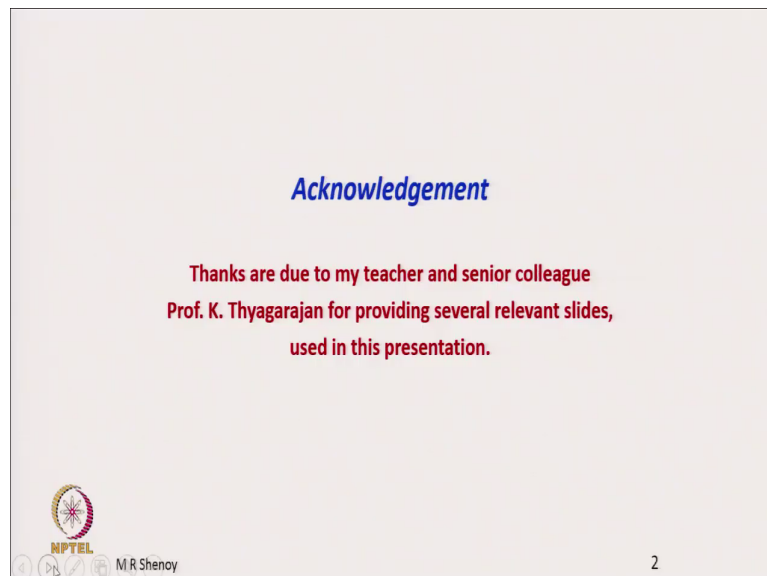


**Introduction to LASER**  
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**Lecture - 37**  
**Lasers in Nonlinear Optics**

Welcome to this MOOC on lasers. We have been looking at some laser systems and applications. So, today I will discuss one of the important applications of Lasers that is in Non-linear Optics, particularly for generation of new frequencies.

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Before I start, I would like to record my thanks which are due to my teacher and senior colleague Professor Thyagarajan for providing several relevant slides, used in this presentation. We have worked together for over three decades, in similar areas alright.

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**Induced Polarization**


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*Nonlinear Optics* is the regime of Optics wherein the induced polarization in the medium has a nonlinear dependence on the electric field associated with the propagating light waves.

**Recall:** In E.M. Theory,  
$$\vec{D} = \epsilon \vec{E} = \epsilon_0 \vec{E} + \vec{P}$$

$\vec{P}$  → Induced Polarization in the medium

For 'small' electric fields  $\vec{P} \propto \vec{E}$   
much smaller compared to the interatomic electric fields



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First Non-linear optics: Non-linear optics is the regime of optics wherein the induced polarization in the medium has a non-linear dependence on the electric field associated with the propagating light waves. So, let me read the definition. So, non-linear optics is the regime of optics wherein the induced polarization in the medium has a non-linear dependence on the electric field associated with the propagating light waves.

We will discuss this and make this clear as we go further alright. Very quickly recall that in the electromagnetic theory the displacement vector D is given by D is equal to epsilon into E, where epsilon is the permittivity of the medium, which can be written as epsilon 0 permittivity of free space into the electric field plus polarization. So, here this P is the induced polarization in the medium. For 'small' electric fields this induced polarization is proportional to E.

When we say small, this small is much smaller compared to the inter-atomic electric fields. This small refers to very small electric fields when compared to the inter-atomic electric fields. I will put some numbers and show what do we mean by this and we will quantify these aspects.

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### Linear Optics

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→ Consider a monochromatic e. m. wave with only one non-zero component of the electric field, so that it can be treated as a scalar:


$$\vec{E} \rightarrow E_0 \cos(\omega t - kz)$$

The instantaneous electric field at an arbitrary point z, say z = 0,

$$E = E_0 \cos \omega t$$

The induced polarization can be written as  $P \propto E$  or  $P = \epsilon_0 \chi E$

$\chi$  → susceptibility, a scalar constant of the medium which represents “response” of the medium to an applied electric field.



$$P = \epsilon_0 \chi E$$

- represents ‘linear response’  
- is the regime of *linear optics*

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Consider a monochromatic wave, to make the discussion very simple consider a monochromatic electromagnetic wave with only one non-zero component of the electric field, so that it can be treated as a scalar. We know that actually electric field is a vector quantity, but considering one component we can treat it as a scalar for simplicity and we can write the plane wave as  $E_0 \cos \omega t - kz$ .

It is a plane wave monochromatic with single frequency  $\omega$ , angular frequency  $\omega$ , propagating in the z direction and  $E_0$  being the amplitude. The instantaneous electric field

therefore, at any arbitrary point  $z$ , say  $z$  equal to 0, without loss of generality we can write  $E$  is equal to  $E_0 \cos \omega t$ .

Now, the induced polarization can be written as  $P$  proportional to  $E$ , I have deliberately dropped the vector sign there because I am considering a scalar component or  $P$  is equal to  $\epsilon_0 \chi E$ .

So,  $\epsilon_0 \chi$  represents the proportionality constant.  $\chi$  is the susceptibility, it is the electric susceptibility which is the scalar constant of the medium and which represents the “response” of the medium to an applied electric field. We will quantify what is meant by this response.

Therefore,  $P$  is equal to  $\epsilon_0 \chi E$ ; this represents a linear response because  $P$  is directly proportional to  $E$ . So, it is a linear relation therefore, it represents a ‘linear response’, which is the regime of linear optics. In other words, in linear optics the normal optics the polarization induced polarization is related to the applied electric field or electric field of the light by this linear relation  $P$  is equal to  $\epsilon_0 \chi E$ .

This is actually valid only when the magnitude of the electric field is much smaller compared to the electric field or the electrostatic field, which is present in the medium that is between atoms the electrostatic field which is present, we will see that they are much stronger than the normal electric fields associated with light.



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**Nonlinear Polarization**

For higher electric fields, the response of the medium becomes nonlinear, and the polarization is expressed as -

$$\rightarrow P = \epsilon_0 \chi E_0 + (\epsilon_0 \chi^{(2)} E^2 + \epsilon_0 \chi^{(3)} E^3 + \dots)$$

$\vec{E} \vec{E} \vec{E}$


$$\rightarrow P = P_L + P_{NL}$$

$\rightarrow \chi^{(2)}$  - 2<sup>nd</sup> order susceptibility;  $\chi^{(3)}$  - 3<sup>rd</sup> order susceptibility, etc.

$$\rightarrow P_{NL}^{(2)} = \epsilon_0 \chi^{(2)} E^2 \rightarrow \chi^{(2)} \text{ (NL) Effects}$$
$$\rightarrow P_{NL}^{(3)} = \epsilon_0 \chi^{(3)} E^3 \rightarrow \chi^{(3)} \text{ (NL) Effects}$$

NOTE:  $\chi^{(i)}$  - are actually Tensors of rank 2, 3, 4, ... etc.

**Where does the Laser come-in in Nonlinear Optics?**



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For higher electric fields, for stronger electric fields the response of the medium becomes non-linear, and the polarization is expressed as P is equal to this is the linear part  $\epsilon_0 \chi$  into E 0 plus  $\epsilon_0 \chi^{(2)}$  into E square plus  $\epsilon_0 \chi^{(3)}$  into E cube plus etcetera a power series. Therefore, P can be written as the polarization can be written as P linear plus P non-linear.

So, P linear is the first part  $\epsilon_0 \chi$  into E 0 whereas, P non-linear is the quantity which is present in the brackets.  $\chi^{(2)}$  is called the 2<sup>nd</sup> order susceptibility,  $\chi^{(3)}$  the 3<sup>rd</sup> order susceptibility and so on. And similarly, the 2<sup>nd</sup> order polarization term which comprises of  $\epsilon_0 \chi^{(2)}$  into E square gives rise to  $\chi^{(2)}$  non-linear effects, and the 3<sup>rd</sup> order term gives rise to  $\chi^{(3)}$  non-linear effects, we will see what are these  $\chi^{(2)}$  effects and  $\chi^{(3)}$  effects.

But, actually in practice note that chi are actually Tensors of rank 2, 3, and 4 depending on because remember that, E is an electric field here actually it is a vector quantity. So, it is E square is actually E into E and E cube is E into E into E so, if we write this vector then, these have to be written in the form of tensors and therefore the susceptibility is actually a tensor, but here I am just considering them as some scalar constant and we are more interested in the magnitude of these susceptibilities.

Now, where does the Laser come-in Non-linear Optics? The title of today's lecture is lasers in non-linear optics. So, we are not really discussing details of non-linear optics, but where does laser fit in or where does laser come as an important tool in the study of non-linear optics. So, where does laser come in non-linear optics, let us see.

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### Higher-order Susceptibilities

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→  $P = \epsilon_0 \chi E + (\epsilon_0 \chi^{(2)} E^2 + \epsilon_0 \chi^{(3)} E^3 + \dots)$

$\chi^{(1)} \sim 1$


→  $\chi^{(2)} \sim 10^{-12} \text{ m/V}$

→  $\chi^{(3)} \sim 10^{-25} (\text{m/V})^2$

→ ∴ The 2<sup>nd</sup> order, 3<sup>rd</sup> order terms will become significant if  $E \gtrsim 10^4 - 10^6 \text{ V/m}$

→ How to estimate the electric field associated with a laser beam?

Using  $E = E_0 \cos \omega t$ , and  $H = H_0 \cos \omega t$ , the magnitude of the Poynting vector gives:



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$$I = \frac{n}{2c\mu_0} |E_0|^2$$

$$I = P/A$$

Power / Area of cross-section

$$\vec{S} = \langle \vec{E} \times \vec{H} \rangle$$

$$\langle E H \rangle$$

$$\langle E_0 H_0 \cos^2 \omega t \rangle$$

$$\frac{1}{2} E_0 H_0$$

$$\frac{1}{2} \sqrt{\frac{\epsilon_0}{\mu_0}} E_0^2$$

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Now, for higher order susceptibilities as indicated the polarization for higher electric fields, for stronger electric fields the polarization is represented as  $\epsilon_0 \chi_1 E$  plus the power series. Typical magnitude of  $\chi_1$ , the linear susceptibility is of the order of 1,  $\chi_2$  is of the order of  $10^{-12}$  meters per volt it could be  $10^{-10}$  to  $10^{-13}$  typically.

So, of the order of  $10^{-13}$  meter per volt and  $\chi_3$  is of the order of  $10^{-25}$  meter per volt square. But note that, the  $\chi_2$  goes with  $E^2$  here and  $\chi_3$  goes with  $E^3$  here therefore, if  $E$  is sufficiently large then only this term becomes significant,  $E^2$  into  $\chi_2$  then only this term can be comparable to the first term. So, normally for moderate electric fields associated with light the polarization is proportional to the amplitude of the electric field.

Whereas, if  $E$  becomes very strong so, this is not  $E_0$ . So, it is  $E$  when,  $E$  becomes strong or high electric fields then although  $\chi_2$  is very small, it is multiplied by  $E^2$  and this quantity could become comparable or non negligible compared to the first term.

In the contribution to the polarization similarly, if  $E$  becomes very large  $E^3$  could be a large number although  $\chi_3$  is a small number and therefore, the 2nd order, 3rd order terms will become significant if  $E$  is greater than or of the order of  $10^4$  to  $10^6$  volts per meter.

For example, if you take the 2nd order coefficient here 2nd order term then  $\chi_2$  is of the order of  $10^{-12}$  and therefore, if  $E$  is of the order of  $10^6$  then it is  $10^{-12}$  is multiplied by  $10^{12}$ . And it comes to the same order as  $\chi_1$  of the order of 1, and that is why we have written if  $E$  becomes  $10^4$  to  $10^6$  of that order then the second order term will become important and similarly the 3rd order term.

Now, how to estimate the electric field associated with the laser beam? If we use  $E$  is equal to  $E_0 \cos \omega t$ , and  $H$  is equal to  $H_0 \cos \omega t$ , then the magnitude of the pointing

vector. So, magnitude of the pointing vector is given by  $E \times H$  here and  $E$  and  $H$  if you assume perpendicular then it is average  $E$  into  $H$ , if  $E$  and  $H$  are perpendicular the magnitude is  $E$  into  $H$ .

So, this is equal to  $E_0$  into  $H_0$  into  $\cos^2 \omega t$ , the average  $\cos^2$  so this is equal to half  $E_0$  into  $H_0$ . Because, average of  $\cos^2 \omega t$  is half and  $H_0$  is related to  $E_0$  the ratio of  $E_0$  by  $H_0$  is  $\sqrt{\epsilon_0 / \mu_0}$ . So, this is equal to half square root of  $\epsilon_0 / \mu_0$  into  $E_0^2$ .

And this can be written in this form here that intensity so, this is nothing but the intensity that is the magnitude of the pointing vector  $S$ . So, the pointing vector  $S$  is given by  $E \times H$ . So, this is nothing but the intensity  $I$  is equal to half square root of  $\epsilon_0 / \mu_0$  into  $E_0^2$ , which can also be written in this form because  $n$  is related to  $\epsilon_0$  and  $\mu_0$ ,  $c$  is equal to  $1 / \sqrt{\epsilon_0 \mu_0}$ .

So, we can write intensity in this form, and intensity is also equal to power per unit area. So,  $P$  here is the power, the output power of the laser beam. So, this is power and this is area of cross section; area of cross section of the beam; cross section. And therefore, we can calculate the electric field corresponding to a certain power of the laser beam.

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### Electric Field of Laser Beam

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→ For a 10 mW laser beam of 1 mm diameter:


$$E_0^2 = \left[ \frac{2c\mu_0 P}{n \cdot A} \right] = \frac{2 \times 3 \times 10^8 \times 4\pi \times 10^{-7}}{1.5} \times \frac{10 \times 10^{-3} \times 4}{\pi(1 \times 10^{-3})^2}$$

$n = 1.5$       for 1 mm

Electric Field amplitude,  $E_0 \approx 2.5 \times 10^3$  V/m → 1 mm spot-size

If we focus the laser beam to 100 μm,  $E_0 \rightarrow 2.5 \times 10^4$  V/m  
 → 10 μm →  $2.5 \times 10^5$  V/m  
 → 1 μm →  $2.5 \times 10^6$  V/m

**- Should be possible to observe nonlinear optical effects!**


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So, let us take a numerical example here, for a 10 milli watt laser typical helium neon laser let us say 10 milli watt laser beam of 1 millimeter diameter, we can calculate  $E_0^2$  is equal to so we are just seeing here  $E_0^2$  is equal to  $2c\mu_0 I$ ,  $I$  is  $P$  by  $A$  divided by  $n$ . So,  $2$  into  $c$  so this is  $2c$ ,  $c$  is  $3$  into  $10$  to the power of  $8$  meters per second  $\mu_0$  is  $4\pi$  into  $10$  to the power of minus  $7$ .

And  $n$  I have assumed  $n$  is equal to  $1.5$  into  $P$  power is equal to  $10$  milli watt. So,  $10$  milli watt  $10$  to the power of minus  $3$  divided by the area of cross section that is  $\pi r^2$  or  $\pi d^2$  square by  $4$ . So,  $\pi d$  diameter is  $1$  millimeter. So, this is  $1$  millimeter diameter. So, if we substitute these numbers we will see that  $E_0$  will come out to be approximately  $2.5$  into  $10$  to the power of  $3$  volts per meter, for a  $1$  millimeter spot for  $1$  millimeter diameter spot.

If we focus the laser beam, the same laser but if we focus the laser beam to 100 micrometer size using a lens then  $E_0$  will become 10 times more. Because,  $E_0$  square will be inversely proportional to square of the diameter so, you decrease the diameter then  $E_0$  square will increase linearly or  $E_0$  will increase inversely with the diameter and therefore,  $E_0$  becomes 10 times more you reduce the diameter further by focusing then, the electric field will become  $2.5 \times 10^5$  volt per meter.

So, all of these are volt per meter so volt per meter. And if we still focus it tightly focus the laser beam to 1 micrometer diameter, it is possible to focus because if you take helium neon laser then the wavelength is 0.6328 micrometer and therefore, you can focus the spot down to the wavelength size.

And that is of the order of 1 micrometer and the electric field associated with the light beam at the focus is  $2.5 \times 10^6$  volt per meter. This tells us that, it should be possible to observe non-linear optical effects using high intensity laser beams or using focused laser beams. So, that is the conclusion that we get by putting these numbers ok.

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**Electrostatic Field in the Medium**


→ If we calculate the electric field of an Hydrogen atom (at the Bohr radius  $\sim 0.53 \text{ \AA}$ ) using

$$E = \frac{1}{4\pi\epsilon_0} \frac{q}{r^2} \approx \underline{10^{12} \text{ V/m}}$$

- between the atoms, i.e. at an interatomic distance of  $\sim 4 \text{ \AA}$ ,

$$E \sim \underline{10^{10} \text{ V/m}}$$

- Electric field of confined/focussed Laser Beams can be comparable!



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If we calculate the what type of electrostatic field is present in the medium, if we calculate the electric field of an hydrogen atom, at the bore radius which is about 0.53 angstrom using  $E$  is equal to  $\frac{1}{4\pi\epsilon_0} \frac{q}{r^2}$   $q$  is the charge which is equal to  $1.6 \times 10^{-19} \text{ C}$   $r$  is  $0.53 \times 10^{-10} \text{ m}$   $E$  is

So, if we substitute the values we will see that it is of the order of  $10^{12}$  volts per meter. And between atoms at an inter atomic distance so, instead of  $r$  is equal to 0.53 angstrom if you put  $r$  is of the order of 4 angstrom or 5 angstrom or 3 angstrom, that is the typical inter atomic distances. Then, we get  $E$  of the order of  $10^{10}$  volt per meter.

This is the kind of electric field which is present in the medium and therefore, the electric field of confined or focused laser beams can be comparable to these and that is where the laser comes. The first one is, it is the intensity of the laser which we can achieve by focusing a

laser beam or by having a high power laser beam, that we can observe non-linear optical effects much easily, alright.

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**Recap: Nonlinear Polarization**

→ For strong electric fields, polarization in a medium is given by

$$P = \epsilon_0 \chi E + \epsilon_0 \chi^{(2)} E^2 + \epsilon_0 \chi^{(3)} E^3 + \epsilon_0 \chi^{(4)} E^4 + \dots$$

Linear Effect

$P = \epsilon_0 \chi E$   
 $= \epsilon_0 \chi^{(2)} E_0^2 \cos^2 \omega t$   
 $(1 + \cos 2\omega t)$   
 d.c. term  
 Second Harmonic

Nonlinear effect

$E = (E_{01} \cos \omega_1 t + E_{02} \cos \omega_2 t)$   
 $\cos \omega_1 t$   
 $\cos(\omega_1 + \omega_2) t$   
 $\cos(\omega_1 - \omega_2) t$   
 $\cos 2\omega_1 t$   
 $\cos 2\omega_2 t$

$\chi$ :  
Linear  
susceptibility

$\chi^{(2)}$ : SHG  
→ SFG  
→ DFG

$\chi^{(3)}$ : THG  
SPM  
FWM

$\chi^{(4)}$ : FHG

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Let us come back to the non-linear polarization here. So, as I mentioned for strong electric fields the polarization in the medium can be represented in this form P is equal to epsilon 0 chi E plus epsilon 0 chi 2 E square and so on. So, a schematic illustration of the linear effect that is if we had only this much, it means polarization is proportional to E then, P versus E would have been a straight line like this.

It is just a schematic illustration, which tells that the relation between P and E becomes non-linear if you consider the higher order terms and which becomes significant when the strength of the electric field becomes sufficiently large. And therefore, chi represents the linear susceptibility which is responsible for the linear effects and the refractive index of the



medium,  $\chi^2$  gives rise to second harmonic generation for example, if we substitute in this term that is  $P$  is equal to  $\epsilon_0 \chi^2$  into  $E$  square.

So, if we substitute for  $E$  is equal to  $\epsilon_0$  into  $\chi^2$  into  $E^2 \cos^2 \omega t$ . So, we have substituted  $E_0 \cos \omega t$  for  $E$  so,  $\cos^2 \omega t$ . This term is  $1 + \cos 2\omega t$ , which means it gives rise to a dc term. So, this is a d.c. term and a term which is at second harmonic.

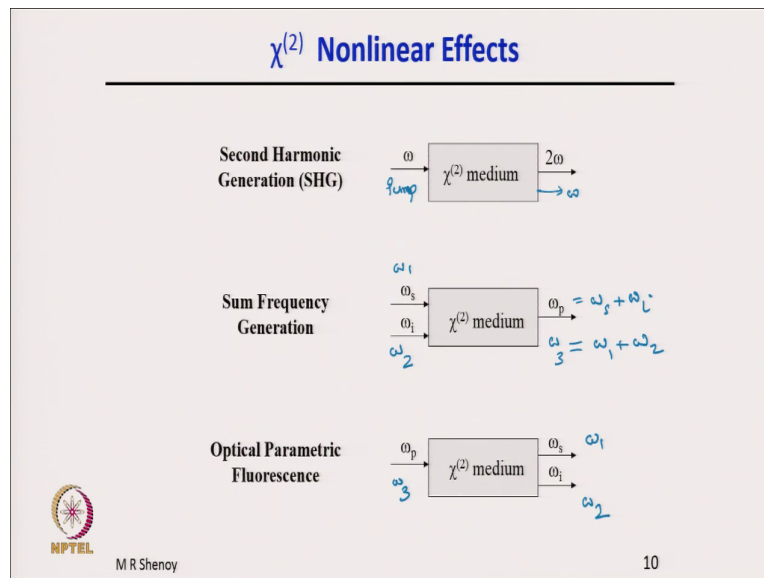
And therefore, we get second harmonic generation due to the  $\chi^2$  term. This represents some frequency generation and difference frequency generation. We had taken here a single monochromatic wave, suppose instead of a single wave if I had  $E$  is equal to  $E_0 \cos \omega_1 t + E_0 \cos \omega_2 t$  and then, we substitute for this  $E$  in the same expression  $\epsilon_0 \chi^2$  into  $E$  square.

We will get we will have  $\cos \omega_1 t$ , we will have  $\cos \omega_1 + \omega_2 t$ , we will have  $\cos \omega_1 - \omega_2 t$  and of course,  $\cos^2 \omega_1$  that is  $\cos 2\omega_1 t$  and  $\cos 2\omega_2 t$ . So, all these terms would come if we substitute this into the expression here.

This expression here tells us that there are two frequencies which are added, it is a some frequency generation one term gives rise to generation of some frequency because, there is a polarization component at the sum of the two frequencies. Similarly, this represents difference frequency and we call this as DFG Difference Frequency Generation, sum frequency generation, and second-harmonic generation.

Similarly, if we substitute the same expression for  $E$  that is  $E_0 \cos \omega t$  in the third term this will give rise to third-harmonic generation, this will also give rise to an intensity dependent refractive index, which is responsible for self-phase modulation and a four-wave mixing that is three waves generating a fourth wave. And the  $\chi^4$  would give rise to fourth harmonic, there are a number of non-linear effects which are possible depending on the dominance of these terms, these higher order non-linear terms.

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So, if we consider  $\chi^{(2)}$  so just if I look at  $\chi^{(2)}$  effect that is  $\chi^{(2)}$  terms then, the three important effects which are shown here that I mentioned second harmonic generation a pump at  $\omega$  so, this is the pump at  $\omega$  interacting with the medium  $\chi^{(2)}$  medium gives rise to the second harmonic  $2\omega$ .

Of course, there will be residual  $\omega$  also which will come so,  $2\omega$  and  $\omega$ . If you have two frequencies input, which are called  $\omega_s$  and  $\omega_i$  or  $\omega_1$  and  $\omega_2$ . Then, we will have at the output  $\omega_p$  which is equal to  $\omega_s + \omega_i$ , or if we call this as  $\omega_1$ , we call this as  $\omega_2$  and denote this as  $\omega_3$  then,  $\omega_3$  will be equal to  $\omega_1 + \omega_2$ . And this is called some frequency generation.

But, if we look at the reverse process that is if we had an input which is  $\omega_3$  here then, an interaction with the medium can result in the generation of the two frequencies  $\omega_1$  and

$\omega_2$ , and this is called optical parametric fluorescence. So, all these effects are possible due to the  $\chi^2$  non-linear effects.

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### Second Harmonic Generation

*Second Harmonic generation (SHG) is the phenomenon wherein an optical beam of frequency ' $2\omega$ ' is generated as a result of the interaction of a high-power laser beam of frequency ' $\omega$ ' with a suitable nonlinear crystal.*

Two Important Requirements for efficient interaction:

1. Phase matching between the interacting waves
2. Good overlap of the interacting waves

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Now, let me pick up just one and highlight the non-linear effect, which is the second harmonic generation. So, second harmonic generation is the phenomenon wherein an optical beam of frequency ' $2\omega$ ' is generated as a result of the interaction of a high-power laser beam of frequency ' $\omega$ ' with a suitable non-linear crystal.

So, there is a non-linear crystal here I will discuss more about this. So, we have a pump a high power pump at frequency  $\omega$ , which gives rise to the second harmonic which is SH at  $2\omega$  and the residual pump. So, this is the residual pump. Because, as we will see that all the pump power cannot be converted to second harmonic and therefore, we will always have residual pump coming out.

There are two important requirements for any non-linear interaction, for efficient interaction. 1st, there must be phase matching between the interacting waves. Phase matching 'phase matching' here refers to the phase velocities of the two waves. The phase velocity of the interacting waves must be equal, if we want to have efficient interaction.

The 2nd one is that there must be good overlap of the interacting waves, what this means is if we have a wave which is at frequency  $\omega_1$   $\psi_1$  and if both of them are propagating in the medium. If they are separated in space for example, if they are separated in space like this,  $\psi_2$  then the overlap the region where they overlap is very small.

On the other hand, if  $\psi_2$  were also travelling along with this, with the same phase velocity then the interaction would be very strong. Because, they are continuously interacting at the same spatial locations and this is what is meant in simple terms by good overlap of the interacting waves. We will discuss this a little bit more.

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### Efficiency of Second Harmonic Generation

Growth of second harmonic field is given by (Plane wave analysis)

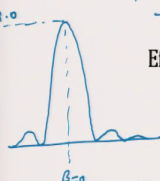
$E_1 \rightarrow \omega$   
 $E_2 \rightarrow 2\omega$

$\rightarrow z$

$$\frac{dE_2}{dz} = -\frac{i\mu_0 \alpha c \omega}{n_2} E_1^2(z) e^{i(\Delta k)z}$$

$n_2 = n(2\omega)$   
 $n_1 = n(\omega)$

$\rightarrow \Delta k = k_2 - 2k_1 = (2\omega/c)(n_2 - n_1)$  is the 'Phase mismatch'



$\beta = 0$

$$\text{Efficiency, } \eta = \frac{P_2}{P_1} = \frac{2c^3 \mu_0^3 \alpha^2 \omega^2}{n_1^2 n_2} z^2 \cdot \left(\frac{P_1}{A}\right) \cdot \left(\frac{\sin \beta}{\beta}\right)^2$$


with  $\beta = \frac{1}{2}(\Delta k)z$

Intensity of pump  
Propagation distance

$P_1 \rightarrow$  Input (pump) power at  $\omega$ ;  $\alpha$  is the nonlinear coefficient;  
 $P_2 \rightarrow$  Output power at  $2\omega$ .

$\alpha = \frac{1}{2} \epsilon_0 \chi^{(2)}$

If  $\Delta k = 0 \Rightarrow$  no phase mismatch



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So, for growth of the harmonic field, if we perform a plane wave analysis, the analysis is beyond the scope of our course. If we perform a plane wave analysis, then we would get an equation which describes the amplitude of the second harmonic  $E_2$  is the amplitude at  $2\omega$  and  $E_1$  is the electric field at  $\omega$ .

Now,  $dE_2/dz$ , they are propagating in the  $z$  direction both of them are propagating in the  $z$  direction, the evolution of the amplitude of the electric field  $E_2$  is the pump which is generating  $E_1$ . The rate of change of  $E_2$  with  $z$  that is the propagation distance is given by a differential equation of this form.

It is a simplified differential equation, where  $\Delta k$  note that, it has an oscillatory term  $e^{i\Delta k z}$ , the power  $i\Delta k z$ ,  $\Delta k$  is  $k_2 - 2k_1$ ,  $k_2$  is the propagation constant at the frequency  $2\omega$ ,  $k_1$  is the propagation constant at the frequency  $\omega$ . So,  $\Delta k$  is  $k_2 - 2k_1$ .

minus  $2k_1$ , which is equal to  $2\omega$  by  $c$ ,  $\omega$  by  $c$  is  $k_0$  that is the propagation constant in the free space into  $n_2$  refractive index.

So,  $n_2$  is the refractive index at  $2\omega$ . Please remember that, refractive index is a function of frequency or wavelength and  $n_1$  is the refractive index so, equal to  $n_1$  of  $\omega$ . So, refractive index at the frequency  $\omega$  so, it is simply  $n$ ,  $n$  of  $2\omega$  and  $n$  of  $\omega$ .

So,  $n$  of  $2\omega$  is designated it is a given medium refractive index  $n$  of the medium at  $2\omega$  frequency is designated as  $n_2$ , refractive index of the medium at  $\omega$  frequency is designated as  $n_1$  and then,  $\Delta k$  the term here is given by this expression. This is called the 'phase mismatch'.

And the efficiency if we calculate the efficiency of second harmonic generation  $P_2$  is the power at  $2\omega$  generated power,  $P_1$  is the power at  $\omega$  that is the pump. Therefore, the conversion efficiency  $\eta$  is  $P_2$  by  $P_1$  this is the input power  $P_1$ , which is given by an expression of this form.

As I mentioned the derivation is beyond the scope of our course, but what we are interested to note is that this efficiency is proportional to  $\alpha^2$ , where  $\alpha$  is given by  $\frac{1}{2}\epsilon_0\chi^{(2)}$ . So, it is proportional to  $\chi^{(2)2}$ . The efficiency is proportional to  $\alpha^2$  means proportional to  $\chi^{(2)2}$ ,  $\chi^{(2)}$  is the non-linear susceptibility of the medium and larger the  $\chi^{(2)}$  larger will be the conversion efficiency, first point.

Second, it is proportional to  $Z^2$   $Z$  is the propagation distance. So, this is because the wave is propagating as indicated here in the  $Z$  direction. So, this is the propagation distance. So, larger the interaction length, larger will be the efficiency. Third  $P_1$  by  $A$ ,  $P_1$  is the pump power that is the input power and  $A$  is the area of cross section.

And therefore, this term here represents intensity of the pump, power per unit area intensity of the pump of the pump. And the fourth term equally important is a sinc square term, that is  $\text{sinc}^2$

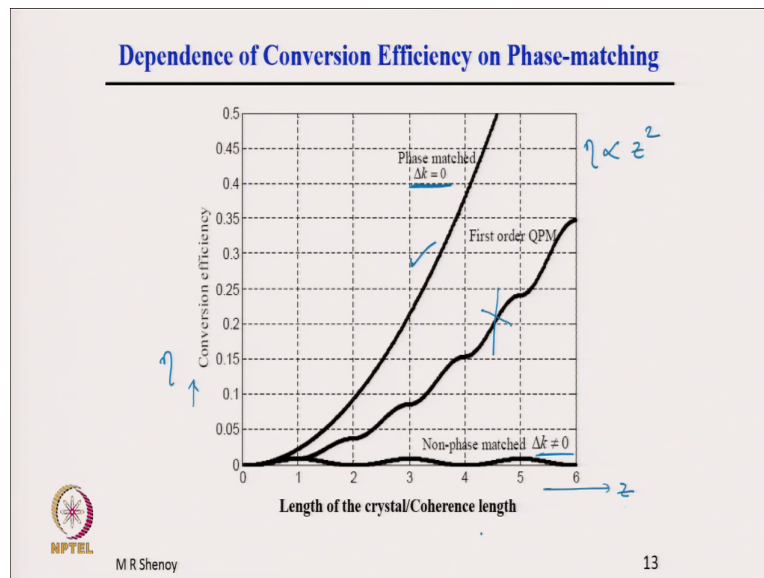
square beta by beta square term if I plot it here so it varies. So, this is sin square beta versus beta and this is at beta is equal to 0 this is maximum.

At beta is equal to 0 this is 1, the sinc square function is 1 at beta equal to 0 and what is beta? Beta is equal to half delta k into z. So, delta k is the expression which is given here so, here is delta k the expression, which is the phase mismatch and beta will be 0 if delta k is equal to 0. That is if delta k is equal to 0, which implies there is no phase mismatch; no phase mismatch which means, the phase velocity at omega is equal to phase velocity at 2 omega no phase mismatch the efficiency will be maximum.

So, the purpose of writing this expression although we have not derived it is to indicate that the non-linear optical efficiency, in this case conversion efficiency that is the fraction of pump power that is getting converted into the second harmonic is proportional to square of the susceptibility alpha square; square of the length of interaction, intensity of the pump which tells you that stronger the pump better will be the efficiency.

And finally, it is maximum when delta k is equal to 0 or when there is no phase mismatch. That is the purpose of giving this expression to recognize that the efficiency of non-linear interaction will be maximum when there is no phase mismatch, or when there is perfect phase matching between the interacting waves, alright.

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Let me discuss this a little bit more and therefore, a graph here shows so, this is the normalized length of the crystal that is normalized length divided by coherence length. So, it is essentially this is  $Z$ , if you plot conversion efficiency as a function of  $Z$  so, this is  $\eta$  then we see that if  $\Delta k$  is equal to 0 the efficiency increases it goes up as square of the distance it is parabolically.

So,  $\eta$  is proportional to  $Z$  square, and if  $\Delta k$  is not equal to 0 forget about this term this curve at the moment. So, this is when  $\Delta k$  is equal to 0 that is perfectly phase matched interaction and if  $\Delta k$  is not equal to 0 then, conversion efficiency will go on oscillating because of the sinc square term.



So, we see that it oscillates, but never builds up. The conversion efficiency does not increase with the length of interaction, but simply oscillates. This clearly shows that phase matching is a very important requirement for efficient non-linear optical interactions.

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### Phase Matching

→ Two methods widely used for 'momentum conservation' or Phase Matching: (discussion w. r. t. SHG)

→ **1. Birefringent phase matching** →  $n_o, n_e$

$o \rightarrow$  ordinary wave  $\vec{k}_o^{(\omega)}$

$e \rightarrow$  extra-ordinary wave  $\vec{k}_e^{(\omega)}$

$\Delta k = 2k_o(n_2 - n_1)$

$\rightarrow 2\vec{k}_o^{(\omega)} = \vec{k}_e^{(2\omega)}$

or  $n_o(\omega) = n_e(2\omega)$

$n_o(2\omega) = n_e(\omega)$   
or  
 $n_o(\omega) = n_e(2\omega)$   
possible

→ **2. Quasi phase-matching (QPM) By using an appropriate Grating**

$n = n(2\omega)$

$n_2 = n(\omega)$

$\vec{k}^{(\omega)} + \vec{k}^{(\omega)} + \vec{K} = \vec{k}^{(2\omega)}$

$= \frac{2\pi}{\Lambda}$

Grating vector

By choosing an appropriate grating period (and hence  $K$ ), phase-matching can be achieved in any material, and at any pump wavelength.

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Now, let us see the phase matching techniques which are used, two most widely used methods for phase matching is birefringent phase matching and quasi phase matching. Birefringent phase matching so here what is written is o this is o, o for ordinary wave. So, this is the propagation constant or propagation vector of the ordinary wave, and e here refers to the extraordinary wave, it is not 0, it is o extraordinary wave.

So, in birefringent phase matching which means, in a birefringent medium a birefringent medium so, birefringent medium is characterized by if we consider a uniaxial medium then, it is characterized by an ordinary refractive index  $n_o$  and an extraordinary refractive index  $n_e$

e. Now, if we go back to the phase mismatch term here. So, we see that  $\Delta k$  is equal to  $2k_0$  into  $n_2$  minus  $n_1$ . So, let me write this again here.

So, note that  $\Delta k$  is equal to  $2k_0$ ,  $k_0$  is in free space that is  $\omega$  by  $c$  into  $n_2$  minus  $n_1$ ,  $n_2$  minus  $n_1$ , what is  $n_2$ ?  $n_2$  is the refractive index at  $2\omega$  and so this is  $n_2$  and  $n_1$  is refractive index at the frequency  $\omega$ . In a normal medium, every medium is a dispersive medium which means the refractive index changes with frequency we can never have  $n_2$  equal to  $n_1$ .

Because, if you take a general medium and if you plot frequency  $\omega$  versus refractive index you see that the refractive index will go on increasing with the increasing  $\omega$ , in a normal dispersive medium or if you plot as a function of wavelength. So, let me show with a different color, if wavelength increases the refractive index decreases like this. For any normal dispersive medium and therefore, we can never have  $n_2$  equal to  $n_1$ . So, this is not possible in any normal medium.

But, if we consider a birefringent medium which is characterized by two refractive indexes  $n_o$  and  $n_e$ . It is possible at some frequency that  $n_o$  of  $2\omega$  is equal to  $n_e$  of  $\omega$  or so,  $n_o$  of  $\omega$  is equal to  $n_e$  of  $2\omega$ . So, this is possible in a uniaxial crystal, birefringent crystal. But it is not always possible at any given frequency, but at certain frequency or by changing the angle we can obtain this condition. So, I want to illustrate this in the next diagram here.

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### Birefringent Phase Matching

**Uniaxial Crystal:**  
 The extraordinary refractive index  $n_e$  depends on the direction of propagation, i. e. angle  $\theta$  between the optic axis and the direction of propagation.

$n_e \equiv n_e(\theta)$

For the chosen direction  $\mathbf{k}$ ,

$n_o^{\omega} = n_e^{2\omega}(\theta_m)$

Normal (index) surfaces for the ordinary and extraordinary rays  
in a negative ( $n_e < n_o$ ) uniaxial crystal.

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So, birefringent phase matching so what is shown here, are index surfaces circles here correspond to the ordinary refractive index  $n_o$ . What it means is, if this is the optic axis then if we propagate in any direction. So, you have what are shown are index surfaces these are called index surfaces. Index surface means, the magnitude the radius here at anywhere.

So, if you are propagating in this direction then the radius gives you the refractive index, if you are propagating in this direction the radius gives you the refractive index, the magnitude of the radius. So, if you plot refractive index as a function of the direction of propagation you get a surface, which is called index surface or normal surface.

Now, the ordinary refractive index is independent of the direction of propagation and therefore, if we plot an index surface it will be a sphere. So, what we have shown here as a circle is in 2d actually it is a sphere, but in 2d it will be a circle. So, the circle indicates that

independent of the direction of propagation the refractive index is the same, but if we plot  $n_e$  then we get either an ellipse like this or an ellipse like this.

So, depending on the direction of propagation the refractive index changes in the medium. So, if we are propagating in this direction this will be the refractive index, if you are propagating in this direction this will be the refractive index, and if we are propagating in this direction this will be the refractive index. If it is an ellipse obviously, it means that if I call this as  $n_o$  then this will be  $n_e$ .

If you are propagating along the optic axis, then it will see a refractive index which is  $n_o$  and if you are propagating along a direction perpendicular to the optic axis then, it will see a refractive index  $n_e$ . So, this is what is shown as index surface. Now, what is important? We can see two circles the first circle here.

So, this is  $n_o$  of  $2\omega$  ordinary index at  $2\omega$  and this circle represents  $n_o$  at  $\omega$   $n_o$  at  $\omega$ . The ellipse represents  $n_e$  at  $2\omega$  and the inner ellipse represents  $n_e$  at  $\omega$ . So, this represents  $n_e$  at  $\omega$  and we see that it is an ellipse which means depending on the direction of propagation refractive index would change.

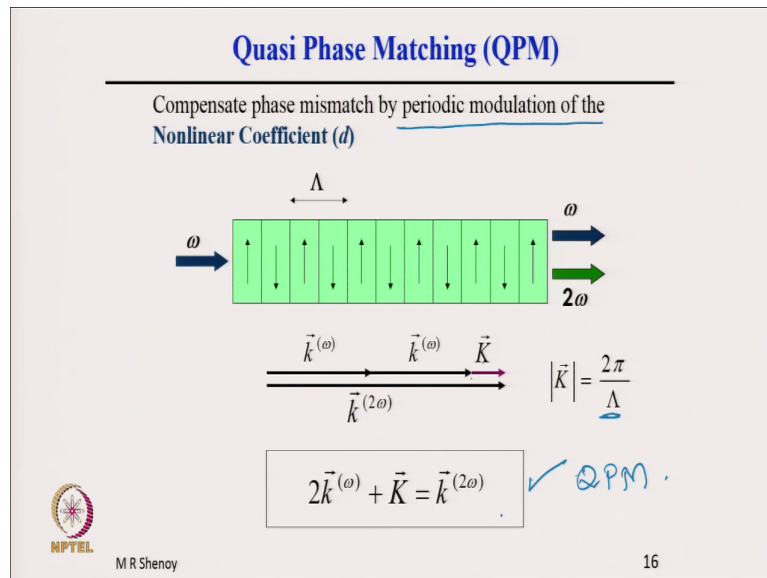
What is important? Important is this point this direction see the direction. And there is a point of intersection, if the wave is propagating in this direction at the point of intersection here; in the point of intersection  $n_e$  of  $2\omega$  is equal to  $n_o$  of  $\omega$ , or we have  $n_o$  of  $\omega$  is equal to  $n_e$  of  $2\omega$ .

Because,  $n_e$  is a function the direction of  $n_e$  is a function of theta that is propagation direction therefore, if we propagate the pump in a specific direction it is possible to achieve phase matching. Phase matching namely the refractive index at the frequency  $\omega$  equal to the refractive index at the frequency of  $2\omega$ .

Of course, in this case at  $\omega$  we will have an ordinary wave whereas; at  $2\omega$  we will have an extraordinary wave. This is what is meant by birefringent phase matching. So,

birefringent phase matching is a technique by using which we can achieve phase matching between the fundamental wave or the omega frequency and the 2 omega frequency alright.

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So, the quasi phase matching very quickly it is an important concept. So, let me discuss here I have skipped this place. The quasi phase matching 2nd one represents here by using an appropriate grating, we can have phase matching at the frequency omega and 2 omega. So, please see here k o of omega plus k o of omega happens to be k e of 2 omega because, that is the phase matching condition.

If they are not equal, if k o of omega and k e are not equal or in a normal medium k o of omega plus k o of omega will always be smaller than k of 2 omega because, n of 2 omega is greater than n at omega. The mismatch, the difference if it can be made up by another quantity this is the propagation vector and therefore, this quantity K is called the grating

vector, what is this grating? We will see. So, the statement is by using an appropriate grating, the magnitude of the grating vector is given by  $2\pi/\lambda$ , where  $\lambda$  is the period.

So, the magnitude of the grating vector is given by  $2\pi/\lambda$  where  $\lambda$  is the periodicity of the grating. And a statement which says which I will illustrate by choosing an appropriate grating period and hence  $K$  because,  $K$  is magnitude is  $2\pi/\lambda$  phase matching can be achieved in any material and at any pump wavelength.

This is a very profound statement we will not have time to go into the details of this, but this is a very important technique by which we can achieve quasi phase matching. So, very quickly illustrated here so, this grating is achieved by periodic modulation of the non-linear coefficient  $d$ , that is by inverting the ferroelectric domains.

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### Second Harmonic Generation (SHG)

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Growth of second harmonic field is given by

$$\frac{dE_2}{dz} = -\frac{i\mu_0\alpha c \omega}{n_2} E_1^2(z)e^{i(\Delta k)z}$$

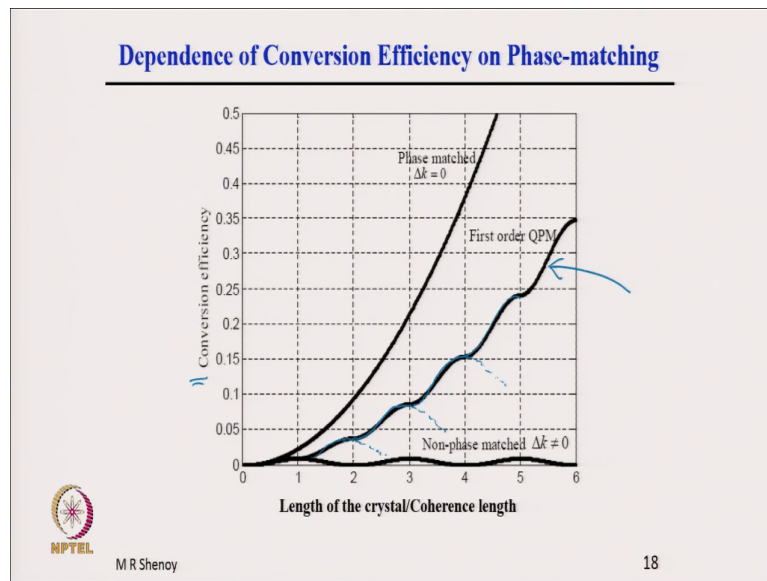
$\Delta k = k_2 - 2k_1 = (2\omega/c)(n_2 - n_1)$  is the 'Phase mismatch'

**QPM - SHG**

Ferroelectric Domains

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### Periodically Poled Lithium Niobate (PPLN)


**LiNbO<sub>3</sub>**

- A ferroelectric crystal
- Very large nonlinear coefficient  $d_{33}$
- Transparent in the 0.4 - 4.5  $\mu\text{m}$  region
- • Periodic Poling: Inverting the ferroelectric domains periodically

*By applying a periodic electric field*

$\Lambda$

$d \ -d \ d \ -d \ d \ -d \ d \ -d$  PPLN crystal

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So, let me show this. So, here if we take lithium niobate here a material which can be periodically poled so, periodic polling refers to inverting the ferroelectric domains periodically. So, this is a ferroelectric crystal, where the ferroelectric domains can be periodically poled by applying an electric field by applying a periodic electric field by applying a periodic electric field.

We can have a grating with period lambda, the moment you have a grating with period lambda. So, this is when you periodically pole the non-linear coefficient  $d$  if it is positive here in this it will be minus  $d$ , here again it is positive and in the next domain it is minus  $d$  and this is possible to achieve. So, it is not a refractive index grating, but it is a grating of periodically inverted non-linear coefficient in the medium, and by using this we can achieve quasi phase matching.

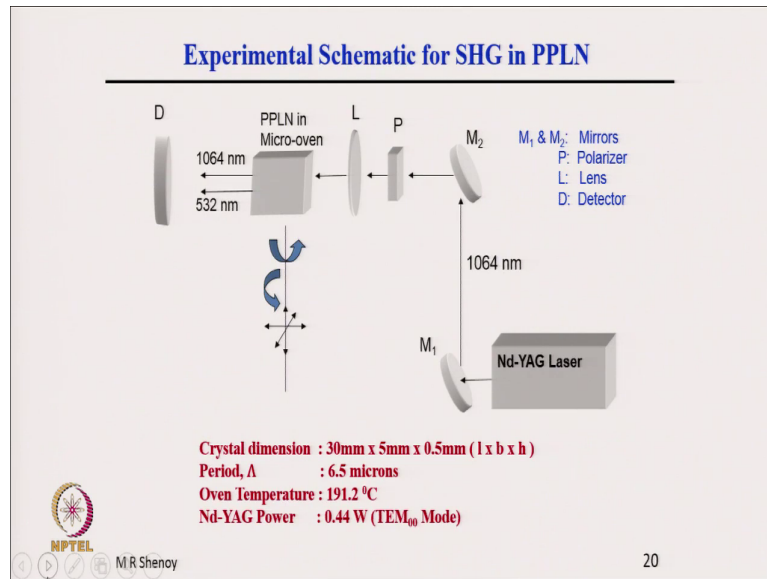


So, let me quickly come back here. So, this is what is illustrated here is the periodic polling and to make up for this phase mismatch. If we choose an appropriate period  $\lambda$  as shown here then, we can have a  $k$  which satisfies this equation and this is called phase matching by quasi phase matching. So, this is quasi phase matching QPM Quasi Phase Matching equation, and the mathematics of evolution is discussed here, but I will skip this.

And the interaction in this case so periodically you invert the domain therefore, the interaction where it would have gone down to 0 oscillating like this, it oscillates and then you flip and then again it starts building up, before it starts coming down you flip again it goes flip, what flip? Flip the non-linear coefficient if we did not flip it would have come down here.

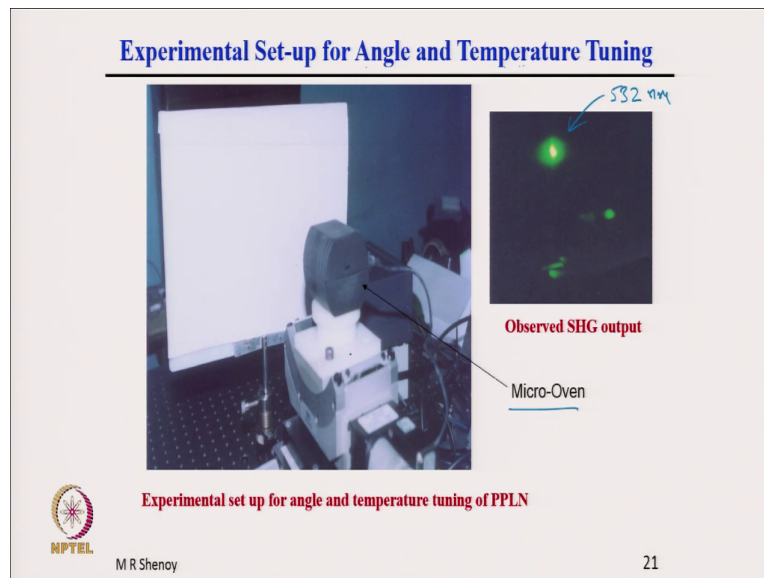
But, if we flip the non-linear coefficient then again the second harmonic starts building up. So, if we did not flip here it would have come down oscillating, but we flip then again it builds up. So, these are multiple sin square curves; multiple sin square increasing leads to an effective increase of the conversion efficiency by quasi phase matching. You may probably have to see more details, but the objective was just to indicate the quasi phase matching technique.

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So, here is an experimental schematic of the second harmonic generation, which we had done in our lab using a Nd-YAG laser here passing through a micro oven, where the crystal is placed and then we achieve second harmonic at the output.

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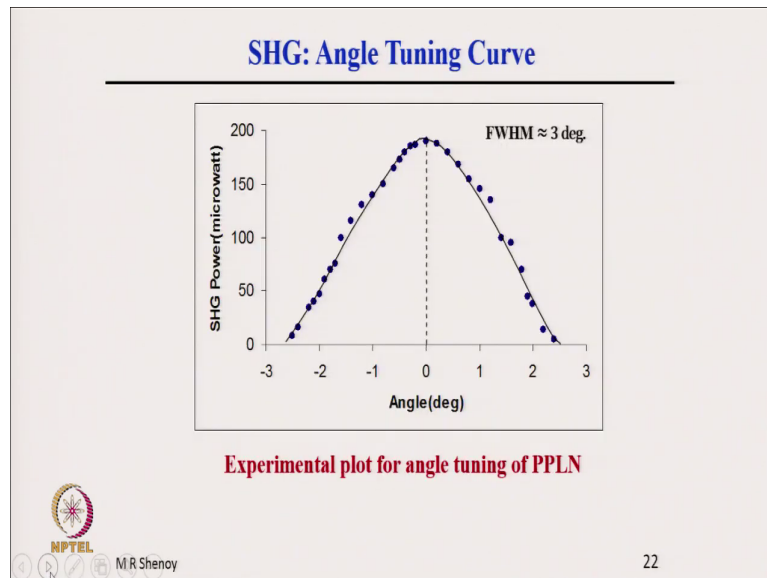
The experimental setup looks like this; it is a photograph of the setup. So, this is the micro oven and we have a small slot here through which the laser light is pumped into the crystal. The crystal is held in this micro oven because the crystal has to be held at a certain temperature for efficient interaction.

And what is shown here is the observed green light at 532 nanometer; 532 nanometer due to pumping by the pump, which is at 1064 nanometer the Nd-YAG laser. The pump is not shown in the diagram, but the arrangement of the micro oven mounted on a goniometric stage this is a goniometric stage and again mounted on a rotational stage for a angle tuning.

So, angle tuning and temperature tuning, temperature tuning the temperature of the crystal can be tuned by changing the temperature of the micro oven. And the angle at which the beam

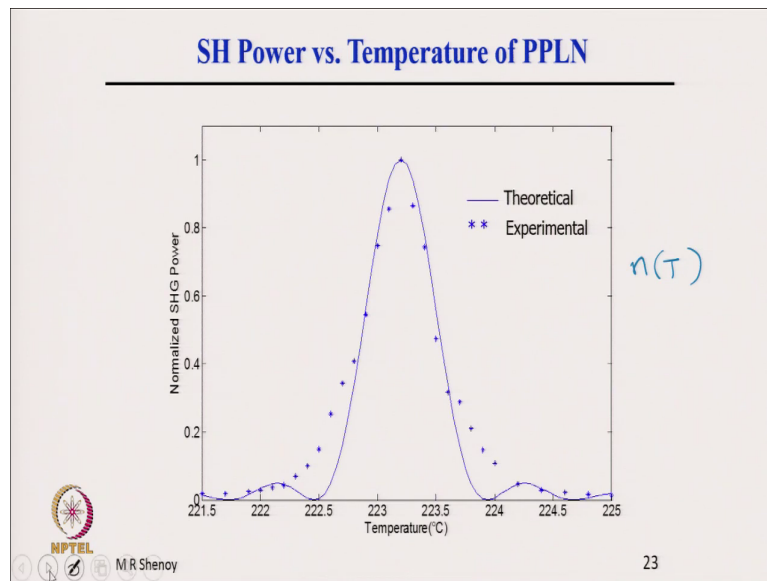
hits the crystal or passes through the crystal can be tuned by adjusting the rotational stage on which the oven is mounted.

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So, these are some angle tuning curves, this is to show that it is very crucial for phase matching to have the specified angle where we get maximum conversion efficiency.

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And similarly, this is a curve which shows the temperature dependence of second harmonic generation. You have to be at the right temperature because, the refractive index  $n$  is a function of temperature. So, by tuning the temperature you can have the refractive index to achieve phase matching, you can obtain the correct refractive index to achieve phase matching that is called temperature tuning in a periodically poled lithium niobate.

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**Nonlinear Optical Materials**

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Widely used materials for NLO:

LiNbO<sub>3</sub>, LiTaO<sub>3</sub>

BBO – Beta Barium Borate ( $\beta$ -BaB<sub>2</sub>O<sub>4</sub>)


ZGP – Zinc Germanium Phosphide

KDP – Potassium Dihydrogen Phosphide (KH<sub>2</sub>PO<sub>4</sub>)

KTP – Potassium Titanyl Phosphate (KTiOPO<sub>4</sub>)

LBO – LiB<sub>3</sub>O<sub>5</sub>

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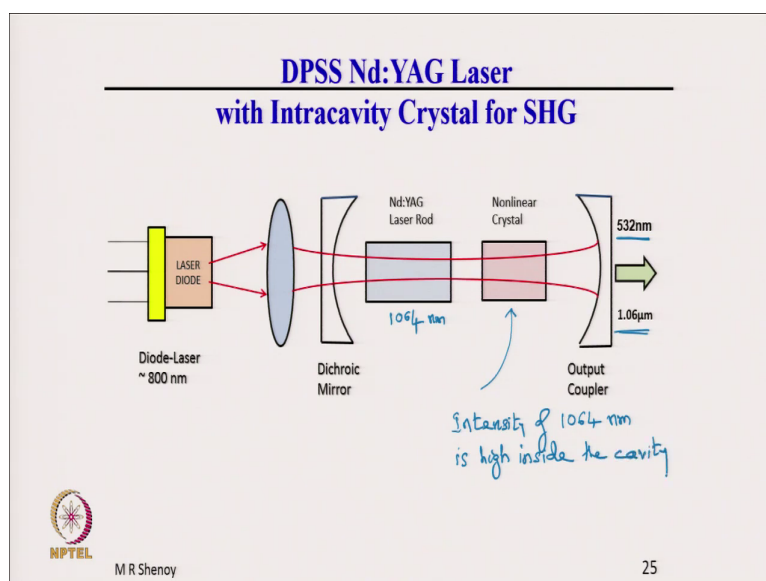


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So, some of the non-linear optical materials, which are widely used Lithium Niobate, Lithium Tantalate, BBO that is Beta Barium Borate, ZGP, KDP, KTP various crystals which are widely used in non-linear optics, there are many many more crystals which are used.

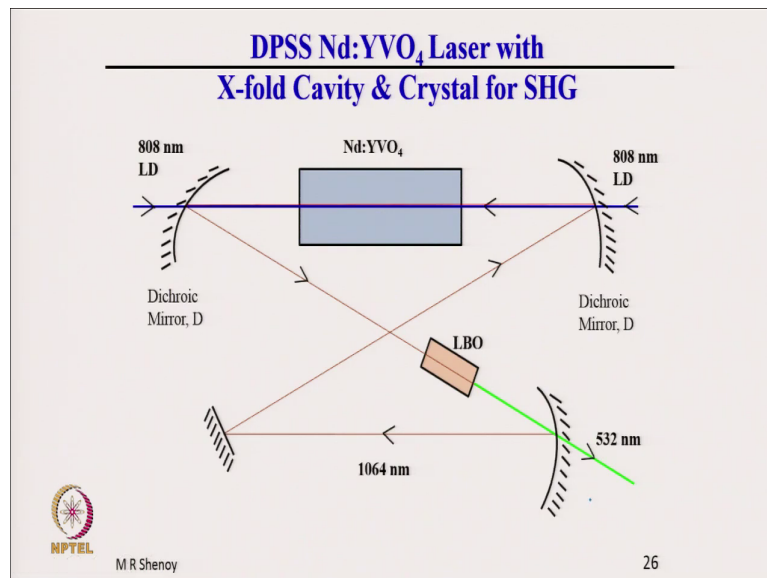
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Now, finally I will just show you the two diagrams which I had shown earlier, one is the DPSS Nd:YAG laser with an intra cavity crystal for second harmonic generation. So, we have the pump diode which is generating 1064. So, this generate the 1064, which pumps the non-linear crystal which gives out 532. So, at the output we have 532 and 1064 nanometer. So, this is the pump and this is the second harmonic.

As I mentioned, why the crystal is placed inside? The crystal is placed inside because, the intensity we have seen that the efficiency of second harmonic generation is proportional to the intensity of the pump beam and the intensity of pump intensity of 1064 nanometer, which is the laser radiation actually is high inside the cavity. That is why we have placed the second harmonic generator, the non-linear crystal inside the cavity rather than outside the cavity; inside the cavity.

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And one more diagram which I had shown is this is a lithium borate, which is also another crystal used for second harmonic generation in the x fold cavity.



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**Summary**

- Basic Nonlinear Optical Effects
- **Role of Laser in Nonlinear Optics**
- Phase Matching Techniques
- **Second Harmonic Generation (SHG)**
- Intra-cavity SHG

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So, to conclude what I had discussed is basic non-linear optical effects, the role of laser in non-linear optics so, this is determined by high intensity possible high intensity and it is a highly monochromatic source because, the phase matching which is an important requirement phase matching critically matching depends on the wavelength depends on the wavelength.

What does this mean? If we do not have a laser, if we have a broadband source then there are many wavelengths, which would pump and which would satisfy phase matching at different wavelengths. And therefore, we need one wavelength to have a scheme of phase matching, which is very efficient and laser meets these two criteria high intensity and phase matching at a specific wavelength.

And as an example I discussed second harmonic generation and we have seen the importance of intra cavity SHG because, the efficiency of second harmonic generation depends on the

intensity of the pump. With that we will stop this non-linear optics is a big subject in it is self, there are large number of non-linear optical effects and it forms indeed a separate course.

Thank you.