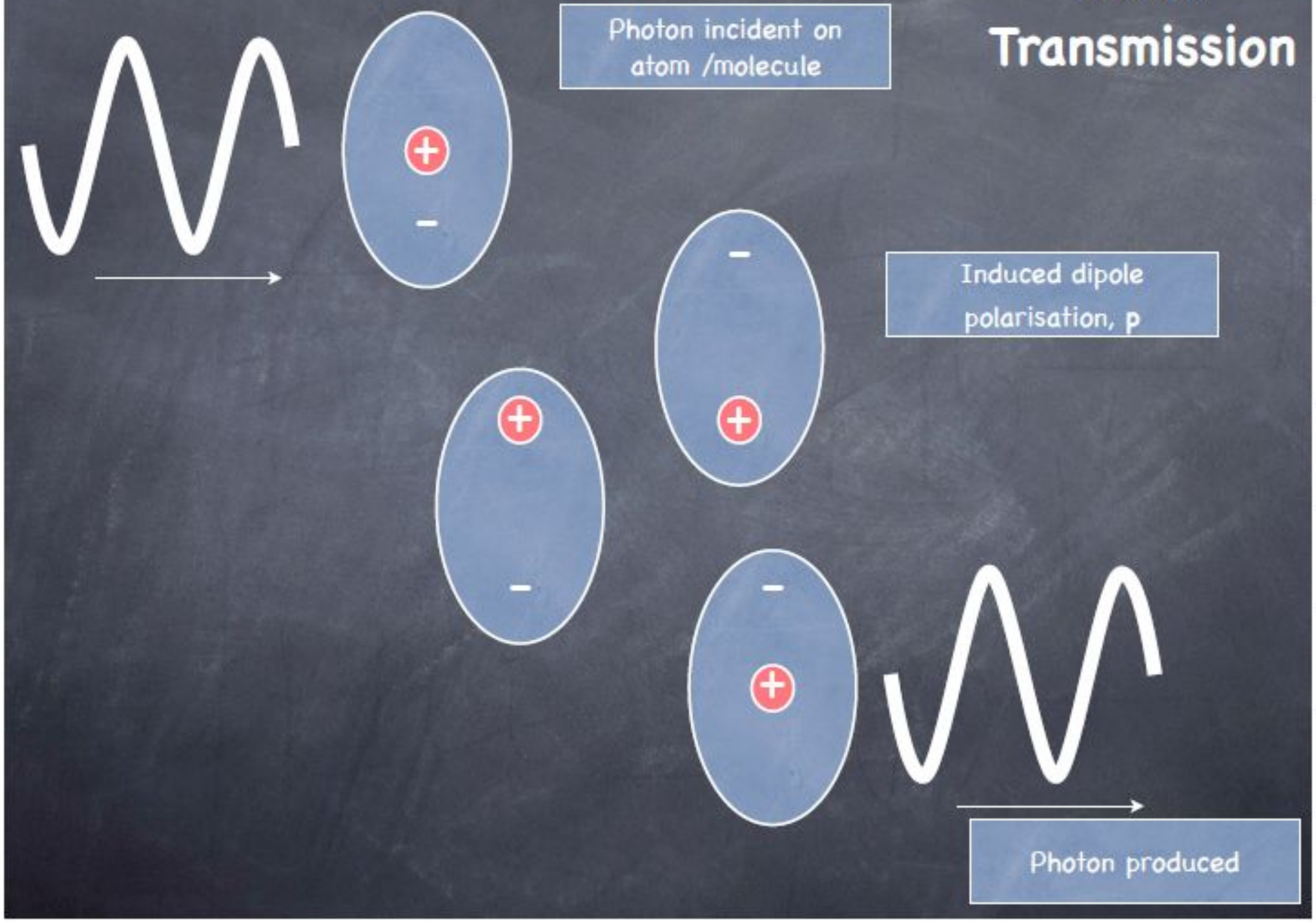
Нелинейные оптические явления

- Первый вопрос, мы должны спросить - каким образом свет проходит через среду?
  Чтобы ответить на этот вопрос, мы должны думать о взаимодействие света с атомами и молекулами. Как вы хорошо осведомлены, свет является электромагнитыми колебаниями.
  Что происходит, когда эта волна попадает на атом или молекулу?
  Один вариант заключается в том, что Фотон может быть поглощен. Возбужденный атом затем релаксирует через фононных процессов на основе в этом случае объект нагревается.
  Другой вариант в том, что Фотон поглощается, а затем излучение проходит в другой длина волны флуоресценции.
  Чаще всего, входящие волна индуцирует колеблющегося диполя электронов молекулы, в результате происходит повторная эмиссия на той же длине волны.

Обратите внимание, что чем ближе к линии поглощения материала, тем дольше задержка переизлучения - (вспомним, о показателе преломления, который существенно возрастает вблизи линий поглощения, т.е. резонансов).
Соответственно прохождение света замедляется через посредство увеличение показателя преломления.

Linear Transmission

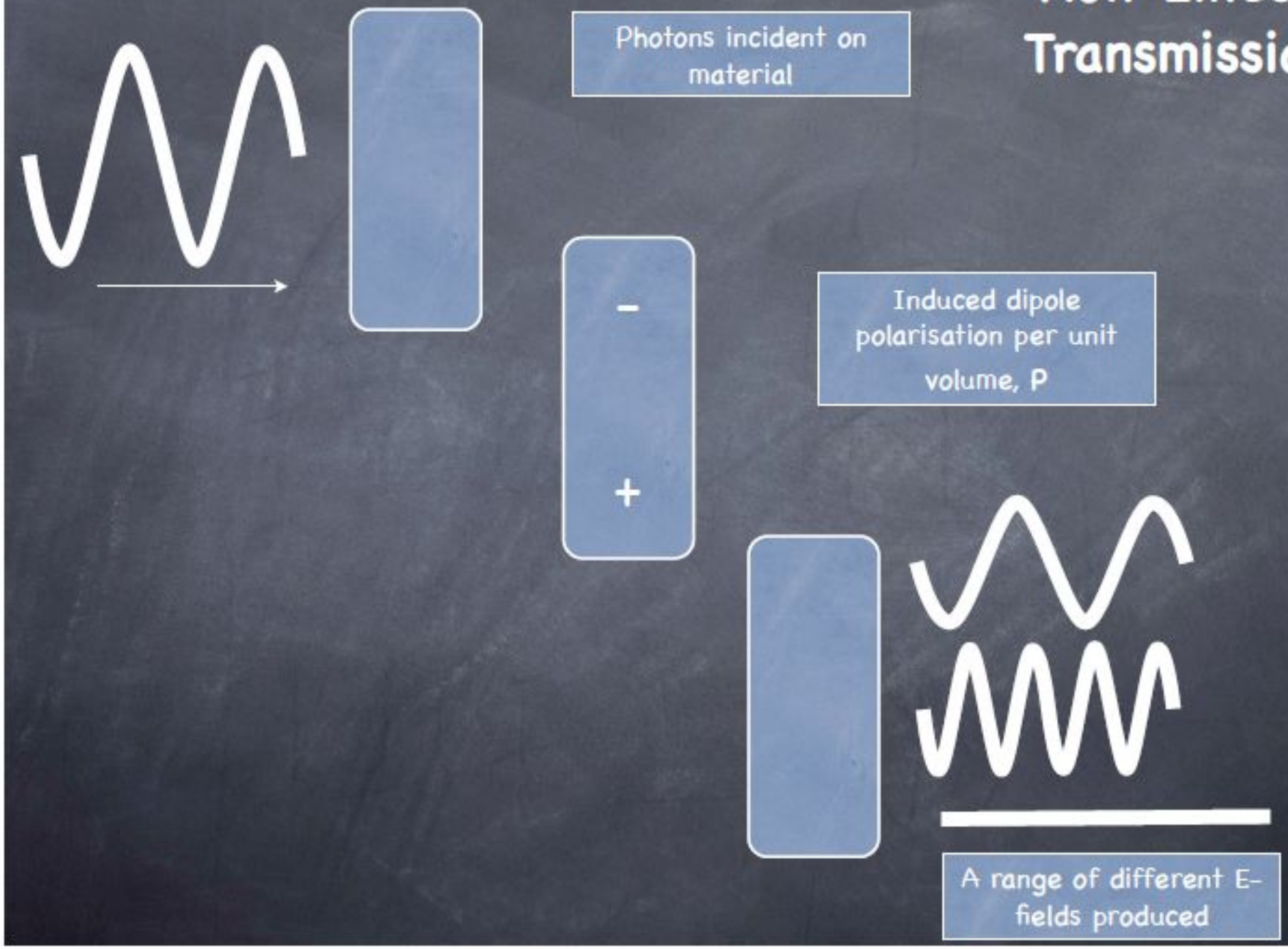


Photon incident on atom /molecule

Induced dipole polarisation, p

Photon produced

Non-Linear Transmission



Photons incident on material

Induced dipole polarisation per unit volume, P

A range of different E-fields produced

Trigonometric Identities

$\sin^2\theta + \cos^2\theta = 1$ $\cos^2\theta = \frac{1}{2}(1 + \cos 2\theta)$

$$\cos A \cos B = \frac{\cos(A + B) + \cos(A - B)}{2}$$

$\cos(-\theta) = \cos(\theta)$

Mathematical formalisation.

For the sake of convenience we will define our light wave as:

$\mathbf{E}(t) = E_{\omega}\cos(\omega t)$

Note here that ω is the angular frequency = $2\pi f$ – we will use the words frequency and angular frequency interchangeably from here on in.

When this wave interacts with an atom / molecule, we obtain **an induced dipole polarisation moment**: $\underline{p}_i$.

We can then write the induced dipole moment per unit volume as:

$$\underline{P} = \sum_i \underline{p}_i$$

Under normal circumstances we can write:

$$\underline{P} = \varepsilon_0 \chi \mathbf{E}$$

ε_0 is the free-space permittivity

χ is the polarizability or dielectric susceptibility

How do we obtain the generated field? Remember back to Maxwell's equations for a dielectric material:

Electric displacement $\underline{D}$:

$$\underline{D} = \varepsilon_0\underline{E} + \underline{P}$$

$$= \varepsilon_0\underline{E} + \varepsilon_0\chi\underline{E}$$

$$= \varepsilon_0(1+\chi)\underline{E}$$

$$= \varepsilon_0\varepsilon_r\underline{E}$$

Where ε_r is the relative permitivity of the medium.

As the light intensity increases, this *linear* model begins to breakdown, instead we now use a power series to model the dipole response:

$$\underline{P} = \varepsilon_0 \left(\chi^{(1)}\mathbf{E} + \chi^{(2)}\mathbf{E}^2 + \chi^{(3)}\mathbf{E}^3 + \ldots\ldots\right)$$

So what does this mean?

Let's imagine our wave interacting with only the first 2 terms of this equation:

$$\underline{P} = \varepsilon_0(\chi^{(1)}E_\omega \cos(\omega t) + \chi^{(2)} \cos^2(\omega t))$$

Key importance here is to know and remember trigonometric identities:

$\cos^2\theta = ½ (1 + \cos 2\theta)$ **You must known this!**

So we can now write:

$\underline{P} = \varepsilon_0(\chi^{(1)}E_\omega \cos(\omega t) + \chi^{(2)}E_\omega{}^2(½ + ½ \cos 2\omega t))$

So now you can see we have 3 terms involved in the induced dipole polarisation:

$½ \chi^{(2)}E_\omega{}^2$ – DC non- oscillating term

$\chi^{(1)}E_\omega \cos(\omega t)$ - Term oscillating at frequency ω (linear propagation)

$½\chi^{(2)}E_\omega{}^2 \cos(2\omega t)$ - Term oscillating at frequency 2ω (non-linear generation)

So our beam, as well as propagating through the medium as before also begins to generate radiation at a frequency of 2ω (wavelength = $\lambda/2$.) This behaviour is known as second harmonic generation and is a key result in non-linear optics. Note that this result increases with E_ω^2 – this is where the expression NON-LINEAR comes from.

We will come back and study this behaviour further as the course goes on.

Let's now turn our attention briefly to $\chi^{(n)}$. $\chi^{(n)}$ is, in general a tensor of rank n+1. A tensor is a mathematical formalism that allows a quantity to be expressed as a multi-element array. A tensor of rank 0 is familiar to us as a scalar, a tensor of rank 1 as a vector – things get more complicated.

This type of notation is required as we have to think about the environment within which the induced dipole polarisation is being formed. Using tensor notation allows us to take the spatial environment into account and this leads to some important conclusions.

The material type now becomes very important. Think about refractive index. In a glass the refractive index IS NOT a function of the direction of propagation (it is a function of λ, material type though.) Why is this?

A glass is an ***isotropic*** material – this means that whatever direction you go in, it appears the same! In this case, the refractive index is a scalar quantity.

Many crystals do not behave like this – the direction of propagation matters – crystals have different symmetries and thus certain material properties change depending on direction.

As an example, we'll consider conductivity σ and Ohm's law.

Let's consider an electric field $\underline{E} = [E_x, E_y, E_z]$ where $E_{x(y,z)}$ are the components in the x, y, z directions respectively.

Now applying Ohm's law: $\underline{j} = \sigma \underline{E}$

In the isotropic case: $j_x = \sigma_x E_x$, $j_y = \sigma_y E_y$ and $j_z = \sigma_z E_z$

This then is simple, vector-like behaviour so σ can be represented as a 1st rank tensor (ie. a vector.)

In the non-isotropic case we also have to consider the coupling between the different axes:

$j_x = \sigma_{xx}E_x + \sigma_{xy}E_y + \sigma_{xz}E_z$
$j_y = \sigma_{yx}E_x + \sigma_{yy}E_y + \sigma_{yz}E_z$
$j_z = \sigma_{zx}E_x + \sigma_{zy}E_y + \sigma_{zz}E_z$

In this case, σ = a tensor of 2nd rank. We can simplify the way that we write this by using the Einstein convention:

$$j_i = \sum_{j=1}^{3} \sigma_{ij} E_j$$

Or more simply still:

$\mathbf{j_l = \sigma_{ij}E_j}$ Where the repeated indicies indicate the variables to be summed over.

So a second rank tensor has 9 terms except that through symmetry argeuments, $\sigma_{xy} = \sigma_{yx}$ etc, so in fact there are only 6 independent terms.

Our example here was for conductivity, but $\chi^{(n)}$ also behaves in an identical way. It's properties are STRONGLY dependent on the material through which the light is propagating. For example in glass, or for that matter any isotropic material, $\chi^{(2)}$ is zero meaning second harmonic generation is impossible!

Alternative Notation for Non-Linear optics

We can write the second order non-linear susceptibility:

$$\underline{P}_j = \varepsilon_0 \chi^{(2)}_{ijk} \underline{E}_j \underline{E}_k \text{, i,j,k = 1,2,3}$$

Now the tensor $\chi^{(2)}{}_{ijk}$ is a tensor with 27 coefficients, but taking into account symmetry leaves us with a a two dimensional 3 x 6 tensor. This is the ***d*** tensor, with coefficients from d_{11} to d_{36}.

Using the d-tensor notation allows us to rewrite the second order non-linear polarisation as:

$$\underline{P} = \varepsilon_0 d \underline{E}\,\underline{E}$$

In fact most crystals only have a few non-zero d coefficients giving us the preferred directions for non-linear generation. Often for a particular direction, we can reduce d to a scalar quantity d_{eff}, the effective non-linear coefficient for a particular direction of propagation for that material.

Генерация 2-й гармоники нелинейность второго порядка

$P = \varepsilon 0 \ (\chi(1)\mathbf{E} + \chi(2)\mathbf{E}2 + \chi(3)\mathbf{E}3 + \ldots\ldots)$

$D = \varepsilon 0E + P$

So intense radiation incident on a particular material can cause ***nonlinear***

terms to be generated. For the next couple of lectures, we are

going to examine the behaviour of non-linear optics based on the $\chi(2)$ tensor – these are called *second order effects*.

E(t) = Eωcos(ωt) into the expression for P gave rise to a number of terms:

S χ(2)Eω

2 – DC non- oscillating term

χ(1)Eωcos(ωt) - Term oscillating at frequency ω (linear propagation)

Sχ(2)Eω

2 cos(2ωt) - Term oscillating at frequency 2ω (non-linear generation)

Remember also that $\chi(2)$ is a tensor and that the value changes depending of the direction of propagation in the material. In isotropic (iso – the same, tropic - in space) materials (eg. glasses, liquids etc) $\chi(2) = 0$ throughout the material so second harmonic generation is impossible. In the case of anisotropic materials this is not the case and the $\chi(2)$ tensor has some non-zero elements resulting in second harmonic generation.

Now we need to consider how the second harmonic radiation is

generated as we travel along the length of our crystal.

Now let's rewrite our wave propagating in the +z direction:

$E(t,z) = E\omega\cos(\omega t - k\omega z)$

Can easily show that the second harmonic wave will have the form:

$E_{2\omega\ \mathrm{gen}}(t,z) = \mathrm{const}\ \cos(2\omega t - 2k_{\omega}z)$

Where kω is the absolute value wave propagation vector:

$k\omega = 2\pi n_{\omega} / \lambda$ (nω is the refractive index, n(ω))

$k\omega = \omega n_{\omega} / c$ (c is the speed of light in a vacuum)

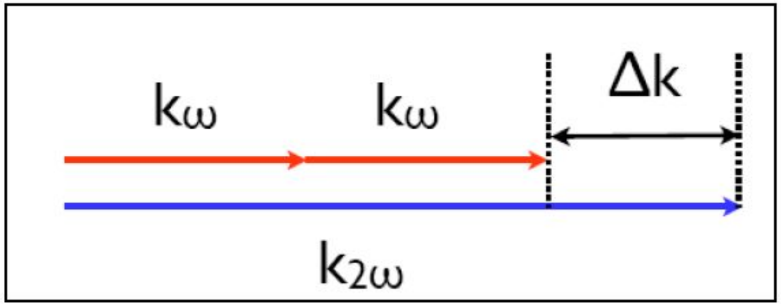
What about the propagating second harmonic wave?

$E_{2\omega\ \mathrm{prop}}(t,z) = \mathrm{const}\ \cos(2\omega t - k_{2\omega} z)$

$k_{2\omega} = 2\omega n_{2\omega}/c$

Key point – in general $\mathbf{n_{2\omega} \neq n_{\omega}}$ this means that there is a PHASE MISMATCH (Δk) between generated and propagating second harmonic!

$\Delta\mathbf{k} = \mathbf{k}_{2\omega} - 2\mathbf{k}_{\omega} = \frac{4\pi}{\lambda}\left[n_{2\omega} - n_{\omega}\right]$ where λ is the fundamental wavelength



What does this mean?

Can show that (see Koechner pp.592-594 if you're interested) :

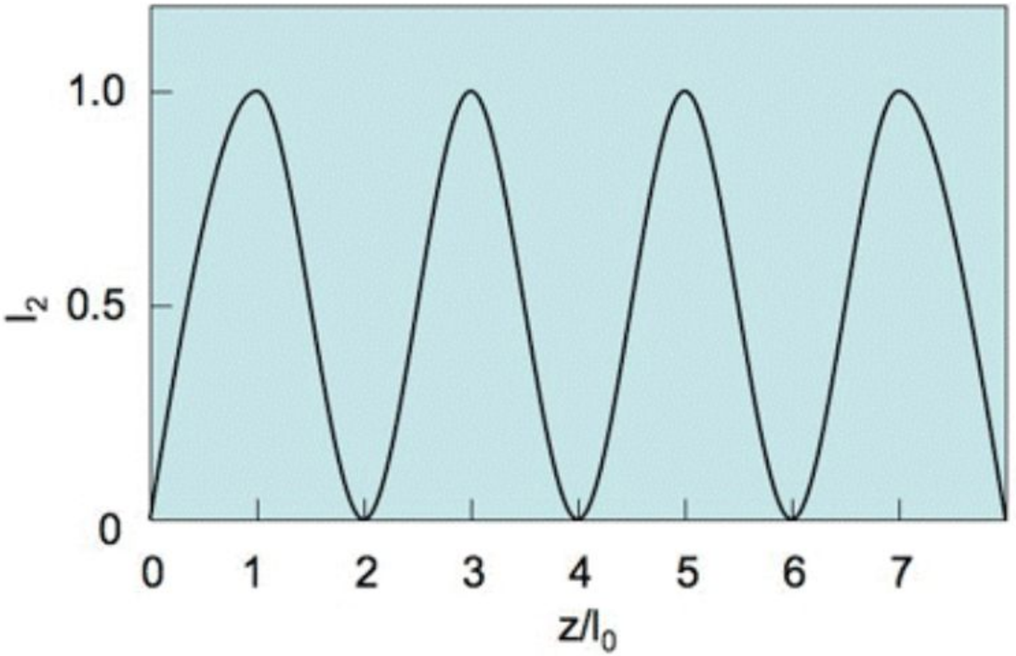
$$I_{2\omega}(z) \propto I_{\omega}^{2}(0)\left(\frac{\sin\frac{\Delta k\, z}{2}}{\frac{\Delta k}{2}}\right)^{2}$$

$I_{2\omega}(z)$ = second harmonic intensity at z
$I_{\omega}(0)$ = incident intensity of fundamental

This function has a maximum where $\Delta k.\mathbf{z} = 0$ and a 0 where $\Delta k.\mathbf{z} = 2n\pi$. This shows that for the most efficient SHG $\mathbf{\Delta k=0}$. This condition is known as ***phase matching*** – ie. a constant phase relationship is maintained between the generated and propagating waves.

Phase matching – constant phase relationship maintained between waves. Ideally for SHG $k_2 = 2k_1$.

In general though, $\Delta k \neq 0$. What does this imply for second harmonic generation as a function of $\mathbf{z}$?



So, the intensity of SHG varies as a function **z** between a maximum (at **z** = rl_c r- odd integer) and 0 (at **z**=sl_c s- even integer). l_c is the coherence length.

Coherence length – the effective interaction length for SHG when $\Delta k \neq 0$.

So, the maximum useful length for the SHG crystal = l_c when no phase matching is present.

For our maximum $I_{2\omega}$, we know that: $\sin \Delta k l_c/2 = 1$

So $\Delta k l_c/2 = \pi/2$

Therefore $l_c = \pi / \Delta k$

Example:

$\lambda = 800nm,\ n_\omega = 1.4,\ n_{2\omega} = 1.5$

$l_c = 800\text{x}10^{-9} / 4(1.5-1.4) = 2 \text{ x } 10^{-6}\text{m}$ (2μm) !!!!!

$\lambda = 800nm,\ n_\omega = 1.4,\ n_{2\omega} = 1.401$

$l_c = 800 \text{ x } 10^{-9} / 0.004 = 0.2\text{mm}$

So the typical coherence length is VERY short – does this make SHG impossible?

No – we have to think up clever ways to do phase matching. One key way to do this is to make $\Delta k=0$ through using birefringence properties of materials.

The Efficiency of SHG

In a simple scenario where we neglect any depletion of the fundamental beam, we can express the generated second harmonic intensity $I_{2\omega}$ as:

$$I_{2\omega} = C^2 L^2 I_\omega^2 \frac{\sin^2\left(\Delta k L/2\right)}{\left(\Delta k L/2\right)^2}$$

Where L is the crystal length, I_ω = fundamental intensity and C is a constant:

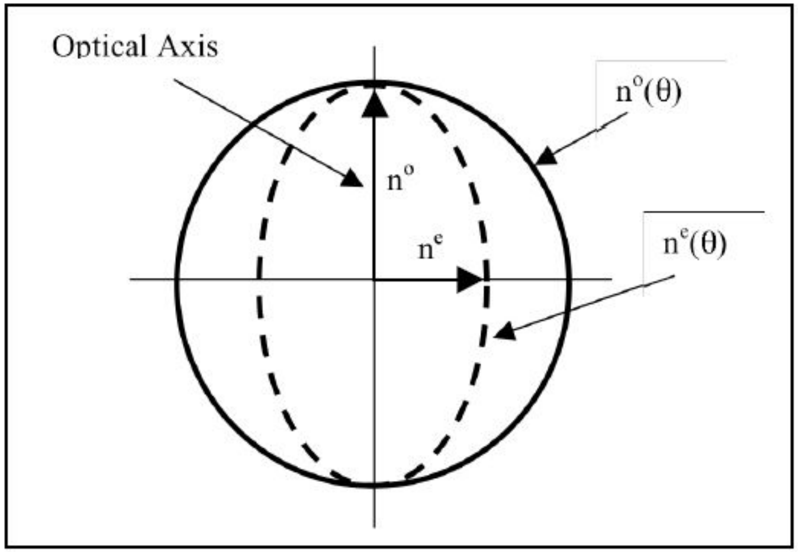
$$C^2 = \frac{8\pi^2 d_{eff}^2}{\varepsilon_0 c \lambda_\omega^2 n_\omega^3}$$

Laser focussed to 10um spot size, d_{eff} = 4.7pm/V, n=2.2, λ=1.06um, L=5mm. Calculate maximum conversion efficiencies. (ie assuming perfect phasematching)

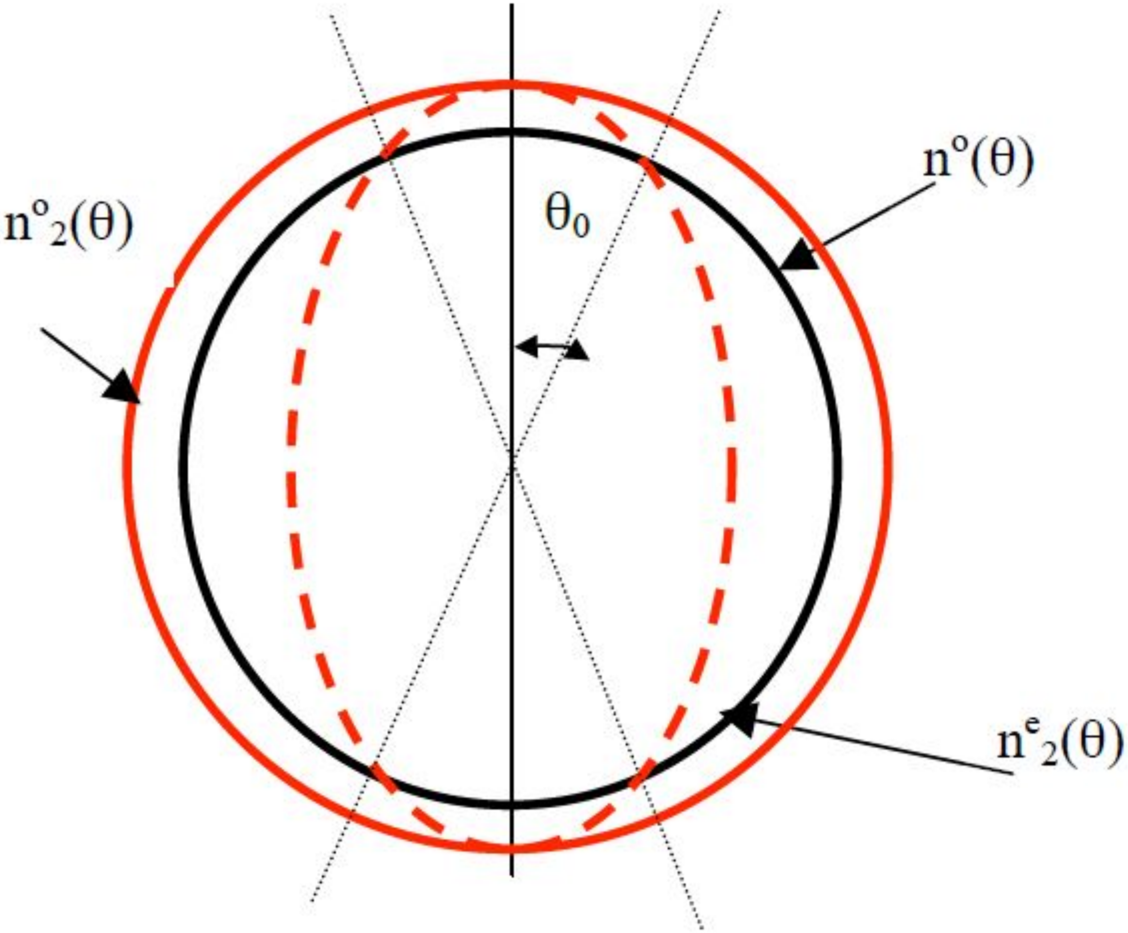
Power	**Intensity**	η_0
1mW	3MW/m^2	4x10^{-6}
1W	3GW/m^2	4x10^{-3}
100W	300GW/m^2	0.4

Phase Matching was of key importance for non-linear optics. Remember that phase matching occurs when a constant phase relationship is maintained between the generated and propagating waves. In general, this does not occur, so a repeated build up and decline of radiation is observed with a characteristic length given by the coherence length l0. In order to make phase matching occur, we need to arrange a circumstance where $n_{2\omega} = n_{\omega}$. In order to do this we can use ***birefringent*** materials. Birefringence is a property of certain materials where different polarisations have different refractive indicies. You may be familiar with the c oncept of double refraction in calcite – this is an example of birefringence where the two rays formed have different polarisations – these are the ordinary, o-ray and the extraordinary, e-ray.

We'll consider the case of the negative uniaxial crystal – $n_o > n_e$. There is a special direction where $n_o = n_e$ – this is called the optical axis. Making a plot of n_e and n_o as a function of angle generates the *refractive index ellipsoids*.



So, the crystal has different refractive indices for the ordina
extraordinary ray. Now let's expand this picture to include the
harmonic index ellipses: n^o_2 and n^e_2:



We can see that there's an angle, θ_0 where the ordinary polarised fundamental and the extraordinary polarised second harmonic have the same refractive index. This implies that when the radiation propagates in the crystal at angle θ_0 relative to the optical axis, perfect phase matching is possible. This technique is sometimes known as ***birefringent phase matching – BPM.***

So in this case:

$n^o(\theta) + n^o(\theta) = 2n^e{}_2(\theta)$ – This is known as ***Type I phase matching.***

There are a range of other non-linear processes that can take place and requiring phasematching and sometimes the following condition is satisfied:

$n^0(\theta) + n^e(\theta) = 2n^e{}_2(\theta)$ – This is known as ***Type II phase matching***.

In order to tune for different wavelengths of operation, we can simple change the angle θ_0 – this gives rise to so-called ***angle tuning***. This technique is very sensitive dependent on the angle of propagation – get the angle only a little bit wrong and no phase matching occurs. This type of phase matching is also therefore known as ***CRITICAL PHASE MATCHING***.

Problems:

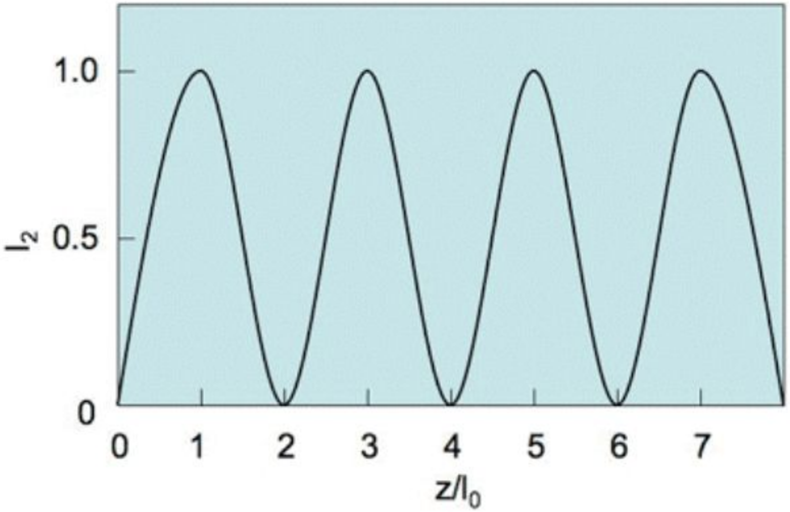
1. Limited range of uni- and bi-axial crystals.
2. Critical dependence on angle.
3. Walk-off occurs between generating beam and second harmonic limiting interaction length.
4. χ^2 varies with angle (it's a tensor remember!) giving variable efficiency with wavelength.

Non-critical phase matching (NCPM)

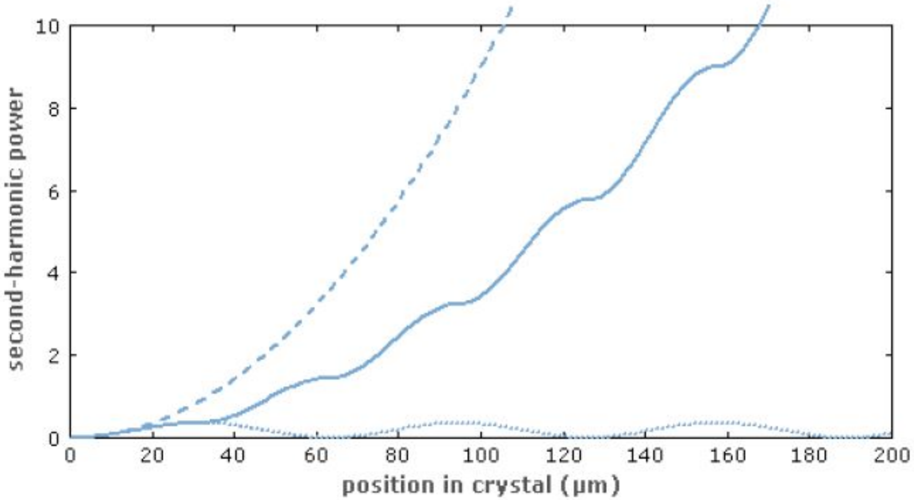
In order to circumvent some of these problems, another approach, non-critical phasematching can sometimes be adopted. In this case, the beams propagate down one of the axes of the non-linear crystal and the temperature of the crystal is adjusted. The propagation direction is normally at 90° to the optical axis, so this techniques is also sometimes known as 90° phase matching. Often, the n^e is much more temperature sensitive than n^o meaning a phase matching condition can be set up.

Quasi Phase Matching (QPM)

But what happens if your material is not birefringent (eg. GaAs) or has a large non-linear coefficient in a direction in which it's impossible to do BPM or NCPM (eg. $LiNbO_3$)? In this case we can physically engineer the material itself to perform phase matching. From before:



Now what would happen if we were to change the sign of the phase every coherence length? Instead of seeing a periodic reduction in non-linear signal, it would continue to grow – this is the basis of quasi-phase matching. Imagine that we do this in a periodic way with a period given by Λ_g, where Λ_g is the period of the grating (= $2l_c$.)



(diagram from http://www.rp-photonics.com/quasi_phase_matching.html)

In the frequency domain, we can represent the grating wave vector:

$k_g = 2\pi / \Lambda_g$

And the phase matching relationship becomes:

$\Delta k = 2\mathbf{k}_{\omega} - \mathbf{k}_{2\omega} - \mathbf{k}_g$

Which allows us to write:

$$\frac{2\pi}{\Lambda_g} = \frac{4\pi}{\lambda}\left[n_\omega - n_{2\omega}\right]$$

Therefore:

$$\Lambda_g = \frac{\lambda_\omega}{2(n_\omega - n_{2\omega})}$$

Example: LiNbO$_3$

For BPM, we are forced to use a direction which has a non-linear coefficient = 4.3pm/V, using QPM allows us to access a coefficient = 27pm/V giving a large enhancement in non-linear optical efficiency.

Let's calculate the grating period required – use a Sellemeir Equation:

$$\left(n^o\right)^2 = 4.9048 + \frac{0.11768}{\lambda^2 - 0.04750} - 0.027169\,\lambda^2$$

Note that for this equation λ is in μm!

λ=1.064μm $\Rightarrow$ n = 2.2364
λ=0.532μm $\Rightarrow$ n = 2.3231

$\Rightarrow$ Grating period = 6.1μm.

Fabrication of QPM structures is complex and has developed in the last 15 years. The simplest approach is to take very thin slices of material and re-orientate them every coherence length. This has done with GaAs where up to 50 layers of GaAs slices have been formed.

Where we want to use a 'standard' non-linear crystal, a different approach is taken. Here we use crystals that are '*ferroelectric*.' Ferroelectric crystals can be given a permanent electrical polarization by the application of electricity. This process is known as '***periodic poling.***' An example of such a crystal is $LiNbO_3$ which has a high non-linear coefficient in a direction that is impossible to access using BPM. To do this, we pattern electrodes onto the top surface of the crystal and apply a sophisticated electric field. This causes a permanent change giving us a 'domain engineered' structure. The electric field required for this in LN is of the order of 21kV/mm !

Advantages of QPM

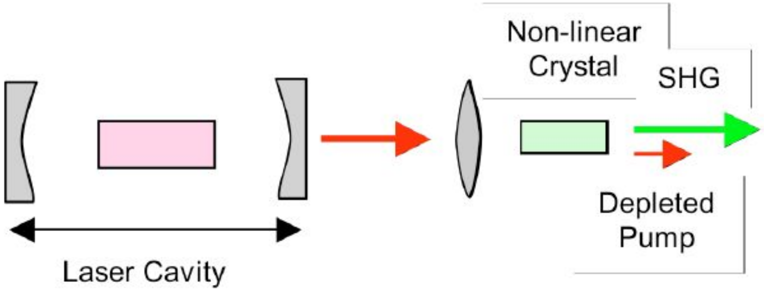
Allows phase matching in non-birefringent materials.
Can access directions with high non-linear coefficients.
Permits non-critical phasematching.
Can be tunable for a range of applications.
Gives direct control over the non-linear materials.

Disadvantages of QPM.

Limited range of materials for e-field poling.
Cost and complexity of fabrication.
Limited widths of grating period (<4μm is almost impossible.)

Practical implementations of non-linear optics.

The simplest implementation is the placing a crystal in the ***extra-cavity configuration***:

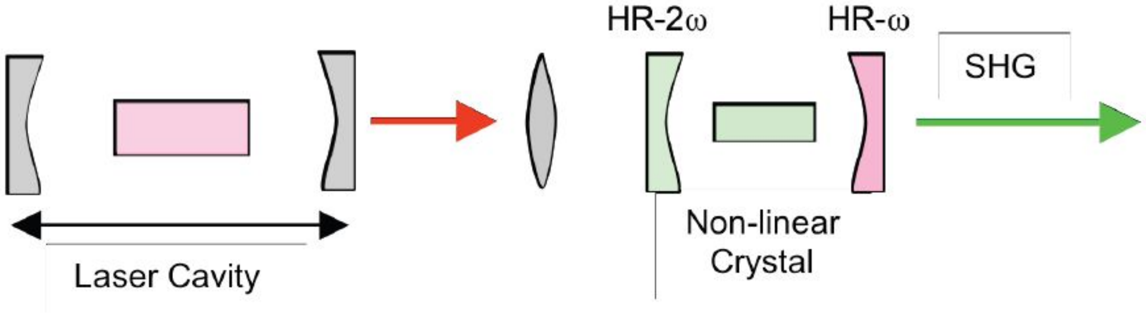


This implementation was first used to demonstrate NLO in the early 1960's. However in this implementation, the efficiency is generally rather low as the incident electric field on the crystal is not high, however using short pulses with high peak powers can work very successfully in this arrangement.

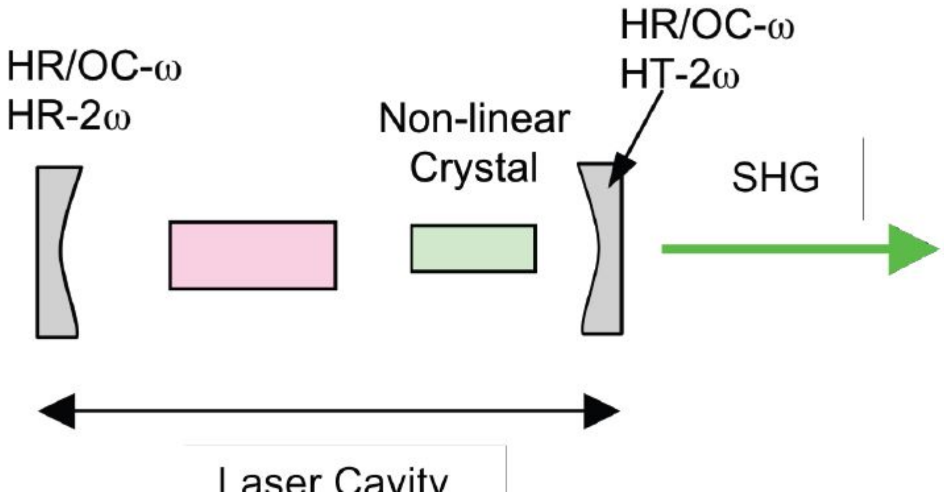
Eg. A Ti:Sapphire laser generates 100fs pulses at 100MHz with a CW average power of 1W – what is the peak power?

So: Pulse energy = $1/100\times10^{6} = 1\times10^{-8}$ J/pulse
$\Rightarrow P_{peak} = 10^{-8}/100\times10^{-15}$ = 100kW! – very handy for NLO!

A refinement on this design can also be undertaken to improve pump conversion to second harmonic:



A final refinement that is often used with CW lasers is to place the non-linear crystal within the laser cavity – the so-called *intra-cavity* configuration.



The key benefit is that the non-linear crystal is now exposed to the circulating power within the laser cavity. ie. A laser with a 1% output coupling that emits 1W of power has a circulating power within the cavity of 100W! This offers a clear enhancement for NLO. This approach can also give very compact systems (think about green laser pointers.)

The disadvantages of intracavity NLO:

- Cavity alignment becomes mores sensitive.
- Mirror coatings become more expensive
- NLO crystal quality must be good to avoid laser losses.
- Cavity design must be changed to incorporate crystal.
- Angle tuning can be difficult.

In general these cavity configurations can be used for all non-linear processes, not just SHG.

What happens if we can to generate 3ω? We could use χ^3 as we will see later, but this is often not very efficient – instead we can carefully cascade two χ^2 processes – second harmonic generation and sum frequency mixing.

Sum frequency mixing.

Now lets imagine a situation where we have two beams incident on our non-linear crystal:

$E = E_1+E_2 =E_{\omega 1}\cos(\omega_1 t)+E_2\cos(\omega_2 t)$

Let's examine the behaviour of the $\chi^{(2)}$ component of the induced dipole polarisation:

$P_{\chi(2)} = \text{const}\ \chi^{(2)} E^2$

$= \text{const}\ \chi^{(2)}\ (E_{\omega 1}\cos(\omega_1 t)+E_{\omega 2}\cos(\omega_2 t))(E_{\omega 1}\cos(\omega_1 t)+E_{\omega 2}\cos(\omega_2 t))$

$= \text{const}\ \chi^{(2)}(\ E_{\omega 1}{}^2\cos^2(\omega_1 t)+ E_{\omega 2}{}^2\cos^2(\omega_2 t)+2E_{\omega 1}E_{\omega 2}\cos(\omega_1 t)\cos(\omega_2 t))$

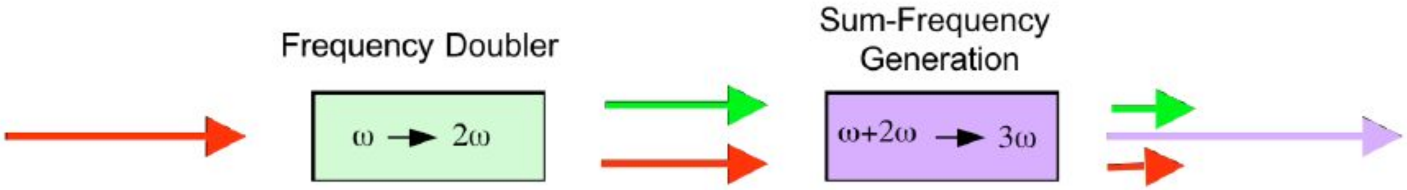
The terms in $\cos^2$ lead to SHG of ω_1 and ω_2 as before, but of interest here is the last term and remembering the trig ID

$\cos A \cos B = ½\ (\cos(A+B) + \cos(A-B))$:

$2E_{\omega 1}E_{\omega 2}\cos(\omega_1 t)\cos(\omega_2 t) = \text{const}\ [\cos(\omega_1+\omega_2)t + \cos(\omega_1-\omega_2)t]$

We have now generated two new waves with frequencies = $\omega_1+\omega_2$ and $\omega_1-\omega_2$. These are the so-called sum and difference frequencies and allow us to look at other uses for $\chi^{(2)}$ non-linearities when appropriately phase matched.

For example, we can now use this to generate efficiently our third harmonic radiation.

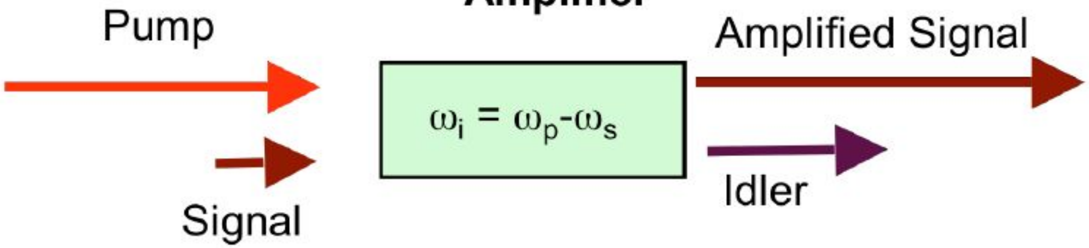


Difference frequency mixing

We can exploit difference frequency mixing to generate waves of different frequencies or to transfer energy from a strong pump wave to a weak input (signal) wave. This is called parametric amplification. In the process, assuming phase matching takes place, a new longer wavelength (idler) wave is generated subject to:

$$\frac{1}{\lambda_{pump}} = \frac{1}{\lambda_{signal}} + \frac{1}{\lambda_{pump}}$$

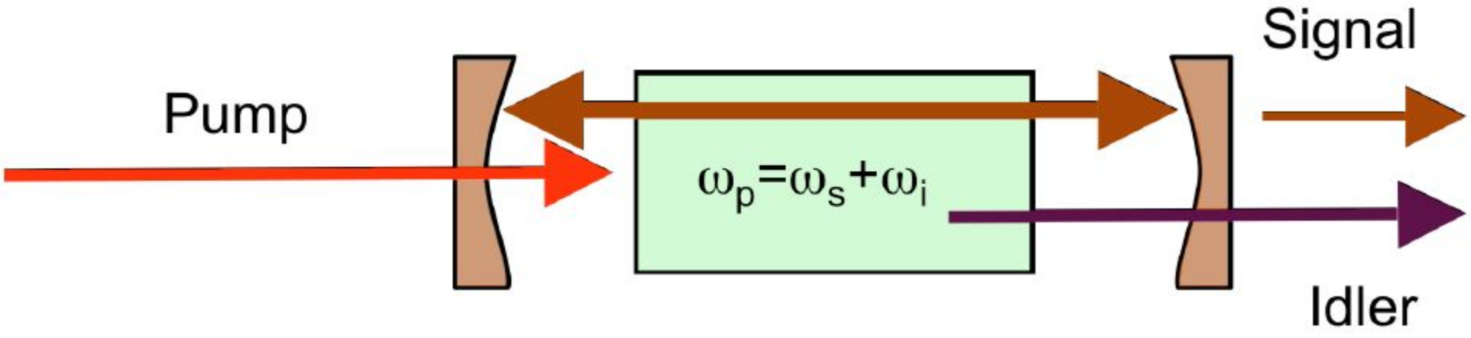
Optical Parametric Amplifier



Eg. if we wish to transfer energy from a wave at 800nm to a wave at 1000nm, we will also generate an idler wave at: 4000nm.

If we now form a cavity where the signal, idler or both are resonated, a device called an optical parametric oscillator can be made. Depending on the phase matching conditions, a wide range of tunable signal and idler wavelengths can be formed.

Optical Parametric Oscillator (Resonant for Signal)



Summary for $\chi^{(2)}$ processes

We have seen that the coupling between waves offered by the χ(2) coefficient has opened a rich seam of non-linear optical effects – from SHG through to OPO’s. Remember also that for all of the processes discussed, phase matching must be achieved whether through BPM or QPM type techniques. As we move to a wider range of wavelengths it’s also very important to consider the transparency range of your crystals, eg. if we wish to generate 7µm