Ultrafast Processes in Chemistry
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Lecture No. 25
Active and Passive Mode locking

So, far we have discussed about titanium sapphire laser and how it gets modelocked by itself by something called kerr lens mode locking. Now, we are not so lucky in all lasers not all lasers would behave like Ti sapphire laser and get self mode locked. Especially when you want to produce picosecond pulses in nanosecond pulses, you have to put in some effort and get the pulses prepared. And one method by which as we have discussed you can get short pulses is mode locking.

So, there are 2 ways in which one can achieve mode locking one is active mode locking one is passive mode blocking. Active mode locking means, we put in some device and by applying a voltage as you are doing something, we try a mode lock the pulses. In passive mode lock in we put in an element to which we did not do anything, but by virtue of some property of this element itself, mode locking takes place. So, in this module, we are going to perform a brief discussion of both these forms of mode locking.

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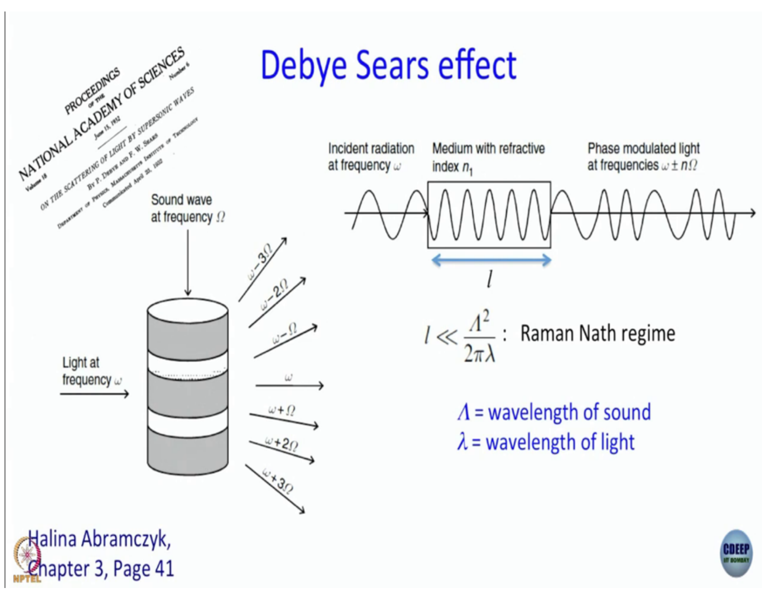

So, active locking is based on what is called Debye Sears effect or something that was known decades before the first laser was made. So, you can see this paper 1932 PNAS is where this was published. Now, the Debye Sears effect essentially means this you take a glass of water and put it on a transducer and then apply some sound wave what you see in the diagram is sound waves have been applied from the top and light goes through in a perpendicular direction.

So, what you see in Debye Sears effect is you see something like a diffraction you did not see 1 spot going through and the sound was applied rather you see several along the vertical axis in this kind of a setup not only that, the beams that are displaced from the mean position, above or below are frequency modulated and the amount by which the frequency is modulated is an integral multiple of omega, where omega is the frequency of angular frequency of the sound wave.

So, you can think like this, you see in this diagram we have alternate layers of light and dark, your subtract layers that are white and some are not white. What does this mean? Why are we drawn it like this? No not constructive interference as yet even before that, all we have done is we have put in some soundwave. Remember sound wave is a little different from your electromagnetic wave, what kind of, there are 2 kinds of wavess transverse wave and longitudinal wave.

What kind of wave sounds? It is longitudinal wave which means it propagates by alternate compression and rarefaction of the medium through which it propagates. So, in this diagram the dark region stands for an amount of medium which has been compacted and the light one is whether medium is rarefied, so, it is very easy to understand in case of a gas. Of course, but then this same mechanism happens in liquids and solids and that is how sound propagates.

So, this is essentially depiction of the soundwave that has been applied. Now, see if you have something like this a cylinder in which, you have alternate regions of high and low density does that remind you of something that we have discussed earlier something that you know about? Well, it is like a grating or you can think it is like a series of slits. Wherever the medium is verified you can think it is a slit wherever if it is compacted, you can think that it is a stop.

So what happens when light goes through? Even 2 slits, you get diffraction not you. And what is the meaning of diffraction? So that is what happens here. So essentially this diffraction, but then since you are applying soundwave, which has a particular frequency, it is not just diffraction, frequency modulation also takes place of course debye sears effect is a lot of mathematics, which I am not going to go into that is too much of optics too much of physics.

But we need to know the end result at least this is actually an undergraduate experiment in physics nowadays, one can buy apparatus like this so what you see in the apparatus in the top panel in this slide is like an aquarium in which you can fill water. On top of the aquarium, you have a transducer and a 90 degrees. Laser goes in in the setup that we have shown here. You can use a red laser or a green laser.

And in the bottom panel, you can see the spots that you can get if you hold a piece of paper or a photographic film on the other side. And you can see as you change the intensity of the sound, you get more or less number of spots. And what you can perhaps not see here is that the spots are all of different colors. can you see the different color of the spots. You can I cannot and I will be surprised if you see I suspect that is that white and red is intensity.

Because see what is the frequency of say red light, say it in hertz forget about angular frequency, how much hertz 10 to the power 4 centimeter inverse right, 10000, 20000, 10 to the power 4 centimeter inverse multiplied by speed of light in centimeter to 10 to power of 8, or 10 to the power 10. So 10 to the power 14 and what is the frequency of sound wave typically megahertz, so, 10 to the power 14, 10 to the power 6.

We are doing 10 to the power 14 +-10 to the power 6 may not be so easy to see. At least I cannot see you might be able to see, I cannot, but there are instruments that can. So, this is very roughly what debye sears effect is and this is the beginning of our attempt to do active mode locking now think why do we see this frequency modulation? Why do we have not only refraction, but also phase modulation? First of all, light is going in from air into whatever that medium is it can be quartz it can be a piece of glass, it can be something some transfer medium.

So, there is definitely a denser medium quartz or glass or water or whatever it is. So, what will happen as a denser medium frequency will change is that right? Why will frequency change because what is lambda nu? Lambda multiplied by nu that is equal to what? Actually C by n is not it? We write C because we always take n to be 1 but it is not the complete statement C by n so since n is changing your frequency and all also have to change, everything changes.

So now, suppose I change this n periodically, what am I doing in this transducer, I am applying a sound wave, and it is not just a block of glass. It is not just a block of quartz. It is a block of glass or quartz in which I am applying a sound wave. So, as an application of sound wave, the frequency is also going to you are sorry, the refractive index is also going to change periodically, because you are applying a sound wave that is periodic.

So that is what leads to phase modulation. The output in time since your periodic variation of n1 by applying a frequency some kind of frequency, the output gets phase modulated. Now, when you want to do mode locking, you have to work in what is called Raman Nath regime. Raman is the well-known C V Raman, Nath was one of his students. Raman Nath regime means L should be much lesser than capital lambda squared divided by 2 Pi lambda. Capital lambda is the wavelength of sound.

Small lambda is the wavelength of light and L is the thickness of the mode locking medium that you want to use. Now, once again a lot of mathematics will not go into we are not going to go into it whoever is interested, there are a lot of discussions about Raman Nath regime in Max Bonds Optics book for example. If you are interested you can read it but it is a non-trivial exercise. If you want to read that book you need to understand all the math.

In Raman Nath regime, this mode locking phenomenon is observed later on in the next module perhaps we are going to come across some other regime is absolutely opposite well L is much larger than capital lambda square by 2 by small lambda that is called Bragg regime. And what we will discuss is in Bragg regime you do not get this kind of an effect you get something else is close, but not the same, that is what is used in what is called cavity damping.

Now, we are discussing mode locking and remember mode locking can only take place in Raman Nath regime. Now, what is the only thing that I have control on I have not only thing perhaps I did not say it correctly. So, I know that when I want to make a modelocker I have to be in Raman Nath regime. So, what are the parameters in my control? First of all capital lambda wavelength of the sound, I can use whatever wavelength like those, it is very easy to produce all wavelengths of sound, second thing is L.

So, when the construction of a mode locker these things have to be taken into consideration. It is not as if you can take any piece of quartz and apply some frequency you will get mode locking is not the case.

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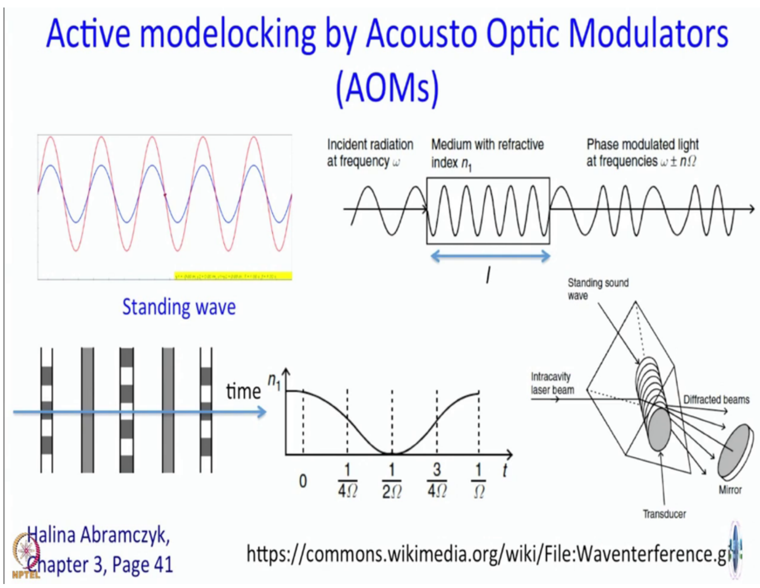

Now, what happens in modelockers occurs is that you work with not traveling waves, but standing waves. And at this point, let me ask, what is the standing wave? So I got this animation. Unfortunately, in the web, the animation is endless. Here it is not let us see if it runs you see so basically, you have this red and blue red you see 2 waves, red and blue propagating in opposite directions. And when they superimpose the give rise to a standing wave, in which the position of the maxima and minima did not change.

In fact, the position of the node also does not change it just goes up and down maximum because whenever minimum becomes maximum, not remains the same. I will do it once again see carefully.

So it is red and blue are traveling waves and the standing wave is in black, big wave. Blue, cannot be blue, Blue, red and what is the other color what you saw? Green, not black, green. So look at the green wave. That is a standing wave see how it changes with time.

So what I have here is that I have one I have a red wave going from one direction to the other. I have an equivalent blue wave traveling in the opposite direction. See what happens to the green one. It is oscillating in in such a way that the nodes remain where they are the maximum displacement, if you did not worry about sine, those positions are also the same. So this is the kind of wave that you have, so, this is what you generate, because what you do is you take quartz crystal or something like that.

You have a transducer at one end and polish the other end. So the sound waves go back and forth, and they set up a standing wave. Now the next question to ask is, very often it is easy for us to understand transverse waves, sometimes we get confused when we talk about longitudinal waves. What is a standing longitudinal wave? This is what the situation would be at different times let us say this here is your mode locker you applied sound wave from this direction.

So, at first instance this is the rarefied portion, this is the dense portion compress portion rarefied dense like that after some time it becomes homogeneous after some other time what happens is what was white now becomes black what was black becomes white that is the other extreme where maxima and minima change maxima and minima interchange. Now, tell me look at this line in time, suppose, a light goes to the middle.

Then what will the refractive index experience be as a function of time to start with refractive index is high, very high, then we will go down then it will be very high in the other direction. Then it comes to the mean position and then it becomes very high again. So, this is how it will change of course, refractive index will never become negative, it will oscillate between a maximum and a minimum value periodically.

And the point to understand here is that the period of oscillation is, it has some relation with period of oscillation with sound waves also. What is the time period associated with a sound wave whose angular frequency is omega, angular frequency is omega. So, what is the time period? So, from this graph can you tell me what the relationship is between angular frequency of refractive index and angular frequency of this sound wave or the time periods if that helps, essentially same so it makes the refractive index change periodically that is what it does.

And this is the experimental setup in an ancient spectra physics laser actually I have given only this book reference. But this is discussed a little better EW Smalls, chapter 1 chapter 2 in Topics in fluorescence spectroscopy volume one. So this is an actual diagram of a spectral physics laser from 3, 4 decades ago, the way they did mode locking is that right in front of the high reflector mirror they put in this prism.

So this is a prism light is incident from this direction the triangular faces of the prism are polished and on 1 side you apply a transducer. So, I hope you can see in the diagram that these circles denote these regions of differing refractive index; there is a direction in which the sound wave is applied. So, what will happen? The omega beam will go in a particular direction, that direction is aligned with the axis of propagation of laser is omega plus minus n delta go in other directions.

So, these directions are not sustained propagation and these durations are not sustained in the lacing action. So, you only have omega that can participate in lasing and since, the refractive index varies sinusoidally will periodically that is what causes mode locking wherever it refractive indexes is the least that is when light propagates most. And see intense light is what will propagate with the highest probability and intense light comes from mode locker.

Not only that, the period of oscillation of the refractive index in a mode locker is said to be exactly equal to the period of oscillation of a pulse in the cavity, so it acts as a gate. It lets a pulse through and then when the pulse does a round trip and comes back there it finds a gate open. Anything that comes in between is not allowed to go through. That is how; that is a mode locking is achieved.

You cannot use this technique to make femtosecond pulses. Picosecond is where this works most efficiently. So, this is active mode locking, and it is achieved by using what are called acousto optic modulator AOM's in short and acousto optic modulator are used in other applications related to lasers as well we will come to that.

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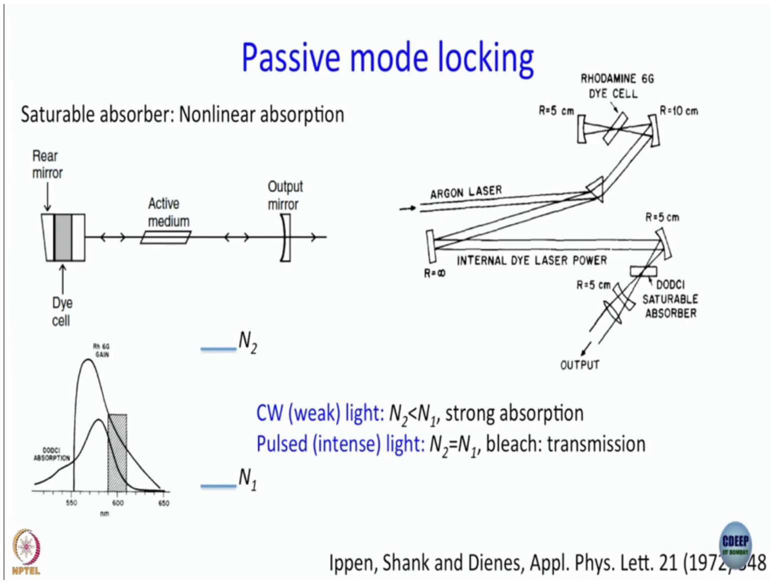

Now, let us talk about passive mode. What is the meaning of passive mode locking? In passive mode locking, what you do is you introduce an element in which you do not apply sound do not apply voltage did not apply anything there is some property of the medium by which mode locking is achieved. And the simplest passive mode locking device is a dye cell very highly considered solution of some dye over the dye has to have some property we will come to that this is called the saturable absorber.

Can someone tell what the meaning of saturable absorber is it has some excellent absorbance, no see everything will have a spectrum anyway, maximum threshold actually is a site that is called nonlinear absorption that will absorb the light until **"Professor – student conversation starts"** what is the meaning of threshold or what is the other way around, low intensity light will be will be blocked and high intensity light will be allowed to pass through and we will see very schematically what we mean by that **"Professor – student conversation ends"**

But before we go that this would be the typical arrangement of getting mode locking using passive mode locker so, you keep a solution of dye and when I say solution or dye, I mean very, very highly concentrated solution of dye. It should be so dark that if you hold it up in front of the light, you should not see any light that is usually kept in front of a mirror and we will see what you can use other than dyes also but that is not the active medium, active medium is something else.

So, now, what you see is a schematic a very general schematic of a laser with a saturable absorber passive mode locker. And this is a schematic of an actual laser once again from many years ago, you can see the year of this paper it was published in 1972 applied physics letters 1972. So, here see carefully what the beam is it is a dye laser and the dye that is used as an active medium is rhodamine 6G. Where does it absorb where does it emit rhodamine 6G, so it absorbs green and it emits in red.

So this is the cavity, here you have the high reflector. While you are here you have one of these is the high reflector, here you have the 2 concave mirrors. And here this is where the argon ion laser is fixed into the cavity by using a prism. And then this here is the cavity in the other end of the cavity, you have DODCI is dye it is a cyanine dye, you did not need to know the full name there is a cyanine dye and which has rather interesting photo physics very short lifetime.

So, in fact, lifetime of the saturable absorber is also very important. It should be short if you want a short pulse. So now, what happens is this you see this is the emission spectrum you can say I have emission spectrum of rhodamine and absorption spectrum of DODCI there is a strong overlap. So, DODCI would absorb the light that comes out from rhodamine the emission of rhodamine. Now see, let us consider 2 levels in DODCI 2 levels involved in absorption process.

And let us say population of the lower one is N1 population of the high level is N2. So, if you have a small intensity then what will happen there they really absorb when does absorption take place when does transmission take place? Absorption will take place as long as N1 is less than N2 if you somehow achieve N1 equal to N2 then you get what is called bleaching, so if you have a weak beam of light it will just get absorbed.

And then the molecule comes down from its excited state to ground state and then it is reset once again once again N1 is very large number and N2 is practically 0. However, what happens when there is a strong beam? If have a strong beam, then it is possible to almost achieve population inversion, almost, but not quite, almost and as you know, and as you worked out yourself, pulsed light is really very intense.

So, let us say we have this laser cavity in which we have CW light propagating and pulsed light propagating as well. CW light will be absorbed by the saturable absorber and therefore stopped. This propagation will be hindered pulsed light, because it is intense, will cause bleaching and we will get transmitted. So, this saturable absorber is going to select pulse light over CW light, so let me show you something.

This is the actual data of pulse width how we measure pulse width we will come to that sometime later, but this is the pulse width measured in 1972 using this absorber using this apparatus using this laser that you see what is the pulse width written there? 2.3 picosecond, so in this laser, you can get almost 2 picosecond was reported in 1972. So, here in this setup, what is happening is the rgon ion acts only as a pump.

If there is no saturable absorber, you are going to get a CW operation of the dye laser. Since the saturable absorber has been inserted, that is the only mode locking device there itself you can get a pulse width that is as narrow as about 2 picoseconds. So, what happens is you can do better than this you can do better if you have pulsing already and you use the saturable absorber only as a selector, but not as the primary mode locking device.

One thing that I want to say before closing this part of the discussion is and I was actually hoping that you are going to say will you agree with me if I say that the pulse gets narrowed as a result of this? See let us consider that there is a pulse somehow mode locking has taken place. That always happens some amount of mode locking will take place. So, that modelocked pulse is there. Now, if you have saturable absorber while going through the leading edge of the pulse is going to be absorbed, is not it?

Do you agree your pulse something like this in time initially at the onset of the pulse intensity is 0, then it goes up gradually, well quickly to a maximum and then falls again. So, what will happen to the leading edge of the pulse that will be absorbed? And that light will be used to produce to increase N2 more and more then there will we will reach a time when bleaching will take place. And from that instant onward the pulse will go through do we agree.

So, this induction time for which the population of the excited state is being prepared, there is a time when even the pulse will not be transmitted it will be absorbed. So, that portion of the pulse is cut off. So, you end up so, let me give you an example, let us say I have a 20 picosecond pulse 20 picosecond full width half maximum. And let us say to keep the discussion simple. It takes 20, picosecond for bleaching to happen what does that mean? What was full width half maximum of the pulse now becomes the base of the pulse.

So, full width and half maximum perhaps becomes 2 picosecond, 3 picosecond something like that, so it is important to understand that passive mode locking leads to narrowing of pulses as well, so we will stop here, and we will continue the discussion in the next module.