Nonlinear Optics

According to the Lorentz model, the response of a bound electron to an external electric field satisfies Newton's 2^{nd} law:

$$\underset{\substack{\uparrow \\ \text{mass}}}{m}\,\ddot{\vec{r}} = -\underset{\substack{\uparrow \\ \text{charge}}}{e}\,\underset{\substack{\uparrow \\ \text{electric field}}}{\vec{E}} - \underset{\substack{\uparrow \\ \text{damping}}}{\gamma}\,\dot{\vec{r}} - \nabla \underset{\substack{\uparrow \\ \text{potential binding the electron.}}}{V(\vec{r})}$$

which must be solved for the electron's position $\vec{r}(t)$.

Let the origin of $\vec{r}$ be at the equilibrium point of $V(\vec{r})$ so that $\nabla V(\vec{0}) = \vec{0}$. ~~Then a Taylor expansion gives~~ If the medium is isotropic,

$$V(\vec{r}) = V(|\vec{r}|) = V(r)$$

~~So~~ and its taylor expansion has the form

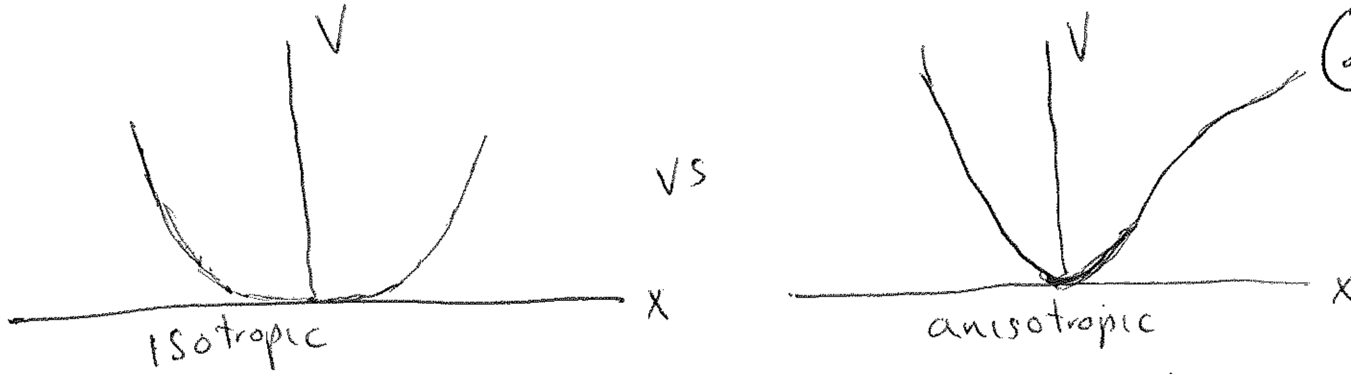
$$V(\vec{r}) = V_0 + \frac{C_2 r^2}{2} + \frac{C_4 r^4}{4} + \cdots$$

Let us fix y & z and look only at x. Then

$$V(x) = V_0 + \frac{C_2 X^2}{2} + \frac{C_4 X^4}{4} + \cdots \quad (1\ \text{iso})$$

If the medium is anisotropic, on the other hand, the section in x of the potential well could be asymmetric, so that

$$V(x) = V_0 + \frac{C_2 X^2}{2} + \frac{C_3 X^3}{3} + \frac{C_4 X^4}{4} + \cdots \quad (2\ \text{aniso})$$



Note that, for the isotropic case the potential in y would look the same but for the anisotropic it could be quite different.

The equation of motion for the electron is then

$$m\ddot{x} = eE_x - \gamma\dot{x} - C_2 x - C_3 x^2 - C_4 x^3 + \ldots$$

For convenience, divide by m and define

$$\Gamma = \frac{\gamma}{m}, \quad a = \frac{C_3}{m}, \quad b = \frac{C_4}{m}, \quad \omega_0^2 = \frac{C_2}{m}, \text{ so that}$$

$$\ddot{x} + \Gamma\dot{x} + \omega_0^2 x = -\frac{e}{m}E_x - a x^2 - b x^3 + \ldots$$

Let the applied field be monochromatic with frequency ω: $E_x = \frac{\mathcal{E}(\omega)}{2} e^{-i\omega t} + \frac{\mathcal{E}^*(\omega)}{2} e^{i\omega t}$.

Consider first the linear case $(a=0, b=0, \ldots)$

$$\ddot{x} + \Gamma\dot{x} + \omega_0^2 x = \frac{e\mathcal{E}e^{-i\omega t}}{2m} + \frac{e\mathcal{E}^* e^{i\omega t}}{2m}$$

Propose the solution

$$X(t) = \frac{\mathrm{X}_1 e^{-i\omega t}}{2} + \frac{\mathrm{X}_1^* e^{i\omega t}}{2}$$

plug into the equation:

$$\frac{1}{2}\left[\omega_0^2-\omega^2-i\Gamma\omega\right]\mathrm{X}_1 e^{-i\omega t} + \frac{1}{2}\left[\omega_0^2-\omega^2+i\Gamma\omega\right]\mathrm{X}_1^* e^{i\omega t}$$

$$= -\left[\frac{e\mathcal{E}e^{-i\omega t}}{2} + \frac{e\mathcal{E}^* e^{i\omega t}}{2}\right] \quad \forall t.$$

So that $\mathrm{X}_1 = \dfrac{-e\mathcal{E}}{m(\omega_0^2-\omega^2-i\Gamma\omega)}$ linear in $\mathcal{E}$

The medium's polarization is

$$P_x = -\underset{\substack{\uparrow \\ \text{density} \\ \text{of electrons}}}{N}\ \underbrace{\frac{e(\mathrm{X}_1 e^{-i\omega t} + \mathrm{X}_1^* e^{i\omega t})}{2}}_{\text{dipole moment}}$$

We define the susceptibility $\chi(\omega)$ such that

$$P_x = \varepsilon_0 \frac{\chi(\omega)\mathcal{E}e^{-i\omega t} + \chi^*(\omega)\mathcal{E}^* e^{i\omega t}}{2}.$$

It is easy to see that $\chi(\omega) = \dfrac{Ne^2}{m\varepsilon_0(\omega_0^2-\omega^2-i\Gamma\omega)}$

The refractive index is $n(\omega) = \sqrt{1+\chi(\omega)}$

Now assume we are in an anisotropic medium (e.g. a quartz crystal) where a contributes appreciably to the potential, but higher order contributions can be ignored:

$$\ddot{X} + \Gamma \dot{X} + \omega_0^2 X \approx -\frac{e}{2m}\left(\mathcal{E} e^{-i\omega t} + \mathcal{E}^* e^{i\omega t}\right) - aX^2$$

For simplicity ignore the damping term $(\Gamma = 0)$.

We can solve this through perturbation if the contribution of aX^2 is small:

$$\ddot{X}^{(n)} + \omega_0^2 X^{(n)} \approx -\frac{e}{2m}\left(\mathcal{E} e^{-i\omega t} + \mathcal{E}^* e^{i\omega t}\right) - aX^{(n-1)2}$$

where $X^{(n)}$ gives the n^{th} order approximation, and $X^{(1)}$ is the linear solution

$$X^{(1)} = \frac{X_1 e^{-i\omega t} + X_1^* e^{i\omega t}}{2}, \quad X_1 = \frac{-e\mathcal{E}}{m(\omega_0^2 - \omega^2)}$$

For our purposes we will only use the next correction, $X \approx X^{(2)}$:

$$\ddot{X}^{(2)} + \omega_0^2 X^{(2)} = -\frac{e}{2m}\left(\mathcal{E} e^{-i\omega t} + \mathcal{E}^* e^{i\omega t}\right) - \frac{a}{2}\left(|X_1|^2 + \frac{X_1^2}{2} e^{-i2\omega t} + X_1^{*2} e^{i2\omega t}\right)$$

Propose the solution

$$X^{(2)} = \mathbb{X}_0 + \underbrace{\frac{\mathbb{X}_1 e^{-i\omega t} + \mathbb{X}_1^* e^{i\omega t}}{2}}_{X^{(1)}} + \frac{\mathbb{X}_2 e^{-i2\omega t} + \mathbb{X}_2^* e^{i2\omega t}}{2}$$

Substituting we get

$$\omega_0^2 \mathbb{X}_0 + \frac{(\omega_0^2 - 4\omega^2)}{2} \mathbb{X}_2 e^{-i2\omega t} + \frac{(\omega_0^2 - 4\omega^2)}{2} \mathbb{X}_2^* e^{i2\omega t}$$

$$= -\frac{a}{2} |\mathbb{X}_1|^2 - \frac{a}{4} \mathbb{X}_1^2 e^{-i2\omega t} - \frac{a}{4} \mathbb{X}_1^{*2} e^{i2\omega t}, \quad \forall t$$

Therefore

$$\mathbb{X}_0 = -\frac{a}{2\omega_0^2} |\mathbb{X}_1|^2 = \frac{-a e^2 |\mathcal{E}|^2}{2m^2 \omega_0^2 (\omega_0^2 - \omega^2)^2}$$

$$\mathbb{X}_2 = +\frac{a}{2(4\omega^2 - \omega_0^2)} \mathbb{X}_1^2 = \frac{a e^2 \mathcal{E}^2}{2m^2 (4\omega^2 - \omega_0^2)(\omega_0^2 - \omega^2)^2}$$

both are quadratic on the field.

In this approximation, the medium's polarization is

$$P_x = N e X \approx N e X^{(2)} = \underbrace{N e \left(\frac{\mathbb{X}_1 e^{-i\omega t} + \mathbb{X}_1^* e^{i\omega t}}{2} \right)}_{P_x^{(L)}}$$

$$+ \underbrace{N e \left[\mathbb{X}_0 + \frac{\mathbb{X}_2 e^{-i2\omega t} + \mathbb{X}_2^* e^{i2\omega t}}{2} \right]}_{P_x^{(NL)}}$$

Exercise: Now suppose that the medium is isotropic ($a=0$) and calculate the nonlinear polarization of the medium by solving through perturbations the equation:

$$\ddot{X} + \omega_0^2 X = \frac{-e}{2m}\left(\mathcal{E} e^{-i\omega t} + \mathcal{E}^* e^{i\omega t}\right) - bX^3$$

We now study the propagation of the field, so we introduce dependence in z. We therefore replace:

$$\mathcal{E}(\omega)\, e^{-i\omega t} \longrightarrow \mathcal{E}(\omega,z)\, \underbrace{e^{iK(\omega)z-i\omega t}}_{\text{plane wave solution of linear problem}}$$

where

$$K(\omega) = \frac{\omega}{c}\sqrt{1+\chi(\omega)}\,.$$

The wave equation is

$$\nabla^2 E_x - \underbrace{\varepsilon_0 \mu_0}_{1/c^2} \frac{\partial^2 E_x}{\partial t^2} = \mu_0 \frac{\partial^2 P_x}{\partial t^2}$$

Let us assume that, for each frequency, $\mathcal{E}(\omega,z)$ varies much slower in z than $e^{iK(\omega)z}$, so

$$\left(\nabla^2 - \varepsilon_0\mu_0 \frac{\partial^2}{\partial t^2}\right) \mathcal{E}(\omega,z)\, e^{iK(\omega)z-i\omega t} = \Big[\underbrace{\frac{\partial^2 \mathcal{E}}{\partial z^2}}_{\text{neglect}} + 2iK\frac{\partial \mathcal{E}}{\partial z} - K^2\mathcal{E} + \frac{\omega^2}{c^2}\mathcal{E}\Big] e^{iKz-i\omega t}$$

$$\approx \left[2iK\frac{\partial \mathcal{E}}{\partial z} - \frac{\omega^2}{c^2}\chi\mathcal{E}\right] e^{iKz-i\omega t}$$

2^{nd} Harmonic generation

Suppose a field of freq. ω enters a nonlinear medium, generating a DC and a second Harmonic field. This can be written as

$$\left(\nabla^2 - \varepsilon_0\mu_0\frac{\partial^2}{\partial t^2}\right)\left[\frac{\mathcal{E}(\omega,z)e^{iK(\omega)z-i\omega t}}{2} + c.c. + \cancel{\mathcal{E}(0,z)} + \frac{\mathcal{E}(2\omega,z)e^{iK(2\omega)z-2i\omega t}}{2} + c.c.\right]$$

$$= \mu_0\left[\frac{\partial^2 P^{(L)}}{\partial t^2}(\omega) + \frac{\partial^2 P^{(NL)}}{\partial t^2}(\omega,\omega) + \cancel{\frac{\partial^2 P^{(NL)}}{\partial t^2}(\omega,-\omega)} + \frac{\partial^2 P^{(L)}}{\partial t^2}(2\omega) + \underbrace{\frac{\partial^2 P^{(NL)}(2\omega,2\omega)}{\partial t^2}}_{\text{Oscillates at }4\omega\text{ neglect.}}\right]$$

From here we get

Terms at frequency ω:

$$2iK(\omega)\frac{\partial\mathcal{E}(\omega,z)}{\partial z} - \cancel{\frac{\omega^2}{c^2}\chi(\omega)\mathcal{E}(\omega,z)} = \cancel{-\frac{\omega^2}{c^2}\chi(\omega)\mathcal{E}(\omega,z)}$$

$$2iK(2\omega)\frac{\partial\mathcal{E}(2\omega,z)}{\partial z} - \cancel{\frac{4\omega^2}{c^2}\chi(2\omega)\mathcal{E}(2\omega,z)} = \overbrace{\cancel{-\frac{4\omega^2}{c^2}\chi(2\omega)\mathcal{E}(2\omega,z)}}^{\frac{\partial^2 P^{(L)}}{\partial t^2}(2\omega)} + \frac{\bar{a}}{c^2}\chi^2(\omega)\chi(2\omega)\mathcal{E}^2(\omega)\,e^{i[\underbrace{2K(\omega)-K(2\omega)}_{\Delta K(\omega)}]z}$$

so $\mathcal{E}(\omega,z) \approx$ constant

$$\mathcal{E}(2\omega,z) \approx \frac{-\bar{a}}{2c^2K(2\omega)}\chi^2(\omega)\chi(2\omega)\mathcal{E}^2(\omega)\frac{e^{i\Delta K(\omega)z}-1}{\Delta K(\omega)}$$

$$|\mathcal{E}(2\omega,z)|^2 = \frac{\bar{a}^2\,|\chi^2(\omega)\chi(2\omega)|^2}{4c^4|K(2\omega)|^2}|\mathcal{E}(\omega)|^2 z^2\left|\frac{\sin\left(\frac{\Delta K(\omega)z}{2}\right)}{\frac{\Delta K(\omega)z}{2}}\right|^2$$

www.ictp.it

Three-wave mixing

Let us apply now a field with two frequencies:

$$E_x = \sum_{j=1}^{2} \frac{\mathcal{E}(\omega_j)\, e^{-i\omega_j t} + \mathcal{E}^*(\omega_j) e^{i\omega_j t}}{2}$$

We find that the polarization contains terms with frequencies ω_1 & ω_2 (linear terms) as well as with frequencies 0, $2\omega_1$, $2\omega_2$ (second harmonic nonlinear terms, and $\omega_1+\omega_2$, $\omega_1-\omega_2$ (sum/difference nonlinear terms.

$$P_x = \underbrace{\sum_{i=1}^{2} P^{(L)}(\omega_i)}_{\text{Linear terms}} + \underbrace{\sum_{i,j=1}^{2} P^{(NL)}(\omega_i,\omega_j) + \sum_{i,j}^{2} P^{(NL)}(\omega_i,-\omega_j)}_{\text{Nonlinear terms}}$$

where

$$P^{(L)}(\omega_i) = \varepsilon_0 \,\mathrm{Re}\left\{\chi(\omega_i)\, \mathcal{E}(\omega_i)\, e^{-i\omega_i t}\right\}$$

$$P^{(NL)}(\omega_i,\omega_j) = \varepsilon_0\, \bar{a}\, \mathrm{Re}\left\{\chi(\omega_i)\chi(\omega_j)\chi(\omega_i+\omega_j)\mathcal{E}(\omega_i)e^{-i\omega_i t} \cdot \mathcal{E}(\omega_j)e^{-i\omega_j t}\right\}$$

with the constant $\bar{a} = \dfrac{m\varepsilon_0^2}{2N^2e^3}\, a$

This is valid in fact for more frequencies too.

Can define the nonlinear susceptibility more generally as

$$P^{(NL)}(\omega_i,\omega_j) = \frac{\varepsilon_0}{2}\chi(-\omega_i-\omega_j,\omega_i,\omega_j)\mathcal{E}(\omega_i)\mathcal{E}(\omega_j)e^{-i\omega_i t}e^{-i\omega_j t}$$

For a single resonance, then:

$$\chi(-\omega_i-\omega_j,\omega_i,\omega_j) = 2\bar{a}\,\chi(\omega_i)\chi(\omega_j)\chi(\omega_i+\omega_j)$$

The expression for the second harmonic generated intensity is then

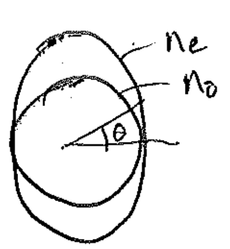
$$|\mathcal{E}(2\omega,z)|^2 = \frac{|\chi(-2\omega,\omega,\omega)|^2|\mathcal{E}(\omega)|^2}{16c^4|K(2\omega)|^2}z^2\left|\frac{\sin\left(\frac{\Delta K(\omega)z}{2}\right)}{\frac{\Delta K(\omega)z}{2}}\right|^2$$

So to enhance SHG, can:

- use ω or 2ω near resonances
- make $\Delta K = 2K(\omega) - K(2\omega)$ small or zero.

This second is called "phase matching".

Recall that for a crystal $K(\omega) = \frac{\omega}{c}n(\omega)$ depends on polarization.



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

If $n_e > n_o$ (Positive uniaxial)

if pump is in e polarization, can find that $\Delta K = 0$ for

$$\sin^2\theta = \frac{1 - n_o^2(\omega)/n_o^2(2\omega)}{1 - n_o^2(\omega)/n_e^2(\omega)}$$

For three-wave mixing, if we have three frequencies ω_1, ω_2, $\omega_3 = \omega_1 + \omega_2$, the wave equation gives approximately

$$2iK(\omega_1)\frac{\partial \mathcal{E}}{\partial z}(\omega_1, z) - \cancel{\frac{\omega_1^2}{c^2}\chi(\omega_1)\mathcal{E}(\omega_1, z)} = -\overbrace{\cancel{\frac{\omega_1^2}{c^2}\chi(\omega_1)\mathcal{E}(\omega_1, z)}}^{P^{(1)}(\omega_1)}$$

$$-\frac{\omega_1^2}{c^2}\chi(-\omega_1,-\omega_2,\omega_3)\mathcal{E}^*(\omega_2)\mathcal{E}(\omega_3)\, e^{i\Delta K z}$$

$$2iK(\omega_2)\frac{\partial \mathcal{E}}{\partial z}(\omega_2, z) - \cancel{\emptyset} = -\emptyset - \frac{\omega_2^2}{c^2}\chi(-\omega_2,-\omega_1,\omega_3)\mathcal{E}^*(\omega_1)\mathcal{E}(\omega_3)\, e^{i\Delta K z}$$

$$2iK(\omega_3)\frac{\partial \mathcal{E}}{\partial z}(\omega_3, z) = -\frac{\omega_3^2}{c^2}\chi(-\omega_3,\omega_1,\omega_2)\mathcal{E}(\omega_1)\mathcal{E}(\omega_2)\, e^{i\Delta K z}$$

where

$$\Delta K = K(\omega_1) + K(\omega_2) - K(\omega_3)$$

Exercise: from these equations, find the "Manley-Rowe" equations:

$$\frac{1}{\omega_1}\frac{d}{dz}\left[\sqrt{\frac{\varepsilon(\omega_1)}{\mu_0}}\,|\mathcal{E}(\omega_1, z)|^2\right] = \frac{1}{\omega_2}\frac{d}{dz}\left[\sqrt{\frac{\varepsilon(\omega_2)}{\mu_0}}\,|\mathcal{E}(\omega_2, z)|^2\right]$$

$$= -\frac{1}{\omega_3}\frac{d}{dz}\left[\sqrt{\frac{\varepsilon(\omega_3)}{\mu_0}}\,|\mathcal{E}(\omega_3, z)|^2\right].$$

that is $\dfrac{d}{dz}\dfrac{I(\omega_1)}{\omega_1} = \dfrac{d}{dz}\dfrac{I(\omega_2)}{\omega_2} = -\dfrac{d}{dz}\dfrac{I(\omega_3)}{\omega_3}$