**Ultrafast Optics and Spectroscopy**
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**Lecture - 08**

Welcome to module 3, of the course "Ultrafast Optics and Spectroscopy".

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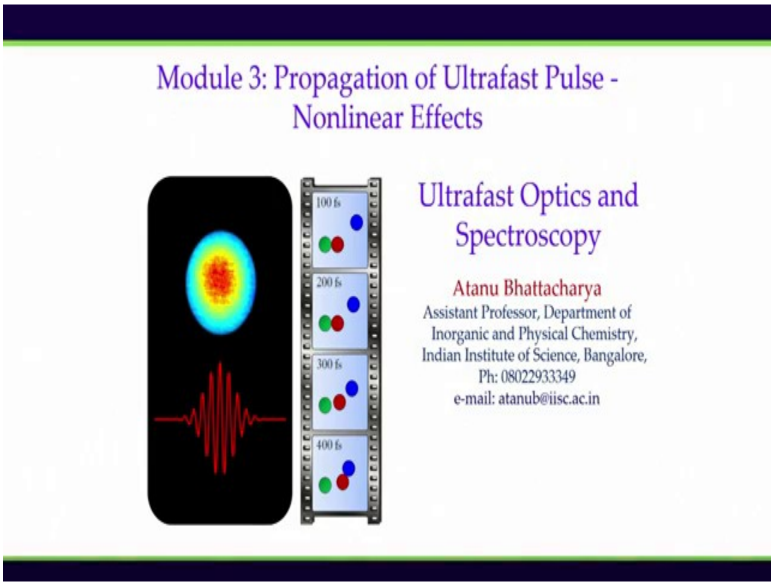


In this module, we will learn different non-linear effects due to propagation of ultrafast pulse, primarily through a dielectric medium.

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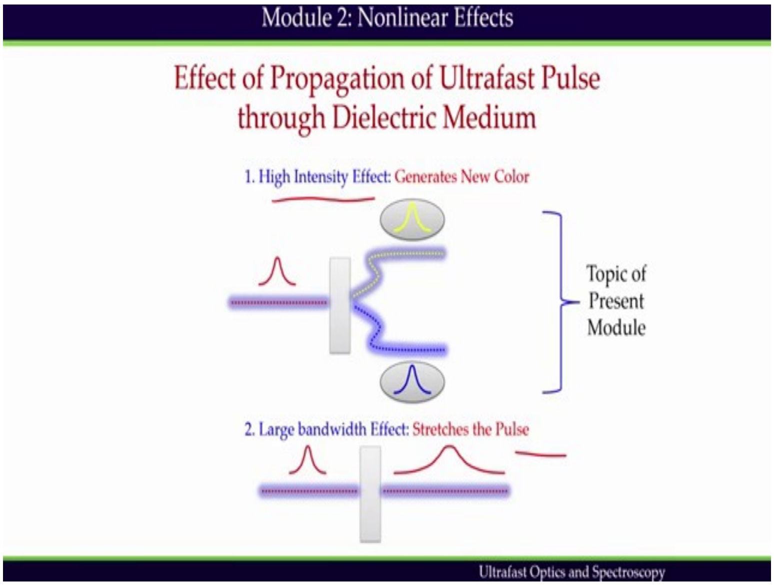


Ultrafast pulse is a propagating electromagnetic wave, which is synthesized by the interference of many plane waves with different colors with a stable phase relationship.

In all ultrafast spectroscopy experiments, ultrafast pulse travels through a medium such as lens, polarizer, wave plate, non-linear crystals. Therefore, it is quite instructive that we study the propagation of an ultrafast pulse in an optical medium. In particular, we must consider the propagation of ultrafast pulse through dielectric medium, because most of the optics such as mirrors, lens, wave plates, polarizer are made of dielectric medium. Primarily, two effects are absorbed; high intensity effect and dispersion effect. The high intensity effect gives birth to the non-linear optical effects, which is the theme of the present module.

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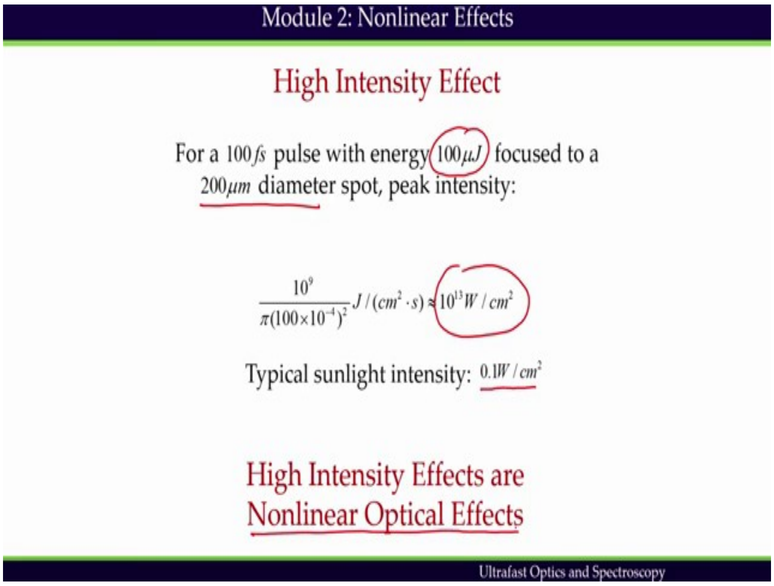


But question is how intense are these ultrafast pulse is? To get a practical feel for it. Let us consider a typical 100 femtosecond ultrafast pulse with 100 micro joule energy. 100 micro joule energy is typically used in many ultrafast experiments. The peak power of this pulse is estimated to be 10 to the power 9 joule per second, which can be converted to peak intensity. If it is focused to 200 micron diameter spot size, then we get the peak intensity 10 to the power 13 watt per centimeter square.

These calculations can be done very quickly. So, what does it suggest? We can now, compare this peak intensity with typical sunlight intensity, which is 0.1 watt per centimeter square, this shows that femtosecond laser pulses are 10 to the power 14 times intense than sunlight intensity. Due to this extremely high peaked intensity ultrafast pulses exhibit high intensity effects, which is called non-linear optical effects.

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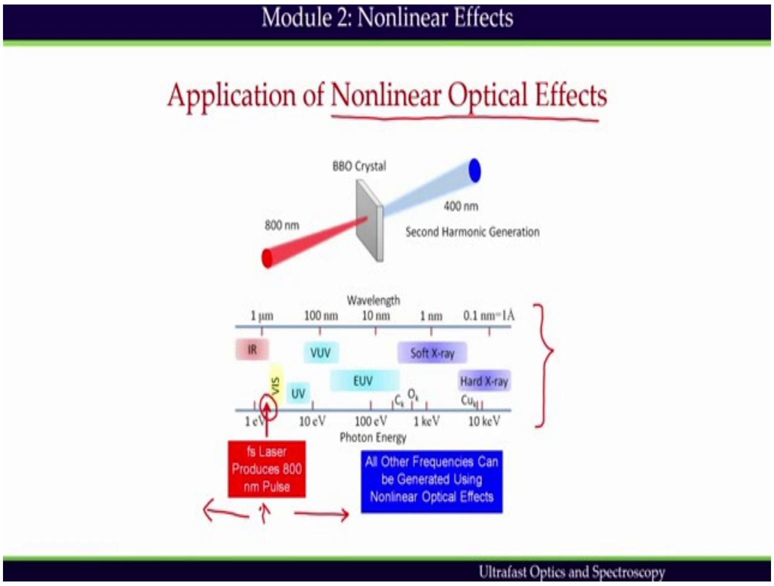


Most of the currently available femtosecond laser oscillators or amplifiers produce pulses with center wavelength, near 800 nanometer in these regime. This is the entire electromagnetic spectrum we have shown and light with 800 nanometer center wavelength light can be converted to any frequency both in IR and in the X-ray or UV regime with the help of this non-linear optical effect and that is why non-linear, non-linear optical effects are very important and it has enormous application in frequency conversion.

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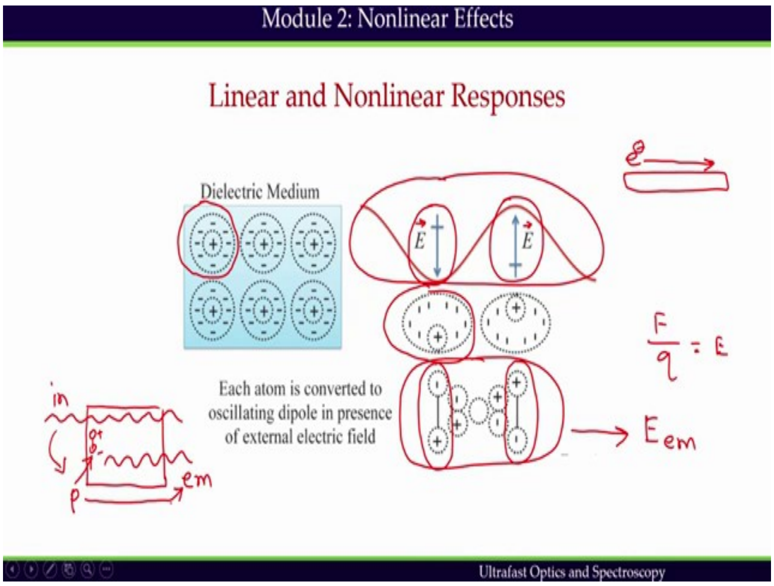

Therefore, it is quite instructive that we understand non-linear effects particularly in dielectric medium, because most of the frequency conversions are done in dielectric medium. A dielectric medium is an electrical insulator that can be polarized by an applied electric field.

According to the classical model, a dielectric medium is made of atoms, it does not have free electron. So, if I have a metal then I can have electron, which is freely moving. And in a dielectric medium, we do not have free electron all the electrons for an example here, are bound to the positive ion core. So, each atom consists of a cloud of negative electrons, negative charge bound to a positive charge at, its center as depicted here.

There is no freely moving conducting electron in dielectric medium in the presence of an electric field. Now, what happens to this electron cloud, the negative cloud is distorted just like this. So, if I have a field acting like this which will be oscillating in every half cycle, it is exchanging the polarity. This is the polarity change of the electric field and due to this electric field, electric field is nothing, but force per unit charge which is field due to this force this negative cloud is distorted and when electric field is reversed, every half cycle polarization of the dipole is also reversed.

So, for an example here dipole looks like this negative positive here, but here is the polarity has reversed. As oscillating dipole is a source of electromagnetic radiation mediums induced polarization, creates another emitted field. So, as this electric field; input electric field oscillates over the time, the polarization or the dipole, induce dipole is also oscillating and any oscillatory dipole is the source of another electromagnetic field that is called E emitted.

Thus, effect of electric field on dielectric medium can be briefed as follows through two step. Step one, the light electric field converts each atom in the medium into oscillating dipole and step two, the dipole oscillation then emits an additional electric field at the same frequency of the oscillating dipole. So, what happens in brief one can say like this, I have a medium here in this medium electromagnetic wave is propagating, we will take an example of plane wave right now, but the same is true for the pulse. When it is propagating here, we are creating dipole and again this dipole is also oscillating. These dipoles collectively are described by the polarization of the medium.

So, input beam is creating a polarization this polarization, because it is auto oscillatory dipole, it is giving birth to another radiation that is called emitted radiation. So, there are two steps input beam is polarizing the medium first and the medium polarization will again emit another electromagnetic field.

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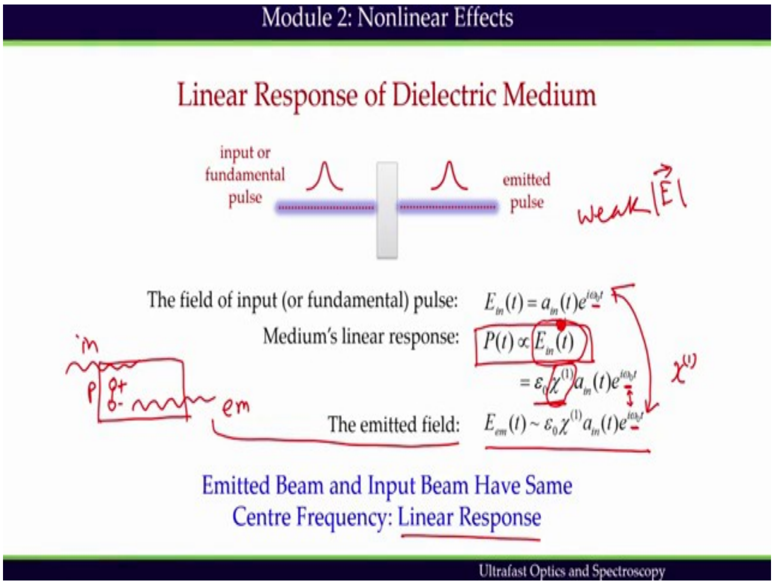


At low external electric field strength in which the field is smaller than the field which binds the electrons to the ion cores, the input beam or sometimes it is also called fundamental radiation, acts as a small perturbation to the system medium.

This produces a polarization that is directly proportional to the electric field strength of the input light. So, this direct proportionality depends when I have very weak E, when the field strength is very weak; then it is linear with respect to the induced polarization is linear with respect to the input electric field, which can be expressed by this proportionality constant and this is the proportionality constant. Epsilon naught is free space permittivity and chi[χ] with 1 within bracket suggests that it is linear susceptibility. (Please look at the slides for mathematical expressions)

The susceptibility chi is a measure of how easily the dielectric medium can be polarized in response to an input electric field. As stated earlier, the induced polarization creates another field which is emitted from the medium. So, there is a two step process; input beam creates a polarization in the medium that polarization oscillates and that is why this polarization now creates another beam that is called emitted beam. This is input beam and this is polarization.

So, this emitted field can be also approximately written as this where we can see that the induced polarization frequency omega naught[$\omega_0$] is same as emitted beam frequency, which means that the emitted field oscillates at the same frequency of the input field. This is called linear response of the medium, which means in linear response of the medium, frequency does not change, frequency remains to be the same. Input beam and outgoing beam frequencies of the same and that is why it is linear response, linear with respect to the polarization depends linearly to the field. Most of the absorption spectroscopy is performed in this regime only.

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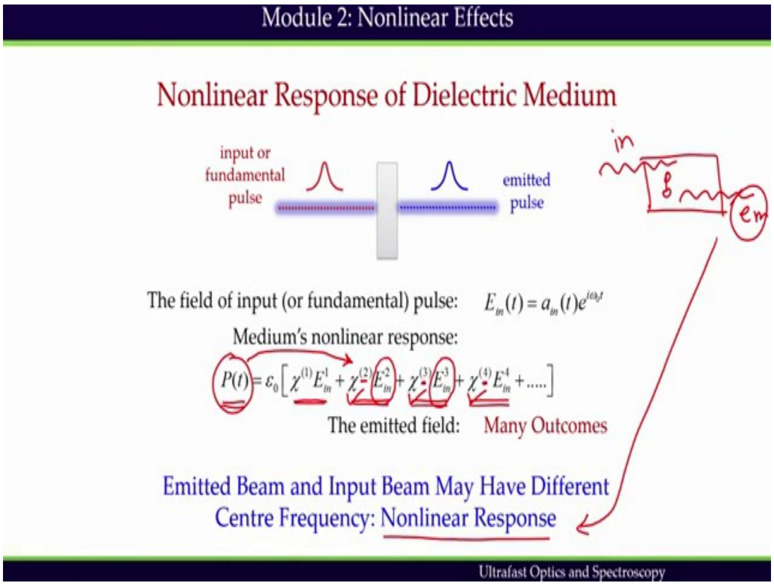


But just think about when the field strength is very-very high, which means that when radiation field is comparable to the atomic field strength, which binds the nuclei and electron.

The relation between polarization and the input radiation field is no longer a linear one. The general relationship between the polarized polarization and the input electric field, then can be expressed with the help of this Taylor series expansion, where we have many terms.

This is induced polarization at very high field strength where I have linear term, second order term, third order term, fourth order term. And what you see is that second order, because it is square of the input electric field, third order, because it is cube of this input electric field and this numbers within the bracket it suggests the order.

So, the susceptibility second order susceptibility is this one, third order susceptibility is this one, fourth order susceptibility is this one. Each polarization term results in different outcomes and we will closely look at a few outcomes here. So, this is called non-linear response. When it is a non-linear, it means that induced polarization has to be non-linear with respect to input electric field. Again, I remind this picture two step model; I have input beam. This input beam is creating polarization in the medium. This polarization now, is creating emitted beam and what kind of emission I can have under non-linear response that is going to be the topic of the next slide. (Please look at the slides for mathematical expressions)

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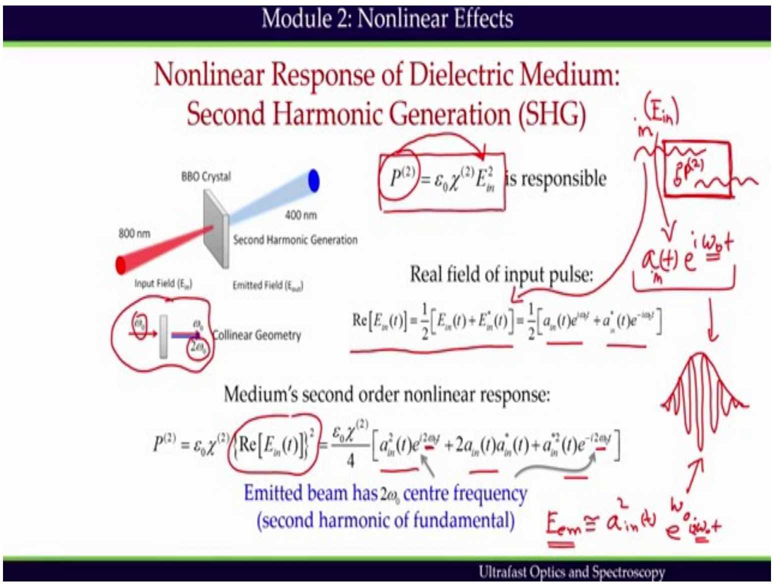


We will begin with second order polarization. Second order polarization, we have taken only this term from Taylor series expansion. Second order, because induced polarization depends on E square of the input beam that is second order. In collinear geometry as depicted here, the real field of an input pulse, in the time domain can be written as this, which means that I have this medium and I have input beam and real part of this input electric field is expressed as this.

So, this electric field we know, we can express this electric field as a(t), which is envelope function e to the power i omega naught t here, omega naught is the center frequency of the pulse. How does this pulse look like? I have an envelope function like this and then I have this carrier wave oscillation this wave has a frequency omega naught, which is the resultant frequency which you get after the interference of all the frequency component present in the pulse. (Please look at the slides for mathematical expressions)

So, that is an expression which is very familiar and this is the complex conjugate of that. So, mediums second order non-linear response can be calculated from this equation, where we take square of this real part and we get this terms. There are three terms we get. This equation, suggest that second order non-linear process creates polarization that oscillates at 2 omega naught frequency; 2 omega naught frequency.

As the emitted field always oscillates at the same frequency of the induced polarization, the second order induced polarization generates another light at 2 omega naught frequency that is why we can write down E field of emitted field. So, it has created a polarization and this polarization is now, this is a second order polarization. This polarization has now created emitted beam and this emitted beam will also oscillate at the same frequency.

So, we can get write down emitted field as a in square t e to the power i 2 omega naught t, which means that the center frequency of the emitted field is going to be 2 omega naught. This process is called second harmonic generation, because it is the 2 omega naught, which we are creating with respect to the fundamental beam input beam. (Please look at the slides for mathematical expressions)

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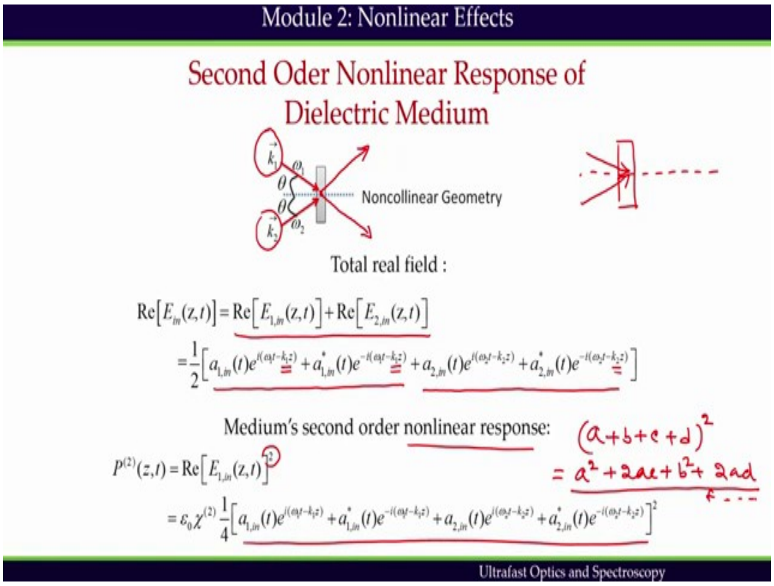


Now, we will discuss second order process in non collinear geometry, where two beams are coming at an angle with respect to this axis to induce second order polarization

This is non collinear geometry, the real resultant field is always additive, fields are always additive that we know. So, at this point where the interaction regime, at this point field is additive and we take the real field. We add them together; this is the first field, this is the second and complex conjugate, this is the second field and complex conjugate.

In the end the second order non-linear response, we get as a square term and we get this equation which is nothing, but a plus b plus c plus d whole square, which will have many terms among them, we will analyze a square term plus 2 ac term plus b square term plus 2 ad term. There are many other terms, but these are the terms which would be very much interesting to analyze here. We note that we have given wave vector as well in the beam and k 1 and k 2, they are the wave vectors for these two beams, which means that the direction of this beam is k 1 along this way and direction of this beam is k 2, along this way, directions are important. (Please look at the slides for mathematical expressions)

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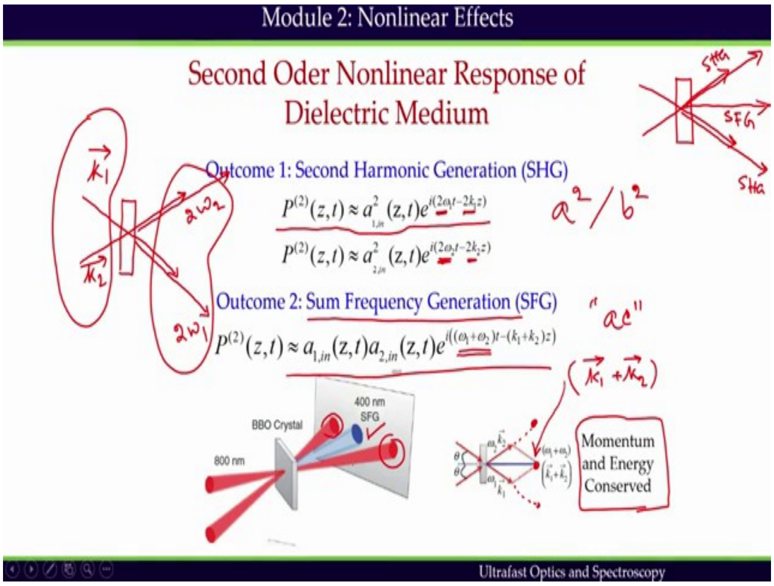


So, first thing we will look at one of the terms which we described previously, the square term and when you look at the square term we see that that is a square term or b square term. The square terms, these polarizations create new emitted pulse at 2 omega 1 and 2 omega 2 center frequencies, this is called second harmonic generation.

So, when I have non collinear configuration two beam interacting like this, this is my k 1, this is my k 2 direction, then it does not change the direction. This polarization directions are the same. So, along this direction only, we will create 2 omega 1 second harmonic, along this direction only will create 2 omega 2 second harmonics. So, each beam each input beam will create second harmonics along the same direction. Next one, we will look at some frequency generation. Some frequency generation is, because of the ac term, which we discussed earlier. (Please look at the slides for mathematical expressions)

Here, polarization is expressed by this equation. This polarization gives birth to the sum frequency generation, a new pulse at central frequency omega 1 plus omega 2, at the center frequency and the direction is going to be very interesting to note. Direction is going to be k 1 plus k 2, along this direction and this direction is represented by this arrow, the blue arrow, which suggests that momentum as well as energy is conserved, when we are undergoing non-linear conversion, photon conversion or the frequency conversion. It is interesting to note that when we have non collinear configuration this is input beam, this is another input beam. Along this beam along this direction only we get another second harmonic generation, along this direction; we get another second harmonic generation.

But some frequency generation is created in between these two beams, some frequency generation and that is why we will be able to identify some frequency generation beam in the middle of two input beams.

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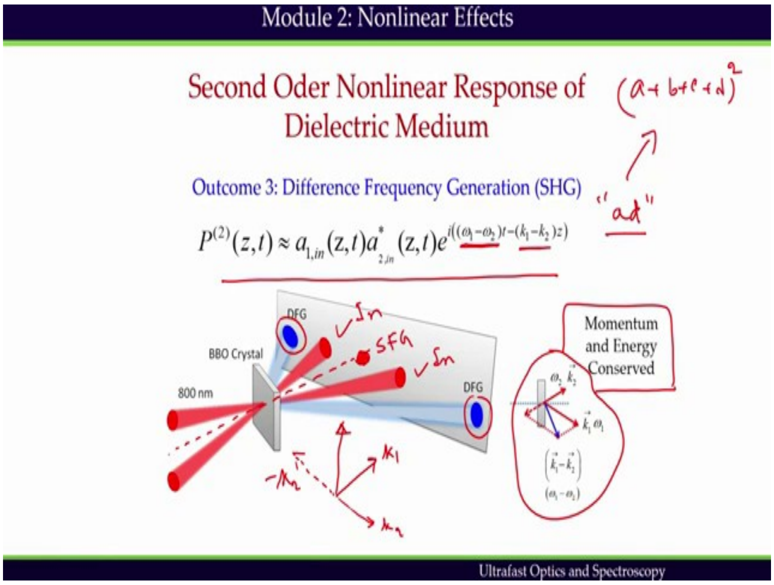


Now, we will talk about different frequency generation. The term which is responsible for this is ad term which we obtained from that equation we have written a plus b plus c plus d whole square and the polarization is given by this. This induced polarization results in different frequency generation. This new pulse is created at omega 1 minus omega 2 that is wise difference frequency generation and it is generated in the direction of k1 minus k2, k1 minus k2 direction. (Please look at the slides for mathematical expressions)

I had k1 direction along this way, k2 direction was this way, this is k1, this is k2, but now k2 has to be negative. So, I will write it like this way, this is minus k2 then add them together, it is along this direction which means that momentum and energy is conserved when we are doing non-linear frequency conversion. So, the position difference frequency generation positions are going to be far away from the input beam.

So, we have created some frequency generation, but different frequency generation is created here and here with respect to the input beam these are the input beams. So, the directions can be identified very easily, if we go over this quick Math.

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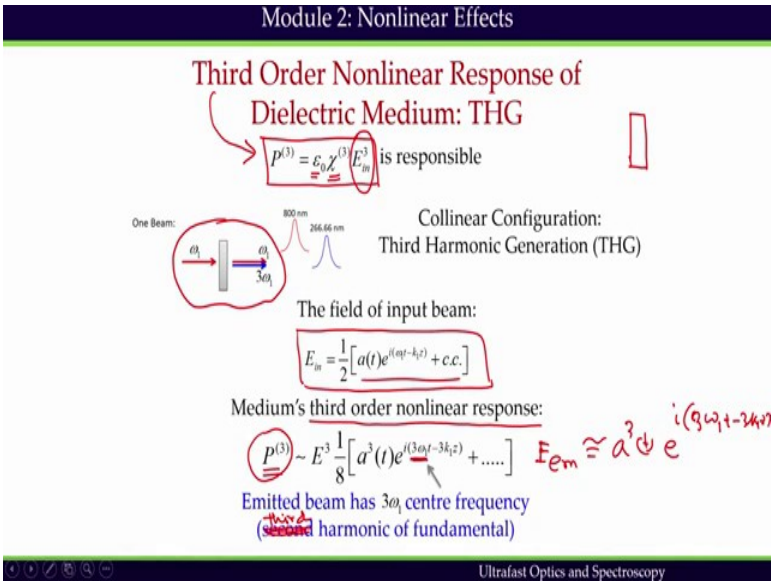


So, so far we have discussed different second order processes, including second harmonic generation some frequency generation and difference frequency generation. Similar discussion on the third order process also deserves some attention. If a single input beam is used just like collinear configuration, then the real field of the input beam is expressed by and we can remember, remind ourselves that third order process is controlled by this polarization, third order polarization where, it depends on E to the power cube that is the input beam to the power input beam electric field and this is third order susceptibility and this is free space permittivity. So, the field we have expressed as this is quite familiar expression.

Now, field in time domain and, because of that medium third order response will be described by the third order polarization, which is nothing, but cube of this function of this function and if we take the cube of the function one of the terms we get is this term, where I have 3 omega 1 as the center frequency. Which means that this polarization is going to create another emitted beam and this emitted, beam electric field will have the center frequency as 3 omega1 t minus 3 k1 z and that is why it is third harmonic generation. This is wrong, this is not second harmonic generation, this should be third harmonic generation.

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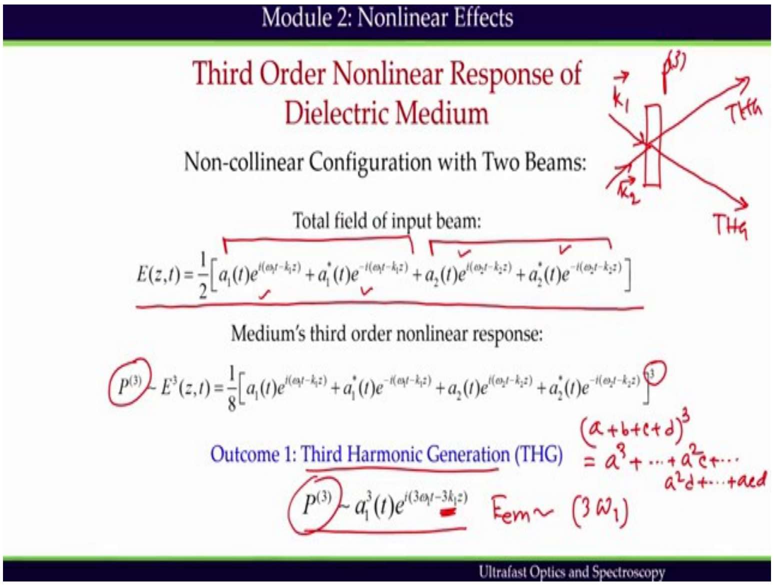


Now, think about non collinear configuration, I have two beams k 1 along this way, k 2 along this if there are two beams and we are considering third order process in this non collinear configuration again resultant electric field, the total electric field is going to be summation, fields are additive. The first field real complex conjugate, second field real complex conjugate, and third order response is nothing, but cube of this total field.

Now, we have an equation like a plus b plus c plus d whole cube, which will have many terms, but among them we are interested in a cube, then a square c, then a square d, then acd. These four terms are interesting and this is why we will go over these four terms. (Please look at the slides for mathematical expressions)

The first term is nothing, but third harmonic generation. So, each beam will create the third harmonics along the same direction. So, this polarization will create another emitted beam and that will have center frequency 3 omega 1 along the direction of k 1. So, along this direction we will have one third harmonic generation. Along this direction also, we will have one third harmonic generation.(Please look at the slides for mathematical expressions)

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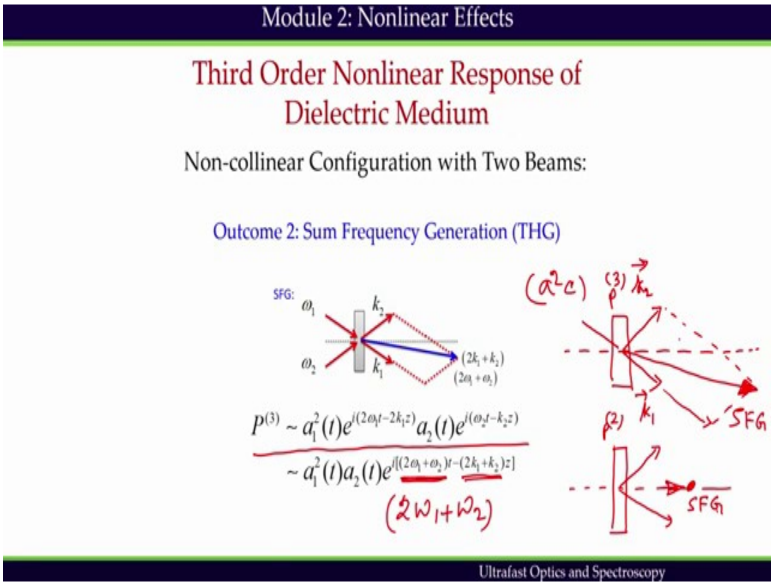


Now, look at some frequency generation. Some frequency generation can be done in a second order medium and also in third order medium. If it is third order, then we have a square c term, a square c and that is why polarization can be written like this way, where a new frequency is created and the new frequency, which is created, the pulse which is created having center frequency 2 omega 1 plus omega 2 along the direction of 2 k 1 plus k 2.

How do we identify, this direction this is also following momentum conservation, I have k1, I have k, this is k1 direction, this is k2 direction, but now I have to take 2 k1, which is this much. So, if this is the axis we see that the some frequency generation, if it is a third order process is going to be off the axis. (Please look at the slides for mathematical expressions)

This is due to third order polarization. Here, we can remind our self that if it is a second order polarization, then it was, some frequency generation was on the axis. In the middle of these two beams and here, we have got some frequency generation, but if it is third order process then some frequency generation is of the axis, but it is still in between these two beams, input beams.

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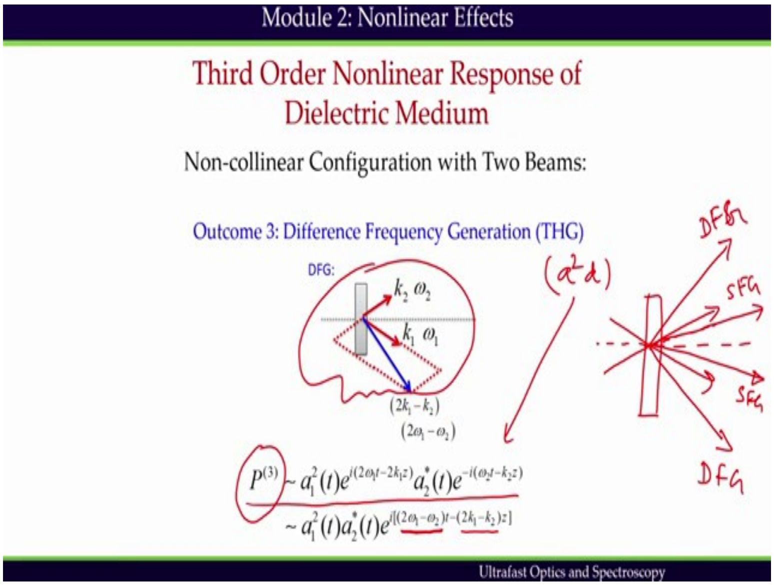


Then we look at differenct frequency generation. The term which is responsible for this is a squared d and that is why we get polarization and if we simplify it, we see that a pulse would be created.

This polarization will create another emitted beam and the beam will have center frequency at 2 omega 1 minus omega 2, along the direction 2 k1 minus k2, 2 k1 minus k2 direction is shown here. Again, we are conserving energy and the momentum; this is called difference frequency generation. (Please look at the slides for mathematical expressions)

So, different frequency generation again it is actually out of this two beam. So, I have and that was true for second order process as well. So, I have input beam these two input beams create difference frequency generation will be along this direction. Another difference frequency generation will be along this direction whereas; some frequency generation is of the axis along this direction and also along this direction. So, these are quite similar to second order process.

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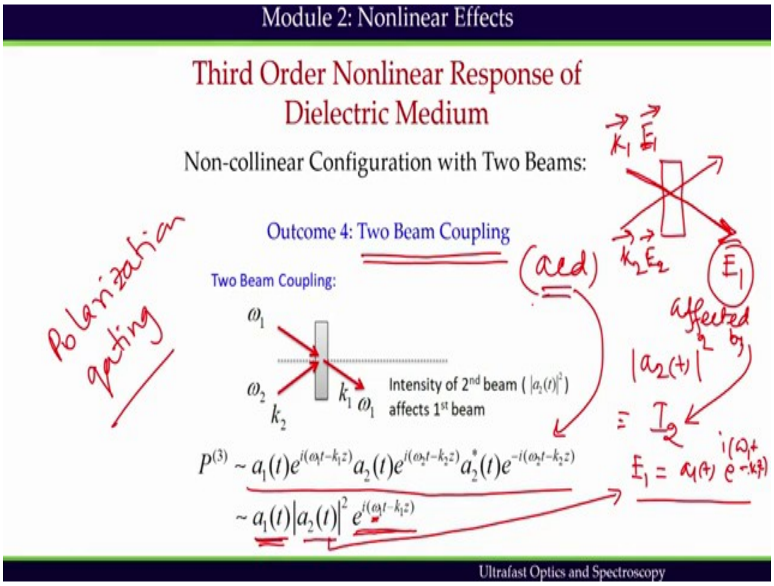


But this is the two beam coupling, this is the fourth one and the term, which is responsible for the for this two beam coupling is acd and accordingly, we get the polarization written like this or in other words, it is a1 (t) a2(t) square modulus of a2(t) e to the power i omega1 t minus k1 z.

So, I have a beam, I have k1 and I have k2, this is k1, this is E1 that is the first beam and then this is the second beam E2. Now, what is going on here, this term a2(t) square modulus of a2( t ) is nothing, but intensity of 2, second beam intensity and the first beam remains to be the same a1. (Please look at the slides for mathematical expressions)

So, the first beam is expressed as E1 equals a1(t) e to the power i omega1 t minus k1 z. So, this is the first beam. It is suggesting that I have still the first beam E1, but it is affected by the intensity of the second beam. This third order non-linear polarization yields emitted pulse that has the same frequency omega1 and the direction as the first beam, but the emitted pulse is affected by the second beams intensity. So, second beam effects the first beam without changing center frequency and direction. (Please look at the slides for mathematical expressions)

This third order process is called beam coupling, these two beams are getting coupled with each other, but their frequency and the direction are the same and implementation of this technique is called polarization getting polarization getting. Polarization getting is a third order process and based on this acd term, which is called two beam coupling.

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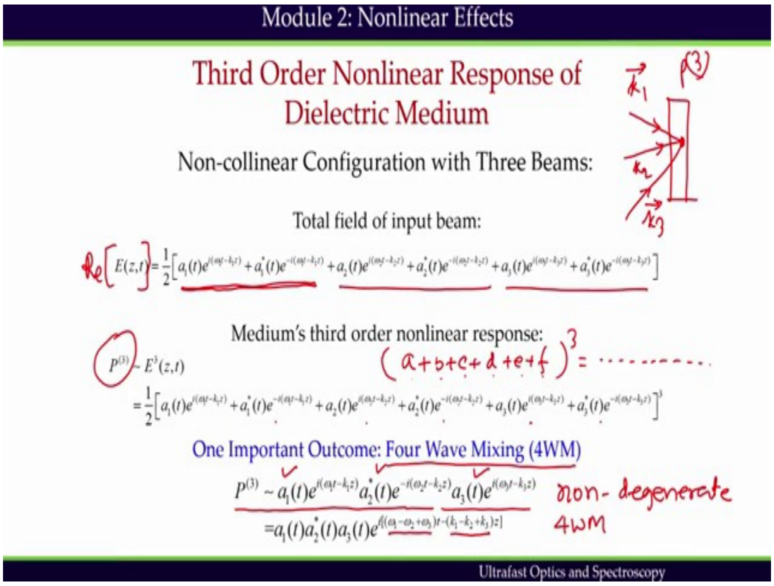


Now, we will think about three beams; I have three beams, let us say I have this P3 non-linear third order non-linear medium and I have three beams; k1 k2 and k3. So, the first beam resultant field is additive again at this point, interaction point.

So, I have first beam, second beam and third beam. We have taken the real part of the resultant field and that is why third order polarization is nothing, but cube of this function. So, we will get many terms here.

So, one of them would be very interesting and it is called four wave mixing. The four wave mixing is given by the polarization written here and effectively, if we reduce this, we get omega1 minus omega2 plus omega3 along the direction k1 minus k2 plus k3. Now, this four wave mixing, this four wave mixing is called non degenerate four wave mixing, non degenerate, because it is called non degenerate four wave mixing, because I have three beams, three different beams, but what about if we get the same beam.

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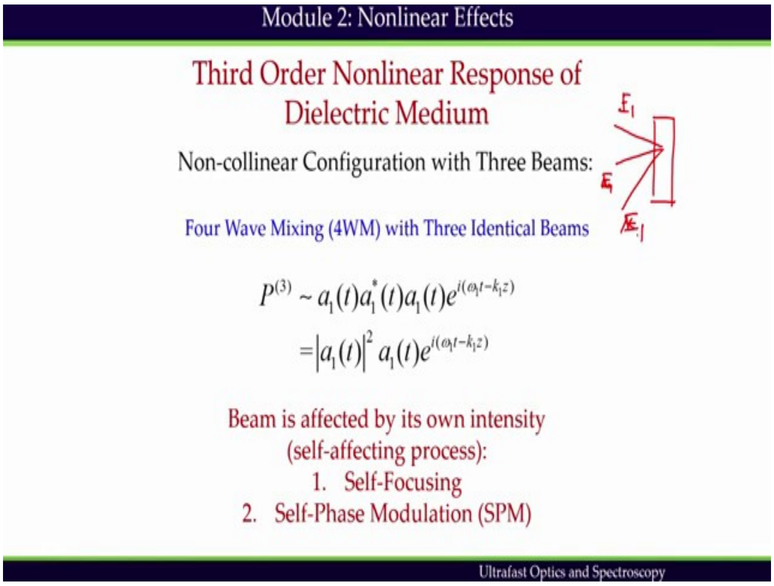


A very interesting variant of four wave mixing is observed if we take identical three input beams. I can take it like this way; 1 2 3 the same beam, this is E1 this is E, this is E1, this is E1.

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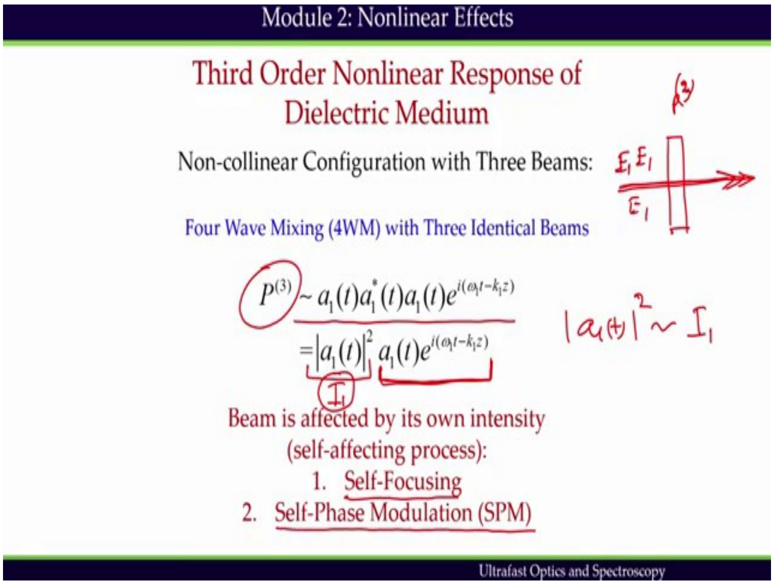


If we take identical or I can also think of like this, three identical beams propagating along the same direction, so three identical beams propagating along the same direction. So, it is experiencing third order non-linear process.

In that case this previous equation, all these three beams will be the same and that is why we get, we end up with getting this expression which is nothing, but intensity of the first beam of its own intensity.

So, what is does it suggest just like beam coupling expression what we have seen you have said in the beam coupling we have said that the second beam is affecting the first beam. Here, one can say that the first beam; the beam is getting affected by it is own intensity and this is the polarization, which is responsible for this.

Therefore, the intensity of the beam effects itself. This polarization results in and emitted pulse at center frequency of the input beam and the emitted beam propagates along the same direction of the input beam, but the emitted pulse gets affected by its own intensity. This is why the process is called self-affecting process. There are two important self-affecting processes are known oneself focusing, another one self phase modulation. We will stop here and will continue the subject in the next class.