Imperial College London

NOTES

IMPERIAL COLLEGE LONDON

DEPARTMENT OF PHYSICS

Nonlinear Optics

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1 Linear oscillator model and the nonlinear polarisation

1.1 Linear optics

We can describe the electric field $\boldsymbol{E}$ of a light wave propagating with an oscillatory function as:

$$\boldsymbol{E}(\boldsymbol{r},t) = A\mathrm{e}^{-i(\omega t - \boldsymbol{k}\cdot\boldsymbol{r})}, \tag{1}$$

with amplitude A, frequency ω and wavevector $\boldsymbol{k}$. Linear optics assumes a linear response where the electric polarisation:

$$\boldsymbol{P} = \varepsilon_0\chi^{(1)}\boldsymbol{E}, \tag{2}$$

$$\boldsymbol{D} = \varepsilon_0\boldsymbol{E} + \boldsymbol{P} = \varepsilon_0(1+\chi^{(1)})\boldsymbol{E} = \varepsilon_r\varepsilon_0\boldsymbol{E}, \tag{3}$$

where $\boldsymbol{P}$ is the electric polarisation and $\boldsymbol{D}$ is the displacement vector. The wave equation in a medium is

$$\boldsymbol{\nabla}^2\boldsymbol{E} = \mu_0\frac{\partial^2}{\partial t^2}\left(\varepsilon_0\boldsymbol{E} + \boldsymbol{P}\right) = \frac{n^2}{c^2}\frac{\partial^2}{\partial t^2}\boldsymbol{E}, \tag{4}$$

where $n = \sqrt{\varepsilon_r\mu_r}$ and $c = 1/\sqrt{\varepsilon_0\mu_0}$.

1.2 The Lorentz oscillator model

The equation of motion for an electron in an oscillating electric field is given by

$$m^*\frac{\mathrm{d}^2x}{\mathrm{d}t^2} + \underbrace{m^*\gamma\frac{\mathrm{d}x}{\mathrm{d}t}}_{\text{Damping}} + \underbrace{m^*\omega_0^2x}_{\text{Restoring}} = \underbrace{-eE_0\exp(-i\omega t)}_{\text{Driving}}. \tag{5}$$

Assume $x(t)$ has the same temporal dependence on the driving field:

$$x(t) = x_0\exp(-i\omega t), \tag{6}$$

and substituting this into Eqn. (5), we find that

$$x(t) = -\frac{e}{m^*}\frac{1}{\omega_0^2 - \omega^2 - i\gamma\omega}E(t). \tag{7}$$

The **polarisation** field is

$$\boldsymbol{P} = -Ne\boldsymbol{x}(t) = \frac{e^2N}{m^*}\frac{1}{\omega_0^2 - \omega^2 - i\gamma\omega}\boldsymbol{E}(t) = \varepsilon_0\chi^{(1)}\boldsymbol{E}, \tag{8}$$

where N is the density of oscillators per unit volume. Therefore, the linear susceptibility $\chi^{(1)}$ and the relative permittivity ε_r can be expressed as

$$\chi^{(1)} = \frac{e^2N}{\varepsilon_0m^*}\frac{1}{\omega_0^2 - \omega^2 - i\gamma\omega} = \frac{\omega_p^2}{\omega_0^2 - \omega^2 - i\gamma\omega}, \tag{9}$$

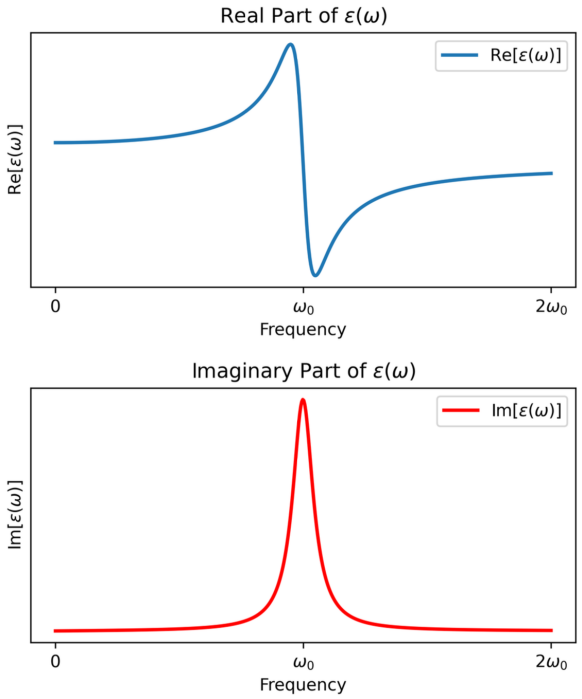

Figure 1: Real and imaginary parts of the dielectric constant ε_r derived from the Lorentz model.

$$\varepsilon_r = 1 + \chi^{(1)} = 1 + \frac{\omega_p^2}{\omega_0^2 - \omega^2 - i\gamma\omega}, \tag{10}$$

where $\omega_p = \sqrt{Ne^2/m^*\varepsilon_0}$ is known as the plasma frequency. The real and imaginary parts of the dielectric constant ε_r are

$$\mathrm{Re}(\varepsilon_r) = 1 + \frac{(\omega_0^2 - \omega)\omega_p^2}{(\omega_0^2 - \omega^2)^2 + \gamma^2\omega^2}, \tag{11}$$

$$\mathrm{Im}(\varepsilon_r) = \frac{\gamma\omega\omega_p^2}{(\omega_0^2 - \omega^2)^2 + \gamma^2\omega^2}. \tag{12}$$

The refractive index n is defined as the ratio of the speed of light in a vacuum to the speed of light in the dielectric

$$n = \frac{c}{v} = \sqrt{\varepsilon_r} = n_r + in_i, \tag{13}$$

which varies with frequency (wavelength). This is known as **dispersion**. Considering the wave propagation, the dispersion in the refractive index has two distinct regimes. When

$$\frac{\partial n}{\partial \omega} < 0, \tag{14}$$

the dispersion is described as **anomalous dispersion**. This occurs when ω is close to ω_0. When ω is far from ω_0

$$\frac{\partial n}{\partial \omega} > 0, \tag{15}$$

which is described as **normal dispersion**. At low absorption, $n_i \to 0, n \to n_r$ and from the Lorenz model:

$$n = a + \frac{b\lambda^2}{\lambda^2 + c}. \tag{16}$$

This is known as the **Sellmeier equation** and it will prove useful when we need values of the refractive index at different wavelengths.

1.3 Nonlinear polarisation

Assume that the electron displacement is not linear with driving field

$$F_{\text{ah}}(x) = -sx - px^2 - qx^3 - \dots, \tag{17}$$

and the potential

$$V = \frac{1}{2}sx^2 + \frac{1}{3}px^3 + \dots. \tag{18}$$

The Nonlinear polarisation then becomes

$$P = \varepsilon_0 \left(\chi^{(1)}E + \chi^{(2)}E^2 + \chi^{(3)}E^3 + \dots\right). \tag{19}$$

We can write the input field in the form

$$E = E_0 + E_\omega \cos(\omega t), \tag{20}$$

and substitute this into the nonlinear polarisation then we get

$$\begin{aligned} P &= \varepsilon_0 \left(\chi^{(1)}E + \chi^{(2)}E^2 + \chi^{(3)}E^3 + \dots\right) \\ &= \varepsilon_0\chi^{(1)}(E_0 + E_\omega\cos(\omega t)) && \text{(linear/first order)} \\ &\quad + \varepsilon_0\chi^{(2)}(E_0 + E_\omega\cos(\omega t))^2 && \text{(second order)} \\ &\quad + \varepsilon_0\chi^{(3)}(E_0 + E_\omega\cos(\omega t))^3 && \text{(third order)} \\ &\quad + \dots. \end{aligned} \tag{21}$$

2 Coupled wave equations for nonlinear optical media

The propagation of an EM wave can be described by the wave equation:

$$\boldsymbol{\nabla}^2 \boldsymbol{E} = \mu_0 \frac{\partial^2}{\partial t^2}(\varepsilon_0 \boldsymbol{E} + \boldsymbol{P}). \tag{22}$$

Restrict this equation to one-dimension then we have

$$\frac{\partial^2 E}{\partial z^2} - \mu_0 \varepsilon_0 \frac{\partial^2 E}{\partial t^2} = \mu_0 \frac{\partial^2 P}{\partial t^2}. \tag{23}$$

The polarisation P can be expressed as

$$P = P^L + P^{NL} = \varepsilon_0 \chi^{(1)} E + \varepsilon_0 \chi^{(2)} E^2 + \varepsilon_0 \chi^{(3)} E^3 + \dots. \tag{24}$$

Take $E(z,t)$ and $P(z,t)$, both as scalar plane waves, to be the sum of discrete frequency components:

$$E(z,t) = \sum_n \frac{1}{2} \left[\tilde{E}_n(z) \mathrm{e}^{i(\omega_n t - k_n z)} + \text{c.c.} \right], \tag{25}$$

$$P(z,t) = \sum_n \frac{1}{2} \left[\tilde{P}_n(z) \mathrm{e}^{i(\omega_n t - k_n z)} + \text{c.c.} \right], \tag{26}$$

where $\tilde{E}_n(z)$ and $\tilde{P}_n(z)$ are the complex amplitudes of the electric field and polarisation at the frequency ω_n. Substitute these equations in to the one-dimensional wave equation,

$$\frac{\partial^2 E_n}{\partial z^2} - \frac{n^2}{c^2} \frac{\partial^2 E_n}{\partial t^2} = \mu_0 \frac{\partial^2 P_n^{NL}}{\partial t^2}, \tag{27}$$

where

$$\frac{\partial E_n}{\partial z} = \frac{1}{2} \left[-i k_n \tilde{E}_n \mathrm{e}^{i(\omega_n t - k_n z)} + \frac{\partial \tilde{E}}{\partial z} \mathrm{e}^{i(\omega_n t - k_n z)} + \text{c.c.} \right], \tag{28}$$

$$\frac{\partial^2 E_n}{\partial z^2} = \frac{1}{2} \left[-k_n^2 \tilde{E}_n \mathrm{e}^{i(\omega_n t - k_n z)} - 2 i k_n \frac{\partial \tilde{E}_n}{\partial z} \mathrm{e}^{i(\omega_n t - k_n z)} + \frac{\partial^2 \tilde{E}}{\partial z^2} \mathrm{e}^{i(\omega_n t - k_n z)} + \text{c.c.} \right], \tag{29}$$

$$\frac{\partial^2 E_n}{\partial t^2} = \frac{1}{2} \left[-\omega^2 \tilde{E}_n \mathrm{e}^{i(\omega_n t - k_n z)} + \text{c.c.} \right], \tag{30}$$

$$\frac{\partial^2 P_n^{NL}}{\partial t^2} = \frac{1}{2} \left[-\omega^2 \tilde{P}_n^{NL} \mathrm{e}^{i(\omega_n t - k_n z)} + \text{c.c.} \right], \tag{31}$$

which we can combine in the following partial differential equation

$$\frac{\partial^2 \tilde{E}_n}{\partial z^2} - 2 i k_n \frac{\partial \tilde{E}_n}{\partial z} - \left(k_n^2 - \frac{n_n^2 \omega_n^2}{c^2} \right) \tilde{E}_n(z) = -\mu_0 \omega_n^2 \tilde{P}_n^{NL}(z). \tag{32}$$

The **slowly varying envelope approximation (SVEA)**, i.e. $\frac{\partial^2 \tilde{E}_n}{\partial z^2} \approx 0$, significantly simplifies the analysis of the equation. Finally we get

$$\boxed{\frac{\partial \tilde{E}_n}{\partial z} = \frac{-i\omega_n}{2\varepsilon_0 c n_n} \tilde{P}_n^{NL}(z).} \tag{33}$$

3 Second harmonic generation

3.1 Coupled wave equations for SHG

Consider the second harmonic generation, the total fields present in the crystal can be expressed as

$$E(z,t) = \frac{1}{2}\left(\tilde{E}_\omega \mathrm{e}^{i(\omega t - k_\omega z)} + \tilde{E}_{2\omega}\mathrm{e}^{i(2\omega t - k_{2\omega} z)} + \text{c.c.}\right). \tag{34}$$

Considering only the second order effects, the nonlinear polarisation is

$$\begin{aligned} P^{NL}(z,t) =& \varepsilon_0 \chi^{(2)} E^2(z,t) \\ =& \frac{1}{4}\varepsilon_0\chi^{(2)}\left(\tilde{E}_\omega \mathrm{e}^{i(\omega t - k_\omega z)} + \tilde{E}_{2\omega}\mathrm{e}^{i(2\omega t - k_{2\omega} z)} + \text{c.c.}\right)^2 \\ =& \frac{1}{2}\left(\tilde{P}^{NL}_\omega \mathrm{e}^{i(\omega t - k_\omega z)} + \tilde{P}^{NL}_{2\omega}\mathrm{e}^{i(2\omega t - k_{2\omega} z)} + \dots\right) + \text{c.c.}, \end{aligned} \tag{35}$$

where the fundamental (pump) term and the second harmonic term are

$$\tilde{P}^{NL}_\omega(z) = \varepsilon_0\chi^{(2)}\tilde{E}_{2\omega}\tilde{E}^*_\omega \mathrm{e}^{-i(k_{2\omega}-2k_\omega)z} = \varepsilon_0\chi^{(2)}\tilde{E}_{2\omega}\tilde{E}^*_\omega \mathrm{e}^{-i\Delta k z}, \tag{36}$$

$$\tilde{P}^{NL}_{2\omega}(z) = \frac{1}{2}\varepsilon_0\chi^{(2)}\tilde{E}_\omega\tilde{E}_\omega \mathrm{e}^{i(k_{2\omega}-2k_\omega)z} = \frac{1}{2}\varepsilon_0\chi^{(2)}\tilde{E}^2_\omega \mathrm{e}^{i\Delta k z}. \tag{37}$$

Here, $\Delta k = k_{2\omega} - 2k_\omega$ is the wave vector mismatch. Substituting the expressions into Eqn. (33), then

$$\frac{\partial \tilde{E}_\omega}{\partial z} = \frac{-i\omega\chi^{(2)}}{2cn_\omega}\tilde{E}_{2\omega}\tilde{E}^*_\omega \mathrm{e}^{-i\Delta k z}, \tag{38}$$

$$\frac{\partial \tilde{E}_{2\omega}}{\partial z} = \frac{-i\omega\chi^{(2)}}{2cn_{2\omega}}\tilde{E}^2_\omega \mathrm{e}^{i\Delta k z}, \tag{39}$$

with the wave vector mismatch

$$\Delta k = k_{2\omega} - 2k_\omega = \frac{(2\omega)n_{2\omega}}{c} - 2\frac{\omega n_\omega}{c} = \frac{2\omega}{c}(n_{2\omega} - n_\omega). \tag{40}$$

For normal dispersion $n_{2\omega} > n_\omega$, so there would be a phase mismatch term between $E_{2\omega}$ and $P_{2\omega}$.

3.2 Low pump depletion approximation

For efficiencies $\eta_{SHG} = I_{2\omega}/I_\omega \ll 1$, the pump $\tilde{E}_\omega$ can be considered constant. The equations become

$$\frac{\partial \tilde{E}_\omega}{\partial z} = 0, \quad \frac{\partial \tilde{E}_{2\omega}}{\partial z} = \frac{-i\omega\chi^{(2)}}{2cn_{2\omega}}\tilde{E}^2_\omega \mathrm{e}^{i\Delta k z}. \tag{41}$$

We can solve the equation by integrating

$$\tilde{E}_{2\omega}(L) = \frac{-i\omega\chi^{(2)}}{2cn_{2\omega}}\tilde{E}^2_\omega \int_0^L \mathrm{e}^{i\Delta k z}\mathrm{d}z = \frac{-i\omega\chi^{(2)}}{2cn_{2\omega}}\tilde{E}^2_\omega \mathrm{e}^{i\frac{\Delta k L}{2}} L\mathrm{sinc}\left(\frac{\Delta k L}{2}\right). \tag{42}$$

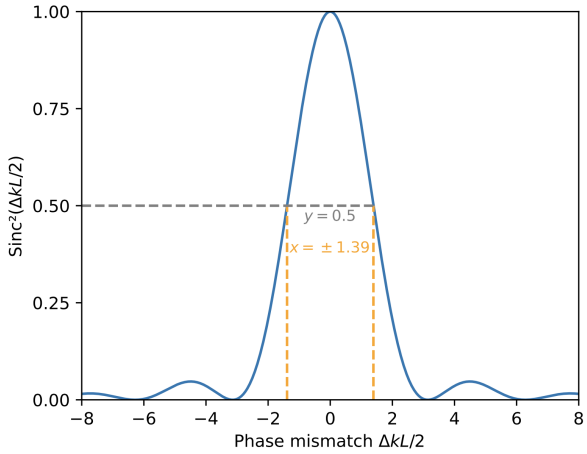

Figure 2: The $\mathrm{sinc}^2(\Delta kL/2)$ function.

Recall the intensity

$$I = \frac{1}{2} n \varepsilon_0 c |E|^2, \tag{43}$$

which allows us to write

$$I_\omega = \frac{1}{2} n_\omega \varepsilon_0 c \tilde{E}_\omega^2, \tag{44}$$

$$I_{2\omega} = \frac{1}{2} n_{2\omega} \varepsilon_0 c \frac{\omega^2 (\chi^{(2)})^2}{4c^2 n_{2\omega}^2} \tilde{E}_\omega^4 L^2 \mathrm{sinc}^2\left(\frac{\Delta kL}{2}\right) = \frac{1}{2} \frac{\omega^2 (\chi^{(2)})^2 I_\omega^2}{\varepsilon_0 c^3 n_\omega^2 n_{2\omega}} L^2 \mathrm{sinc}^2\left(\frac{\Delta kL}{2}\right), \tag{45}$$

which can also be considered in terms of efficiency

$$\eta_{SHG} = \frac{I_{2\omega}}{I_\omega} = \frac{1}{2} \frac{\omega^2 (\chi^{(2)})^2 I_\omega}{\varepsilon_0 c^3 n_\omega^2 n_{2\omega}} L^2 \mathrm{sinc}^2\left(\frac{\Delta kL}{2}\right) \propto L^2 \mathrm{sinc}^2\left(\frac{\Delta kL}{2}\right) = \frac{\sin^2(\frac{\Delta kL}{2})}{(\frac{\Delta k}{2})^2}. \tag{46}$$

If phase matched, i.e. $\Delta k = 0$, the process is described as being *phase matched*. If the process is not phase matched, the first maximum in the sinusoid occurs when

$$\frac{\Delta kL}{2} = \frac{\pi}{2}. \tag{47}$$

We use this to define the **coherence length**, L_{coh}, which is the shortest length of the crystal which will *maximise* the second harmonic generated for a given wavevector mismatch, that is:

$$L_{coh} = \frac{\pi}{\Delta k}. \tag{48}$$

3.3 Photon picture and Manley-Rowe relations

Here two photons at frequency ω are annihilated and one photon at frequency 2ω is created. The energy conservation and momentum conservation give

$$\hbar\omega + \hbar\omega = \hbar 2\omega, \quad \hbar k_\omega + \hbar k_\omega = \hbar k_{2\omega}, \tag{49}$$

which corresponds to the phase matching requirement $2k_\omega = k_{2\omega}$ that we have already identified. The photon flux is also conserved:

$$\frac{\mathrm{d}\Phi_\omega}{\mathrm{d}z} = -2\frac{\mathrm{d}\Phi_{2\omega}}{\mathrm{d}z}, \tag{50}$$

where Φ_ω is the photon flux of the fundamental, and $\Phi_{2\omega}$ is the photon flux of the second harmonic. The intensity

$$I_i = \Phi_i \hbar\omega_i, \tag{51}$$

So we can write the relation

$$\boxed{\frac{\mathrm{d}I_\omega}{\mathrm{d}z} = -\frac{\mathrm{d}I_{2\omega}}{\mathrm{d}z},} \tag{52}$$

which is known as the **Manley-Rowe relations**. This relation can also derived from classical field:

$$\frac{\mathrm{d}I_\omega}{\mathrm{d}z} = \frac{\mathrm{d}}{\mathrm{d}z}\left(\frac{1}{2}n_\omega\varepsilon_0 c\tilde{E}_\omega\tilde{E}_\omega^*\right) = \frac{1}{2}n_\omega\varepsilon_0 c\left(\tilde{E}_\omega\frac{\partial\tilde{E}_\omega^*}{\partial z} + \tilde{E}_\omega^*\frac{\partial\tilde{E}_\omega}{\partial z}\right). \tag{53}$$

Combined with couples equations we can get the equivalent results.

3.4 SHG pump depletion

The coupled wave equationa are

$$\frac{\partial\tilde{E}_\omega}{\partial z} = \frac{-i\omega\chi^{(2)}}{2cn_\omega}\tilde{E}_{2\omega}\tilde{E}_\omega^*\mathrm{e}^{-i\Delta kz}, \tag{54}$$

$$\frac{\partial\tilde{E}_{2\omega}}{\partial z} = \frac{-i\omega\chi^{(2)}}{2cn_{2\omega}}\tilde{E}_\omega^2\mathrm{e}^{i\Delta kz}. \tag{55}$$

If we have high levels of SHG, we can no longer ignore the corresponding reduction in $\tilde{E}_\omega$. Let's start with $\Delta k = 0$, and take $\chi^{(2)}$ and n to be equal for each frequency.

$$\frac{\partial\tilde{E}_\omega}{\partial z} = -i\kappa\tilde{E}_{2\omega}\tilde{E}_\omega^*, \quad \frac{\partial\tilde{E}_{2\omega}}{\partial z} = -i\kappa\tilde{E}_\omega^2, \tag{56}$$

where $\kappa = \frac{\omega\chi^{(2)}}{2cn}$. $\tilde{E}$ is a complex amplitude

$$\tilde{E}_\omega = E_\omega\mathrm{e}^{i\phi_\omega}, \quad \tilde{E}_{2\omega} = E_{2\omega}\mathrm{e}^{i\phi_{2\omega}}, \tag{57}$$

and substitute these into coupled equations giving

$$\frac{\partial E_\omega}{\partial z} = -i\kappa E_{2\omega}E_\omega^*\mathrm{e}^{-i(2\phi_\omega-\phi_{2\omega})}, \tag{58}$$

$$\frac{\partial E_{2\omega}}{\partial z} = -i\kappa E_\omega^2\mathrm{e}^{i(2\phi_\omega-\phi_{2\omega})}. \tag{59}$$

E_ω and $E_{2\omega}$ are real, so the exponent terms $\mathrm{e}^{i(2\phi_\omega-\phi_{2\omega})} = i$.

$$\frac{\partial E_\omega}{\partial z} = -\kappa E_{2\omega}E_\omega, \quad \frac{\partial E_{2\omega}}{\partial z} = \kappa E_\omega^2. \tag{60}$$

The Manley-Rowe energy conservation gives

$$\frac{\mathrm{d}}{\mathrm{d}z}(I_\omega + I_{2\omega}) = 0, \tag{61}$$

so we have

$$I_0 = I_\omega + I_{2\omega} = \frac{1}{2}c\varepsilon_0 n\left(E_\omega^2 + E_{2\omega}^2\right) \quad \Rightarrow \quad E_\omega^2 = \frac{2I_0}{c\varepsilon_0 n} - E_{2\omega}^2. \tag{62}$$

Therefore,

$$\frac{\partial E_{2\omega}}{\partial z} = \kappa\left(\frac{2I_0}{c\varepsilon_0 n} - E_{2\omega}^2\right). \tag{63}$$

For a nonlinear crystal of length L, integrating the equation gives

$$\int_0^{E_{2\omega}(L)} \frac{\mathrm{d}E_{2\omega}(z)}{\left(\frac{2I_0}{c\varepsilon_0 n} - E_{2\omega}^2\right)} = \int_0^L \kappa \mathrm{d}z, \tag{64}$$

and the solution is

$$E_{2\omega}(L) = \sqrt{\frac{2I_0}{c\varepsilon_0 n}}\tanh\left(\sqrt{\frac{2I_0}{c\varepsilon_0 n}}\kappa L\right). \tag{65}$$

So the intensity

$$I_{2\omega} = \frac{1}{2}n\varepsilon_0 c E_{2\omega}^2 = I_0\tanh^2(\Gamma L), \tag{66}$$

where the coupling coefficient Γ is

$$\Gamma = \sqrt{\frac{2I_0}{c\varepsilon_0 n}}\kappa, \quad \kappa = \frac{\omega\chi^{(2)}}{2cn}. \tag{67}$$

Finally the pump expression is

$$I_\omega = I_0 - I_{2\omega} = I_0\left(1 - \tanh^2(\Gamma L)\right). \tag{68}$$

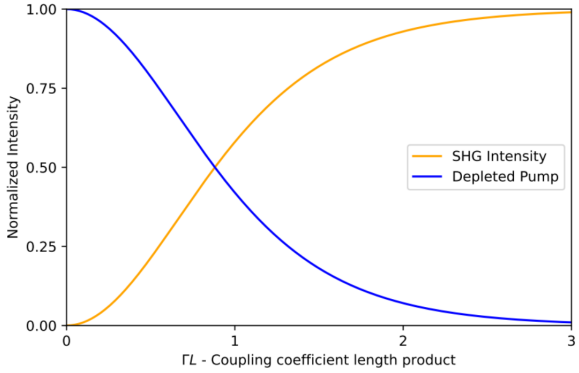

Figure 3: Pump depletion solution.

4 Crystal optics

4.1 Light in Birefringent media

We need to find a way of achieving $\Delta k = 0$. In SHG phase matching states

$$k_{2\omega} - 2k_{\omega} = 0 \quad \Rightarrow \quad n_{2\omega} = n_{\omega}. \tag{69}$$

Yet dispersion tells us that $n_{2\omega} > n_{\omega}$ for a given material (assuming normal dispersion). To get around this we need to consider whether a material could have more than one value of refractive index for a given ω. This was touched on earlier when it was noted that materials might have different refractive indices for light with its electric field polarised in different directions. This property is known as **birefringence**.
In general $n_x \neq n_y \neq n_z$, but we consider the slightly simpler situation in a *uniaxial* crystal where two of the values are equal. The unique direction is called the **optic axis** (denoted c) with principal extraodinary refractive index $\tilde{n}_e$, while the refractive index for light polarised orthogonally is n_o. A uniaxial crystal can be categorized as *positive* or *negative*:

$$\tilde{n}_e > n_o \quad \text{positive uniaxial}, \tag{70}$$
$$\tilde{n}_e < n_o \quad \text{negative uniaxial}. \tag{71}$$

If we designate the optic axis c to be in the z-direction and suppose the situation when light is propagating along the direction of the wavevector $\boldsymbol{k}$ at an angle θ with respect to the optic axis c, the refractive index satisfies

$$n_e^2(\theta)\left(\frac{\cos^2\theta}{n_o^2} + \frac{\sin^2\theta}{\tilde{n}_e^2}\right) = 1, \tag{72}$$

which gives

$$n_e(\theta) = \left(\frac{\cos^2\theta}{n_o^2} + \frac{\sin^2\theta}{\tilde{n}_e^2}\right)^{-\frac{1}{2}}. \tag{73}$$

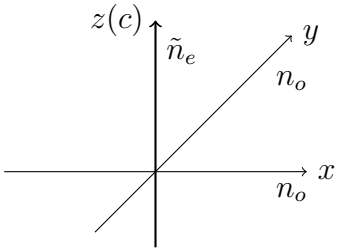

Figure 4: An uniaxial crystal with optic axis designated to be in the z-direction.

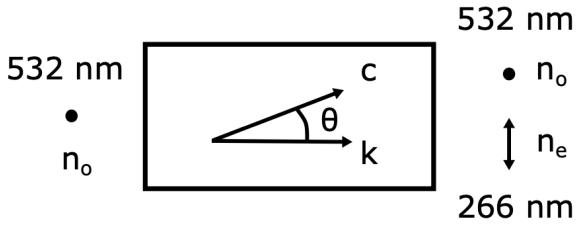

Figure 5: Schematic for Type I phase matching SHG of 532 nm light in a β-BBO crystal. Indicating the wavelengths involved, the relative polarisations of the beams, the optic axis c and the cut angle θ relative to the wavevector k.

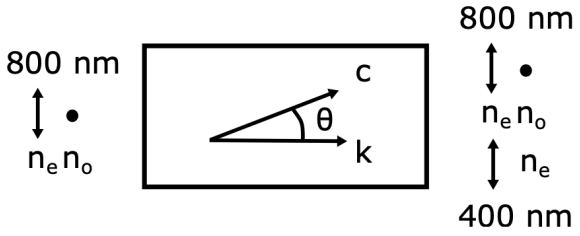

Figure 6: Schematic for Type II oee phase matching for SHG of 800 nm light in an ADP crystal. Indicating the wavelengths involved, the relative polarisations of the beams, the optic axis c and the cut angle θ relative to the wavevector k.

4.2 Phase matching in Birefringent media

4.2.1 Type I phase matching - SHG in BBO

In Type I phase matching for SHG the pump experiences the higher refractive index:

(1) In a positive uniaxial crystal ($\tilde{n}_e > n_o$), the pump experiences $n_e(\omega, \theta_I)$, the SH experiences $n_o(2\omega)$.

(2) In a negative uniaxial crystal ($\tilde{n}_e < n_o$), the pump experiences $n_o(\omega)$, the SH experiences $n_e(2\omega, \theta_I)$

Most nonlinear crystals we will encounter happen to be negative uniaxial, therefore for phase matching we want

$$n_e(2\omega, \theta_I) = n_o(\omega). \tag{74}$$

4.2.2 Type II phase matching - SHG in ADP

Type II phase matching is subtly different, with the pump having both ordinary and extraordinary components. This means we can have a positive crystal (oeo) or a negative crystal (oee). Considering the negative (oee) situation the phase matching requires

$$\Delta k_{II} = k_e(2\omega) - (k_e(\omega) + k_o(\omega)), \tag{75}$$

which is equivalent to

$$n_e(2\omega, \theta_{II}) = \frac{n_e(\omega, \theta_{II}) + n_o(\omega)}{2}. \tag{76}$$

4.2.3 Beam walk off

Walk-off arises from the fact that the energy flow is given by the Poynting vector:

$$\boldsymbol{S} = \boldsymbol{E} \times \boldsymbol{H}, \tag{77}$$

which is not the same as the direction of propagation

$$\boldsymbol{k} = \boldsymbol{D} \times \boldsymbol{H}. \tag{78}$$

Beam walk off can not be avoid in critical (angular) phase matching, and has some negative side effects:

(1) Reduction in conversion efficiency due to spatial overlap reduction with increasing walk-off (reduced interaction length).

(2) The spatial profile of the beam produced will be wider than that of the original beams.

The walk-off angle is given by

$$\rho = -\frac{1}{n_e}\frac{\partial n_e}{\partial \theta}. \tag{79}$$

4.2.4 Non-critital phase matching (NCPM)

The phase matching considered so far (Type I and Type II) is known as critical phase matching. These techniques rely on achieving phase matching through angular tuning, but as we saw, they have the disadvantage of beam walk-off. We will now discuss an alternative approach, known as **non-critical phase matching**, to achieve $\Delta k = 0$ and avoid walk-off.
Consider the situation where in Type I phase matching we have $\theta_I = 90^\circ$ and $n_e(2\omega, 90^\circ, T) = \tilde{n}_e(2\omega, T)$. Then we can tune the refractive indices with temperature. For example Lithium Niobate ($LiNbO_3$) has the following:

$$n_o(\lambda) = n_o(23^\circ) + \frac{\partial n_o}{\partial T}(T - 23^\circ) = 2.239 + 1 \times 10^{-5}(T - 23^\circ), \tag{80}$$

$$\tilde{n}_e(\lambda/2) = \tilde{n}_e(23^\circ) + \frac{\partial \tilde{n}_e}{\partial T}(T - 23^\circ) = 2.235 + 6.67 \times 10^{-5}(T - 23^\circ). \tag{81}$$

5 Nonlinear optics with real beams

5.1 A pulsed laser source

Real beams are Gaussian laser pulses with a temporal width of $\Delta\nu$ and a frequency width of Δt. The bandwidth theorem tells us that:

$$\Delta\nu\Delta t = 0.441. \tag{82}$$

The pump and SH pulses (both made up of a range of optical frequencies) propagate through the crystal at different speeds given by the group velocity:

$$v_g(\omega_0) = \left.\frac{\partial\omega}{\partial k}\right|_{\omega_0}. \tag{83}$$

If we consider the limit that (after propagating through a crystal of length L) the pulses are out of step by the pulse width (Δt) then

$$\Delta t = \frac{L}{v_g(2\omega)} - \frac{L}{v_g(\omega)} = L \times \text{GVM}, \tag{84}$$

where GVM is the **group velocity mismatch** and

$$\text{GVM} = \frac{1}{v_g(2\omega)} - \frac{1}{v_g(\omega)}. \tag{85}$$

The **phase matching bandwidth** is

$$\Delta\omega = 2\pi\Delta\nu = \frac{2\pi\ 0.441}{\Delta t} = \frac{2\pi\ 0.441}{L \times \text{GVM}}, \tag{86}$$

or equivalently in wavelength

$$\Delta\lambda = \frac{\lambda_0^2}{c}\Delta\nu = \frac{0.441\lambda_0^2}{cL \times \text{GVM}}. \tag{87}$$

5.2 Focussed beams

Focused Gaussian beams have a wavevector spread Δk. Recall that the SHG efficiency has the form

$$\eta_{SHG} \propto L^2\text{sinc}^2\left(\frac{\Delta kL}{2}\right), \tag{88}$$

and we discussed that this falls to 50% when

$$\frac{\Delta kL}{2} = \pm 1.39 \quad \Rightarrow \quad |\Delta k| = \frac{2 \times 1.39}{L}. \tag{89}$$

We anticipate that the changes will be small so we can expand Δk as a Taylor Series around the phase matching angle θ_{PM} as follows:

$$\Delta k(\theta) = \Delta k(\theta_{PM}) + \left.\frac{\partial\Delta k}{\partial\theta}\right|_{\theta_{PM}}(\theta - \theta_{PM}) + \frac{1}{2}\left.\frac{\partial^2\Delta k}{\partial\theta^2}\right|_{\theta_{PM}}(\theta - \theta_{PM})^2 + \ldots. \tag{90}$$

As the system is assumed to be phase matched at normal incidence, $\Delta k(\theta_{PM}) = 0$. Then

$$|\Delta k| = \left|\frac{\partial \Delta k}{\partial \theta}\right|_{\theta_{PM}} (\theta - \theta_{PM}). \tag{91}$$

So the full cone angle

$$\Delta\theta = 2(\theta - \theta_{PM}) = \frac{2|\Delta k|}{\frac{\partial \Delta k}{\partial \theta}\big|_{\theta_{PM}}}. \tag{92}$$

So from our criterion for the acceptance angle (efficiency $\leq 50\%$) we get

$$\Delta\theta_{ACC} = \frac{4 \times 1.39}{L\frac{\partial \Delta k}{\partial \theta}\big|_{\theta_{PM}}}, \tag{93}$$

where

$$\begin{aligned} \frac{\partial \Delta k}{\partial \theta} =& \frac{\partial}{\partial \theta}\left[\frac{2\omega}{c}(n_e(2\omega,\theta) - n_o(\omega))\right] = \frac{2\omega}{c}\frac{\partial n_e(2\omega,\theta)}{\partial \theta} \\ =& \frac{\omega}{c} n_e^3(2\omega,\theta)\sin(2\theta)\left(\frac{1}{n_o^2(2\omega)} - \frac{1}{\tilde{n}_e^2(2\omega)}\right). \end{aligned} \tag{94}$$

6 Other second order processes

6.1 Three-wave mixing processes

Three waves in a crystal gives the polarisation

$$P^{NL}(z,t) = \varepsilon_0 \chi^{(2)} (E_1(z,t) + E_2(z,t) + E_3(z,t))^2, \tag{95}$$

where

$$E_j(z,t) = \frac{1}{2}\left(\tilde{E}_j \mathrm{e}^{i(\omega_j t - k_j z)} + \tilde{E}_j^* \mathrm{e}^{-i(\omega_j t - k_j z)}\right). \tag{96}$$

Assuming E_3 is a weak field, so the nonlinear polarisation

$$P^{NL}(z,t) = \varepsilon_0 \chi^{(2)} E^2_{\text{total}}(z,t), \tag{97}$$

where

$$\begin{aligned} E^2_{\text{total}}(z,t) =& \frac{1}{4}\left(\tilde{E}_1 \mathrm{e}^{i(\omega_1 t - k_1 z)} + \tilde{E}_1^* \mathrm{e}^{-i(\omega_1 t - k_1 z)} + \tilde{E}_2 \mathrm{e}^{i(\omega_2 t - k_2 z)} + \tilde{E}_2^* \mathrm{e}^{-i(\omega_2 t - k_2 z)}\right)^2 \\ =& + \frac{1}{4}\left(\tilde{E}_1^2 \mathrm{e}^{(i(2\omega_1 t - 2k_1 z))} + \tilde{E}_2^2 \mathrm{e}^{(i(2\omega_2 t - 2k_2 z))} + \text{c.c.}\right) \quad \text{SHG} \\ &+ \frac{1}{4}\left(2\tilde{E}_1 \tilde{E}_2 \mathrm{e}^{i((\omega_1+\omega_2)t - (k_1+k_2)z)} + \text{c.c}\right) \quad \text{SFG} \\ &+ \frac{1}{4}\left(2\tilde{E}_1 \tilde{E}_2^* \mathrm{e}^{i((\omega_1-\omega_2)t - (k_1-k_2)z)} + \text{c.c}\right) \quad \text{DFG} \\ &+ \frac{1}{2}|\tilde{E}_1|^2 + \frac{1}{2}|\tilde{E}_2^2|. \quad \text{DC} \end{aligned} \tag{98}$$

6.2 Sum frequency generation (SFG)

Let us know look at SFG in more detail. First recall the general form of the coupled nonlinear equations:

$$\frac{\partial \tilde{E}_n}{\partial z} = \frac{-i\omega_n}{2\varepsilon_0 c n_n} \tilde{P}^{NL}(z). \tag{99}$$

Taking $\omega_3 = \omega_1 + \omega_2$, and we can write

$$\frac{\partial \tilde{E}_3}{\partial z} = \frac{-i\omega_3}{2\varepsilon_0 c n_3} \varepsilon_0 \chi^{(2)} \tilde{E}_1 \tilde{E}_2 \mathrm{e}^{i\Delta k z} = \frac{-i\omega_3 \chi^{(2)}}{2c n_3} \tilde{E}_1 \tilde{E}_2 \mathrm{e}^{i\Delta k z}, \tag{100}$$

where the wave vector mismatch $\Delta k = k_3 - (k_1 + k_2)$. Integrating this equation and we get

$$\tilde{E}_3 = \frac{-i\omega_3 \chi^{(2)}}{2c n_3} \tilde{E}_1 \tilde{E}_2 \mathrm{e}^{i\frac{\Delta k L}{2}} L \mathrm{sinc}\left(\frac{\Delta k L}{2}\right), \tag{101}$$

Then the intensity

$$I_3 \propto (\chi^{(2)})^2 I_1 I_2 L^2 \mathrm{sinc}^2\left(\frac{\Delta k L}{2}\right). \tag{102}$$

For the phase matching $k_3 = k_1 + k_2$, we have

$$n_3\omega_3 = n_1\omega_1 + n_2\omega_2, \quad \text{or,} \quad \frac{n_3}{\lambda_3} = \frac{n_1}{\lambda_1} + \frac{n_2}{\lambda_2}. \tag{103}$$

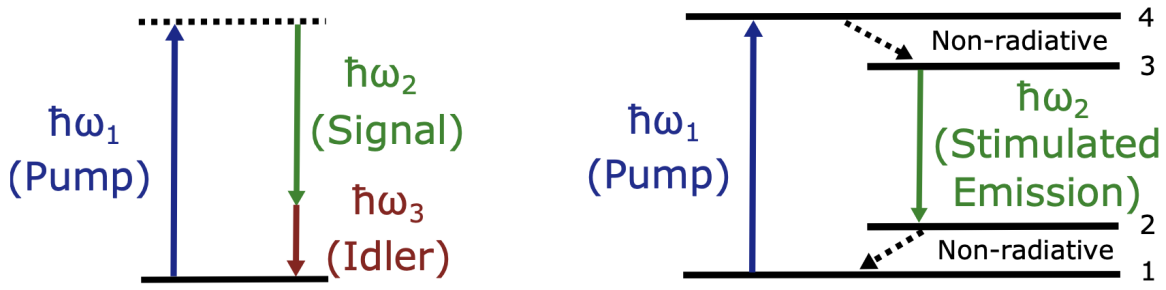

Figure 7: Comparison between (a) an OPA providing optical gain based on nonlinear process and (b) an optical amplifier providing optical gain based on stimulated emission. Both can provide optical gain, but with very different properties.

6.3 Difference frequency generation (DFG)

Similarly with SFG, the third field and the intensity

$$\tilde{E}_3 = \frac{-i\omega_3\chi^{(2)}}{2cn_3}\tilde{E}_1\tilde{E}_2^* e^{i\frac{\Delta kL}{2}} L\text{sinc}\left(\frac{\Delta kL}{2}\right), \tag{104}$$

$$I_3 \propto (\chi^{(2)})^2 I_1 I_2 L^2 \text{sinc}^2\left(\frac{\Delta kL}{2}\right), \tag{105}$$

where the wave vector mismatch is $\Delta k = k_3 - (k_1 - k_2)$. Using DFG, we can build very useful and versatile light sources, such as optical parametric amplification (OPA) and optical parametric oscillator (OPO).

6.4 Optical parametric amplification (OPA)

The OPA process based on DFG in particular can be summarised as follows:

(1) Introduce a **signal** beam at ω_2.

(2) Provide a **pump** beam at ω_1.

(3) The pump photon at ω_1 annihilates.

(4) A new photon is created at ω_3 which is phase matched with the signal beam. This is called the **idler**. Often (but not always!), the idler is only present to phase and energy match the three wave process, and it may be discarded and not used. (Be careful this is not always true!)

(5) A new photon at ω_2 is created - the signal beam is amplified!

In the OPA process, the signal beam

$$\tilde{E}_2(z) = \tilde{E}_2(0)\cosh(\kappa z), \quad \kappa \propto \chi^{(2)}\tilde{E}_1. \tag{106}$$

Note that $\cosh(\kappa z)$ has the following approximations:

$$\kappa z \ll 1, \quad \cosh(\kappa z) \to 1 + \kappa^2 z^2, \tag{107}$$

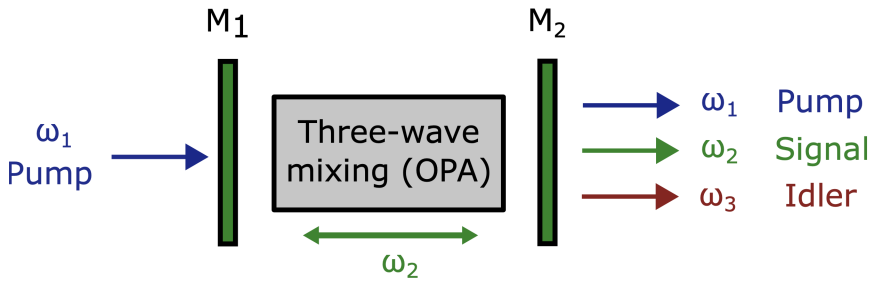

Figure 8: A general schematic of an OPO resonant at the signal frequency ω_2. The mirror M_1 is completely transparent to the pump and highly reflective to the signal. M_2 will also be highly reflective of the signal, but slightly less than M_1 to allow for the output coupling of the required signal light.

$$\kappa z \gg 1, \quad \cosh(\kappa z) \to \mathrm{e}^{\kappa z}. \tag{108}$$

Hence in an optimised, phase-matched arrangement the intensity of the light at ω_2 grows as

$$I(\omega_2, z) = I(\omega_2, 0)\mathrm{e}^{\gamma z}, \quad \gamma \propto \chi^{(2)}\sqrt{I(\omega_1)}. \tag{109}$$

For the OPA:

- Conversion via a virtual state - no energy is stored.
- No energy is lost to the medium (energy conservation) - except through absorption/scattering present in all materials.
- Bandwidth depends on phase matching bandwidth (and transparency of the crystal).
- Pump and signal must overlap temporally and spatially.

For the stimulated emission amplifier:

- Pump to upper energy level - energy stored in medium.
- Non-radiative transitions - energy absorbed, heating the medium.
- Bandwidth limited by transition linewidth in the host material.

6.5 Optical parametric oscillator (OPO)

The gain from the OPA and the feedback from the cavity cause it to oscillate, providing coherent radiation like a laser. This is called an optical parametric oscillator (OPO). Within the cavity there are the three beams necessary for the OPA process:

(1) The **pump** - providing the energy that will provide the gain.

(2) The **signal** - this is resonant in the cavity and will form the mode.

(3) The **idler** - this completes the phase matching in the OPA/DFG process.

The resonant spacing is

$$\Delta\omega = 2\pi\Delta\nu = \frac{2\pi c}{2n_\omega L}. \tag{110}$$

The system will oscillate at the frequency of the longitudinal mode closest to the phase matching condition.

7 Third order processes

7.1 Third harmonic generation (THG)

When we consider the third order processes, the polarisation

$$P = \varepsilon_0 \chi^{(1)} E + \varepsilon_0 \chi^{(2)} E^2 + \varepsilon_0 \chi^{(3)} E^3 \tag{111}$$

In centrosymmetric media, the $\chi^{(2)}$ tern would disappear. In this case

$$P^{NL} = \varepsilon_0 \chi^{(3)} E^3. \tag{112}$$

Focusing on the case where only one pump beam is present which we will write as $E = E_\omega \cos(\omega t)$, the nonlinear polarisation

$$P^{NL} = \varepsilon_0 \chi^{(3)} E_\omega \cos^3(\omega t) = \frac{1}{4} \varepsilon_0 \chi^{(3)} E_\omega \left(3\cos(\omega t) + \cos(3\omega t)\right). \tag{113}$$

The 3ω term is third harmonic generation (THG).

7.2 Intensity dependent refractive index (IDRI)

From the discussions above, the polarisation is

$$P = \varepsilon_0 \left(\chi^{(1)} + \frac{3}{4}\chi^{(3)} E_\omega^2\right) E_\omega \cos(\omega t) + \text{THG}. \tag{114}$$

If we neglect THG for now, then

$$P = \varepsilon_0 \left(\chi^{(1)} + \frac{3}{4}\chi^{(3)} E_\omega^2\right) E_\omega \cos(\omega t) = \varepsilon_0 \chi_{\text{eff}} E_\omega \cos(\omega t), \tag{115}$$

which is known as **Kerr effect**, and the effective susceptibility

$$\chi_{\text{eff}} = \chi^{(1)} + \frac{3}{4}\chi^{(3)} E_0^2 \tag{116}$$

So the refractive index n satisfies

$$n^2 = 1 + \chi_{\text{eff}} = 1 + \chi^{(1)} + \frac{3}{4}\chi^{(3)} E_0^2 = n_L^2 + \frac{3}{4}\chi^{(3)} E_0^2 = n_L^2 \left(1 + \frac{3}{4}\frac{\chi^{(3)}}{n_L^2} E_0^2\right), \tag{117}$$

then

$$n = n_L \left(1 + \frac{3}{4}\frac{\chi^{(3)}}{n_L^2} E_0^2\right)^{\frac{1}{2}}, \quad n_L^2 = 1 + \chi^{(1)}. \tag{118}$$

When $\chi^{(3)} \ll 1$, we can use binomial expansion:

$$n = n_L + \frac{3}{8}\frac{\chi^{(3)}}{n_L} E_0^2 = n_L + \frac{3}{4}\frac{\chi^{(3)}}{n_L^2 c \varepsilon_0} I_0 = n_L + n_2 I_0, \quad n_2 = \frac{3}{4}\frac{\chi^{(3)}}{n_L^2 c \varepsilon_0}. \tag{119}$$

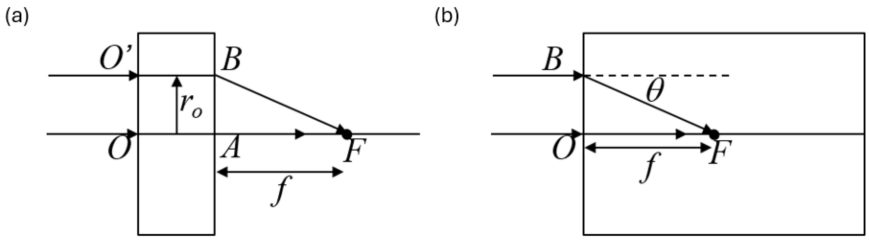

Figure 9: Self focussing arising within (a) a thin and (b) a thick block of material.

7.3 Self-focussing

The fundamental mode of a laser has a Gaussian profile of radius w (at $1/\mathrm{e}^2$) is described by

$$I = I_0 \exp\left(-\frac{2r^2}{w^2}\right). \tag{120}$$

Close to the axis of a Gaussian beam the intensity profile is approximately quadratic in r

$$I = I_0 \exp\left(-\frac{2r^2}{w^2}\right) \approx I_0 \left(1 - \frac{2r^2}{w^2}\right). \tag{121}$$

We can substitute this back into our equation we found for the IDRI as follows

$$n = n_L + n_2 I_0 \left(1 - \frac{2r^2}{w^2}\right). \tag{122}$$

7.3.1 Thin block

The optical path, OP, is given by

$$OP(r) = n(r)L. \tag{123}$$

In a thin block (thickness d of material exhibiting an IDRI through n_2), the optical path

$$OAF = (n_L + n_2 I_0)\, d + f, \tag{124}$$

$$\begin{aligned} O'BF &= \left(n_L + n_2 I_0 - n_2 I_0 \frac{2r_0^2}{w^2}\right) d + \sqrt{f^2 + r_0^2} \\ &= \left(n_L + n_2 I_0 - n_2 I_0 \frac{2r_0^2}{w^2}\right) d + f + \frac{r_0^2}{2f}. \end{aligned} \tag{125}$$

Bringing these equations together and equating the optical paths gives the focal length

$$f = \frac{w^2}{4 n_2 I_0 d} \tag{126}$$

7.3.2 Thick block

If we consider a thick block the situation is slightly different. We assume that the ray incident at B is at the extreme and only experiences the linear refractive index n_L, The optical paths

$$OF = (n_L + n_2 I_0) f, \tag{127}$$

$$BF = \frac{n_L f}{\cos\theta} = n_L f \left(1 + \frac{\theta^2}{2}\right). \tag{128}$$

Equating paths gives the focussing angle

$$\theta = \sqrt{\frac{2n_2 I_0}{n_L}}. \tag{129}$$

For a thick block the focussing of the beam means a greater IDRI which leads to tighter focussing until the spot becomes very small spot with very high intensity. The material will then damage, either through intense heating (thermal damage) or ionisation, both leading to permanent optical damage. This is a problem for high intensity (pulse) propagation in materials exhibiting IDRI.

7.4 Self-phase modulation (SPM)

A Gaussian pulse of full-width half maximum (FWHM) Δt described by

$$I(t) = I_0 \exp\left(-\frac{t^2}{\Delta t^2}\right). \tag{130}$$

If the material exhibits an IDRI then the refractive index experienced by the pulse depends on time and is given by:

$$n(t) = n_L + n_2 I_0 \exp\left(-\frac{t^2}{\Delta t^2}\right). \tag{131}$$

The phase of this wave is described by

$$\phi(t) = \omega_0 t - kz = \omega_0 t - \frac{\omega_0}{c} n(t) z \tag{132}$$

the intensity dependent refractive index changes the phase of the the wave itself as a function of time - this is **self-phase modulation (SPM)**. The frequency

$$\omega = \frac{\partial\phi(t)}{\partial t} = \omega_0 - \frac{\omega_0}{c} z \frac{\partial n(t)}{\partial t}. \tag{133}$$

Therefore, for a nonlinear material of length L, the new spread of frequencies present at the output is given by

$$\omega(t) = \omega_0 + \frac{2\omega_0 n_2 I_0}{c\Delta t^2} L t \mathrm{e}^{-\frac{t^2}{\Delta t^2}}. \tag{134}$$

This tells us that the frequency changes relative to the central frequency ω_0 across the pulse width in time. This is called a frequency chirp. When t close to zero, $\omega(t) - \omega_0 \propto t$, that is to say, a **linear** chirp. For $|t| \gg \Delta t$, then $\omega \to \omega_0$. In between these two limits, there are turning points

$$\frac{\partial^2 \phi(t)}{\partial t^2} \propto \mathrm{e}^{-\frac{t^2}{\Delta t^2}} \left(1 - \frac{2t^2}{\Delta t^2}\right) = 0, \tag{135}$$

which gives

$$t = \pm \frac{\Delta t}{\sqrt{2}}. \tag{136}$$

We then define the bandwidth generated by SPM ($\Delta\omega_{SPM}$) as the range between the turning points:

$$\Delta\omega_{SPM} = \omega(\Delta t/\sqrt{2}) - \omega(-\Delta t/\sqrt{2}) = \frac{2\sqrt{2}\omega_0 n_2 I_0 L}{c\Delta t\sqrt{\mathrm{e}}} \propto \frac{n_2 I L}{\Delta t}. \tag{137}$$

7.5 Four-wave mixing (FWM)

Now we consider a more general framework for any four-wave mixing process.

$$P^{NL}(z,t) = \varepsilon_0 \chi^{(3)} (E_1(z,t) + E_2(z,t) + E_3(z,t) + E_4(z,t))^3. \tag{138}$$

This results in the generation of waves with frequencies given by $\omega = \pm\omega_1 \pm \omega_2 \pm \omega_3$ in addition to third harmonic generation waves with $\omega = 3\omega_1, 3\omega_2, 3\omega_3$. Now we consider the following $\omega = \omega_1 + \omega_2 - \omega_3$ and $\boldsymbol{k} = \boldsymbol{k}_1 + \boldsymbol{k}_2 - \boldsymbol{k}_3$, then the terms at ω are

$$P^{NL}(\boldsymbol{r},t) = \frac{3}{2}\varepsilon_0 \chi^{(3)} \tilde{E}_1 \mathrm{e}^{-i(\omega_1 t - \boldsymbol{k}_1\cdot\boldsymbol{r})} \tilde{E}_2 \mathrm{e}^{-i(\omega_2 t - \boldsymbol{k}_2\cdot\boldsymbol{r})} \tilde{E}_3^* \mathrm{e}^{i(\omega_3 t - \boldsymbol{k}_3\cdot\boldsymbol{r})} \tag{139}$$

Restrict to common geometry $\boldsymbol{k}_1 = -\boldsymbol{k}_2$ and $\boldsymbol{k}_4 = -\boldsymbol{k}_3$. Assume E_1 and E_2 are very strong, much stronger than E_3 and E_4. Recall the coupled equations

$$\frac{\partial \tilde{E}_n}{\partial z} = \frac{-i\omega_n}{2\varepsilon_0 c n_n} \tilde{P}_n^{NL}(z), \tag{140}$$

we have

$$\frac{\partial \tilde{E}_1}{\partial z} = i\kappa(|\tilde{E}_1|^2 + 2|\tilde{E}_2|^2)\tilde{E}_1, \tag{141}$$

$$\frac{\partial \tilde{E}_2^*}{\partial z} = -i\kappa(|\tilde{E}_2|^2 + 2|\tilde{E}_1|^2)\tilde{E}_2, \tag{142}$$

$$\frac{\partial \tilde{E}_3}{\partial z} = i\kappa\left[(|\tilde{E}_1|^2 + |\tilde{E}_2|^2)\tilde{E}_3 + \tilde{E}_1\tilde{E}_2\tilde{E}_4^*\right], \tag{143}$$

$$\frac{\partial \tilde{E}_4^*}{\partial z} = -i\kappa\left[(|\tilde{E}_1|^2 + |\tilde{E}_2|^2)\tilde{E}_4 + \tilde{E}_1\tilde{E}_2\tilde{E}_3^*\right]. \tag{144}$$

If we assume E_1, E_2, E_3 are undepleted, i.e., $\partial\tilde{E}_{1,2,3}/\partial z = 0$, then

$$I_4(L) = \frac{L^2}{4}(\chi^{(3)})^2 I_1 I_2 I_3 \mathrm{sinc}^2\left(\frac{\Delta k L}{2}\right). \tag{145}$$

If we assume E_1, E_2 are undepleted, E_3 is now depleted, then

$$\frac{\partial \tilde{E}_3}{\partial z} = i\kappa \left[(|\tilde{E}_1|^2 + |\tilde{E}_2|^2)\tilde{E}_3 + \tilde{E}_1 \tilde{E}_2 \tilde{E}_4^* \right], \tag{146}$$

$$\frac{\partial \tilde{E}_4^*}{\partial z} = -i\kappa \left[(|\tilde{E}_1|^2 + |\tilde{E}_2|^2)\tilde{E}_4 + \tilde{E}_1 \tilde{E}_2 \tilde{E}_3^* \right]. \tag{147}$$

The equations can be simplified with a change of variable $\tilde{E}_3 = \tilde{E}_3' \exp(2i\kappa z)$ and $\tilde{E}_4 = \tilde{E}_4' \exp(-2i\kappa z)$, leading to

$$\frac{\partial \tilde{E}_3'}{\partial z} = i\kappa \tilde{E}_4'^*, \quad \frac{\partial \tilde{E}_4'}{\partial z} = i\kappa \tilde{E}_3'^*. \tag{148}$$

The trial solution is

$$\tilde{E}_4' = B \sin(\kappa z + C) + D \cos(\kappa z + E). \tag{149}$$

7.5.1 Optical phase conjugation

Assuming phase-matching and undepleted pump

$$E_4(L) = \frac{i\omega L}{2nc} \chi^{(3)} E_1(0) E_2(0) E_3^*(0). \tag{150}$$

E_4 is therefore the phase-conjugate of E_3:

$$E_3(z,t) = E_0 \mathrm{e}^{i(\omega t - kz)} \quad \Rightarrow \quad E_4(z,t) \propto \mathrm{e}^{-i(\omega t - kz)}. \tag{151}$$

Phase conjugate mirror inverts the sign of the phase:

$$\phi = \omega t - \boldsymbol{k} \cdot \boldsymbol{r} \quad \Rightarrow \quad -\phi = \omega(-t) - (-\boldsymbol{k}) \cdot \boldsymbol{r}, \tag{152}$$

while normal mirror reflects only direction of $\boldsymbol{k}$. One application of optical phase conjugation is real time holography:

(1) Hologram is written by E_1 and E_2 through the optical Kerr effect.

(2) Probe E_3 interacts with the hologram and diffracts into E_4.

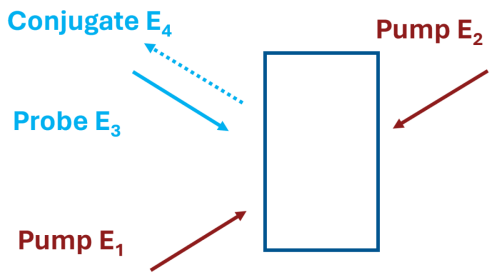

Figure 10: Phase conjugation geometry.