

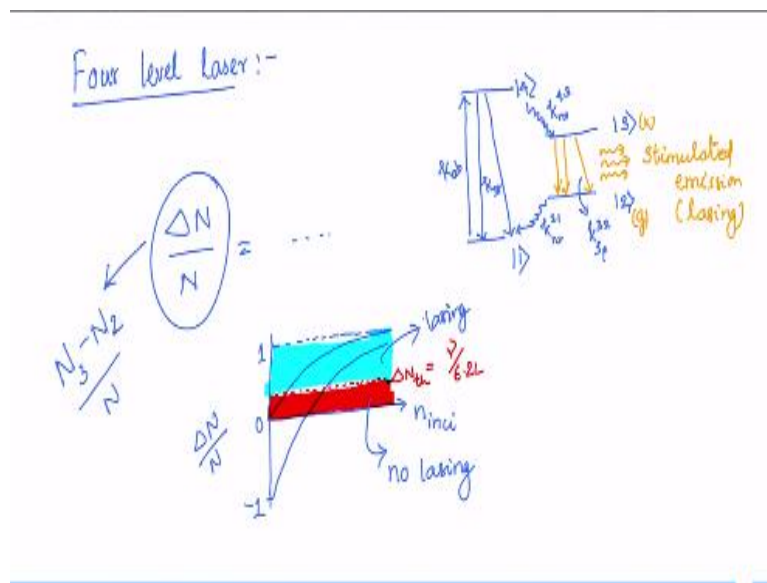
Optical Spectroscopy and Microscopy : Fundamentals of Optical Measurements and Instrumentation
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Lecture - 28

Hello and welcome to the course on Optical Spectroscopy and Microscopy. In the previous lecture what we have seen is that there are for populations inversion to exist which is the precondition for our being able to see a net emission of light through stimulated I mean through stimulated emission, net light coming out from the device through stimulated emission we need a population inversion.

For that we looked at different possibilities of the electronic state configurations in the lasing media itself and then we said okay look, each of them requires a different, I mean sets a different condition for the existence of a population in motion. And then towards the end, we were actually looking at the four level system.

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Or we call that as a four level laser, wherein we describe these four levels as level 1, 2, 3 and 4 in increasing order of energy and the processes we indicate through the arrows. And then I said okay, we can actually go ahead and work out the expression for $\frac{\Delta N}{N}$, the population fraction of the population existing as a higher population, where the ΔN here is actually the ΔN here is really number of molecules in the N_3 minus number of molecules in state two.

So because the lasing is happening between the levels 3 and 4 okay, well this is where the stimulated emission we are trying to hone in on, this is the stimulated okay. So we can stimulated emission. We can call that as lasing. All right, so we want to amplify this, right. That is the whole idea here. So in this process, we can actually, what we said was that if we can actually write down the expression for delta N by N.

Hopefully you guys went ahead and obtained an expression for this. I will, so when you do that, your expression will look something similar to this okay.

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$$\frac{\Delta N}{N} = \frac{(k_{nr}^{21} - k_{sp}^{32}) \cdot N_{ini}}{k_{abs}^{23} (k_{21} + k_{32}) + k_{sp}^{32} k_{21} + k_{21} k_{43}}$$

non-radiative $|2\rangle \rightarrow |1\rangle$

$k_{nr}^{21} > k_{sp}^{32}$

$\Delta = N_3 - N_2$
70

So where the notations are exactly similar to what we have described before, where the subscripts represent the kind of transitions that we are actually looking at and the superscript talks to you about the levels from where to where the transition is happening. For example, here this would mean a non-radiative is a rate constant for non-radiative process happening non-radiative transition happening from state 2 to state 1.

And the rate constant of that we are actually representing as k_{nr}^{21} . Now let us look at the energy level diagram what it actually means is that we are talking about this transition. This is a leap rate at from which the ground lasing level is being depleted to the ground level of the molecule itself, okay. So now so that minus so the you see that the delta N goes as that minus the k's spontaneous rate of the level 32.

So that is this spontaneous rate decay. So what it tells you is that, as long as you are going to have a faster rate right that is $k_{21} n_r$, see in this whole expression, we are looking for ΔN to be positive because ΔN again I am writing it, ΔN is our $N_3 - N_2$, the number of molecules in the third level minus the second level. So if you have $k_{21} n_r$ be larger than I mean we want to have this greater than 0 as much as possible at all times.

So for us to do that, then what we are seeing is that you will need to be able to have this of course, this times the n incidence is going to happen. So it will, you will have the positive fraction as long as $k_{21} n_r$ is larger, $k_{21} n_r$ of 21 is larger than, okay. So that essentially would mean that this rate is larger than the spontaneous decay rate. That is so this is our $k_{spontaneous}$.

Remember the angled I mean the arrows but still straight, going from higher level to lower level is we represent the spontaneous emission through that. So what we are saying is, as long as this rate is faster, you always see a residual amount here, which means it will be at the best be 0 or more, right. Because the depletion rate is larger than the supply rate. So it is this is the rate determining step.

So you end up having some molecules or no molecules at all. It is never the case where you have more molecules here, unlike our three level laser, where there is more number of molecules that at the ground state because they were these two states were overlapping. So now as long as this is ensured in your molecule, you will always have this ΔN be 0 or greater than 1, I mean greater than 0.

And that is why you see that the if you plot the this fraction ΔN by N , it starts with 0 and goes up and at the max it can go to about 1 and let us see when it will be close to 1. It will be close I mean we can increase that by increasing the n incidence, right. This increasing this is going to increase this, right.

That goes directly and you will see that being higher when you are able to basically $k_{21} n_r$ if you assume make the assumption $k_{21} n_r$ is large than spontaneous rate. So as long as this number is much larger than the entire denominator. Now the entire denominator

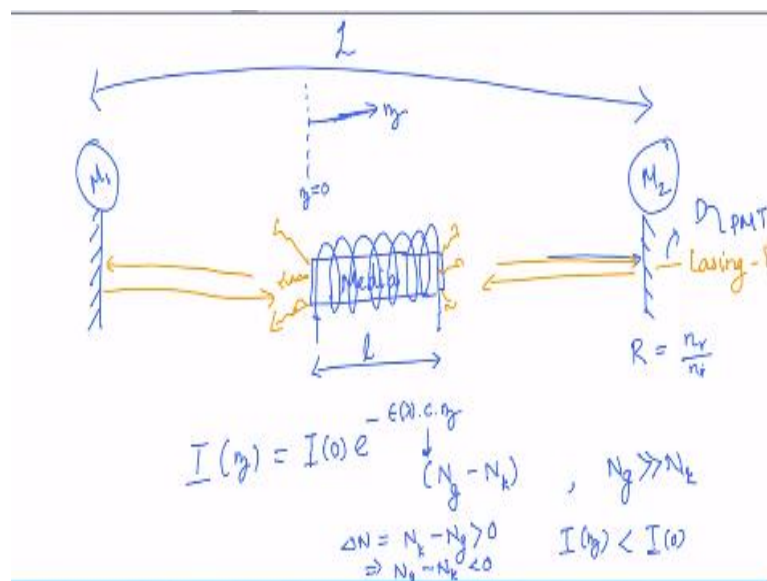
is basically summation of quite a few rates. If you carefully look at it, what it will tell you is that it is a basic you would have seen this is common here 21 and the thing.

So what it will tell you is that you are if you are this through portrayed the pumping rate is high compared to the decay rates. Then you are going to and then of course, k_{43} is larger, so k_{nr43} , right. So let us look at that. And so that is figuring in these two places as long as we are actually pumping it at a speed that is larger compared to k_{21} , right.

That k_{21} is we just now saw which is our spontaneous emission rate, right. So as I mean not just spontaneous emission, but the clearance rate, right. So if you, actually if your pumping rate is larger than this denominator rates of the combined process of this denominator, then you are going to have this as close to n as possible. When you do that mean the point is that this is under your control.

You can actually do that right. So if you do that, because these are all constant for a given molecule then actually even at some point you can actually reach that. That is the crux of the matter. So you do see that the four level laser beam more efficient.

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Now that does not guarantee that if you take a material, even its four level system, and then let us we will call this as media, right like just as before, and then place it between two conductors or reflective surfaces, mirrors. We can call it as M_1 and M_2

2. And we need to have a mechanism to actually impart energy into the system. Here we are talking about optical pumping.

So typically that is the way it is done is by having a discharge tube alright. That is like a flash lamp that this wound around this media. And then what is happening is that the light flash that is generated in this discharge tube is going to energize this media. And then the media is going to fluoresce, the things that the substance that is present in the media is going to fluoresce.

And it is so this fluorescence we are tapping on to okay, and it is this fluorescence we are tapping on to and some of these guys are going to make this round trips and extract more and more molecules in the excited, bring down the more and more molecules from the excited state to the ground state. Remember now we have we are talking about the situation where we have created higher number of molecules and excited states.

So these guys the fluorescent photon when they come back, they actually are going to bring down more molecules and generate photons of their own kind, okay. So now would that result in lasing? Lasing is nothing but the light amplification through stimulated emission of radiation. So what you are doing is you are amplifying the number density of the stimulated emission, photons that are created through stimulated emission, okay.

So now would it ensure that we have lasing? The answer is no. So let us do a little analysis of what is happening here. So since this is the gain media right, this is the media through which we are hoping to gain or convert the light energy that is of one kind right, which is incoherent lights. I mean, it is a light energy that is existing as a incoherent light in terms of the discharge lamp or a flash lamp.

And that is being used to generate fluorescence. Now we want to, we want to make sure that this energy delivered through this flash lamp is efficiently or is converted into energy of stimulated emitted, I mean, the photons of stimulated emission. So now in order to do that, we will go ahead and write down gain equations or the equations

that describe what happens to the photon density when the photons pass through a set up something like this.

Now for a moment if let us forget the fact that we are actually trying to amplify the number density of the stimulated emission photons. Let us think about light being incident on a medium, right. The medium is going to interact with this light and we know from our initial classes that when you have a medium of some length l , then we know what to do or to describe the intensity of the light that is traversing across that media when the media is going to interact with the light.

So that is given by our very simple Beer-Lambert's law. So I am going to write this as my z axis. So intensity as a function of z is given by so my I_0 starts from here. This is z equal to 0. So if I were to write that so then this will be and the intensity will be basically $I_0 e^{-\epsilon c l}$, what is the intensity at the beginning of the medium e^0 to the power minus epsilon function of the frequency or lambda.

So let us think in terms of lambda times the number density or the concentration of the interacting substance that is present times the length of interaction, okay. So now this concentrations of the substance that will be present let us look at it a little bit more closely. So now here, the concentration that we are talking about essentially is the normally we do not have to do this because we are going to now talk about two kinds of interaction here, right.

One is absorption other is a stimulated emission. So in a regular absorption spectrometer I told you before that we do not have molecules in the excited state. So we ignore the stimulated emission term, but here we are going to talk about a stimulated emission eventually. So what we want to take do is to recast this concentration in terms of number densities of the molecules present in the ground state and the excited state, okay.

So if you go ahead and write it, it is actually so we can write this c in terms of number density in the ground state minus the number density that is present in the excited state, okay. So for a four level atom, then we can actually replace the actual numbers

that we have been using so that we do not get confused. So we are talking about the numbers 2 state here as the ground state right for the stimulated emission.

And this number 3 state as the our excited state okay. In our TDBD link it would be the k state. So if you preserve that number then preserve that notation then it will be talking about it $N_g - N_k$. So normally N_k is 0 so that we take the c to be N_g . That is the whole point. So it is a it perfectly fits. Now as you can see, when you have created a population inversion, so this I mean, a step back.

So as you can see, normally at the room temperature what we see is N_g is far greater than N_k . So what you see is I_z is progressively less as the light travels through the media. So I_z goes down okay I_z is lesser than I_0 . So we say the light is being absorbed by the media through interaction.

However, in our case right in the special case of the four level atom and then our way of generating more number of atoms or more number of molecules at the higher energy state of the emission among the emission levels, what we have is the population in motion essentially means ΔN is in our case in $N_k - N_g$ and this is greater than 0 implies $N_g - N_k$ is lesser than zero.

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$$\begin{aligned}
 I(\lambda, z) &= I(\lambda, 0) e^{-\epsilon(\lambda)(N_g - N_k)z} \\
 &= I(\lambda, 0) e^{\epsilon(\lambda)(N_k - N_g)z} \\
 I(\lambda, z) &= I(\lambda, 0) e^{-\epsilon(\lambda)(N_k - N_g)z} \quad \begin{array}{l} \nearrow \text{length of} \\ \text{media} \end{array} \\
 &\quad \downarrow \Delta N \\
 @ \text{ each reflecting surface } (1-R) &\rightarrow \gamma \quad \begin{array}{l} \nearrow \\ \text{(losses)} \end{array} \\
 &\quad \gamma \rightarrow f(1-R, \epsilon(\lambda), \sigma_{\text{scat}}) \\
 I &= I_0 e^{-\gamma z}
 \end{aligned}$$

So we can put this into the system into the equation. So I_z equals I_0 e to the power minus of epsilon a function of lambda right. So still maintain. So clearly the intensity

will also be a function of lambda. So there are different lambdas that gets absorbed. So to be complete, we should actually write this as lambda comma z and so is this.

For a given lambda at z equal to zero, whatever the intensity from that point onwards, we are actually the intensity actually goes down. So epsilon a function of lambda and times $N_g - N_k$ times l. Now for in a during population inversion we just saw this is negative, so you can actually write down this as e to the power epsilon of lambda times N_k , sorry, $N_k - N_g$ times l.

So what you see is that there is a gain in intensity. There is an increase in intensity and it is precisely because of this right if you can actually create a population inversion state, the media that you are talking about becomes a gain media because when you actually normally pass the light through a media it is going to absorb but here, because of the population in motion, you are actually gaining the intensity that is coming out from the other end.

So you could see that the intensity at lambda of z is actually supposed to be z. I should correct everywhere, alright. So alright. So now this time z. So for the entire length we can actually write it as $I(\lambda, 0) e^{\text{minus epsilon of lambda and } N_k - N_g \text{ times l}}$ and this is nothing but our population inversion that we have actually talked about all the time, right.

So now this tells you given a population in motion, how the, how much of the intensity gain we will have as a function of the length of the gain media. But the key here is that we need to consider few more things here which is as the light travels through this setup, what you will see is that the entire light that is hitting on the M 2 is not reflected back okay.

So the amount of light that is reflected by M 2 is depending on the reflection coefficient of the mirror. So if R being the reflection coefficient which is the fraction of the light that is so defined as number of photons that are reflected by the total number, okay. So which is the I mean number of photons that are reflected by the number of photons that are incident.

You will see that this number is at the best be 1 but it is never equal to 1. So at every such reflection there will be a loss okay. So we need to take into account the losses. So the first loss we are talking about is the loss due to reflection. So the loss due to reflection is given by at each of the mirror, reflecting surface we have a loss of $1 - R$ okay. And now what we will do is we will look at this gain per round trip.

So what we are going to do is we would imagine we would take a photon, okay that originated from here, a fluorescent photon right. Initially, to start with what you have is that you have the media and you excited the media such that everybody has gone to the excited state. And there is because of that there is some amount of spontaneous emission that is possible from the level three to I mean level three to two.

During this spontaneous emission one of the photon, that is what the fluorescence says. And one of the fluorescent photon actually may take makes a trip from this media the position here, all the way down to this mirror and then comes back goes through this media and then comes all the way back here. So we are going to talk about one full round trip. In one full round trip, that is twice the length of the entire cavity.

Let us call this the distance between the two mirrors capital L. So we are going to estimate how much of the loss do you actually does the photon, what is the fraction of loss that the photon will encounter, when it goes through this entire trip. The idea here is I know the gain that the photon will have when it passes through the media.

And I also know the probability of loss or the fractional loss a bunch of photons will encounter. So in the whole system will set to gain only if the mean the whole system will set to operate in a to generate an output only if you have a gain higher than the loss. So we are going to equate that to tell you the to arrive at the threshold at which you will start seeing lasing. That is a very simple idea.

So now we have written down the expression for the gain. Now we are actually calculating the loss per round trip. So at the reflection losses for round trip would be two mirrors because the photon is experiencing reflection at mirror the photons that we started with is experiencing the reflection loss at M 2 followed by M 1. So what

we are going to have is $1 - R$ times I mean, again you will have another reflection loss.

So it is two surfaces. In general, we could what we could actually do is that we could capture all of this losses with a coefficient let us say γ . So this there is a reflection loss. Apart from this there is also some amount of loss because of absorption of the, so the γ basically encapsulates. This is a function of $1 - R$ epsilon lambda.

This is because of the absorption of the emitted light, right that we know that, that can also happen, epsilon. And also loss of these photon due to scattering and other such limitation right. So we will call scattering coefficient as some σ_s scattering. So all of this we will encapsulate into coefficient, let us call it as γ . This tells you per unit length, how much of the loss that are that the photon will suffer.

So if you formulate in that way then pretty similar to the Beer-Lambert's law that we wrote for gain, we can actually write this expression as I the intensity before at the start I mean intensity after round trip would be equivalent to $I_0 e$ to the power minus γ . So and so now we can actually combine these two expressions because this is because of the gain and this is because of the loss and to get the net result output right.

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$$I(\lambda, z) = I(\lambda, 0) \cdot e^{(\epsilon(\lambda) \Delta N \cdot 2L - \gamma)}$$

$$I(\lambda, 2L) > I(\lambda, 0)$$

$$\epsilon(\lambda) \cdot \Delta N \cdot 2L - \gamma > 0$$

$$\Delta N > \frac{\gamma}{\epsilon(\lambda) \cdot 2L}$$

Population inversion: $(N_k - N_g)$

loss coefficient of cavity

So when we do that, what we have is, I of lambda z we will write in terms we can write it as I of lambda at z equal to zero, okay. So it just amounts to taking the photon

from this end or we defining the 0 at the other end, it does not so we do that. Then can be written as $e^{-\gamma L}$ to the power minus of minus our actually plus $\epsilon \lambda \Delta N$. Let us write the gain. So $\epsilon \lambda \Delta N$ the population inversion.

The L here, see the, this L previously was representing the length of the media. But to be able to couple it with the γ , what we can actually do is that we can think of that is the length anyway for the interaction, assuming there is no interaction in the outside of the media, so you can generalize this L to be that of the entire cavity length. So you could write that as ΔN times two times the L .

Because it is one L is one half of the trip. We are talking about a round trip. So you start from a point and we come back to the point in the same pointing in the same direction. So it will be $2L$ and then times $2L$ minus γ , all right. So this whole thing is can be written like this. So you can see that one only if you are I mean for there needs to be a net gain. That is the whole point of this, right?

We want to have net gain. So which means I of λz or actually z equal to two times L , right. So for per trip right, so per trip we actually want to write it as two times the L . If you want to have this intensity greater than the intensity that we actually started with, so that can only happen if this number is greater than γ . So that tells you that sets you the condition for lasing.

$\Delta N 2L$ minus γ needs to be greater than 0 or in other words $\epsilon \lambda \Delta N$ or we can straight away write down the expression for ΔN . So ΔN needs to be greater than γ over $\epsilon \lambda 2L$ okay. So now this that this is the we know this is ΔN is the population inversion okay. Now this gives you the final condition. That is the ΔN that we are looking at.

It is not only sufficient to have a ΔN positive, but it also should be greater than this number. γ is the loss coefficient right of the cavity of cavity. This encapsulates the percent the fractional loss that the light suffers when it passes through the when it completes one round trip, okay. And ϵ of λ is the absorption coefficient and length L is the length of the cavity.

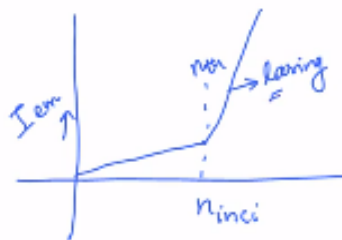
So here we are actually looking for ΔN which is the $N_k - N_g$. So only if you build up the population inversion above this γ by $\epsilon 2L$, you would see the lasing. So or in other words, let us go back to our graph describing the population inversion, the fraction of the population inversion as a function n_{inci} .

You can actually see the lasing will not happen as soon as the things cross over above 0, but there is a threshold and only if ΔN right this is equivalent to our γ over $\epsilon 2L$, okay. Only if the ΔN goes above this and only in this region you have lasing that is I am going to take the highlighter. Only in this region you have lasing and not, not when the light is present I mean where the ΔN is here.

No lasing here, alright. So unless so this is still no lasing even though lasing. So even though you cross, you have a positive ΔN , you do not see lasing because the loss dominates. I mean you are the gain is just about or less than the loss suffered in the cavity. Now this has an important consequence that is, if you have to actually plot the output of this measure the photon density that is coming out from this device alright.

So put it onto a photomultiplier tube or any photo detector and plot this output as a function of incident intensity, what you see is this threshold behavior.

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That is the intensity will kind of slowly increase until the intensity that you are measuring here is intensity of emission, okay. Now what you are measuring is that is basically that of the fluorescence. It will keep increasing until you reach a threshold at

which point you will suddenly start seeing a change in the slope which is very steep increase.

And then you call this as lasing and apart from this there are some certain unique properties to this entire laser light and we will talk about these properties in the forthcoming class. Thank you.