## Ultrafast Passes in Chemistry Prof. Anindya Dutta Department of Chemistry Indian Institute of Technology-Bombay

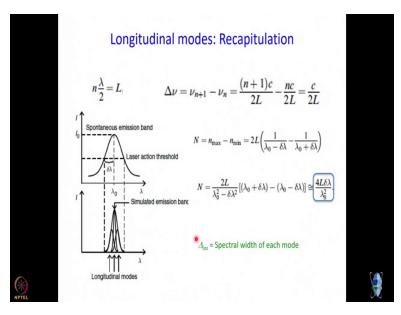
## Lecture No. 21 Modelocking for Short Pulses

Slowly going towards understanding how ultra-short pulses are produced, and on our way, we have discussed what longitudinal modes are whatever discussion we are doing now is completely.



From this introduction to laser spectroscopy this is the cover of the book it is not very costly 1500 rupees now used to be 8000 at one point of time. So, this is what we are going to follow by and large for next few modules.

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So, to remind ourselves what we have learned is that a laser is we always think that laser is monochromatic, but what we have discussed is that it is strictly speaking most lasers are not because they comprise of longitudinal modes, what is the meaning of longitudinal modes, we said that to set up a standing wave, the essential condition that has to be fulfilled by a wave is that the cavity length l must be an integral multiple of half wavelength n lambda by 2 equal to L.

So, there can be many such modes that are that can be there in the laser. As we discussed earlier, we are not really dealing with n = 123, we are dealing with n = 123 equal to very large number 10 to the power 6 to the power 4. So, 10 to the power 4 + 1 is almost into the power 4. So, wavelengths of modes that are adjacent to each other do not really vary too much. That is the first thing to understand.

Secondly, we have worked out the expression for delta nu where delta nu is the difference in frequencies of 2 successive modes? Mode number n and mode number n + 1. And that turned out to be C divided by 2L. And the C divided by 2L is actually an interesting quantity. What is it? See what is 2L by C? It is a round trip time so it turns out that delta nu the difference between 2 modes, 2 successive modes, is equal to round trip time, inverse of round trip time rather.

If you do not remember this, you better write it down. We are going to need this later on in this module or next. And this is another thing that we said that if you think of the spontaneous emission

band, spontaneous emission spectrum, and if you think of stimulated emission spectrum, we discussed why stimulated emission spectrum is narrower. We said that let us say that this laser action threshold is somewhere here.

And we said it is about at half the intensity, we said if the half width at half max is delta lambda, then delta lambda is going to be the half width at the base of the stimulated emission band. So, now we are really talking about not full width half maximum, but pulse duration, which is the time for a pulse to go from 0 to a maximum to 0, well, in this case it is not really time, we are talking about lambda, but as you know frequency and time are interconvertible by Fourier transformation. We are going to invoke it in this module or the next one is another thing that we have studied.

We asked that if this is a spectrum, off stimulated emission how many longitudinal modes are there and the answer was capital N equal to approximately 4 l delta lambda divided by lambda 0 squared. What is lambda 0 squared? Lambda is the wavelength where the spectral maximum occurs and what is delta lambda? Half width that have maximum of spontaneous emission. So, it is basically half width at base of stimulated emission band and what was that capital delta nu that we talked about a little while ago delta nu what was the delta nu is the free difference between frequencies of 2 successive modes.

So, please do not confuse that capital delta nu with this small delta lambda. This delta lambda is for the entire spectrum for the entire simulated emission band that capital delta nu is the difference in frequencies between 2 successive longitudinal modes. I have this bad habit of saying normal modes all the time. So if I say normal mode by mistake, I really mean longitudinal modes, I don't really mean normal modes of vibration or any such thing.

So this is another important quantity that we are going to use. So if you do not remember you do not take I do not expect you to remember it, but better write it down is going to come handy later on total number of longitudinal modes is four L delta lambda divided by lambda 0 squared. And then one thing that I would like to draw your attention to which we did not say in the last couple of modules is do not think that the longitudinal modes have spectra that look like delta functions.

So far, what are we saying we are saying that the nth longitudinal mode has some frequency nu n. The next one in (n+1)th longitudinal mode has frequency nu n+1. So, that might give you the idea that these are delta functions, but that is not true. Each longitudinal mode has a spectrum, which has finite width. Do you agree or do not agree with this? Let me rephrase the question. Is it ever possible to have a spectrum which is a delta function in the truest sense, no, why not why can we never get for any system a spectrum which is a delta function.

And here I am talking about energy domain spectrum. So, this takes us back to the semi classical treatment once again, we keep referring to it again and again in this discussion, time dependent perturbation theoretical treatment of interaction of radiation with matters. So there if you work it out, you will see that of course, you have a spectrum, there are many line broadening mechanisms that can take place, some of them are homogeneous, some of them are inhomogeneous we are going to talk about homogeneous and inhomogeneous line broadening mechanisms later in this course.

Because you cannot have a course, talking about lasers and not talk about line widths and in the next couple of modules, we get sort of get an idea why. So, can we name some line broadening mechanisms? I will tell you 1 collisional broadening right, molecules that collide with each other, the transfer energy and so, energy staying and that is why spectral line gets broad, Doppler broadening somebody said Doppler broadening is also correct so what is the doppler broadening.

It is best understood in terms of emission, your system emits light and it goes towards the detector. Now, if the molecules are also moving towards the detector or away from the detector, then the effective length changes and effective frequency changes. Very much like what we studied in class 11 and 12 physics, whether the train comes towards you the whistle sounds more shrill, more shrill means frequency is higher, and when the engine goes away from you, it sounds less shrill, because there is more space in that time and the wave is important that is called Doppler broadening.

So, Doppler broadening is an important issue in spectrum in spectroscopy or suppose I say that I have a situation where I completely stop collisions. If I freeze the sample, nothing is moving. And since nothing is moving, Doppler broadening also does not arise. Still there is some mechanism

no matter what you do, you cannot get rid of one thing that will invariably add to line width. Let

say I have quenched vibration, of course I can never quench vibration because it will be in that

v=0 state.

So, the mechanism that you cannot do anything about is natural broadening or lifetime broadening

right It sounded like you can if I put it very qualitatively ground state has a lifetime of infinity let

us say and excited state has us has some lifetime. So, of course, there is some uncertainty in time

and we know uncertainty principle uncertainty time multiplied by uncertainty and energy is a

constant.

So, what is the maximum answer in time allowed associated with the excited state cannot be more

than its lifetime. My height is 5 feet + - 30 feet. Does it make any sense? It does not. If I say my

height is 5 feet + - 5 feet, then also it does not make sense to me for me, but it does make sense for

quantum mechanical objects is you can only do so much. So, that is the maximum limit on

uncertainty.

So, since that uncertainty is limited, you will have some uncertainty and energy also, you cannot

have delta you almost equal to 00. So speaking very qualitatively, that is what gives rise that is

why it is called lifetime broadening, so some broadening will be there. So, it is important to

recognize that each mode each longitudinal mode also has a spectrum that is not a delta function,

at least natural line broadening will be there. Let us say that is delta las. My problems is that I use

such a color I myself cannot read. Anyway, so delta las spectral width of each mode.

It is important to recognize that it is there Now, next what I will do is I will take a little bit of a

detour. We will come back to this mode locking business mode locking is a technique we want to

discuss today by which you can produce ultra-fast pulses. But there is another method by which

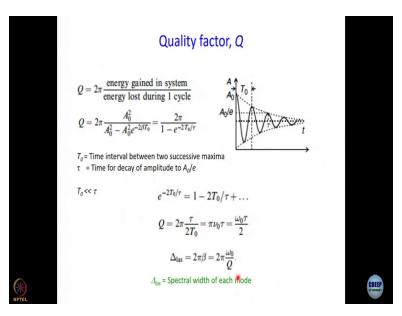
you cannot perhaps produce an ultra-fast passes but you can produce nanosecond pulses or so. So

it is called Q switching your light. So what is this Q where did it come from? Is it james bond Q

or what is it? So, most likely will have to come back to this issue of Q. But let me for the next 4

minutes at least provide a glimpse of what it is. Q is quality factor.

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And it is defined like this 2 pi multiplied by energy gained in the system divided by energy lost during 1 cycle why will energy again be there in a laser, we are talking about laser of course, what is the mechanism by which energy gain can take place and when I talk about Q I am talking about the cavity, so, what happens is if we go back to the very basics of lasers, you have these active medium that is giving out light right and it is doing round grips in every round trip the number of photons is getting increased.

So, in other words energy is increasing right that is called gain and how do I get lost? I mean, I do not get lost how do I lose energy? How does the cavity lose energy? One thing you can think is suppose now light goes out of the cavity, there is actually a loss. That is what we want, what as far as the system is concerned, it is a loss, but for now, let us not even talk about light going out from there.

If you even put 200% reflecting mirrors will it just keep on gaining energy or will there be some will invariably be some loss why because system will get heated up heating is a very big problem in laser. So, you see, you have to use things like chillers and all many times when you use powerful lasers right there can be defraction there can be scattering. So, there can be many mechanisms.

So, this Q is given by two pi into energy gained in system by energy lost and when I say Q switching What do I mean then I mean that for some time I let the Q go up that was energy is

getting built in the system, then by some mechanism I switch the energy out. So, what will happen at that instant Q will fall so, Q will switch from a very high value to very low value then again Q will start building up.

So, if you can somehow achieve this that is one way of producing pulsed laser, Q switching how we achieve this we are going to discuss when we talk about the actual instrumentation part. But the point is, since there is some loss, it is no longer fair to think of this wave that I have as just a plain wave. It is good to sort of damped oscillation if there is a loss. So do something like this, do not worry too much.

The purpose is not to do every bit of math that is associated, but rather to get a working idea. So, we will take things axiomatically, but these are not all that difficult also. So something like this oscillation and there is a decay. And this figure here is not really to scale. It is not to scale. Because this t0 that you see t0 is basically the time difference between 2 maxima time interval between 2 successive maxima.

Time for decay of amplitude from A0 to A0 by e, does that ring a bell? it sort of something like lifetime time constant. Now generally t 0 is actually much smaller than tau otherwise you are very bad laser if it decays completely, even before doing a cycle, how will it work. So, this figure is just is not to scale is done in such a way so that you can see all the quantities involved that is all, but is it is actually much smaller than tau and we are going to use it shortly.

But let me write something, let me write this 2 pi then what is energy gain in the system A0 squared. What is energy lost in 1 cycle it will be A0 squared - A0 squared into e to the power - 2 beta T0. For now do not worry about beta is we will have to come back to that we can go to the final expression is A0 square will cancel between numerator denominator, you are left with 2 pi divided by 1 minus e to the power - 2 T0 by tau, which is a very familiar form of the equation for us. Now, we can simplify this expression a little bit precisely because T0 is so much smaller than tau. So, what will happen if T0 smaller than tau how do I expand e to the power - 2 T0 by tau.

And this is a technique that we have used in almost all physical chemistry courses. If you do

quantum mechanics, statistical mechanics, whatever, you always use this kind of an approximation

t 0 is much, much smaller than tau. So how do I expand e to the power - 2 T0 by tau? It is about -

x when x is very small. One is correct. Second write something like this of course, there are higher

terms but as it always happens are we are saying that this itself is very small.

So, I higher terms are even smaller. So, we neglect them. So, we can just put e to the power minus

2 T0 by tau = 1 - 2 T0 by tau and if I take this expression and plug it in into the expression of Q.

What will I get? Very convenient 1 and 1 will cancel each other what am I left with something like

this now, so, 2 and 2 will cancel of course, 1 by t 0 what is t 0 is time period right what is 1 by t

01 by time period frequency and of course the answer is also in front of you. pi nu 0 tau.

pi nu 0 is the angular frequency we are going to use angular frequency time and again here because

we are dealing with periodic functions, they are going to repeat after regular intervals. So, angular

frequency very often turns out to be an easier parameter to use. So you get omega 0 tau by 2. Now,

next part I am not going to derive I will just tell you that delta las what is delta las spectral width

of each mode is given by 2 pi into omega 0 by q into omega 0 by q.

So, this is the expression for spectral width of each mode. The purpose of this discussion is twofold.

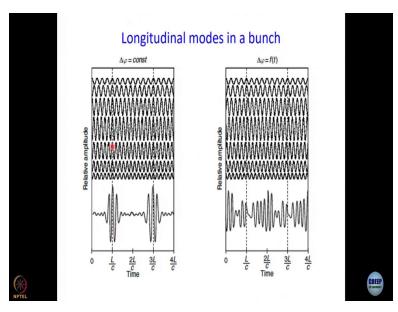
First, to emphasize the fact that longitudinal modes do not have delta function spectra. Second, to

introduce this important parameter quality factor Q, which plays a very important role in producing

larger pulses. And also, as we will see later on, the same kind of device is used when you want to

amplify your laser. So, Q factor is something that will come back again and again.

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Now, let us come back to our discussion where we stopped in the last module. We said when we take longitudinal modes in a bunch, what do you expect to get and we have talked about 2 situations, 1 in which delta phi is constant, what is delta phi? Phase difference between 2 successive longitudinal modes constant means it is independent of time and second one where delta phi is a function of time. So, let us take the second one first. If phase difference is a function of time, then you know very chaotic situation there is no correlation.

Now it is there now it is not there. So you are going to get something like this. You are going to get a free running laser with inherent fluctuations that might be there. However, if delta phi is constant then what happens? We did not discuss it in the last module, but we did not really spend too much of time on it. So, for the benefit of those who might be new to something like this, it is not very difficult to understand first of all, it is understood quite is simply by using simple day to day analogs.

Think of A team of runners, 5 runners and they are running around the field running around, they start together and they start together. They are in phase when they start running, somebody runs faster, somebody run slower, what will happen? There will be a spread right then it is possible that they are going to come back altogether. If they keep on running at the same speed for a long, long time, there will be times when they come together.

There will be times when the move away from each other. It might be difficult to believe if I am

talking about 10 runners, but it is absolutely easy to understand if I talk about two. right suppose

souradip and I when I run a race around gymkhana ground what will happen? Of course, he will

win. As he can run faster, he will not win. So let us set an agenda. They will definitely win. So

what will happen we will start off together then we are in phase? And then what happens Tanuja

starts taking a lead, and I start lagging and then if our speeds are sufficiently different.

What will happen? Eventually she will catch up. I will have done 5 laps, she would have done 6

and she would have caught up with me. So, that time we are again in phase whenever we come

together we are in phase when we start moving away from each other be facing starts and when

we are diametrically opposite in the field that is absolute destructive interference. So, now just

extend this to many runners, that is exactly what happens when delta phi is constant. Well, what I

discussed is when delta phi becomes 0, delta phi = 0 is easier to understand perhaps, what will

happen is, at this time.

Let us say, all the waves are in phase all the maximum match, then what will happen? X axis is

time, they are different frequencies. So, when you go away from here, some of these amplitudes

will decay slower, some of these will decay faster. So, phase difference is there but then what will

happen is you do not have constructive interference anymore. So if you look at the resultant

amplitude that is going to fall, it might do some beats up and down and then it becomes 0. After

some time, they will come together once again.

This is something that you encounter very frequently, when you discuss things like NMR

spectroscopy in time domain, then you talk about interferograms. So, the important thing here that

we are going to use is Y axis is amplitude. What is the relationship between amplitude and

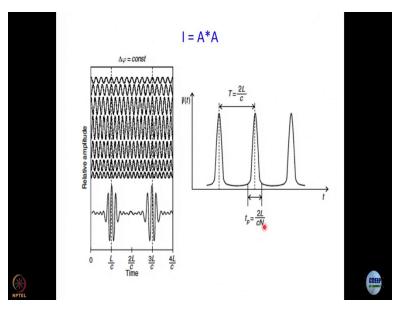
intensity? What is the relationship between amplitude and intensity? Actually mod squared feel

does have an imaginary component. So, mod squared if I take mod square or something like this,

what will I get? Everything will be positive, right it will become sharper. So we get something like

this.

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And as we are going to see in the next module, we do not exactly get something like this we get a little more structure in each of the pulses. So, what the crux of the matter is you get pulsed operation and we are going to show in the next module that separation between 2 pulses is going to be to 2L by C. So, what I read into this may be a little confusing. Did I say the opposite. A\* means complex conjugate. So, separation is going to be 2L by C roundtrip time which is great.

This is why we can do mode looking actually the separation between pulses actually a round trip time. And then pulse duration as we are going to see in the next module is 2L divided by c. What is the meaning of n? capital n? number of longitudinal modes that are locked, this is what brings us to mode locking, if we can lock a large number of modes, then we get short pulses, because see n comes in the denominator if n is large, then TP the pulse duration is going to be small. If n is small then pulse duration is going to be large.

So, what do you need? Do you need a single mode laser or do you need a multimode laser? what is the meaning of a multimode laser? if I put it in simpler terms, the bandwidth should be large. And it is the bandwidth here I basically mean the spec spectrum should be as broad as possible. The spectrum is too narrow then that will mean you will have lesser number of longitudinal modes there set. So, that is 1 essential condition.

That is why it is not so easy to make a an ultra-fast laser using a gas as an active medium because it has very sharp lines, you need something that has a broad spectral band, otherwise it will just not work. And so, we see that they are related to each other and ultrashort laser is always going to be a broader band laser. So, if you we are going to go back to the lab and we are going to demonstrate the way we know that our laser has become pulsed is that we were going to discuss how you can measure the pulse width also using something called auto correlator.

But we do not even do that. All we do is we look at the spectrum is the spectrum is very narrow, then we know that it is not pulsed. If the spectrum is broad, then we know it is pulsed right. So that in the next module, which is going to be perhaps a little smaller than what it is, what our models usually are, we are going to sort of derive these expressions, even though T equal to 2L by C might sound an obvious conclusion.

But we will still derive it but while deriving it we are going to jump stairs, we are going to take things actually axiomatically. The idea is we want an overall picture of why things are, how they are. And another thing that we learned is that this the way we have drawn it here is actually a simplification. You get a picture like this, when you lock small number of modes. When you lock a large number of modes, you do not get something, you get something that looks a little different. That is what we are going to discuss.