

Ultrafast Optics and Spectroscopy
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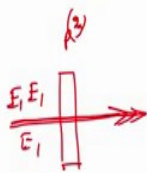
Lecture - 09

We are continuing module 2. In this module, we are discussing different non-linear effects and so far we have discussed second order process, third order process. And in the third order process, we have seen two self affecting processes.

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Module 2: Nonlinear Effects

Third Order Nonlinear Response of Dielectric Medium

Non-collinear Configuration with Three Beams: 

Four Wave Mixing (4WM) with Three Identical Beams

$$P^{(3)} \sim a_1(t) a_1^*(t) a_1(t) e^{i(a_1 t - k_1 z)}$$

$$= \underbrace{|a_1(t)|^2}_{\textcircled{I}} \underbrace{a_1(t) e^{i(a_1 t - k_1 z)}}_{\textcircled{II}}$$

Beam is affected by its own intensity (self-affecting process):

1. Self-Focusing ✓
2. Self-Phase Modulation (SPM)

$|a_1(t)|^2 \sim I_1$

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The first one is self focusing and the second one self phase modulation.

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Module 2: Nonlinear Effects

Third Order Nonlinear Response of Dielectric Medium

Self-Focusing

Three Identical Beams Propagating along the same direction collectively represent a single beam

$$P^{(3)} \sim a_1(t)a_1^*(t)a_1(t)e^{i(\omega_1 t - k_1 z)}$$

$$= |a_1(t)|^2 a_1(t) e^{i(\omega_1 t - k_1 z)}$$

(a) A lens focuses a beam using law of refraction (linear refractive index) and surface's curvature

(b) A rectangular medium can also focus a beam using non-linear refractive index: self-focusing

(3)

E_1

$E_{in} = E_1$

$= a(t) e^{i(\omega_1 t - k_1 z)}$

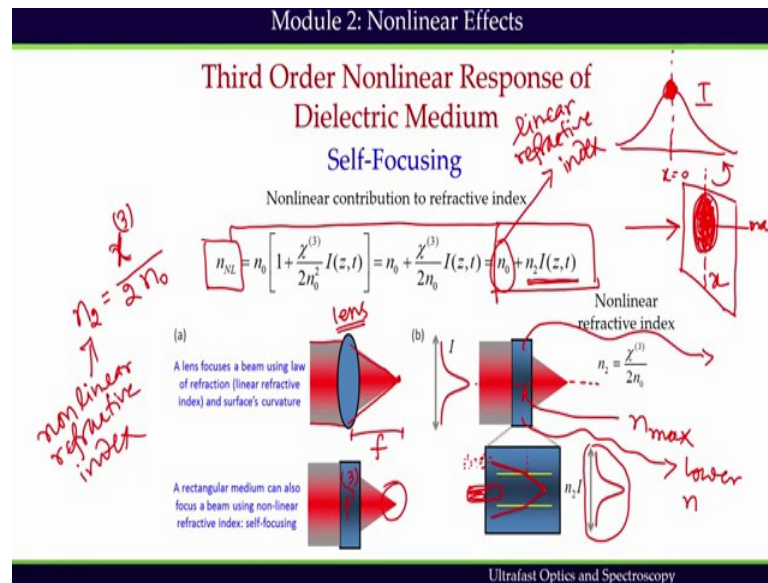
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When all three input beams are identical in this equation and if they propagate along the same direction then these three input beams can be collectively considered to be a single input beam.

So, I have this medium which expressed by this third order non-linear polarization. And we can consider as a single beam propagating along this direction in the end effectively. This input beam is expressed as which is nothing, but E_1 which is expressed as a t envelope function e to the power $i \omega_1 t - k_1 z$. So, that is the very familiar expression for an ultrafast pulse, which is having an envelope function and the carrier wave component with center frequency ω_1 .

So, this non-linear polarization expression suggests that, the beam we get back the beam E_1 , but it is affected by its own intensity a_1 . When pulses own intensity forces it is spontaneous focusing it is called self focusing.

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We all know that a lens is a transmissive optical device that forces a light beam to focus. So, this is a lens and it focuses the beam at the focal point. So, this is called focal length. Similar to the lens a medium having third order non-linear polarization, a rectangular non-linear medium can also behave like a lens and can focus a light beam.

This occurs through the non-linear contribution to the refractive index. Generally every medium has its own refractive index that is n_0 . But the non-linear expression for refractive index is this, which depends on the intensity, n_0 plus n_2 multiplied by its own intensity I . So, it is intensity dependent refractive index. Which means that if I take on beam, we have not discussed this beam the transverse electromagnetic mode of the beam. When the beam is propagating along this direction and if I place one piece of paper then we see a circular spot and that circular spot is called transverse electromagnetic mode. (Please look at the slides for mathematical expressions)

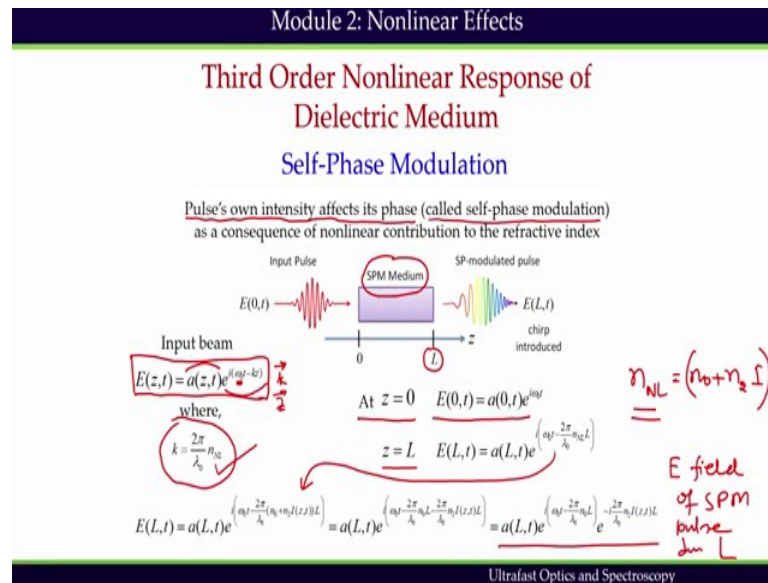
And if we take a cross section of that spot, then what we get a Gaussian distribution of the intensity. This is not the Gaussian pulse in time domain, this is the Gaussian distribution of the intensity is a spatial distribution. So, this is your x with respect to. So, this is the axis with respect to this axis we see that in the middle of the beam. So, this is the cross section. If the beam is propagating along this way and then if I place one piece of paper, then I get this cross section and the middle of the beam intensity would be maximum, this is the maximum intensity.

And that is why and we have taken a cross section like this and we are representing this one, this is my x axis let us say. So, middle of the beam is maximum which suggests that I will have this non-linear refractive index maximum here, maximum refractive index and the wings will have lower refractive index. This intensity dependent refractive index is called optical Kerr effect. This is a direct consequence of non-linear contribution to the refractive index which depends on intensity.

Here, n_2 is the non-linear refractive index which is expressed by χ^3 third order susceptibility divided by $2 n_0$; n_0 is the linear refractive index this is linear refractive index and n_2 is non-linear. So, because of these non-linear contributions to the refractive index, we get self focusing of the beam. How does it happen? It is shown here in this figure let us say, the beam is trying to come from lower refractive index regime to the higher refractive index regime. (Please look at the slides for mathematical expressions)

And it will definitely bend and due to this bending it will start focusing the beam. And we know that this regime would be higher refractive index because the intensity is high here. And this regime would be lower refractive index because intensity is low here because of this Gaussian distribution of the intensity of a Gaussian beam. So, for a Gaussian laser beam the spatial intensity profile results in spatial variation of refractive index which is higher in the center than at the wings. This variation in refractive index acts as a lens, self-focusing lens and the beam converges and eventually gets tightly focused.

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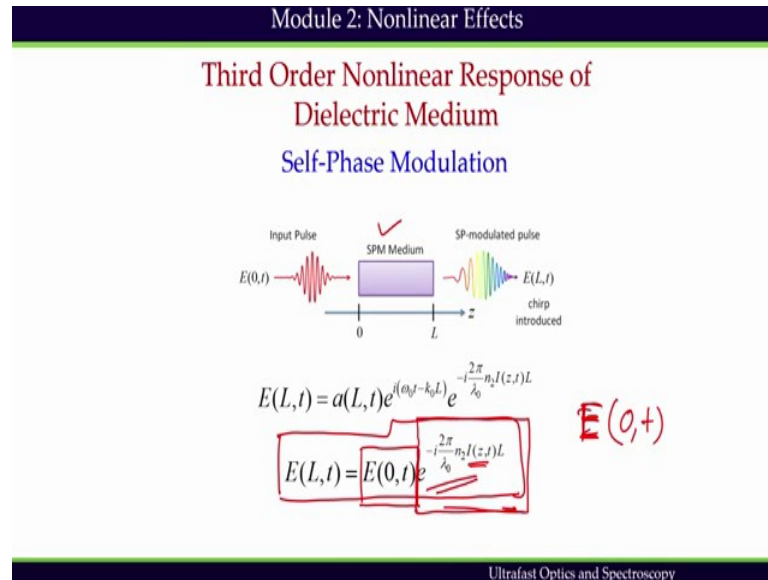
So, this is called self focusing. On the other hand the intensity of the pulse also affects its phase and that is why it is called self-phase modulation. Intensity dependent refractive index imposes an additional phase shift to the pulse envelope during the propagation. We will find out how this kind of additional phase can be introduced to the pulse very soon. To visualize the effect of the self phase modulation, we will first express the electric field of an incident pulse propagating in z direction as shown in slide.

This is again the very familiar expression for a pulse. This is envelope function, this is carrier wave part we center frequency ω_0 along propagating along k direction which is z direction here. We assume that, we have a non-linear medium that is self phase modulation medium with thickness L and we have to express k in terms of its non-linear refractive index. So, at z equals 0, when the pulse is just entering the medium I can express electric field as this where z equals 0 that is why kz term is gone from this equation. And at z equals L distance I can write down $\omega_0 t - \frac{2\pi}{\lambda_0} n_{\text{eff}} L - \frac{2\pi}{\lambda_0} n_2 I(z,t) L$. (Please look at the slides for mathematical expressions)

This k has been replaced by this non-linear refractive index. So, if we express like this way, then we know that non-linear refractive index is nothing but $n_0 + n_2 I$ intensity dependent refractive index and that is exactly what we have inserted here. Then little bit of rearrangement we get the final expression for the electric field. This electric

field is E field of Self Phase Modulated pulse due to propagation of L distance in the medium.

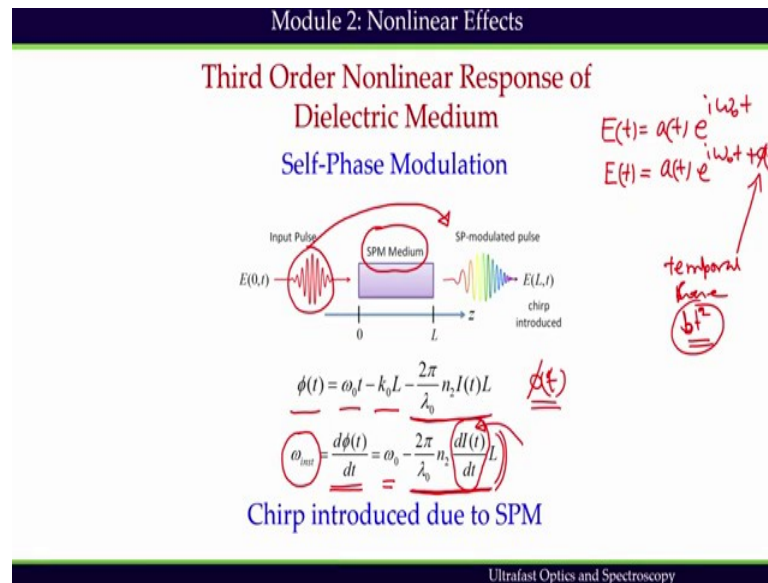
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Once we get the electric field, then we can write down that this expression as this where $E(0, t)$ is the field without the medium. If we did not have the medium then the pulse would have propagated L distance. And due to the propagation the field should have been like this, but because the medium is present here, we have introduced additional phase, complex phase in the medium.

This mathematical expression for self phase modulated pulse after propagating L distance in third order non-linear medium. Suggests that the time dependence of the additional temporal phase more specifically the complex temporal phase, complex because it is in the complex notation due to self phase modulation comes from the time dependent intensity, this part. The induced phase changes the behavior of the pulse.

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Now, we can remind ourselves in module 2, we have seen quadratic temporal phase. So, our transform limited pulse was expressed as $E(t)$. And we said that if it is a chirp pulse due to second order non-linear, second order temporal phase as shown in slide, the additional temporal phase. (Please look at the slides for mathematical expressions)

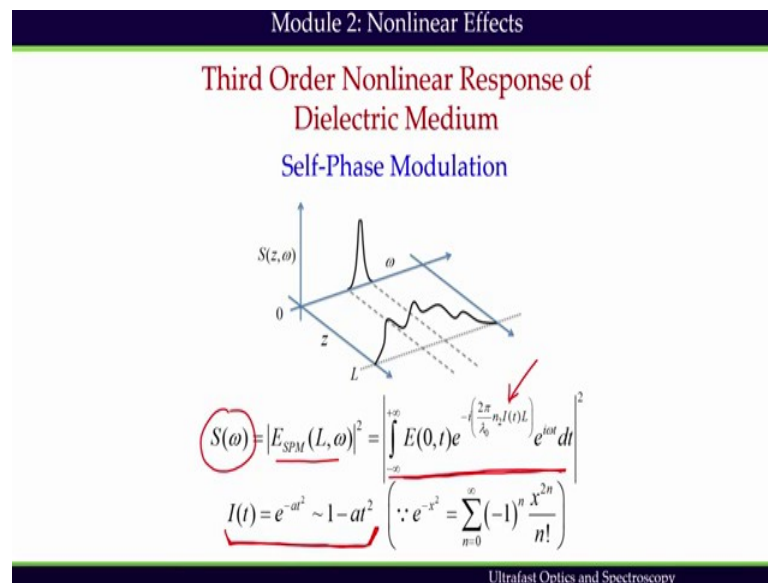
This is called temporal phase. And we have take an example of bt square at some point in module 2. And we have seen that that represents quadratic temporal phase which gives chirp in the pulse. Similarly, here total phase if we look at the total phase ϕ total phase which is obtained for the self phase modulated pulse. We get $\omega_0 t - k_0 L$ minus this expression which means I have now additional phase temporal phase, which is also called complex temporal phase. (Please look at the slides for mathematical expressions)

And whenever we introduce complex temporal phase to a pulse, we have chirp. In this case chirp is very complicated and the way we realize chirp by considering instantaneous omega. Instantaneous omega if it is varying with respect to time, in module 2 we have studied then it is chirp. So, if we get the instantaneous omega which is nothing, but the first derivative with respect to t we get ω_0 minus this expression and this has time dependent part $I(t)$. (Please look at the slides for mathematical expressions)

Time dependent part in the temporal phase is coming due to the intensity of the pulse itself. This suggests that if I have self phase modulation in the medium then definitely I

am going to introduce chirp into the input beam. And chirp is not as simple as bt square, it is extremely complicated chirp depending on the intensity. And whenever we are introducing chirp, we know that the pulse is going to elongate in time. (Please look at the slides for mathematical expressions)

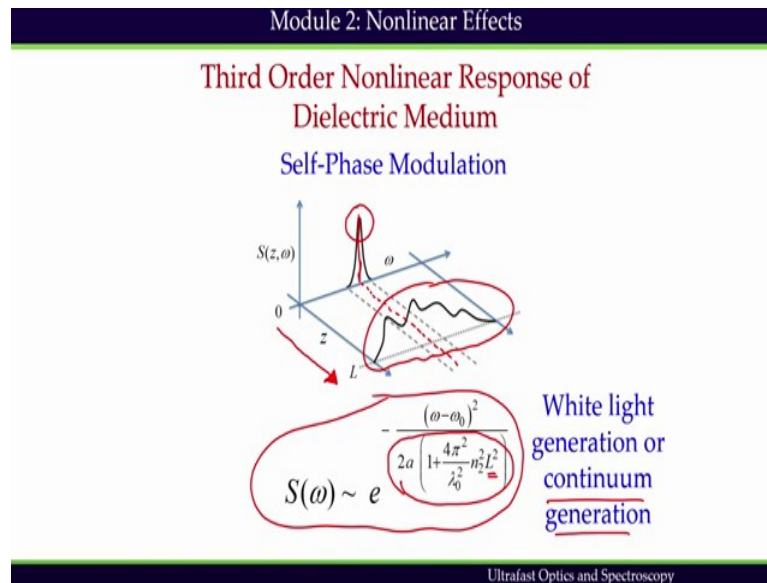
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Now, we look at the frequency domain expression. We have already seen the time domain expression and we know how to convert to frequency domain. The corresponding spectrum of the output pulse which is self phase modulated pulse, is expressed as square modulus of the Fourier transform of the time domain field. And this is the spectrum.

We can assume a Gaussian intensity profile for this $I(t)$. And in that case, we can write down with the first order approximation this is a very simple approximation we are making to get an analytical solution, otherwise we have to use numerical solution. But to get analytical solution, we have this $I(t)$ expressed in exponential series up to first order.

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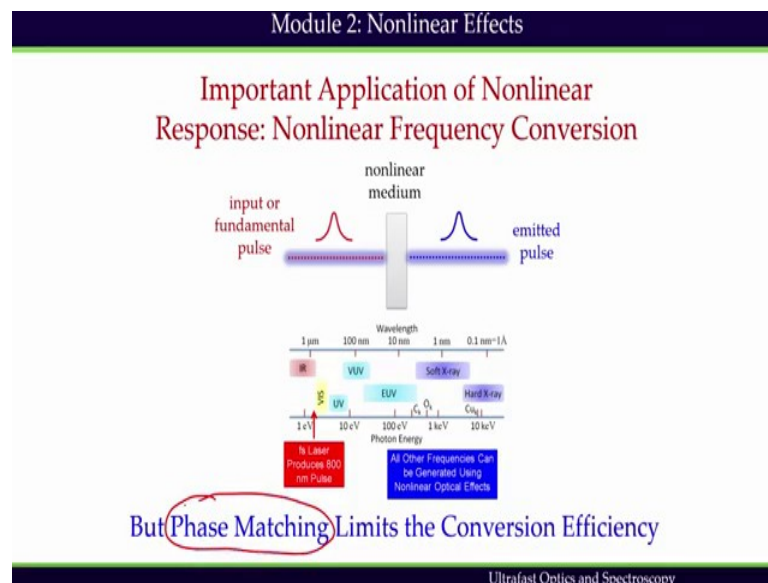


And if we do that then with the help of a little bit of math one can show that, the spectrum expression for the spectrum of self phase modulated spectrum, looks like this. And the term given here suggests that, it will always spread as a function of L which is shown in this figure. So, this is the thickness of the crystal L distance I started with a transform limited pulse, a Gaussian nice looking spectrum we had. As it is propagating along z direction L direction we are seeing that new frequency components are created and the pulse is broadening.

So, this equation clearly shows as a spectrum will broaden as a function of the crystal thickness L which is depicted in this figure. New frequency components are generated due to self-phase modulation. Extra frequency components brought in the spectrum on both side of the center wavelength. So, here the center wavelength was at this point and it is broadening on the both sides of the center wavelength. The broad spectrum generated in this manner is called continuum generation or white light generation.

If an ultrafast pulse centered at 800 nanometer is focused into transparent non-linear material which can potentially exhibit self phase modulation the emitted pulse will appear as a pulse of white light. And it will be broad as well. Broadening is appearing because of the introduced temporal phase. Most common medium which is used for white light generation is sapphire crystal.

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So, thus far all the examples of second order third order processes which we have discussed thus far, suggests that non-linear effects are very interesting and can be very useful pathway to achieve frequency conversion. In non-linear frequency conversion process; however, we have to remember that, one limiting factor is phase matching.