Ultrafast Processes in Chemistry
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Lecture # 20
Longitudinal Modes

Okay, now we will begin our discussion on longitudinal modes. But before we do so, we have a question, what is the question? The question is how to find out people to find out peak power. It is important to realize that what we see here, it is the power of energy per pulse, it is basically area under the curve. So, from there suppose the pulses triangular it is easier to understand triangles as I am saying triangular it will come to gaussian suppose a triangular pulse so it happens then your area is half into base into altitude.

So from there if you know that we are taking 200 femtosecond to be the width, there is a base, and you can find out what altitude is and here at the point to remember is that the base is really very small. So half into base into altitude, base is small. So altitude will automatically become very large. And now for a more realistic situation where we have a gaussian kind of distribution there, what we will have is we are not going to have half into base into altitude.

But even gaussian has a line shape it is I 0 intensity at the maximum position multiplied by a gaussian distribution, e to the power minus x squared by 2 or something like that. So from there, we will still be able to find out I 0. That is how you find peak power and for small pulses, and same kind of energy peak power is really, really high. Damage threshold is usually peak power, not average power.

Because damage is a result of one to one interaction of light and molecule. So, sometimes what happens is that you know what kind of laser you are using so in that case, your peak power is proportional to the average power and proportionality constant is known, the system is known, then average power may be mentioned. But what you really have to worry about is if you are short pulses, peak power is the killer.

If you are worried about damage of your system, sometimes damage is good because you might want to do lithography or you might want to do laser cutting or something like that. In a favorite example of mine, nothing to do with spectroscopy to be honest. I do not exactly remember the year most likely sometime in 2003 or 4 or something like that, there was this interesting paper in which somebody had manufactured a bull.

Bull the animal that is rampant in our campus bull, he cow a bull. Now, of course being an IIT Bombay, one might wonder what is the need of manufacturing bull there, God has manufactured plenty, and they have provided plenty of problem for us all the time. But this bull was special, this bull was a Nano bull it would not provide too much of a trouble to you. And the way they walked, they just wanted to demonstrate there is no reason for making a bull otherwise you cannot even see that unless you put it under a microscope.

But the way this bull was prepared was that a pulse laser was focused on to this monomer solution and the idea is that the light that you put in it is going to cause polymerization. So, wherever the focal point is, that is where polymer will be formed. And then you move the stage carefully. And the polymer bull is formed. It is nice, very, very good, nice, good looking bull, actually, you can read that paper.

But then the point that I am trying to make here is that it is focused right. And you talk about damaged threshold, this is not damaged, this is the threshold that is required for the reaction to happen. And then if you want a very good resolution, so here, the trick that was played was 800 nanometer light was focused, but actually 400 nanometer light was required for polymerization to take place. So polymerization took place as a result of two photon excitation.

Now two photon cross section is actually square of intensity right, so it is narrower than the actual pulse. That is why better spatial resolution was obtained by focusing light not a 400 nanometer but of 800 nanometer. I did not, perhaps we digress a little bit to answer that question. But intrinsically they are related something that can destroy can also build if it is used properly. Now, let us come back to a point of discussion, we want to talk about longitudinal modes.

Now, you might think what is going on here? What is where did this come from? We were talking about lasers, what about pulse lasers, you are saying that you pack too many photons in all this discussion, what is the meaning of not longitudinal mode and what is the relevance we cannot say what the relevance is right now, you have to wait maybe 15 minutes or 20 minutes to arrive there so, let us first understand what these modes are as we mentioned in the previous Module.

There is something called transverse modes, transverse mode means spacially, what does the laser beam look like? If you take the laser beam laser spot on a piece of paper? Do you get a circle? Do you get a dumbbell do you get something like a d orbital and they are depending on the number of nodes along x & y direction, you call them TEM00 01 10, so on and so forth. Most, but right now, well, they are important. We will come to them later. Now we want to talk about longitudinal modes longitudinal means modes along the direction of propagation of laser light.

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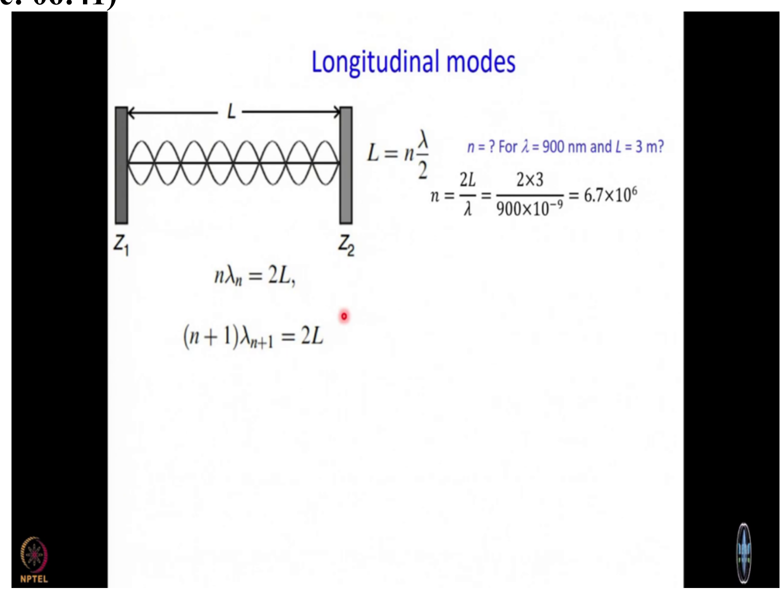

So now, it is important to understand that for a given cavity, not all wavelengths are going to be sustained. Because in order to get a beam where light is going back and forth, you must have constructive inference right standing wave has to be generated and condition of standing wave is that the cavity length l, this one must contain an integral number of half wavelengths l equal to n lambda by 2 this is very well known I think all of us understand that so, the point we are trying to make is that you make a laser l is defined you decide already that a particular set of wavelengths can be sustained.

In principle is number of wavelengths is in finite because n can go from what is the smallest value of n cannot be 0,1 and get in can go from 1 to infinity, but then also you have some active medium, which has a special bandwidth. It is not going to give you 1 to infinity. So, how many modes are there in the bandwidth that we are using, that is something that we learn first while we are learning all this will see by the end of this module hopefully.

So before going further, I want to calculate this ‘n’. And the reason why I want you to do this is it is very easy to now think that, l equal to n lambda by 2 take a Ti-Sapphire laser, I, since I have used 900 nanometer here, I will take that number 900 nanometer is a wavelength that is sustained in Ti-sapphire laser perhaps that is n equal to 1. So n equal to 2, what will be lambda? Lambda be,it will be half of that, ‘n’ equal to three will become one third, and so on and so forth.

But that is actually not the case we have, as we will see. So we want to dispel that notion. That is why we have to once again get a sense of numbers in this business. So my question is, for lambda equal to 900 nanometer and l equal to 3 meter. This is rough number that I am using here, what is the value of n can you calculate? 6.6 into 10 to the power of 6, I have written 6.67 and 6.6666 6.7 10 to the power 6, that is the first thing to understand.

We are not dealing with ‘n’ equal to one, we are not dealing with ‘n’ equal to two we are not dealing with ‘n’ equal to three, we are dealing with n equal to a very, very, very large number. If we are dealing with ‘n’ equal to 123 we would actually have single mode lasers. Because n equal to 1 and n equal to 2 would differ largely in the wavelength is not it? To be very easy to cut out one and use another one using optical coupler use an output coupler that is a dichroic mirror everything other than n equal to 1 or n equal to 2 and n equal to 3 would be cut off.

Here however, it is important to understand first of all, the ‘n’ value that we use is actually very large. So if n is equal to this 6.7 into 10 to the power of 6, what is the value of n plus 1, 7.7 into 10 to the power of 6, 6.7 into 10 to the power of 7. You do not even want to answer that question because it is best to say 6.7 into 10 to the power of 6 in bracket plus one. Otherwise, you actually have to write down all those zeros 670000 and then finally one zero becomes one. point we are trying to make here is ‘n’ an ‘n plus 1’, delta n by n is a very small number.

Delta n is of course one, when I am talking about n and n plus one but delta by n by n is a very small number how does that matter this is all that matters n into lambda n equal to 2L. n plus 1 into lambda n plus one is also equal to 2L. And what we just said is that this n and n plus 1 are practically the same if n is equal to one the n plus 1 only twice in what n equal to 6.7 into 10 to the power of 6 that plus one is practically equal to 6.7 into 10 to the power of 6.

If your salary is one crore rupees you do not really mind whether 10 rupees are added or subtracted right something like that so, now, what will be delta lambda let us do a rough calculation first, lambda into n by lambda n minus lambda n plus 1 what will it be it will be a very small number right, is going to be basically to 2L divided by n which will be longer lambda n plus 1 is higher energy. So, the difference is going to be not that much. So and that tells us why it is so difficult to make multimode lasers.

So, you have we 900 nanometer and 900 and 900.5 nanometers right these are 2 successive modes, how will you separate them? That is why multimode lasers. Usually lasers we have a not multimode lasers and multimode lasers are available but they are costly. For our purpose this is a blessing in disguise. Because if you want to do ultra-fast spectroscopy, you do not want a multimode laser as we will see. For now, it is important to understand that the difference between the wavelengths of successive modes is really very small.

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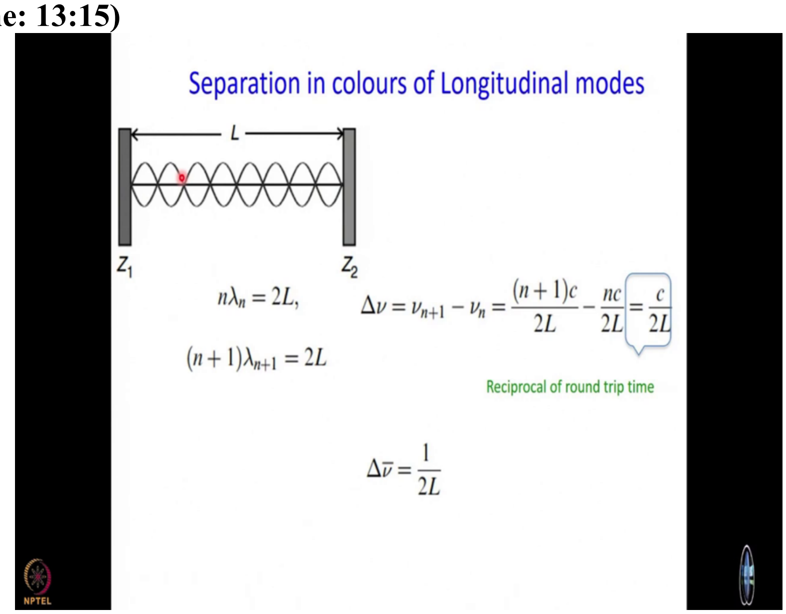

That is best understood. If we take this problem of separation colors of longitudinal modes, if we call them colors in terms of not lambda, but nu, because energy is proportional to lambda right, energy is proportional to nu, reciprocal with lambda. So, nobody works to delta lambda generally delta nu is better. Can you tell me, what is delta nu? When Delta nu is nu of n plus one th mode minus nu of Nth mode, can you work it out? delta lambda equal to nu n plus 1 minus nu n what will it be ?to convert this expression of lambda to nu of course, lambda nu equal to c.

So lambda is c by nu? Very small is already established that I am not asking for that I am asking for an expression. I have 2 answers. One is C by 2L. Another one is more complicated. What is the right answer? See c by 2L well, delta nu, which is the difference in frequencies of 2 successive modes, it adds up to c by 2L. Well very interesting. Why is it interesting? It is interesting because a constant you are working with some laser L is defined and C is defined any way unless you change the medium or something. So C by 2L is a constant.

That is the first amusing information that comes out, delta nu is constant and the only thing that determines what delta how big or small delta nu will be is L all right. So if you want delta nu to be double the value that it is it right now, what do you do to the cavity? To increase to decrease? Do you make it double do make it half I want delta nu to become double, then l has to become half and I want delta nu to become half.

That means I want more energetically closer energetically more closer modes. Then, do I need a shorter cavity length? Or do I need a larger cavity length? I need a longer cavity length. This is an important issue. And this is an important issue for both depending on your requirement, do you want more modes do you want less modes? There is a fast control. As you see, we want more modes if you want an ultra-fast pulse.

So for that, you have to increase the cavity length. Where was the first femtosecond pulse produced in India? Bangalore is an answer Bangalore No, in this case no. I give you a hint. I am asking the question. But of course it is not our lab. It was produced in IIT Bombay. It was producing Department of Physics Prof. BP Singh’s lab. So if you go to his lab even now it is there you can see it. They made a very strange arrangement he opened up the laser and then he added a vertical invar rod on which he mounted optics that is how increased the cavity length.

So you can actually buy the Ti-sapphire lasers that will never give you pulse. Always CW and you find that there will be small. Ti-sapphire laser is first of all big secondly it has folded cavity. We are going to open it up and we are going to demonstrate that is because you need a certain cavity length in order to get pulsed and then if you want to be a state cavities simple two mirror cavity then you need a huge space your table is going to be from there to here.

So it is better to fold it. By folding is done by using mirrors. So C by 2L is a constant for a constant cavity. And if you want more modes more modes means smaller separation between modes. You want the cavity to be bigger. There is a first point, what is the second point C by 2L there is a very special number what is the C by 2L. Ok I will make it easier. What is 2L by C is a round trip time. You start at this point, let us say hit this mirror, go back, hit this mirror, come back that is called a round trip.

So the total distance traveled by the photon is 2L right and is a photons it travels at the speed of light. So to 2L by C is the time for a photon to do a round trip. Not you or me to do a round trip photon. So C by 2L essentially is the reciprocal of round trip time. Incidental but interesting important. Now I have written in terms of delta nu. nu is something that I am not really comfortable with or you understand wavelength or wave number better if I want to write delta nu bar, what will it be? Yeah. And this is an easy question that the nu bar that should not take so much time. Yes, divided by C, what is it? 1 by 2L? Well, you are hesitant to give the answer because it is so simple but sometimes we get simple answers to difficult questions.

What is the problem with that? So delta nu bar is one by 2L? The only thing that matters is cavity length. Now, let us once again do a quick bit of math, we are working with 900 nanometer, let's stick to 900 nanometer. Can you tell me what is nu bar for 900 nanometer? What is the wave number corresponding to 900 nanometer. 1 by 900 into 10 to the power 9 of course. If I want to do mental arithmetic, it is easier to make it 1000.

So 1000 into 10 to the power of 3, 10 to the power 3 into 10 to the power minus 9 is 10 to the power minus 6 was that the right? Yes, reciprocal of that is 10 to the power of 6. So that is your nu bar for some 'n'th longitudinal mode. What is delta nu bar? So nu bar what did you get 10 to the power of 6, what is the unit? Meter inverse right? See, once again I was going wrong by a factor of 10 to the power two this is my problem. So 10 to the power 6 per meter, meter inverse.

And what is a typical cavity length? Meter, 1 meter, 2 meter, 3 meter does not matter. We also taken 3 meter earlier, is not it? Lets say 3 meter so, what is delta nu bar? 1 by six per meter is that right if l equal to 3, 1 by 2L is 1 by 6, what is 1 by 6. So, now, you see this is what we are trying to say nu bar for that 900 nanometer light is 10 to the power 6 per meter and nu bar for the next line is 10 to the power 6 plus or if you take on the other side minus 1 by 2L very close ok. Successive modes are actually very close in energy, because we are dealing with high end values we are not dealing with an equal to 123, here we go ahead.

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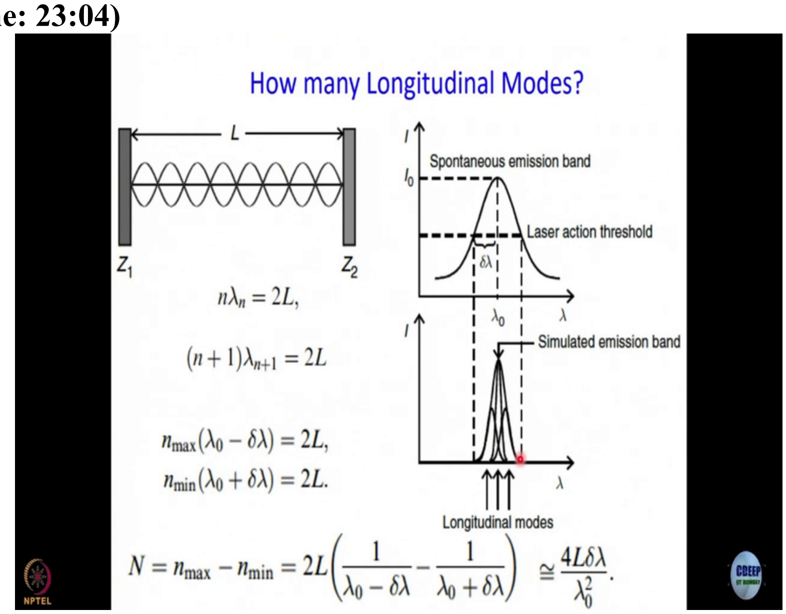

Now, we want to know how many longitudinal modes are there why are we doing all this? Because we are going to see eventually that we want to take these modes and bunch them together to get the pulses, so you want to know before going there, how many longitudinal modes are there? To do that, let us have a lecture all this is from that book whose author's name I cannot pronounce at shown the book in the first module I hope.

Introduction to laser spectroscopy Halina and I cannot pronounce a second name. Here, let us consider this this is the laser cavity we have discussed many times here let us say this is the spontaneous emission band intensity versus lambda let us say spectral maximum is that lambda 0 and let us say delta lambda is half width that half maximum see what will happen is stimulated emission band is always narrower than spontaneous emission band. when, energy density matters limited energy density of this light is going to be very small and energy density of this is maximum.

So, this is sort of like the, the story of the bull that we are discussing little early Nano bull, they are also there was sharpening in space because you required a 2 photon cross section here we get a spectral sharpening because of variation of energy density. So let us say for this may not be correct for all systems. Let us consider that this this is where onset of stimulated emission takes place half width at half maximum of the spontaneous emission band defines the half width of spectral base of this stimulated emission band.

It is not necessary that it is half it can be one third it can be one for something, but for now it has worked with half let us say that this spectral which is delta lambda. So, what is the base of the spectrum two into delta lambda? So, once again we are back to the question that we started with, but in a different domain that was in time domain this is in wavelength domain. So, now n multiplied by lambda n equal to 2L into n plus 1 multiplied by lambda and plus one equal to 2L that we know let us say you are in nmax is the mode number.

So giving roll numbers to modes, mode number 10 mode number 11 mode number 12 is just that we know it is not 10,11,12 it is ten to the power 6 plus minus 1234. So, let us say N max is the mode number in this position where will n max be here or here it will be on the lower wavelength side right where lower wavelength is higher energy let us say n min is the mode number for this end of the spectrum. So, we can rewrite these equations then n max multiplied by lambda zero minus delta lambda should be equal to 2L. N max is the mode number here.

What is lambda? Lambda is this lambda zero minus delta lambda. So, n max multiplied by lambda zero minus delta lambda in bracket is equal to 2L. Similarly, in min multiplied by lambda zero plus delta lambda in bracket is equal to 2L once again have you understood? What is the total number of modes n max minus n min and when you might want to do in max minus n min minus one but then n min minus one n min are not very different from each other.

n max minus n min what is that? n max will be equal to 2L divided by lambda zero minus delta lambda. n min will be 2L divided by lambda zero plus delta lambda. So, basically you are subtracting one from the other, you will get a denominator of lambda zero square minus delta lambda square, and then you will have lambda zero plus delta in the numerator you have lambda zero plus delta lambda minus lambda zero minus minus delta lambda.

So, the numerator lambda zero will cancel you will be left is this what you got? And what is the denominators lambda zero square minus delta lambda square, but then what we have established so far is that the lambda is really a small value. So you might as well neglected square for a small value square will be even smaller. So what will the denominator be? Lambda zero square right? This is my answer. four L delta lambda by lambda zero squared, is this an absolute relation or does it change from case to case see this relationship is obtained with certain considerations. First consideration is the onset takes place at lambda zero minus delta lambda, and lambda zero plus delta lambda.

Where delta lambda half width half maximum that may or may not be corrected can vary, it depends on the shape of the spectrum and so on and so forth. Roughly, you get something like this, you will definitely get l delta lambda in the numerator, you will definitely add lambda zero squared in the denominator that four instead of 4 it can be 4.12 or 3.89. It can vary a little bit depending on what kind of spectrum you have, and what kind of system you have. But roughly this constant multiplied by l delta lambda by lambda zero squared will hold.

Now, we are in a position to look at this and think what n going to be will actually work out given some typical values, but if l increases n is going to increase okay that may or may not have occurred to us if we did not see it would not have occurred to me for sure. If delta lambda increases n will actually increase I would not have guessed it to be honest and lambda zero square in the numerator the denominator that also would n to increase.

If lambda zero decreases n will increase, now let us work it out. For our Ti-Sapphire laser maximum is at 800 nanometers so lambda zero is 800 nanometer l what is the value of l let us take four meter? Will it help? It will help to some extent. So, let us say l equal to 4 meter what should what is a good value of delta lambda where does lasing start for Ti-sapphire laser say 700 nanometer let us say 100 on each side 200 no hundred.

So, let us the delta lambda is hundred nanometers can we work out what the number of modes is under the spectrum four multiplied by l is 4 meter multiplied by delta lambda is 100 into 10 to the power minus 9 meters divided by divided by is too much what is this divided by what is the numerator. Just 800 nanometer square. So, 4 into 4 is good that will get will take care of multiplied by 10 to the power 2 square is 10 to the power 4 multiplied by nanometer squared. So 10 to the minus 18 will be there, this fours all take care of each other, you are left with 10 to the power minus seven in the numerator.

And in the denominator it is 10 to the minus 14 is that right what is the number that is coming? How did you get 25, 4 L delta lambda by z lambda zero square data lambda is hundred nanometers. So, what is your answer Vikas. Do we get that answer? How much 2.5 into 10 to the power six large number. So, we can expect therefore, something like titanium Sapphire the emission is very highly stimulated emission is very highly multimodal 10 to the power 6 modes are there. Now, let us see what happens in two cases, one in which these modes are have no phase relationship, one in which they have some phaserelationship.

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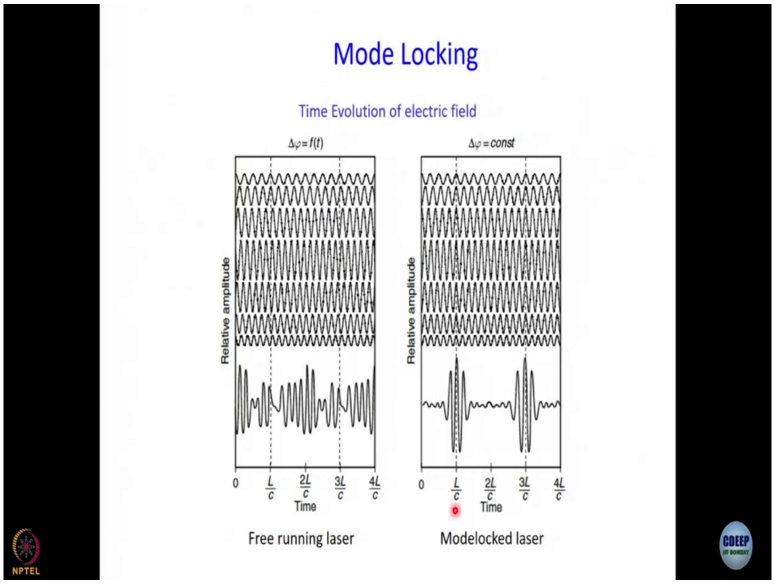

This is what happens here to keep things simple, we are not plotting intensity, and we are plotting the time evolution of electric field. So, I am showing a large number of waves here. Waves whose frequencies are different from each other, just a little. So, this is a free running laser in which these different modes have no relationship, let us say. And here this is called a mode lock laser. And we will see why it is called a mode locked laser, where delta phi is equal to constant which means some phase relationship is maintained, the easier thing to work out here is at some points, let us just say at this point, phase difference is zero.

Now what happens, you see this kind of an oscillation here. And here, you see that at this point, everything is in phase. So if this is in phase, all the electric fields will add up, you will get constructive interference. The moment you move x axis is time, remember, the moment you move out in any direction, the waves start getting out of step, this something that we might have studied when we studied time domain spectroscopy.
So, they start getting out of phase until they have complete control and destructive interference. And then after some time, they start getting back in phase. So, you are going to get something like these are called interferograms, So, you have packets of energy at regular intervals, intervals shown here we are going to derive this next day, but just see what the interval is. 3L by c minus L by C What is that ? 2L by C same 2L by C that we encountered earlier and this is telling us a story? What is the story?

The story is like this. Suppose you are shutter in the cavity, you open it for a very short duration and close it again what will happen in the short duration is that some of these some waves will go through let us say you somehow managed to ensure that at the time of going through, all these waves are in phase. They come back when they come back to the shutter. Again, you open it what you do then you sustain this mode locked operation rather than free running operation.

So basically bundling together waves that are in phase at that point of time of crossing the shutter and then let them evolve by themselves so that they will go to zero very quickly and then come back. That is how you inch you prepare phases. This is only an introduction. In the next couple of modules, we are going to do the math. And then we are going to say how it is actually experimentally done. That will be our discussion of mode locking. If it is not ultra-fast, there is perhaps easier to do. If you want nanosecond pulses, it is done by something called Q-switching. If it is femtosecond pulses. Then things become difficult.

And fortunately as we will see, nature provides us a way out by which femtosecond pulses are produced by themselves. You only have to perturb the system a little bit and the system model locks itself. That is what we want to do after this week, we may or may not discuss how to mode lock immediately right away after this, but rather after we have developed the concept of how to prepare, how to make pulses, we want to talk about how to amplify them. And then we come to the concept of chirped pulse amplification. Then we will take it from there.