Non Linear Dielectric Response

Abbreviated sketch

$$P = \chi^1 E + \chi^2 EE + \chi^3 EEE$$

$$E = \cos \omega t$$

$$\Rightarrow EE = \cos^2 \omega t = \tfrac{1}{2}(\underbrace{\cos 2\omega t}_{\text{frequency doubled}} + \underbrace{1}_{\text{static}})$$

Thus P has terms like $\cos \omega t$ — large

and $\frac{1}{2}(\cos 2\omega t + 1)$ — small

That gives the same terms in E (oscillating dipole broadcasts an oscillating E-field)

These recombine

$$e^{\pm i\omega t} \times e^{\pm i\omega t} \rightarrow 1,\ e^{\pm 2i\omega t}$$

$$e^{\pm 2i\omega t} \times e^{\pm i\omega t} \rightarrow e^{\pm i\omega t},\ e^{\pm 3i\omega t}$$

$$e^{\pm 3i\omega t} \times e^{\pm i\omega t} \rightarrow e^{\pm 2i\omega t},\ e^{\pm 4i\omega t}$$

Generally these terms stay small.

If and only if a certain phase-matching condition is met, then the material will amplify the higher harmonics

~~..~~ 1. Essentials of Complete Theory

Dipole in material comes from a nonlocal, non instantaneous, anisotropic response:

$$P_\alpha(\mathbf{r},t) = \sum_\beta \int \chi^{1}_{\alpha\beta}(\mathbf{r}-\mathbf{r}', t-t')\, E_\beta(\mathbf{r}',t')\, d\mathbf{r}'\, dt'$$

$$+ \sum_{\beta\gamma} \int \chi^{(2)}_{\alpha,\beta\gamma}(\mathbf{r}-\mathbf{r}_1, t-t_1; \mathbf{r}-\mathbf{r}_2, t-t_2)$$

$$\times\, E_\beta(\mathbf{r}_1, t_1)\, E_\gamma(\mathbf{r}_2, t_2)\, d\mathbf{r}_1 dt_1 d\mathbf{r}_2 dt_2$$

$$+ \sum_{\beta\gamma\delta} \int \chi^{(3)}_{\alpha,\beta\gamma\delta}(\mathbf{r}-\mathbf{r}_1, t-t_1; \mathbf{r}-\mathbf{r}_2, t-t_2; \mathbf{r}-\mathbf{r}_3, t-t_3)$$

$$E_\beta(\mathbf{r}_1\ t_1)\, E_\gamma(\mathbf{r}_2\ t_2)\, E_\delta(\mathbf{r}_3, t_3)\, d\mathbf{r}_1 dt_1\, d\mathbf{r}_2 dt_2\, d\mathbf{r}_3 dt_3$$

We were forced to take time delay in χ^1 into account, but we ignored nonlocality in χ^1.

We are forced to consider tensor aspect of χ^1 to get phase matching. We are also forced to consider tensor aspect of χ^2, because if the material is isotropic, $\chi^2 = 0$.

Frequency Space

Define $\chi^{(2)}_{\alpha,\beta\gamma}(\mathbf{k}_1\,\omega_1\;\mathbf{k}_2\,\omega_2) = \int e^{-i(\mathbf{k}_1\rho_1-\omega_1\tau_1)}\,e^{-i(\mathbf{k}_2\rho_2-\omega_2\tau_2)}$

$\rho_1 = \mathbf{r}-\mathbf{r}_1 \qquad \tau_1 = t-t_1 \qquad \chi_{\alpha,\beta\gamma}(\rho_1\,\tau_1\;\rho_2\,\tau_2)\;d\rho_1 d\rho_2$
$\rho_2 = \mathbf{r}-\mathbf{r}_2 \qquad \tau_2 = t-t_2 \qquad d\tau_1 d\tau_2$

Then [thm]

$$P_\alpha(\mathbf{r},t) = \int e^{i(\mathbf{k}_1+\mathbf{k}_2)\mathbf{r}-(\omega_1+\omega_2)t}$$

$$\chi^{(2)}_{\alpha\;\beta\gamma}(\mathbf{k}_1\omega_1,\;\mathbf{k}_2\omega_2)$$

$$E_\beta(\mathbf{k}_1\omega_1)\;E_\gamma(\mathbf{k}_2\omega_2)$$

$$\frac{d^3k_1\,d^3k_2\,d\omega_1\,d\omega_2}{(2\pi)^8}$$

Thus $E_\beta = e^{\pm i(\mathbf{k}_\beta\mathbf{r}-\omega_\beta t)}$
$E_\gamma = e^{\pm i(\mathbf{k}_\gamma\mathbf{r}-\omega_\gamma t)}$

gives P_α with $e^{i(\mathbf{k}_\beta+\mathbf{k}_\gamma)\cdot\mathbf{r}-(\omega_\beta+\omega_\gamma)t}$
$e^{i(\mathbf{k}_\beta-\mathbf{k}_\gamma)\cdot\mathbf{r}-(\omega_\beta-\omega_\gamma)t}$

sum and difference terms.

Note $\chi^{(2)}_{\alpha\;\beta\gamma} = \chi^{(2)}_{\alpha\;\gamma\beta}$

Size and
Frequency dependence of $\chi^{(2)}$:

$\chi^{(2)}$ is generally small:

$$P^{(1)} = \chi^{(1)} E \qquad \chi^{(1)} = \mathcal{O}(1)$$

$$P^{(2)} = \chi^{(2)} E E$$

units of $\chi^{(2)}$ = units of $\chi^{(1)}/E$
typical size of $\chi^{(2)}$ = size of $\chi^{(1)}$/ atomic unit of electric field

$$\sim 10 V/Å$$

$$\sim 10^{9} V/cm$$

Thus in ESU $\quad \chi^{(1)} \sim 1$

$\chi^{(2)} \sim 3\times10^{-8}$ to 3×10^{-7} esu

$\chi^{(3)} \sim 10^{-14}$ to 10^{-13} esu

$\vdots \qquad \vdots$

However $\chi^{(2)}$ can increase greatly near resonances, if either ω or 2ω is near a resonance of the system.

NLDR 5

Amplifying the frequency-doubled wave — Simplify by going to scalar theory

Look at some elementary dif. eqs

a) Driven harmonic oscillator

$$\frac{d^2\psi}{dt^2} + \omega_0^2 \psi = f e^{i\omega_d t}$$

[out of ~~reson~~ resonance]

$$\psi = c_+ e^{i\omega_0 t} + c_- e^{-i\omega_0 t} + a e^{i\omega_d t}$$

$$a = \frac{f}{\omega_0^2 - \omega_d^2}$$

in resonance

$$\frac{d^2\psi}{dt^2} + \omega_0^2 \psi = f e^{i\omega_0 t}$$

$$\psi = a\, t\, e^{i\omega_0 t}$$

$$a = \frac{f}{2i\omega_0}$$

real combinations: if driver is $\cos\omega_0 t$

response is $\frac{f}{2\omega_0} t \sin\omega_0 t$

- oscillator amplifies signal from driver
- velocity is in phase with force $\Rightarrow$ oscillator absorbs power without limit.

b. Classical Wave Eq.

$$\left(\frac{\partial^2}{\partial x^2} - \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right)\psi(x,t) = 0$$

$$\psi = e^{i(kx-\omega t)}, \quad \omega = ck$$

c) CWE including time delays

$$\left[\frac{\partial^2}{\partial x^2} - \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right]\psi(x,t) - \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\int \chi(t-t')\,\psi(x,t')\,dt' = 0$$

If we Fourier transform in both x and t we get

$$\left(-k^2 + \frac{\omega^2}{c^2}\right)\tilde{\tilde{\psi}}(k,\omega) + \frac{\tilde{\chi}(\omega)}{c^2}\tilde{\tilde{\psi}}(k,\omega) = 0$$

i.e.

$$-k^2 + \frac{n(\omega)^2\omega^2}{c^2} = 0$$

$$n(\omega)^2 = 1 + \tilde{\chi}(\omega) \equiv \tilde{\varepsilon}(\omega)$$

If we transform t but not x, we obtain

$$\frac{d^2}{dx^2}\tilde{\psi}(x,\omega) + \frac{n(\omega)^2\omega^2}{c^2}\tilde{\psi}(x,\omega) = 0$$

Solutions are $e^{ik_0(\omega)x}$ $\quad k_0(\omega) = \frac{n(\omega)\,\omega}{c}$.

d. ~~The~~ A driven wave equation including time-delays

$$\left(\frac{\partial^2}{\partial x^2} - \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right)\Psi(x,t) - \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\int \chi(t-t')\,\Psi(x,t')\,dt' = S(x,t)$$

$S(x,t)$ is a source fn, that creates waves everywhere in space x at ~~time~~ all times t.

Transform time to frequency

$$\left[\frac{\partial^2}{\partial x^2} + \frac{n(\omega)^2\omega^2}{c^2}\right]\tilde{\Psi}(x,\omega) = \tilde{S}(x,\omega)$$

Suppose the source looks like $f e^{ik_s x}$

We have

$$\left[\frac{\partial^2}{\partial x^2} + \underbrace{\frac{n^2(\omega)\,\omega^2}{c^2}}_{k_0(\omega)^2}\right]\tilde{\Psi}(x,\omega) = f e^{ik_s x}$$

But this is just the driven harmonic oscillator

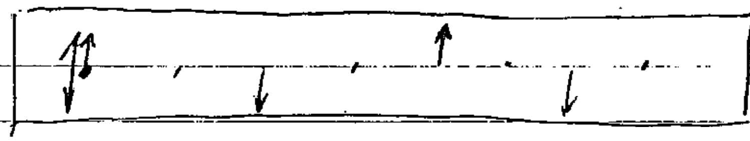
$$\omega_d \rightarrow k_s$$

$$\omega_0 \rightarrow k_0$$

Solutions are $C_+ e^{ik_0 x} + C_- e^{-ik_0 x} + a\, e^{ik_s x}$

$$a = \frac{f}{k_0^2 - k_s^2}$$

The amplitude of the resulting wave is ~~equal to~~ proportional to the strength of the driver f divided by $k_0^2 - k_s^2$ where $k_0 = \sqrt{\frac{n^2(\omega)\omega^2}{c^2}}$ is the natural wave number of the substance at that frequency, and k_s is the wave number of the driver.



Distributed source. At every point it is oscillating at frequency ω. However the various points are not in phase with each other.

The substance carries waves of any frequency; their wavelength is $k_0^2 = n^2(\omega)\omega^2/c^2$.

If the wavelength of the driver does not match the natural wavelength of waves at that frequency for that substance, then the substance carries those waves without amplifying them.

b. Now suppose the wave number of the driver matches the ~~wave~~ natural wave number of the substance at that frequency: $k_s = k_0(\omega)$.

This is called a "Phase Matching Condition": the phase of ~~each pa~~ the driver at each location matches the phase of a naturally-carried wave at that location.

Then the solution will GROW

$$\tilde{\hat{\Psi}}(x, \omega) = \frac{f}{2 i k_0(\omega)} x e^{i k_0(\omega) x}$$

We will show that a nonlinear medium will give frequency-doubling of ANY incident signal, but the harmonic wave will in general not grow.

If a phase-matching condition is met, then the harmonic wave will be amplified at the expense of the incident wave.

red → $\boxed{}$ → blue

But now we have more success than we really want.

One constant amplitude wave in $E_0 e^{i(kx-\omega t)}$

$$P = \chi^{(2)} E E \rightarrow \chi^{(2)} E_0^2 e^{2i(kx-\omega t)}$$

twice the freq
half the wavelength

Produces a growing wave

$$E = \text{const} \cdot x \, e^{2i(kx-\omega t)}$$

$\Rightarrow$ Creation of energy.

What's next:

Amplitude of red must decrease as amplitude of blue grows.