Lecture 17: Kramers Krönig Relations

The behavior of the complex susceptibility $\tilde{\chi}(\omega)$ is contrained by the laws of physics, namely causality. As we will see, this imposes a relation between the real and imaginary part of $\tilde{\chi}(\omega)$ know as Kramers-Krönig Relations. Such relations are general for linear, causal response.

Consider "Black Box" which determines an output given an input signal

$$S_{in}(t) \longrightarrow \boxed{\;G\;} \longrightarrow S_{out}(t)$$

For S_{out} linearly related by S_{in}, the most general relationship is

$$S_{out}(t) = \int_{-\infty}^{\infty} dt' \; G(t,t') \; S_{in}(t')$$

$$G(t,t') \equiv \text{"Green's Function"} = \text{Response function}$$

Stationarity: $G(t,t') = G(t-t')$ (Response independent of time origin)

Causality: $G(\tau) = 0 \quad \tau < 0$

Output @ t depends on Input @ $t' < t$

Convolution: $S_{out}(t) = \int_{-\infty}^{\infty} dt' \, G(t-t') \, S_{in}(t')$

If $S_{in}(t) = \delta(t-t_0)$ (Unit impulse @ $t = t_0$)

$\Rightarrow S_{out}(t) = G(t-t_0) \Rightarrow G(t-t_0)$ is the output of the black box to an impulse @ t_0

$\Rightarrow$ $G \equiv$ "Impulse response"

The input signal is a superposition of impulses

$$S_{in}(t) = \int_{-\infty}^{\infty} dt' \, \underset{\substack{\uparrow \\ \text{strength of impulse @ } t_0}}{S_{in}(t')} \, \delta(t-t')$$

$\Rightarrow$ $S_{out}(t)$ = superposition of impulse responses

$$= \int_{-\infty}^{\infty} dt' \, S_{in}(t') \, G(t-t') \checkmark$$

In frequency domain

$$\tilde{S}_{out}(\omega) = \int_{-\infty}^{\infty} dt \, S_{out}(t) \, e^{i\omega t}$$

$$S_{out}(t) = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \, \tilde{S}(\omega) \, e^{-i\omega t} = G(t) \underset{\substack{\Downarrow \\ }}{\circledast} S_{in}(t)$$

Convolution

$$\Rightarrow S_{out}(\omega) = \tilde{G}(\omega) \, \tilde{S}_{in}(\omega)$$

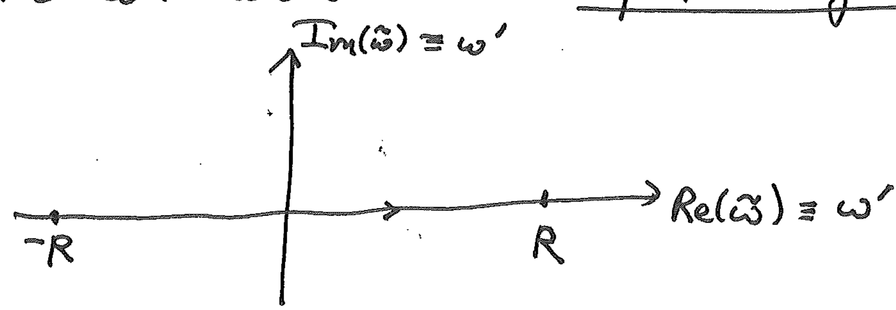
$\Uparrow$ response function = "filter" in frequency domain

The Constraint of causality

Given $G(\tau) = 0$, $\tau < 0$, the frequency domain function $\tilde{G}(\omega)$ is constrained

$$G(\tau) = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \, \tilde{G}(\omega) \, e^{-i\omega t}$$

Analytic continuation to complex omega space, $\tilde{\omega}$



Evaluate integral on Re($\tilde{\omega}$) axis using contour integration.

Recall $\oint dz \, f(z) = 2\pi i \sum_j (\text{Residues})_j$

complex function on complex plane

Residue = Coefficient of Laurent Series

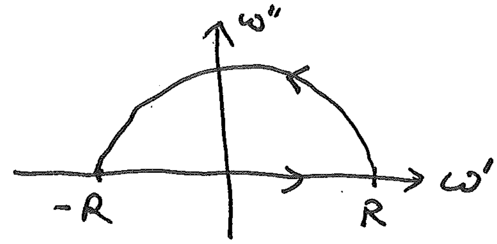
$$f(z) = \sum_{n=-\infty}^{\infty} a_n (z - z_0)^n$$

If z_j is a pole of order n

$$\text{Residue } a_j = \lim_{z \to z_j} (z - z_j)^n f(z)$$

Now, integral on ~~the~~ real(ω) axis = Closed contour integral if the extra piece vanishes

Consider the contour



$$\lim_{R\to\infty} \oint \frac{d\tilde{\omega}}{2\pi} \tilde{f}(\tilde{\omega})\, e^{-i\tilde{\omega}\tau} = \int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} \tilde{f}(\omega')\, e^{-i\omega'\tau}$$

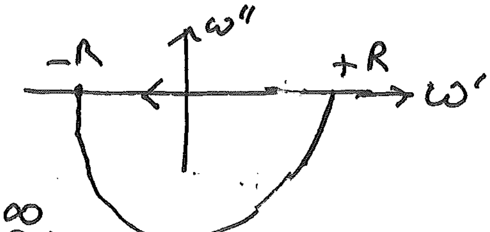
$$+ \lim_{R\to\infty} \int \frac{d\tilde{\omega}}{2\pi} \tilde{f}(\tilde{\omega})\, e^{-i\omega'\tau}\, e^{+\omega''\tau}$$

$\therefore$ If $\tau < 0$ $\quad e^{\omega''\tau} \to 0$ as $\omega'' \to \infty$

$$\Rightarrow \text{for } \tau<0 \quad \int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} \tilde{f}(\omega')\, e^{-i\omega'\tau} = \lim_{R\to\infty} \oint \frac{d\tilde{\omega}}{2\pi} \tilde{f}(\tilde{\omega})\, e^{-i\tilde{\omega}\tau}$$

$= 2\pi i \sum$ Residue in upper complex $\tilde{\omega}$ plane

Similarly Consider



$$\lim_{R\to\infty} \oint \frac{d\tilde{\omega}}{2\pi} \tilde{f}(\tilde{\omega})\, e^{-i\tilde{\omega}\tau} = -\int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} \tilde{f}(\omega')\, e^{-i\omega'\tau} + \lim \int \frac{d\tilde{\omega}}{2\pi} \tilde{f}(\tilde{\omega})\, e^{-i\omega'\tau}\, e^{-\omega''\tau}$$

If $\tau > 0$ $\quad e^{-\omega''\tau} \to 0$ as $\omega'' \to \infty$

$$\Rightarrow \text{for } \tau>0 \quad \int_{-\infty}^{\infty} \frac{d\omega'}{2\pi} \tilde{f}(\omega')\, e^{-i\omega'\tau} = 2\pi i \sum \text{Residues in lower half plane}$$

Implications: Given <u>Causal</u> Greens function

$$G(\tau) = 0 \quad \tau < 0$$

$\Rightarrow$ $\tilde{G}(\tilde{\omega})$ has no poles in the upper complex ω plane

$\Rightarrow$ $\tilde{G}(\tilde{\omega})$ <u>analytic</u> function for $\omega'' > 0$

<u>Example:</u> <u>Lorentz oscillator model</u>

Input $E(t)$
Ouput $p(t)$ dipole response

$$\tilde{p}(\omega) = \tilde{\alpha}(\omega)\,\tilde{E}(\omega)$$

$$\tilde{P}(\omega) = \tilde{\chi}(\omega)\,\tilde{E}(\omega), \qquad \tilde{\chi}(\omega) = N\,\tilde{\alpha}(\omega)$$

(no local field corrections)

$$\Rightarrow \tilde{\chi}(\omega) = \frac{Ne^2}{m} \sum_j \frac{f_j}{(\omega_j^2-\omega^2) - i\omega\Gamma_j}$$

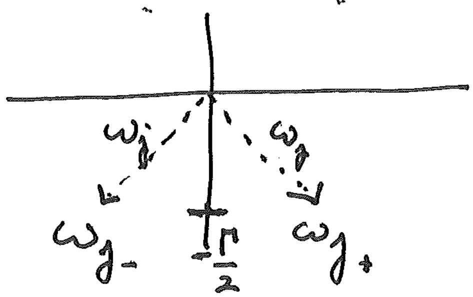
$\frac{Ne^2}{m} = \frac{\omega_p^2}{4\pi}$ (plasma frequency)

Poles: $\omega_j^2 - \omega^2 - i\omega\Gamma_j = 0$

$$(\omega - \omega_{j+})(\omega - \omega_{j-}) = 0$$

where $\omega_{j\pm} = -i\frac{\Gamma}{2} \pm \underbrace{\sqrt{\omega_j^2 - \frac{\Gamma_j^2}{4}}}_{\text{real if underdamped}} \equiv -i\frac{\Gamma}{2} \pm \Omega$

$\Rightarrow$ Poles only in lower $\tilde{\omega}$ plane



~~Eam~~ Response function in time domain

for $\tau > 0$ $\quad \chi(\tau) = \int_{-\infty}^{\infty} \frac{d\tilde{\omega}}{2\pi} \tilde{\chi}(\tilde{\omega}) e^{-i\tilde{\omega}\tau} = -2\pi i \sum_{\substack{\text{lower} \\ \text{half}}} \text{Residues}$

Aside $\quad \frac{\tilde{\chi}(\omega)}{2\pi} e^{-i\omega\tau} = \frac{1}{2\pi} \sum_j \frac{\omega_p^2/4\pi \;\; f_j}{-(\omega-\omega_{j+})(\omega-\omega_{j-})} e^{-i\omega\tau}$

$\Rightarrow$ Residue at ω_{j+} $\quad \frac{e^{-i\omega_{j+}\tau}}{2\pi} \quad \frac{\frac{\omega_p^2}{4\pi} f_j}{-(\omega_{j+}-\omega_{j-})}$

Residue at ω_{j-} $\quad \frac{e^{-i\omega_{j-}\tau}}{2\pi} \quad \frac{\frac{\omega_p^2}{4\pi} f_j}{-(\omega_{j-}-\omega_{j+})}$

$\Rightarrow$ Greens function for dipole response
(susceptibility in time domain)

for $\tau > 0$ $$\chi(\tau) = \frac{\omega_p^2}{4\pi} \sum_j f_j \left(i \frac{e^{-i\omega_{j+}\tau}}{\omega_{j+} - \omega_{j-}} + i \frac{e^{-i\omega_{j+}\tau}}{\omega_{j-} - \omega_{j+}} \right)$$

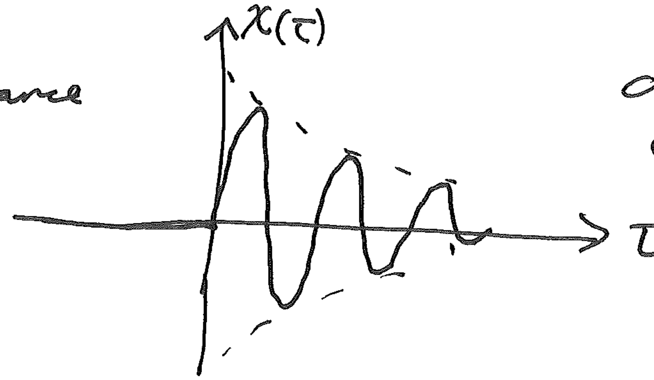
$$= \frac{\omega_p^2}{4\pi} \sum_j f_j \left(i \frac{e^{-i\Omega_j \tau} e^{-\frac{\Gamma_j}{2}\tau}}{2\Omega_j} - i \frac{e^{+i\Omega_j \tau} e^{-\frac{\Gamma_j}{2}\tau}}{2\Omega_j} \right)$$

$$= \frac{\omega_p^2}{4\pi} \sum_j f_j \left(\frac{\sin \Omega_j \tau}{\Omega_j} e^{-\frac{\Gamma_j}{2}\tau} \right)$$

$\chi(\tau) = 0$ for $\tau < 0$

Single resonance

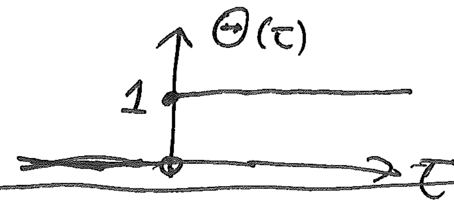
(impulse response)



decaying oscillation at $\sqrt{\omega_0^2 - \frac{\Gamma^2}{4}}$

Formally, define Heavyside Step function

$$\Theta(\tau) = \begin{cases} 0 & t \leq 0 \\ 1 & t > 0 \end{cases}$$



$$\Rightarrow \boxed{\tilde{\chi}(\tau) = \Theta(\tau) \sum_j \frac{\omega_p^2}{4\pi} f_j \left(\frac{\sin \Omega_j \tau}{\Omega_j} \right) e^{-\Gamma_j \tau/2}}$$

Kramers – Krönig Relations

Given Causal response: $\chi(\tau) = \Theta(\tau)\,\chi(\tau)$

$$\therefore \quad \tilde{\chi}(\omega) = \frac{1}{2\pi}\, \tilde{\Theta}(\omega) \circledast \tilde{\chi}(\omega)$$

$\Uparrow$ Fourier transform of step-function

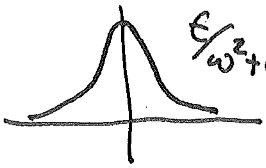
$$\tilde{\Theta}(\omega) = \int_{-\infty}^{\infty} \Theta(t)\, e^{i\omega t}\, dt = \int_0^{\infty} e^{i\omega t}\, dt$$

$$= \left.\frac{e^{i\omega t}}{i\omega}\right|_0^{\infty} \rightarrow \underline{\text{Undefined}}$$

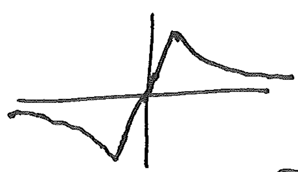
Distribution rather than true function
Regularize, then take limit

$$\tilde{\Theta}(\omega) = \lim_{\epsilon \to 0} \int_{-\infty}^{\infty} \Theta(t)\, e^{i\omega t} e^{-\epsilon t} = \lim_{\epsilon \to 0} \int_0^{\infty} e^{i(\omega + i\epsilon)t}\, dt$$

$$= \lim_{\epsilon \to 0} \left[-\frac{1}{i\omega - \epsilon} \right] = \lim_{\epsilon \to 0} \left[\frac{\epsilon}{\omega^2 + \epsilon^2} + i\, \frac{\omega}{\omega^2 + \epsilon^2} \right]$$



$\epsilon / (\omega^2 + \epsilon^2) \rightarrow \pi\, \delta(\omega)$ as $\epsilon \to 0$



$\dfrac{\omega}{\omega^2 + \epsilon^2}$

$\epsilon \to 0 \rightarrow \mathbb{P}\left[\frac{1}{\omega}\right]$

$\uparrow$ Cauchy's Principal Value

$$\boxed{\tilde{\Theta}(\omega) = \pi\, \delta(\omega) + i\, \mathbb{P}\left(\frac{1}{\omega}\right)}$$

Cauchy's Principal value:

Like the delta function, only make sense in integral

$$\mathbb{P}\left(\frac{1}{\omega}\right) = \frac{1}{\omega}, \text{ except at } \omega = 0$$

(cuts out singularity)

$$\mathbb{P}\int_a^b \frac{f(x)}{x}\,dx \equiv \lim_{\epsilon\to 0}\left[\int_{+a}^{-\epsilon} + \int_{\epsilon}^{b}\right]\frac{f(x)}{x}\,dx$$

↑ interval including origin

Thus:

$$\tilde{\chi}(\omega) = \frac{1}{2\pi}\tilde{\Theta}(\omega) \circledast \tilde{\chi}(\omega)$$

$$= \frac{1}{2\pi}\Big(\pi\underbrace{\delta(\omega) \circledast \tilde{\chi}(\omega)} + i\,\mathbb{P}\left[\frac{1}{\omega}\right] \circledast \tilde{\chi}(\omega)\Big)$$

$$= \int d\omega'\,\delta(\omega-\omega')\,\tilde{\chi}(\omega') = \tilde{\chi}(\omega)$$

$$= \frac{1}{2}\tilde{\chi}(\omega) + \frac{i}{2\pi}\mathbb{P}\left[\frac{1}{\omega}\right] \circledast \tilde{\chi}(\omega)$$

$$\Rightarrow \quad \tilde{\chi}(\omega) = \frac{i}{\pi}\mathbb{P}\left[\frac{1}{\omega}\right] \circledast \tilde{\chi}(\omega)$$

$$\Rightarrow \quad \tilde{\chi}(\omega) = \frac{i}{\pi}\mathbb{P}\int_{-\infty}^{\infty} d\Omega\,\frac{\tilde{\chi}(\Omega)}{\omega - \Omega}$$

Equate real and imaginary parts: $\tilde{\chi}(\omega) = \tilde{\chi}'(\omega) + i\,\tilde{\chi}''(\omega)$

$$\Rightarrow \tilde{\chi}'(\omega) = -\frac{1}{\pi}\mathbb{P}\int_{-\infty}^{\infty}\frac{\tilde{\chi}''(\Omega)\,d\Omega}{\omega-\Omega} = \frac{1}{\pi}\mathbb{P}\int_{-\infty}^{\infty}\frac{\tilde{\chi}''(\Omega)\,d\Omega}{\Omega-\omega}$$

$$\tilde{\chi}''(\omega) = \frac{1}{\pi}\mathbb{P}\int_{-\infty}^{\infty}\frac{\tilde{\chi}'(\Omega)\,d\Omega}{\omega-\Omega} = -\frac{1}{\pi}\mathbb{P}\int_{-\infty}^{\infty}\frac{\tilde{\chi}'(\Omega)\,d\Omega}{\Omega-\omega}$$

Since $\chi(\tau)$ is <u>real</u>

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

(on real ω axis)

$\Rightarrow \tilde{\chi}'(\omega) = \tilde{\chi}'(-\omega)$ symmetric function

$\tilde{\chi}''(\omega) = -\tilde{\chi}''(-\omega)$ antisymmetric function

$$\Rightarrow \boxed{\begin{aligned} \tilde{\chi}'(\omega) &= \frac{2}{\pi}\mathbb{P}\int_{0}^{\infty}\frac{\Omega\,\tilde{\chi}''(\Omega)}{\Omega^2-\omega^2}\,d\Omega \\ \tilde{\chi}''(\omega) &= -\frac{2}{\pi}\mathbb{P}\int_{0}^{\infty}\frac{\omega\,\tilde{\chi}'(\Omega)}{\Omega^2-\omega^2}\,d\Omega \end{aligned}}$$

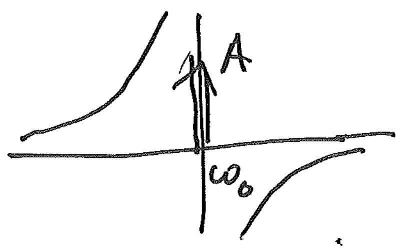
— Kramers Krönig Relations

Useful for understanding general properties of dispersion and propagation

Example: $\tilde{\chi}''(\omega) = A\,\delta(\omega-\omega_0)$ + slowly varying terms
(resonance @ ω_0 of ~~the~~ strength A)

$\Rightarrow$ Near ω_0 $\quad \tilde{\chi}'(\omega) \cong \frac{2A}{\pi}\frac{\omega_0}{\omega_0^2-\omega^2} \sim -\frac{2\omega_0 A}{\pi}\frac{1}{\omega^2}$

off resonance

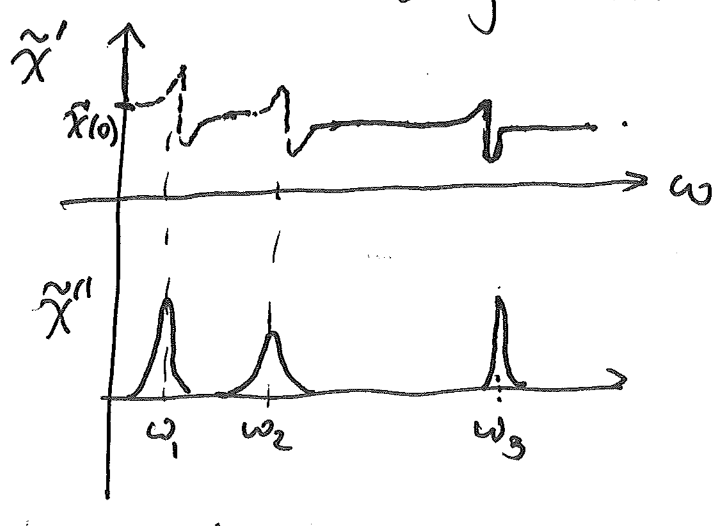


For $\omega \ll \omega_0$ $\quad \tilde{\chi}'(\omega) \cong \frac{2}{\pi\omega_0}A$ $\quad$ (D.C. polarizability related to lowest absorption resonance)

zero frequency sum rule

$$\tilde{\chi}'(0) = \frac{2}{\pi}\int_0^\infty \frac{\tilde{\chi}''(\Omega)}{\Omega}\,d\Omega$$

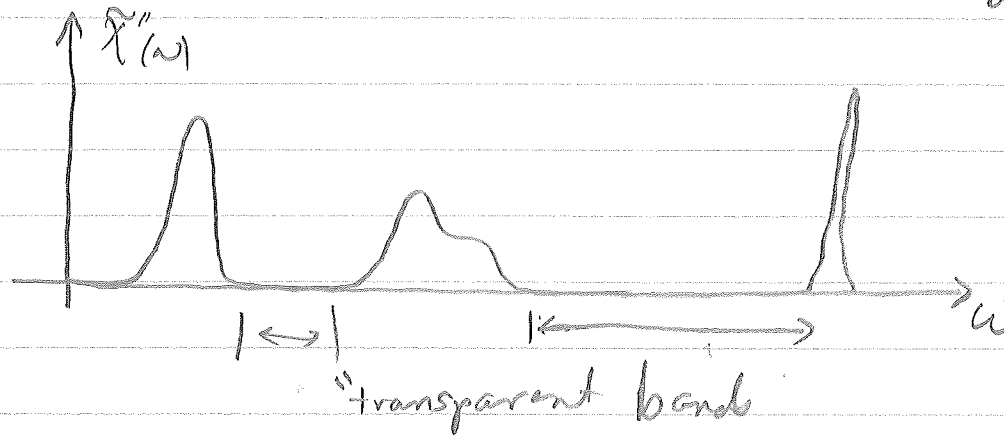
Dominated by resonance @ low freq.



Normal Dispersion

Consider a medium that does not amplify the field

$\Rightarrow \quad \tilde{\chi}''(\omega) \geq 0 \quad \forall \omega$: Absorption only (passive medium)



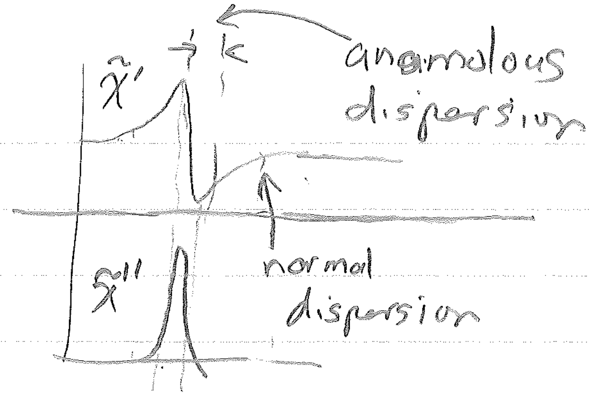
For the range of frequencies for which $\tilde{\chi}''(\omega) \simeq 0$ the medium is transparent, with negligible absorption. In such bands,

$$\tilde{\chi}'(\omega) = \frac{2}{\pi} \mathbb{P} \int_0^\infty \frac{\Omega \tilde{\chi}''(\Omega)}{\Omega^2 - \omega^2} \approx \frac{2}{\pi} \int_0^\infty \frac{\Omega \tilde{\chi}''(\Omega)}{\Omega^2 - \omega^2}$$

Since $\tilde{\chi}''(\omega) \simeq 0$, so no pole @ $\Omega = \omega$

$$\Rightarrow \frac{d}{d\omega} \tilde{\chi}'(\omega) = \frac{4\omega}{\pi} \int_0^\infty \frac{\Omega \tilde{\chi}''(\Omega)}{\Omega^2 - \omega^2} \geq 0$$

$\Rightarrow$ For a passive medium in a transparent band, $\frac{d}{d\omega} \tilde{\chi}'(\omega) > 0$



$\frac{d\tilde{\chi}'}{d\omega} > 0$: "normal dispersion"

$\frac{d\tilde{\chi}'}{d\omega} < 0$: "Anamolous dispersion"

In a transparent band the index of refraction

$$n(\omega) = \sqrt{1 + 4\pi \tilde{\chi}'(\omega)} > 0$$

$$\Rightarrow \frac{dn}{d\omega} = \frac{2\pi}{n} \frac{d\tilde{\chi}'(\omega)}{d\omega} > 0$$

$\Rightarrow$ Normal dispersion n increases with ω

Recall group velocity $\frac{1}{v_g} = \frac{dk}{d\omega} = \frac{1}{c}\frac{d(\omega n)}{d\omega}$

$\Rightarrow$ "Group index" $n_g \equiv \frac{d}{d\omega}(\omega n) = n + \omega \frac{dn}{d\omega}$

$\Rightarrow$ For a passive medium with normal dispersion

$$\boxed{\begin{aligned} &\omega \frac{dn}{d\omega} > 0 \\ &\Rightarrow \quad n_g > n \end{aligned}}$$

More stringent bound on n_g

Aside: $$\frac{d}{d\omega}\left(\omega^2 \tilde{\chi}'(\omega)\right) = 2\omega \tilde{\chi}'(\omega) + \omega^2 \frac{d\tilde{\chi}'}{d\omega}$$

$$= \frac{2}{\pi}\int_0^\infty d\Omega\, \tilde{\chi}''(\Omega)\left[\frac{2\omega\Omega}{\Omega^2-\omega^2} + \frac{2\omega^3\Omega}{(\Omega^2-\omega^2)^2}\right]$$

$$= \frac{4\omega}{\pi}\int_0^\infty d\Omega\, \frac{\Omega^3 \tilde{\chi}''(\Omega)}{(\Omega^2-\omega^2)^2} \geq 0$$ for passive medium

$$\therefore \frac{d\tilde{\chi}'}{d\omega} > -\frac{2}{\omega}\tilde{\chi}'(\omega)$$: Normal dispersion w/ $\tilde{\chi}''(\omega) > 0$

Now, $$\frac{d}{d\omega}\tilde{\chi}' = \frac{d}{d\omega}\left(\frac{n^2-1}{4\pi}\right) = \frac{1}{4\pi}\left(2n\frac{dn}{d\omega}\right)$$

$$\geq -\frac{2}{\omega}\left[\frac{n^2-1}{4\pi}\right]$$

$$\Rightarrow \omega\frac{dn}{d\omega} \geq \frac{1}{n} - n \Rightarrow n + \omega\frac{dn}{d\omega} \geq \frac{1}{n}$$

$\Rightarrow$ Normal Dispersion and $\tilde{\chi}''(\omega) > 0$

$$\boxed{V_g \geq \frac{1}{n}}$$

We thus have for a passive medium in a transparent band

$$n_g \geq n \quad \text{and} \quad n_g \geq \frac{1}{n}$$

$$\Rightarrow \quad n_g \geq 1$$

$$\Rightarrow \quad \boxed{v_g \leq c} \quad \text{Subluminal}$$

Even if $n < 1$ and phase velocity $v_p = \frac{c}{n} > c$

For a transparent / passive med $v_g \leq c$

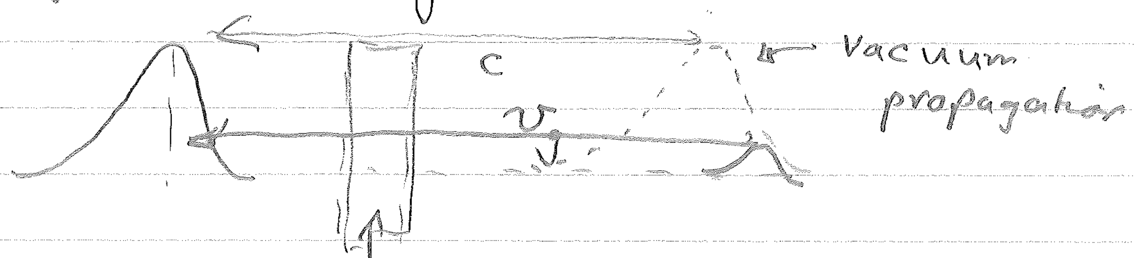
<u>Anomalous dispersion</u>

When $\frac{dn}{d\omega} < 0$, for an absorptive medium we can have $v_g > c$! <u>superluminal</u> !

Have no fear, this does not violate Einstein causality

(i) In a region of anomalous dispersion / absorption we often have a very rapid change of the index with ω, and the whole slowly-varying envelope approximation

(ii) For a sufficiently broad absorption band and smooth Gaussian pulse Garrett and McCumber predicted that pulse reshaping could lead to an efficient/effective superluminal $v_g > c$ (Phys Rev A. 1 305, 1970)



absorptive medium w/ anomalous dispersion

The pulse that emerges is Gaussian but highly attenuated. The information is in the tail, and no information travels faster than c. This was seen in experiment by Chu & Wong (Phys. Rev. Lett. 48 738 (1982)

Active medium

A very interesting development of the last decades was the study of dispersion in active media with gain. Then the no-go theorems of Kramers-Krönig we derived don't apply. One can have anomalous dispersion and transparency. This leads to negative group velocity (see Homework)

- Theory: R.Y. Chiao in "Amazing Light", Springer 1996
- Experiment: Wang, Kuzmich, & Dogariu, Nature 406 277, (2000)