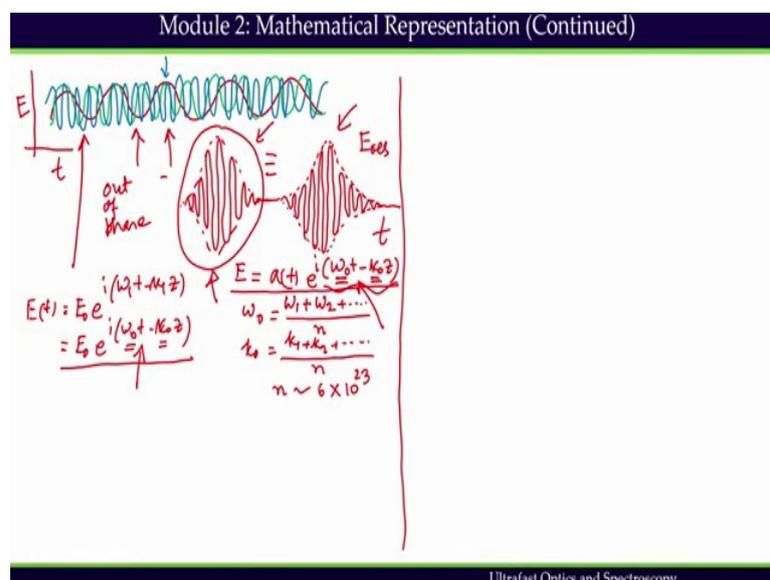


**Ultrafast Optics and Spectroscopy**  
**Dr. Atanu Bhattacharya**  
**Department of Inorganic and Physical Chemistry**  
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**Lecture - 05**

Welcome back to the module 2 of this course “Ultrafast Optics and Spectroscopy”. We are continuing this module and what we have studied so far different characteristics of ultra fast pulses using mathematical formulation. So, we will continue this module. So, far what we have seen that a short pulse can only be constructed using optical interference of plane waves.

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We will draw plane waves, we have one plane wave like this and we will consider slightly different frequency represented by different color. So, we are drawing two plane waves, they are interfering with each other, we will select one more color; one more frequency component.

So, there are multiple frequency components we can consider and what we see here is that at this point we have constructive interference, at this point we have destructive interference and then there are points where we have destructive interference. So, this is going to be in phase and this one is going to be out of phase and due to this interference

what we are plotting here is the electric field with time. Similarly, the resultant field can be shown like this way, this dot line just to guide our eye.

So, what we see is that the resultant field is varying like this way which is nothing, but a pulse. Now, this field variation will repeat after a certain time again there will be in this position, there will be another pulse in time and it will go on like this way. So, I am plotting here again resultant electric field due to this interference and often we have pointed out that this field can be expressed isolated pulse can be expressed as  $a(t)$  multiplied by  $e$  to the power  $i \omega_0 t - k_0 z$ . (Please look at the slides for mathematical expression)

Here,  $\omega_0$  is the resultant or the average frequency component, I have so many frequency components let us say I have Avogadro number of frequency components, so I get the average of it. On the other hand, this  $k_0$  is representing the average wave vector component wave vector magnitude of the wave vector  $k_1$  plus  $k_2$  plus blah blah blah divided by  $n$  and  $n$  could be Avogadro number which is approximately  $6 \times 10^{23}$ .

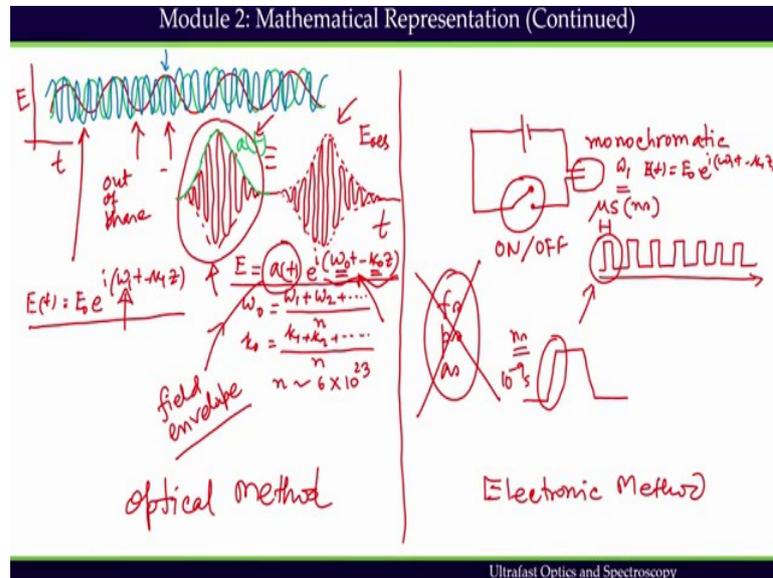
So, let us say I have this many numbers  $10^{23}$  numbers of plane waves interfering with each other, if they are interfering with each other in time we will have localized electromagnetic energy or the burst of electromagnetic energy. Each pulse can be expressed with the help of under SEVA we can express by this equation; this is then equation representing an isolated pulse not a pulse train. And  $\omega_0$  is not a single frequency component  $k_0$  is not associated with single frequency component they are representing the average value of the frequency angular frequency and wave vector.

On the other hand, if we think about one plane wave can be written as  $E$  which is function of  $t$  equals  $E_0 e^{i(\omega_0 t - k_0 z)}$ . So, to avoid confusion sometimes we also write like this way also  $e^{i(\omega_0 t - k_0 z)}$ , when we write  $\omega_0$   $k_0$  for an plane wave and when we write  $\omega_0$  and  $k_0$  for a pulse; we should remember that the meaning of this  $\omega_0$  and  $k_0$  in pulse are different. (Please look at the slides for mathematical expression)

In the pulse, it is representing the average values and in plane wave, it is representing its characteristic frequencies and the wave vector. So, generally to avoid this confusion we

generally use, but we do not use  $\omega$   $k$   $z$  we use  $\omega$   $1$ ,  $k$   $1$   $z$  to represent plane wave. (Please look at the slides for mathematical expression)

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And, for plane wave it is single frequency component and for a pulse it is actually multiple frequency component and this  $a(t)$  is the field envelope, this is called field envelope; how the modulation of the magnitude will occur and this field envelope is nothing, but this dotted line. So, how this modulation will occur that will be defined by this envelope function. So, this envelope function if we add this tip of this, then we get this envelope function and this is represented by  $a(t)$  here.

So, what we have seen here that interfering waves having slightly different frequencies produce pulse. The resultant field can then be represented by a pulse which is nothing, but localization of electromagnetic energy in time. Here, one can also argue that a pulse does not need to be always synthesized using optical method which is interference, what we are showing here in order to produce a pulse I need many frequency components that we have shown.

But one can argue that I really do not need many frequency components, I can use an electronic method. So, this is an optical method to produce a pulse, but I can have electronic method as well and in this method what we can consider I can write down an circuit I have a source, then it is connected to a light source and then I am using a switch let us say.

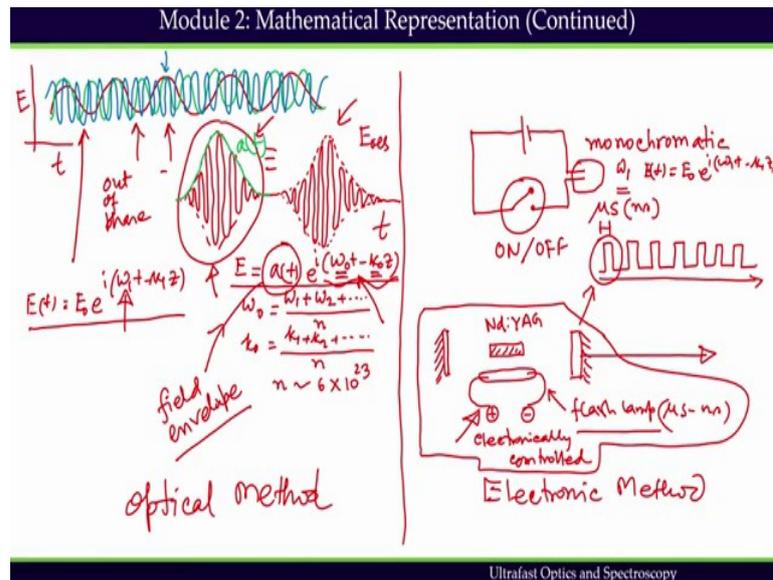
So, if I have this circuit then I can switch on and off this is a switch, so I can make it on or off and I will assume that this is a light source rather than a monochromatic light source let us say. It is very difficult to get a monochromatic light source, but we will consider that we have a monochromatic light source. Question is with this configuration can we prepare a pulse, can you synthesize a pulse? Monochromatic which means it gives only one frequency component which means that I have only this  $E(t) = E_0 \cos(\omega t - kz)$ , whenever the circuit is on. (Please look at the slides for mathematical expression)

So, if I switch on and off then as a function of time if I plot everything what I get is that a pulse again a pulse; again a pulse; again a pulse on off; on off like this way we will continue and we will be able to control that. So, this way we can actually create a pulse and a temporal localization or temporarily localize bursts of energy with single frequency  $\omega$ , it can be synthesized using electronics that is true we can do that, but its pulse duration can be mostly microsecond. So, the duration of the pulse can only be microsecond it cannot be femtosecond; it cannot be femtosecond, it cannot be picoseconds, it cannot be attosecond it is not possible.

The pulse duration is going to be microsecond at best it can be nanosecond, if it is not microsecond if we can improve much then it can be of the order of nanosecond not less than nanosecond. It is because the response time of electronics is nanosecond when we switch on and off, if we look at this pulse it is nothing, but I am switching on and off. So, this is 0 voltage suddenly I have something then again certain value and then I am closing it. So, the time by when I can switch on a particular device with an electronics it is going to be nanosecond because the response time of all electronics is going to be nanosecond  $10^{-9}$  second, it cannot be faster than a nanosecond.

So, the ultimate time resolution of this kind of idea synthesizing a pulse will depend on how quickly we can switch on and off. And the fastest time scale we can achieve with the help of electronics is going to be nanosecond that is why the pulse duration at best we can produce with the help of this kind of methodology is going to be nanosecond pulses or microsecond pulses. In fact, electronically controlled flash lamp is used in the construction of nanosecond pulsed lasers such as Nd YAG lasers, we can draw Nd YAG laser cavity here and I will draw the cavity to show you how this electronic approach is used.

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We need a cavity optical, cavity we will learn all this optical cavity very soon in this course. This optical cavity is prepared with the help of two mirrors facing each other and then I have this lasing medium which is nothing, but in Nd YAG rod, this Nd YAG rod is very common laser medium which we use in the synthesis of nanosecond pulses.

And then this rod has to be excited this medium has to be excited, we do this excitation with the help of flash lamp and this flash lamp is nothing, but a electronically controlled flash lamp and this is called flash lamp; a flash lamp and in flash lamp, what we do? We do is this on and off. So, basically it is flashing, but the time scale by which we can do this flash is going to be microsecond to nanosecond. And this is purely electronically controlled flash lamp; electronically controlled flash lamp.

So, it is very important that we realize that a short pulse such as picosecond, femtosecond and attosecond can only be synthesized using optical interference method only, this is why often we say that an ultrafast laser cannot be externally triggered. See if we think about this Nd YAG nanosecond laser this can be externally triggered; external triggered which what does it mean? It means that I can operate this externally, I can operate this change of on and off for this flash lamp externally or electronically and then I can get the output.

So, I can trigger it externally, but ultrafast pulses cannot be ultrafast laser sources cannot be triggered externally just because there is no electronics available currently which can

trigger with at that kind of time scale femtosecond or picosecond time scale is way too slow all electronics are way too slow for the for the ultrafast laser system. That is why, we have to always use its internal trigger to externally trigger rest of the devices such as pulse nozzle counting electronics and etc. Ultrafast laser system cannot be triggered in the counting experiment it cannot be triggered externally it has to be triggered internally.

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Module 2: Mathematical Representation (Continued)

### A Plane Wave and a Pulse

A Plane Wave

$$E_0 \cos(\omega t - k_1 z)$$

$$\vec{E}(z,t) = \vec{E}_0 e^{i(\omega t - kz)}$$

A Pulse

$$E = a(t) e^{i(\omega_0 t - k_0 z)}$$

avg. freq. comp.  
 $\frac{\omega_1 + \omega_2 + \omega_3 + \dots}{n}$

Under the Slowly Varying Envelope Approximation

$I \sim E E^* = (a(t))^2$

$a(t) = e^{-at^2}$

$I \sim e^{-2at^2}$

Ultrafast Optics and Spectroscopy

So, these are the concepts which we have already developed in this module. And this is the plane wave we are showing here one plane wave propagating along z direction and one pulse propagating along z direction both are propagating along z direction. As pointed out previously a plane wave will be represented by this equation which is nothing but a cosine modulated oscillation, the real part of this is going to be in scalar quantity it is going to be  $\cos(\omega_1 t - k_1 z)$ . This will be expressed as 1, to point out that this belongs to a plane wave. (Please look at the slides for mathematical expression)

On the other hand, a pulse propagating along the z direction having similar kind of equation except for this envelope function this is called field envelope, it is associated with the field and the average frequency component, I said previously that average frequency component is also called carrier wave, this is the average frequency component.

Average frequency component how do you get that? All the frequency components which are interfering they will be summed and divided by its number. So, that we get

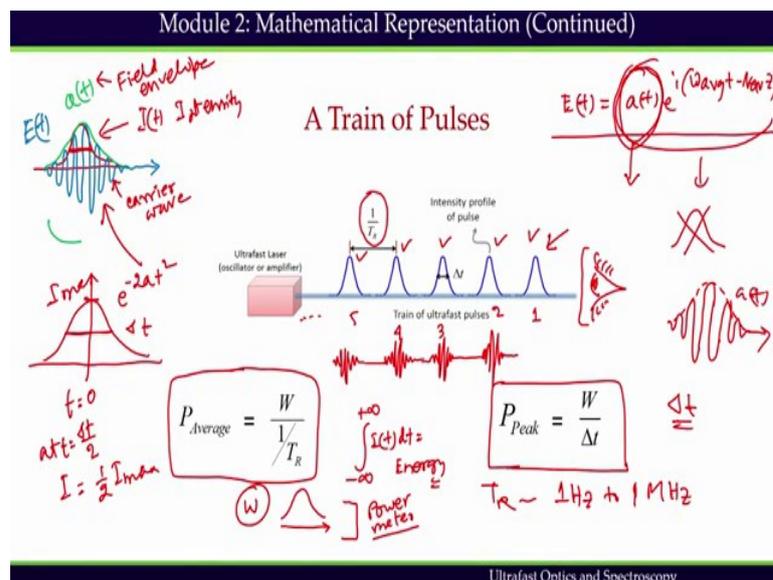
this average frequency component and that is called carrier wave of a pulse and field envelope is the modulation the amplitude by which we will be able to modulate it. But this is under slowly varying envelope approximation this expression suggesting that we have an isolated propagating pulse, propagating along the z direction.

Now, the pulse here we are representing varies and also we have seen that in general when you talk about the pulse we also present its intensity profile and intensity profile goes like the way given in slide. It is not exactly field envelope, it is slightly different from field envelope because intensity is nothing, but  $EE^*$  which is nothing, but  $a(t)^2$ .

So, if  $a(t)$  is represented by a Gaussian function, then its intensity is going to be represented by its square that is why we have the intensity profile inside the field envelope it looks like this. If we consider the normalized functions then intensity profile is going to be like this and its field envelope would like would be would like this as shown in slide.

So, their widths are slightly different and mostly we talk about intensity. In intensity profile we should remember that there is no oscillation, it is just an envelope function intensity envelope function.

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Now, this pulse is an isolated propagating pulse. In general however an ultrafast laser source generates a train of pulses; a train of pulses is represented by here we are showing

intensity profile. Generally when we represent a pulse we just show like this and what does it mean? It just we actually try to show its intensity profile.

If this is intensity profile we should remember that actual electric field would be like following, actual electric field would be in a different color I will present it, actual electric field would be like this is the field envelope and then field carrier wave is would look like this; carrier wave would be something like this; this is going to be the carrier wave. And so when quickly we are representing a pulse we do not show all these oscillations, generally we just show the intensity profile and we should not get confused by looking at different representations.

Now, any ultrafast laser source will produce a train of pulses. And for train of pulses if we have an observer sitting here, then what will happen, what I will see as an observer? I will see one pulse, then another pulse, then another pulse, then another pulse, then another pulse and so on they are coming to the observer at a particular repetition rate. And why we get this repetition rate, we will be able to understand all these details when we will talk about the construction of ultrafast laser system.

But what we have shown here so far is that the expression which we have written  $E(t)$  equals  $a(t) e^{i(\omega t - kz)}$  the power is  $\langle |E|^2 \rangle$ . I will write down  $\langle |E|^2 \rangle$  again  $k$  average  $z$  this is the expression I have represented, this is an isolated pulse propagating along the  $z$  direction. And how do I represent this pulse? The pictorial representation is not going to be like this it is not like this, it is actually a field envelope which is  $a(t)$  and then its carrier wave field envelope and its carrier wave. So, this is the actual representation of this expression. (Please look at the slides for mathematical expression)

Now, this is representing one isolated propagating pulse, this does not represent a train of pulses. To represent train of pulses, we have to explicitly write down the expression for  $a(t)$  which we have shrink to  $a(t)$  there is an expression for the train of pulses, we have shrinked to  $a(t)$  just because we used SVEA: Slowly Varying Envelope Approximation. But, if we want to represent train of pulses we have to get an explicit expression for the slowly varying component.

Now, we note here that each pulse is represented by its intensity profile this one pulse 1, pulse 2, pulse 3, pulse 4, pulse 5 and so on. All these pulses are quickly represented with

by its intensity profile not by its carrier wave oscillation; this just simplifies the representation that is all. However, we must note that each pulse is nothing, but a localized amplitude modulated electromagnetic wave and that each pulse represents an amplitude modulated wave like the one which we have shown here. So, each pulse is actually look like it should look like this one pulse, then another pulse, then another pulse, then another pulse and like this way it should look like.

Now, it is quite clear that what is intensity profile, what is field envelope profile and what is a carrier oscillation? We should also remember that when we talk about the pulse duration, what we actually mean by pulse duration?  $I(t)$  is intensity profile, then green color is going to be field envelope function and  $E(t)$  is blue color representing the carrier wave.

So, this is carrier wave which is the average frequency, then  $a(t)$  is the field envelope and when we say that the pulse duration is  $\Delta t$  we mean the full width half max of the intensity profile. We should remember this is full width half max of intensity profile, not the full width half max of the field envelope.

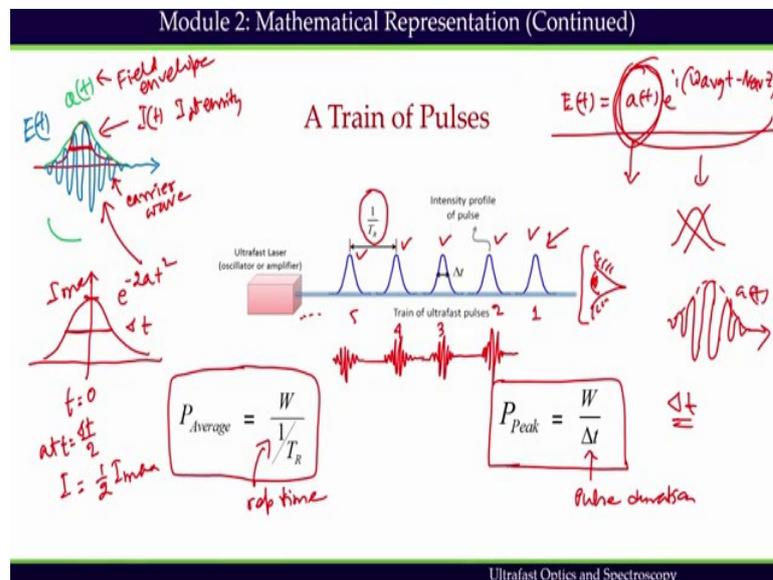
And, if we have an intensity profile like this which can be represented by  $e^{-\frac{1}{2}(\frac{t}{\Delta t})^2}$  that we have shown then it is the full width half max  $\Delta t$ . And, we remember that this is centered at  $t = 0$  which means that at  $t = \pm \frac{\Delta t}{2}$ , the intensity will drop down to half of its maximum value. So,  $\Delta t$  is related to its full width half max of the intensity profile not to the field envelope profile. We have already seen that each pulse is characterized by its temporal width  $\Delta t$  that is the full width half max of intensity profile. (Please look at the slides for mathematical expression)

In a train of pulses repetition rate how often these pulses are repeating for an observer sitting here that repetition rate decides the separation between two nearest neighbor pulses. If  $T_R$  is the repetition rate, then  $\frac{1}{T_R}$  is the repetition time which is represented by this; this temporal separation of neighboring pulses is the repetition time. Most of the ultrafast laser systems work at a repetition rate ranging from 1 Hertz to 1 mega Hertz, these are the options available right now.

If  $W$  is the energy per pulse which means the energy content for each pulse how can I get that, if I direct this pulse to a power meter, then power meter what it does? It gets an integrated intensity, which is the energy content I have in each pulse. If the total energy

content each pulse having  $W$  that can be expressed in Joule, then we can define two quantities. One is peak power that is energy which is localized within this  $\Delta t$  timescale and  $\Delta t$  is the full width half max of the intensity profile. So, within that femtosecond time scale how much energy  $I$  have concentrated? Another one is called average power; average power is energy divided by repetition time; repetition time is nothing, but  $1/T_R$  where  $T_R$  is the repetition rate. So, these are the two important terminologies for the laser system. Peak power depends on the full width half max of the each intensity profile which means that it depends on the pulse duration  $\Delta t$  and average power depends on the repetition time.

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So, let us say we have these two quantities and we often use these two quantities to define an ultrafast laser source.



milli Joule energy, power meter will give me 5 milli Joule energy per pulse each pulse containing 5 milli Joule energy.

And average power, there is an average power section here and if we see that average power we can calculate because we know P average is nothing, but W divided by 1 by repetition rate which is nothing, but for this we have for the specification we have selected it is 5 milli Joule per pulse energy divided by 1 by  $T_R$  which is nothing, but 1 by 1000 second inverse Hertz which gives me 5 Joule per second which is nothing, but 5 Watt. (Please look at the slides for mathematical expression)

So, this 5 Watt is connected to 5 milli Joule energy for a repetition rate 1 kilo Hertz. So, what does it mean? It means that in every 1 millisecond I will have 1 pulse coming out of this source and this is within 1 millisecond timescale; 1 millisecond timescale I have been able to localize the energy W amount of energy. And mostly in the experiments average power will contribute to the total thermal heating, on the other hand peak power will contribute to the non-linear effects which we see during the propagation of the ultrafast pulse through the medium.

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Module 2: Mathematical Representation (Continued)

### Limit of Temporal Duration of a Pulse

Center wavelength 800 nm

$$v = \frac{c}{\lambda} = \frac{3 \times 10^8 \text{ m/s}}{800 \times 10^{-9} \text{ m}}$$

Optical period ( $\frac{1}{v}$ ) 2.7 fs

An optical pulse cannot be shorter than 2.7 fs at 800 nm centre wavelength. In order to synthesize an attosecond pulse, we have to reduce the wavelength which reduce the optical cycle of the pulse.

$\lambda \ll \lambda_{min} \rightarrow t$

So, whatever we have seen so far is a spectra physics spitfire which can produce a pulse as short as 35 femtosecond. Many other systems can be manufactured by coherent and Game Labs and many other vendors. Perhaps, right now there are many vendors available, they can also produce similar short pulses, but one question is obvious right

now. When you say that I have a pulse centered at 800 nanometer, this is my pulse spectrum and the temporal width of this time domain representation is going to let us say 35 femtosecond. So, this is the specification.

Given the specification one question is obvious is there any limit of the shortest pulse duration which can be synthesized at a particular center wavelength? Let us say I have center wavelength 800 nanometers, when I say center wavelength it does not mean that pulse is made of one single frequency component it just suggests that center frequency component is nothing, but the average frequency component.

So, I have many other frequency components present here they all produce this constructive destructive interferences and then we get the pulse and, but we name it with the help of central frequency component we say that it is 800 nanometer pulse. Now, question is at the 800 nanometer pulse if I try to reduce the pulse duration, then I definitely need more frequency component because we know that time bandwidth product  $\Delta \nu \Delta t$  is going to be constant.

So, in order to produce short pulse I need broad spectrum, broad  $\Delta \nu$  and  $\Delta \lambda$  has to be long. Now, if I think of a situation that I have enough bandwidth to produce which will help me reduce the pulse duration. Question is can I achieve the shortest duration pulse, what would be the shortest duration pulse I can achieve at 800 nanometer?

So, in order to answer this question we have to think about the oscillatory part, at 800 nanometer center frequency I have these oscillations and there are so many oscillations present in the 35 femtosecond pulse. 35 femtosecond which means the intensity profile would be like this, the dotted line is representing the field envelope and this would represent the intensity profile and 35 femtosecond means that this width is 35 femtosecond.

Now, in 35 femtosecond there are so many oscillations presents, if I assume that I have been able to compress a pulse which means I am reducing number of oscillations and I am trying to compress a pulse and if I compress a pulse, I can assume that in the end I may have only single oscillation present. I cannot imagine a pulse which is having less than 1 oscillation, I should have at least 1 oscillation in the pulse.

Now, if I have only 1 oscillation in the pulse at 800 nanometer, then I can find out the frequency for associated with 800 nanometer and the optical period frequency can be calculated very easily  $c$  by  $\lambda$ ,  $c$  is  $3 \times 10^8$  meter per second,  $\lambda$  is 800 nanometer we can directly calculate it and then from  $\nu$  I can get the optical period. Optical period what does it mean? This time scale from here to here, what is the optical period of the oscillation, one oscillation how long does it take? And we see that at 800 nanometer center frequency the optical period is going to be 2.7 femtosecond.

What does it mean? It means that in order to sustain the shortest duration pulse which I can produce at 800 nanometer is going to be 2.7 femtosecond, it cannot be less than 2.7 femtosecond because if we make it less than 2.7 femtosecond, then we would not have one single oscillation even in the pulse which is not possible.

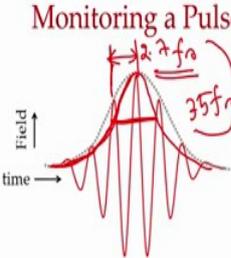
So, it is quite clear that an ultrafast pulse at 800 nanometer center wavelength cannot be shorter than 2.7 femtosecond. In order to obtain a pulse shorter than 1 femtosecond, we need to shorten the wavelength itself or optical cycle, this is why an attosecond pulse cannot be generated in near infrared or visible regime. What I need to do; if I want to reduce this pulse duration further here, what I need? I need to reduce the optical cycle.

And short optical cycle means you have  $\lambda$  which is much less than 800 nanometer, I have to reduce the  $\lambda$ . So, I have to go for the X-ray regime. So, attosecond pulse can be produced only in the extreme ultraviolet or X-ray regime of electromagnetic radiation, it cannot be produced in the visible range or the near IR because optical cycles are too long for attosecond pulses.

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Module 2: Mathematical Representation (Continued)

### Monitoring a Pulse with Electronics



Center wavelength 800 nm

$$v = \frac{c}{\lambda} = \frac{3 \times 10^8 \text{ m/s}}{800 \times 10^{-9} \text{ m}}$$

Optical period ( $\frac{1}{v}$ ) 2.7 fs

Response time of time sensitive photodetectors  
(photodiode, PMT, Power Meter): ns

They are too slow

$$V_{\text{signal}} = \int_{-\infty}^{+\infty} I(t) dt$$

Energy/pulse

We have already seen that a pulse with center wavelength 800 nanometer exhibits optical period of 2.7 femtosecond. On the other hand pulse duration is represented by the full width half max of the intensity profile. Duration of a typical ultrafast pulse which is frequently used in lab, in the lab measurements is let us say 100 femtosecond.

So, we have seen that this oscillation period is at 800 nanometer this oscillation period is 2.7 femtosecond, this is why in a 35 femtosecond pulse I have 35 femtosecond is the full width half max of intensity profile. So, in 35 femtosecond pulse I have many such oscillations present in the pulse and 2.5 femtosecond is the minimum oscillation I should have.

On the other hand the pulse duration is 35 femtosecond. So, both oscillation as well as the time by which this envelop function is changing both are too short, too fast with respect to any electronics. Within this time scale intensity profile variation occurs within this 35 femtosecond intensity variation occurs, but response time of all time sensitive detectors such as photodiode, photomultiplier tube, CCD camera etc. is mostly nanosecond.

So, if we look at this time scale and compare with the response time of electronics it is going to be nanosecond which is 10 to the power minus 9 and these are femtosecond 10 to the power minus 15 second. They are way too fast as compared to the timescale response time of electronics. Therefore, time sensitive detectors are way too slow to

measure optical cycles, optical cycles cannot be measured. As well as we cannot measure the intensity profile as well with the help of time sensitive detectors such as photomultiplier tube, CCD camera or photodiode etcetera.

Available time sensitive detectors are way too slow to measure the ultrafast intensity profile and field end oscillation. They are slow, they can only measure time integrated intensity. It is more like when you look at a rotating fan, our eye is too slow to capture every blade of the fan. And what we get? We get a blur image and that blur image mathematically will be represented by a time integrated signal. And this is nothing, but closely related to energy per pulse energy content of the pulse energy per pulse which is nothing, but time integrated intensity.

We will stop here and we will continue this module in our next lecture.