

Second Order Nonlinear Optics

In a nonlinear medium, the induced polarization is written

$$P = \varepsilon_0 [\chi^{(1)} E + \chi^{(2)} E^2 + \chi^{(3)} E^3 + \dots]$$

For this lecture we will focus on 2nd order nonlinearities.

Illustration: consider an incident electric field

$$E = \frac{1}{2} [\hat{E}(\omega_1) e^{-i\omega_1 t} + \hat{E}(\omega_2) e^{-i\omega_2 t} + \text{c.c.}]$$

This “perturbative” approach is valid for intensities less than $\sim 10^{14}$ W/cm², where the laser field is below the atomic field

The induced polarization is

$$P_{NL}^{(2)} = \frac{\varepsilon_0}{4} \left[\chi^{(2)}(2\omega_1 : \omega_1, \omega_1) \hat{E}^2(\omega_1) e^{-i2\omega_1 t} + \chi^{(2)}(2\omega_2 : \omega_2, \omega_2) \hat{E}^2(\omega_2) e^{-i2\omega_2 t} \right. \\ + 2\chi^{(2)}(\omega_1 + \omega_2 : \omega_1, \omega_2) \hat{E}(\omega_1) \hat{E}(\omega_2) e^{-i(\omega_1 + \omega_2)t} \\ + 2\chi^{(2)}(\omega_1 - \omega_2 : \omega_1, -\omega_2) \hat{E}(\omega_1) \hat{E}^*(\omega_2) e^{-i(\omega_1 - \omega_2)t} \\ \left. + \chi^{(2)}(0 : \omega_1, -\omega_1) \hat{E}(\omega_1) \hat{E}^*(\omega_1) + \chi^{(2)}(0 : \omega_2, -\omega_2) \hat{E}(\omega_2) \hat{E}^*(\omega_2) \right] \\ + \text{c.c.}$$

Second Harmonic Generation
Sum Frequency Generation
Difference Frequency Generation
Optical Rectification

Vector Fields and Tensor Susceptibilities

In general the fields and polarization must be treated as vectors and the nonlinear susceptibility is a tensor.

Consider the case of sum frequency generation.

Write

$$\mathbf{E} = \frac{1}{2} \sum_{i=1}^3 \hat{u}_i \left[\hat{E}_i(\omega_1) e^{-i\omega_1 t} + \hat{E}_i(\omega_2) e^{-i\omega_2 t} + \text{c.c.} \right]$$

and

$$\mathbf{P} = \frac{1}{2} \sum_{i=1}^3 \hat{u}_i \left[\hat{P}_i^{(2)}(\omega_1 + \omega_2) e^{-i(\omega_1 + \omega_2)t} + \text{c.c.} \right]$$

the components of \mathbf{P} can be written in terms of the susceptibility tensor as

$$\hat{P}_i^{(2)}(\omega_1 + \omega_2) = \frac{\epsilon_0 D}{2} \sum_{j,k=1}^3 \chi_{ijk}^{(2)}(\omega_1 + \omega_2 : \omega_1, \omega_2) \hat{E}_j(\omega_1) \hat{E}_k(\omega_2)$$

where D is a degeneracy factor equal to number of distinguishable permutations.
For SFG $D = 2$, for SHG $D = 1$

Properties of $\chi_{ijk}^{(2)}$

In general $\chi_{ijk}^{(2)}$ has 27 components, however symmetry greatly reduce number in independent components

In a transparent (lossless) material $\chi_{ijk}^{(2)}$ is real.

If nonlinearity is dispersionless (instantaneous) $\chi_{ijk}^{(2)}$ is independent of ω .

If the material has inversion symmetry $\chi_{ijk}^{(2)} = 0$

There are typically only a few independent components

“Engineering notation”:

$$d_{ijk} = \frac{1}{2} \chi_{ijk}^{(2)}$$

d_{ijk} is in the range of 10^{-11} m/V to 10^{-13} m/V

Propagation

Recall from the second lecture the wave equation within SVEA:

$$2ik \frac{\partial \hat{E}(z', t')}{\partial z'} = \mu_0 \omega^2 \hat{P}(z', t')$$

We will generalize this slightly to include dispersion (first order polarization) as an explicit group velocity (only considering dispersion between ω and 2ω , for example)

$$\frac{\partial \hat{E}(z, t)}{\partial z} + \frac{1}{v_g} \frac{\partial \hat{E}(z, t)}{\partial t} = \frac{i\mu_0 \omega_0 c}{2n} \hat{P}_{NL}(z, t) (\hat{\mathbf{e}} \cdot \hat{\mathbf{p}}) e^{-ik_0 z}$$

We can also express this in the frequency domain where dispersion is fully included in the $k(\omega)$ term

$$\frac{\partial \hat{E}(z, \omega)}{\partial z} = \frac{i\mu_0 \omega c}{2n} \hat{P}_{NL}(z, \omega) (\hat{\mathbf{e}} \cdot \hat{\mathbf{p}}) e^{-ik(\omega)z}$$

Note that this equation is derived using a SVEA with respect to space, i.e., the change in the envelope due to the non-linear polarization is small, thus it is valid even for very short pulses

CW Second Harmonic Generation

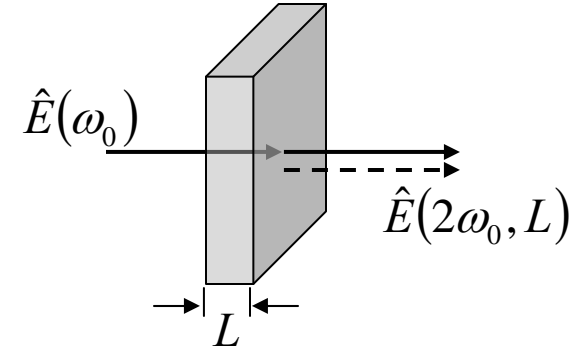
First review second harmonic generation for CW beams

$$E(z, t) = \frac{1}{2} \left[\hat{E}_\omega(z) e^{-i(\omega_0 t - k_\omega z)} + \hat{E}_{2\omega}(z) e^{-i(2\omega_0 t - k_{2\omega} z)} + \text{c.c.} \right]$$

$$P_{NL}(z, t) = 2\varepsilon_0 d_{\text{eff}} E^2(z, t)$$

where we've gone to scalars and lumped vector effects in d_{eff}

$$\begin{aligned} P_{NL}(z, t) &= \frac{1}{2} \left[\hat{P}_{NL}^{(\omega)}(z, t) e^{-i\omega_0 t} + \hat{P}_{NL}^{(2\omega)}(z, t) e^{-2i\omega_0 t} + \text{c.c.} \right] \\ &= \frac{\varepsilon_0 d_{\text{eff}}}{2} \left[\hat{E}_\omega^2(z) e^{-2i(\omega_0 t - k_\omega z)} + 2\hat{E}_{2\omega}(z) \hat{E}_\omega^*(z) e^{-i[\omega_0 t - (k_{2\omega} - k_\omega)z]} + \text{c.c.} \right] \end{aligned}$$



Insert this in time-domain wave equation to get

$$\frac{\partial \hat{E}_\omega}{\partial z} = \frac{i\omega d_{\text{eff}}}{n_\omega c} \hat{E}_{2\omega} \hat{E}_\omega^* e^{i\Delta k z} \qquad \frac{\partial \hat{E}_{2\omega}}{\partial z} = \frac{i\omega d_{\text{eff}}}{n_{2\omega} c} \hat{E}_\omega^2 e^{-i\Delta k z}$$

Where the phase mismatch is $\Delta k = k_{2\omega} - k_\omega$

Phase matched

Assume

Phase matching $\rightarrow \Delta k = 0$

Then for no pump depletion $\rightarrow |\hat{E}_{2\omega}(z)| \ll |\hat{E}_\omega(0)|$ and $\hat{E}_\omega(z) \approx \hat{E}_\omega(0)$

$$\hat{E}_{2\omega}(z) = i\kappa \hat{E}_\omega^2(0)z \quad \text{with} \quad \kappa = \frac{\omega_0 d_{eff}}{cn}$$

or, in terms of intensity

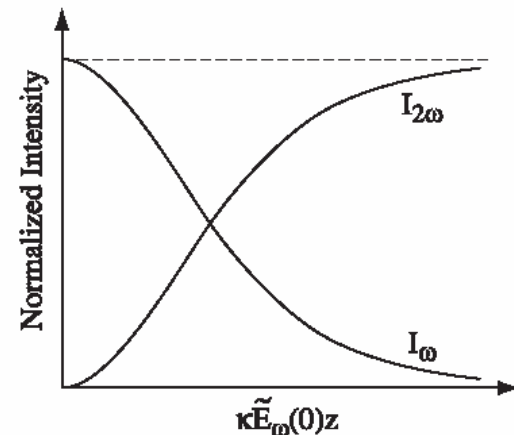
$$I_{2\omega}(L) = \Gamma^2 I_\omega^2 L^2 \quad \text{with} \quad \Gamma = \frac{2\kappa^2}{\epsilon_0 cn} = \frac{2\omega_0^2 d_{eff}^2}{\epsilon_0 c^3 n^3}$$

the conversion efficiency, $I_{2\omega}/I_\omega$, is proportional to input intensity and length squared

for perfect phase matching, even with arbitrary pump depletion, solvable

$$\hat{E}_\omega(z) = \hat{E}_\omega(0) \operatorname{sech}(\kappa \hat{E}_\omega(0)z)$$

$$\hat{E}_{2\omega}(z) = i\hat{E}_\omega(0) \tanh(\kappa \hat{E}_\omega(0)z)$$



Phase matching

For non zero phase mismatch

$$\Delta k = k_{2\omega} - 2k_{\omega} = \frac{2\omega_0 n_{2\omega}}{c} - 2 \frac{\omega_0 n_{\omega}}{c} = \frac{2\omega_0}{c} (n_{2\omega} - n_{\omega})$$

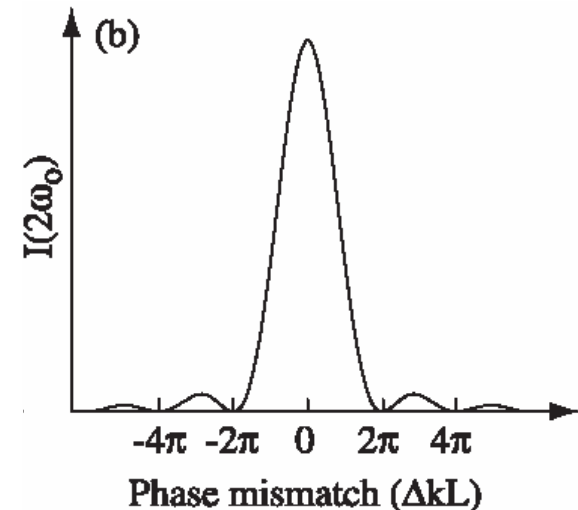
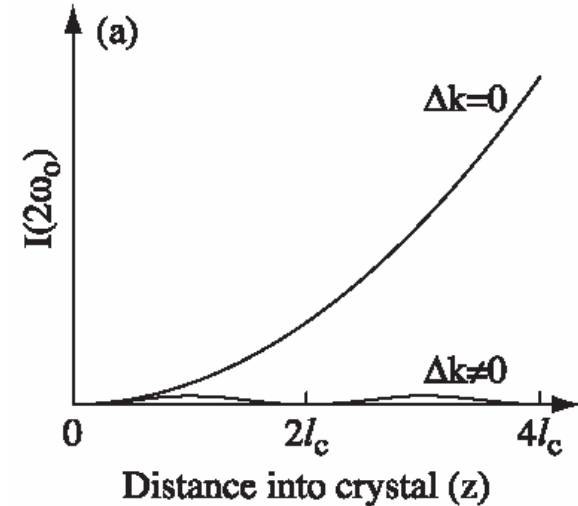
In the non-depleted pump approximation, the solution is

$$\hat{E}_{2\omega}(z) = \frac{2i\omega_0 d_{\text{eff}}}{n_{2\omega} c \Delta k} \hat{E}_{\omega}^2 e^{-i\Delta k z/2} \sin\left(\frac{\Delta k z}{2}\right)$$

Intensity:

$$I_{2\omega} = \Gamma^2 I_{\omega}^2 \left[\left(\frac{2}{\Delta k} \right) \sin\left(\frac{\Delta k z}{2}\right) \right]^2$$

$$I_{2\omega}(L) = \Gamma^2 I_{\omega}^2 L^2 \frac{\sin^2\left(\frac{\Delta k L}{2}\right)}{\left(\frac{\Delta k L}{2}\right)^2} = \Gamma^2 I_{\omega}^2 L^2 \text{sinc}^2\left(\frac{\Delta k L}{2}\right)$$



Birefringent material

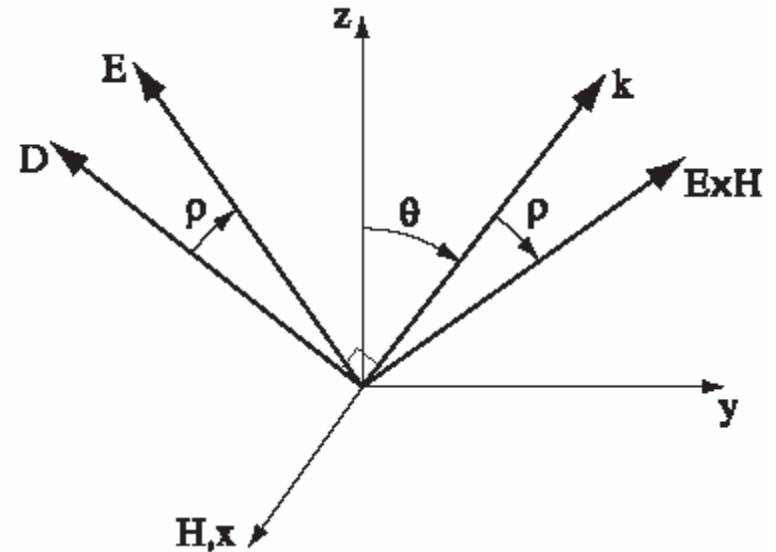
In birefringent material, the index of refraction can be adjusted by polarization and propagation direction

$$D_i = \sum_{j=1}^3 \varepsilon_{ij} E_j \quad \text{where} \quad \varepsilon_{ij} = \varepsilon_0 (\delta_{ij} + \chi_{ij})$$

For isotropic media, \mathbf{D} and \mathbf{E} are parallel, but for anisotropic material they do not point in the same direction

$$\mathbf{k} \times \hat{\mathbf{E}} = \mu_0 \omega \hat{\mathbf{H}} \quad \mathbf{k} \times \hat{\mathbf{H}} = -\omega \hat{\mathbf{D}}$$

Note that the Poynting vector $\mathbf{E} \times \mathbf{H}$ is not parallel to \mathbf{k} : walkoff at angle ρ



Ordinary and Extraordinary Waves

The wave equation in birefringent material

$$k^2 \tilde{\mathbf{E}} - \mathbf{k}(\mathbf{k} \cdot \tilde{\mathbf{E}}) = \omega^2 \mu_0 \tilde{\mathbf{D}}$$

rewritten in terms of vector components

$$k^2 \tilde{E}_i - k_i k_j \tilde{E}_j = \omega^2 \mu_0 \varepsilon_{ij} \tilde{E}_j$$

or equivalently

$$(k^2 \delta_{ij} - k_i k_j - \omega^2 \mu_0 \varepsilon_{ij}) \tilde{E}_j = 0$$

this is quadratic in k , hence for a specific direction, we expect 2 solutions.

specialize to a uniaxial crystal

$$\varepsilon_{ij} = \varepsilon_0 \begin{pmatrix} n_o^2 & 0 & 0 \\ 0 & n_o^2 & 0 \\ 0 & 0 & n_e^2 \end{pmatrix}$$

positive uniaxial: $n_e > n_o$

negative uniaxial: $n_e < n_o$

Solutions:

1. ordinary wave (o-wave)

$$k = \frac{\omega n_o}{c}$$

no walkoff

2. extraordinary wave (e-wave)

$$k = \frac{\omega n_e(\theta)}{c}$$

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

$\theta = 0$: \mathbf{k} along \mathbf{z}

$\theta = 90^\circ$: \mathbf{k} in x - y plane

walkoff

Walkoff

Walkoff causes o- and e-waves to eventually separate for beams of finite size

$$\mathbf{D} = D_o[-\cos\theta\hat{\mathbf{y}} + \sin\theta\hat{\mathbf{z}}]$$

$$\mathbf{E} = E_o[-\cos(\theta + \rho)\hat{\mathbf{y}} + \sin(\theta + \rho)\hat{\mathbf{z}}]$$

Since these are related by the dielectric tensor, we can write

$$\mathbf{D} = \varepsilon_o E_o[-n_o^2 \cos(\theta + \rho)\hat{\mathbf{y}} + n_e^2 \sin(\theta + \rho)\hat{\mathbf{z}}]$$

Which then gives

$$n_o^2 \tan\theta = n_e^2 \tan(\theta + \rho)$$

which implicitly gives the walk off angle as function of propagation direction.

The walkoff angle goes to zero for propagation along a principal axis.

Birefringent phase matching

In a positive uniaxial (sketched at right), there are two schemes for achieving phase matching:

Type 1: Input e-wave at fundamental and generate o-wave output.

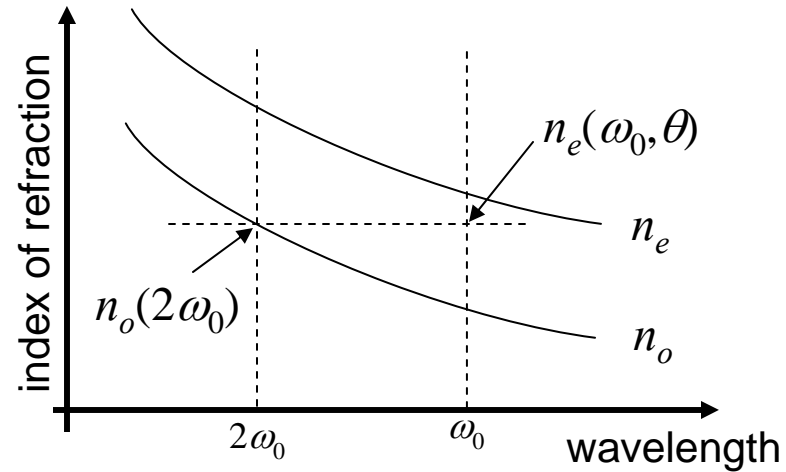
$$\sin^2 \theta_p = \left[\frac{1}{n_o^2(2\omega_0)} - \frac{1}{n_o^2(\omega_0)} \right] / \left[\frac{1}{n_e^2(\omega_0)} - \frac{1}{n_o^2(\omega_0)} \right]$$

Type 2: Input e-wave & o-wave at fundamental and generate o-wave output.

$$2n_o(2\omega_0) = n_o(\omega_0) + n_e(\omega_0, \theta)$$

Negative uniaxial *Type 1:*

$$\sin^2 \theta_p = \left[\frac{1}{n_o^2(\omega_0)} - \frac{1}{n_o^2(2\omega_0)} \right] / \left[\frac{1}{n_e^2(2\omega_0)} - \frac{1}{n_o^2(2\omega_0)} \right]$$



Gaussian beam analysis of second harmonic

The driven paraxial wave equation for the second harmonic Gaussian beam is

$$\left\{ \frac{-i}{2k_{2\omega}} \nabla_T^2 + \frac{\partial}{\partial z} \right\} \left(\tilde{E}_{2\omega}(z) u_{2\omega}(x, y, z) \right) = \frac{i\omega_0 \mu_0 c}{n_{2\omega}} \tilde{P}_{NL}(\hat{\mathbf{e}} \cdot \hat{\mathbf{p}}) e^{-ik_{2\omega} z}$$

Since $u_{2\omega}$ is a solution to the undriven paraxial wave equation, this can be simplified

$$u_{2\omega} \frac{\partial \tilde{E}_{2\omega}}{\partial z} = \frac{i\omega_0 \mu_0 c}{n_{2\omega}} \tilde{P}_{NL}(\hat{\mathbf{e}} \cdot \hat{\mathbf{p}}) e^{-ik_{2\omega} z}$$

The nonlinear polarization is written

$$\tilde{P}_{NL}^{(2\omega)} = \varepsilon_0 d_{eff} \tilde{E}_\omega^2(0) u_\omega^2(x, y, z) e^{2ik_\omega z}$$

which gives

$$\frac{\partial \tilde{E}_{2\omega}}{\partial z} = \frac{i\omega_0 d_{eff}}{n_{2\omega} c} \tilde{E}_\omega^2(0) \frac{u_\omega^2(x, y, z)}{u_{2\omega}(x, y, z)} e^{-i\Delta k z}$$

to have a valid solution we must have

$$w_{0(2\omega)} = \frac{w_{0(\omega)}}{\sqrt{2}} \quad \text{which means that both } \omega \text{ and } 2\omega \text{ beams have same } z_0$$

the final propagation equation is

$$\frac{\partial \tilde{E}_{2\omega}}{\partial z} = \frac{i\omega_0 d_{eff}}{nc} \frac{\omega_{0(\omega)}}{\omega_{(\omega)}(z)} \tilde{E}_\omega^2 \tilde{E}_\omega^2(0) e^{-i\phi_\omega(z)} e^{-i\Delta k z}$$

Weak focusing, negligible walkoff

For a large focused spot, the depth of focus ($b = 2 z_0$) is longer than the crystal (length L). In this case w and ϕ are approximately constant and the propagation equation can be integrated analytically to get

$$\tilde{E}_{2\omega}(L) = \frac{i\omega_o d_{eff}}{nc} \tilde{E}_\omega^2 L$$

The power in the second harmonic beam is calculated by integrating the intensity over the beam cross-section, giving

$$P_{2\omega} = \frac{(\Gamma P_\omega L)^2}{\pi w_{o(\omega)}^2} = \gamma P_\omega^2 \frac{L^2}{b} \quad \text{where} \quad \gamma = \frac{2n\Gamma^2}{\lambda} = \frac{4\omega_o^2 d_{eff}^2}{n^2 c^3 \epsilon_o \lambda}$$

which is basically the same as the result for plane waves.

The output power can be increased by focusing more tightly, decreasing b , until $b \sim L$, in which case the weak focusing approximation fails.

Strong focusing, negligible walk off

For strong focusing, $b \ll L$, most of the second harmonic is generated near the focus. A good estimate is to simply replace L by $\pi b/2$ in the previous result:

$$P_{2\omega} \approx \left(\frac{\pi}{2}\right)^2 \gamma P_{\omega}^2 b$$

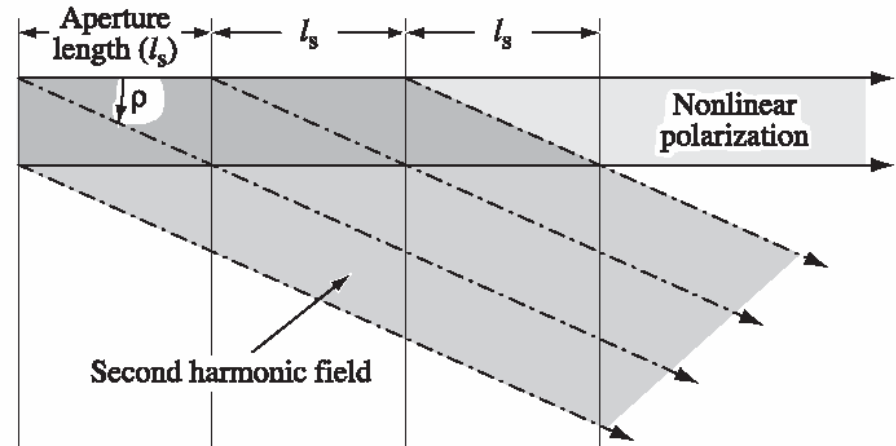
This actually slightly underestimates the second harmonic power. An exact treatment gives an optimum $L = 2.84b$, for which

$$P_{2\omega} \approx 1.068 \gamma P_{\omega}^2 L$$

Walkoff

Walk off causes the second harmonic to walk off of the nonlinear polarization produced by the fundamental. It walks off by one beam diameter in a distance

$$l_s = \frac{\sqrt{\pi} w_o(\omega)}{\rho}$$



This is significant if $l_s \ll L$.

In the **weak focusing** regime, $L \ll b$, the output is the sum of that generated by each segment with length l_s , there are L/l_s such segments

$$P_{2\omega} \approx \gamma P_\omega^2 \left(\frac{l_s^2}{b} \right) \left(\frac{L}{l_s} \right) = \gamma P_\omega^2 \frac{L l_s}{b}$$

In the **strong focusing** regime, just replace L with $\pi b/2$ as before

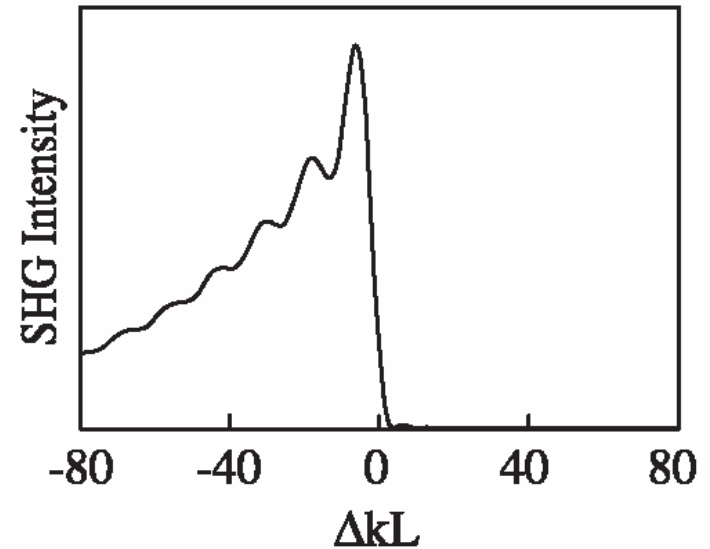
$$P_{2\omega} \approx \gamma P_\omega^2 \left(\frac{l_s^2}{b} \right) \left(\frac{\pi b}{2 l_s} \right) = \frac{\pi \gamma}{2} P_\omega^2 l_s$$

Phase matching and strong focusing

For weak focusing, phase matching is unchanged from plane waves.

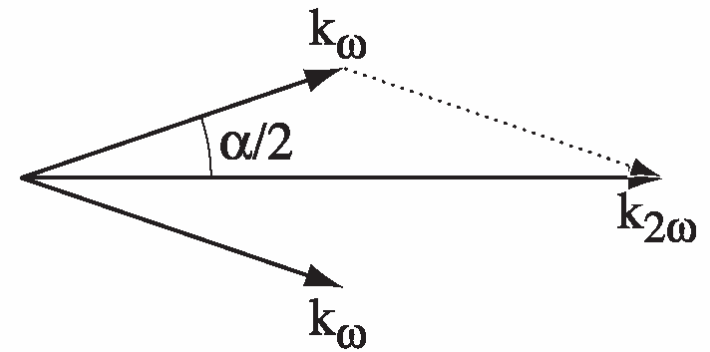
For strong focusing, the phase matching is shown at right for $L/b = 50$.

The asymmetry is due to noncollinear phase matching, which can only happen for negative Δk



$$2|k_{\omega}| \cos\left(\frac{\alpha}{2}\right) = |k_{2\omega}|$$

This can distort the spectrum.



Second Harmonic from ultrashort pulses

Perform analysis in frequency domain, let

$$E_{\omega}(t) = \frac{1}{2} \left[\frac{1}{2\pi} \int_0^{\infty} d\omega \tilde{E}_{\omega}(\omega) e^{-i[\omega t - k_{\omega}(\omega)z]} + c.c. \right]$$

$$E_{2\omega}(t) = \frac{1}{2} \left[\frac{1}{2\pi} \int_0^{\infty} d\omega \tilde{E}_{2\omega}(\omega) e^{-i[\omega t - k_{2\omega}(\omega)z]} + c.c. \right]$$

$$P_{NL}(t) = \frac{1}{2} \left[\frac{1}{2\pi} \int_0^{\infty} d\omega \tilde{P}_{NL}(\omega) e^{-i\omega t} + c.c. \right]$$

the nonlinear polarization responsible for the second harmonic is

$$P_{NL}(t) = 2\varepsilon_0 d_{eff} E_{\omega}^2(t)$$

Substituting for the input field and letting $\Omega = \omega + \omega'$

$$P_{NL}(t) = \frac{\varepsilon_0 d_{eff}}{2} \int_0^{\infty} \frac{d\Omega}{2\pi} e^{-i\Omega t} \int_0^{\infty} \frac{d\omega'}{2\pi} \tilde{E}_{\omega}(\omega') \tilde{E}_{\omega}(\Omega - \omega') e^{i[k_{\omega}(\omega') + k_{\omega}(\Omega - \omega')]z} + c.c.$$

Comparing to above, we identify

$$\tilde{P}_{NL}(\Omega) = \varepsilon_0 d_{eff} \int_0^{\infty} \frac{d\omega'}{2\pi} \tilde{E}_{\omega}(\omega') \tilde{E}_{\omega}(\Omega - \omega') e^{i[k_{\omega}(\omega') + k_{\omega}(\Omega - \omega')]z}$$

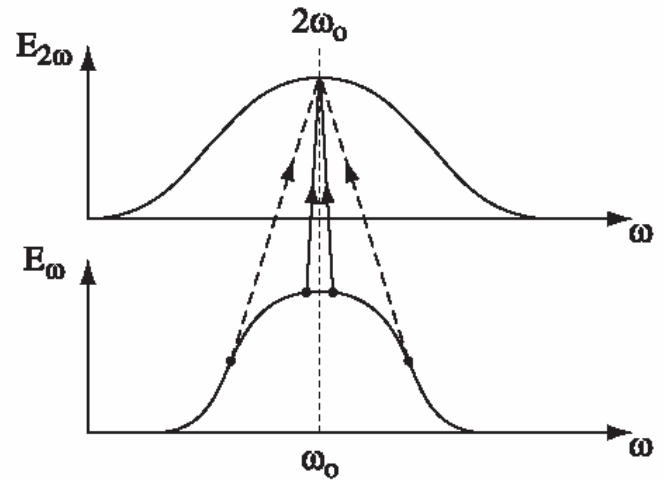
Second Harmonic Spectrum

The second harmonic spectrum $\tilde{P}_{NL}(\Omega) = \varepsilon_0 d_{eff} \int_0^\infty \frac{d\omega'}{2\pi} \tilde{E}_\omega(\omega') \tilde{E}_\omega(\Omega - \omega') e^{i[k_\omega(\omega') + k_\omega(\Omega - \omega')]z}$

shows that various pairs of the input combine to drive frequency components

Substitute into driven wave equation

$$\frac{\partial \hat{E}(z, \omega)}{\partial z} = \frac{i\mu_0 \omega c}{2n} \hat{P}_{NL}(z, \omega) (\hat{\mathbf{e}} \cdot \hat{\mathbf{p}}) e^{-ik(\omega)z}$$



To get

$$\frac{\partial \tilde{E}_{2\omega}(\Omega)}{\partial z} = \frac{i\omega_0 d_{eff}}{n_{2\omega}} \left[\frac{1}{2\pi} \int_0^\infty d\omega' \tilde{E}_\omega(\omega') \tilde{E}_\omega(\Omega - \omega') e^{i\Delta k z} \right]$$

where $\Delta k = k_{2\omega}(\Omega) - k_\omega(\omega') - k_\omega(\Omega - \omega')$

the solution is

$$\tilde{E}(L, \Omega) = \frac{i\omega_0 d_{eff}}{n_{2\omega} c} \int_0^\infty \frac{d\omega'}{2\pi} \tilde{E}_\omega(\omega') \tilde{E}_\omega(\Omega - \omega') \left[L \exp\left(\frac{-i\Delta k L}{2}\right) \text{sinc}\left(\frac{\Delta k L}{2}\right) \right]$$

Attributes of Second Harmonic

$$\tilde{E}(L, \Omega) = \frac{i\omega_0 d_{eff}}{n_{2\omega} c} \int_0^\infty \frac{d\omega'}{2\pi} \tilde{E}_\omega(\omega') \tilde{E}_\omega(\Omega - \omega') \left[L \exp\left(\frac{-i\Delta k L}{2}\right) \text{sinc}\left(\frac{\Delta k L}{2}\right) \right]$$

if we assume phase matching at ω_0 , then $k_{2\omega}(2\omega_0) = 2k_\omega(\omega_0)$ and we can write

$$\Delta k(\Omega) = \left[\frac{\partial k_{2\omega}}{\partial \omega} \Big|_{2\omega_0} - \frac{\partial k_\omega}{\partial \omega} \Big|_{\omega_0} \right] (\Omega - 2\omega_0) = [v_g^{-1}(2\omega_0) - v_g^{-1}(\omega_0)] (\Omega - 2\omega_0) = \Delta(v_g^{-1}) (\Omega - 2\omega_0)$$

- 1) Autoconvolution of input field \rightarrow broader in spectrum for bandwidth limited pulses
- 2) Phase matching acts as effective filter on SH with bandwidth

$$\Delta\nu = \frac{0.88}{|\Delta v_g^{-1}|} L$$

- 3) Even in the presence of group velocity mismatch, all frequencies participate, yielding high efficiency
- 4) We've only considered phase matching between fundamental and second harmonic, broadening of each alone has been neglected

Second Harmonic in time domain

In the time domain, the second harmonic is

$$E_{2\omega}(L, t) = \frac{1}{2} \left[\frac{i\omega_0 d_{eff}}{n_{2\omega} c} \frac{e^{i[2\omega_0 t - k_{2\omega}(2\omega_0)L]}}{\Delta(v_g^{-1})} \int_{t - \Delta(v_g^{-1})L}^t dt' a_{\omega}^2(L, t') + c.c. \right]$$

where $a_{\omega}(0, t)$ is the envelope of the fundamental and

$$a_{\omega}(L, t) = a_{\omega}(0, t - v_g^{-1}(\omega_0)L)$$

this integral is taking into account the temporal walk-off between fundamental and second harmonic. Consider two limits:

1) $|\Delta(v_g^{-1})L| \ll \Delta t_{eff}$

the group delay difference is much smaller than the pulse width, Δt_{eff} , and can be neglected. Equivalent to phase matching bandwidth \geq pulse bandwidth. Output field in time is square of input.

2) $|\Delta(v_g^{-1})L| \gg \Delta t_{eff}$

the group delay difference is much larger than the pulse width. Equivalent to narrow phase matching bandwidth. Output pulse in time is essentially a square profile.

Vis/NIR: $\Delta(v_g^{-1})$ is 10's to 100's fs/mm

Second Harmonic based pulse measurements

How are techniques such as autocorrelation and FROG affected by group velocity differences? Recall:

$$G_2(\tau) \sim \int dt |E_\omega(t)E_\omega(t-\tau)|^2 \quad I_{FROG}(\Omega, \tau) \sim \left| \int dt E_\omega(t)E_\omega(t-\tau)e^{i\omega t} \right|^2 \quad G_2(\tau) \sim \int d\Omega I_{FROG}(\Omega, t)$$

For type-I phase matching, the second harmonic field in either case is

$$E_{2\omega}(t) \sim \int_0^\infty d\Omega H(\Omega) \int dt' E_\omega(t')E_\omega(t'-\tau)e^{i\Omega t'} + c.c.$$

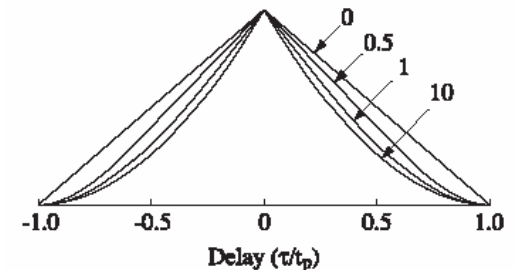
where $H(\Omega)$ is a filter function that accounts for finite phase matching bandwidth

Which gives

$$I'_{FROG}(\Omega, \tau) \sim |H(\Omega)|^2 \left| \int dt E_\omega(t)E_\omega(t-\tau)e^{i\omega t} \right|^2 \quad G'_2(\tau) \sim \int d\Omega I'_{FROG}(\Omega, t)$$

Showing that 1) for $H(\Omega)$ sufficiently broadband the traces are undistorted and 2) the phase of the filter function does not matter

The effect on autocorrelation depends on pulse shape, for a chirped Gaussian, no effect, for a square pulse:



For FROG an ideal FROG can be estimated:

$$I_{FROG}(\Omega, t) \sim I'_{FROG}(\Omega, t) \frac{\int d\omega |\tilde{E}_\omega(\omega)|^2 |\tilde{E}_\omega(\Omega - \omega)|^2}{\int d\tau I'_{FROG}(\Omega, \tau)}$$

Three wave interactions

In general 2nd order processes allow 3-wave interactions that satisfy

$$\text{energy conservation: } \omega_1 + \omega_2 = \omega_3$$

$$\text{momentum conservation: } \mathbf{k}_1 + \mathbf{k}_2 = \mathbf{k}_3$$

also called “phase matching”,
but different than what we
discussed before

Practically two cases:

- 1) Sum frequency generation (second harmonic is special case)
- 2) Difference frequency generation (optical parametric amplification is closely related)

Interactions between three waves with field amplitude a_i and frequencies ω_i are given by

$$\frac{\partial a_1}{\partial z} + \frac{1}{v_{g,1}} \frac{\partial a_1}{\partial t} = i\kappa_1 a_3 a_2^* e^{i\Delta k z}$$

$$\frac{\partial a_2}{\partial z} + \frac{1}{v_{g,2}} \frac{\partial a_2}{\partial t} = i\kappa_2 a_3 a_1^* e^{i\Delta k z}$$

$$\frac{\partial a_3}{\partial z} + \frac{1}{v_{g,3}} \frac{\partial a_3}{\partial t} = i\kappa_3 a_1 a_2 e^{-i\Delta k z}$$

where

$$\kappa_i = \frac{\omega_i d_{eff}}{2n_i c}$$

$$\Delta k = k_3 - k_1 - k_2$$

Sum Frequency Generation

Input fields at ω_1 and ω_2 , assume field at ω_3 remains weak and $\Delta k = 0$

$$\frac{\partial a_3}{\partial z} + \frac{1}{v_{g,3}} \frac{\partial a_3}{\partial t} = i\kappa_3 a_1 \left(t - \frac{z}{v_{g,1}} \right) a_2 \left(t - \frac{z}{v_{g,2}} \right)$$

For pulses long enough that group velocity walk-off is negligible, result is similar to second harmonic:

$$I_3(L, t'_3) = \frac{\omega_3^2 d_{eff}^2}{2c^3 \epsilon_0 n_1 n_2 n_3} I_1(t'_3) I_2(t'_3) L^2 \quad \text{where} \quad t'_3 = t - \frac{z}{v_{g,3}}$$

If group velocity walk off is significant

$$\frac{\partial a_3(z, t'_3)}{\partial z} = i\kappa_3 a_1(t'_3 - \eta_{13}z) a_2(t'_3 - \eta_{23}z) \quad \text{where} \quad \eta_{ij} = \frac{1}{v_{g,i}} - \frac{1}{v_{g,j}}$$

The general result is

$$a_3(L, t'_3) = i\kappa_3 \int_0^L dz a_1(t'_3 - \eta_{13}z) a_2(t'_3 - \eta_{23}z)$$

Sum Frequency with one delta-function pulse

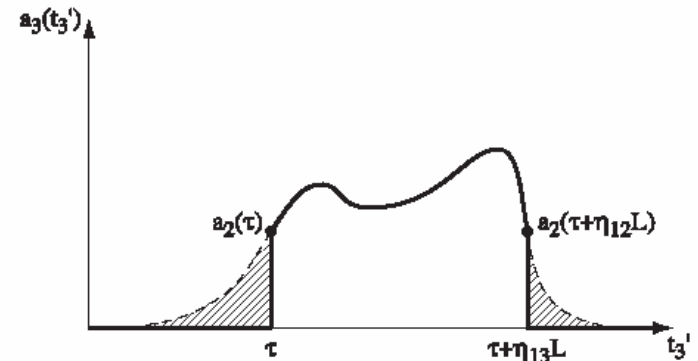
Consider SF when one input pulse is infinitely short, let

$$a_1(0, t) = A_1 \delta(t - \tau)$$

Then

$$a_3(L, t'_3) = \frac{i\kappa_3 A_1}{|\eta_{13}|} \text{sq} \left(\frac{t'_3 - \tau}{\eta_{13} L} - \frac{1}{2} \right) a_2 \left(\frac{\eta_{12} (t'_3 - \tau)}{\eta_{13}} + \tau \right)$$

The output (at right) is a temporally stretched (or compressed) version of a_2



This shows that for SF cross-correlation measurements, there is a minimum time resolution of $\eta_{12} L$

This can also be used to estimate the efficiency of up conversion, the output fluence is

$$F_3 = \left[\frac{\omega_3^2 d_{eff}^2}{2c^3 \epsilon_0 n_1 n_2 n_3} I_1(t'_3) I_2(t'_3) l_T^2 T_1 \right] \times \left(\frac{L_{eff}}{l_T} \right) \quad \text{where} \quad l_T = T_1 / |\eta_{13}| \quad L_{eff} = T_2 / |\eta_{12}|$$

This shows that the efficiency is independent of crystal length

Difference frequency generation

Difference frequency generation is analogous to sum frequency, just interchange fields

$$a_2(L, t'_2) = i\kappa_2 \int_0^L dz a_1^*(t'_2 - \eta_{12}z) a_3(t'_2 - \eta_{32}z)$$

Again consider the special case of pulse 1 being infinitely short

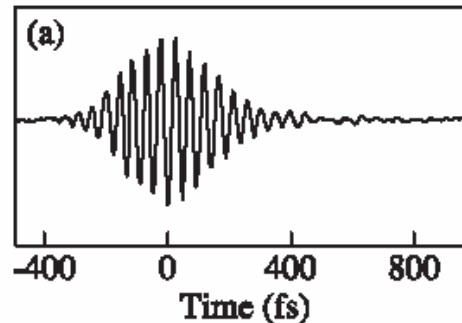
$$a_2(L, t'_3) = \frac{i\kappa_2 A_1}{|\eta_{12}|} \text{sq}\left(\frac{t'_2 - \tau}{\eta_{12}L} - \frac{1}{2}\right) a_3\left(\frac{\eta_{13}(t'_2 - \tau)}{\eta_{12}} + \tau\right)$$

Experimental demonstration of DFG is shown at right.

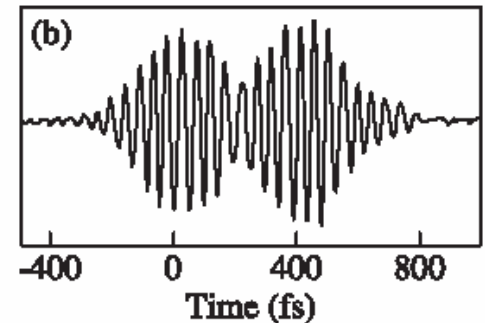
Note that for two input pulses separated by 60 fs, the output shows 400 fs separation

Due to walkoff

One input pulse



Two input pulses separated by 60 fs



Optical Parametric Amplification

In Optical Parametric Amplification, the high frequency wave, a_3 , is a strong pump

Consider phase-matched CW case in lossless material

$$\frac{\partial}{\partial z} \begin{pmatrix} a_1 \\ a_2^* \end{pmatrix} = \begin{pmatrix} 0 & i\kappa_1 a_3 \\ -i\kappa_2 a_3^* & 0 \end{pmatrix} \begin{pmatrix} a_1 \\ a_2^* \end{pmatrix}$$

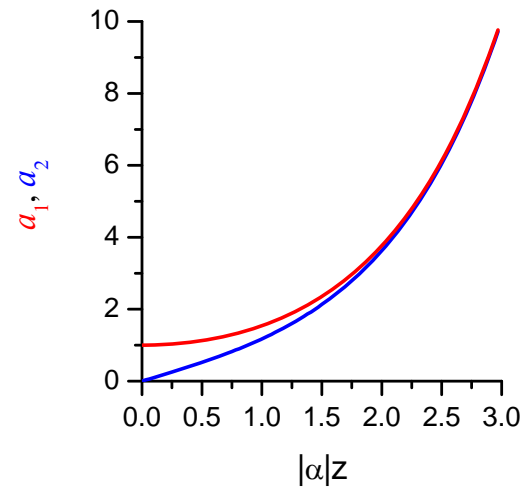
Solutions are $a_1 = \bar{a}_1 \exp(\alpha z)$ and $a_2 = \bar{a}_2 \exp(\alpha z)$ back substitute to get

$$\begin{pmatrix} -\alpha & i\kappa_1 a_3 \\ -i\kappa_2 a_3^* & -\alpha \end{pmatrix} \begin{pmatrix} a_1 \\ a_2^* \end{pmatrix} = 0$$

setting determinant to zero yields

$$a_1(z) = a_1(0) \cosh(|\alpha|z)$$

$$a_2(z) = i \sqrt{\frac{\omega_2 n_1}{\omega_1 n_2} \frac{a_3}{|a_3|}} a_1^*(0) \sinh(|\alpha|z)$$



Note the phase of a_2 reflects the phase of a_3 and a_1 . In terms of intensities:

$$I_1(z) = I_1(0) \cosh^2(|\alpha|z)$$

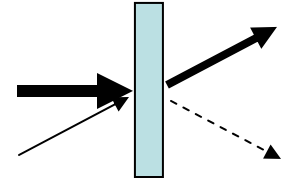
$$I_2(z) = \frac{\omega_2}{\omega_1} I_1(0) \sinh^2(|\alpha|z)$$

Optical Parametric Devices

Optical Parametric Amplifier (OPA)

Input: strong pump at ω_3 and weak signal at ω_1

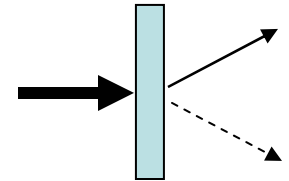
Output: amplified signal and idler at ω_2



Optical Parametric Generator (OPG)

Input: strong pump at ω_3

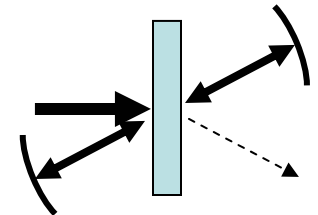
Output: signal at ω_1 and idler at ω_2 seeded by vacuum fluctuations



Optical Parametric Oscillator (OPO)

Input: strong pump at ω_3

Signal is resonant in cavity, builds up from vacuum fluctuations



OPO vs. OPA

	OPO	OPA
Pump source	fs oscillator	double fs amplifier
Pump power or energy	1 W (avg.)	100 μ J
Pump duration	100 fs	100 fs
Pump repetition rate	100 MHz	1 KHz
Pump peak power	10^5 W	10^9 W
Beam diameter	20 μ m	250 μ m
Pump Intensity	10^{10} W/cm ²	10^{12} W/cm ²

Optical Parametric Amplifier

