Nanobiophotonics: Touching Our Daily Life
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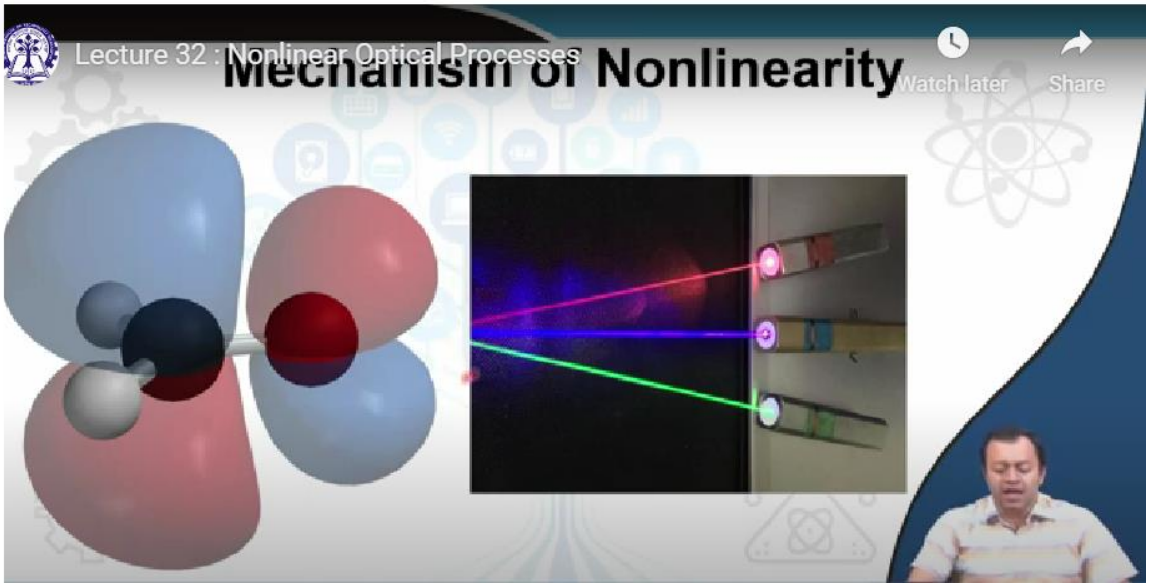
Lecture No. 32
Nonlinear Optical Processes

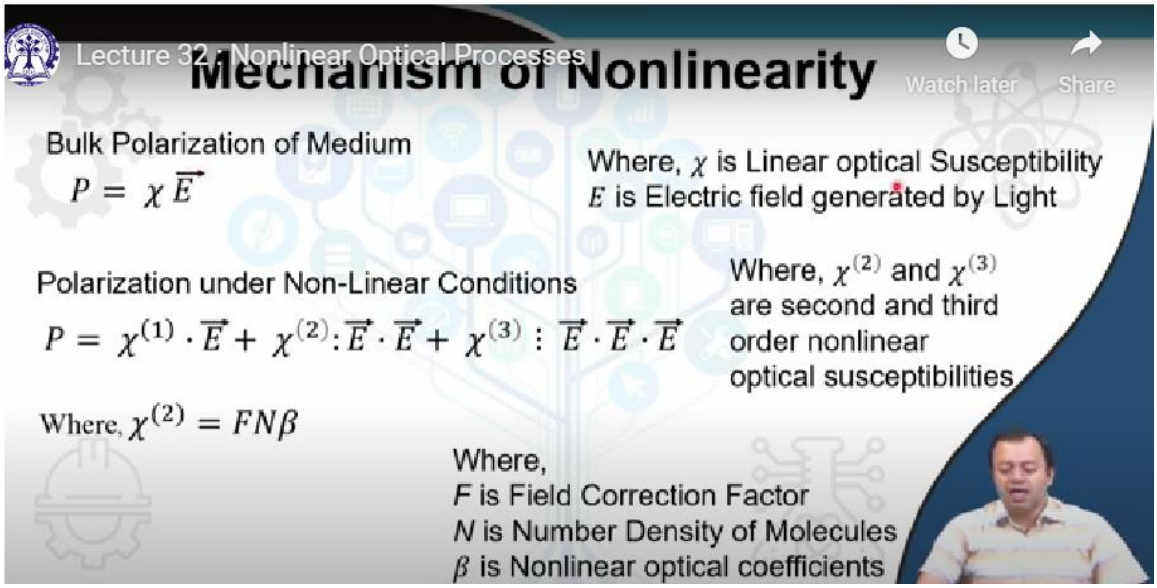
Welcome back. We are still discussing about lasers and today we are going to discuss in this particular topic a very interesting phenomena that again theoretically was known for a long period of time, but by the advent of laser and by finding out some exotic material exotic minerals got into prominence. So, today I am going to discuss the non-linear optical processes ok. This is part of the module lasers for biophotonics.



So, in complex systems you know that when there are two complicated atoms connected with other not so complicated atoms by complicated I mean higher elements higher atomic number masses compared with lower atomic number masses metal organic frameworks or some kind of ah say complex compound with their electron cloud. So, this is the electron cloud this is the electron cloud this electron cloud is overlapping in a specific specific manner so that there is a stability into the system so that the attractive forces win so that the ah atom could combine to form a compound ah an element forms compound the atomic elements forms compound forms molecules and then the electron have enough space to move around and thereby overall the energy gets reduced.

What happens when you subject it to intense photons intense source of light of large number of photons of a coherence a specific energy specific source specific wavelength

right. We start seeing the concept of the effect of non-linearity we start seeing the effect of non-linearity. Now, you will ask what exactly is non-linearity? Non-linearity simply means the rearrangement the rearrangement of the electron clouds the rearrangement of the electron cloud under the interaction upon the interaction by intense electromagnetic field produced by laser light. So, laser light has an electromagnetic field the electron moving has a particular charge particle produce electromagnetic fields.

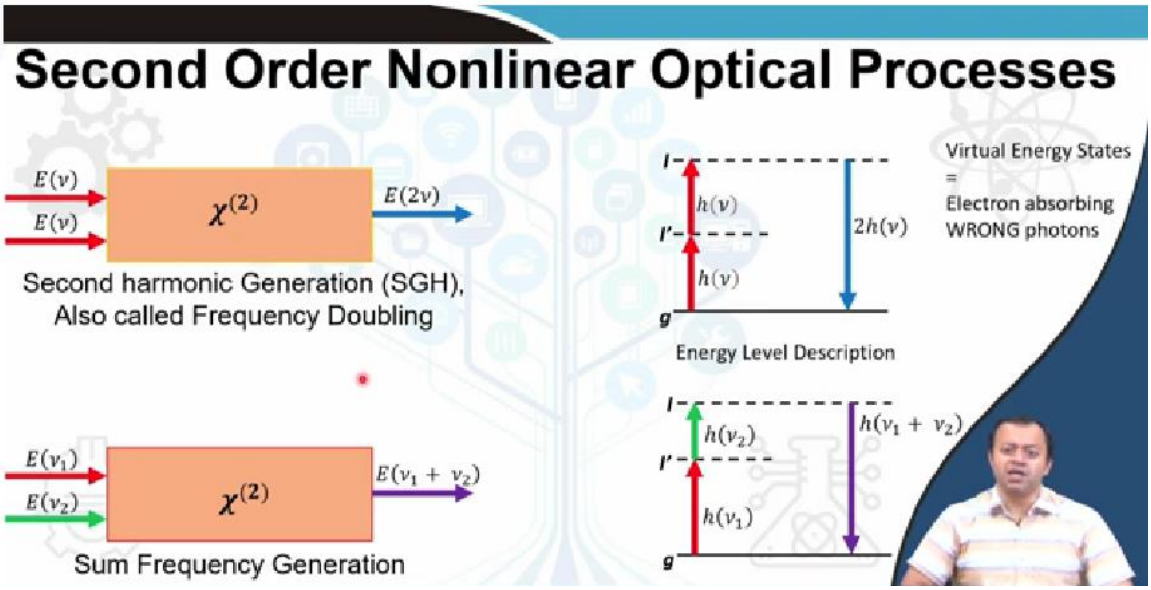


So, these charge particles will be interacting with the intense electric field produced by the laser and they will be rearranging themselves they will be rearranging themselves and that is given by the process of polarization of the medium. So, the polarization is given by it is not x it is actually Greek letter χE is the electric field is the electric field generated by light chi is the linear optical susceptibility susceptible. So, polarization polarization the electrons or the charge particles take a particular distance move a particular distance they align themselves you know in magnet whenever the magnet can be the domains can be in random direction whenever there is a magnetic field all of them align themselves into a specific direction towards the magnets. So, that is that is polarization they are polarizing they are aligning towards a specific specific direction. How quickly or what by what effect they will align themselves polarization will takes place upon having this particular electric field generated by light is given by this factor called susceptibility susceptible.

How quickly how much what is the amount not just necessarily time by how much amount how much amount the material this complex complex molecule has the capacity to rearrange rearrange it is electron cloud when it is subjected when it is subjected to an intense electric field as generated by laser light it is given by this particular formula p is equal to χE is the electric field p is the polarization this is the alignment and the amount of alignment amount of alignment how much what amount the alignment will happen is given by susceptibility. We talk about susceptible how someone is you know susceptible a person

is susceptible to a particular disease someone is susceptible to ah bad advice. So, this is optical susceptibility how it will react to how much it will react when subjected to optical susceptibility. Now this thing becomes complicated in non-linear conditions where not just this the second and third order non-linear optical susceptibility starts coming up where $\chi 2$ and $\chi 3$ starts coming up where $\chi 2$ is given by this particular formula where f is the field correct factor n is the number density of molecules beta is the non-linear optical coefficient what does this mean is almost all molecule and their electric electron cloud get somehow affected by electric field generated by light. So, this susceptibility is common in almost all materials right this rearrangement somewhat amount of rearrangement here and there in the electron cloud when subjected to electromagnetic field intense electromagnetic field is common.

However there are certain materials where the formula this formula is not enough where you have to use $\chi 2$ and $\chi 3$ second order and third order non-linear values which has its own own own value which has its own ah formula and that starts affecting the material deeply meaning in simple term meaning in simple term the rearrangement the rearrangement of the electron cloud in certain materials upon subjected to intense light has the potential not necessarily it will happen has the potential has the potential to generate additional energy states.



What does that mean? You have a band gap yes you have a band gap higher energy state lower energy state what does the band gap depends on? Band gap depends on how the electrons are arranged how the atom has formed the molecule the distribution of the electron cloud and so on and so forth. The intense realignment intense realignment of the electron cloud will can change the band gap and and can create additional energy states within the band gap and that is a non-linear process linear means anything that is linear is

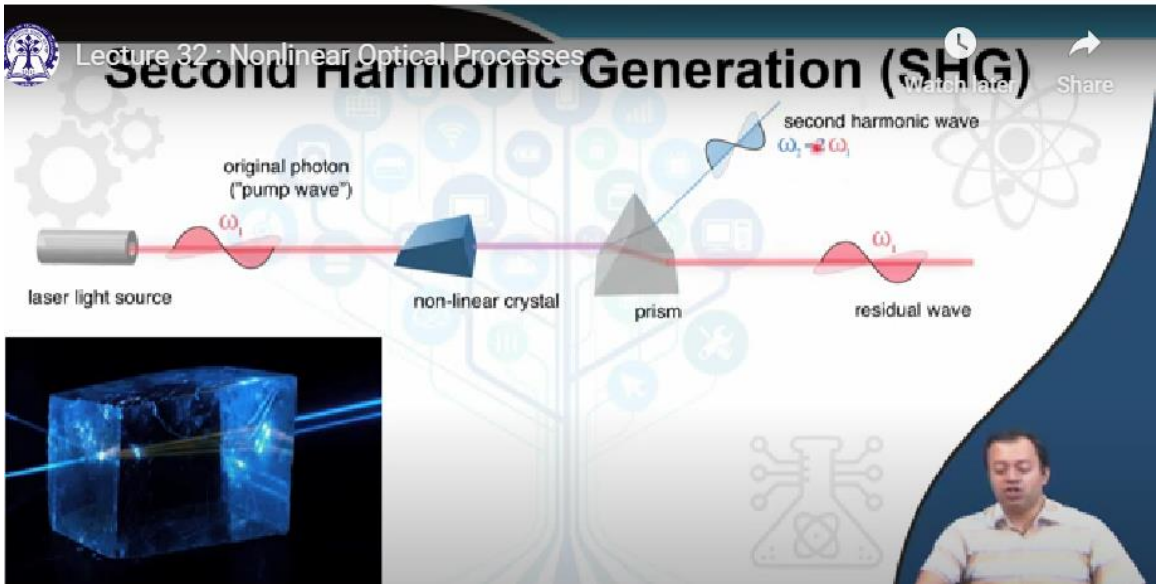
simply directly proportional indirectly proportional non-linear where square cube all other terms coming up where simply input is not directly or indirectly indirectly proportional or directly proportional to the output there is a complicated relationship between them. How does that matter? It produces a bizarre thing I told you previously that only that photon will be absorbed which matches the band gap. I told you these energy states are discrete you can only sit in step 1 or step 2 or step 3 there is no step 2.

5 in the ladder in the ladder there are rungs rung 1 rung 2 rung 3 rung 4 there is nothing in between 2 steps of a ladder you cannot put your feet in between 2 steps of ladder it has to be discrete. Turns out in non-linearity when you have subjected it to intense energy state intense energy electric field to some exotic materials common example lithium niobate lithium niobate lithium niobium oxygen lithium niobate this complex structure lithium is how much atomic number very small atomic number niobium is very high in a atomic number oxygen is less, but not as less as lithium this complex structure has formed this complex structure has formed it does something bizarre it starts absorbing the wrong photons it starts absorbing the wrong photons wrong photons it creates intermediate energy states in between the band gap it creates intermediate eigenstates intermediate energy states in between the band gap. So, previously I told you previously I told you if it has consumed say 500 nanometer photon it will emit 500 nanometer photons if it is a very simple 2 level system ah silicon or any other kind of ah not silicon silicon is 1.14 eV 500 nanometer any any other ah compound semiconductor as such. In this particular case in a non-linear material you do not give a 500 nanometer photon and get a 500 nanometer photon back you give 2 photons 250 nanometer each this is just a lambda is not directly converted into e, but 2 photons of half the energy h nu and h nu nu and nu and you get 2hν back.

Second order non-linear optical process there is a virtual energy state that is only present that is only present when this material this non-linear material is subjected to intense wavelength of light intense laser light it start absorbing the wrong photon which is half its band gap which is different from its band gap half I have said it is easy h nu h nu combine together and produce 2hν the opposite some frequency generation is also possible where you have ν1 and ν2 input the output is ν1 plus ν2 nu mean the frequency. This is the bizarre thing that comes up in non-linear optical processes that falls flat on our idea that energy states are discrete yes energy states are discrete 99.9 percent cases. The 0.1 percent case is where artificial or virtual energy states are created in certain exotic materials where where the material starts absorbing the wrong photon it is not supposed to absorb this it is not supposed to absorb this it cannot absorb something in between and then you make nu 2 which takes it to further.

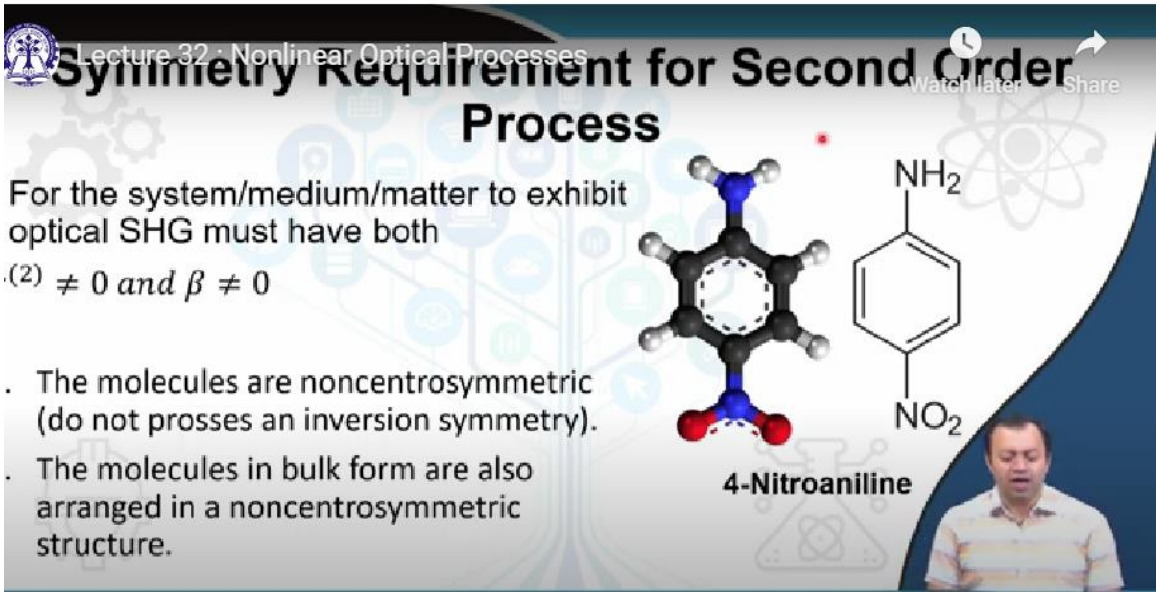
We do not know what happened to the electron that is going from here to here does it stop

here in between you can say that there is a defect state we have seen metastable state, but this is different defect state is a permanent state you have added dopant into the your semiconductor and it is showing defect fine granted here there is no dopant and this is purely temporary we cannot measure it this is only under certain conditions when you have subjected to intense laser light this does not happen when you remove the light. So, this state is non-permanent metastable state as we have seen previously having defect having impurity those are permanent you have given it defect you have added dopant into your semiconductor ah arsenic or phosphorus or p-type n-type we have not removed those. Here it is only when light is being subjected and that is non-linear optical processes. Non-linear optical processes mean a rearrangement a polarization of the electron cloud in such a bizarre manner that some additional energy states start setting up start setting up within the existing band gap. These energy states are temporary energy states cannot be measured they are called virtual energy states eigen value not at all like your normal eigen value of energies the normal energy states and they can be utilized to produce some frequency some frequency input 1 plus input 2 is output.



If you have the same input then the output is 2 times the input 2hν or hν 1 plus ν2 and this is what had happens this is the crystal is a representative image do not take it literally you have a light source it forms into that exotic material as I said lithium niobate or even silicon at certain time shows non-linear effect this omega 1 large number of omega 1 are produced that that passes through the prism the prism you know divides low and behold θ 2 is 2 θ 1 this θ 1 is adding another θ 1 addition has taken place and some amount of θ 1 the residual wave which has not been absorbed which has not done anything is simply taken out you have a second harmonic generation which is the sum the sum frequency the sum frequency of the light source omega 1 plus omega 1 is omega 2 the previous case is also possible θ 1 and θ 2 gives rise to θ 1 plus θ 2 you can consider it as θ 3. So, non-linear optical processes is the one in which you can generate some frequency think of it

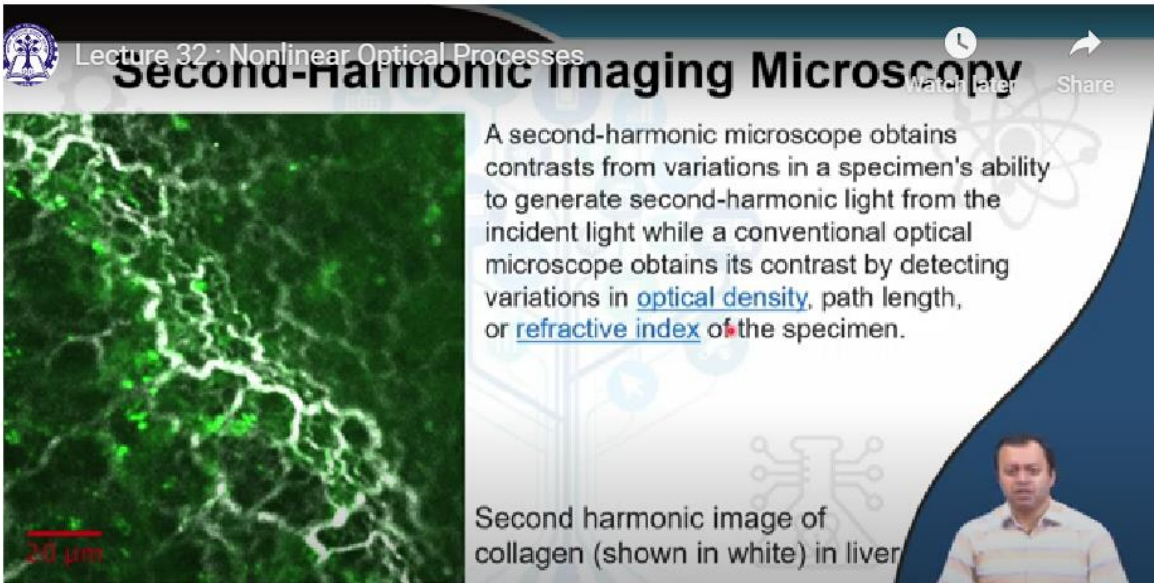
for ah electronics engineers as creating an op amp an optical op amp you remember your first year lecture where you solved operational amplifiers adder yeah 2 or 3 voltages are added together this is exactly the case where the 2 or 3 energies wavelengths are added together frequencies are added together and you generate the output is V1 plus V2 plus V3 remember your adder from ah operational amplifier op amp first chapter first first year electronics engineering ah medical student do not worry about it is an internal joke of ah electronics engineers.



So, what is the symmetry requirement I mean what creates I mean what are the condition that you will need to have ah the non-linear process for a system medium matter to exhibit optical second harmonic generation that is χ 2 to be prominent must have both χ 2 to be 0 and β to be 0 where β is this particular value is a non-linear optical coefficient meaning that the molecules are non centro symmetric it is not symmetric across the center this is not symmetric across the center. Across the center if you bifurcate it if you bifurcate it if you divide it 2 halves will not create it back it is anisotropic it is different in different direction. So, the molecules are non centro symmetric the molecules do not have symmetry across the center they are asymmetric molecules the molecules are different they are complicated molecules complicated molecules having different arrangement of electron cloud in different direction it is not a sphere or an elliptical electron cloud it is a complex electron cloud that is asymmetric in different direction.

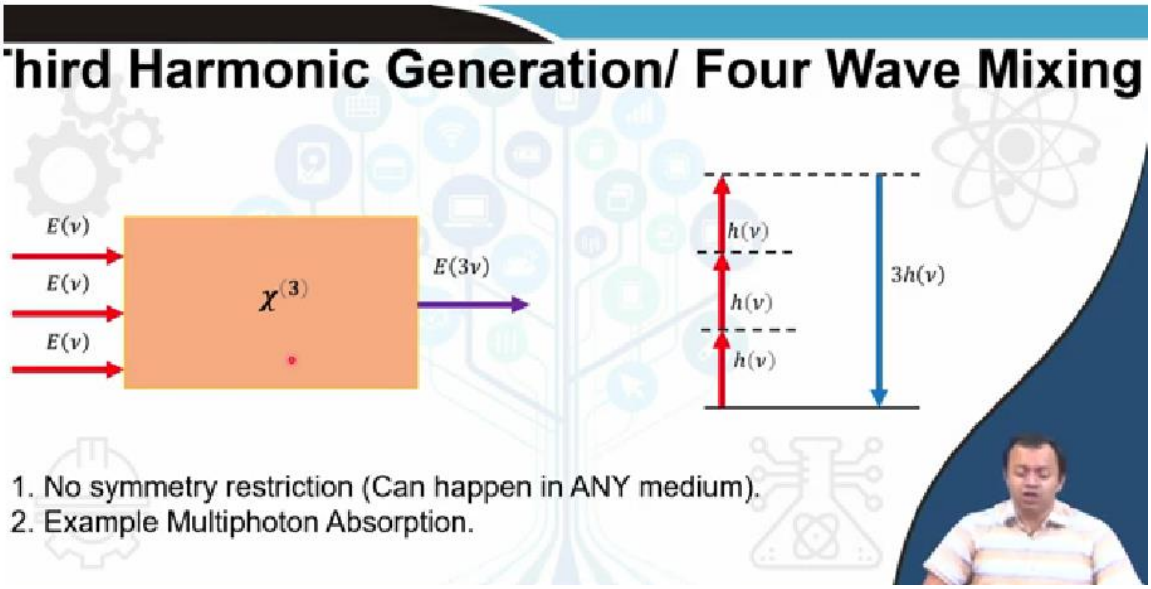
So, that when you have subjected to intense light the rearrangement is also asymmetric the molecule in bulk form are also arranged in a non centro symmetric structure sometimes it happen that something is arranged randomly, but so many is like glass so many of them are arranged. So, the overall average symmetry comes up something specific specific material has to be there whose bulk form bulk arrangement is also non centro symmetric

it is throughout asymmetric both in it is short form and it is long form like this nitro aniline etcetera the molecules do not have inversion symmetry if you invert them it will not overlap with it is previous part like our hand our hands are chiral like our hands our hands are chiral it is non centro symmetric they are not symmetric. They are not symmetric across the center you cannot simply break them into half and arrange this 2 half across the center they are asymmetric arrangement asymmetrically arranged complicated molecules produce this bizarre effect that breaks that breaks this notion of discrete energy states it produces nonlinearity where virtual states virtual Eigen states get generated under subject to intense optical light intense optical light and and it can be utilized for addition of 2 input energy frequencies it can be utilized as an example of a 2 input optical operational amplifier if you can add can you subtract if you can add can you subtract if you have addition and subtraction the rest 2 operation multiplication division can you thereby create create a precursor of an arithmetic and logic unit working on the 3rd order of computing only on photons have you heard of quantum computing what do they do can it be utilizing nonlinearity do we have nonlinear material enough with us lithium niobate do you have it why do we do it or where bio photonics people utilizes it.

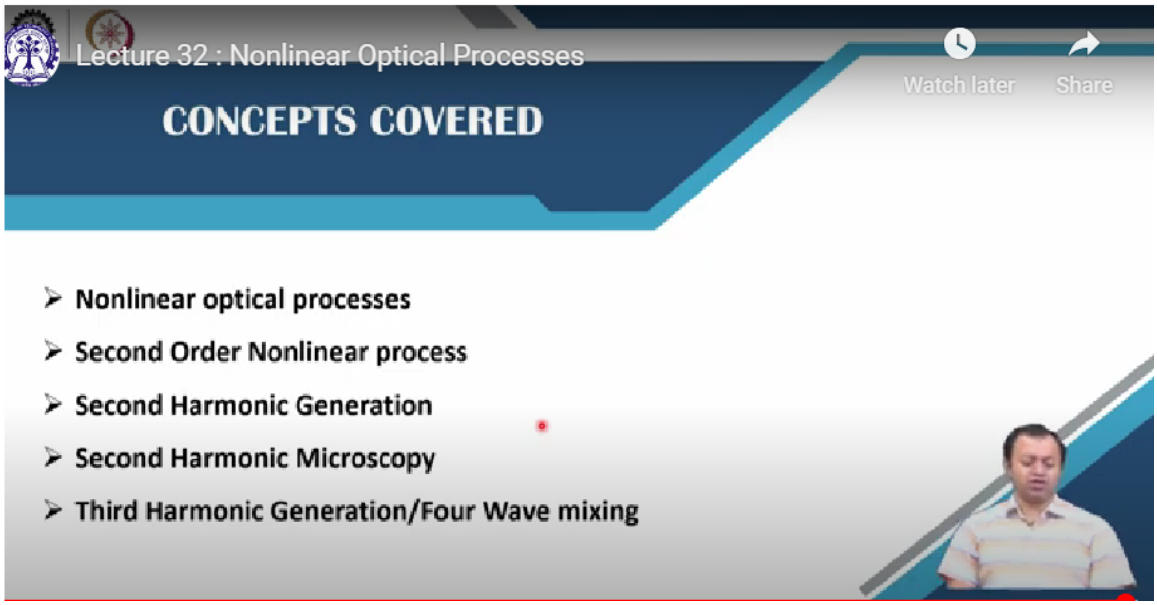


Second harmonic generation is quite common in some imaging techniques a second harmonic microscopes obtain contrast from variation in specimens ability to generate so your body has complex molecules right nothing is more complex than those molecules that are present in your body proteins etcetera several of them has the capacity to produce nonlinearity effect meaning if you produces if you give an input of 2 photon some of them may add those photon up and emit omega 3 which is equal to omega 1 plus omega 2 omega being the frequency frequency 1 and frequency 2 you have given it inside a molecule inside a protein inside some kind of a biological matter they have added those 2 up and emitted a specific photon which is twice which is the sum frequency our bodies collagen

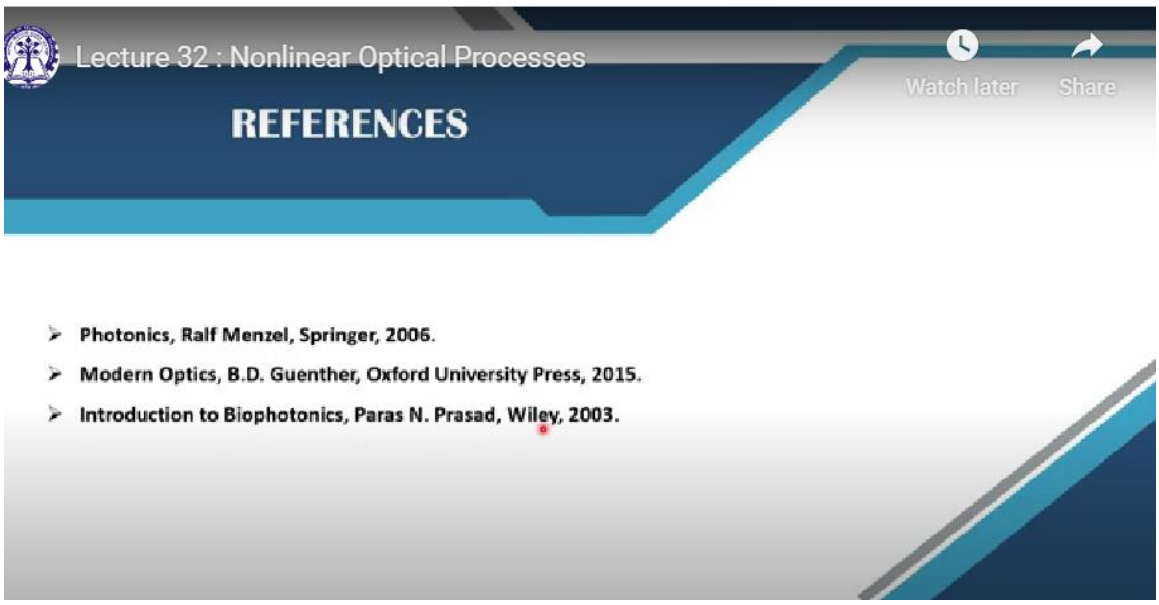
produces second harmonic generate second harmonic. So, if you image a liver you can see the actual you can see the actual collagen distribution collagen will be 1 which will be emitting a specific photon which will be the sum of the 2 inputs which you have used to illuminate if you have given sum you get a specific frequency rest of them will not give non collagen part will not give and here by you will see the distribution of collagen where is collagen present in our body other than liver or what is collagen used for remember we have discussed this you can see the distribution of collagen in liver using second harmonic microscopes it can conventional optical microscope as compared from conventional optical microscopes is obtains it contrast by detecting variation in optical density path length or refractive index of specimen second harmonic generation utilizes to see ah collagen distribution in different parts of body because collagen is such a complex compound which shows which has the capacity to sum the frequencies there is also third harmonic generation though these are quite weak



technically second harmonic generation can also be present in other materials but its effect is very very weak so almost negligible third harmonic generation where E 1 E 2 and E 3 nu 1 nu 2 and nu 3 are added together to produce nu 4 which is sum frequency nu 1 nu 2 nu 3 if you have the same frequency then instead of nu 1 nu 2 nu 3 you get 3 nu 3 nu frequency no symmetry restriction can happen multi photon absorption takes place but this effect third harmonic generation or 4 wave mixing 1 2 3 and 4 wave mixing is very most of the time quite weak quite of a weak signal but it simply shows that laser opened up several new field which were previously only theoretically accepted that this thing might be possible the rearrangement but laser proved the existence of such things previously practically.



So, these are the concepts covered for today non-linear optical processes second harmonic generation second harmonic microscopy and these are my ah references



I will see you in next class. Thank you very much.