Wave equation for nonlinear optics:

$$\vec{\nabla}^2 \vec{E} - \frac{1}{c^2}\frac{\partial^2 \vec{E}}{\partial t^2} = \mu_0 \frac{\partial^2 \vec{P}_{NL}}{\partial t^2}$$

Exponential notation:

Convenient to use $E = E_0 e^{i\omega t}$
rather than $E = E_0 \cos\omega t$

have E = real field = $\mathrm{Re}\{E_0 e^{i\omega t}\} = \mathrm{Re}\{\tilde{E}\}$

However $P_{NL}(t) \neq \mathrm{Re}\{2d\tilde{E}^2\} = 2dE_0^2 \cos 2\omega t$
wrong amp, missing dc term

Exponential trick breaks down with nonlinear terms
need to write out.

if $E(t) = \mathrm{Re}\,\tilde{E}(t)$ ← complex form

$$= \frac{1}{2}(\tilde{E} + \tilde{E}^*)$$

so

$$E^2 = \frac{1}{4}(\tilde{E}^2 + \tilde{E}^{*2} + 2\tilde{E}\tilde{E}^*)$$

$$= \frac{1}{2}\left[\frac{1}{2}(\tilde{E}^2 + \tilde{E}^*\tilde{E}) + c.c.\right]$$

so take

$$P_{NL}(t) = \mathrm{Re}\{\tilde{P}_{NL}\} = \frac{1}{2}(\tilde{P}_{NL} + \tilde{P}_{NL}^*)$$

$$\tilde{P}_{NL} = 2d \cdot \frac{1}{2}(\tilde{E}^2 + \tilde{E}^*\tilde{E}) = d(\tilde{E}^2 + \underset{\sim}{\tilde{E}^*\tilde{E}})$$

For example: if $\tilde{E} = E_1 e^{i\omega_1 t} + E_2 e^{i\omega_2 t}$

$$\tilde{P}_{NL}(t) = d\,[E_1^2 e^{2i\omega_1 t} + E_2^2 e^{2i\omega_2 t} + 2E_1E_2 e^{i(\omega_1+\omega_2)t} + |E_1|^2 + |E_2|^2 + \underbrace{E_1^* E_2 e^{-i(\omega_1-\omega_2)t}}_{\text{can replace with c.c.}} + E_1E_2^* e^{i(\omega_1-\omega_2)t}]$$

$P_{NL}(2\omega_1) = dE_1^2$
$P_{NL}(2\omega_2) = dE_2^2$] second harmonic generation

$P_{NL}(\omega_1+\omega_2) = 2dE_1E_2$
$P_{NL}(\omega_1-\omega_2) = 2dE_1E_2^*$] 3 wave mixing

From $\tilde{E}\tilde{E}^*$ term
$P_{NL}(0) = d(|E_1|^2 + |E_2|^2)$

Source at 5 different frequencies

Generally write $P_{NL}(\omega_1+\omega_2) = g\,d\,E(\omega_1)E(\omega_2)$

g : symmetry factor $= 2$ if $\omega_1 \neq \omega_2$
$= 1$ if $\omega_1 = \omega_2$

$E(-\omega_1) = E(\omega_1)^*$

For third order, more complicated:

$$P_{NL}(\omega_1+\omega_2+\omega_3) = g\,\chi^{(3)} E(\omega_1)E(\omega_2)E(\omega_3)$$

g_1 = # of unique orderings of $(\omega_1, \omega_2, \omega_3)$

$g = 6$ $\omega_1 \neq \omega_2 \neq \omega_3$
$= 3$ $\omega_1 = \omega_2 \neq \omega_3$
$= 1$ $\omega_1 = \omega_2 = \omega_3$

Also, in reality, d depends on frequencies

$$d = d(\omega_1, \omega_2; \omega_3)$$

so $P(\omega_1 + \omega_2) \neq P(\omega_1 - \omega_2)$ in general
→ dispersion

Tensor Character

Really $P_i(\omega_3) = \sum_{jk} g\, d_{ijk} E_j(\omega_1) E_k(\omega_2)$

d_{ijk} = 2nd order nonlinear tensor.

must have $d_{ijk} = d_{ikj}$; so write as d_{iJ}
$J = 1$ to 6 as before.

But before had r_{Ij}, $I = 1$ to 6

$$\Delta \eta_{ij} = \sum_k r_{ijk} E_k$$

But d_{iJ} has same crystal symmetry dependence as r_{Ij}

KPP $r_{Ij} = \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \\ r_{41} & 0 & 0 \\ 0 & r_{41} & 0 \\ 0 & 0 & r_{63} \end{bmatrix}$ $\quad d_{iJ} = \begin{bmatrix} 0 & 0 & 0 & d_{14} & 0 & 0 \\ 0 & 0 & 0 & 0 & d_{14} & 0 \\ 0 & 0 & 0 & 0 & 0 & d_{36} \end{bmatrix}$

Also an additional symmetry approximately holds

$$d_{ijk} \simeq d_{jik} \simeq d_{jki}$$

called kleinmen symmetry

So in KDP $d_{14} = d_{xyz} \simeq d_{36} = d_{zxy}$
$= 4.1 \times 10^{-24} \frac{C}{V^2}$ $= 3.8 \times 10^{-24} \frac{C}{V^2}$

Note, this gives yet more factors of 2!

Say have $E_y(\omega_1)\ E_z(\omega_2)$

Then in KDP get $P_x(\omega_3) = g\, d_{xyz}\, E_y(\omega_1)\, E_z(\omega_2)$
$= 2 d_{14}\, E_y(\omega_1)\, E_z(\omega_2)$

don't add. $d_{xzy}\, E_z(\omega_2)\, E_y(\omega_1)$
already counted in g

But if $\omega_1 = \omega_2 = \omega$:
$P_x(2\omega) = d_{xyz}\, E_y(\omega)\, E_z(\omega) + d_{xzy}\, E_y(\omega)\, E_z(\omega)$
$= 2 d_{14}\, E_y(\omega)\, E_z(\omega)$

No factor if using say d_{13}

$$P_x(2\omega) = d_{xzz}\, E_z(\omega)\, E_z(\omega)$$

For 2^{nd} order, get factor of two if two different input fields, not if two identical fields
(in polarization & frequency.)

For simplicity, we will write

$$P(\omega_1+\omega_2) = 2d' E(\omega_1) E(\omega_2)$$

d' modified by these effects
& includes multiple d_{ijk}'s as needed.