Optical Spectroscopy and Microscopy : Fundamentals of Optical Measurements and Instrumentation Prof. Balaji Jayaprakash Centre for Neuroscience Indian Institute of Science-Bangalore

Lecture - 26

Hello and welcome to the lecture series on the Optical Spectroscopy and Microscopy course. And so far what we have seen is we have looked at the fluorescence and some of its properties. And one of the goal of this whole course is to be able to build an equipments and not just build equipment, I mean instruments, but be able to do that with the fundamental understanding of the principles on which they operate on.

So in that spirit, towards the end of the course what we will be doing is we will be looking at how to put together a microscope often used in a biological, to study biological systems. And when you are putting together a microscope one of the quintessential part of that would be a light source. So not just a microscope, even any present day optical instrument if you take, then fundamental part of such an equipment would be light source.

And in this respect, we would actually extend our analysis of what happens when the light interacts with the matter. And from there how we can actually generate and light, okay. So light of a particular kind. In here, we are going to be talking about a light that is originating from stimulated emission, okay. Let us look at it initially about some of the properties and how do we actually generate the stimulated emission itself, we know that already, right.

When we looked at the second conversation or the creation and annihilation operators of the photonic states and then let the interact, let the, we write down the way they interact with the atomic system.

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What we encountered is that the emission process itself is given by rate of emission, right. This is the transition from 2 to 1, okay, 2 to 1 transition. This is given by n k + 1 h cross omega k times R 12 I mean modulus square, right. All right. So from here, we I mean it is directly proportional to, we observed some of this constants and stuff like that. So let us write down as directly proportional.

So from here we said the spontaneous emission or the fluorescence originates because of this term, alright? Because even without any photons, you still have some finite probability or finite rate, which is I mean greater than zero of seeing this transition from 2 to 1 right because it is this is nonzero. On the other hand, we also looked at n k and then I said this is giving rise to stimulated emission, okay.

So with n k set to zero and then looking at the spontaneous emission, we said it is the origin of the fluorescence and then we looked at several properties of the fluorescence itself, right. So the isotropicity anisotropy of the polarization and nature of the spectrum what determines the intensity of various different transitions, so on and so forth. Here, what we will do is we will go into the other part.

Just I call I mean, you have two parts to it one is the spontaneous emission part and other is the n k dependent part, which is the stimulated emission part. What does it tell you? So if you have n k photons present, and who are resonating with the omega k and k is present, then the property of this is that the photon density of the n k goes up by 1, the photon number goes up by 1. That is the whole point here, right.

This is the point of stimulated emission, right. When you have 1 it becomes 2. But 2 of what kind? 2 of the n k kind. Means you can write down where k is the mode of the photon, right? Okay. K here we talked about the mode of the photon, okay. What do we mean by mode of the photon?

Essentially it means you can classify the photon in whichever the category that you would like to categorize them into in terms of polarization in terms of energy, which is the color of the photon in terms of the directionality at which with which it is coming in, and so on and so forth. When you do that, you club all of them with the similar properties into one mode and that mode, we call it as k.

And the fact that the stimulated emission goes up by 1 of that particular photon that mode means that the newly generated photon matches with the photon that actually stimulated the emission in every single aspect taken the polarization, the direction of propagation, the energy, everything is matched dot to the fluorescent photon that is actually, I mean the stimulated emission the photon that was incident on it, okay?

Thus the photon when you stimulate an emission by shining in the photon, then the photon number not only increases just like just like that, but it also increases in a very specific way. That is the resultant photon is exactly identical to that of the incident photon and their number goes up by 1. That is the consequence of this. Which means, in some sense, you would be able to sort of generate a bunch of photons of a similar nature all together.

Now that is very powerful, right? Because that is what it allows you to do is that you can I mean when you are generating such a bunch then that can have very peculiar properties and that is really how laser light, so to speak is different from any other light that is generated from the incoherent light source. So we will talk about the, I mean, you know in a little bit more depth later on the course, about the nature of the light, laser light itself.

But before that, what I want to stress on is that the since it is going to increase the number in the kth mode and nowhere else and the kth mode is very precisely defined

you have identical photon that is generated because of this stimulated emission. Now how can we utilize this to generate more and more photons of the same kind. That comes down to the issue of okay now let us suppose we have a lasing let us call it as a medium, okay?

The media here is essentially it is a media that is capable of absorbing the light and emitting it, it is basically you should be able to see fluorescence. This is the spontaneous emission. And now the idea here is somehow in this media, and if this media I am going to I mean again go into representation in terms of electronic states as a two level state, right? I mean, there is the ground state and excited state.

Now the point here is that somehow, if I were to populate if I had to generate if I am able to generate a large number of molecules, okay present in the exciter state and then I come in with a light radiation, okay that resonates with the energy gap okay within the resonance band of the energy gap, then what I end up generating is that I generate I increase the intensity of this by 1 for every interaction that is every photon that actually interacts with it, okay.

Okay, so now what I could do is that I could take this media and then let us say I can put them between two reflecting surfaces. So I am going to represent the reflecting surfaces as something like this, okay the media has been classified too. So now when you do that, okay, what we are able to do is what we are able to create is that the photon the fluorescence that is being generated from the media because of higher population in the state, I mean, state 2.

Some of the photons are somehow I am able to not only generate but also continuously be able to keep it there let us say in a hypothetical case. Some of these photons are naturally going to go and come back again, okay? So because the angle of incidence is such that, it is going to reflect back into the and take back the same path. So in which case, these photons end up increasing the number by one.

While the other guys who are not in line with this are not going to be able to engage in this kind of a process. They do not increase the number by one, but not just by one, that is one for a single pass, single pass of interaction. So the increased photons then keep I mean goes and gets reflected by this.

Remember, what you have done is that you have actually given a selective increase, selective amplification, selective increase in number for photons of which are having a particular direction, right. The direction that sends them back into the media again. So which means when you have a reflector on the other side, which is identical to the first one, so then what is going to happen is it is going to reflect it back.

So thereby the photon that originated and that is aligned along this axis, either this way or that way, either ways, is going to selectively increase its number further and further, thereby resulting in extracting more amount of energy so to speak, increase its density by bringing the molecules down from level 2 to 1.

And this increase in density and then the resultant creation of a homogeneous photons of a mode k or mode l whichever mode that you like to call gives rise to a special kind of a light, we call it as laser light. Now this requires that you be able to create a state where the state 2 the higher energy state is it is more populated than the ground state, okay.

If that is not happening, then what we know that already that if you have a predominantly larger ground state as we normally see in a room temperature, then you do not see this happening because since there are more number of molecules in the ground state, the photon that you are shining on to this media is busy taking the molecule from the ground state to the excited state.

And then the one that is on the excited state can either come back by interacting with another light photon of the incident light or it can come back spontaneously, but that does not necessarily increase its increase the number of the incident light photon because it is just the probability that it will take the probability that the light photon will be absorbed by the ground state is larger, because simply because there are more number of ground states here, right. I mean sorry. So as a result, what you will see is that it is equally likely to interact and the interaction probability is exactly the same. However, the number of molecules or number of molecules are higher in the ground state. So you will see a net absorption process. So to defeat that what you need is you need a higher population on the excited state.

Now that scenario is not always possible, particularly when you are pumping through optical means, when you are trying to generate this population by using light itself. So now let us investigate that process a little bit more closely.

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Rate
$$d$$
 $absasp^{2} = k_{abs} N_{1}$
 $V_{abs}^{abs} N_{1}$
 $V_{abs}^{abs} N_{2}$
 $V_{abs}^{abs} N_{1}$
 $V_{abs}^{abs} N_{2}$
 $V_{abs}^{abs} N_{2}$

So what we are going to do is we are going to set up these two state system. We will call that as 1 and state 2. The number of molecules in the state 1, we are going to call it as N 1. Number of molecules in state 2, we are going to call it as N 2. And the total number of the molecule that is available to us is N T which needs to be N 1 + N 2, alright. Now the N 1 and N 2 are a function of time. Why?

Because when we start shining in incident light radiation, say h nu matching with where the nu matches with that of the delta 12 or 21. Both of them are the same. So E 12. So then what is going to happen is that the molecules are going to is going to get excited, alright. And the rate of that process, rate of the forward excitation process, we can write down is given by, is equal to k absorption times N, okay.

N 1 is the number of molecules in the excited state alright. Now k absorption is the rate constant, molecular rate constant. So it is naturally dependent on the matrix element squared where we have written we have done it before, which is R 1 2 square times the number of photons that are incident on it, right. There is an intensity dependence.

So if you increase the intensity you can actually modulate the number of molecules that are going from the ground state to the excited state by modulating k absorption. Good. So now similarly, we can write down an expression for rate of emission. Now this we know that there are two kinds of emission. So we are going to write down the emission rate of total emission. Let us, we call it as the stimulated emission.

Is given by this term plus spontaneous emission, again goes as N 2. However, this is again given by R 2 1 modular square times n inci, okay. So now what we can do is that we can, what we are interested in is to ask the question so once I turn on the light and then keep shining the light, would there be any situation no matter for whatever the intensity of the light I keep increasing the intensity or whatever it is, is there an I mean, but once I increase I wait there for some time for the system to reach a steady state.

Is there a possibility of me getting a steady state where the population N 2 is larger than N 1, right? That is the key here right, the key for observing stimulated emission, laser emission is that we need to have some we need to have more number of molecules on the excited state or the state 2 than 1. So what we are asking is, is there for any intensity n in s, is there any way I can get more N 2 than N 1, right.

So why incident intensity because that is one of the governing fact. Given a system R 1 2 is fixed. So the only way I can change the number of molecules in the N 2 is by increasing my n incidence, but that has its way of governing the population in a slightly non trivial manner because the when I increase the n inci, the rate of absorption increases no doubt about it, but also the rate of emission. So now how are they going to play out?

And would we ever have a situation where N 2 is greater than N 1? That is the question that we are asking. So how do we ask that? We can actually looking at steady state population for a given n inci. So what do you mean by a steady state? Steady state is when n in dN by dt equals sorry, not n in but N 1 is when these two become equal, then we say basically the rate of efflux and influx both of them are same.

And we say we have reached the steady state. So let us represent that in the picture. So these straight arrows are intensity dependent processes, while this is fluorescence. So we can write down that as 1 over tau f. So given this, we can actually go ahead and solve for the population of the states are the steady state right. So we can write down N 1or other N 2 steady state population, we would like to know what it is.

And also N 1 steady state, okay. So in order to do that what we are going to do is we are going to use this to write down the expression for the rate of change of population of the first state N 1. So we have -k a N 1 k st N 2 tau f where 1 divided by tau f, why the tau f is the fluorescence light time. And hence 1 over tau f is nothing but k the rate of spontaneous emission. Okay, so now that is for the dN 1.

So we want to make sure that at the steady state, this is equal to 0, right? S means steady state. So how do we do that? So we set it to 0 solve for N 1 and N 2. When you do that, what you will see is that you get an expression N 2 s steady state at steady state is given by k st plus 1 over tau f equals k ab N 1.

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 $\frac{N_{2}^{58} - N_{1}^{55}}{N_{1}^{59}} = \left(\frac{k_{ab}}{k_{st} + k_{sp}} - 1\right) < 0$ $\frac{1}{N_{1}^{59}} \times \frac{1}{N_{2}^{55}} \times \frac{1}{N_{1}^{55}} = \frac{1}{N_{1}^{55}}$ $\frac{1}{N_{2}^{55}} < N_{1}^{55} \times \frac{1}{N_{1}^{55}} = \frac{1}{N_{2}^{55}}$ $\frac{1}{N_{2}^{55}} < N_{1}^{55} \times \frac{1}{N_{1}^{55}} = \frac{1}{N_{2}^{55}} \times \frac{1}{N_{1}^{55}} = \frac{1}{N_{2}^{55}}$ $\frac{1}{N_{2}^{55}} < N_{1}^{55} \times \frac{1}{N_{1}^{55}} = \frac{1}$

Or in other words we can rearrange that for N 2 at steady state minus N 1 at steady state divided by N 1 at steady state which can be given as equal to K ab divided by k st plus k spontaneous minus 1 which would imply that N 2 see now you can actually see that the k ab, so this whole term that N 2 - N 1 by N 1 so which would imply that the N 2 has some limits, right. What is it?

The limit is that you see that we know k st the molecular rate constant for the stimulated emission is equal to ab okay. And certainly, the k st plus k sp the spontaneous emission rate plus the stimulated emission rate can at the best be equal to the k ab or more. It is never going to be less than k ab. As a result, this whole expression will always be lesser than 0, this is -1. So this whole expression will always be lesser than 0.

Which would imply N 2 s will be lesser than N 1 at steady state, or in other words, optically if you were to pump a two level system and expect to have a larger population at N 2 now that is not going to happen. It is at the best, the both the ground state and the excited state might reach the same number of, might have same number of molecules or same population level.

But it can never be the situation where the higher level is more populated than the ground state ruling out the possibility of an optically pumped two level system. Of course, you can always generate a two level lasing system where which is not

optically pumped. If you are pumping through some other means, then you do not have this problem of you do not necessarily have this problem of stimulated emission.

If you do not, if you again end up having the same situation where the interaction is such that it the interaction does not differentiate between the absorption and the emission process then this is not possible at all, okay. So, or in other words, you cannot have an optically pumped two level system or optically pumped two level laser. That is not possible. Given that then how mean what is the next step?

What do we do then is that we can actually look at a three level system and ask if in this system we are able to generate higher density of the excited state. So now for that we can exactly do the same thing, right.

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That is to say, okay I am going to write down my laser media as a three level system, okay. Now when you do that we are talking about levels 1, 2 and 3. The idea is that the absorption on stimulated emission happens between the levels 1 and 3, okay. Okay, and the lasing per se happens between level 2 and 1 and the molecules do come back from the level 3 through a non radiative transfer, okay.

So now for this, we can actually set up the same rate equations. So N 1 being the number of molecules in the ground state N 2 the second state and N 3 the third state. So what we can do is that we can actually set up the rate equation describing the

population dynamics of these three level system. We will write down this rate equations in the next class.

And then what I am going to do is I am going to state you solution of this system. And then we will look at the properties, interesting properties that emerge from the system and then later compare with one other I mean, four level system too. All right, I will see you in the next class, next lecture.