Plan :

(1) Relationship between Re + Im part of χ

(2) Susceptibility of a two level system

(3) Polaritons

New Homework, due 3 Feb.

7.4, 7.5, 7.6, 7.7, 7.8

Reminder from last time

From last time :

$$P(t) = \epsilon_0 \int_{-\infty}^{\infty} dt' \ \chi(t-t') E(t')$$

We want causality $\Rightarrow$ $E(t')$ can influence $P(t)$ only when $t > t'$.

Hence, for all susceptibilities [at least for equilibrium systems] we have:

$$\Rightarrow \chi(t) = \theta(t) \chi(t)$$

$\Rightarrow$ All poles of $\chi(\omega)$ are in the upper half-plane.

As an example, for the driven + damped classical oscillator

$$\chi(\omega) = \frac{C}{\omega_0^2 - \omega^2 + i \Gamma \omega} \qquad \chi(t) = C \left[2i e^{-\Gamma t/2} \frac{\sin\left[\frac{1}{2} t \sqrt{4\omega_0^2 - \Gamma^2}\right]}{\sqrt{4\omega_0^2 - \Gamma^2}} \right] \theta(t)$$

where $C = \frac{q^2}{\epsilon_0 m} \left(\frac{N}{V} \right)$

An extended example is a medium composed of a bunch of oscillators, each with susceptibility χ_α.

In this case, we can simply sum their susceptibilities [use for HW]

$$\chi = \sum_\alpha \chi_\alpha$$

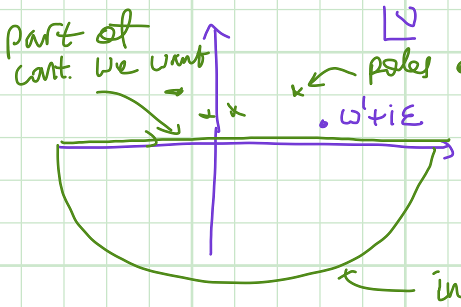
Kramers – Kronig: relation between χ' and χ''

Consider the integral

$$\int_{-\infty}^{\infty} d\omega \, \frac{\chi(\omega)}{\omega' - \omega + i\varepsilon}$$

where $\varepsilon \to 0^+$ and $\omega' \in \mathbb{R}$

We can perform this integral by contour integration



integral over this contour is zero

[generically $\chi(\omega) \sim \frac{1}{\omega}$ for $|\omega| \to \infty$]

Choosing the lower contour we observe that the integral is zero.

$$\int_{-\infty}^{\infty} d\omega \, \frac{\chi(\omega)}{\omega' - \omega + i\varepsilon} = \oint d\omega \, \frac{\chi(\omega)}{\omega' - \omega + i\varepsilon} = 0$$

Now let's apply the Dirac formula

$$0 = \int_{-\infty}^{\infty} \frac{d\omega \, \chi(\omega)}{\omega' - \omega + i\varepsilon} = P\int_{-\infty}^{\infty} d\omega \, \frac{\chi(\omega)}{\omega' - \omega} + i\pi \int_{-\infty}^{\infty} d\omega \quad \delta(\omega - \omega') \, \chi(\omega)$$

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

Hence: $\chi(\omega) = \frac{1}{i\pi} P\int_{-\infty}^{\infty} d\omega' \, \frac{\chi(\omega')}{\omega' - \omega}$

Providing a relation between the real and imaginary parts of χ

Explicitly:

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

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

If we add the assumption that

$$\chi'(\omega) = \chi'(-\omega)$$
$$\chi''(\omega) = -\chi''(-\omega)$$

we can derive a somewhat easier to use relation:

$$\chi'(\omega) = \frac{1}{\pi}\left[\underset{\substack{\parallel \\ P\int_{\infty}^{0} \frac{-d\tilde\omega\, \chi''(-\tilde\omega)}{-\tilde\omega - \omega} = P\int_{0}^{\infty} \frac{d\tilde\omega\, \chi''(\tilde\omega)}{\tilde\omega + \omega}}}{P\int_{-\infty}^{0} \frac{d\tilde\omega\, \chi''(\tilde\omega)}{\tilde\omega - \omega}} + P\int_{0}^{\infty} \frac{d\tilde\omega\, \chi''(\tilde\omega)}{\tilde\omega - \omega} \right] = \frac{2}{\pi}\int_0^\infty \frac{d\tilde\omega\, \chi''(\tilde\omega)}{\tilde\omega^2 - \omega^2}\,\tilde\omega$$

similar Reasoning leads to

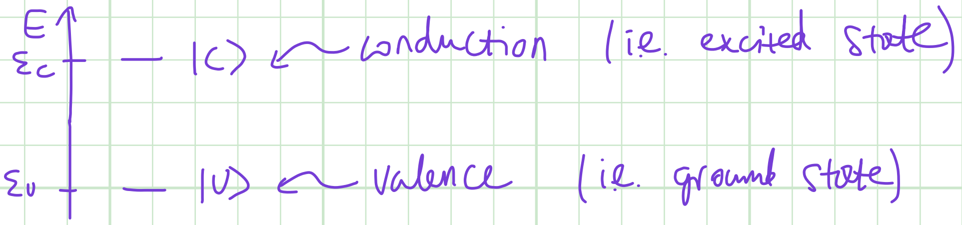
$$\chi''(\omega) = -\frac{2}{\pi} P\int_0^\infty d\tilde\omega \,\frac{\chi'(\tilde\omega)\,\omega}{\tilde\omega^2 - \omega^2}$$

Note ⇒ if we have χ' we can compute χ'' and vice-verse

⇒ Relation between absorbtion + reflection spectrum

⇒ often difficult to use as we need χ' or χ'' over a large range of ω (typically stitch together several measurments + model $\omega \to 0$ + $\omega \to \infty$ parts ...)

Susceptibility of a 2-level system:



How to write the Wave function?

$$|\Psi\rangle = \Psi_v |v\rangle + \Psi_c |c\rangle$$

[We will need to modify this slightly for interaction picture, but we need to define $\mathcal{H}$ first.]

What is the Hamiltonian?

$$\mathcal{H} = \underset{\substack{\uparrow \\ \text{2-level system}}}{\mathcal{H}_0} + V_{int} \leftarrow \text{coupling to } \vec{E}\text{-field}$$

$$\mathcal{H}_0 \equiv \mathcal{E}_v |v\rangle\langle v| + \mathcal{E}_c |c\rangle\langle c| = \begin{array}{c} \begin{array}{cc} |v\rangle & |c\rangle \end{array} \\ \begin{pmatrix} \mathcal{E}_v & 0 \\ 0 & \mathcal{E}_c \end{pmatrix} \end{array}$$

$$V_{int} = -\hat{x}\, qE = -qE \begin{pmatrix} \langle v|x|v\rangle & \langle v|x|c\rangle \\ \langle c|x|v\rangle & \langle c|x|c\rangle \end{pmatrix} = -\begin{pmatrix} 0 & \alpha \\ \alpha^* & 0 \end{pmatrix} E \Leftarrow \text{assume } \langle v|x|v\rangle = \langle c|x|c\rangle = 0$$

} notice, these are almost same!

This is typical

What is the polarization?

$$P \equiv q\hat{x} = q \begin{pmatrix} \langle v|x|v\rangle & \langle v|x|c\rangle \\ \langle c|x|v\rangle & \langle c|x|c\rangle \end{pmatrix} = \begin{pmatrix} 0 & \alpha \\ \alpha^* & 0 \end{pmatrix}$$

Evolution of polarization:

$$P(t) = \langle \psi_0 | \hat{P}(t) |\psi_0\rangle = \langle \psi_0 | e^{i\int_{-\infty}^{t} H(t')dt'} \hat{P}(t) e^{-i\int_{-\infty}^{t} H(t')dt'} |\psi_0\rangle$$

$$= \langle \psi_0 | \left(1 + i\int_{-\infty}^{t} V_{int}(t')dt'\right) P(t) \left(1 - i\int_{-\infty}^{t} V_{int}(t')dt'\right) |\psi_0\rangle$$

[this is in the interaction picture]

$$= \langle \psi_0 | P(t) |\psi_0\rangle + i\int_{-\infty}^{t} \langle \psi_0 [V_{int}(t'), P(t)] \psi_0\rangle dt' + O(V_{int}^2)$$

Assume we start from the ground state $|\psi_0\rangle = |v\rangle$. By assumption on matrix elements of $\hat{x}$, $\langle \psi_0 | \hat{P}(t) | \psi_0 \rangle = \langle v | e^{iH_0 t} \hat{P} e^{-iH_0 t} |v\rangle = 0$

So let us now look at the first order in V_{int} correction

$$P(t) = i\int_{-\infty}^{t} \langle v | [V_{int}(t'), P(t)] | v\rangle dt'$$

$$= i\int_{-\infty}^{t} \langle v | e^{+iH_0 t'} \hat{V}_{int}(t') e^{-iH_0 t'} e^{+iH_0 t} \hat{P}(t) e^{-iH_0 t} - e^{+iH_0 t} \hat{P} e^{-iH_0 t} e^{+iH_0 t'} \hat{V} e^{-iH_0 t'} |v\rangle \, dt'$$

$$= -i\int_{-\infty}^{t} e^{+i\mathcal{E}_v t'} \alpha E(t') e^{-i\mathcal{E}_c (t'-t)} \alpha^* e^{-i\mathcal{E}_v t} - e^{+i\mathcal{E}_v t} \alpha e^{-i\mathcal{E}_c (t-t')} \alpha^* E(t') e^{-i\mathcal{E}_v (t')} \, dt'$$

$$= -i\int_{-\infty}^{t} e^{-i(\mathcal{E}_c - \mathcal{E}_v)(t'-t)} |\alpha|^2 E(t') - |\alpha|^2 e^{-i(\mathcal{E}_c - \mathcal{E}_v)(t-t')} E(t') \, dt'$$

$$= \int_{-\infty}^{t} \underbrace{+2|\alpha|^2 \sin[(\mathcal{E}_c - \mathcal{E}_v)(t-t')]}_{\chi(t-t')/\rho} E(t') \, dt'$$

~ density of oscillators.

$e^{ix} = \cos(x) + i\sin(x)$

$-i\,[i\sin(+x) - i\sin(x)] = -2\sin(x)$

By causality $\chi(t) = \rho\,\theta(t) \left\{ 2|\langle v|x|c\rangle|^2 q^2 \sin([\mathcal{E}_c - \mathcal{E}_v]t) \right\}$

This result looks very similar to the classical one, except no damping.

Taking the fourier transform we obtain

$$\chi(\omega) = \int_{-\infty}^{0} dt \quad e^{i\omega t} \left[\theta\, 2|q\langle v|x|c\rangle|^2 \sin(\omega_0 t)\right]$$

$\omega_0 \equiv \varepsilon_c - \varepsilon_v$

compute the $\int \Rightarrow \int_{-\infty}^{0} e^{i\omega t} \frac{\left(e^{i\omega_0 t} - e^{-i\omega_0 t}\right)}{2i} dt = \frac{1}{2i}\left[\frac{e^{i(\omega+\omega_0)t}}{i(\omega+\omega_0)} - \frac{e^{i(\omega-\omega_0)t}}{i(\omega-\omega_0)}\right]_{-\infty}^{0} = -\frac{1}{2}\left[\frac{1}{\omega+\omega_0-i\varepsilon} - \frac{1}{\omega-\omega_0-i\varepsilon}\right]$

$$= P\frac{1}{2}\left[\frac{+2\omega_0}{\omega^2-\omega_0^2}\right] - i\pi/2\ \delta(\omega+\omega_0) + \frac{i\pi}{2}\delta(\omega-\omega_0)$$

ω must have a small Im part for convergence

$$\chi(\omega) = 2|q\langle v|x|c\rangle|^2 \left\{P\left[\frac{\omega_0}{\omega^2-\omega_0^2}\right] + \frac{i\pi}{2}\left[\delta(\omega-\omega_0) - \delta(\omega+\omega_0)\right]\right\}$$

$\Rightarrow$ this matches 7.32

Polaritons:

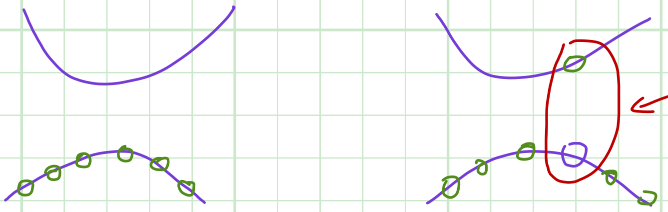
A quasi-particle formed by "mixture" of:

polar: an excitation that polarizes the material

ton: a photon

What sort of excitations polarize the material?

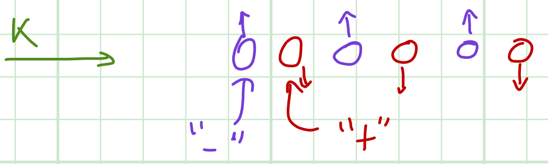
Excitons $\Rightarrow$



this is really the 2-level system from earlier in class

$\Rightarrow$ "exciton polaritons"

optical phonons $\Rightarrow$ must have solid with at least 2 distinct atoms per unit cell



"TO"

this transverse optical phonon will induce polarization in the crystal

$\Rightarrow$ "phonon polaritons" or just "polaritons"