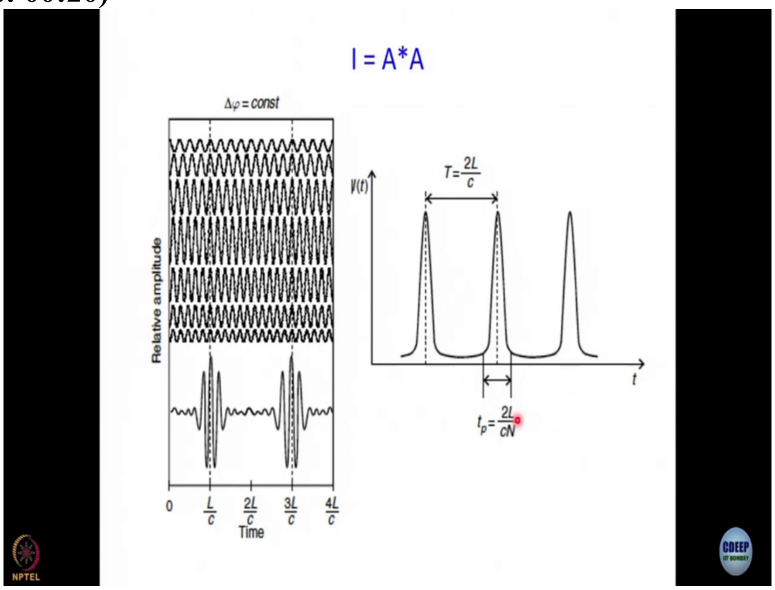
Ultrafast Processes in Chemistry
Prof. Anindya Dutta
Department of Chemistry
Indian Institute of Technology-Bombay

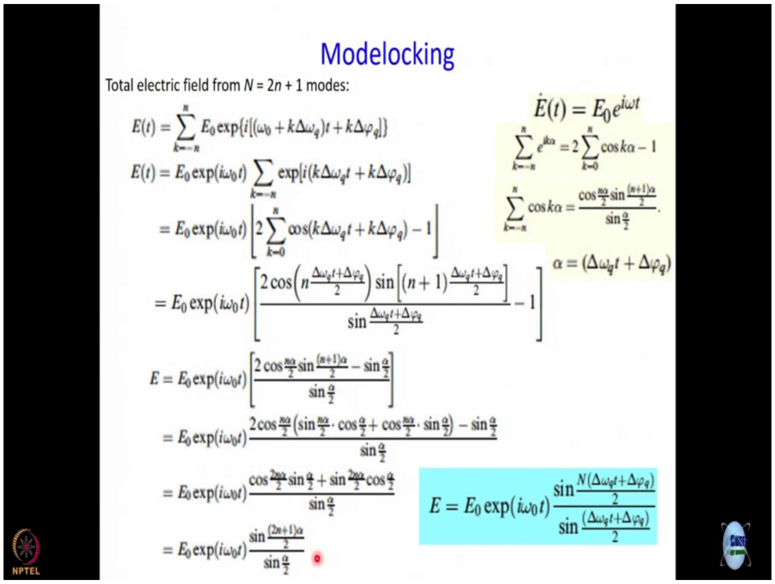
Lecture # 22
Modelocking for Short Pulses (contd.)

(Refer Slide Time: 00:20)



Right, here's where we have stopped. We said that intensity is A star into A and we said that provided the result first, we said that when you lock n number of modes together so, that the phase difference is not a function of time, then you get pulse operation repetition rate is not a function of number of pulses that are locked, pulse width is that is an important take home message. So, repetition rate is 2L / C all that is important there is what is L. L tells you what the repetition rate will be right, but pulse width that is where number of modes coming. So now, let us see how.

(Refer Slide Time: 01:09)



So we start our somewhat sketchy discussion of mode looking, I apologize for the quality of slides. Several uploaded will hopefully be better. I tried to do things quickly and there is another moral lesson that you do when you try to do things quickly. They do not work out too very good. So, as I said in the previous module, we do not really have plane waves we have damped oscillation, but then eventually, the way we go from time domain to frequency domain is Fourier transformation. And the beauty of Fourier transformation is that to get the correct information about frequencies, it does not matter.

If you take a plane wave or a damped wave because they get separated. So that makes the mathematics a little easier. Otherwise we would have another damping factor here. So we are going to work with plane waves where we will say that this electric field the function of time, and is given by E0 the amplitude maximum amplitude into e to the power i omega t. What is omega? Yes angular frequency omega is angular frequency, what is the relationship between omega and nu, frequency and Angular frequency 2 pi.

Now, just all you have to know is whether to multiply by 2 pi or divide by 2 pi. Not very difficult. I think we know it. So now what I am saying is, I have locked these modes together. And I am saying the number of modes that I am working with capital N is equal to 2n + 1. So working with an odd number of modes, because the math becomes a little easier as you will see in the next step.

If I have an odd number of words, it does not matter even if it is an even number of modes, you can conveniently neglect 1. Remember how many modes are we going to have 5000 6000 10,000 20,000 something like that. So, it does not matter whether it is 20,532 or 20,531. So, we formulate using 2n + 1 number of modes. So, here, we are saying that from 1 mode to the next, there is a difference in frequencies. And we have worked out the expression earlier. What is delta nu? delta nu remember delta nu what is delta nu it is difference between the frequency of nth and the n + 1 th longitudinal mode.

What is it? What is the expression? C/2L be handy remember do not forget that that is very important. C/2L which is inverse of round trip time right C by 2L. So what is going to be delta omega? What is delta omega going to be? You know, delta nu, delta nu is C/2L, what will delta omega be that factor of 2 pi will come. We are going to use this so, do not get confused. So, this is what we are writing E0 into e to the power i omega 0 + k delta omega q, ok? What is k, k is a number that varies from - n to + n.

So, now, you already see why you want to 2n + 1 number of modes, it becomes nice symmetric, symmetrical distributed about a mean. About a central position not mid central position + we are saying k into delta phi Q What is the delta phi q phase difference and that is independent of time let us remember that that is very important. So, how do I expand it, I can well e to the power a + b is equal to e to the power A multiplied by e to the power b.

So, I can use that and I can take this E0 e to the power i omega 0 t outside the summation. And then inside I am left with e to the power i k delta omega q t + k delta phi Q, if you do the summation, and now, the advantage of summation here it is not showing what advantage of summing k from - n to + n is that I know what the answer is. So, these summations i mean I am sure we might have done many of these in 11,12th maybe in BSc or something?

These summations are all nicely worked out. And it is known that some over k equal to - n to n, e to the power ik alpha is equal to 2 multiplied by sum over k equal to 0 to n cosk alpha - 1. And then this you might think one summation is leading to another summation. So, what is the big deal?

The big deal is that this summation is also known as you are going to see, but let us first write it, this is what it will be.

You have this outside the bracket inside the summation of exponential terms becomes summation of cosine and also some over this domain changes, right? It was - n to + n. Now, it is going to be equal to 0 to n. So, if you took only capital N and then we did have this trouble you cannot even write n by 2. So that is why we want 2n + 1 modes. , so now all we need to know to go further is what is the summation of cosine terms.

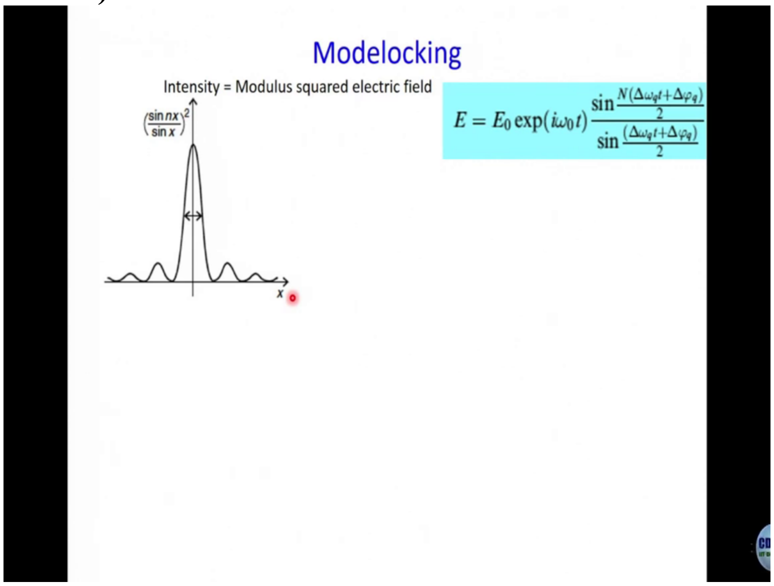
This is a summation of cosine terms. Does it look very nice, but that is what it is what can we do? Alright, so I am going to rush through the next steps. You can do the math if you want. It is not all that difficult. So, this is what we substitute, substitute for the summation. And then we put alpha equal to delta omega q t + delta phi q. So, in terms of alpha, this is the expression that we get, you get something like E0 e to the power i omega T.

And you understand that the moment finally we do not want to work with field, , we want to work with intensity. The moment we want to work with intensity, this e to the power i omega t is just not going to be there. Because we multiplied by its complex conjugate that term is not going to appear. So eventually, we do not do not have to worry about it. We do have to worry about this, Sin(2n + 1) alpha by 2 divided by sine alpha by 2 this term.

So, let us write it in a little easier form what is 2 n + 1, 2 into small n + 1 what is that 2 n + 1 what is that number? The answer is there in the slide. Total number of longitudinal modes right. So, the other way we write it is capital N So, now we have the job of the small 2n is small n is done. Let us write in terms of capital N and this is what we get and here on the quality of slide also starts getting a little better.

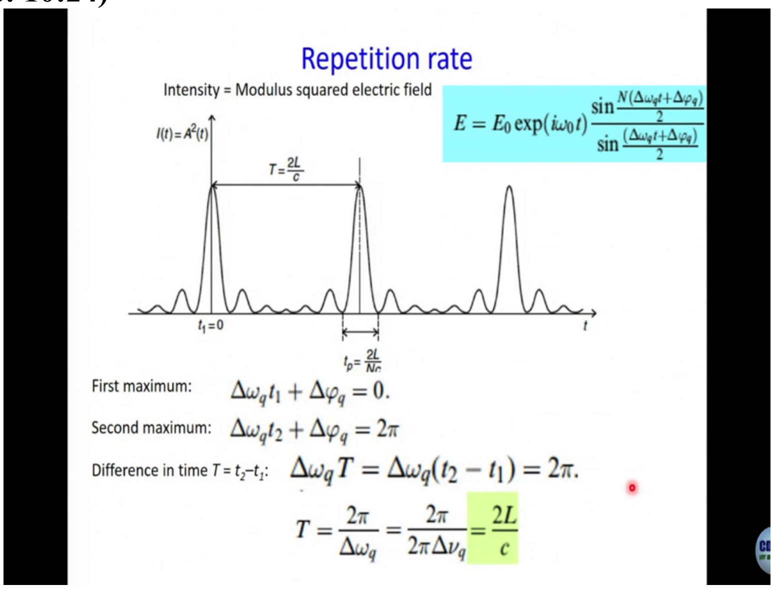
so we get E equal to E0 e to the power i omega 0 t multiplied by sine capital n multiplied by sum of the sum of delta omega and delta phi divided by 2. Yes, as I said eventually it will not matter because you want to do a fourier transformation right, you can add on the amplitudes at that time that is true, but then there is a way out there. So, this is what we end up with.

(Refer Slide Time: 09:44)



The form of function is something like if you are talking about intensity sine nx by sine x whole square what is x? x is this delta omega t + delta phi divided by 2, and a plot of sine nx by x sine x, I encourage you to do it using whatever graph plotting software you want to use. You see, this is what you get. And this is a kind of function that we encountered everywhere. Whenever we use optics, 1 thing we should not forget is that this is a periodic function. I have done only 1, but it is not going to be 1 it is going to repeat.

(Refer Slide Time: 10:24)



So you are going to get this and this is what I was saying. That picture we drew earlier was not completely correct. In the sense that you do not just get a pulse, you get the small you can call after pulse if you want. But the issue is, even this picture is drawn using a comparatively smaller number of smaller values of capital N and when we really work with ultra-short pulses, this duration is so small that in the same scale, it is very difficult to see capital T and tp. And you get to see tp only when you zoom in into 1 pulse. So this is what it is. So do not forget what is the x axis what is the y axis here? This is what you get.

Now I have given you the answer already. Now, let us see if we can derive it. So first let us worry about repetition rate. What is the meaning of repetition rate? We can say it is time between 2 maximum Where will the first maximum occur sine squared in x divided by sine square x where we will let her maximum value of course for that we need to work out limits and so it is difficult because it is something it might sound like there is a discontinuity when x = 0, but it is removable discontinuity ? This point here, if you think that point is going too actually 0, is not it? 0 by 0 kind of situation not 0 sorry, 0 is what you will get.

But the discontinued is something that you can remove. So, first maximum will occur where delta omega q t 1 + delta phi Q equal to 0. Where will the next 1occur second maximum well give you the answer when it is 2 pi does it make sense? right, plug this this into this expression, and you will get the answer at 0 and at 2 pi. So, now, if I ask you, what is capital T, capital T you can get by, capital T is basically t2-t1.

And since it is a periodic function, this separation will be same for any pair of consecutive pulse you take so, what is the t2-t1, then subtract delta omega q is constant. So, what do you get if you subtract 1 from the others you get delta omega q multiplied by t2-t1 and now see delta phi q would cancel each other the moment you take a difference that is exactly what will not happen if delta phi is also a function of time.

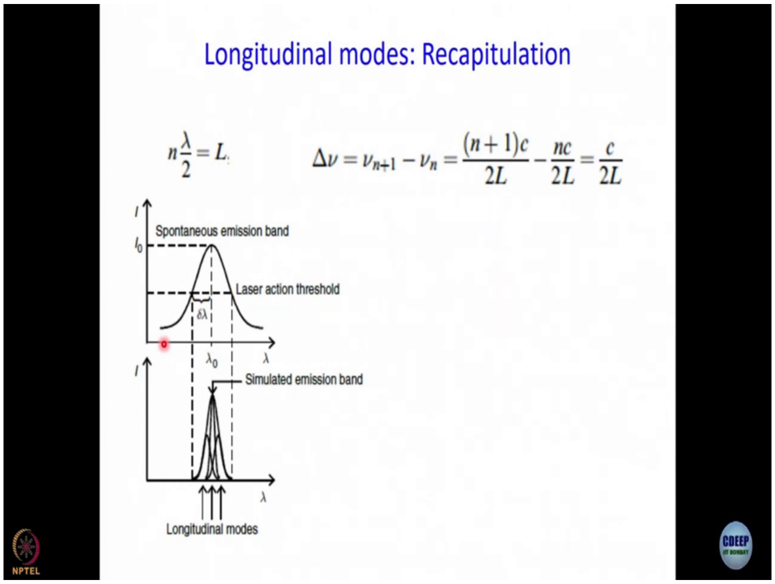
You can do this only because delta phi is not a function of time and it is constant everywhere. So, what do you get delta omega q multiplied by t2 - t1 is equal to 2 pi and what is the t2-t1 that is capital T time between 2 pulses. This one occurs at time t2 this one occurs at time t 1. X axis is time multiplied by something from there you can always convert to time. So, t2-t1 is equal to capital T.

So, I can do it like this difference in time capital T = t2 - t1. So, you get delta omega q multiplied by capital T = 2 pi. So, what is T? 2 pi divided by delta omega q. Now, again we come back to what we promised is going to be useful later on the later on time has come, what is delta omega q? Delta is difference, omega is angular frequency. So, it is difference in angular frequency between what and what 2 consecutive longitudinal modes very good. And we already know some quantity associated with it. We know delta nu and what is the relationship between delta nu and delta omega we know that already.

So, what we can say is we can write something like this 2 pi divided by 2 pi into delta nu q. because delta omega = 2 pi into delta nu when you go from frequency to angular frequency, that factor of 2 pi comes, so you are left with 1 by delta nu q. What is delta nu q? Now from there, you get 2 L by C. So, we see that the repetition rate that we get has nothing to do with how many modes are locked. That is the nice thing that comes up because otherwise it would have been a problem.

How do you decide how many modes are available for you? Can you decide one thing that is there is what is the spectral bandwidth. Say, to start with, let us say you are spontaneous emission spectrum that is independent of anything else spontaneous emission. Then remember, we have drawn laser threshold where half width was delta lambda. ? So where that laser threshold will be who determines that? If I can somehow decrease the laser threshold, then I will have more modes that will get locked. Are you following what I am saying? this figure.

(Refer Slide Time: 16:34)

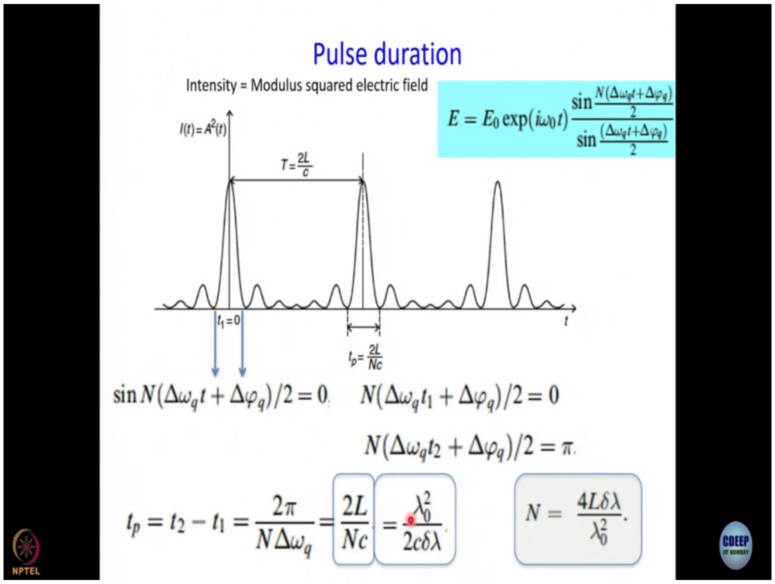


This here is your laser action threshold, now what I am saying is that my first thing that is important is with substance what is your active medium that would better have a broad enough spontaneous emission band. Then what I am trying to say is, here I have placed a laser action threshold here. Is it in any way possible to push it further down? Remember laser action means you should have stimulated emission. Is there any way for a given substance that I can increase the stimulated emission bandwidth? How can I do it?

Exactly pump harder, then this threshold can actually go down. There is more molecules will be excited. So, do not forget the stimulate emission is like a biomolecular reaction by molecule elementary the action between light and matters. So, you could think of doing that. So that way you can get more longitudinal modes and you can try to get shorter pulse that is why Ti-sapphire laser, you want to pump with a high energy you want to pump if possible by 5 watt instead of 3 watt you want to pump if possible by 8 watt instead of 5 watt to get shorter and shorter pulse.

Of course there are other issues, damage is an issue so you have to take care of those. But the problem is if the repetition rate also depended on number of modes, they would have been an issue, is not it? Then you, you do not want the repetition rate to change. otherwise syncronous electronics everything will change. So, it is good. That reputation rate is just 2L by c. Nothing else matters. How hard you pump nothing. Not so for pulse duration.

(Refer Slide Time: 18:56)



So pulse duration, it is not very difficult to understand that, what happens, let us start thinking a little qualitatively, let us go back to this analogy of people running around the field. So, when they come together that is a pulse maximum? If you want a pulse of short duration then they have to scatter fast. Now, if we have more and more people running with different speeds, will you agree with me that scattering is more efficient there? In fact, if you look at this interferograms, where you mix many waves, you see that more the number of waves, sharper is the fall off that is why you need more pulses to get a shorter pulse duration there is a qualitative understanding. Now, let us see what we get from here. How do I define pulse duration as we are see, practically we always talk about full width half maximum. And when we finish this discussion today, we will be talking about full width half maximum.

But for now, it is easier for us if we consider all the normal modes that are there. Otherwise again pulse shape and all that will come if you want to consider all the longitudinal modes that are there, you want to go from here to here. So, let us see what is, the time about this big maximum where your intensity becomes 0 before and after difference between these 2 times is going to give me pulse duration. Remember this pulse duration is not full width half maximum. So this is 200 picosecond, full width at half maximum is perhaps 100, perhaps less depending on the shape of the pulse.

And this is why shape of the pulse also becomes important? If you want to go full width, do you agree with me that ratio of full width at half maximum and full width at lets say 10th maximum is not going to be the same for a gaussian pulse and samples? They will be different. So right now, we do not worry about all let me just talk about the pulse duration. So, when will they become 0 when the numerator is equal to 0 and the denominator is not, so that condition is sine N delta phi t + delta omega t + delta phi by 2 = 0.

This is 1 condition sine x = 0 when x = 0 Next time, when does it become 0, yeah pi. So I can put time as the t1, I guess this is the t1 this thing is equal to 0, for t2, the next one, it is equal to pi. Again from here you can find t2-t1 very easily, that is going to be tp the pulse duration. So, do it, this is what you get tp is equal to t2-t1 the subtract what will you get? You will get two pi divided by n delta n delta omega is that right, this simple algebra.

Again, delta omega what does by now we are sensitized to this issue? Whenever we see delta omega, what should we do? And here this is 2 pi divided by delta wave is even more tempting go over to delta nu and you know the expression for delta nu, it turns out to be 2L by Nc. So, Just this n is different. So, we have indicated you need more number of pulses. So that this more number of or not pulses sorry more number of longitudinal modes because it this n comes to the denominator.

Larger denominator smaller tp that is what you want. ? But then we might as well as stopped here, if you wanted to have only theoretical finally, we want to go over to something that is more tangible, who is going to count the number of longitudinal modes it may not be all that easy, but there may be something else that is easier. So, let us use another expression. And you might have forgotten.

Because we have talked about it couple of modules earlier remember this in fact, we talked about it at the beginning of the previous module. As a recap, we worked out that number of total number of longitudinal modes capital N is given by 4 l delta lambda divided by lambda 0 squared. So might as well substitute this. So, in substitute what do you get? You get pulse duration = lambda 0 square divided by 2 c delta lambda.

What is delta lambda in the treatment that we have followed delta lambda is half width at half maximum of the spontaneous emission band, but what is more important for us is that it is the half width at the base of the stimulated emission band half width at base. Ya. So now we have something tangible? We can measure spectrum, when we measure spectrum, we know what lambda 0 is, we know what delta lambda is.

So it is not very difficult to figure out what the pulse width is going to be. That is why in our lab, we do not bother about trying to measure the pulse width in time domain said shortly we are going to discuss how to do it. You see what the problem is. We are talking about short pulses, 100 femtosecond, 50 femtosecond 6 femtosecond how will you measure you cannot see it on an oscilloscope directly. So you have to apply some kind of a trick.

And the trick that is applied invariably depends on some instrument that was made long ago, let me digress a little bit. What is the speed of light? Is that large is a small how do you know it is correct? Who told you that that is the speed of light? And it takes as I told you How does it know somebody must have measured it. So if I want to measure what is the speed of soumyodipto that is easy?

I can do it using perhaps my wristwatch who what is a wristwatch or what is a stopwatch that allowed somebody to measure the speed of light? Again everybody has studied in the 9 10 11 12 sometime in physics, Michelson interferometer, the moment I say it you will know it is just that you might know it as Michelson interferometer. So immediately interferometer if you remember, monochromatic light was used and there was a relationship from which you could find out the from path length.

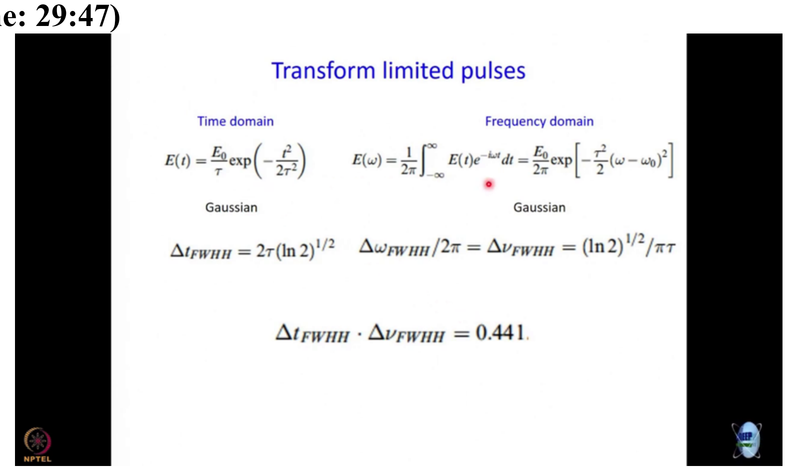
So basically what just Michelson interferometer does is it does generate some map, like we generate a map of the fluorescence decay that takes place in femtosecond by monitoring second harmonic intensity as a function of delay time. So Michelson interferometer also generates a map, and it sort of stretches out. The stage out it behaves as if it stretches out and creates a wave whose wave is correlated with the wavelength that falls on it.

There is a relationship it is not very difficult to work out. So that is how speed of light was determined. So now once you know the speed of light, you can determine small times using Michelson interferometer kind of arrangement. We will talk about that but it is a little cumbersome and I do not know how many chemistry ultrafast labs in India at all have what is called an auto correlators. It is called an auto correlators the instrument by which you measure the pulse width is called an auto correlator and we are happily working in this field without ever having had 1.

Why? Because this is our saving grace. tp is four l delta lambda divided by lambda 0 squared. So if delta lambda because if delta lambda is of some value in principle we can calculate what the pulse width is? If you do not want to calculate what the pulse width is, we can get some qualitative value. We know from our experience that for 100 femtosecond pulse laser, you are full width at half maximum has to be 12 nanometers. For the amplifier, which is shorter pulse laser full width at half maximum has to be more.

This is a relationship that 1 can use and work happily in an ultra-fast lab without ever having to buy an auto correlator. But then it may not be as simple as I have proposed it to be because do not forget what this delta lambda is. It is the basal width right. 1 thing that we have not considered so far at all is the spectral shape that is actually important. So, let us close todays discussion by talking about it. So, what we are essentially talking about is transform limited pulses and I have goofed up a little bit on this slide. Unfortunately, bear with me.

(Refer Slide Time: 29:47)



See, what are we saying? We are saying that a broader spread in wavelength and therefore energy corresponds to a shorter spread in time, that is essentially something that is absolutely correlated with uncertainty principle once again greater uncertainty in time is associated with lesser uncertainty in energy and vice versa. So these are called transform limited pulses. Of course, this is the best case scenario, you might have a bandwidth, full width half maxima of 12 nanometer but not hundred femtosecond pulse if something else is wrong.

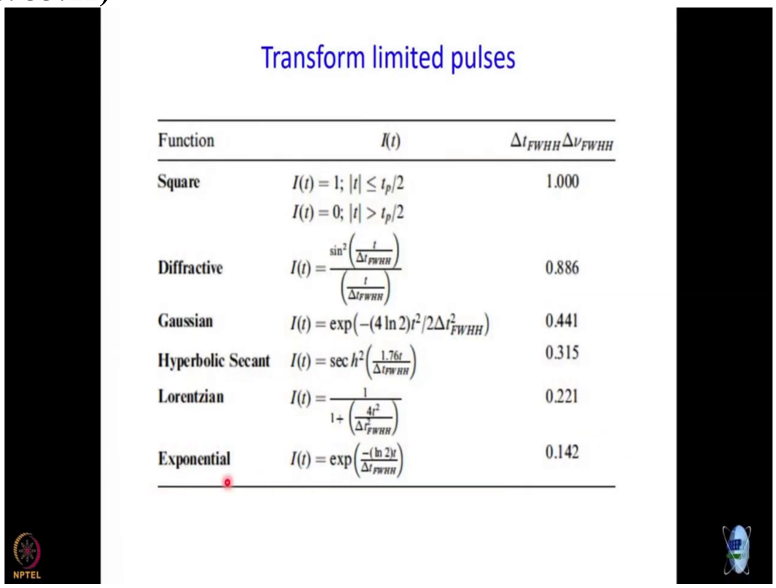
Now we are talking about the best case scenario. So, let us see why they are called transform limited pulses. Let us start talking about a gaussian pulse in time domain. This is a gaussian function. If I want to know its spectrum, if I want to know what that pulse looks like in frequency domain, what do we do? We do Fourier transformation and when you do fourier transformation, what kind of function do you get? another gaussian and this thing that I had by mistake written earlier is that full width half maximum delta omega full width half maximum that is given by this kind of a function.

What is the spreading time for this gaussian function in time the spread is given by 2 tau square root of ln2. All right, now if I multiply the 2 together full width half maximum in time domain multiplied by full width half maximum in frequency domain what do I get? I get 0.441 all this tau and everything cancel off, ln2 is a number. So, a transform limited pulse which is gaussian is going to be obey this so, if your pulse is gaussian then it is very nice, again no need of an auto correlator.

The only thing to remember is you should talk about delta nu not delta lambda ok, lambda is an inverse scale that is why that lambda 0 square factor comes. So, delta t full width half maximum in time multiplied by full width half maximum in frequency gives .4414 Gaussian for a transform limited pulse. So, what is it? Why do people buy auto correlators in the first place? So, far I have told you that we are happily working without an auto correlator without having to spend those few lakhs of rupees that are required to buy it or build it. Why do people buy at all?

They buy it because not all pulses are transform limited pulses, this is sort of the theoretical limit, and then let us now close by showing you something else. This number .441 that we got, why did we get 0.441 and we are not 0.8, because we worked with gaussian function . So, similarly, this has been worked out for other pulses as well.

(Refer Slide Time: 33:11)



Transform limited pulses

Function	$I(t)$	$\Delta t_{FWHH}\Delta\nu_{FWHH}$
Square	$I(t)=1;\ \lvert t\rvert \le t_p/2$ $I(t)=0;\ \lvert t\rvert > t_p/2$	1.000
Diffractive	$I(t)=\dfrac{\sin^2\left(\frac{t}{\Delta t_{FWHH}}\right)}{\left(\frac{t}{\Delta t_{FWHH}}\right)}$	0.886
Gaussian	$I(t)=\exp\left(-(4\ln 2)t^2/2\Delta t_{FWHH}^2\right)$	0.441
Hyperbolic Secant	$I(t)=\sec h^2\left(\frac{1.76t}{\Delta t_{FWHH}}\right)$	0.315
Lorentzian	$I(t)=\dfrac{1}{1+\left(\frac{4t^2}{\Delta t_{FWHH}^2}\right)}$	0.221
Exponential	$I(t)=\exp\left(\frac{-(\ln 2)t}{\Delta t_{FWHH}}\right)$	0.142

And these are the numbers in forget about this for now, for square pulse, the product has to be 1 for a diffractive pulse .886. Gaussian we have discussed already for hyperbolic second, these are all different shapes, and there is no guarantee that 1 shape that always you are going to work with gaussian kind of pulse. Lorentzian .221, see Gaussian and Lorentzian we are very familiar with see what the differences.

1 is half of the other. exponential, 2 sided exponential, and the product is .142. So depending on what kind of pulse you have, what kind of spectrum you have, in principle you can work out the pulse width if you just look at the spectrum provided you are working with transform limited pulses. So that is what we wanted to discuss today. Next, let us see how to achieve we have talked about mode locking, we have talked about the theory. How do you do it? That is what we are going to discuss in the next few modules.