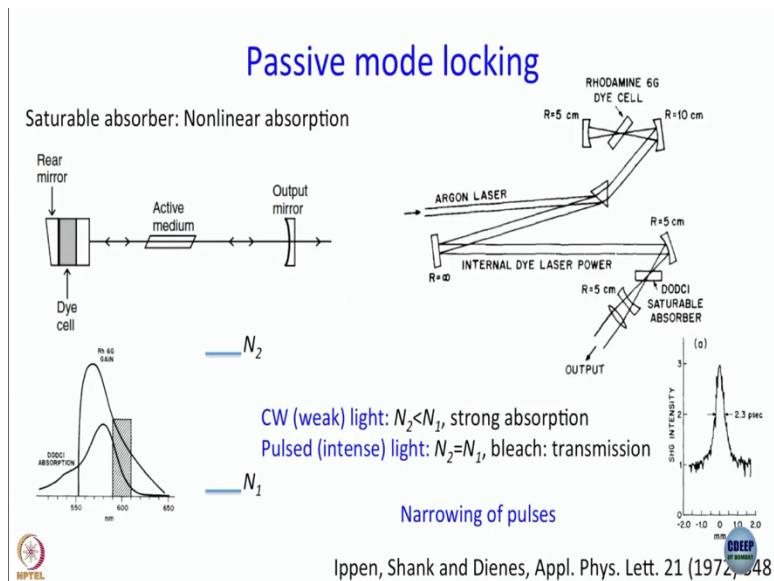


**Ultrafast Processes in Chemistry**  
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**Lecture No. 26**  
**Modelocking and Cavity Dumping**

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We will continue our discussion of mode locking and then go on to something called cavity dumping. This is where we have got until the last module, which we have discussed active mode locking using a mode locker by applying sound frequency using Debye Sears effects within the Raman Nath regime. And then we moved on to talk about passive mode locking, where what is used is saturable absorbers, so, and we have finished our discussion saying saturable absorber can not only produce pulses.

But they can also narrow them down now, what I want to say is, saturable absorber works even better if it is not given the work of producing pulses as such, but pulses are produced already and the only job of the saturable absorber is to narrow the pulses down.

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## Synchronous pumping

**Pump:** pulsed laser like modelocked Nd: YAG

**Cavity length:** Same as that from pumped laser

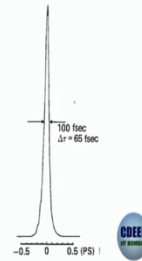
Ausschnitt and Jain, Appl. Phys. Lett. 32 (1978) 727:

Pulse duration of dye laser = Square root of that of pump laser

Kafka and Baer, Proc. SPIE 0533 (1985) doi: 10.1117/12.946538

**Saturable absorber:** Further narrowing of pulse

Seikai and coworkers, Optics Lett. 12 (1987) 681



And that is achieved very commonly in dye lasers especially and also in some kinds of solid state lasers by what is called synchronous pumping. In synchronous pumping, I pump the dye laser with not a CW laser, but with a pulsed laser. So, what happens if I pumped with a pulsed laser what kind of output do I get from the dye? We are talking about rhodamine 6G dye lasers, I have a rhodamine 6G dye lasers I pump it with green light 532 nanometer but the 532 nanometer light itself is say 20 picosecond pulses 80 megahertz or something.

What should the output of the dye laser should be, I will give options will the output be CW or will it be pulsed that was a very simple question. But I am exciting with the pulse laser if I excited with pulse it should be output also not be pulsed, why the pulses at 20 picosecond lifetime of the dye is say 10 nanosecond, say 5 nanosecond and repetition rate is such that the difference between 2 pulses is something like 20 nanosecond should I not get pulses like this.

Every pulse of pump is going to produce a burst of red light and when the pump is off, then what will happen? There is nothing to produce a burst, is not it? So, you get an inherently pumped laser. But the problem is these pulses that you produce, are going to exactly they are not going to be sustained unless the cavity length of the dye laser is exactly the same as the cavity length of the pump laser.

So, you have to carefully match the cavity length of the pump laser and the dye laser, this is called synchronous pumping. Pumping is by a mode locked in the laser or something like that and cavity length is the same as that of the not pumped laser, I made a mistake there pump laser that is used for pumping, so already the pulses there and then without going into the math once again there is this 1978 paper which says that in synchronous pumping pulse duration of the dye laser is square root of that of the pump laser.

So, in synchronous pumping you can actually get shorter pulses than the pump laser itself that is why this was a very common technology for a couple of decades before the advent of titanium sapphire lasers, so square root of that of pump laser. So the good thing is it is not very difficult to produce a pump laser which has say 20, 30, 50 picosecond full width max but let us see what 25 picosecond pulse width laser that is used for pumping.

Automatically the output of the dye laser is going to be 5 picoseconds, square root of that of laser and I am asking you to believe me on this will not do the math. If you are interested, please read this Apl papers from 1978. Now, I should show you some result. This is a result Kafka and Baer I think where engineers working in spectra physics, we remember correctly. And this is what they produced in Nd YAG, pulsed Nd YAG pump.

Synchronous pump rhodamine 6G laser, what is the full width of maximum with you read here, 220 femtosecond. So, if you pump laser itself as a short enough pulse width, then you can actually go down to 220 femtosecond just by synchronous pumping that is how good it is, but then, later on the modified the cavity a little bit Kafka and Bear not be from meta physics they might have made a mistake. This optic slater is from spectral physics.

So, this is an again a laser, do you see the cavity, let us look at the cavity carefully. Here we can right away see some elements that we have discussed earlier we see our familiar 4 prisms and that is what will give you tunability anyway. Here this is the output coupler, the partial reflecting beam through which the light will go out of force in the other end will be the high reflector. And this is what the pump is and we have discussed Ti sapphire already.

Do you see a major difference between pumping geometries of Ti sapphire laser and dye laser? Remember in titanium Sapphire laser, the pump green light went through 1 of the mirrors. He said there is a dichroic mirror inside Ti sapphire. Why because in case of solid state lasers, you get the best result if the pumping is coaxial with the laser, with the lasing axis. Not so in case of dye laser. In dye laser pumping can at some angle and that angle is also optimized.

Another thing I would like draw your attention to is what is written here. What does it say? Gain jet. What does that mean? Gain of course, gain medium rhodamine 6G dye, what is the meaning of jet so as you know, when very short pulses go through any medium they get broadened to do chirping and all. So, in order to get short pulses which are picosecond or lesser you did not want any extra component to come in the path.

So, gain jet means, in these lasers what they do is the dye would be stored in as a small bucket and it has a lot of dye, 1 liter or something, a lot of dye solution rather, and typically the solution would be in glycerol some such very highly viscous solvent and there used to be a pump which would circulate this dye. So it would circulate and bring it out through a jet. Jet means a metal nozzle, which would be flattened.

So, the output of the dye through that nozzle would be a flat jet, spherical usually if you take a hose pipe, what kind of jet do you get? The cross section will be circular, that will not work here you want it to be flat and as thin as possible. That is why the jet was meant it was very, very thin and then it would go into a scotched tube which would be connected to the receiver to go back there, this was the state of the art for again over 20 years or so, in picosecond, then femtosecond lasers, one would use jets.

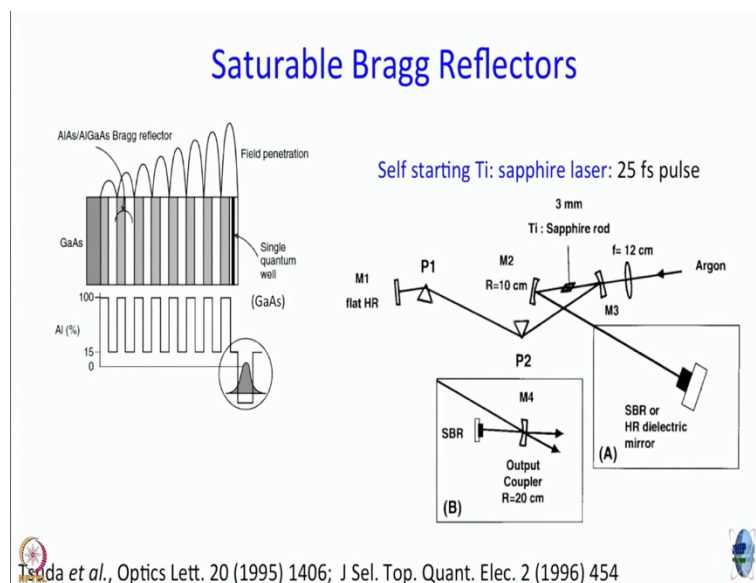
And in fact, even now, a lot of people do femto experiment where the sample is not a rotating sample or translating sample or anything but a jet because then there is no quartz coming in the path. No further broadening of the pulse, we really want to go down to small timescales jet is the way out, but I digress too much. And then here you have this gain jet. And here do you see there is a saturable absorber jet? What is that DODCI once again circulated in the same manner.

So, now you see, rhodamine 6G already pulsed and you know how good the pulse can be? Or what do you say 220 femtosecond or something like that. Now, the question is by introducing this; saturable absorber is there any improvement? And this is the answer. What is the full width half max now? 65 femtosecond? No, the lower number is correct. I did not know why they are written 100 there but I checked with the text 65 femtosecond is what they say from 220 femtosecond it has gone down to 65 femtoseconds because now think 220 femtosecond pulse.

That means the base would be something like we 400 femtosecond from there to say 200 femtosecond or so, for that time that that is the time required for the light to actually get absorbed even of the intense pulse and to have an increasing amount of bleaching. When the threshold is reached, then only the pulse propagates that is why the pulses narrower. So synchronous pumping with saturable absorber for a long time, used to be the state of the art for producing short pulses.

Of course now it is a thing of the past. Nobody would do anything other than titanium sapphire laser. But it is important that we know the principles, at least qualitatively. That is why we are discussing this.

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But everything is said and done nobody likes to use dye solution and all that anymore because first of all, they are messy Secondly, dyes degrade very fast, especially saturable absorber. Rhodamine 6G is stout dye. Not much happens, but DODCI is extremely flaky. I worked with this kind of a

setup we would make a DODCI solution today and it would be very intense dark color within a matter of weeks. There will be no color in the jet at all. It will bleach completely.

Because if you leave DODCI solution on the table under tube light, it will get bleached so that is a very big problem and then when you are done with it, where do you throw so much of solvent and there is 100 other paraphernalia there. Whenever you are solid, the pump can go bad, the solvent chamber can leak you have to cool it. It is really messy. So, that is really a thing of past now. So, right now what everybody wants to use is your solid state solutions.

And something that is used now, very often is a passive mode locker made of; will not made of is perhaps not the correct term. Something what is used is a saturable Bragg reflector, which act as saturable absorber and therefore passive mode locker, but I should explain what a saturable Bragg reflector is even before going there. I should tell you what a Bragg reflector is does anybody know what is Bragg reflector?

Whenever we say Bragg, what do you think? Diffraction? Whenever you say diffraction, what comes to your mind is alternate layers a lot of apertures and all that. So, a bragg reflector is something like this you have sort of a mirror and in the example that we discussed here the reflective surface is gallium arsenide and then your alternate layers of high and low refractive index. So, when light falls on it what will happen?

Light goes in first of all, let us not worry about this thing on the top from this directional light goes in just reflected and comes back. But then when it comes back, since you have different layers, what kind of light will come out? Let me take the question down to an easy level. You have grating or you have this crystal and all, light falls on it then light goes back. Everybody knows the rule, which tells that at which angle it will come out what is the rule? Bragg's law?

What is it  $n\lambda = 2d \sin \theta$  I am asking an easy question I never asked difficult questions. So, where does the  $n\lambda = 2d \sin \theta$  come from? Again answer is easy. So, remember what you studied in childhood, you are 1 reflecting layer another reflecting layer.

So, you get reflected the ray from the first and another reflected ray from the second. And then what you do is you have to say that you have to have constructive interference.

So path difference must be integral multiple of  $\lambda$  by 2. That is how you derived  $n\lambda$  is equal to  $2d \sin \theta$ . So in other words, so this is a Bragg reflector, is not it? What I just discussed your crystal are whatever it is a Bragg reflector and what is the property of the light that comes out? All the rays that constitute that beam are in phase. So now if you use a reflector like this light that goes in, and then light comes out, whatever light is able to come out of this Bragg reflector has to have components that are completely in phase or in other words mode locked.

So, when you use a Bragg reflector, automatically you get mode locking. So, this is the state of art we have it is a little later in the slide, where these are used in auto start Ti sapphire lasers. You might remember that in 1 of the previous modules as I told you that when we work with homemade Ti sapphire lasers to get the mode locking started, we had to tap a meter in the tsunami that we have in our lab, there also to get the mode locking started we have to disturb the cavity a little bit.

However, in MaiTai the new laser that we have the compact laser do we did not have to do anything. Why? Because Maitai uses a saturated reflector we have not even go to the saturable part Bragg reflector. So automatically light that comes out is mode locked. So, that is why one wants to use Bragg reflector and if you want it to be a saturable Bragg reflector, what you do is you introduce what is called a quantum well the substance is returned here once it is used, what is the meaning of quantum well?

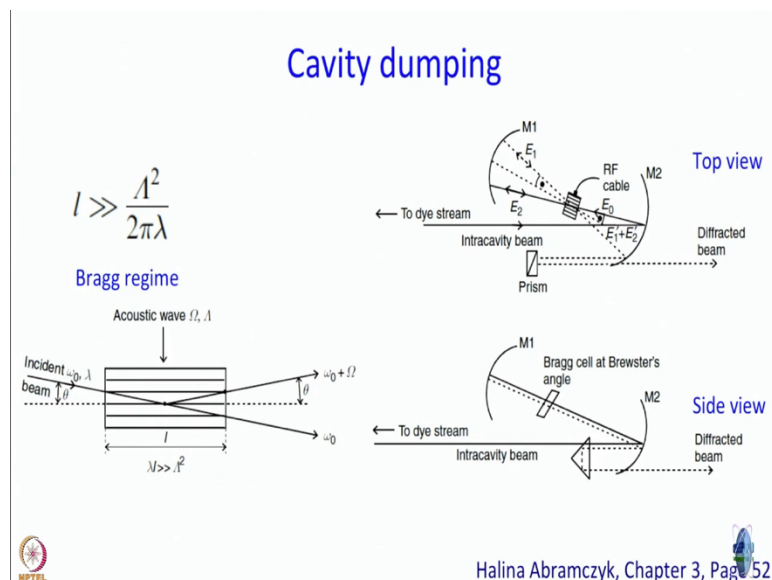
Your substance is small bandgap sandwiched between 2 layers of another substance which has large band gap once they excite on gets tap that is not so easy to come out what does that mean? When will it come out when intense light falls on it? So that is the saturable absorbing action. So Bragg reflector with a layer of quantum well is this saturable absorber of choice in present day nobody uses DODCI anymore. DODCI sales must have fallen worldwide might; of course this is made by layer by layer assembly and this is an example.

And what I want to draw your attention to today is how old the technology is 1995, 1978 I told you is when femtosecond pulses are already there 1995 is when you have the use of such saturable Bragg reflectors. The diagram you see here is diode pump solid state laser and I think you can more or less make out all the components what is going on this here is a saturable reflector SPL and that is what causes passive mode locking.

What is the full width half maximum here 100 femtosecond not as good as 65 femtosecond but this as hassle free nobody cares and also the comparison may not be fair because there you had a synchronously pump dye laser which as saturable absorbers, here you have a diode pumped solid state laser. Inherently pulse width is going to be more but then now I show you another one from references from there in the same paper and it is there in a laser focus world article as well. This is a self-starting Ti sapphire laser.

Once again you see there are 2 different cavity designs. But the crux of the matter is that this saturable Bragg reflector is used as 1 of the mirrors. Now see what the pulse width is 25 femtosecond. So good thing is first of all, Ti Sapphire itself gets mode locked by KLM into it you are introducing a saturable absorber. So the output is as good as it gets. So in all, not all, but in most of the modern day compact, self-starting Ti sapphire lasers this technology is used.

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Now we move on to something that is a little different but it is very easy to get confused between this so far we have been talking about mode locking how to produce a short pulse. Maybe not an ultrashort pulse, but short pulse nevertheless and why not an ultrashort pulse just showed you that you can go down to 25 femtosecond by mode locking. Next question is how do you get the pulse out? You can get the pulse out by using an output couplers.

But especially in the dye laser era, the question that people ask is; is it possible to do better, is it possible to use some other mechanism by which there will be further amplification whereas further narrowing of pulse and we can change the repetition rate. So, I will give you an example. This all those dye lasers, rhodamine 6G dye laser that I showed you, they are synchronously pump by green lasers which have a repetition rate of about 70, 80 something like that.

As you understand, if you want to do say fluorescence experiments TCSPC experiments 70 megahertz is too fast you have to cut down the repetition rate, how do you cut down repetition rate? So, as long as dye lasers rule the market cavity dumping was the technology. Unfortunately, titanium sapphire laser cannot be cavity dumped, it would have been great if it could be. So, that is why you have to use something else called pulse picking. But let us talk about cavity dumping first.

If there is time we will talk very briefly about pulse picking and then we will pick it up next day. So, in cavity dumping, you use the same thing you use an acousto optic modulators you take a quartz crystal or something and use the transducer but now, you did not work in Raman Nath regime anymore. This is the main difference. You work in what is called the Bragg regime and Bragg regime condition is exactly opposite of Raman Nath regime here  $L$  is much greater than  $\frac{1}{\lambda^2}$  that is where cavity dumping is observed best.

So, first of all, what do you have to do compared to mode locker you have to use a higher frequency of acoustic wave, because  $L$  much larger than  $\frac{1}{\lambda^2}$ , you have no control over  $\lambda$  depends on what kind of laser you are working with. You do have control over  $L$ , which is the wavelength of light. So,  $L$  has to be much larger than  $\frac{1}{\lambda^2}$  divided by some constant.

Then, that capital  $\lambda$  should be small or in other words, the frequency of the acoustic wave should be large also, the other thing have in your hand is the length. So you can use a longer interaction length. You can use thicker crystals. Now, of course, there is a limit to that, because if you use something that is too thick, then again passes will get burden so, you always have to find the optimal length.

So, what happens when you work in Bragg regime is this, remember what happened in Raman Nath regime, light passing through was diffracted on the 2 sides and was phase modulated by amount of  $n\omega$  here that does not happen? All the higher order diffraction is eliminated, you did not get it. Why you did not get it? We have to read the original papers, but we, for our purpose, you did not even need to know.

You did not get higher order deflection at all only 2 rays are sustained the zero order that is unmodulated  $\omega$  and the first order  $\omega$  zero plus capital  $\omega$ . So this is what happens in a Bragg regime, are we have we understood? What happens? Can I go ahead only 2 beams now and that is important for cavity dumping and this is what a cavity dumping is made up off. There is some math after this maybe I will skip all of that and just show you the final result you can go through the math if you want.

So, here what happens is first of all, now, here we have an interesting situation. All this time we have been saying that in a laser you have a high reflector and we have an output coupler in these lasers is no output coupler all mirrors are high reflector so in absence of cavity dumper what kind of a laser is it? You pump it lasing will start there will be gain but there will be no loss it will never come out.

So there is no output, can we do cavity dumping is a device that is put into a laser like that to get the pulse out to switch the pulse out how the same thing is a part of the laser cavity. On this side you can understand there will be the gain medium and all. And the other end other high reflector. Here you can think this is the last mirror M1 in this case is last mirror, first mirror in this whatever it is one you can see it is first mirror, last mirror is somewhere else.

But the crux of the matter is no output coupler. Here you have M2 both are concave mirrors, focusing mirrors. And we have this AOM at the focus of both center of the acousto optic modulator is that the focus of these two mirrors, maybe I will just take this and stop and start from here next day. So, think of this horizontal line, the horizontal black line is the lasing access comes here goes through. Now, this acousto optic modulator is being operated in Bragg regime.

So what will happen? Zeroth order one will go through and there will be the first order line which is frequency modulated by just a little bit. So, now you will have 2 rays then since it is constructed in such a way that these rays hit well, I did not even have to say that this one is at the center. So, these rays are made to retrace their path. Now, what happens when the retrace the path on this side also you will have 2 rays understood one of these will go back to the laser cavity.

The other one which takes a different path will not go through as a cavity is going somewhere else. You put in a prism there and it goes out, so that is how dumping is done. What is the advantage? We will start discussion from here next day but what is the advantage of this. See, we have used the pulse picker. What is the difference between cavity dumper and pulse picker? In pulse picker the acousto optic modulator is outside the cavity.

So, what are we doing? We operate the laser at 80 megahertz and we operate the acousto optic modulator at 8 megahertz. So, we are taking 10% of the pulses from the dumping the remaining 90% of the pulses. We will discuss later again what is happening in cavity dumping either the light goes out and you use it or it goes back to the cavity nothing is wasted. And much of photons that go back to the cavity is going to get amplified in the next round trip.

So, in cavity dumping, this is the attractive feature of cavity dumping. You take out the light at the same time you have further amplification so whereas your power is going to get cut down, if you do pulse picking, you mentioned the power before pulse picker and after a pulse picker, if you operate, it is a 1 - 10th ratio. Your power is also 1 10th, is not it? Because you are taking only 1 - 10th of the pulses.

But in dye laser first of all, if there is no cavity dumping, you cannot even measure anything no light will come out. But due to cavity dumping, what you see is there is an increase in power because, while the light is taken out, only one part is taken out, the remaining part is sent back to the cavity. No photon is wasted. Nothing is dumped; this is where we will start from next day.