Optical Spectroscopy and Microscopy : Fundamentals of Optical Measurements and Instrumentation
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Lecture - 27

Hello and welcome to this lecture series on Optical Spectroscopy and Microscopy. In the last lecture what we saw was that how do we analyze the population dynamics in a two level laser system in the hope of seeing if we can generate a optically pumped two level laser system, okay. So I told you that when we did our analysis at the max what we see is that the population can be equally distributed between the level one and level two and not any more.

Often it is lesser. So as a result, you never exceed or you never generate more number of molecules in the excited state. Then if you do not generate then it is not possible to have a laser system. And this process of having more number of molecules in the excited state than the lower state for our emission, we call that as population inversion. And then all we are saying is that in a two level system, that is not possible.

And we went to a three level system and then there we were saying is it possible intuitively speaking, when you look at the energy level diagram, now what we have done is this is the following.

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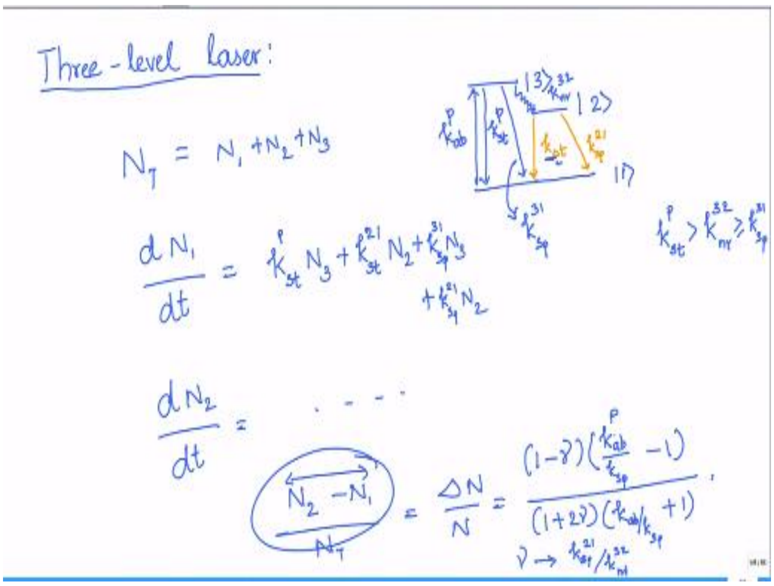

So in a two level system, what is happening is that the light radiation that we are actually using to pump is exactly the same as that of the light radiation that is actually inducing the stimulated emission. As a result, it is constantly competing the molecules is constantly competing to take the molecules from up to down or down to up.

They are, so as a result, what you have is that I mean the higher population in the level two being not feasible. In the three level system what it allows you to do is it allows you to decouple the state. So here what we are doing is that we are actually stimulating remember k ab is dependent on the incident light radiation. So is k st.

This is on the pump laser right the laser that is taking or the light that is actually taking the molecules from the ground state to the excited state, okay. So that pump light so to speak. So to differentiate that from the laser emission, we will call that as k p stimulated emission arising from the incident pump light. This is of a higher energy or higher intensity higher energy density.

So that the light in which is incident on this media can actually take the molecule to the excited state. Okay, good. But then the lasing per se right that also requires remember that also requires the stimulated emission that is the whole idea of lasing right. But that lasing takes place between level two and one. So what effectively it what it does is that, see this is also case stimulated emission.

Okay, there are spontaneous emissions I am not actually showing you in the picture. But you can actually think of this, think of them being here. So let us to be consistent with my convention I am going to draw this as a straight line right? I need to draw this as a perpendicular line to the energy states that is a stimulated emission. And of course the spontaneous states, right they are there, okay. The spontaneous emission is happening.

So what you can actually think of is, I mean, so there are different kinds of spontaneous emission. So one that is going like this the orange color sorry, the orange represents it is actually stimulated. So we do not have to write k st k st twice okay. That is k st while sp represents a spontaneous. Similarly, there is also a spontaneous emission that is happening from that can happen from here to here. Okay, so now this we call it as k sp.

Now there are two kinds of spontaneous and the stimulated emission, right. We want to differentiate these two. So what I am going to do is that, I am going to differentiate this in terms of marking it by the state from which they are actually making the transition. So here, this spontaneous emission is happening from three to state one, and this spontaneous emission the orange spontaneous emission is happening from the state two to one, okay.

So when you look at the literature or the books they typically where you see this numbers representing the levels to really capture where the transition is happening. So now we have listed out all the possible processes. And of course, this wiggly arrows represents non radiative decay from our level three to level two. And so we can call it as k nr and that means non radiative and it is happening from level three to two.

These are direct application or direct use of an abstracted Jablonski diagram you can think of because we are actually representing only the electronic levels not the in between vibrational and rotational levels. However, the process arrows, the rules following that we are actually sticking to that. So you can see how useful it is to actually not having to draw the potential energy diagrams.

But you can actually draw the lines and then see this state transitions. Now using this then we can actually write down the expression for the rates of these processes. And the first thing that comes to that we should keep in mind is the total now is distributed across three states, N 1, N 2, and N 3. And we can write down the expression for each of these rate process dN 1 by dt is going to be the influx minus the I mean the outflux.

So we can write here as k p stimulated times N 3 plus k st that thing in the orange right so I am going to switch back to that stimulator being written as subscript. So k st but now here the k st is going from 2 to 1. As a result have it going to N 2. And then of course the spontaneous rate constants come into picture, right? The spontaneous emissions going from N 3 and spontaneous emission that is 3 to 1, 2 to 1 of N 2.

Similarly, we can actually write down the rate equation for dN 2. What are we after? What we are after, blah, blah, but what we are after is actually this number, right N 2 – N 1. This number is what we are actually after and actually this N 2 – N 1 to the total fraction N T. See that is exactly what we had actually calculated right? N 2 – N 1 by N 1 it could be N T or N 1. This N T is constant.

So but you can always replace that N 1 as from through N T. But that is exactly what we are after. Because it is this number that we, that determines the number of atoms that can actually potentially undergo this lasing action. So now if you go ahead and solve for it, right, so I am going to just state it okay. So what we are going to do is delta N, which is N 2 – N 1 by N we can, the logic is exactly the same.

You will write down the expression for dN 2 dt and then say that at steady state, we are going to follow this dynamics. Then when you write down this expression is clearly is going to have as we would expect will have more terms and the terms I am going to just state the result here. You can please go ahead and verify it for yourself.

One minus gamma, I will tell you in a minute what gamma is, k absorption by k spontaneous minus 1 divided by 1 + 2 gamma k absorption by spontaneous plus 1, okay. So now that gamma here is oh, this is case spontaneous. So okay. Now the gamma is essentially ratio between case spontaneous between the levels 2 and 1 by k nr between the levels 3 and 2. Now let us look at this little bit more closely.

First thing we want to know is that can you actually generate can you have a positive delta N that is N 2 being larger than N 1. Is it possible at all? So now it turns out, yes, you can have the delta N to be positive. For you, for that to remain positive one thing we want to make sure is that this number k absorption by k spontaneous need to be greater than one.

So that is no problem because the absorption here we are talking about the pumping rate and then this is the spontaneous emission rate. So this can this is just a spontaneous emission rate right. So this definitely can we can actually increase the incident light intensity and make it 1. However, the important thing here is that you want to make sure that gamma is less than 1.

If gamma is 1, I mean or greater than 1, then the whole process is I mean you are not going to generate a delta N being positive, right? Because and what is gamma? Gamma is the ratio of the rates of the spontaneous emission from the between the lasing levels, like the 2 and the 1 and divided by the inflow rate the rate constant k, the non radiative rate constant, right.

So all it means is that, so you want that to be such that it is lesser than 1 which means the k nr needs to be greater than.

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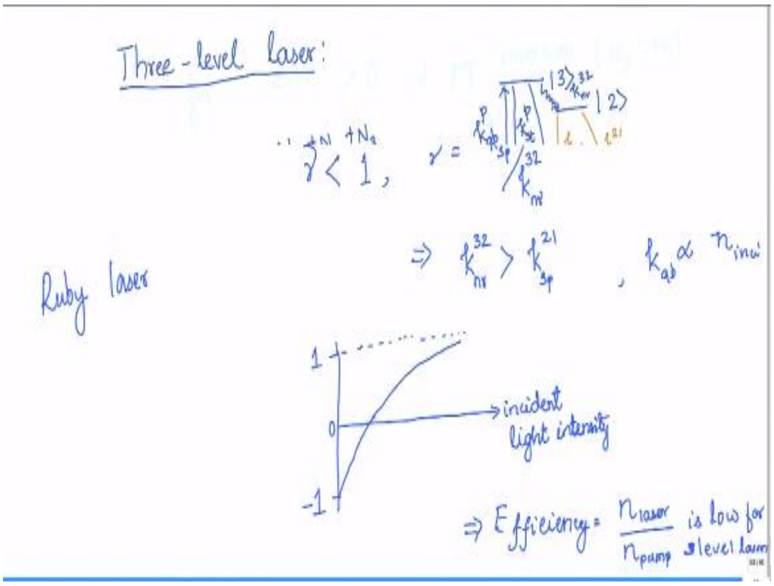

So for delta N to be, in the above equation for delta N to be greater than 0 right, which would imply population inversion right. That is N 2 greater than N 1. So which is essential for our generating net output or the stimulated emission out of the laser. So for that to be happening we see that the gamma need to be gamma needs to be less than one, less than one and gamma per se is equal to k spontaneous emission happening between level two and one divided by k the non radiative rate of 3 to 2, okay.

So for you to that that would imply the non radiative rate constant need to be larger than right that would make sense right. If you look at it, look at the energy level diagram what basically it says is you need to be able to populate this level two faster than the spontaneous emission, right? The k sp to 1 is basically the rate at which the molecules in the level two are coming down. Okay? As the I mean it is a rate constant.

So it is proportional to the rate at which the molecules are coming down. So you, when you say that the k nr needs to be larger than the k sp, what it means is that you need to be able to that the inflow here needs to be larger than k sp. So and the inflow can be larger by making the number of I mean, since it is a rate constant now it is a it is really a molecular property.

But essentially, what you are trying to tell here is that the net flow here that is happening has to be larger than the net outflow, that I mean the flow that is going out. So that you can have accumulation of the molecules here okay? But then the problem is that if it is a, we know that the spontaneous emission rate constant on the stimulated emission rate constants are related, right.

The, you remember the either the Einstein's coefficients or even our second quantisation right, any of them. So then you know that the both of them are related through the molecularly they are related through the r 12 the matrix element. So any change I mean, that actually reduces this is going to somehow affect the stimulated emission rate too. So we have to be a little careful about modifying this.

But then what you are looking for is to have the media where the k nr the molecules to transition from 3 to 2 is really high. You can actually do that by cleverly changing the matrix in which these molecules are embedded in a solid state laser, or in a if you are talking about a solution, then it is actually by changing the solvent properties, you can actually play around with the k nr and stuff.

But the bottom line here is that you can actually by changing by choosing the media in a specific way, you can actually make sure that the delta N is greater than 1. Provided again there is this k ab by k sp where the k ab needs to be larger than k sp. Where you have to as you keep increasing the incident power, you will see that it is only beyond certain incident power, you will start to populate the more number of molecules in 2 than 1.

That is the key here. Okay, delta N being greater than 0 is not just any population N 2 but the population here should be greater than population at 1. If that condition, you can see why it is not achieved straightaway, because the population in 2 is fed by population 3. So to feed the population 3, you need to take the molecules from ground one.

I mean from the ground level or level one. So as you keep increasing the intensity, they will populate here and then some of the molecules will go down and stay there before it can come down by stimulated emission or spontaneous. Now if you do it for a sufficiently long enough period of keep doing it for a sufficiently long enough, I mean period or I mean sorry, sufficiently high enough intensity, then you can actually end up having the molecules at the level two.

So the interesting phenomena here is that one can actually plot this inversion right or to say that the ratio that is delta N by N or delta N itself as a function of the incident light radiation. So now what you will see is to start with right, to start with all the molecules are on the ground state, right. So we are plotting delta N, okay delta N as a function of incident light radiation, okay.

Or let us say we are plotting the delta N by N as a function of incident light radiation. So to start with, what do we have is that N 2 – N 1 is basically –e N because N 1 is equal to N to start with very low intensities, extremely low intensity. So 0 in this axis incident intensity. As you will see that the delta n will be -1 because basically delta N by N will be -1 because what you are starting with as the everybody in the ground state. So it is N 1 = N T.

So as you keep increasing the incident intensity at one point, it crosses the 0 and then approaches closely I mean it is never going to reach there but approaches towards 1, okay. It will never be able to reach it because it is only a small, it is only a the k 3 2 okay k 3 2 by k st or k sp you will see that the molecule has three pathways to follow. And k st is larger than k nr typically than k stp. I mean than k sp 3 1, okay.

So in other words, you would see that k stimulated p, the order would be somewhat like this either equal or greater than okay. So now what it means is that you have to, you have to really increase the intensity, pretty high. So that for you to get any considerable number of molecules here, but then at that point in time, whatever the molecules that are present here need to be lesser than this, right.

That is what the population N 2 needs to be lesser than N 1. So you will never be able to take everybody every single molecule down to level 2. It is very hard, it is extremely hard, but so you would say that, okay, it never reaches about 1. It tends to reach but it never reaches 1, okay. And so the dependence of the incident intensity comes from k ab, right just to make sure that we understand what we are plotting and why it is dependent. So let me state it explicitly here.

We know that k ab is function of n incidence, okay. And that is what we are actually plotting here. So you would say that you would see that a large amount of light is used towards actually bringing the molecule or bringing the number of molecules equal between the ground state and the excited state.

So or in other words, you will see that the efficiency of this process efficiency defined as the amount of the pump light intensity that you are actually putting in to the amount of the emitted light intensity that you get out, okay. So efficiency you can think of n pump is the ratio between the other way round; n lasing or laser by n pump, okay.

If you see if you define the efficiency, you will see that the amount of pump intensity that you require to get the laser output is very low is low for this three level for three level lasers. Nevertheless, there are lasers practical, I mean, there are lasers with a three level system that can be approximated to three level system. In fact, we will see a little bit more in detail little later.

The first laser to be discovered, called as the Ruby laser from made out of the ruby crystal ruby gem is actually a three level laser, okay. But then the point is that, as we are saying it is actually very inefficient because lot of energy is lot of incident light energy is not I mean does not go into converting into the laser itself. On the other hand, I mean, the question that you would like to ask is, is there a way to improve?

Okay, clearly yes, there is a way to improve. The idea becomes very simple. And moment you realize the reason why you are having this trouble is because you are fighting against the number of molecules in the ground state. Yes, what you have done is you have created an extra state, thereby you can actually start accumulating so that your incident light does not compete with the laser light.

So I mean, that is accomplished with the stimulated emission. So you can actually start accumulating the molecules in the excited state. However, what is happening is that you are actually the laser emission is measured by the difference between the number of molecules in the excited state and the ground state and the lower energy state. So between which the lasing is happening.

So you are always having a larger number of molecules in the ground state. Naturally, because it is being supplied by the stimulated emission, spontaneous emission from here and then the spontaneous emission from here as well as this stimulated emission.

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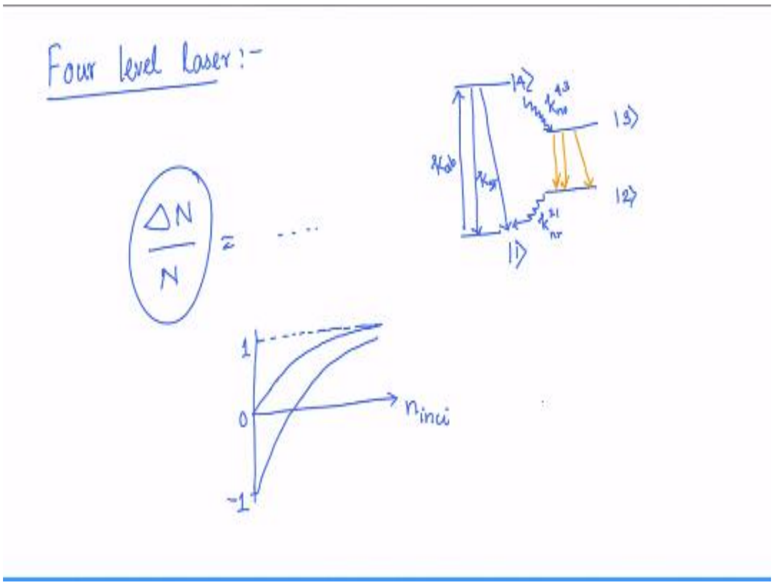

So if the natural extension to this would be suppose if we had a laser system wherein the lasing occurs between levels that is not inclusive of the original ground state, okay? So we could think of a laser system something like this where the lasing okay happens here, okay? Okay, and if that were to be the case, then you can actually see practically you can maintain this level, the level two population at 0, okay?

There is no restriction of having to have more molecules here because they are it is at the level. It is not the ground state it is a higher than the ground state. And it is been fed, the other the higher energy state is not the higher excited state to which the incident light is being pumping. So as a result, you are segregating that competition as well as you are segregating the level to which it is actually lasing on to.

So you have what is called as a four level lasers. So in this four level system, we can action the same thing happens. You have the pump laser that is operating between the one and level one and level four. So naturally there is this stimulated emission because of this pump okay. And there is also spontaneous emission supplying to this and in a non radiative manner. Why non radiative?

Because we are actually there is no light that can we make sure that there is no light that is matching this energy that is in the system, available in the system. So this non radiative manner we are feeding on to the higher level of the lasing electronic states, alright. So across which the lasing happens, okay. 4, 3, and then these are the stimulated emission. And here, you also have your non radiative, I mean the spontaneous and the non radiative I am kind of ignoring between the emission levels.

So let us not worry about that. And then once this comes, what we do is and like that, we can actually go ahead and write down the rate constants and describe this whole system alright? When you do that, and what are we after? We are actually after the same expression right delta N by N. And then we are actually asking how does this compare with respect to the three level system?

And what we are going to do is we are going to so please go ahead and get this expression down. So but what we will see is that the bottom line is when you plot this as a function of n incident, right? How does it look? Remember, for a three level system, we started with 1, -1 across the 0, and it was trying to reach 1 at some point, okay?

But what you will see is because this level, level two is kept close to 0 because of, so that demands the k nr 2 1 needs to be larger. Since it is kept close to 0, what you will see is that this in this level, you straightaway start from 0 and it can the delta N by N can only be positive, it does not go negative at all. And then it really reaches 1. It can actually effectively take everybody to level three okay.

So there are many lasers that are of four level that are can be approximated into a four level system. And we will see I mean, we will see these examples later in the course, but then I hope this gives you a flair of where I mean why we cannot take any material and hope to generate a laser but why we need to pay special attention to the nature of the electronic states that are available in the molecule and how what kind of analysis or what kind of properties are we actually looking at.

And once we say that, okay, this is what we are looking at, and this is how we can actually generate I mean this, this passes the this kind of serves as a quality check whether it can or cannot act as a lasing medium for a optically pumped laser. You can generate something else. You can pump it through some other means that is a different story.

But if you are using a light to generate a light of a particular kind, then we need to pass this quality check. So in the coming classes what we will see is that we will see the property of this light radiation itself and then we will talk about an important category of lasers, the lasers that categorization of lasers in terms of the light intensity as a function of time.

Light, the behavior as a function of time and compare it with other light sources, what is its special property and why it is useful in our equipments, that equipment that we use for scientific study. Alright. I hope I will see you in the next lecture.