III. Linear Dielectric Response of Matter

Before getting to frequency doubling (which requires nonlinear anisotropic response of samples) we need to understand linear isotropic response.

Possible types of behavior

instantaneous linear local response

$$\mathbb{P}(\mathbb{r}, t) = \chi \quad \mathbb{E}(\mathbb{r}, t)$$

$$\mathbb{D} = \mathbb{E} + \mathbb{P} = (1 + \underset{\uparrow \text{ dielectric susceptibility}}{\chi}) \mathbb{E} = \underset{\uparrow \text{ dielectric constant}}{\varepsilon} \mathbb{E}$$

Heaviside Lorentz

[Gaussian $\mathbb{D} = \mathbb{E} + 4\pi \mathbb{P}$ SI $\mathbb{D} = \epsilon_0 \mathbb{E} + \mathbb{P}$]

anisotropic response

$$\mathbb{P}_\alpha(\mathbb{r}, t) = \sum_\beta \chi_{\alpha\beta} \quad \mathbb{E}_\beta(\mathbb{r}, t)$$

time-delayed response

$$\mathbb{P}_\alpha(\mathbb{r}, t) = \int_{-\infty}^{t} \chi_{\alpha\beta}(t-t') \quad \mathbb{E}_\beta(t') \, dt'$$

Nonlinear response

$$\mathbb{P}_\alpha = \chi^{[1]}_{\alpha\beta} \, E_\beta + \chi^{[2]}_{\alpha\beta\gamma} \, E_\beta E_\gamma + \chi^{[3]}_{\alpha\beta\gamma\delta} \, E_\beta E_\gamma E_\delta + \cdots$$

Nonlocal:

The dipole moment $\mathbb{P}$ might depend on the electric field ELSEWHERE, SOME TIME AGO.

$$P_\alpha(\mathbb{r},t) = \sum_\beta \int dt'\, d\mathbb{r}'\ \chi_{\alpha\beta}(\mathbb{r}-\mathbb{r}', t-t')\ E_\beta(\mathbb{r}', t')$$

here + now — How Far Away, In What Direction — How Long Ago — there + then

[Mills 2.8]

χ depends only on $\mathbb{r}-\mathbb{r}'$ $\Leftrightarrow$ sample is uniform.
What dependence on $\mathbb{r}-\mathbb{r}'$ corresponds to an isotropic sample?

For now we examine

local isotropic linear time-delayed response

$$\mathbb{P}(\mathbb{r},t) = \int_{-\infty}^{t} \chi(t-t')\ \mathbb{E}(\mathbb{r},t')\, dt'$$

$$\mathbb{D} = \mathbb{E} + \mathbb{P} = \int_{-\infty}^{t^+} \varepsilon(t-t')\ \mathbb{E}(t')\, dt'$$

$$\varepsilon(t-t') = \delta(t-t') + \chi(t-t')$$

Essential Concepts

- The material may respond with a time-delay. That gives a dielectric response function $\varepsilon(t - t')$ (t: effect, t': cause)

- The Fourier transform of this quantity is the frequency-dependent dielectric constant $\tilde{\varepsilon}(\omega)$. This gives dispersion

- $\tilde{\varepsilon}(\omega)$ is complex. Its im part gives absorption, while its real part is related to the index of refraction.

- There are integral relationships between $\tilde{\varepsilon}^{re}(\omega)$ and $\tilde{\varepsilon}^{im}(\omega)$. (Kramers-Kronig relationships)

- Simple models give an approximate formula for $\tilde{\varepsilon}^{im}(\omega)$. Quantum theory gives a comparable result.

- From $\tilde{\varepsilon}^{im}(\omega)$, the integral relationship gives $\tilde{\varepsilon}^{re}(\omega)$

- $\tilde{\varepsilon}^{re}$ can be negative.

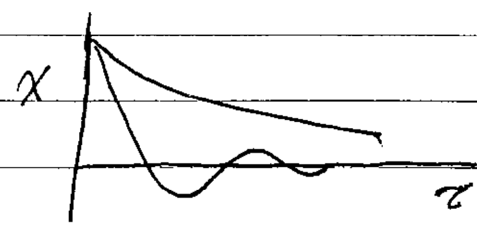
The polarization here and now may depend on the electric field in the past, but NOT on the electric field in the future.

(The little man in the material might remember what we have done in the past, and he may still be responding to that; however he does not know what we are going to do in the future.)

Then $$P_\alpha(\mathbf{r},t) = \int_{-\infty}^{t} dt'\ \chi(t-t')\ E_\alpha(\mathbf{r},t')$$

set $\tau = t - t'$ $$\underbrace{P_\alpha(\mathbf{r},t)}_{\text{now}} = \int_{\underbrace{0}_{(\text{now})}}^{\infty\ (\text{distant past})} \underbrace{\chi(\tau)}_{\text{how long ago}}\ E_\alpha(\mathbf{r}, \underbrace{t-\tau}_{\text{then}})\ d\tau$$

$\chi(\tau) = 0 \quad \tau < 0.$



$\chi(\tau)$ typically decays to zero on a time scale that represents the "relaxation time" of the material. This may be the time required to obtain thermal equilibrium. (picoseconds to days).

If we change $\mathbf{E}$ on a slower time-scale, then

$$P_\alpha(\mathbf{r},t) = \int_0^\infty \chi(\tau)\ E_\alpha(\mathbf{r}, t-\tau)\, d\tau \simeq \int_0^\infty \chi(\tau)\ E_\alpha(\mathbf{r},t)\, d\tau$$

$$= \underbrace{\int_0^\infty \chi(\tau)\, d\tau}\ \ E_\alpha(\mathbf{r},t)$$

$$P_\alpha(\mathbf{r},t) = \chi\ \ E_\alpha(\mathbf{r},t)$$

Define Fourier Transforms:

$$\tilde{\mathbb{E}}(\mathbb{r}, \omega) = \int_{-\infty}^{\infty} e^{i\omega t}\ \mathbb{E}_\beta(\mathbb{r}, t)\ dt$$

$$\tilde{\mathbb{P}}(\mathbb{r}, \omega) = \int e^{i\omega t}\ \mathbb{P}(\mathbb{r}, t)\ dt$$

$$\tilde{\chi}(\omega) = \int_{-\infty}^{\infty} e^{i\omega t}\ \chi(\tau)\ d\tau$$

<u>Theorem</u>

$$\tilde{\mathbb{P}}_\alpha(\mathbb{r}, \omega) = \tilde{\chi}(\omega)\ \tilde{\mathbb{E}}_\beta(\mathbb{r}, \omega)$$

Therefore

$$\mathbb{D}(\mathbb{r}, \omega) = \tilde{\varepsilon}(\omega)\ \mathbb{E}(\mathbb{r}, \omega)$$

$$\tilde{\varepsilon}(\omega) = 1 + \tilde{\chi}(\omega)$$

Proof:

Special Case: Local Isotropic Noninstantaneous

$$P_\alpha(t) = \int_0^\infty \chi(\tau)\, E_\alpha(t-\tau)\, d\tau$$

extend to $-\infty$ with $\chi(\tau)=0$ for $\tau<0$.

$$\tilde{P}(\omega) = \int_{-\infty}^{\infty} e^{i\omega t}\, P(t)\, dt$$

$$= \int_{-\infty}^{\infty} dt\, e^{i\omega t} \int_{-\infty}^{\infty} \chi(\tau)\, E(t-\tau)\, d\tau$$

$$= \int_{-\infty}^{\infty} d\tau\, e^{i\omega\tau} \chi(\tau) \int_{-\infty}^{\infty} dt \quad e^{i\omega t} e^{-i\omega\tau} E(t-\tau)$$

$$= \quad \tilde{\chi}(\omega) \quad \tilde{E}(\omega)$$

Convolution $\Leftrightarrow$ Product

Exercise: Show that this also holds for nonlocal anisotropic case

[pages III. 8-13 do not exist in this version]

a. Analytic

Properties of $\tilde{\varepsilon}(\omega)$ and $\tilde{\chi}(\omega)$ for complex ω.

- They are defined for $\operatorname{Im}\omega > 0$, i.e. in the upper half ω-plane.

- $$[\tilde{\chi}(\omega)]^* = \left[\int_0^\infty e^{+i\omega\tau}\chi(\tau)\,d\tau\right]^*$$
  $$= \left[\int_0^\infty e^{-i\omega^*\tau}\chi^*(\tau)\,d\tau\right]$$

but $\chi(\tau)$ is real for real τ, so

$$[\tilde{\chi}(\omega)]^* = \int_0^\infty e^{+i(-\omega^*)\tau}\chi(\tau)\,d\tau$$

$$= \chi(-\omega^*)$$

Also

$$[\tilde{\varepsilon}(\omega)]^* = \varepsilon(-\omega^*)$$

$\Rightarrow$ • On the positive imaginary ω axis, $\tilde{\varepsilon}(\omega)$ is real.

• On the real ω axis,

$\operatorname{Re}\tilde{\varepsilon}(\omega)$ is a sym fn of ω

$\operatorname{Im}\tilde{\varepsilon}(\omega)$ is an antisym fn of ω

b. Causality and the Kramers-Kronig relationships

Causality condition: $\chi(t-t') = 0 \qquad t < t'$.
(the medium responds to fields from the past, but not from the future)
This implies certain relationships for $\tilde{\varepsilon}(\omega)$

$\tilde{\varepsilon}(\omega)$ is complex. Write $\tilde{\varepsilon}(\omega) = \tilde{\varepsilon}_r(\omega) + i\tilde{\varepsilon}_i(\omega)$

Proposition: the causality condition implies certain integral relationships between $\tilde{\varepsilon}_r(\omega)$ and $\tilde{\varepsilon}_i(\omega)$. We claim

$$\tilde{\varepsilon}_r(\omega) = \left[1 + \frac{2}{\pi} \mathcal{P}\int_0^\infty \frac{\Omega}{(\Omega^2-\omega^2)} \tilde{\varepsilon}_i(\Omega)\, d\Omega\right]$$

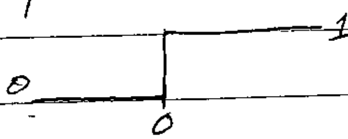
$$\tilde{\varepsilon}_i(\omega) = \left[\frac{2\omega}{\pi} \mathcal{P}\int \frac{\tilde{\varepsilon}_r(\Omega)-1}{(\omega^2-\Omega^2)}\, d\Omega\right]$$

Mills normalization

Proof: since $\varepsilon(\tau) = 0$ for $\tau < 0$, and $\chi(\tau) = 0$ for $\tau < 0$, we can write $\chi(\tau)$ as a product

$$\chi(\tau) = \chi(\tau) H(\tau)$$

where $H(\tau)$ is the Heaviside step fn



It follows that $\tilde{\chi}(\omega)$ is the convolution of itself with the F.T. of the Heaviside fn. Set

$$\tilde{\chi}(\omega) = \int_0^\infty \chi(\tau) e^{i\omega\tau} d\tau = \int_{-\infty}^{\infty} \chi(\tau) H(\tau) e^{i\omega\tau} d\tau$$

$$\tilde{\chi}(\omega) = \int \chi(\tau)\, H(\tau)\, e^{i\omega\tau}\, d\tau$$

$$= \int \chi(\tau')\, \delta(\tau-\tau') \qquad H(\tau)\, e^{i\omega\tau}\, d\tau\, d\tau'$$

$$= \int d\tau d\tau' \quad \chi(\tau') \left[\int \frac{e^{-i\omega'(\tau-\tau')}}{2\pi}\, d\omega'\right] H(\tau) e^{i\omega\tau}$$

$$= \int \frac{d\omega'}{2\pi} \quad \int d\tau'\, e^{i\omega'\tau'} \chi(\tau') \quad \int d\tau\, e^{i(\omega-\omega')\tau} H(\tau)$$

$$= \frac{1}{2\pi} \int \tilde{\chi}(\omega')\ \tilde{H}(\omega-\omega')\ d\omega'$$

$$\tilde{H}(\omega) = \int_{0^-}^{\infty} e^{i\omega\tau}\, H(\tau)\, d\tau$$

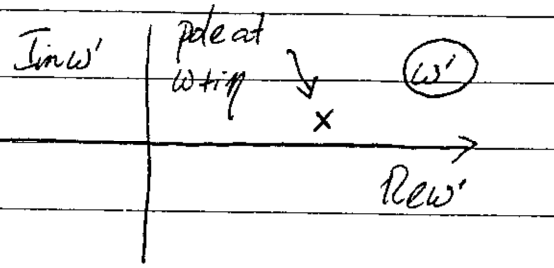
The integral converges if ω has a small + im part:

$$\tilde{H}(\omega) = \left.\frac{e^{i\omega\tau}}{i\omega}\right|_{0^-}^{\infty} = \frac{i}{\omega+i\eta}$$

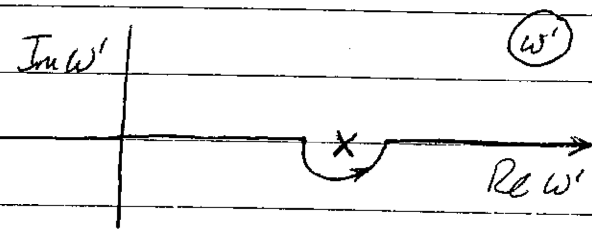
$$\tilde{H}(\omega-\omega') = \frac{i}{\omega-\omega'+i\eta}$$

$$\tilde{\chi}(\omega) = \frac{1}{2\pi}\int_{-\infty}^{\infty} \tilde{\chi}(\omega') \frac{i}{(\omega + i\eta - \omega')}\, d\omega'$$

$$= \frac{1}{2\pi}\int_{-\infty}^{\infty} \tilde{\chi}(\omega') \frac{-i}{\omega' - (\omega + i\eta)}\, d\omega'$$



take limit $\eta \to 0$



$$= \frac{1}{2\pi}\left\{ \mathcal{P}\int_{-\infty}^{\infty} \tilde{\chi}(\omega') \frac{-i}{(\omega' - \omega)}\, d\omega' \right. \qquad *$$

$$\left. + \pi i\,(-i)\,\tilde{\chi}(\omega) \right\}$$

$$\tilde{\chi}(\omega) = \frac{1}{2}\tilde{\chi}(\omega) - i/2\pi\, \mathcal{P}\int_{-\infty}^{\infty} \tilde{\chi}(\omega') \frac{1}{\omega' - \omega}\, d\omega'$$

$$\tilde{\chi}(\omega) = 2 \left(\frac{-i}{2\pi}\right) \mathcal{P}\int_{-\infty}^{\infty} \tilde{\chi}(\omega') \frac{1}{(\omega' - \omega)}\, d\omega'$$

Therefore, writing $\tilde{\chi}(\omega) = \tilde{\chi}_{re}(\omega) + i\,\tilde{\chi}_{im}(\omega)$

$$\tilde{\chi}_{re}(\omega) = -\frac{1}{\pi}\, \mathcal{P}\int_{-\infty}^{\infty} \tilde{\chi}_{im}(\omega')\, \frac{1}{\omega'-\omega}\, d\omega'$$

$$\tilde{\chi}_{im}(\omega) = \frac{1}{\pi}\, \mathcal{P}\int_{-\infty}^{\infty} \tilde{\chi}_{re}(\omega')\, \frac{1}{\omega'-\omega}\, d\omega'$$

$$\tilde{\epsilon}^{real}(\omega) = 1 + \frac{1}{\pi}\, \mathcal{P}\int_{-\infty}^{\infty} \tilde{\epsilon}^{im}_{Mills}(\omega')\, \frac{1}{\omega'-\omega}\, d\omega'$$

$$\tilde{\epsilon}^{im}(\omega) = -\frac{1}{\pi}\, \mathcal{P}\int_{-\infty}^{\infty} \left[\tilde{\epsilon}^{real}_{Mills}(\omega') - 1\right] \frac{1}{\omega'-\omega}\, d\omega'$$

Now $\dfrac{1}{\omega'-\omega} = \dfrac{\omega'+\omega}{\omega'^2-\omega^2}$ Denominator is sym in ω'

ϵ^{im} is anti so $\omega\,\epsilon^{im}$ term gives zero

ϵ^{re} is sym so $\omega'\,\epsilon^{re}$ term gives zero.

Therefore

$$\varepsilon^{re}(\omega) = 1 + \frac{2}{\pi} \mathcal{P} \int_0^\infty \varepsilon^{im}(\omega') \frac{\omega'}{\omega'^2 - \omega^2} \, d\omega'$$

(Right)

$$\varepsilon^{im}(\omega) = -\frac{2}{\pi} \mathcal{P} \int_0^\infty \left(\varepsilon^{re}(\omega') - 1 \right) \frac{\omega}{\omega'^2 - \omega^2} \, d\omega'$$

Mills 2.24 a and b

These integral relationships follow from the facts that ε is real and causal.

Now there are two possibilities:

(1) $\epsilon^{re}(\omega) = 1$ for all ω and $\epsilon^{im}(\omega) = 0$ for all ω.
Such a material acts like the vacuum.

(2) $\epsilon^{re}(\omega) \neq 1$ for some ω. It follows that
$\epsilon^{im}(\omega) = 0$ except possibly for isolated points.

Thus all materials absorb radiation at almost all frequencies. We call the medium "transparent" if $\epsilon^{im}(\omega)$ is small.

Behavior of ε for resonant absorption

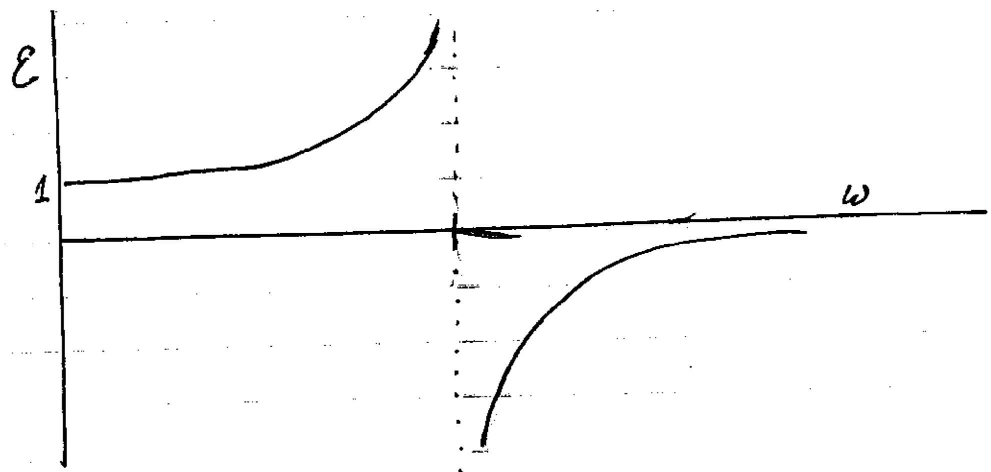
$$\varepsilon^{im} = B\left[\delta(\omega-\omega_0) - \delta(\omega+\omega_0)\right]$$

[must be anti in ω]

$$\varepsilon^{re} = 1 + \frac{2}{\pi}\mathcal{P}\int_0^\infty B\,\delta(\omega'-\omega_0)\,\frac{\omega'}{\omega'^2-\omega^2}\,d\omega'$$

$$= 1 + \frac{2}{\pi} B \frac{\omega_0}{\omega_0^2-\omega^2}$$

$$= 1 + \frac{\Omega^2}{\omega_0^2-\omega^2}$$



For a broadened δ-fn, the princ part is finite

Note: just above resonance, ε can be negative.

"Negative index of refraction" $\Rightarrow$ interesting effects.

They occur if BOTH dielectric const and corresponding magnetic constant are negative.

Dispersion Relation

$$\nabla(\nabla\cdot E) + \left(-\nabla^2 + \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right)E + \frac{1}{c^2}\frac{\partial^2 P}{\partial t^2} = 0$$

$\tilde{P} = \tilde{\chi}\,\tilde{E}$ $\qquad \tilde{D} = \varepsilon\,\tilde{E}$

$\nabla\cdot D = \rho = 0 \Rightarrow \nabla\cdot E = 0$

$$\left[-\nabla^2 - \left(\frac{1+\chi}{c^2}\right)\omega^2\right]\tilde{E}(r,\omega) = 0$$

Traveling waves $\tilde{E}(r,\omega) = e^{i\vec{k}\cdot\vec{r}}$

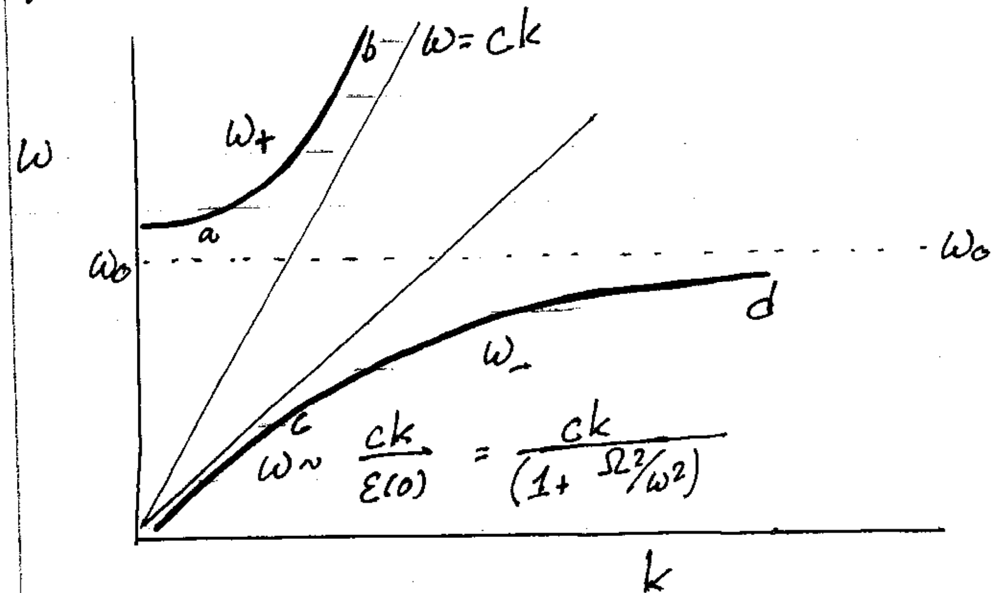
$$+k^2 - \left(\frac{1+\chi}{c^2}\right)\omega^2 = 0$$

$$c^2k^2 = \varepsilon(\omega)\,\omega^2$$

away from resonance

$$= \left(1 + \frac{\Omega^2}{\omega_0^2 - \omega^2}\right)\omega^2$$

Exercise: Show



Properties

a) phase velocity $\to \infty$ grp vel ~ small

b) $v_\phi \sim v_g \sim c$

c) $v_\phi \sim v_g \sim \frac{c}{\varepsilon(0)}$

d) $v_\phi < c$ $v_g \sim 0$

Group velocity is always $\leq c$.
This is NOT a law of nature.

Signal-propagation velocity $\Leftrightarrow$ grp velocity as $\omega \to \infty$ must be less than c.