fulfil $\Delta k = 0$
4. longer crystal

Which we denote as $I_2 \omega$, and this is given by I will give you that expression and one doesn't have to remember this one, this is second order susceptibility, the effective susceptibility multiplied by the square of the path length of the fundamental light through the crystal, multiplied by the square of the incident photon that is we write as $I \omega$ and square.

So, this is the expression for the second harmonic intensity through a non-linear optic crystal, and from here what are the conditions that we can find out that will help maximizing the second harmonic generation. So, what are they so first you can think about this quantity, so second harmonic intensity depends, the square of the incident power? So, you can imagine that, if I increase this power, I can increase the SHG intensity. So, first place, so conditions for achieving large SHG from this formula, so one is increase incident power.

Now, that is very obvious from this equation, but there are something to note I cannot continue increase in the fundamental laser power, why? Because this laser power is beam is intended and if I increase the intensity of this laser beam too much, it can actually damage the material. So, this increase we will suddenly increase the SH power, but up to a certain limit. So, this is limited I cannot go on increasing the instant power infinitely, so limited by, so called damage threshold of the crystal.

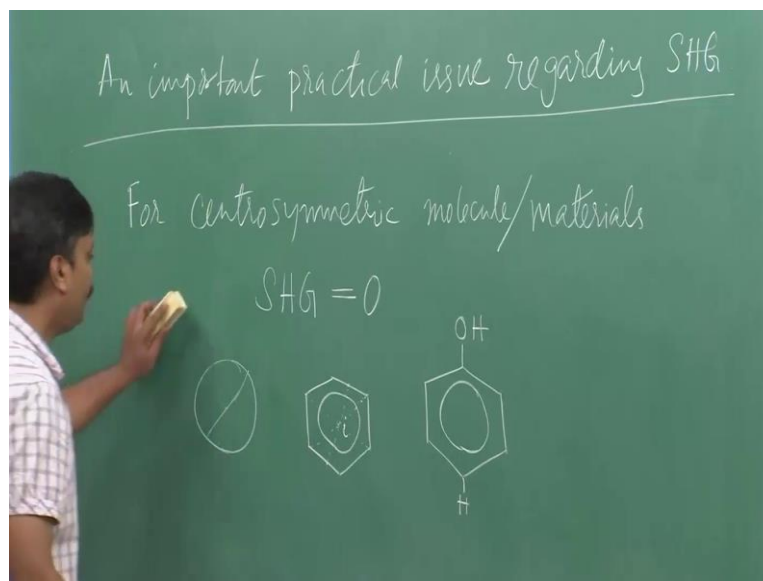
So, below damage threshold, damage threshold is that optimum power beyond which the crystal will get damage or the material will get damage. So, I have to be always below that power. So, below that up to this damage threshold I can keep on increasing the incident power and we can get high SH power ok.

So, the second thing that we can think of the quantity is this one for a given particular material this is fixed, now if I am searching for some material, which will give me very high second harmonic generation then one condition is that it should very high second order susceptibility, if I go for microscopic scale, if I am looking microscopically then it should have very high value for the second order polarizability ok.

So, we need material with large second order susceptibility. So, this need actually has opened of huge field of research, while lot of chemists or actively and physics is are actively involved in preparing material designing and preparing material that may and that will have large second order susceptibility.

And next is we must, that material must fulfill, the condition for phase matching Δk equal to zero, and what is my forth variable that I can play with is this quantity, that is l which is the path length through the crystal. So, if I have a l larger; obviously, $I 2\omega$ also will be larger. So, longer crystal will increase the second harmonic generation more and more.

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So, these are pretty much the ways to increase enhance the second harmonic generation capabilities now we will consider a few practical issues about second harmonic generation regarding SHG. So, what is this particular important thing that we should know, it is that for Centro symmetric molecule or material SHG 0, under electric dipole approximation show all this formalism that we are looking at they are under electric dipole approximation.

So, if we do not say specifically, everything is on the electric dipole approximation, but you should know that there are formalism based on electric quadruple or electric what (Refer Time: 13:08) or magnetic dipole are possible, here we are not going to discuss about that. So, on the electric dipole approximation SHG is 0 for Centro symmetric molecular materials, so for those who do not know, what is Centro symmetric molecule or material? Let me just you know state that quite briefly, so let us take molecule.

So, this is benzene and says this is faunal. So, out of these two molecules, this molecule is known as Centro symmetric molecule while this is non-Centro symmetric, why?

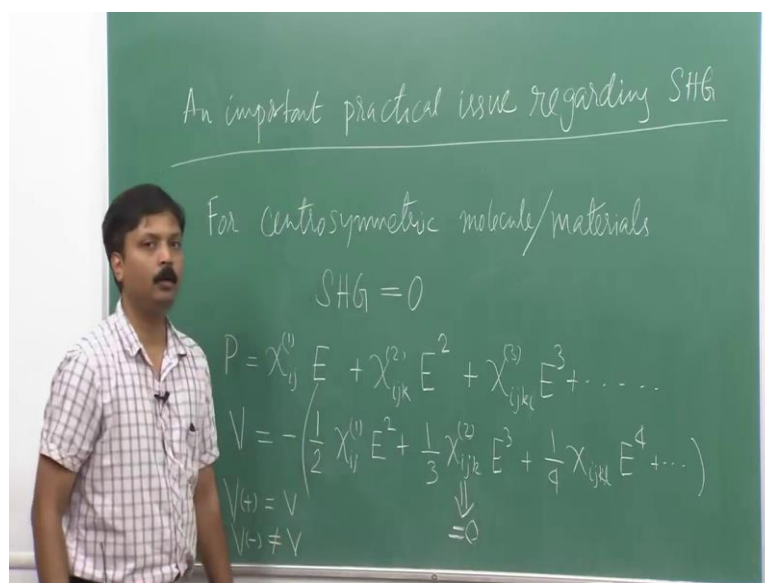
Because for this molecular structure there is a symmetry element known as centre of inversion or I, such that if I start from any part of the molecule passed through that particular point and go an equal amount I will find exactly identical point like from where we have started. So, suppose I start from this particular region and it pass through this one I reach an equivalent point. So, the use of the word identical is not completely correct, I must say this is an equivalent point ok.

So, that is true for any point that will start with and pass through this one I reach an equivalent point this is true for this particular point also. So, if I pass through this end point I get another you know similar kind of point or equivalent point. So, that is why whenever any molecule or material, if I take for example, as sphere. So, if I take a sphere, so doesn't matter from where I start, if I pass through its centre and what equal amount I will reach an equivalent point.

So, this kind of molecular material which has the centre of inversion is known as Centrosymmetric molecule, for material which doesn't have a centre of inversion its non-Centrosymmetric for example, this molecule. So, this molecule if I start from here where there is a hydrogen I will not reach the in another liquid hydrogen, here instead I will see an OH group. So, this is not a Centrosymmetric molecule.

So, now what is so special about Centrosymmetric molecule that we do not observe second harmonic generation from that, so you can easily find that.

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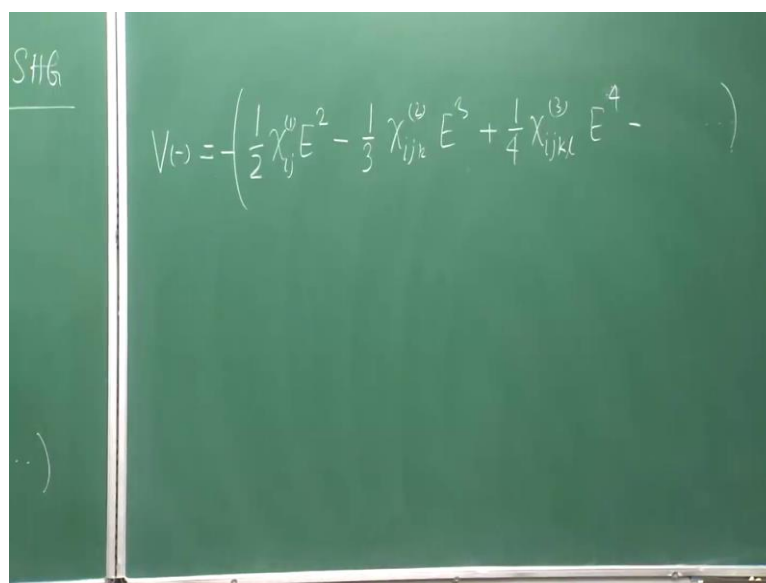
If you look at the polarization, so in general, so polarization is given by χ_{ij} . So, for a moment if I for simplicity remove this subscript, then I can write it this is written quite in a much simpler form. So, now, consider a Centro symmetric molecule, for a Centro symmetric molecule, if I you know look from any side it should be identical very similar to you know the condition where we look at it from a different angle or different position.

So, now if I come with an electric field and you know part of the electronic distribution, so it will get polarized. So, for a Centro symmetric molecule if I look at the potential energy due to this you knows electronic disturbance created by a laser field will be the same irrespective of which way the electric field is acting on. So, for example, if I say I am considering a particular direction say x . So, I can come either from plus x direction or negative x direction, so for that so for say I look at the potential energy for this one. So, how we can write it, so it will be like a half χ_{ij} and so, on with a negative sign on this one.

Now, if I come with an electric field from plus x direction what will happen say all these are positive. So, if I considered V plus that will be equal to this v ; now if I have my electric field coming from positive direction that is my minus E . So, all this is E^2 or E^4 , E^8 , this terms will be un altered while this e is having odd power will become negative.

So, in that case why V minus is not equal to v , right this is easy to understand. So, if I write explicitly.

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$$V(-) = - \left(\frac{1}{2} \chi_{ij}^{(2)} E^2 - \frac{1}{3} \chi_{ijk}^{(3)} E^3 + \frac{1}{4} \chi_{ijkl}^{(4)} E^4 - \dots \right)$$

So, V plus is exactly this and my V minus will be minus half χ E square, then minus 1/3 χ E cube and again plus a minus, other quantities and it will keep going. So, this is definitely not equal to V plus. So, therefore, I will get two different values of the potential energy, when I use whether E or minus E .

Now, if we are considering Centro symmetric molecule. So, it doesn't matter, what is the you know direction of polarization the potential energy is also should be very symmetric, it should be an even function, but if I look at the expression of potential energy here, I see that that is not the case. So, this totally in contradiction the contradiction will be you know removed if and only if this quantity is equated to 0; that means, χ_{ijk} , which is associated with the second order process, if it is 0, then in the potentialized term all the odd power of electric field will be vanished.

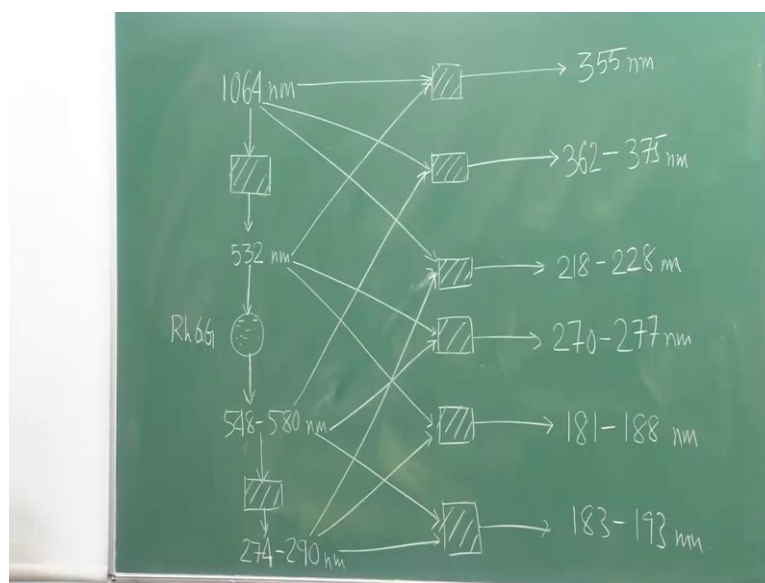
Let me correct it slightly, I talked about only for second order process it is valid for any even order process. So, if I considered the next even order process that is E four or E six. So, those susceptibilities connected to those even order E terms, should vanish for Centro symmetric molecule, in order to avoid the violation of this case. So, this is the region we take χ_2 equals to 0, for Centro symmetric molecule and this is true for all even order non-linear process.

So, if you consider a Centro symmetric molecule on the other hand. So, what will be the you know case there of course, there is no burden on the potential energy to be identical for different electric fields.

So, if I take plus E or minus E, it is not necessary that the potential energy will be different, because if when the electron you know oscillates in one direction, in case of non-Centro symmetric molecule this oscillation doesn't have to be the identical for in non-symmetric molecule, when we come with two different electric field E and minus E. So, that is why in general for Centro symmetric molecule harmonic generation is 0 because the χ^2 is 0.

Let us quickly have a look at the capabilities of these second order non-linear optical processes, to generate various different wavelengths. So, suppose I start with 1064 nanometer ok.

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And if I sent is light to through a crystal, which is a non-linear optical crystal then what we it will do, it will give 532 nanometer due to SHG, now if I same this 532 nanometer through die solution. So, say or example I take rho Damien 6 G, then what will happen it will generate several possible laser wavelength because die laser can you know it gives large emission band and thereby you can you know tuned laser wavelength selecting particular wavelength within that emission bandwidth.

So, this can give rise to a wavelength range, I will give you say 548 nanometer, to 580 nanometer, this is a typical wavelength range that you can get for a Nd:YAG laser, when this is pumped with 532 nanometer, light which again is generated from the SHG, and any wavelength from this range if it is again passed through a non-linear optical crystal, then it will give you another range say 270 to 290 nanometer. So, you can see that you know various wavelengths you can generate.

Now, this is not everything, so if I suppose have another crystal here again and if I send this light, and if I also send this light, they will mix there within the non-linear medium and then I will get another output and you should be able to guess what output it will give, in the last class we mentioned about this one and this should give you approximately 355 nanometer right.

Now, if I put another crystal, here and I mix this 1064 nanometer, and wavelengths from this range I can select any one wavelength from this range, and I can mix it here of course, I can get several different wavelengths depending on which one I am choosing out of this range, and I can get in that way range of wavelengths laser wavelengths which is 362 nanometer to 375 nanometer, alright and if I take another crystal here and if I allow the mixing of 1064 nanometer, and sorry this here then again I can generate huge range which is 218 to 228 nanometer and that is you can see is an ultraviolet region ok.

And if I combined 532 nanometer and this die outputs. So, in a non-linear optic crystal I can again get a range of 270 to 290 nanometer again in the UV, if I take another crystal and if I mix 532 and 532 nanometer, and this output 274 to 290 nanometer, then I can actually generate very unique set of wavelengths, this is below 200 nanometer and it falls in the vacuum ultraviolet region, which is really otherwise difficult to get and one needs to take proper care of this particular wavelengths, because it is observed by pretty much everything ok.

So, also we can you know combined this to die outputs in a non-linear optical crystal and I can get another set of wavelength in the vacuum ultraviolet like this. So, you can see in this whole chart that utilizing, the second harmonic process or other some frequency generation process, I can create. So, many different wavelengths and by you know selecting proper non-linear optical crystal or other medium I can get a highly efficient

second harmonic generation or some frequency generation. So, by which I can create coherent lights of quite narrowband and with high power ok.

So, these are few you know important thing that you should you know look at, and also you should be able to you know appreciate this table, by choosing a particular wavelength and finding is part are combining them and then calculating what will be the resultant you know frequency.

So, if you can take any know you know laser, so I have used 1064 nanometer, which is a fundamental wavelength of into yag laser in the inferred, you can take any other laser say for example, ti sapphire laser, you know you can take you know for the matter (Refer Time: 32:01) 4 and select the wavelength and you will be seeing that how you can use non-linear optics and you can generate various different others type of wavelengths.

And I was saying that this is not the only advantage of this non-linear optics that you can make different wavelengths, but you can use it in various other processes. So, there are vast application of non-linear optics, and second order non-linear optics is simplest yes the most useful one of the you know immediate application, I can tell you it is used to probe the you know pulse with of at a very short or ultra short pulse laser ok.

So, now what I am going to talk about is well application of this you know non-linear optical process, particularly the second harmonic generation. So, this non-linear optics of course, you know gives you the ability to generate various different frequencies having the properties of laser light that is coherent, narrowband, high peak power, everything high average power. So, that is one of the you know finest application of course, of lasers in non-linear optics.

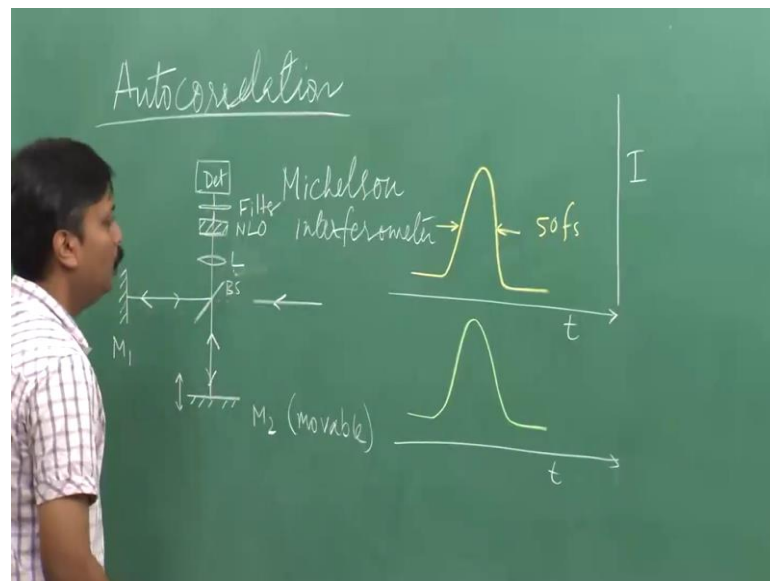
Apart from that there are so many other you know applications of the non-linear optical properties, which is driven by laser. So, I will sight you one example of that one. So, we discussed about ti sapphire laser which is you know laser having ultra-short positive reaction.

Now, how short those pulses maybe they are in the order of 10 power minus 15 second. So, it may be like you know 10 femto second, it may be you know 50 femto second, 100 femto second, now this numbers are really, really small and if you have a little bit of

knowledge about electronics, you know that it is not possible for any electronic device to record information in that short period.

If you consider say a nanosecond pulse is 10^9 times longer than a femtosecond pulse duration, then you can measure it, but not a femtosecond pulse. So, there we need to think a little differently in order to measure them.

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So, how it is done, this is done by using something called autocorrelation. So, what does autocorrelation mean? I will try to be quite brief in this part; you have definite pulse duration for a single pulse. So, let me use two different colours, so that it is quite visible. So, laser we will have this pulse duration and suppose, I am saying that this is something like a 50 femto second pulse, and from the same laser I can have another pulse also, so which should be identical to the first pulse.

Now, if we can allow 2 pulses from the same laser to interact with each other, I may get some information about the pulse duration, how that is what I am going to show you. So, now, we have learnt about the process SHG, where 2 photons of same frequency interacts with each other within a non-linear medium and generate a new third frequency having double frequency or half of the wavelength, that is my second harmonic generation process.

So, if I can put 2 pulses from the same laser, on two different arms somehow and allow them to interact in a non-linear medium as a function of their time delay. So, the matter will be clear, if I can give a schematic. So, the schematic is as follows, so suppose I have a laser light coming in, in this direction. Now if I put an optic, which is known as beam splitter meaning it will split the beam into 50/50 %, how this side it will reflect in this direction and 50 % will transmit in this direction alright.

So, what I have created, I have created 2 beams out of a single beam having 50 % of intensity, now if I put 1 mirror here I call it M1, and then I put a second mirror here, let me call it M2 out of these 2 mirrors, if my M2 is movable, meaning it can be translated back and forth.

So, now these 2 lights, which I will split here they will go hit the mirror and again come back and meet here. So, this light after meeting here it will go in this direction and this light of course, it will transmit through this and go in this direction alright. So, this double-headed arrow indicates that light goes and comes back. So, this geometry is known as interferometer, Michelson interferometer many of you already know about this one.

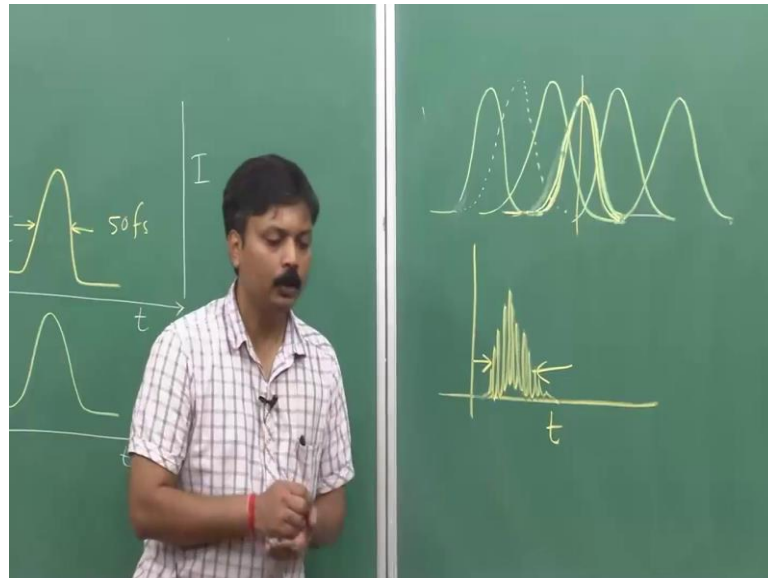
So, this will lead to interference constructive or destructive depending on the relative distance between the 2 beams which are meeting here in time that is the amount of path that they know each arm is having depending on that there will be a phase lag between those 2 beams and I will have the corresponding interference.

Now, if I put if I focus the beam using a lens. So, this is my beam split here and this is my lens here and then I could non-linear crystal NLO crystal, and then I put particular filter which will take only the second harmonic light. So, this is filter and then ultimately it will go to some detector. So, in this case what will happen these 2 lights will go and it will be focused into the non-linear crystal and then it will generate the second harmonic light, fundamental light is also their fundamental light will be filtered here and the only the second harmonic light will come here alright.

So, now for second harmonic generation, so this I showed this is a pulse with rate so; that means, in this axis I have intensity the number of photons. So, second harmonic generation or any other non-linear process takes place; when these 2 or more photons then are mixed in within a non-linear medium. So, the efficiency of the process will

depend on how many photons are taking part there; now let us look at this process very clearly.

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So, to start with I have 1 pulse going to the detector, now I have moved this mirror such that the second pulse is totally outside this envelope; that means, 2 pulses are not interacting. So, there is no mixing between this and this pulse if the situation is like this fine, now if I move this mirror toward this one. So, in the next step this will now come here, right if I move it further it will go like this if I go even further at some pointer time they will overlap and then we move further it moves out and then totally it go out.

So, when this 2 pulses are quite a part there is no SHG, due to the mixing of those 2 pulses, there may be background SHG the movement if starts overlapping with this white one. So, let me differentiate this one earlier we are using this yellow. So, this is pretty much overlapped. So, when this green pulse is totally overlapped then there will be maximum SHG, and when it will pass this pulse there will be very little SHG. So, if I follow the SHG intensity as a function of time, which is coming from this delay that this mirror can, mirror movement can give us then I will see, I will see the intensity pattern like this.

So, essentially this green pulse will map the overall you know trace of the yellow pulse correct, because as it goes from this tail to the other tail it is producing SHG, and for

each part I am getting the SHG intensity maximum being when they are overlapped. So, in that way I get my time with all the pulse.

So, for the autocorrelation pulse with one can actually go and find out what is the exact pulse with of the laser knowing, the you know the particular shape of the pulse. So, what was really difficult to achieve using an electronic device, using second harmonic generation technique and allowing the autocorrelation of 2 pulses by creating a geometric michelson interferometer one can trace the pulse with of a very short pulse or ultra-short pulse laser ok.

So, there as I said there are many, many, many other such techniques such applications of these non-linear optical processes are there. So, we may not be able to discuss about all those things in the short span of light, but whatever we have showed here they are quite basic yet extremely important application of process like SHG.

Thank you for your attention.