Introduction to Non-Linear Optics and Its Applications
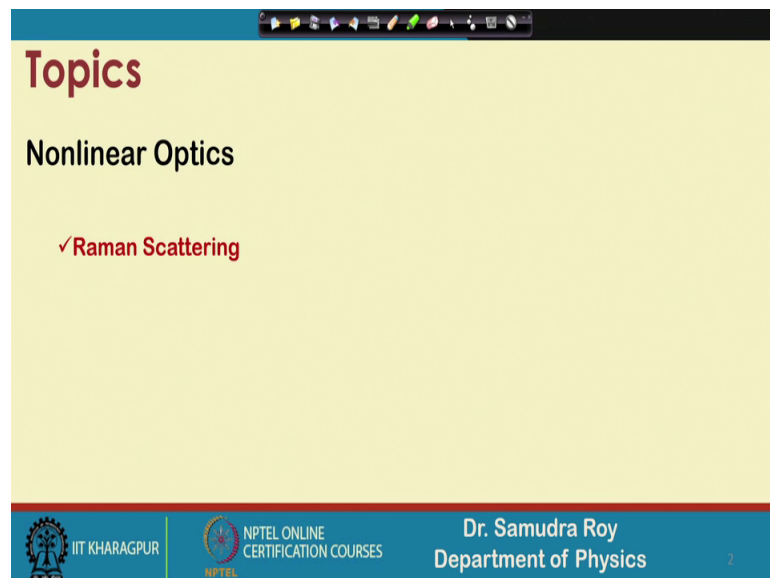**Prof. Samudra Roy**
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Lecture - 54
Raman Scattering

So, welcome back student to the next class of Non-Linear Optics and Application.
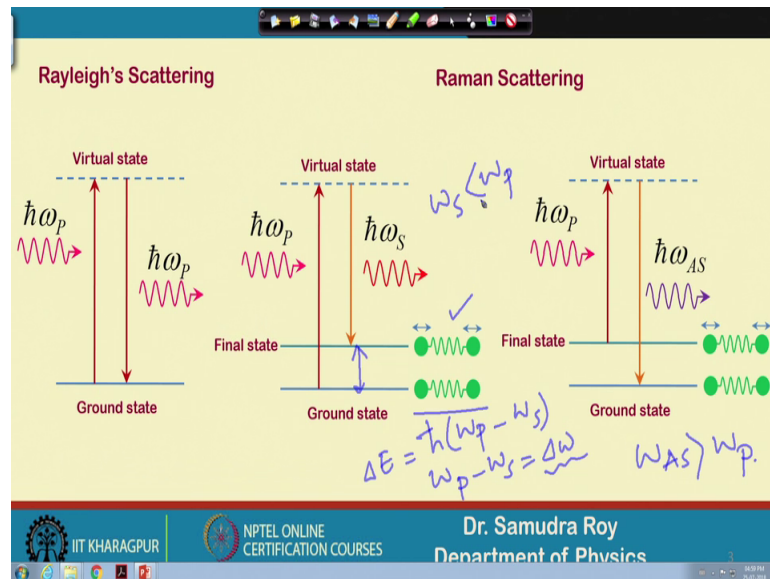
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So, today we will going to learn a very important topic in non-linear optics which is Raman scattering and then gradually we learn the Raman amplification and stimulated Raman scattering etcetera.

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So, what is Raman scattering so, let us give you a very rough idea about that. So, we know that in Rayleigh's scattering what happened in if some light wave is coming or the photon is coming to the material. So, the atoms can go to ground state to some virtual state and then return back to the ground state.

And, we will have a similar when it go through the ground state it emits and if it is some sort of elastic kind of scattering then we will going to have the same wavelength. So, there is a no change of wavelength. So, this is a simple Rayleigh's scattering kind of phenomena, where due to some elastic scattering process there is no change of wavelength or frequency.

On the other hand in Raman scattering process, if you launch a photon, if the photon is interacting with the material then whatever after interaction whatever is coming out is may not be the same wavelength that is launched to the system. So, the wavelength may be greater than to the launched wavelength or the less than to the launched wavelength depending on the system. And here since the energy is exchanged so, the energy conservation; obviously, the energy conservation should be there, but input and output wavelength or frequency is not same.

What happened here so, the molecule can go from ground state to some virtual state and return back to some final state, this final state may not be the ground state. So, there is a difference between the energy from ground state and final state. So, the ground state, the molecule have one vibrational state or rotational state. When it absorb some kind of

energy from the input wave what happened that it will start vibrating or rotating some frequencies some of the molecular vibration or rotational state is now change.

So, this is now in higher energy state, since it is absorbing some sort of energy whatever is coming out from the system should be less than of the energy that is launched to the system. So; obviously, the amount of energy difference delta E is nothing, but h cross omega p minus omega s. Now, if omega p minus omega s is delta omega so, delta omega is the frequency at which this molecule is vibrating or rotating.

So, this is the corresponding frequency; in a similar way if the molecule is already in some excited state and then some energy is falling. So, this molecule can go to some virtual state and then go back to the ground state where, in the ground state it can lose some kind of energy and we will have some amount of added energy here and the wave that is coming out from the system or the material is more than whatever is launched.

So, here omega as is greater than omega p. So, we have a web we have a frequency which is greater here omega s was less than omega p. So, two different procedure different systems can be possible; in one case it is a called stroke system and in other case it is called anti stroke system. So, we all are aware of this these two processes.

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So, today we will going to learn in detail exactly what is going on in Raman scattering process in terms of nonlinear optics.

So, before going to these things, there are few a the few notation that we need to understand that is a generalized coordinate. So, in classical theory of Raman scattering what happened so, this is the left hand side of this process, the left hand side of this slide gives the process of the classical Raman scattering. So, the classical treatment suggests that a monochromatic wave which is E 0 cos omega p t omega p is the frequency of monochromatic wave, it is launched to the system and as a result what happened that the molecule is start.

The molecule is now vibrating with that particular frequency and microscopic polarization can generate because of this vibration the average position of the molecule can change and since there is a charge separation we can have a charge separation multiplied by the distance. And this charge separation multiplied by distance can give rise to some microscopic polarization. So, this microscopic polarization can be represented in this way E 1 is a field that is launched epsilon 0 is as usual there and this alpha basically called microscopic susceptibility. If you look carefully the form is exactly whatever the form we use normally; so, it is used like this way chi 1 this, this is the normal way to write the polarization. Here we are writing the same thing, but we write microscopic polarization.

Because the susceptibility thing is only for this particular molecule for the time being later consider that. Now, this microscopic susceptibility is a function of generalized coordinate; that means, the molecules are vibrating since this is vibrating is generalized coordinate is changing. So, we consider that since the generalized coordinate is changing this is a weak function of this generalized coordinate Q.

So, alpha is a function of Q if it is a weak function of Q we know that any function can be expanded in terms of Taylor series. So, if I expand this as a Taylor series we have a first order term and a second order correction term and since it is since it is the function of Q it should be the derivative of that quantity multiplied by Q. If I go to higher and higher order it should be the double derivative of alpha with respect to Q at central point which is 0 and then Q square with a half term and So on. So, it is simply a Taylor series, but since we say that this is a weak function of Q. So, Q square Q cube and all this term you are not going to consider.

So, this is the microscopic susceptibility and now microscopic susceptibility is written explicitly in terms of Q. Now, what is Q let us try to understand very quickly what is generalized coordinate. So, generalized coordinate is something where, say this is the molecule and this molecule is sitting in some coordinate system one dimensional coordinate system. So, this point I can write x 2 this point I can write x 1; now if it is stretched this position is changing; so, now, x 2 become x 2 prime x 1 become x 1 prime.

So, now, the change of length is x 2 prime minus x 1 prime minus of x 2 minus x 1; that means this length minus this length as simple as that. So, if I do then I can readily find that this difference is nothing, but this quantity whatever we have if it is q 1 and this q 2 is related to these two parameter.

So, q 1 and q 2 if I combine I will have if one coordinate q. So, all this coordinate can be combined in such a way that I can I can write a simple generalize coordinate to understand the dynamics of this system. Here Q is a generalized coordinate so, this Q is a function this microscopic susceptibility is a function of Q. So, since microscopic susceptibility is a function of Q I can expand this in this way.

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Well next same system monochromatic field is launched and because of this monochromatic field the molecules are now vibrating. So, since the molecular vibrating; so, it may have some generalized coordinate, which can which can give the idea how this

position of this molecule is going to change etcetera. So, if it is a sinusoidal field then we can write Q as a sinusoidal also.

But note that I write a different frequency it is may not be the same frequency the way molecules are vibrating. And it is quite obvious that molecule because of this monochromatic field radiation the molecule can go from lower energy to higher energy and return back to some final state and then start vibrating to that final state.

So, omega is the vibration of or the frequency of this final state I am talking about. So, p is a microscopic polarizability; microscopy polarizability can be now expanded in terms of Q because alpha is a weak function of Q. So, alpha is alpha 0 plus d alpha dQ at 0 point multiplied by Q and E i E i is electric field. Now, E i is E 0 cos omega p t, I just simply write this quantity and then I multiply these things to and make it two different terms.

One term is epsilon 0 alpha 0 both are constant E 0 is constant and cos omega p t ; that means, microscopic polarizability is now vibrating with one frequency which is omega p t. But other term is still here which is multiplied by Q and E 0 cos omega p t Q is vibrating with this frequency. So, it is important because we need to take care of that also.

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So, now, the polarizability has few frequency terms epsilon 0 alpha 0 E 0 cos of omega p t usual term already there. Next term is Q multiplied by E i is E 0 cos omega p t and Q is also. So, we know that Q we have already represented this Q as cos of omega t. So, I write explicitly Q as Q 0 cos of omega t.

So, now, if I simply manipulate this term cos omega t and cos omega p t cos b b omega t rather cos omega p t, then if I take a half term, then half of cos omega t into cos omega p t can be represented at cos of omega big omega plus omega p t and cos of big omega minus omega p of t.

So, now you can see quite easily that this microscopic polarizability is having three different frequency components. Original frequency and two other frequencies here and here; and here if I write in terms of frequency the microscopic polarizability is now having three frequency component, original frequency original frequency plus big omega which is the vibration of the molecule and original frequency minus omega which is; that means, some sort of energy that is reduced from the original frequency.

So, microscopic polarizability is a source of field microscopic field. So, 1 microscopic field can due to this term which should have a frequency omega p one field can be generated due to this source term whose frequency can be omega plus omega p and another field can still generate be generated and whose frequency is omega p.

Minus omega mind it is a cos so, it does not make any difference if I write cos of omega p minus omega. Because cos of omega minus omega p is same as cos of omega p minus omega since, omega p is much much greater than omega it is better to write this notation, but anyway since it is a cos. So, there should not be any difference.

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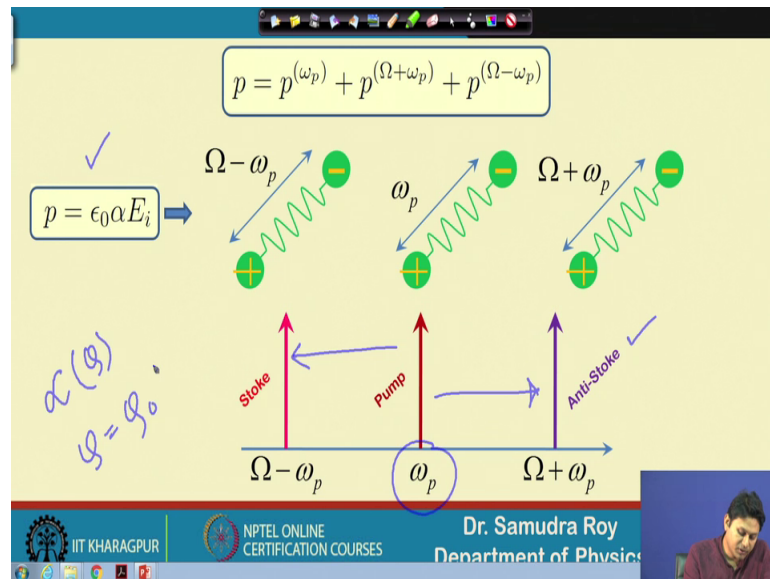
Well if I now try to understand that exactly what is going on and this is a pictorial figure for that, that as I mentioned this microscopic polarizability now vibrating three different frequencies. So, these 3 different frequencies give rise to three different waves and these three different waves if I write in the output if I try to find out what is the spectrum of this three different wave.

I will find apart from this central frequency if I write this pump frequency as central frequency one frequency will be more than central frequency which is anti stoke wave and another frequency will be less than omega p which is some sort of stoke waves. So, this microscopic polarizability because microscopic susceptibility is a function of generalized coordinate, generalized coordinate is vibrating with some frequency generalized coordinate is related to the coordinate of the molecule. So, try to understand once more what is happening here one electric field is launched.

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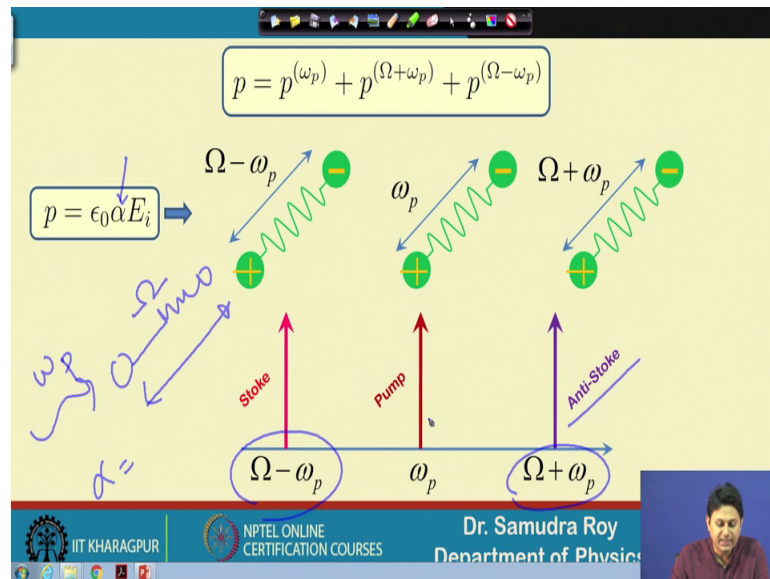
Because of this, electric field what happened the molecules here is now vibrating. This vibration of the molecule can be represent with a frequency say omega the launched electric field has a frequency omega p; because of this non-linear because of this polarization microscopic polarization we have a microscopic susceptibility. So, this microscopic susceptibility is a function of q when they multiplied with the input field then we have the interaction of the molecule infield this interaction of the molecule infield give rise to two different frequencies on.

One is omega p plus omega which is higher than the launched wave or launched frequency we call this anti stoke and another is stoke which is less than the frequency that is launched.

So, two different frequencies can be generated and this is happening because the electric field is directly interacting with the molecule. So, the light metal interaction is directly here and because of this light metal interaction we are getting the frequency, which is higher and lower than the frequency here of the launched frequency.

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Stimulated Raman Scattering

$$E^{(\omega_p)} = \frac{1}{2}\left[E_p e^{i(k_p z - \omega_p t)} + c.c\right]$$

$$E^{(\omega_s)} = \frac{1}{2}\left[E_s e^{i(k_s z - \omega_s t)} + c.c\right]$$

$$E = E^{(\omega_p)} + E^{(\omega_s)}$$

$$p = \epsilon_0 \alpha(Q)E$$

$$P = Np = N\epsilon_0 \alpha(Q)E$$

Microscopic Susceptibility

$$\alpha(Q) = \alpha_0 + \left.\frac{d\alpha}{dQ}\right|_0 Q + ...$$

So, after having a very simple straightforward thing now we try to understand stimulated Raman scattering. So, what is stimulated Raman scattering? So, we already find that if omega p is launched. So, there is a possibility that omega p plus something and omega p minus something can be generated. So, let us try to understand if I generate say anti stoke; that means, omega p minus something if I write it is omega s. So, how this omega s can be amplified?

So, in order to amplify in this omega s what happened I launched omega p to the system, this system I have say chi 3 not equal to 0. Then along with omega p I also launched omega s in the output we have omega p and also omega s, but omega s is amplified you can see it is amplified it was a weak wave, but there is an amplification. So, what is the process here? So, our treatment will be exactly the same E p the field with omega p frequency can be represented in this way E p e to the power i k p z minus omega p t.

The field associated with the stokes waves or the stokes frequency can be represented at e a c to the power k z minus omega t omega s t plus complex conjugate total field is E of omega p plus e of omega s. Microscopic polarizability p is epsilon 0 microscopic susceptibility multiplied by the total electric field E and here we consider the total polarization.

So, when we consider the total polarization we make an approximation and you multiply the P with the number of molecule per unit volume N or the number of dipole per unit volume N. So, multiplying N we have the total polarization. So, microscopic polarization

can be represented in term terms of total polarization, but microscopic susceptibility is still a function of Q.

Which is the most important thing here in classical treatment and I can write this explicit form of alpha which is a function of Q. So, all the things are now in our hand and try to understand how the stimulated process can be generated. So, E p, E s is in our hand, microscopic polarizability can be represent it terms of total polarizability. So, total polarizability is now in in terms of microscopic susceptibility; microscopic susceptibility is represented in terms of Q. So, Q should also be in our hand.

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So, now see what happened, when you try to calculate the stimulated Raman process. So, as I mentioned P is N alpha epsilon 0 and E alpha is a function of Q. So, I can write alpha in this way alpha is a function of Q weak function of Q rather. So, I write alpha 0 plus d of alpha d q at 0 point multiplied by q. So, this is the thing. So, total p is this quantity plus this.

So, now, I divide the total P into 2 part this is a linear part, and this is a non-linear part. So, far in the, we know that total polarization can be represented in terms of p linear plus p non-linear. P linear we know how to represent it is chi 0 epsilon 0 chi 1 E and p non-linear if second order effect is there then it should be epsilon 0 chi 2 E square if third order effect is there it is epsilon 0 chi three E cube this is the way we represent p non-linear.

So, here since chi 2 chi 3 is not directly it is not coming here because we are dealing with microscopic susceptibility. So, we simply write this into 2 parts.

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One is this which is a linear part and another is this which is a non-linear part. So, non-linear part is now in our hand and in non-linear part we have n epsilon 0 d alpha dQ 0 all these things are constant multiplied by Q and E, mind it u is a total field. So, u should be omega p plus e omega is fine.

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Next, next we need to find out what is the differential equation for the molecule. So, when the electric field is launched what happened the molecules goes from ground state to virtual state and returned back to some final state with a vibration and this vibration should have frequency. Delta omega which is the difference between omega p and omega s. S

o, energy conservation is total valid here. So, since this energy conservation is valid we can write that this molecule is now vibrating with a frequency delta omega. Now, we try to understand the dynamics of this vibration because our aim here is to find out this Q, our aim here is to find out the form of this generalized coordinate. In order to find out this generalized coordinate so, I need to write the equation of motion and solve it.

So, the equation of motion is for this molecule which is vibrating as a frequency delta omega is simply a forced m oscillator model, because I am launching some electric field which is because of that it is vibrating and when it is vibrating it should have some sort of resonance frequency and some sort of damping. So, m multiplied by so normally this equation is mx dot double dot plus gamma x dot plus say omega square m gamma x dot multiplied by m omega r x is equal to some force term.

So, we have been using this equation I mean we have used this equation in earlier classes. So, we are using the same equation here, but instead of x we are using the generalized coordinate because here everything is represented in terms of generalized coordinate. So now, if I try to understand what is the equation so, this equation is simply forced damped oscillation and this forced damped oscillation model, if I use for this particular molecule this equation is simply comes out to be this force is something that we need to calculate here.

So, what should be the force on these things it is nothing, but D dot E. So, if a electric field is launched so, some kind of potential energy that is stored here. So, this potential energy that is stored in to the system is nothing, but d dot e with a half term and a negative sign since it is stored which is half epsilon 0 half epsilon E square because D is epsilon E. So, if I put it here and if d and e is in the same direction for isotropic system say.

So, it will be minus of half epsilon 0 E square. So, now, we know what is my potential, which should be a function of Q by the way. So, this potential term now if I make a

derivative of this potential term then we then only we can find out the forced term in our hand which is important. So, we need to know what is the force experienced by the system.

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Well how to find out this. So, in this slide we will going to understand. So, total D is epsilon 0 E P we know, P here is the polarization we know this polarization can be represented in this way epsilon 0 N alpha E because alpha is a microscopic polarization everything. So, far we are doing in is in terms of microscopy polarization. So, d is epsilon 0 1 plus n alpha E and this is epsilon E. So, epsilon I can extract from this equation.

And epsilon is 1 plus N alpha Q epsilon 0 alpha Q is something which we know because this is an explicit function or the expansion that we are using. So, alpha Q we are using that and it is not this is alpha 0 plus d alpha dq and then Q. So, once we have these we put this here. So, V is now written completely in terms of Q. So, V is a function of Q, so, Q was inside this epsilon. So, I now expand this epsilon.

So, when I expand this epsilon V is now represent in terms of Q. So, once we write it is epsilon 0 is square. So, f is the derivative term because F was a grad of this thing. So, only one variable is here Q. So, it should be minus of del q and v of Q.

So, now, if I make a derivative it should be simply half epsilon 0 n this quantity and E square because this E will, because of derivative this term will be affected other two terms will going to vanish. So, F is calculated so, once F is calculated we can have F m which is forced per molecule. The force per molecule we just simply divide the total number of molecule per unit volume which is n and finally, we have F m n in this form.

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So, once we have fm in our hand. then the differential equation can be a little bit simpler because in the right hand side we know everything in terms of electric field and alpha which is our, which is the information that we have so far.

So, in the right hand side we can so, this is the total equation the equation of molecule that is vibrating as a frequency delta omega as I mentioned. So, this equation is now is having a compact form. So, today we will not going to solve this equation because it will take some time again to solve this equation it is easy, but it will take some time to solve. But important thing is that in the right hand side the total field is in the right hand side the force is written in terms of the external or the total field E and the corresponding alpha value, directly alpha is not involved here, but the derivative of alpha is here.

So, in the next class we will start from this part. So, we have derived so far that if I launch an electric field the molecule will be excited and vibrating in some frequencies. If it is vibrating some kind of frequencies then the important thing that what should be the generalized coordinate because the generalized coordinate is important to know what

should be the non-linear polarization associated with that molecule which is vibrating with certain frequencies.

So, first we need to solve the differential equation of the vibration of this molecule. So, then we use the damped force oscillation model here and first find out what is the corresponding field and what is the corresponding force in terms of field that is experienced by the molecule and we have derived a differential equation.

So, next day we will going to solve this differential equation and find out what is the generalized coordinate Q, once we know the Q next we find out what is the corresponding polarization so polarization, non-linear polarization value explicitly we can know once we solve this equation. So, with this note so let me conclude here so.

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