Lect 9: optical phonon — polariton, Huang's equation

Conside ionic crystal. In each unit cell, there exist a pair of cation and anion. We define $\underbrace{\vec{u}_+ - \vec{u}_-}_{\text{the relative displacement}}$: the polarization field.

$\Rightarrow$ optical phonon couples strongly with EM field.

We define $\vec{W} = \rho^{1/2}(\vec{u}_+ - \vec{u}_-)$, where $\rho = \frac{M}{V_2}$ and $M = \frac{M_1 M_2}{M_1 + M_2}$.

Then the kinetic energy density $\boxed{T = \frac{1}{2}\dot{\vec{W}}\dot{\vec{W}}}$

The potential energy $\phi = \phi_{elastic} + \phi_{polar}$

$$\phi_{elastic} = \frac{1}{2}\gamma_{11}\vec{W}\cdot\vec{W}$$

$$\phi_{polar} = -\int_0^E \vec{P}\cdot d\vec{E}$$

$$\boxed{\begin{aligned} d\mathcal{E} &= \vec{E}\cdot d\vec{P} \\ d(\mathcal{E} - \vec{E}\cdot\vec{P}) &= -\vec{P}\cdot d\vec{E} \end{aligned}}$$

Gibbs free energy

plug in $\vec{P} = \gamma_{12}\vec{W} + \gamma_{22}\vec{E}$ ← $\vec{E}$ is the macroscopic field after averaging ore microscopic configurations.

relative displacement of ions

electronic polarization on ions

$$\Rightarrow \phi_{polar} = -\left(\gamma_{12}\vec{W}\cdot\vec{E} + \frac{1}{2}\gamma_{22}\vec{E}\cdot\vec{E}\right)$$

$$\Rightarrow \boxed{\mathcal{L} = T - \phi = \frac{1}{2}\dot{\vec{W}}\cdot\dot{\vec{W}} - \left(\frac{1}{2}\gamma_{11}\vec{W}\cdot\vec{W} - \gamma_{12}\vec{W}\cdot\vec{E} - \frac{1}{2}\gamma_{22}\vec{E}\cdot\vec{E}\right)}$$

In this Langrangian, we neglected $(\nabla\vec{W})^2$, because optical phonons are gapped. The effect of $(\nabla\vec{W})^2$ is weak, and thus we can neglect it band width.

Do variation: $\delta L = -\frac{\partial^2}{\partial t^2}\vec{W}\cdot\delta\vec{W} - \gamma_{11}\vec{W}\cdot\delta\vec{W} + \gamma_{12}\vec{E}\cdot\delta\vec{W}$

$$= \delta\vec{W}\left[-\ddot{\vec{W}} - \gamma_{11}\vec{W} + \gamma_{12}\vec{E}\right]$$

$$\Rightarrow \boxed{\begin{aligned}\ddot{\vec{W}} &= -\gamma_{11}\vec{W} + \gamma_{12}\vec{E} \\ \vec{P} &= \gamma_{12}\vec{W} + \gamma_{22}\vec{E}\end{aligned}}$$

forced harmonic oscillator, $\sqrt{\gamma_{11}}$ is intrinsic frequency.

Huang Kun Equation for ionic crystals.

γ's can be fitted by observables.

- Di-electric function for ionic crystals

Long-wave length limit: $k \to 0$, try $\begin{cases} \vec{E} = \vec{E}_0 e^{-i\omega t} \\ \vec{P} = \vec{P}_0 e^{-i\omega t} \\ W = \vec{W}_0 e^{-i\omega t} \end{cases}$

$$\Rightarrow (-\omega^2 + \gamma_{11})\vec{W}_0 = \gamma_{12}\vec{E}_0$$

$$\Rightarrow \vec{P}_0 = \left\{\gamma_{22} + \frac{\gamma_{12}^2}{\gamma_{11} - \omega^2}\right\}\vec{E}_0$$

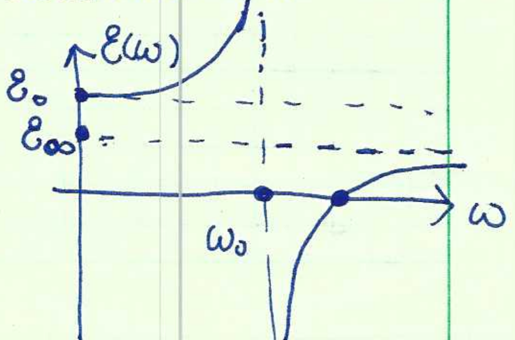
$$\Rightarrow \vec{D}_0 = \vec{E}_0 + 4\pi\vec{P}_0 = \varepsilon\vec{E}_0 \Rightarrow \boxed{\varepsilon(\omega) = 1 + 4\pi\left(\gamma_{22} + \frac{\gamma_{12}^2}{\gamma_{11} - \omega^2}\right)}$$

it has a pole at $\omega = \omega_0 = \sqrt{\gamma_{11}}$, which can determine γ_{11}.

at $\omega \gg \omega_0$, $\varepsilon = \varepsilon_\infty = 1 + 4\pi\gamma_{22}$

(high frequency)

$\omega \ll \omega_0$, $\varepsilon = \varepsilon_0 = 1 + 4\pi\left(\gamma_{22} + \frac{\gamma_{12}^2}{\gamma_{11}}\right)$



$$\Rightarrow \begin{cases} \gamma_{11} = \omega_0^2 \\ \gamma_{12} = \left(\dfrac{\varepsilon_0 - \varepsilon_\infty}{4\pi}\right)^{1/2} \omega_0 \\ \gamma_{22} = \dfrac{\varepsilon_\infty - 1}{4\pi} \end{cases} \quad \text{and } \varepsilon(\omega) = \varepsilon_\infty + \frac{\varepsilon_0 - \varepsilon_\infty}{\omega_0^2 - \omega^2}\,\omega_0^2$$

- Longitudinal wave: (no external field)

$$\nabla\cdot\vec{D}_L = \nabla\cdot(\vec{E}_L + 4\pi\vec{P}_L) = 0 \quad \Rightarrow \quad \vec{E}_L = -4\pi\vec{P}_L$$

$$\Rightarrow \begin{cases} \ddot{\vec{W}}_L = -\gamma_{11}\vec{W}_L + \gamma_{12}\vec{E}_L \\ \vec{P}_L = -\dfrac{1}{4\pi}\vec{E}_L = \gamma_{12}\vec{W}_L + \gamma_{22}\vec{E}_L \quad \Rightarrow \quad \vec{E}_L = \dfrac{\gamma_{12}}{-\frac{1}{4\pi} - \gamma_{22}}\vec{W}_L \end{cases}$$

$$\Rightarrow \ddot{\vec{W}}_L = -\left(\gamma_{11} + \frac{\gamma_{12}^2}{\frac{1}{4\pi} + \gamma_{22}}\right)\vec{W}_L$$

$$\Rightarrow \boxed{\omega_L^2 = \gamma_{11} + \frac{4\pi\gamma_{12}^2}{1 + 4\pi\gamma_{22}}} \quad \Rightarrow \boxed{\frac{\omega_L^2}{\omega_0^2} = \frac{\varepsilon_0}{\varepsilon_\infty}}$$

$$\boxed{\varepsilon(\omega) = \frac{\omega^2 - \omega_L^2}{\omega^2 - \omega_0^2}\,\varepsilon_\infty}$$

ω_L - mode is an intrinsic mode without external E-field
$\Rightarrow$ ω_L is the zero point of ε.
ω_0 is the frequence of the elastic restoring force, $\longrightarrow$ pole of dielectric function.

- transverse wave — light is also transverse
- thus they can hybridize.

① $\begin{cases} \nabla \times \vec{E}_T = -\frac{1}{c}\frac{\partial}{\partial t}\vec{B} \\ \nabla \times \vec{H} = \frac{1}{c}\frac{\partial \vec{D}_T}{\partial t} = \frac{1}{c}\frac{\partial}{\partial t}(\vec{E}_T + 4\pi\vec{P}_T) \end{cases}$

non-magnetic material

no free current

$\Rightarrow \vec{B} = \vec{H} + 4\pi\vec{M}$

$= \vec{H}$

② $\begin{cases} \vec{P}_T = \gamma_{12}\vec{W}_T + \gamma_{22}\vec{E}_T \\ \ddot{\vec{W}}_T + \omega_0^2\vec{W}_T - \gamma_{12}\vec{E}_T = 0 \end{cases}$

$$① \Rightarrow \quad \nabla \times \nabla \times \vec{E}_T = -\frac{1}{c}\frac{\partial}{\partial t}\nabla \times \vec{B} = -\frac{1}{c^2}\frac{\partial^2}{\partial t^2}(\vec{E}_T + 4\pi\vec{P}_T)$$

$$\Rightarrow +\nabla^2\vec{E}_T = \frac{1}{c^2}\frac{\partial^2}{\partial t^2}(\vec{E}_T + 4\pi\vec{P}_T) \Rightarrow \left(k^2 - \frac{\omega^2}{c^2}\right)\vec{E}_T(k,\omega) = \frac{4\pi\omega^2}{c^2}\vec{P}_T(k,\omega)$$

$$\Rightarrow \left(k^2 - \frac{\omega^2}{c^2}\right)\vec{E}_T(k,\omega) = \frac{4\pi\omega^2}{c^2}(\gamma_{12}\vec{W}_T + \gamma_{22}\vec{E}_T)$$

$$\begin{cases} \left[k^2 - \frac{\omega^2}{c^2}(1 + 4\pi\gamma_{22})\right]\vec{E}_T - \frac{4\pi\omega^2\gamma_{12}}{c^2}\vec{W}_T = 0 \\ ② \rightarrow \quad \gamma_{12}\vec{E}_T + (\omega^2 - \omega_0^2)\vec{W}_T = 0 \end{cases}$$

$$\Rightarrow \det\begin{bmatrix} k^2 - \frac{\omega^2}{c^2}\varepsilon_\infty, & \frac{4\pi\omega^2}{c^2}\gamma_{12} \\ \gamma_{12} & \omega^2 - \omega_0^2 \end{bmatrix} = 0 \Rightarrow \left[k^2 - \frac{\omega^2}{c^2}\varepsilon_\infty\right](\omega^2 - \omega_0^2) - \frac{4\pi\omega^2}{c^2}\gamma_{12}^2 = 0$$

$$(\omega^2-\omega_0^2)(\varepsilon_\infty\omega^2-c^2k^2) + 4\pi\omega^2 \frac{\varepsilon_0-\varepsilon_\infty}{4\pi}\omega_0^2 = 0$$

$$\varepsilon_\infty\omega^4 - (k^2c^2+\omega_0^2\varepsilon_0)\omega^2 + \omega_0^2(kc)^2 = 0$$

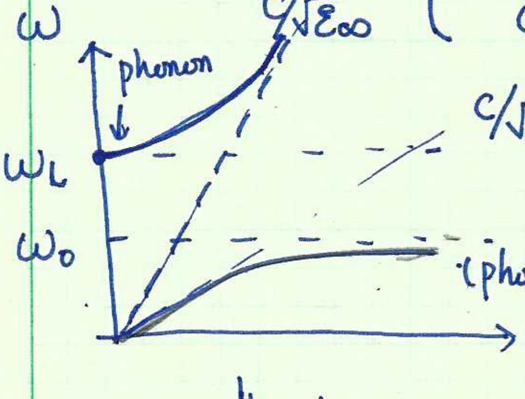
$$\varepsilon_\infty\omega^4 + \left[(kc)^2+\omega_L^2\varepsilon_\infty\right]\omega^2 + \omega_0^2(kc)^2 = 0$$

$$\omega_\pm^2 = \frac{1}{2}\left[\omega_L^2 + \frac{(kc)^2}{\varepsilon_\infty} \pm \left\{\omega_L^4 + \left(\frac{k^2c^2}{\varepsilon_\infty}\right)^2 + \frac{2(kc)^2}{\varepsilon_\infty}(\omega_L^2-2\omega_0^2)\right]^{1/2}\right\}$$

① $k\to 0$, $\omega_\pm^2 = \begin{cases}\omega_L^2 \\ \dfrac{\omega_0^2(kc)^2}{\varepsilon_\infty\omega_L^2} = \dfrac{(kc)^2}{\varepsilon_0}\end{cases}$

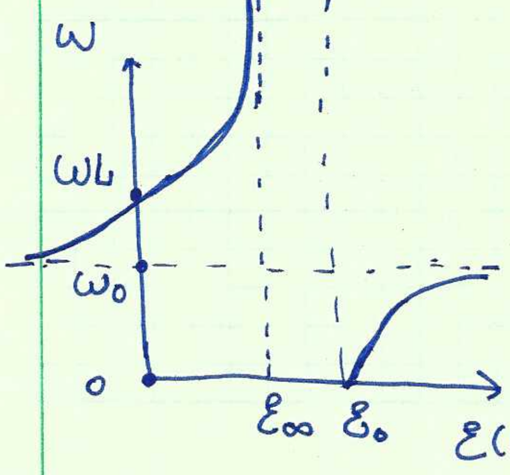
use $\boxed{\omega_+^2\omega_-^2 = \dfrac{\omega_0^2(kc)^2}{\varepsilon_\infty}}$

② $k\to\infty$ $\omega_\pm^2 = \begin{cases}\dfrac{k^2c^2}{\varepsilon_\infty} & \text{(photon)} \\ \omega_0^2\end{cases}$



in this regim $\varepsilon(\omega) < 0$

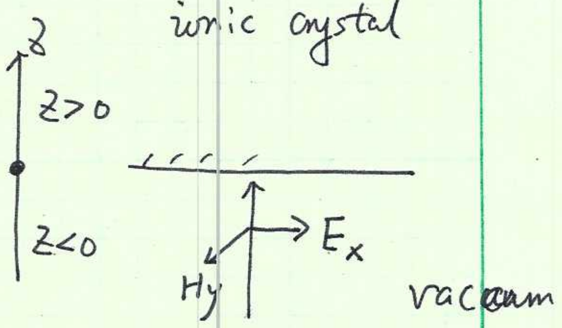
no-propagating mode



$$\boxed{\frac{k^2c^2}{\omega^2} = \varepsilon_\infty\frac{\omega_L^2-\omega^2}{\omega_0^2-\omega^2} = \varepsilon(\omega)}$$

Please prove!

Reflectivity:



at $z<0$: incident $E_{i,x} = H_{i,y} = A\, e^{i\omega\left(\frac{z}{c} - t\right)}$

reflected $E_{r,x} = - H_{r,y}$. (

at $z>0$ $\quad E_{t,x} = E_{x\,t,}\, e^{i(kz-\omega t)}$ where $k = \frac{\omega}{c}\sqrt{\varepsilon(\omega)}$

$$+\frac{\omega}{c} H_{t,y} = k E_{t,x} = \frac{\omega}{c}\sqrt{\varepsilon(\omega)}\, E_{tx} \Rightarrow H_{t,y} = \sqrt{\varepsilon(\omega)}\, E_{x\,t,}$$

Continuity equation:

$$\begin{cases} E_{i,x} + E_{r,x} = E_{t,x} \\ H_{i,y} + H_{r,y} = H_{t,y} \Rightarrow E_{ix} - E_{r,x} = \sqrt{\varepsilon(\omega)}\, E_{t,x} \end{cases}$$

$$\Rightarrow \frac{E_{i,x} + E_{r,x}}{E_{ix} - E_{r,x}} = \frac{1}{\sqrt{\varepsilon(\omega)}} \Rightarrow \frac{E_{r,x}}{E_{ix}} = \frac{1-\sqrt{\varepsilon(\omega)}}{1+\sqrt{\varepsilon(\omega)}}$$

$$R(\omega) = \left|\frac{E_{r,x}}{E_{i,x}}\right|^2 = \left|\frac{\sqrt{\varepsilon(\omega)} - 1}{\sqrt{\varepsilon(\omega)} + 1}\right|^2$$

if $\varepsilon(\omega) > 1$

$\Rightarrow \frac{E_{r,x}}{E_{i,x}} < 0$

"π"-phase shift