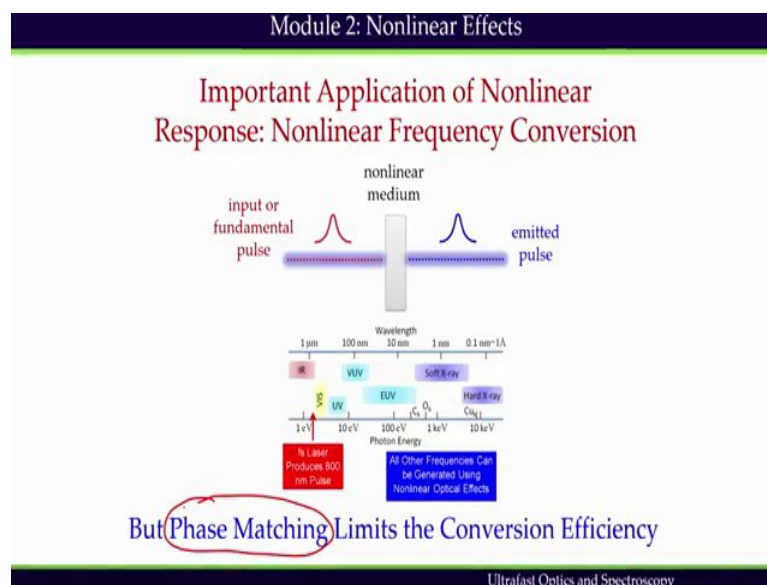


Ultrafast Optics and Spectroscopy
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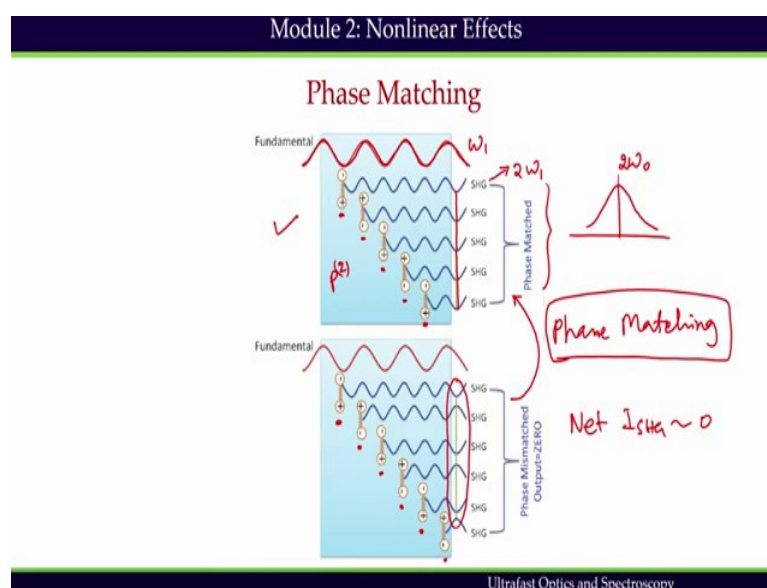
Lecture - 10

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What is phase matching? Is an important concept which we will understand before we end this module.

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So, what is phase matching? Let us demonstrate the meaning of phase matching in non-linear frequency conversion process, for simplicity we shall use second harmonic generation. Propagation of the input beam with ω frequency induces second order polarization in the dielectric medium. So, I have this input beam fundamental beam propagating through the medium.

When is propagating through the medium, I am creating non-linear polarization at different points of the medium, I am creating non-linear polarization and each dipole is going to oscillate and that is why each dipole is going to emit another frequency because it is second harmonic generation that is why it is creating 2ω . So, this is ω that is why the emitted frequency is going to be 2ω because it is SHG process which is involved.

Now, in this figure we have shown that every point, every dipole in the medium is creating emitted beam and all the emitted beam are in phase. If they are in phase, then they will constructively interfere and we know that the result of constructive interference is nothing, but creating a pulse, in that central frequency 2ω central frequency.

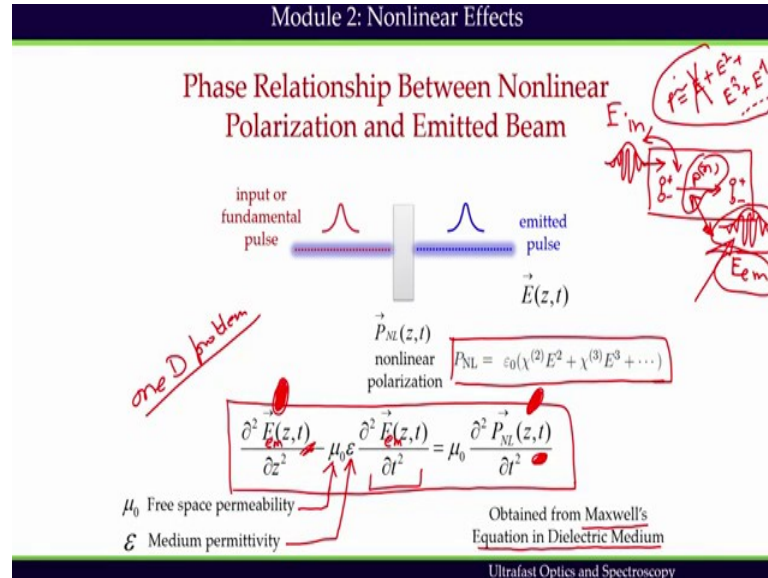
But it may so, happen that each dipole in the medium is emitting SHG field, but they are out of phase. And if they are out of phase net SHG field is going to be 0, net output is going to be 0. The process by which we try to bring this figure to this figure, this process is called phase matching. And this is not very easily achieved in any medium we have to work on it.

Phase matching is the only limiting factor in non-linear conversion process. So, to summarize what does not mean by phase matching, the frequency of the induced polarization is ω equals 2ω in this case. As oscillating dipole is the source of electromagnetic radiation and new light at ω equals 2ω is created at the same frequency of the induced polarization. However, the phase of the induced polarization and emitted field may be different.

If the phase of the induced polarization and emitted field is the same, then we say that phase matching condition is achieved. If the induced polarization creates new light that becomes out of phase with the light it has created earlier two contributions cancel out and consequently intensity of the emitted beam become 0. On the other hand if the two

contributions interfere constructively, intensity of the emitted beam is amplified and thus phase matching is achieved.

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Because phase matching ultimately depends on the phase relationship between non-linear polarization and the emitted beam, it is quite instructive to look at the equation which relates non-linear polarization and the emitted beam. Here we will remind our self that two step process that will help us understand it quickly. I have an input beam going through the medium this is input beam.

Input beam is creating a non-linear polarization here as well as here. Because input beam is propagating through the medium, which means that polarization is also propagating through the medium. And due to this polarization let us say this is I; I created nth order polarization, this polarization will create another emitted beam. Now, whether we will get this emitted beam or not that depends on the phase relationship. So, this is emitted beam, this is input beam.

So, whether we will get any intensity or any field strength for this emitted beam, that depends on the phase relationship between the induced polarization and the emitted beam. We know how this electric field, input electric field and the polarization is related. That is related by E plus E square plus E cube plus E 4 like this, the Taylor series expansion. What we need to know now how the polarization induced polarization and emitted field they are related. (Please look at the slides for mathematical expressions)

They are related by the non-linear equation of optics. The derivation of this equation will be given at later stage of this class not now, we will directly look at the equation. Here it is the second derivative with respect to z we are considering only z direction one dimensional problem in space. This E is emitted field which means, this is also emitted field with respect to and we have taken second derivative with respect to time.

This is free space permeability and this one medium's permittivity and this P_{NL} is the non-linear polarization which is expressed by this. We have taken out the linear contribution, this one we have taken out and the rest of the part is called non-linear polarization, this is non-linear polarization inserted here.

So, which term is now familiar to us and this equation is obtained from Maxwell's equation in dielectric medium. We will find out the derivation for this equation later, but this equation will be very much useful to correlate electric field generated due to induced polarization P_{NL} . This equation also suggests that non-linear polarization this one, acts as a source for emitted field.

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Module 2: Nonlinear Effects

Phase Relationship Between Nonlinear Polarization and Emitted Beam

$$\frac{\partial^2 \vec{E}(z,t)}{\partial z^2} - \mu_0 \epsilon \frac{\partial^2 \vec{E}(z,t)}{\partial t^2} = \mu_0 \frac{\partial^2 \vec{P}_{NL}(z,t)}{\partial t^2}$$

To obtain solution, Let us consider

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

$$\vec{P}_{NL}(z,t) = b(z,t) e^{i(\omega t - k_p z)}$$

Under weak polarization and slowly varying envelope approximations, simplified form of nonlinear equation of optics

$$\frac{\partial a(z,t)}{\partial z} = -i \frac{\mu_0 \omega_0^2}{2k_0} b(z,t) e^{i(k_p - k_0)z}$$

Phase relationship between polarization and emitted beam

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In order to solve this equation, we shall present emitted field in the time domain first. So, this is the emitted field which is an expression of a pulse as I have told you that non-linear polarization is also like a pulse. Because it is propagating, it is generated due to the input beam, but what is the difference between these two expressions?

The difference between these two expressions are following; we have two different envelope functions we have taken. So, the envelope functions can be different, but their frequency non-linear polarization frequency and emitted beam frequency they are the same. And this is always we are saying in this in this module that induced polarization frequency is going to be the same as the input sorry output or emitted beam frequency; However, it is not necessary that we should have wave vector same.

So, under weak non-linear polarization and slowly varying there are two approximations we have to take in order to solve this problem. So, we have taken a general solution for the emitted field and the non-linear polarization. And then we can plug that in, this equation. And under weak non-linear polarization and slowly varying envelope approximations this equation it can be simplified to this equation. The mathematical derivation for this simplification will also be given at a later stage, not now.

So, if you look at this simplified equation, what we get here is that, the emitted field envelope is related to the polarization field envelope as well as the phase difference, the phase relationship between the emitted beam and the outgoing beam, this should be k naught z. So, what we see is that final expression has a phase information. And this information contains the phase relationship between the emitted field and the polarization field, k naught is the wave vector associated with emitted beam and kp is the wave vector associated with the polarization, non-linear polarization.

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Module 2: Nonlinear Effects

If Phase Matched $k_0 = k_p$ *easy achieved*

$$\frac{\partial a(z,t)}{\partial z} = -i \frac{\mu_0 \omega_0^2}{2k_0} b(z,t) e^{i(k_0 - k_p)z}$$

$I = |a|^2$

$$\checkmark \frac{\partial a(z,t)}{\partial z} = -i \frac{\mu_0 \omega_0^2}{2k_0} b(z,t)$$

$$\boxed{a(z,t)} = -\int i \frac{\mu_0 \omega_0^2}{2k_0} b(z,t) dz = -i \frac{\mu_0 \omega_0^2}{2k_0} z b(z,t)$$

$$I(z,t) \sim |a(z,t)|^2 = \frac{\mu_0^2 \omega_0^4}{4k_0^2} z^2 |b(z,t)|^2$$

$I(z,t) \propto z^2$ Emitted beam intensity grows quadratically as a function of medium's thickness

But not easy to achieve.


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Now, if k this should be k naught z . Now there are two conditions we can have, the first condition can be k naught equals k_p , if k naught equals k_p , then the phase factor goes down, you do not have this phase factor anymore, we get this equation, simple integration we will get this equation. And intensity so, this is your envelope function for the emitted beam. So, intensity is nothing, but the square modulus of the field envelope which is given by this expression, which suggests that the intensity of the emitted beam will be proportional to z square. (Please look at the slides for mathematical expressions)

So, if non-linear process holds k naught equals k_p condition that is phase matched condition, the emitted field grows linearly with material thickness. The field is here grows linearly with respect to material thickness z . When a non-linear process holds k naught equals k_p condition the non-linear process is called to be phase matched, but we have to remember that this condition is not easily achieved. (Please look at the slides for mathematical expressions)

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Module 2: Nonlinear Effects



If Not Phase Matched $k_0 \neq k_p$ common

$$\frac{\partial a(z,t)}{\partial z} = -i \frac{\mu_0 \omega_0^2}{2k_0} b(z,t) e^{i(k_0 - k_p)z}$$

Where, phase mismatch is given as

$$\Delta k = (k_0 - k_p)$$

$$\frac{\partial a(z,t)}{\partial z} = -i \frac{\mu_0 \omega_0^2}{2k_0} b(z,t) e^{i\Delta k z}$$

$$\int_0^L \frac{\partial a(z,t)}{\partial z} dz = -i \frac{\mu_0 \omega_0^2}{2k_0} b(z,t) \int_0^L e^{i\Delta k z} dz$$

$$a(L,t) = -i \frac{\mu_0 \omega_0^2}{2k_0} b(z,t) \left[\frac{e^{i\Delta k z}}{i\Delta k} \right]_0^L$$

where, we have used SVEA to take $b(z,t)$ out of the integral

b(z,t) vary very slowly w.r.t z

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Mostly this is going to be again k naught z sorry for this wrong printing. If this they are not equal and that is very common, when a pulse is propagating through the medium, it is hard to bring them in phase. That is why it is hard to get the condition k naught equals k_p full filled. So, when they are not equal then we can write down Δk as the difference between k naught minus k_p and this is a very common and general tendency. When the emitted wave propagates through the medium it easily becomes out of phase

So, previously when k_{naught} equals k_p we had, we have seen that intensity of the emitted beam is proportional to z^2 . So, this in general does not work in most of the practical scenario. That is the induced polarization creates new light that becomes out of phase with the light it creates earlier, this is why instead of making more such light the two contributions cancel out.

In this integration, we have taken this envelope function out assuming the fact that $a(z, t)$ varies very slowly with respect to z . That is called slowly varying envelope approximation and based on that approximation, we can take this term out of the integral and we get this integration done.

Module 2: Nonlinear Effects

If Not Phase Matched $k_0 \neq k_p$

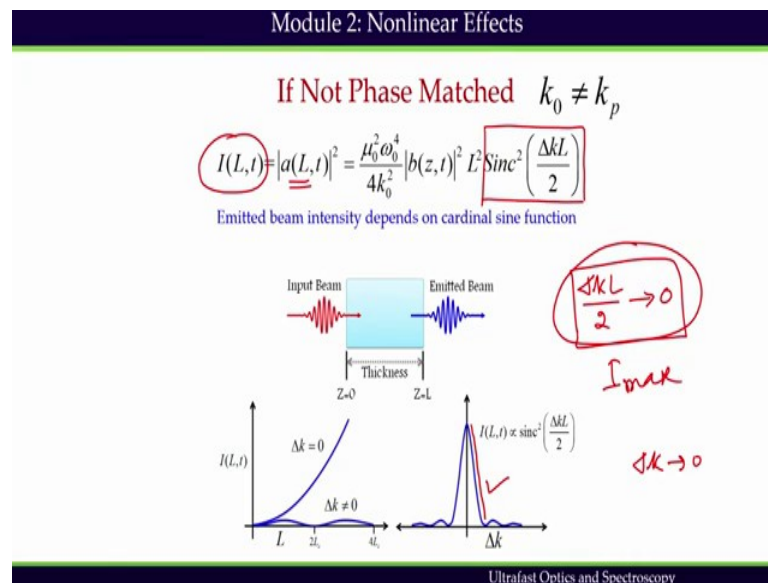
$$\begin{aligned}
 a(L, t) &= -i \frac{\mu_0 \omega_0^2}{2k_0} b(z, t) \left(\frac{e^{i\Delta k L}}{i\Delta k} - \frac{1}{i\Delta k} \right) \\
 &= -i \frac{\mu_0 \omega_0^2}{2k_0} b(z, t) \left(\frac{e^{i\Delta k L} - 1}{i\Delta k} \right) \\
 &= -i \frac{\mu_0 \omega_0^2}{2k_0} b(z, t) L e^{i \frac{\Delta k L}{2}} \left(\frac{e^{i \frac{\Delta k L}{2}} - e^{-i \frac{\Delta k L}{2}}}{2 \cdot \frac{i\Delta k L}{2}} \right) \\
 &= -i \frac{\mu_0 \omega_0^2}{2k_0} b(z, t) L e^{i \frac{\Delta k L}{2}} \left(\frac{i2 \cdot \sin\left(\frac{\Delta k L}{2}\right)}{i2 \cdot \left(\frac{\Delta k L}{2}\right)} \right) \\
 &= -i \frac{\mu_0 \omega_0^2}{2k_0} b(z, t) L e^{i \frac{\Delta k L}{2}} \left(\frac{\sin\left(\frac{\Delta k L}{2}\right)}{\left(\frac{\Delta k L}{2}\right)} \right) \\
 \alpha_{\text{eff}}(f) &= -i \frac{\mu_0 \omega_0^2}{2k_0} b(z, t) L e^{i \frac{\Delta k L}{2}} \boxed{\text{sinc}\left(\frac{\Delta k L}{2}\right)}
 \end{aligned}$$

$\text{sinc} x = \frac{\sin x}{x}$

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And finally, after giving this limit we get an expression. The expression suggests that, emitted beam envelope would depend on definitely L that is the thickness of the crystal, but at the same time it depends on a cardinal sin function. Cardinal sin function is nothing, but something like $\sin x$ by x .

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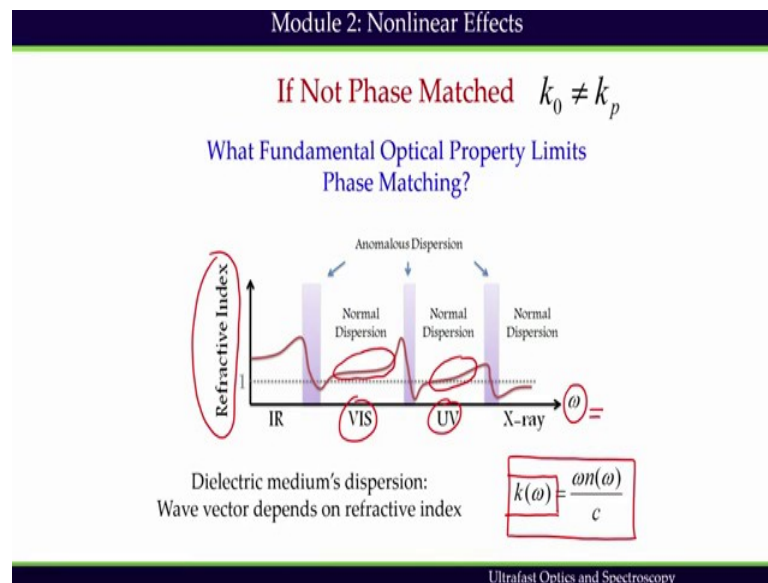
So, if the field is known, we will be able to get intensity is a square modulus of the field, which we get square of this cardinal function. So, intensity will depend on the square of cardinal function. This function, cardinal sin function has maximum at limit x tends to 0. So, if this $\Delta k L$ by 2 tends to 0, then we get I_{\max} . And it is also highly peaked near this, which is shown here in this figure.

So, the non-linear optical effect of interest will experience much greater efficiency if Δk tends to 0. This occurs when the induced polarization and the light it creates remain in phase throughout the non-linear medium and that is called phase matching. But if little bit of phase mismatch will drop down the intensity very quickly to 0.

So, phase matching effectively suggesting that, I have a medium and through this medium the beam is propagating like this way, when the beam is propagating like this way, in this medium I create non-linear polarization in the beginning and in the end of the medium. What we need to remember here, the polarization created in this regime will create emitted beam which is also propagating. And the polarization created in the end of the medium will also create emitted beam which is propagating.

This two emitted beams the beam created later, the beam created earlier they have to be in phase, if they are out of phase then the intensity will go down to 0, but if they are in phase then intensity would be maximum. And we have to bring them in phase to get the maximum intensity of the non-linear frequency conversion.

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But the question is what is the fundamental optical property that contribute to the phase matching problem? And it is the dielectric mediums dispersion which will contribute to the phase matching problem. Dispersion effects have not been discussed so, far in this course we will do it in the next module. But it is the variation of refractive index as a function of the wavelength or the frequency.

And because we are looking at UV visible regime we see that most of the time, we see refractive index is increasing with respect to frequency and that is called dispersion. And there is a very simple equation which represents dispersion k is related to refractive index and k depends on frequency. So, each frequency component in the pulse will have different k .

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Module 2: Nonlinear Effects

If Not Phase Matched $k_0 \neq k_p$

What Fundamental Optical Property Limits
Phase Matching?

An Example: SHG Due to propagation of input beam with ω_1 centre frequency, SHG polarization is expressed as

$$P \sim E_1^2 \sim e^{i(2\omega_1 - 2k_1 z)}$$

Therefore, $k_p = 2k_1$

and, for emitted beam (SHG): $k_0 = \frac{\omega_0 n(\omega_0)}{c} = \frac{2\omega_1 n(2\omega_1)}{c}$

For phase matching, $k_p = k_0$

$$\frac{2\omega_1 n(\omega_1)}{c} = \frac{2\omega_1 n(2\omega_1)}{c}$$

Handwritten notes and diagrams:

- Diagram of a crystal with input field $E_1 \sim e^{i(\omega_1 t - k_1 z)}$ and output field $E_{em} \sim e^{i(\omega_0 t - k_0 z)}$.
- Equation for emitted field: $E_{em} \sim e^{i(\omega_0 t - k_0 z)}$
- Equation for polarization: $P \sim E_1^2 \sim e^{i(2\omega_1 - 2k_1 z)}$
- Equation for wave vector: $k_p = 2k_1$
- Equation for wave vector: $k_0 = \frac{\omega_0 n(\omega_0)}{c} = \frac{2\omega_1 n(2\omega_1)}{c}$
- Equation for phase matching: $k_p = k_0$
- Equation for phase matching: $\frac{2\omega_1 n(\omega_1)}{c} = \frac{2\omega_1 n(2\omega_1)}{c}$

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So, with this little information about dispersion, if we one more time take a look at second harmonic generation process as an example, to understand the fundamental property which contributes to the phase matching problem. We see that, let us consider the centre frequency of the input pulse is ω_1 . So, I have a SHG medium and I have input beam expressed as in slide.

So, second order polarization can be written as, if we forget about this envelope function for now, we can write down second order polarization as this. So, k_p is nothing, but $2k_1$ that is the definition of k_p . And for the emitted beam at k_0 we said that k_0 the emitted field is going to be similar to the polarization field. So, I can write down $E_{em} \sim e^{i(\omega_0 t - k_0 z)}$ that is my emitted field, where k_0 is going to be this expression. Now, for phase matching I have to have this full field, which means I should hold this relationship.

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Module 2: Nonlinear Effects

If Not Phase Matched $k_0 \neq k_p$

What Fundamental Optical Property Limits Phase Matching?

An Example: SHG

For phase matching, $k_p = k_0$ ✓

$n(\omega_1) = n(2\omega_1)$ Must be fulfilled

$n(\omega) \neq n(2\omega)$

But cannot be fulfilled due to dispersion

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We suggest that in order to have k_p equals k_0 , this condition must be fulfilled. Refractive index at ω_1 and refractive index at $2\omega_1$ must be the same. And we have already stated, due to dispersion refractive index will always change as a function of frequency. So, this condition can never hold, because they are not equal in any medium. And because of that we cannot get this condition to be fulfilled easily. (Please look at the slides for mathematical expressions)

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Module 2: Nonlinear Effects

If Not Phase Matched $k_0 \neq k_p$

How do we bypass the dispersion problem to achieve phase matching?

An Example: SHG

For phase matching, $k_p = k_0$

$n(\omega_1) = n(2\omega_1)$ Must be fulfilled

Birefringent crystal shows two refractive indices, n_e n_o

SHG field of ordinary beam in phase with the polarization created by extraordinary beam

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So, what is the solution then? The solution is to use Birefringent crystal. In birefringent crystal, when light propagates it undergoes refraction, but it undergoes double refraction there are two different refracted beam we get. In general when we think about refraction, the light is propagating from lighter refractive index to the higher refractive index medium, then the light bends and this is called refraction. In a particular type of crystal is called birefringent crystal, here also light bends, but they have two different lights now. One of them is called ordinary light another one is called extraordinary light.

And refractive index for birefringent crystal for ordinary and extraordinary beam they are different. So, what happens? Refractive indices are different from for the ordinary and extraordinary beams and therefore, it is possible to achieve phase matching in a following way. It is achieved because the SHG field of one ordinary beam will be in phase with the polarization created by other extraordinary beam as shown here. Ordinary and extraordinary beams can be overlapped by selecting correct incident angle.

So, what happens here if you look at this figure at ω I get refractive index for the extraordinary beam. And at 2ω I get another refractive index for ordinary beam. So, if they are overlapped with each other one can help other to become equal and that is why this relationship can be hold and I can have this phase matching condition fulfilled.

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The slide features a dark blue header bar with the text "Module 2: Nonlinear Effects" in white. Below this, the main content area has a light blue background. It contains the text "In Brief: Propagation of Ultrafast Pulses – Nonlinear Effects" in red, followed by "SHG, THG, SFG, DFG, 4WM, SPM, Phase Matching Controls" in blue. At the bottom, there is a dark blue footer bar with the text "Ultrafast Optics and Spectroscopy" in white.

Module 2: Nonlinear Effects

In Brief: Propagation of Ultrafast Pulses –
Nonlinear Effects

SHG, THG, SFG, DFG, 4WM, SPM,
Phase Matching Controls

Ultrafast Optics and Spectroscopy

So, with this we have come to the end of this module. In this module we have gone of our different optical non-linear optical effects. When ultra fast pulse propagates through

the medium it has high intensity and the high intensity shows non-linear optical effects. We have gone over second harmonic generation, third harmonic generation, fourth harmonic generation, difference frequency generation, four way mixing, self phase modulation and also we have studied phase matching conditions. With this we will end this module we will meet again for the next module.