Contracted notation

If Kleinman's symmetry is valid, each of the three matrices are symmetric:

$$\begin{bmatrix} \cdot & \cdot & \cdot \\ & \cdot & \cdot \\ & & \cdot \end{bmatrix} \quad \begin{bmatrix} \cdot & \cdot & \cdot \\ & \cdot & \cdot \\ & & \cdot \end{bmatrix} \quad \begin{bmatrix} \cdot & \cdot & \cdot \\ & \cdot & \cdot \\ & & \cdot \end{bmatrix} = 18 \text{ elements.}$$

contract second pair of indices to 1:

jk	11	22	33	23, 32	31, 13	12, 21
l	1	2	3	4	5	6

$$\rightarrow d_{il} = \begin{bmatrix} \cdot & \cdot & \cdot & \cdot & \cdot & \cdot \\ \cdot & \cdot & \cdot & \cdot & \cdot & \cdot \\ \cdot & \cdot & \cdot & \cdot & \cdot & \cdot \end{bmatrix}$$ now just one matrix.

not all are independent: can permute indices $\rightarrow$ max 10 independent
- see slide

Crystal symmetry further reduces the # of independent and non-zero matrix elements.

32 possible crystal point groups

21 are non-centrosymmetric, $\rightarrow$ no second-order response.

w/ inversion symmetry, $\chi^{(2)} \rightarrow 0$

SHG example

$$P(t) = \chi^{(2)} E(t)^2 \qquad E(t) = \mathcal{E}_0 \cos \omega t$$

if $E(t) \rightarrow -E(t)$

$P(t) \rightarrow -P(t)$ <u>with</u> inversion symmetry.

but $P(t) = \chi^{(2)}(-E(t))^2 \overset{?}{=} -P(t)$

$\therefore \chi^{(2)} = 0$

similar arguments for crystal symmetries

Effective d:

actual orientation of fields is restricted by phase matching:

for $\omega_3 = \omega_1 + \omega_2$

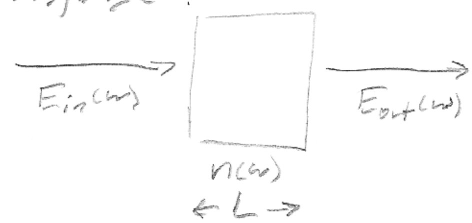
$$k_3 = k_1 + k_2 \quad \text{or} \quad \Delta k = k_1 + k_2 - k_3 = 0$$

within this restriction → cone of angles.

- value of d_{eff} determines best choice.
- we'll return to this when we treat phase matching.

Time vs. frequency domain

Linear response :



$$E_{out}(\omega) = E_{in}(\omega)\, e^{ik_0 n(\omega) L}$$

linear propagation is easiest to describe in freq. space.

- linear systems : $H(\omega)$ = transfer fcn, freq. response

$$F_{out}(\omega) = H(\omega)\, F_{in}(\omega)$$

$$f_{out}(t) = \mathcal{F}^{-1}\{H(\omega) F_{in}(\omega)\}$$

$$= h(t) \otimes f_{in}(t) \qquad \text{convolution theorem}$$

where $$= \int_{-\infty}^{\infty} h(\tau) f_{in}(t-\tau)\, d\tau \qquad \text{is convolution.}$$

if input is $f_{in}(t) = \delta(t)$ (impulse)

$$f_{out}(t) = \int_{-\infty}^{\infty} h(\tau)\, \delta(t-\tau)\, d\tau = h(t)$$

$\therefore$ $h(t) = \mathcal{F}^{-1}\{H(\omega)\}$ is impulse response.

At a microscopic level, $p(t)$ is the induced dipole when $E(t)$ is in.
and $P(t) = N_a p(t)$ is the collective response.

When we solve for $\chi^{(1)} = N_a P/E$, our method gives $\chi^{(1)}(\omega)$

recall steps: $E(t) = E_0 e^{-i\omega t} + c.c.$

$$x^{(1)}(t) = x_0^{(1)} e^{-i\omega t} + c.c.$$

$$x_0^{(1)} = -(e/m)\, E_0 / D(\omega)$$

$$\chi^{(1)}(\omega) = N_a e^2/m \,/\, D(\omega)$$

So what is the impulse response? For a range of input freq:

$$P^{(1)}(\omega) = \chi^{(1)}(\omega)\, E(\omega)$$

Alternative method: take FT of 2nd order eqn.

note that $\mathcal{F}\{\partial f/\partial t\} = -i\omega F(\omega)$

$\rightarrow$ eqn for $\chi^{(1)}(\omega)$ with $E(\omega)$ driving

Now in time domain,

$$P^{(1)}(t) = \mathcal{F}^{-1}\{\chi^{(1)}(\omega)\, E(t)\}$$

Recognize $\chi^{(1)}(\omega)$ as a transfer function.

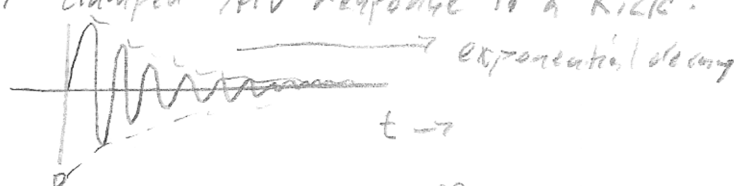
let $R^{(1)}(t) \equiv 2\pi\mathcal{F}^{-1}\{\chi^{(1)}(\omega)\} \equiv$ impulse response

Then $P^{(1)}(t) = \int_{-\infty}^{\infty} R^{(1)}(\tau)\, E(t-\tau)\, d\tau = R^{(1)} \otimes E$

(normally have $\frac{1}{2\pi}$ in front, this is absorbed into defn of R)

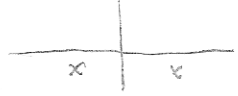
What is the nature of $R^{(1)}(t)$?

- causality requires $R^{(1)}(t) = 0$ for $t<0$
- expect? damped SHO response to a kick:



- Proof: 1) $\mathcal{F}^{-1}\{\chi^{(1)}(\omega)\} = \frac{1}{2\pi}\int_{-\infty}^{\infty} \frac{N(e^2/m)}{\omega^2-\omega_0^2-2i\omega}\, e^{-i\omega t}\, d\omega$

requires contour integration: poles are off real axis:



for $t<0$ close on upper half $\rightarrow 0$

for $t>0$ close lower.

Time dependent NL response.

in NL case, we have multiple input fields.

- can arrive at different times.

e.g.

$$P^{(2)}(t) = \int_0^\infty d\tau_1 \int_0^\infty d\tau_2\, R^{(2)}(\tau_1, \tau_2)\, E(t-\tau_1)\, E(t-\tau_2)$$

$R^{(2)} = 0$ for either $\tau_1 < 0$ or $\tau_2 < 0$ (causality)

put $E(\omega)$ in

$$\rightarrow \int_{-\infty}^{\infty} \frac{d\omega_1}{2\pi} \int_{-\infty}^{\infty} \frac{d\omega_2}{2\pi} \int_0^\infty d\tau_1 \int_0^\infty d\tau_2\, R^2(\tau_1, \tau_2)\, E(\omega_1)\, e^{-i\omega_1(t-\tau_1)} \times E(\omega_2)\, e^{-i\omega_2(t-\tau_2)}$$

$$= \int_{-\infty}^{\infty} \frac{d\omega_1}{2\pi} \int_{-\infty}^{\infty} \frac{d\omega_2}{2\pi}\, \chi^{(2)}(\omega_3; \omega_1, \omega_2)\, E(\omega_1)\, E(\omega_2)\, e^{-i\omega_3 t}$$

where $\omega_3 = \omega_1 + \omega_2$

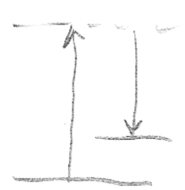
$$\chi^{(2)}(\omega_3; \omega_1, \omega_2) = \int_0^\infty d\tau_1 \int_0^\infty d\tau_2\, R^{(2)}(\tau_1, \tau_2)\, e^{i(\omega_1\tau_1 + \omega_2\tau_2)}$$

similar for $\chi^{(3)}$

Note that if response is assumed to be instantaneous, there is no dispersion in χ

In some important systems, response is delayed.

- e.g. Raman:



→ molecule left in excited state.

change in refr. index must wait for rotation or vibration to start.