

Module 2 : Nonlinear Frequency Mixing

Lecture 13 : Phase Matching

Objectives

In this lecture we will look at

- Phase Matching by birefringence.
- Quasi Phase Matching (by periodic compensation of phase mismatch).
- Some examples.

Recall from Lecture 4, that in birefringent crystals for any given direction of propagation linear wave equation has two independent solutions. For a given \vec{k} , \hat{a} has two possible solutions one corresponding to the ordinary ray and the other corresponding to the extra ordinary ray. In uniaxial crystals

$$\varepsilon = \begin{pmatrix} \varepsilon_{\perp} & 0 & 0 \\ 0 & \varepsilon_{\perp} & 0 \\ 0 & 0 & \varepsilon_{\parallel} \end{pmatrix} \quad (13.1)$$

in the frame of the principal axes of the crystal. If the direction of propagation makes an angle θ with the optic axis ε_{\parallel} , the o-ray has the refractive index $\eta_o = \sqrt{\varepsilon_{\perp} / \varepsilon_0}$ and its polarization vector \hat{a} is perpendicular to the optics axis and the direction of propagation. For the e-ray, the polarization vector \hat{a} lies in the plane containing the optic axis and the direction of propagation. It makes an angle $(\pi/2 - \alpha)$ with the direction of propagation such that

$$\tan \alpha = \frac{1}{2} \eta^2 \sin 2\theta \left(\frac{\varepsilon_0}{\varepsilon_{\parallel}} - \frac{\varepsilon_0}{\varepsilon_{\perp}} \right) \quad (13.2)$$

and the refractive index η_e is given by

$$\frac{1}{\eta_e^2} = \frac{\varepsilon_0 \cos^2 \theta}{\varepsilon_{\perp}} + \frac{\varepsilon_0 \sin^2 \theta}{\varepsilon_{\parallel}} \quad (13.3)$$

In the normal dispersion region the refractive index varies like $\eta(\omega) = \eta(0) + A\omega^2$, so that the $\eta(2\omega) > \eta(\omega)$ for a given mode (or ordinary (o) or extraordinary (e)).

For a positive uniaxial crystal $\eta_e(\theta)$ lies between $\sqrt{\varepsilon_{\parallel} / \varepsilon_0} = \eta_{\parallel} > \eta_o$ and η_o

Thus, if we chose the polarization such that the fundamental beam propagate as the e-wave and the second harmonic wave as o-wave, it is possible to achieve $\eta_e(\omega) = \eta_o(2\omega)$ for some value of θ , the angle between the optic axis and the deviation of the propagation, provides the birefringence $\eta_e(\omega) - \eta_o(\omega)$ is sufficiently large.

Example 1: The first experiment on SHG was reported by Frank et al (PRL 7, 118(1961)). They observed SHG of a commercial ruby Laser in a quartz crystal. The refractive indices of quartz at the wave length of the Ruby Laser (6943 Å) and its second harmonic is given by:

$$\begin{aligned} n_1^e &= 1.55 & n_2^e &= 1.57 \\ n_1^o &= 1.54 & n_2^o &= 1.56 \end{aligned}$$

Since the birefringence here is much smaller than the dispersion, phase matching is not possible in this time

Example 2: Phase Matched SHG of a Ruby Laser was first observed in potassium dihydrogen phosphate crystal by Giordimaine (PRL 8, 119(1962)). For this crystal

$$\begin{aligned} n_1^e &= 1.466 & n_2^e &= 1.487 \\ n_1^o &= 1.505 & n_1^o &= 1.534 \end{aligned}$$

Thus the fundamental wave propagation as ordinary wave, its refractive index is 1.505 independent of the direction of propagation. That, for the second harmonic is 1.534 for the ordinary wave independent of the direction of propagation. The refractive index of the e-wave at SH varies between 1.487 and 1.535 depending on the angle θ between e direction of propagation and the optic axis. This can be made equal to 1.505, the refractive index of the fundamental wave if the angle between the optic axis and the direction of propagation is chosen such that

$$\frac{1}{1.505} = \frac{\cos^2 \theta}{1.534} + \frac{\sin^2 \theta}{1.487}$$

or

$$\theta = 50^\circ$$

Other experimental issues:

- **Walk-off:** As shown in Figure 1.4 the direction of the Poynting vector may differ from the direction of propagation which is normal to the planes of constant phase. Since practical light waves always have finite lateral size, this leads to reducing overlap between the fundamental frequency wave and the second harmonic wave because one of them is ordinary and the other is extra-ordinary wave. This effectively reduces the interaction length. The angle between \vec{E} and \vec{D} is called the walk off angle.
- **Noncritical Phase-Matching:** If phase matching occurs at $\theta = \pi/2$, the \vec{E} and \vec{D} are again parallel and there is no walk off. This is called noncritical phase matching and is desirable for obtaining high conversion efficiency.
- When birefringence is sufficient some times phase matching can be obtained for the process in which fundamental beam is split into an ordinary wave and an extraordinary wave. This is called **type 2 phase matching**. Since the two fundamental frequency waves now travel with different phase velocities they have to be treated as independent. So we now need to write 3 coupled equations –one for the second harmonic and two for the two polarizations of the fundamental wave this is a 3 wave mixing process and will be discussed in that lecture.
- For increasing the efficiency of second harmonic generation we need to increase the intensity. This can be done by focusing the fundamental wave but the tighter the focusing, faster the spread of the focal spot due to diffraction. This again limits the effective interaction length of the crystal. There is thus an optimum focusing condition.
- Phase matching only determines the angle between the optic axis and the direction of propagation. The azimuthal angle is chosen to maximize $\chi_{eff}^{(2)}$, the effective coupling coefficient.

Quasi Phase Matching:

For isotropic crystals BPM is not possible. Since some crystals like semiconductors have large nonlinearities as well as other desirable properties like availability in good quality and relatively large sizes, alternate methods of compensation of phase mismatch were tried. We discuss these methods all originally proposed by ABDP. The most important of them is the quasi phase matching in which the sign of $\chi_{eff}^{(2)}$ is reversed after each coherence length. Since

$$\chi_{eff}^{(2)} = \sum_{i,j,k} \chi_{ijk}^{(2)}(-2\omega, \omega, \omega) a_{2i} a_{1j} a_{1k} \quad (13.4)$$

it follows that the sign of $\chi_{eff}^{(2)}$ can be reversed if the crystal is rotated by 180° about the direction of propagation. This is because such a rotation reverses the direction of every vector normal to it and all the polarization vectors are usually chosen to lie in a plane normal to the direction of propagation by cutting the crystal suitably.

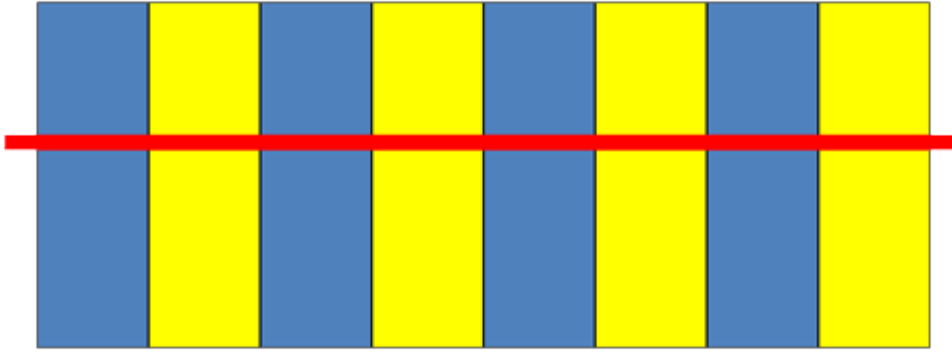


Figure 13.1 $\chi_{eff}^{(2)}$ in a quasi-phase matched stack. Alternate segments are rotated by 180° about the direction of propagation shown by red line.

We can understand the QPM process in two ways.

First following the discussion in Lecture 9 we note that the rate at which the polarization radiates energy in the field at frequency Ω is $\sim \text{Im}(P^{NL}(\Omega) \cdot E^*(\Omega))$ which oscillates with a period $2\pi / |\vec{k}_{NL} - \vec{k}|$ like where the denominator is the mismatch between the propagation vector for the nonlinear polarization and that for the field at the frequency Ω . The polarization travels with a wave vector determined by the fields that create this polarization. Thus the second harmonic grows till distance in the crystal becomes $l_{coh} = \pi / |\vec{k}_{NL} - \vec{k}|$ when the direction of energy flow reverses. If now the sign $\chi_{eff}^{(2)}$ is reversed the second harmonic will continue growing for one more distance of $\pi / |\vec{k}_{NL} - \vec{k}|$. The key idea, first proposed by ABDP is that phase mismatch can be compensated periodically. Next, let us see this more quantitatively in the small signal approximation.

In the first crystal of length $l_{coh} = \pi / |\vec{k}_{NL} - \vec{k}|$, the amplitude of the second harmonic wave is given by eq (9.21)

$$A_2 = z K' e^{-(i\Delta k z / 2)} \frac{\sin(\Delta k z / 2)}{(\Delta k z / 2)} \quad (13.5)$$

In the second crystal, the starting amplitude is $A_2(0) = -i(2/\pi)l_{coh}K'$ and the equation it follows is

$$\frac{\partial A_2}{\partial z} = -K' e^{-i\Delta k z} \quad (13.6)$$

since the sign of $\chi_{eff}^{(2)}$ is reversed and the right hand side is proportional to it. Solving this gives

$$A_2 = -i 2(2/\pi)l_{coh}K' e^{-i\Delta k z} \quad (13.7)$$

at the end of second crystal of length $z = l_{coh}$. Similarly at the end of the third crystal amplitude A_2 is 3 times that in one coherence length.

This shows that the growth of the second harmonic in a QPM stack of crystals is like that in a BPM with $\chi_{eff}^{(2)}$ reduced by a factor $2/\pi$.

This can be seen in yet another way which yields greater insight into this equivalence.

We again solve the equation of evolution of the second harmonic but assume now that the coupling constant K' is a periodic function of z . It can then be expanded into a Fourier series.

$$K'(z) = \sum_{n=-\infty}^{\infty} c_n e^{ik_n z} \quad (13.8)$$

with $k_n = 2n\pi / \Lambda$ where Λ is the period.

Substituting this in eq (9.20) we get

$$\frac{\partial A_2}{\partial z} = K'(z) e^{-i\Delta k z} = \sum_{n=-\infty}^{\infty} c_n e^{i(k_n - \Delta k)z} \quad (13.9)$$

Integrating this we obtain

$$A_2 = \sum_{n=-\infty}^{\infty} c_n e^{i(k_n - \Delta k)z/2} \frac{\sin((k_n - \Delta k)z/2)}{(k_n - \Delta k)/2} \quad (13.10)$$

If now the period of variation of $\chi_{\text{eff}}^{(2)}$ is chosen to be $\Lambda = 2l_{\text{coh}}$, $k_1 = \Delta k$ and only one term will dominate as z increases and we can write

$$A_2 = c_1 z \quad (13.11)$$

For the case when $\chi_{\text{eff}}^{(2)}$ is just changing sign in alternate segments $c_n = K'(2/n\pi)$ and so a QPM stack behaves exactly like a phase matched stack with $\chi_{\text{eff}}^{(2)}$ reduced by a factor of $2/\pi$. This simple derivation also tells us more

- Higher order quasi phase matching is possible in which the sign of $\chi_{\text{eff}}^{(2)}$ is changed after an odd number of coherence lengths but the $\chi_{\text{eff}}^{(2)}$ is reduced by $2/m\pi$ if m th order QPM is used. This is useful when the coherence length is too small for convenient fabrication. This is depicted in the Figure below.

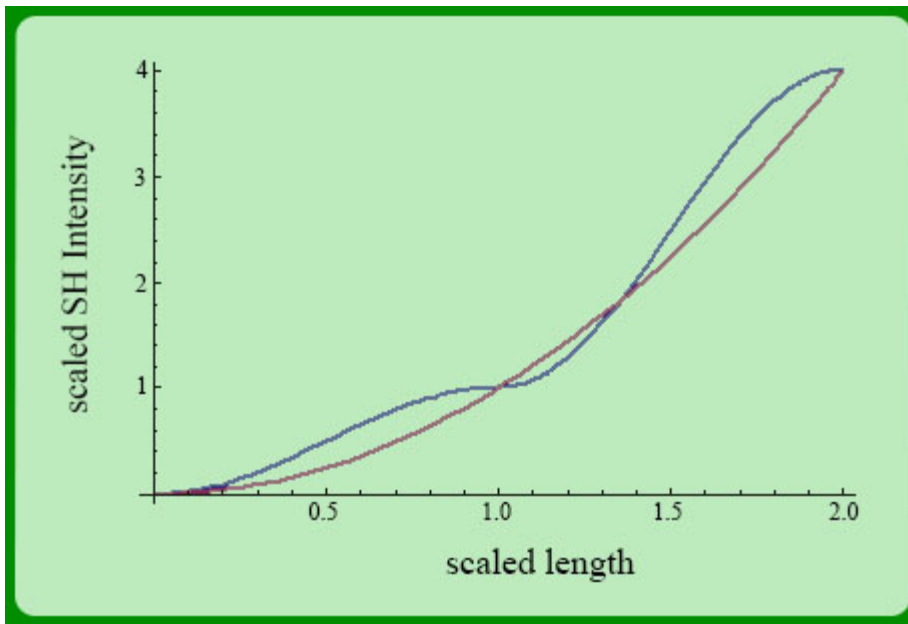


Figure 13.2 The second harmonic growth in the first two segments of the quasi-phase-matched stack. Length is scaled to the coherence length and the second harmonic intensity to that produced in the first segment. The blue curve corresponds to the solutions depicted by eqs(13.5) and (13.7). The magenta curve corresponds to the approximation given by eq (13.11). The difference between the two is due to neglect of higher Fourier components in eq (13.11).

- The periodicity of $\chi_{eff}^{(2)}$ is of importance and not its abrupt change of sign. Although at present it seems simpler to fabricate QPM structures with abrupt change of sign of $\chi_{eff}^{(2)}$ with epitaxial techniques other structures may be possible.
- when the period of $\chi_{eff}^{(2)}$ does not match any odd multiple of $2l_{coh}$, other Fourier components become important.

PUMP DEPLETION EFFECTS IN QUASI PHASE MATCHING:

All the above discussion has been within the small signal approximation. Obviously with high efficiencies of conversion pump depletion effects become important. Recognizing that the ABDP coupled wave solutions discussed in earlier lectures, the following procedure can be used to calculate the nonlinear response of QPM stacks. Starting with a given initial value of the fractional power in the second harmonic v and the relative phase angle θ obtain Γ from eq (10.32) and then the three roots of eq (10.34) which is a cubic in v^2 . By requiring that the initial value of v^2 at $z = 0$ satisfies the condition,

$$v^2(0) = v_a^2 + (v_b^2 - v_a^2) \operatorname{sn}^2 \left(\sqrt{v_c^2 - v_a^2} (\zeta_0), \gamma \right) \quad (13.12)$$

we obtained $|\zeta_0|$. The sign of ζ_0 needs to be determined by requiring that v^2 given by eq (10.35) should increase or decrease with z as determined by the initial value of the relative phase θ , through eq (10.29). Since ζ is proportional to $\chi_{eff}^{(2)}$, its sign depends on the sign of $\chi_{eff}^{(2)}$. Thus, in a stack of crystals with changing sign of $\chi_{eff}^{(2)}$ it is most convenient to work in terms of $|\zeta|$ which is directly proportional to z , the distance traveled in the crystal. We can then rewrite eq(13.12) as

$$v^2 = v_a^2 + (v_b^2 - v_a^2) \operatorname{sn}^2 \left(\sqrt{v_c^2 - v_a^2} (|\zeta| \pm |\zeta_0|), \gamma \right) \quad (13.13)$$

where, the + sign is chosen if $\partial v / \partial z$ is positive and the - sign if it is negative.

The main results of such an analyses are summarized below:

- The crystal length in which the relative phase angle q changes by p depends on the intensity due to phase change induced by the nonlinear coupling which manifests as the dependence of the *coherence length* on $\Delta\mathcal{S}$. In an ideal stack the coherence length was defined as the distance over which the relative phase q changes by p . Consequently, the crystal lengths in such an ideal QPM structure are not constant but have an intensity dependent variation. This dependence is negligible for $|\Delta\mathcal{S}| > 100$ and becomes significant for $|\Delta\mathcal{S}| \leq 10$. In most cases it does not matter but we note that there are many situation when we have birefringence insufficient for phase matching but the phase mismatch can be quite small. More important, the coherence length then changes as we proceed along an ideal quasi phase matched stack –after each crystal.
- when the conversion in each crystal is small ($|\Delta\mathcal{S}| \gg 1$), the second harmonic generation in an ideal quasi-phase-matched stack of crystals behaves like that in an equivalent birefringence phase-matched (BPM) crystal with $\chi_{eff}^{(2)}$ reduced by a factor of $2/\pi$. This is a generalization of result in the small signal limit.
- However, this equivalence is not valid if both the beams are initially present with given phases. For example, if the initial value of the relative phase angle is $\pi/2$, a QPM stack will transfer almost no energy between the waves while a perfectly phase matched system will do so efficiently. Interestingly if we have a BPM crystal and a QPM in tandem only the first one will be effective in SHG unless we manipulate the relative phase between the two crystals.
- For periodic stacks, we investigated the effect of period differing from the coherence length. In such stacks, we found, that the SH amplitude would continue to increase till the relative phase becomes close to $\pi/2$. In the figure below we reproduce the behavior of the fractional power in the SH wave and the relative phase difference for a stack with $\Delta\mathcal{S} = 100$ and in which the length of each crystal is 0.8 of the coherence length.

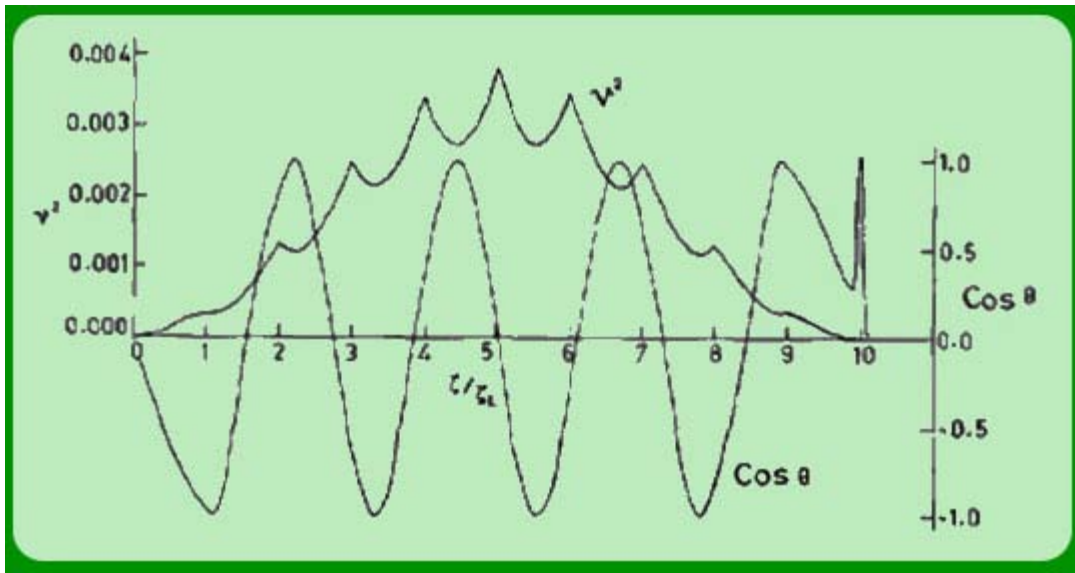


Figure 13.3 The variation of second harmonic intensity and relative phase in a QPM structure with length 20% shorter than coherence length

Clearly for the SH intensity this stack behaves quite like single crystal with coherence length equal to 5 times the coherence length of each crystal. The behavior of the relative phase θ is however quite remarkable. The relative phase oscillates as expected in the small signal approximation for most of the range. However, we see a very sharp variation of phase in the last segment when the second harmonic amplitude becomes small and the relative phase is not yet $\pi/2$. From eq (40) we see that the nonlinear phase change can be very large when SH amplitude is small and the relative phase away from $\pi/2$.

- We found that as shown in the figure below such mismatch in crystal length can be easily compensated by periodically introducing a layer of double the length. More important, our treatment of doubly periodic stacks showed that our method allowed us to obtain the phase and amplitude in all piecewise continuous microstructures. It will be interesting to see how the doubly periodic stack works out in the nonlinear diffraction or Fourier decomposition scheme
- The effect of random variations in crystal lengths around a mean value is important. First simulating experimental errors in a designed stack the individual crystal lengths vary randomly over a small range around a mean value. In the small signal limit analytical results can be obtained. For a nearly periodic stack, deviation of the mean value reduces the conversion efficiency much more than random deviations of similar magnitude. In the second case, simulating twinned crystals the crystal lengths vary randomly over the interval 0 and $2l_{coh}$. In this case the SH intensity averaged over many samples would rise like N , the number of segments. However, note that it is not reasonable to compare this with experimental result for an individual twinned crystal since the standard deviation too increases proportional to the mean. Since the information of practical importance is the behavior of SHG in a typical twinned crystal, we also calculated the SHG in typical distributions of lengths. Results of one such calculation are shown below by solid circles. Clearly the fluctuations are important. This is still a topic of current research.

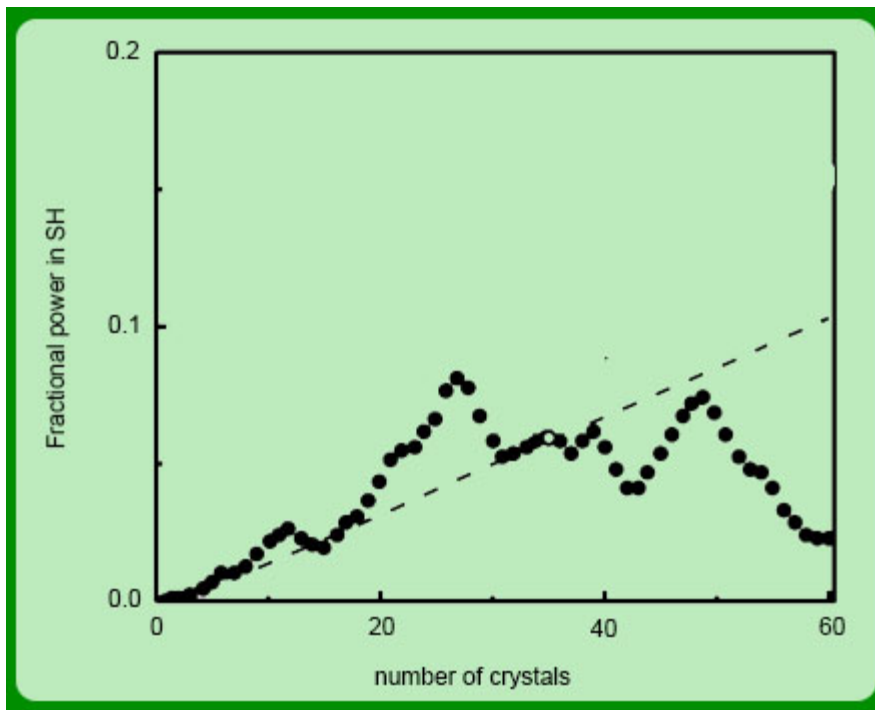


Figure 13.4 Growth of second harmonic intensity for a QPM structure with crystal lengths varying randomly.

Some examples of experimental implementation of Quasi Phase matching are discussed in the next lecture.

Recap:

- For substantial power in nonlinear frequency conversion, phase matching is required.
- In crystals with sufficient birefringence, it can be used to compensate the phase mismatch due to dispersion. Details of this method are discussed for uniaxial crystals.
- Phase mismatch can also be compensated by changing the sign of the nonlinear coupling coefficient every coherence length.