Ultrafast Optics and Spectroscopy
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Lecture - 11

Welcome back to the module 3 of the course Ultrafast Optics and Spectroscopy. In this module, we are studying different non-linear effects, which can be generated due to propagation of ultrafast powers through a dielectric medium.

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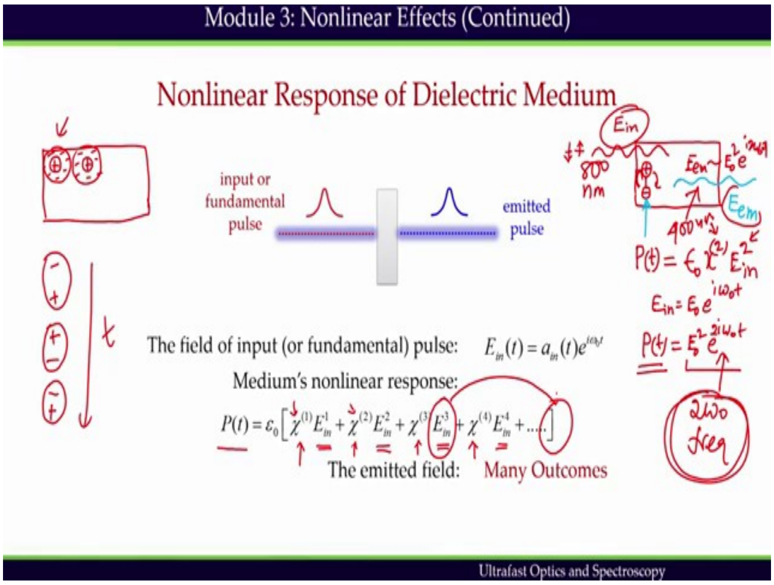

We know that ultrafast pulse is a propagating electromagnetic wave. Propagation of an ultrafast optical pulse, in an optical medium particularly in a dielectric medium, results in many non-linear effects. We have already seen that at high electric field strength, when radiation field is comparable to the atomic field strength, which binds nuclei and electrons. The relation between the polarization and the input radiation field no longer remains a linier one.

The general relationship between the polarization and the input electric field is then expressed by Taylor series expansion, which is given here, this polarization and these are the input field and it depends on this powers. The terms proportional to E square, E cube and E to the power 4 and so on, represent the non-linear polarization of second third and fourth order respectively. And this χ, associated χ, they represent frequency dependence second third and fourth order electric susceptibility is respectively.

We have seen that each polarization results in several outcomes. So, what happens; we will draw this figure which we are familiar with now this input beam entering the medium. This input beam will say E is changing the polarity all the time in every half cycle and this is why we are creating a dipole in the dielectric medium. A dielectric medium is a medium where there is no free electron, just like metal, it has an iron core and then surrounding we have electrons bound to that core.

Similarly, here I have an iron core and then electrons bound to this ion core there is no free electron or the electron, which can move freely in the medium. So, what happens, when due to the input electric field, we distort this electronic cloud. We distort it like this way; let us say we distort it with like this way. This is positive, this is negative again, but this distortion is not static distortion. This distortion is dynamical distortion, because it is changing over the time and it is changing over the time, becauseelectric field is changing the direction in every half cycle and that is why this distortion will also change the polarity in every half cycle.

So, I will get at different time I will have this changes, this as a function of time. So, what is going on in the end dielectric medium is behaving like a like an oscillatory dipole, and any oscillatory dipole will create new radiation and that is the imitate radiation, which we see. So, this oscillatory dipole will oscillate, this dipole will oscillate and due to this oscillation, I will create a new beam that is called emitted beam and this emitted beam is due to the, this emission occurs due to this dipole oscillation, this is a $E_{emitted}$.

So, these are the three step mechanism, we have been discussing in this module and we represent the non-linear effect, non-linear frequency, generation with the help of this figure. Now, we said if this oscillation of the dipole is manifested in the medium by the new polarization, time dependent polarization which will be controlled by the power law, let us say I have second order process.

So, in that case I can write down polarization as $\chi^{(2)}$ E^2. This is within bracket, we are saying it is an order and without bracket, that is a power. So, this is input beam So, if I express input beam in a following way $E_0e^{i\omega t}$. Let us say I have monochromatic plane wave.

The same is true for the pulse as well, but if I have monochromatic plane wave then I have this equation and then P(t) can be written as E_0 square e to the power 2i omega naught t. So, what does it mean? It means that the polarization, which I will create due to propagation of this input beam the second order of polarization, which I will create, which will also oscillate at 2 omega naught frequency.

So, omega naught cannot is the angular frequency for the input beam and the polarization which I have created will have a oscillation with 2 omega naught frequency.We know that what is the frequency of the dipole oscillation, that is the same frequency we get for the emitted beam. So, E emitted will have similar characteristics of E square e to the power 2i omega naught t.

There will a phase matching conditions which needs to be fulfilled as well, but from a simplified point of view, we create a new frequency which will oscillate at 2 omega naught. That is why, if you use 800 nano meter as a input beam we get an output beam 400 nano meter. So, this is all about frequency conversion, we can have different outcomes and we have seen many outcomes. Here, in today's talk, in today's lecture will be focusing only on the third order process and the higher order processes. So, these are the process which we are interested in this lecture.

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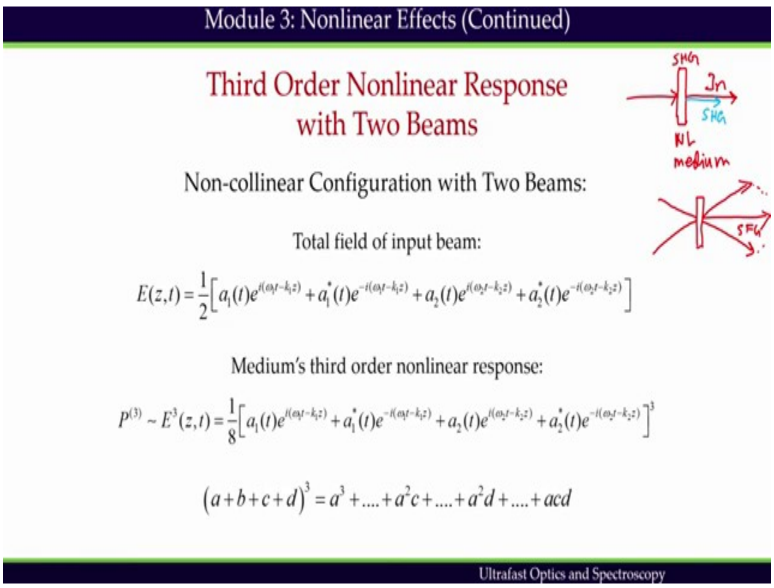

First we will consider third order process with two beams in non collinear configuration. Non collinear and collinear configuration we have seen it, already we have the non-linear medium. In this medium we can have beam propagating along this direction. If the beam is propagating along this direction, then if it is SHG medium second harmonic generation medium, then I will have the parent beam going, but let us say 10 percent of this parent beam will be converted to the second harmonic beam. And second harmonic beam I am representing and they are propagating co linearly with the input beam.

So, this is the SHG beam and this red one is the parent beam, this is the input beam or sometimes we call it fundamental beam. So, this is collinear configuration. In non collinear configuration when you think of it we have let us say two beam non collinear configuration. I have one beam coming like this way another beam coming like this way, they are interacting in the medium non-linear medium. And due to this interaction, we can have some frequency generation and we have seen the some frequency generation can be shown like this way.

This way we create some frequency generation (SFG). So, this is non collinear configuration, we will assume that we have non collinear configuration and if we have non collinear configuration then each beam will draw this non collinear configuration, one more time here to show the fields.

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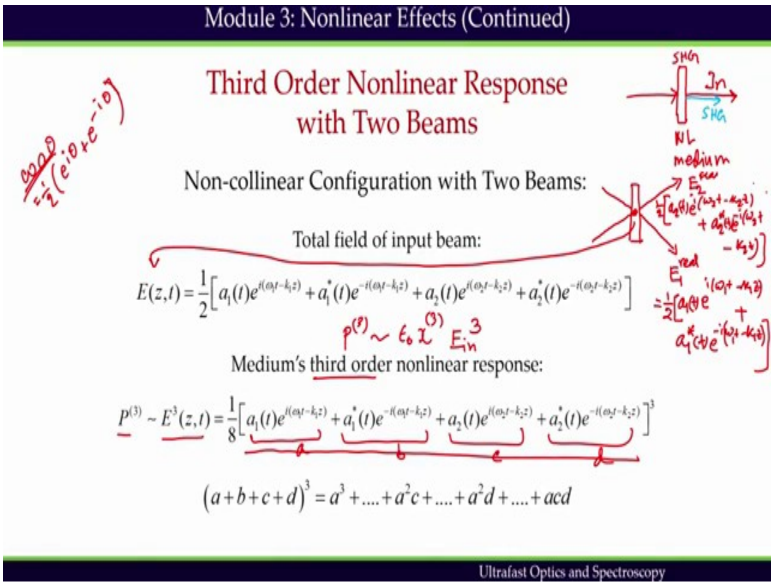

So, I have this medium and this one input beam going like this way, another input beam going like this way and I can write down this input beams as with the complex notation is going to E1 and this is going to be E2, which is nothing, E1 can be written as if it is the real field. I can write down as 1/2[a_1t (that is the envelope function) × e to the power iω_1t minus k_1z plus a_1^*t e to the power minus iω_1t minus k_1z].

So, this is the way we represent the real field in the, this is the real field of the first beam and similarly, real field of second beam can be written as /2[a_2t (that is the envelope function) × e to the power iω_2t minus k_2z plus a_2^*t e to the power minus iω_2t minus k_2z].

So, this complex conjugate is nothing, but our principal, which suggest that $\cos(\theta)=1/2[e^{i\theta}+e^{-i\theta}]$. So, this is this is exactly what we have $\cos(\theta)$ is the representing the real field.

So, what we have given is the at this point, where two beams are interacting in non collinear configuration total electric fields are additives. So, electric field is of this kind. Now, if I consider medium as third order non-linear response which means that I am consider only third order process which is nothing $\chi_0\varepsilon^{(3)}$, that's a third order susceptibility multiplied by E input beam to the power cube. And if it is so, then I can express approximately third order process as the cube of this full the total electric field and if I cube it, then I can consider this term to be a, this term to be b, this term to be c and this term to be d.

This equation is reduced to 12th standard equation or 10th standard equation of a plus b plus c plus d whole cube which can give many terms and we know each term is interesting and give rise to certain non-linear effect, we have seen that before. Now, we will quickly go over each one and then come back to the term, which we are interested in this lecture.

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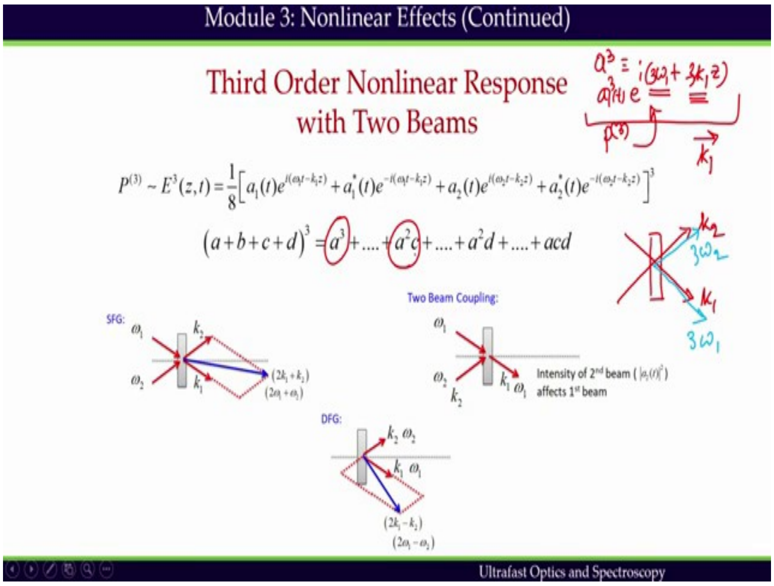

Now, if we look at the cube term, let us say I am interested in a^3term. If I am interested in a cube term is nothing but then a_1 this is a cube term a cube, which is equivalent to a_1 cube t into e to the power i then $3\omega_1$t minus $3k_1$z. So, what does it suggest? It suggests that if I have a cube term, if I consider a cube term then the polarization will depend on this oscillatory function.

This induces polarization and this gives you the third harmonic generation and new emitted pulse entered at $3\omega_1$ along the direction of the k_1. So, $3k_1$ suggesting that I am following still k_1 direction and $3\omega_1$, suggesting that the new frequency which will be generated is at $3\omega_1$. So, what does it mean it means that when I have non collinear configuration and I have two beams going like this way. Each direction, this is your k1 direction, this is your k2 direction. So, this cube, a cube term will produce third harmonious along the same direction so, long that direction I will produce another beam, which is going to be this one, this is going to be $3\omega_1$ and this is going to be another beam, this is going to be $3\omega_2$ along the input beam. We are going to create this third harmonics.

The polarization associated with this a square c term, we can take a look at it and the polarization can be written in a following way. We can write down the polarization of with the term a cube a square c.

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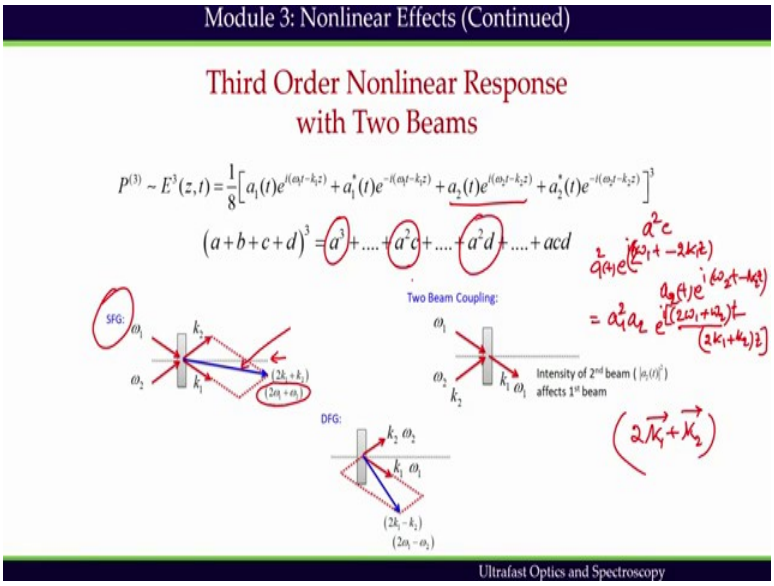

So, a square c will give me a1 t square e to the power i2 omega$(\omega)_1$ t minus $2k_1z$ multiplied by c; c is this one. So, a_2t, e to the power i omega 2t minus k_2z. So, if I simplify this I get a1 square a2 e to the power i then 2 omega 1 plus omega 2 minus, I have t and then minus I have 2 k1 plus k2 z, this is the term we get. So, polarization; this polarization will induce another beam, it will produce another beam at 2 omega 1 plus omega 2 frequency 2 omega 1 plus omega 2 frequency in this frequency, but which direction? Direction is going to be 2 k1 plus k2 direction. So, this is the direction, I have to follow and 2 k1 plus k2 direction is given is shown it here.

It is not along the perpendicular axis, which is shown through this grey line is not along that axis is of the axis, but the direction is shown here, 2 k1 plus k2, this is called some frequency generation, third order some frequency generation. Some frequency generation can happen in second order as well as third order and higher order, is just is combining them. On the other hand, I can have different frequency generation which is shown it here, difference frequency generation is due to a square d term.

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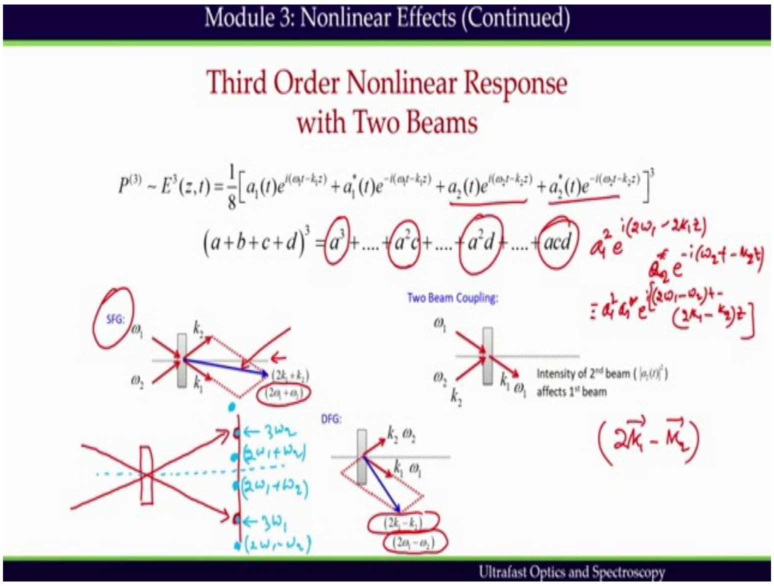

We can take a look at a square d term as well in a square d term, we have a1 square e to the power i 2 omega 1 minus 2 k1 z multiplied by a2 star, because it is d; so, this one is d; so, a2 star e to the power minus i omega 2 t minus k2 z. So, if you simplify it then what I get is that a1 square a2 star multiplied by e to the power i 2 omega 1 minus omega 2 t minus 2 k1 minus k2 z. So, what I get is the difference frequency generation, this polarization will create and new beam at 2 omega 1 minus omega 2 frequency and the direction is going to be along this 2 k1 minus k2 direction, which is shown here.

So, if we project that so let us say I have a non-linear medium and I am seeing it from the top. So, I have two beams going along this way these are the two beams, input beams. So, on a screen I will have one spot here, one spot here, due to the input beam. Now, in addition to that I will have one more spot here at the same position, these are the third harmonic generation. Then I will have, if this is the perpendicular axis, then I will have one more blue spot, which is a new beam created at this point that is called some frequency generation 2 omega 1 plus omega 2.

So, this blue part is 3 omega 1 and this blue is 3 omega 2. So, I will have two beams here and here, there both is going to beam 2 omega 1 plus omega 2. On the other hand here, we will have one more beam and here we will have one more beam, this beam is difference frequency generation 2 omega 1 minus omega 2. This is called difference frequency generation. So, both different frequency generations so, looking at that the position, on a screen, we will be able to identify each beam on the screen and one can identify where is the difference frequency generation occurred and where the some frequency generation occurred.

Now, in the end the last term, which we are interested in here is this term, this is ac d term and we quickly went over this term in our previous lectures acd term is nothing, but two beam coupling acd term. I will write down the expression associated with a c d and that will clarify immediately, why it is two beam coupling, a c d is an interesting two beam coupling term.

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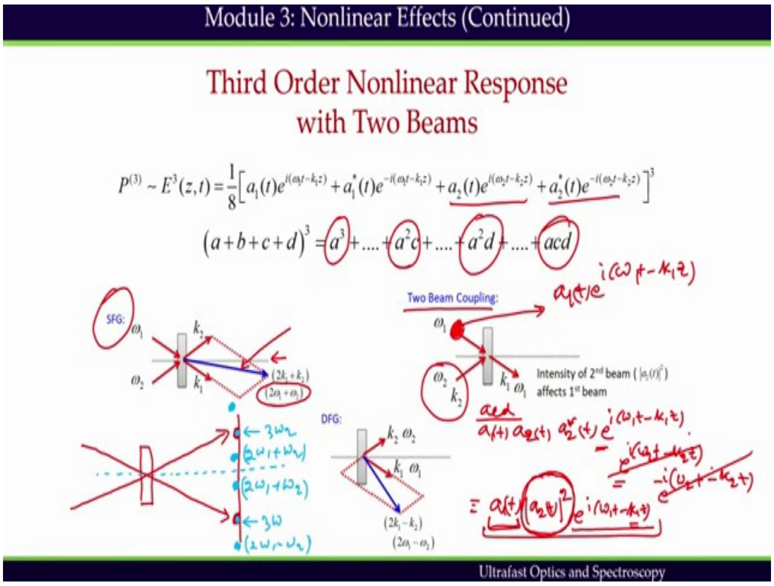

Now, a c d can be written as a 1 t this is a c d so, a 1 t and then c is going to be a 2 t and then d is going to be a 2 star t then e to the power I can write down e to the power i omega 1 t minus k 1 z multiplied by c is a 2 sorry, a 2 is already written so, multiplied by e to the power i omega 2 t minus k 2 z multiplied by e to the power i plus i omega 2 t I am sorry, minus omega 2 t and minus k 2 z.

So, this is your a term, this is c term and this is d term. If I multiply then what we get finally, if we simplify this one then we get, a 1 t then a 2 t square modulus and then e to the power i omega 1 t minus k 1 z. These two terms cancel out and we get finally, this expression. So, what does it mean? It means that I will get one more beam which will be propagating along k 1 direction, but that beam see a 1 t e to the power i omega 1 t minus k 1 z is representing the first beam, this beam, because this input beam was nothing, but a 1 t e to the power i omega 1 t minus k 1 z. So, the same beam I get back, but it is getting affected by the intensity of the, this is expressing the intensity of the second beam.

So, the first beam I am getting back here with the same frequency, but it is amplitude or it is overall shape is getting affected by the second beam intensity. And that is why it is called two beam coupling and implementation of this technique is called polarization getting. So, we will go for the polarization getting in detail.

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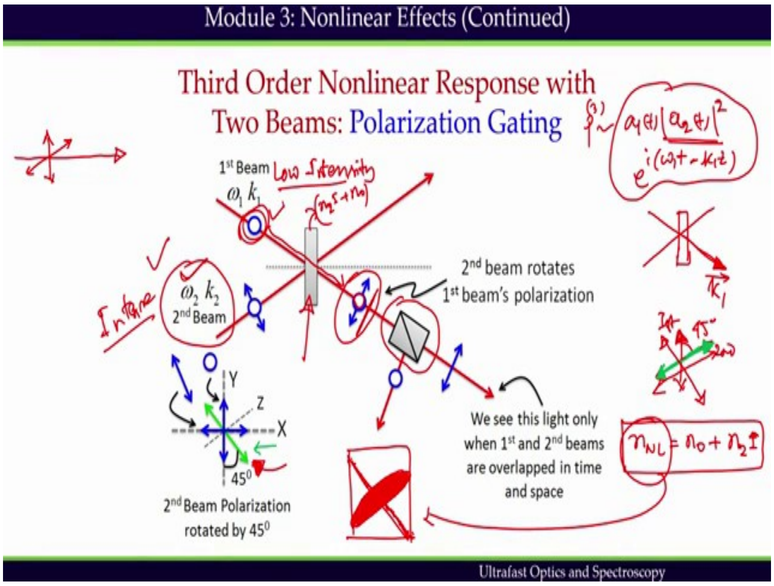

The third order polarization term, which is responsible for two beam coupling is expressed as, we have seen that it can be expressed as that is associated with a c d term third order polarization, which is responsible for this polarization gating is a(t) then a 2 t that is the intensities multiplied by e to the power i omega 1 t minus k 1 z. This is the term which is responsible.

This third order polarization yields emitted pulse that has the same frequency and direction as the first beam, but allows the first beam to get affected by the intensity of the second beam. So, the first beam is getting affected by the intensity of the second beam, that is the basic idea of this term. Polarization getting involves two input beam propagating in non collinear geometry, one beams polarization is rotated at 45 degree to the other.

So, what we are doing here is that, that is the configuration we take, because both beams. So, the beams which is we said that polarization in the two beam coupling we are saying we have said that the new beam which is created is going to be travelling along the same direction of the input beam this k 1 input direction.

So, this is very hard to differentiate or separate these two beams, the output beam which has been created the emitted beam, which is being created by this polarization and the corresponding input beam. Because, they are propagating along the same direction very difficult to separate them. So, we have to use some optical trick to separate them. The optical trick is that the first beam we have taken to be vertically polarized, this sign indicates that is vertically polarized. Vertically polarized, what does it mean? Let us say this is the direction of the propagation direction, along this propagation direction, this is the vertically polarized beam and this is horizontally polarized beam.

So, the vertically polarized beam; it means that it is the arrow, the electric field, locus of the electric field is pointing towards us, if you are looking at the picture from the top. And on the other hand, the other beam the second beam we have taken, the second beam polarization has been rotated at 45 degree which is shown here by this green color arrow, double headed arrow. So, 45 degree, the moment we rotate it at 45 degree then I can have components along the y and z direction.

So, what we have is that the first beam is vertically polarized and relatively low intensity as well, this has low intensity. So, this beam does not induce non-linear polarization in the medium itself; however, the polarization of the second beam is rotated at 45 degree with respect to the first beam. So, if the first beam is vertically polarize like this, then second beam is going to be rotated at 45 degree. So, polarization looks like this, this is the second beam and this is the first beam. Furthermore, second beam is intense enough to induce non-linear third order response, which will affect the weak field of the first beam that is the way we would expect.

So, this is intense enough and this is low intensity. Now, the weak field can be decomposed into orthogonal field components so that one stays parallel to another stays perpendicular to the strong field. So, this one is the strong field I will draw, I will change the colour, the strong field colour will make it as shown here, we will make it green. So, this is the strong field intense field second beam and the weak field is shown by this red color and both the polarizations are different, they are rotated by 45 degree. Now, what will happen if I have this situation then I can decompose my first beam along this direction as well as perpendicular to that direction. So, I can have two different components for the weak beam.

The one component would be parallel to the intense beam strong field, another one would be perpendicular to the strong field. Strong field when we induce that strong field in the medium, it induces optical kerr affect. We know optical kerr effect right now, we know that non-linear refractive index depends on its intensity such as and following this equation.

So, this is non-linear refractive index, so what happens when we use of field, strong field at this at the 45 degree angle we create a big non-linearity along this line along this direction and that depends on the intensity, because intensity is very high. But along this direction perpendicular to it, we have we do not induce that effect, because it depends on the intensity is localized along this direction only. But because intensity is not localized along this direction we do not create appreciable Kerr effect or non-linear refractive index.

So, what happens in the medium we see that along a particular direction, I have high intensity and this high intensity is giving higher non-linear refractive index. So, along this direction the medium is giving different refractive index as compared to this direction, this perpendicular direction. And, because of that difference, the medium can behave like anisotropic medium is not and it can show non-linear birefringence. This means that non-linear refractive index seen by weak field component parallel to the strong field is different from that seen by the weak field component perpendicular to the strong field.

This non-linear birefringence experienced by weak field components leads to polarization rotation of the weak field. This weak field when it is propagating through this due to this non-linear birefringence created by the second beam into i plus n naught. This is the non-linear refractive index. This beam polarization, this input beam polarization will be rotated, does medium acts as wave plate wave plate. We know that wave plate can rotate the polarization. So, medium acts like a wave plate, when this two pulse are as interacting with each other and due to the strong field created in the medium by the second beam. And with sufficient intensity of the second beam and with an optimum medium length a full 90 degree rotation of the polarization of the first beam can occur.

So, what we are showing is that after propagation through the medium, I have both vertical polarization given by this circle and horizontal polarization given by this double headed arrow, both are there. So, vertical polarization representing the input beam; input beam, it is not possible that 100 percent of the input being will be converted in a non-linear conversion process. It is just fraction of that input beam will be converted.

So, fraction of the input beam polarization has been rotated by 90 degree and most of the beam still vertically polarized. But now, we have two different polarizations; one is horizontal polarized light that is the beam which we have created due to this polarization getting. On the other hand, the vertical polarization is remaining to be the input beam polarization. Thus, final emitted beam and the weak field although they are propagating along the same direction have orthogonal polarization; they can now be separated by a polarized polarizing beams splitter.

So, this kind of beam splitter is available, which can separate two polarizations in along two different direction. This arrangement is called polarization getting, which is used in polarization spectroscopy or popularization getting spectroscopy and is used in non-linear optical spectroscopy and non-linear pulse measurement. Phenomenological interpretation can be easily given here, birefringence can only be seen in an isotropic medium that we know already. However, transient anisotropy can be introduced in the medium by utilizing strong field, strong field here we have used for the second beam.

Here, electrons oscillate with the incident field at 45 degree, but due to high intensity the electron cloud in the medium gets distorted, which introduces transient anisotropy in the medium. Then the medium transiently acts like wave plate, optical device which is used to rotate the polarization and rotates polarization of the first beam.

So, the medium; this medium behaves like a wave plate at a particular intensity of the second beam and that is why, it can rotate part of this input beam, first beam to the by 90 degree and we can now, separate it. And this rotation is possible only when they are specially and temporary overlapped, if they are not overlapped this transient anisotropy cannot be realized. So, this is a beautiful demonstration of the a c d term which we have already shown this is two beam coupling. We will stop here and we will continue this module in the next lecture.