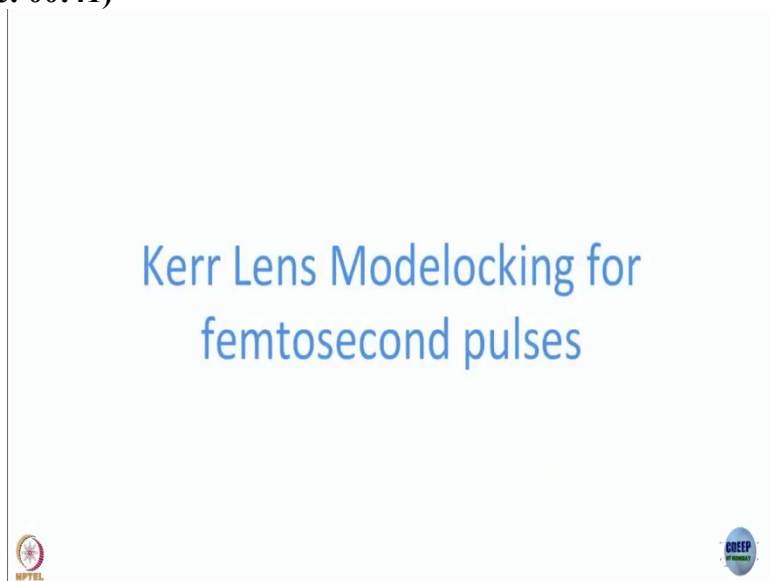


**Ultrafast Processes in Chemistry**  
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**Lecture No. 23**  
**Kerr Lens Modelocking for Femtosecond Pulses**

Today we go a little further with our discussion of how to actually do mode locking. So far we have studied the theory. Today, we are going to talk about at least one case in which mode locking is done and how it is done. If you read any standard textbook, at this stage, what people will discuss is active mode locking, passive mode locking and acousto-optic modulators.

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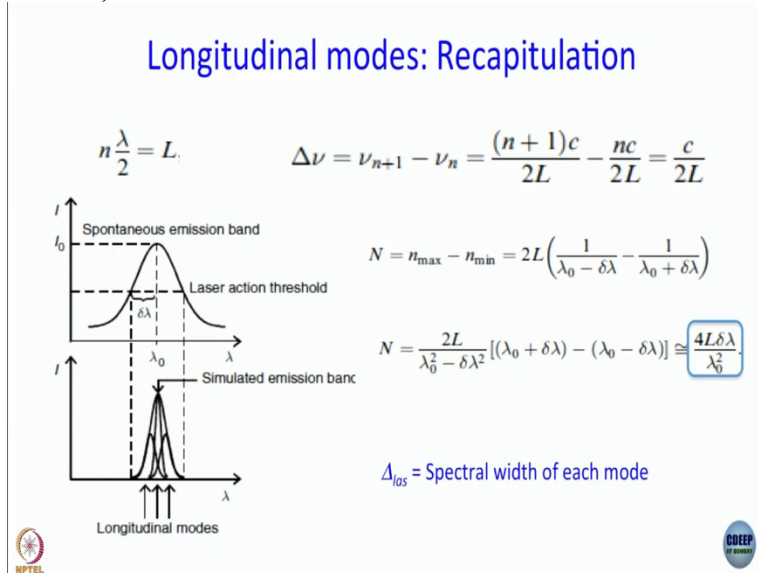


We will take a rain check on that will not do it right now, because our primary goal is to understand how a Ti-sapphire laser works at this point of time. So, we will skip that for a moment. We will come back and do it in the module after next after we have shown you Ti-sapphire laser but today we want to learn about a very strange way in which mode locking gets done and femtosecond pulses are produced in titanium Sapphire laser. And the way it is done is called Kerr lens mode locking. And in fact, the in fact, Kerr lens mode locking happens in a manner in which you do not even perhaps understand that it is happening.

So it is almost like it happens by itself. So the question is, what is this magic? What is the mystery by which this femtosecond pulses that we were looking for so desperately get produced by

themselves without us having to do much? And is there anything that we can do to improve the situation and that is what we will discuss in this module. In the next module. We will go and talk about an actual Ti-sapphire laser.

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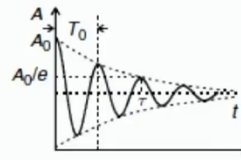
But before that, let us quickly recapitulate what we have discussed. In the last couple of modules, we have talked about longitudinal modes in detail. So now I hope we all understand that a laser can have many modes, many modes mean many frequencies that can be sustained in the cavity. And the kind of cavities we have and the kind of color of light that we deal with. We always deal with things like ten to the power sixth, 10 to the power (6 + 1)th mode and so on and so forth. So these modes are only slightly different from each other as far as frequency or wavelength is concerned. So, and we have also said that this spectral width of each mode is something that we have also calculated and given the modal wavelength, and given the width of the spectrum.

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## Quality factor, $Q$

$$Q = 2\pi \frac{\text{energy gained in system}}{\text{energy lost during 1 cycle}}$$

$$Q = 2\pi \frac{A_0^2}{A_0^2 - A_0^2 e^{-2\beta T_0}} = \frac{2\pi}{1 - e^{-2T_0/\tau}}$$



$T_0$  = Time interval between two successive maxima

$\tau$  = Time for decay of amplitude to  $A_0/e$

$$T_0 \ll \tau$$

$$e^{-2T_0/\tau} = 1 - 2T_0/\tau + \dots$$

$$Q = 2\pi \frac{\tau}{2T_0} = \pi \nu_0 \tau = \frac{\omega_0 \tau}{2}$$

$$\Delta_{\text{las}} = 2\pi\beta = 2\pi \frac{\omega_0}{Q}$$

$\Delta_{\text{las}}$  = Spectral width of each mode

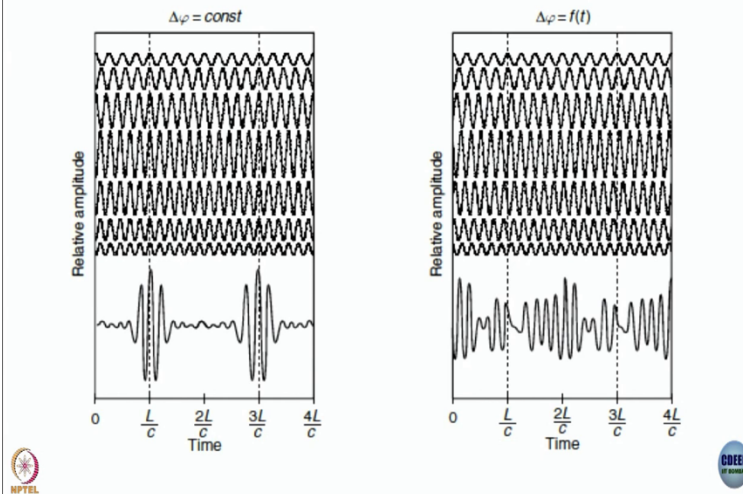


We know what the total number of modes is and then we introduce another term which is very important as far as discussion of laser is concerned not just ultra-fast lasers any kind of laser that is quality factor is essentially a weighted ratio of energy gain in the system and energy lost during one cycle energy gain in the system means, what is a buildup. How was your population inversion in achieve and that is usually achieved by pumping it with an intense laser.

And energy lost we not just by a mission of light, but also due to things like heat and so on and so forth. And then, without derivation, we have given you the relationship of between the spectral width of each mode and the quality factor you very soon will come to a discussion of what is called Q switching where we can generate pulses, not very short pulses, but pulses nevertheless, by switching the quality within the cavity.

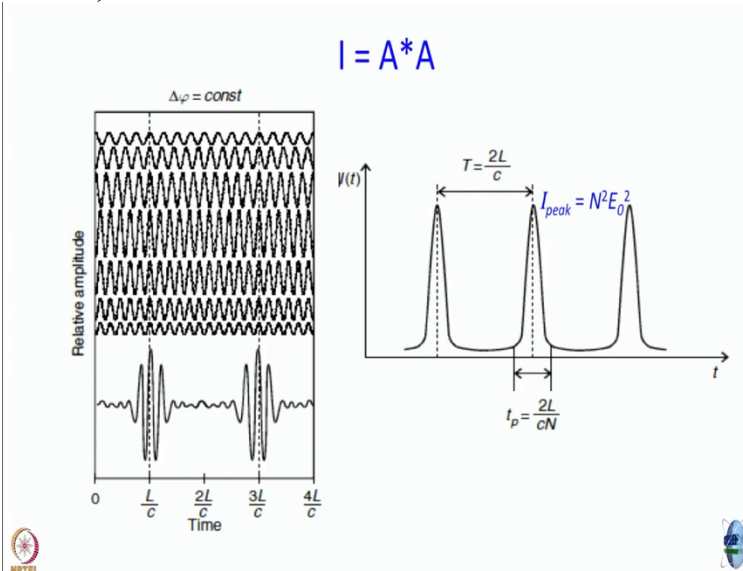
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## Longitudinal modes in a bunch



And then this is a figure that we have shown maybe three four times already what happened bins when you have many modes. First of all, when they have no phase correlation, secondly, when they do have phase correlation and what we have said is that when the phase relationship is a function of time, then you get a random fluctuation and essentially you get a CW output. However, when you have a phase relationship which is independent of time, then you achieve what is called mode locking.

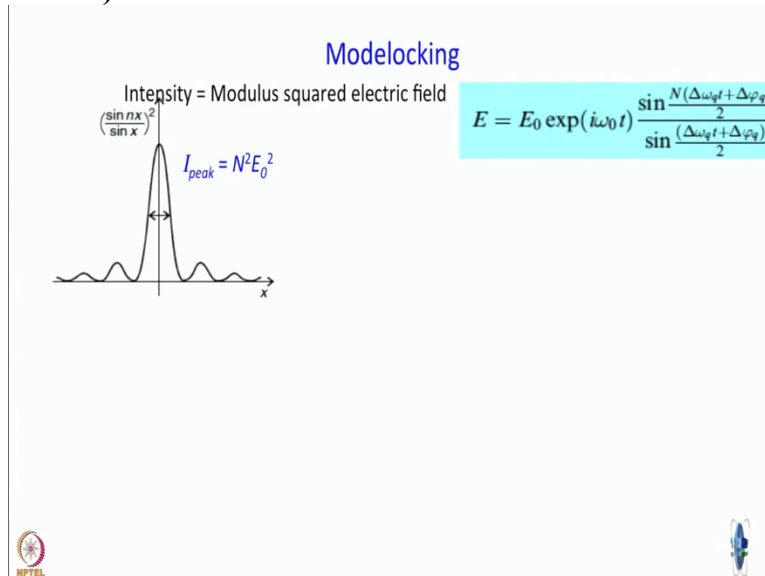
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And that is when you generate pulses and we are talking about pulses we also said that you get the intensity by taking square of the field. So, when you combine a number of pulses is something I do not think I said earlier, when we combine 'n' number of pulses essentially, the intensity at the

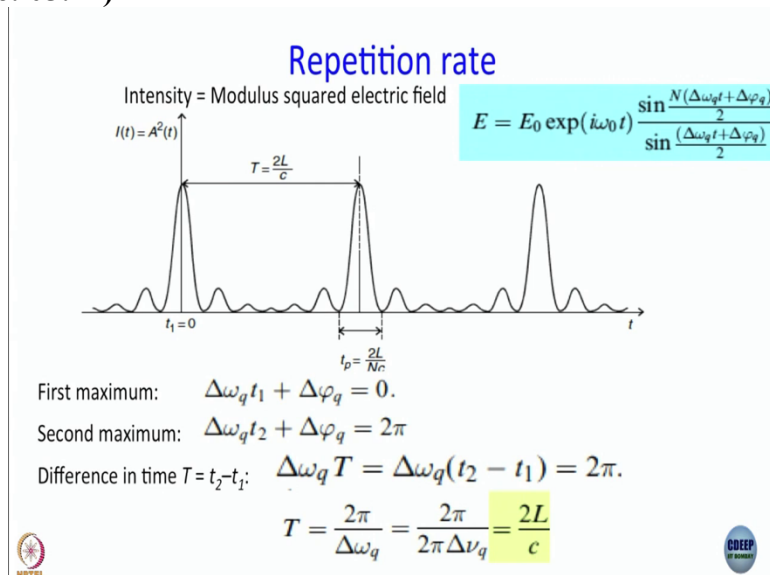
peak turns out to be  $n^2$  multiplied by  $E_0^2$ , but  $E_0^2$  is the maximum amplitude of field associated with the normal modes. So you see you couple 5 modes. Let us say  $E_0$  is same, you couple 5 modes you get 25 multiplied by  $E_0^2$  you couple 500 that you 500 square multiplied by the same  $E_0^2$ . So, that is how peak intensity grows very significantly as you increase the number of nodes, number of modes that are coupled.

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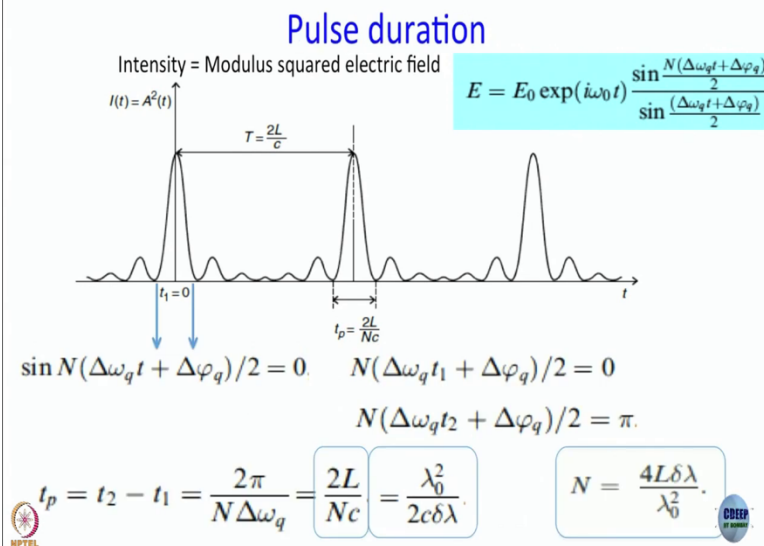
And we said that this is the shape of the intensity of each pulse. If you zoom into it, you have 1 major pulse and you have very short side sidebands and the widths are very small here.

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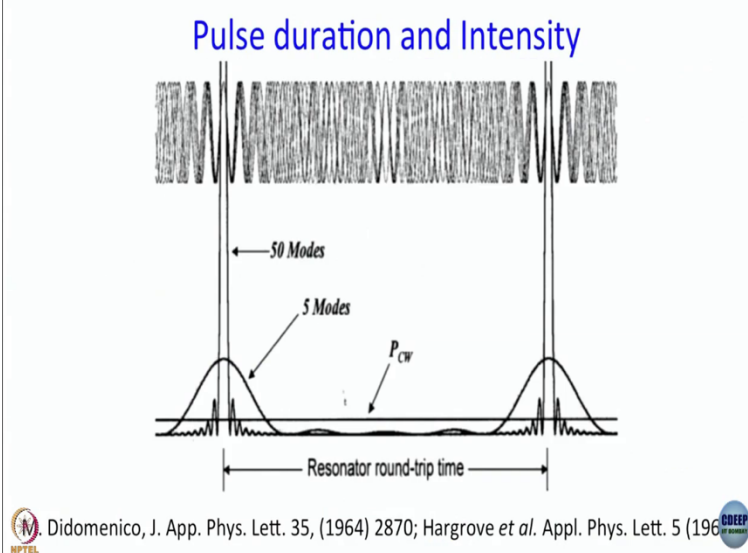
We also talked about repetition rate, repetition rate turns out to be  $2L$  by  $C$ , which is essentially round trip time within the cavity of length  $L$ .

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Pulse width however, turns out to be something that is depending then not only on the length of cavity, but also on the number of modes that are coupled together. And since we know the relationship between number of modes and  $\lambda_0$  the modal wavelength and  $\delta\lambda$ , we worked out this relationship that pulse duration  $t_p$  is not really full width half maximum pulse duration turns out to be  $\lambda_0^2$  squared divided by to see  $\delta\lambda$ .

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And this is something that have not shown earlier this is so, you can see when this was published 1964 in the very beginning of the invention of lasers itself, so, this is what you get, this is what I was talking about, you couple of 5 modes. These are the kind of pulses you get quite broad and less intense. You couple 50 modes, you get sharp and intense pulses. So this is the essence of mode locking and production of pulses.

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### Transform limited pulses

Time domain

$$E(t) = \frac{E_0}{\tau} \exp\left(-\frac{t^2}{2\tau^2}\right)$$

Gaussian

$$\Delta t_{FWHH} = 2\tau(\ln 2)^{1/2}$$



Frequency domain

$$E(\omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} E(t)e^{-i\omega t} dt = \frac{E_0}{2\pi} \exp\left[-\frac{\tau^2}{2}(\omega - \omega_0)^2\right]$$

Gaussian

$$\Delta\omega_{FWHH}/2\pi = \Delta\nu_{FWHH} = (\ln 2)^{1/2}/\pi\tau$$

$$\Delta t_{FWHH} \cdot \Delta\nu_{FWHH} = 0.441$$






Then you said that the best case scenario you can get is when you have transformed limited pulses. So, product of the pulse width, full width half maximum and product of full width half maximum of the spectrum turns out to be a constant, which is 0.441, for gaussian pulses.

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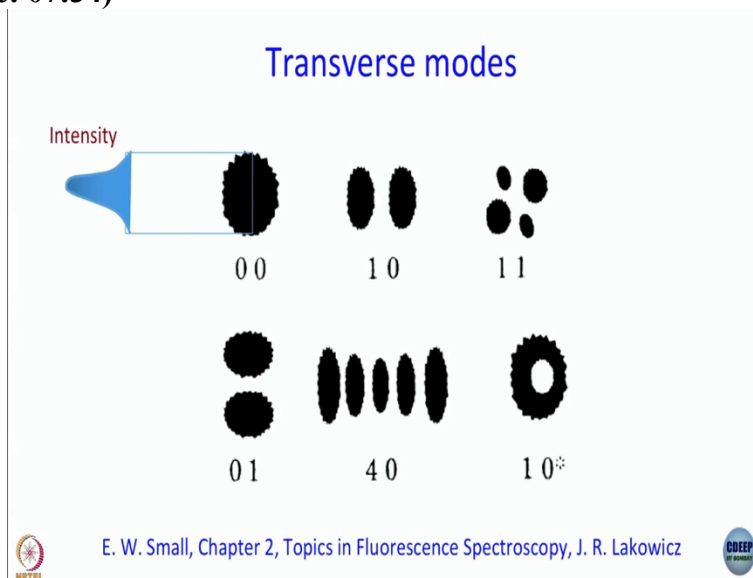
### Transform limited pulses

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

And different numbers for pulses of different shapes. But remember, this is the best case scenario you can get. Just because you have a certain bandwidth did not necessarily mean that you have a pulse width that is as small as you really want it to be. You really have to have good alignment in order to achieve transform limited pulses.

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Before we move on, it is time to talk formally once about transverse modes. What is the Transverse mode? You take a laser beam and put it on a surface maybe expand it and put it on a surface, you are going to see a spot. The question is what does this spot looks like? If the spot looks like a circle roughly a circle without any dark spot anywhere, it is just a patch of light circular patch of light.

Then this mode is called TEM 00 mode TEM means transverse electromagnetic mode T for transverse E for electromagnetic M for mode. 00 means there is no mode in any direction this one is called TEM 1 0 mode that means there is 1 node in a particular direction, this is 1 1 mode, 1 node along this direction, now the problem is this is 1 0 mode. This one also has 1 node, this is called 01 mode which 1 is 10 which one is 01? There is no good answer to that because the directions are all arbitrary.

You might say that this surface is xy. So, this is x axis. So if there is a node along this I will call it 10 if there is a node along this direction, I will call it 01. But then X and Y they are all relative right it depends on how you define the directions. So as E. W. Small says in this chapter of J R lakowicz's, topics in fluorescence spectroscopy, this is not the introduction to fluorescence



spectroscopy book, Lakowicz also has several volumes of topics in fluorescence spectroscopy published long ago there in this chapter 2 E W small said, one man is 1 0 mode is another man is 0 1 mode. So, it depends you look at different books. The crux of the matter is this number 1 or 2 or whatever it is tells you how many nodes there are in a particular direction.

Now, which direction comes first which direction comes second is absolutely a matter of convention. This 1 is 4 0 or you might want to say 0 4 mode. Before coming to this 1, do these modes remind you of something that you have studied in first year something? They remind us of orbitals right the shapes are very much like orbitals. In fact, when I taught this course, actually in a classroom, one of the students sent me these pictures of orbitals and said do not they look very similar.

Ok now, last. So, you can on and on , these modes. What about this? is there a node here? you have what looks like a 00 mode, but then there is a hole punched in it. This is called in colloquial terms a donut mode. 1 0 star now, one thing I have not drawn here is you can also have nodes that are radial. So here I have drawn nodes as planes, but you can also have a spot then a circle of darkness, and then another spot of light only 2 of them 3 of them 4 of them.

So radial nodes are also there again, you call them 1020, so on and so forth. Now this donut mode, 1 0 star is of particular importance in things like super resolution microscopy. So in I do not think we will have scope to discuss super resolution microscopy in this course, it was sometime later I think we know right, what is the smallest spot that I can get by focusing light of wavelength  $\lambda$ ,  $\lambda$  by 2.

Why is it so? it is called the diffraction limited spot so you cannot make it any tighter you can think that will do tight focusing and the spot will become smaller, but then it cannot become 0. They will always be a finite size because below that diffraction will set in and broaden the spot a little bit. So diffraction limited spot so if I use what kind of I want to do say visible microscopy, visible light microscopy, 600 nanometer, 500 nanometers, something like that, let us say I am using a 500 nanometer laser to do microscopy.

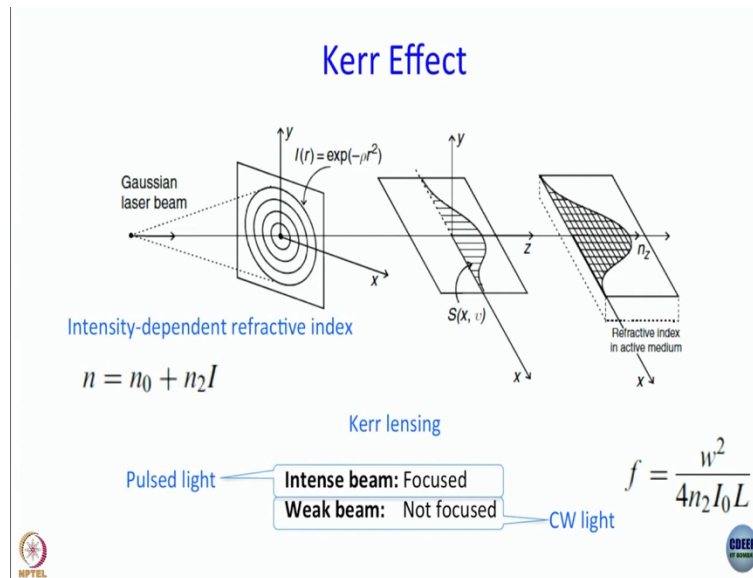
So then the best resolution I can get is to 250 nanometer right. So you cannot do any better by usual microscopic techniques. As you know, I think 2014 Nobel Prize was for Super resolution microscopy, who are the people who got it? Stephen Hill was one Betzig was another one. And Moerner was a third. Moerner actually got it not for Super resolution microscopy, but first single molecule microscopy and spectroscopy, Betzig and Stephen Hill got Nobel Prize for Super resolution microscopy. Super resolution microscopy means doing microscopy using visible light with a resolution that is better than diffraction limit.

How do you do it? There are a well-Stephen Hill's method is you take 2 pulses 1 with a TEM 00 mode another 1 with 1 0 star mode and you overlay them and then you can play around with this and have destructive interference between the 2 pulses. At the instance when you have destructive interference what happens is this, wherever there is light in this 1 0 star mode, in the composite beam in the composite spot that becomes dark, so the beam becomes smaller in radius.

So of course when I say radius, what I mean by radius we will come to that. So once you start do not think it is just for our purpose when we do ultra-fast spectroscopy in bulk mode, we do not want 1 0 star mode. We will see why, but it is useful in Super resolution microscope. But for now, let us only focus on this TEM 00 mode in TEM 00 mode, if I want to plot intensity, this is what it looks like. So, intensity is maximal at the center and it falls off on the 2 sides.

So, in the best case scenario, you have a gaussian kind of distribution, my diagram does not look gaussian that is due to my inability to draw very nicely. But in TEM 00 mode, you do have distribution of intensity spatially and now when I say gaussian, is it two dimensional or three dimensional, there are 2 axis right when you take a spot in 1 axis and you are another axis here, so, it is gaussian with respect to this and gaussian with respect to this also. So, you start at the center intensity falls off as you go out radially. So, this is your TEM 00 mode and you will see why we are certainly discussing these are actually useful in Kerr lens for mode locking.

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Now, let us talk a little bit about Kerr effect says that if you have an intense beam, then it can modulate the intensity of the medium on which it is incident. And without going into further this is a nonlinear optical phenomenon. We are going to talk about nonlinear optics a little bit later on. But today, let us just take it axiomatically the refractive index of the medium on which an intense beam falls changes in this way in  $n_0$  is the refractive index when intense beam is not impinging on it.

When it falls then there is a second term which depends on the intensity of the beam  $n_2$  into  $I$ . Now, one thing one needs to be careful about is  $n_2$  is not what is the unit of refractive index refractive index no unit right it is the ratio but do not think that this  $n_2$  just because I have written it  $n_2$  do not think it is devoid of unit, because see the the equation has to be dimensionally consistent. And here it is not  $n_2$  multiplied by intensity does have a unit.

So  $n_2$  has the dimensions of inverse of intensity, now see what happens. You have this gaussian beam, I hope you all understand contours right? This as the contours get smaller that it represents then we are going to the peak higher intensity. So, as this is the center of the beam as you go out what will happen, intensity will fall off in this manner, as we have discussed and this is from printed book, this is a prettier diagram than I could have drawn.

What will happen to him the refractive index intensities maximum at the center right and it falls off on the 2 sides. So, what will happen is for a sufficiently intense beam the refractive index will also fall off in the same manner the refractive index will not be the same across the beam profile in the medium. Are we clear? Is there a question? So going to have this kind of a gaussian variation of refractive index in that medium this is called Kerr lensing.

Why is it called Kerr lensing? Because think of lens think of convex lens how does it work for a convex lens the any light impinging or it goes through more of the medium. So, more refraction takes place. Here, we do not have convex or concave medium we have let us say a cubicle block, what we due to intensity of the beam, we are bringing in a gradient of refractive index. So, effectively, this block which had no reason to act as a lens is now going to act as a lens. Are we clear about that this region refractive index is less in this region refractive index is more.

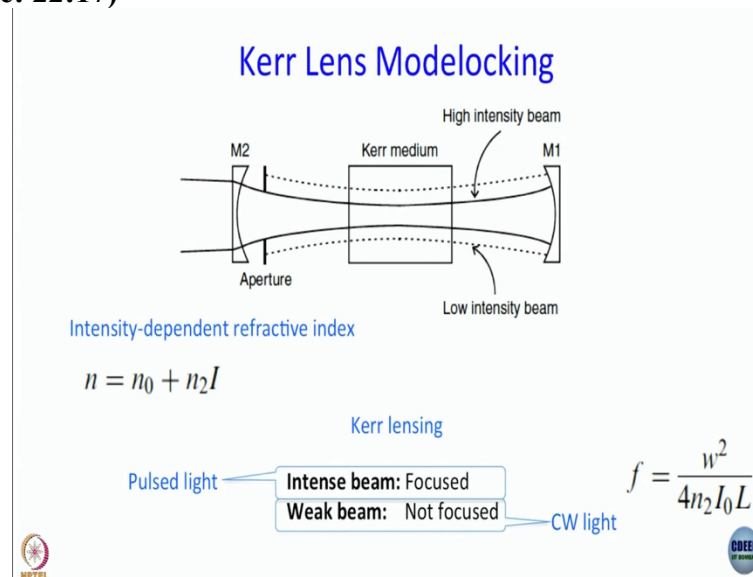
So, what happens let me put it in another way, what happens when a parallel when a collinear beam of light goes through here it goes through a region of smaller refractive index rather smaller change in reflective index from what it actually is so, it is more or less go straight as you go inside what will happen? It will bend more because effective index is more. So, eventually what will happen is this beam that was going straight will get focused.

What kind of a beam will get focused and intense beam only this is the tricky important part here. If you take torch light, make it collimated beam out of it and make it fall on a medium. You will not see kerr lensing because it is not intense. If you take the output of say helium neon laser, put it on a medium you will not see kerr lensing because it is not intense. You take a continuous wave Ti-sapphire laser make it incident on this some medium you will not see kerr lensing. You take the same laser but pulse mode you are going to see kerr lensing because remember what intensities it is  $n^2$  multiplied by  $\epsilon_0$  squared right more than number of modes better it is so mode lock laser mode locked light is what is going to produce this kerr lensing effect. So an intense beam will get focused and a weak beam will not get focused. There is do not forget the beam itself is bringing in Kerr lensing. this is the part that we really need to understand, it may not be so easy to follow when you hear it for the first time.

And then it is going to get more interesting as we go further. So, what we said is, pulsed light gives you intense beam. So, in a mixture of pulsed light and continuous wave light, what will happen you have a medium through which your pulse light and mixture of pulsed light and continuous a light of the same wavelength range is going through, the pulse light will get focused and the part will not get focused.

So if we show it, dramatically, but before that, this is just for the record. Again, since you are not deriving is not much of a fun but then just for the record, the focal length of kerr lens is given by 'w' square where w<sub>0</sub> is the beam waist, beam waist is one again is full width half maximum of a gaussian beam by four into I<sub>0</sub> multiplied by l, where L is the thickness of the medium length of the medium.

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so now with this understanding, we suddenly have something called Kerr Lens Mode locking. What is that about? Let us see, what did we say? We said that your induced refractive index depends on the intensity. So, we are starting with this that we have a mixture of CW and pulsed light in a laser somehow, so we can think like this. You started a laser, and some of the pulses have got mode locked maybe 5 pulses, and there are other pulses that are not mode locked.

This mode locked pulsed light is going to be intense. So we will get focused now thing that we have a laser consisting of your Ti-sapphire crystal nothing else. Ti-sapphire crystal and 2 lenses will introduce something very shortly that Ti-sapphire crystal itself can act as a Kerr medium is it light is going through it, it is a solid crystal. So any light that is passing through the Ti-sapphire crystal itself is going to show this Kerr lensing effect.

So this is what will happen your mixture of CW and in fact it is not a simple mixture of CW and pulsed maybe, maybe there is a gradation. So intense light will get focused like this and CW light which is not intense will not get focused or will get focused to a lesser extent. So the dotted lines denote the CW light the solid lines denote the intense light is there any reason why the solid lines are inside and the dotted lines are outside.

Do not forget that it is a gaussian beam. So intensity is actually maximum at the center. So it is a sort of a synergistic effect. So, you are going to have this kind of a Kerr medium, your pulsed light will get focused, CW will not get focused. Now, if you introduce an aperture here what will happen? So, this one near M2 here these black lines denote the section of an aperture, pinhole. The CW light in the fringe will get blocked.

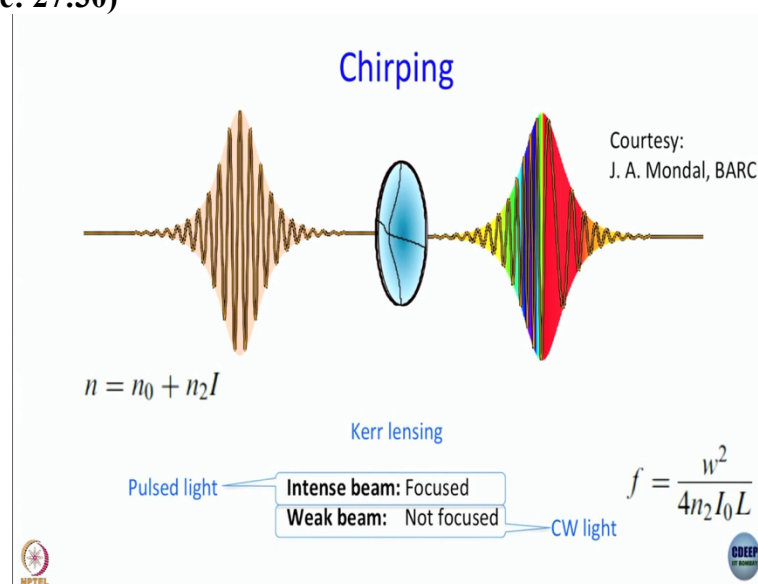
And pulsed moves towards the center will pass through. So combination of 2 mirrors, and the pinhole can give you a laser whose output is pulsed. Any question? So what I am saying is this, you understood that this titanium Sapphire laser crystal that itself can act as a Kerr medium. You are pumping it and then you have this light passing through it, the stimulated emission. To start with let us say, some modes are blocked, somehow that will happen. It is there is always a possibility of happening. So those that modelocked beam of light is going to get focused and ummodelocked CW light will not get focused.

Now, if you introduce an aperture here then the CW light which is on the outer side that is going to get cut and the intense mode lock light which is at the center will not get cut. So, he goes through so, only the first operation will be sustained CW operation will not be sustained. Now, let me say something more is that a aperture even required to understand to start with it is good to have the

aperture there. Suppose I don't have an aperture, will this thing still happen? See, you have curved mirrors right.

So, something that is focused and something that is not you make the cavity in such a way that you have curved mirrors, which support this kind of focusing here. So, anything that goes straight will not come back here. So it automatically go out of the cavity. Remember the terminal mirrors are never plain mirror they are always curved mirror. So in fact that aperture is not even required. You can get pulsed operation without the aperture this is called Kerr Lens Mode locking.

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Now, Kerr lensing gives you a pulse operation that is great, but it always makes things a little difficult for you because it introduces what is called chirp. What is the meaning of chirp? Through that, let me say that nicely animated slides that you are going to see I did not make them these are made way J. A. Mondale, BARC many years ago, she was kind enough to give me the slide. So useful. So, the thing is this in Kerr lensing you have this pulsed light incident on it.

I hope it is not very difficult to recognize this as a pulse, I am drawing the electric field not the intensity then it induces Kerr lensing, so effectively even though it is not even though it is just a slab as far as the light is concerned, the slab behaves like a convex mirror like this, what you see here is sort of a heat map for your refractive index becomes a convex mirrors and then focusing takes place all that is good. But the problem is, what is the meaning of drawing a pulse like this is

a mixture of many modes, is not it and more modes the better it is. So different frequencies and you have a lens. What is a common problem that is encountered when you have polychromatic light being focused by lens. secondary level optics different colors that is a hint the problem yes what different the speed is different exactly. So what happens and what is that called it is called chromatic aberration and you corrected by using achromatic doublets right so that sort of negates the chromatic aberration.

So here, since there are so many different wavelengths involved, do not you think chromatic aberration will happen? So as a result of chromatic aberration, what you will get is you will get a chirped pulse. some color will go ahead, some color will go behind and that will cause a broadening of us. So, before you can actually start making a laser, one needs to correct for this chromatic aberration wants to correct for chirping how we do that in the next module.