Lecture 12: Surface phonon polaritons and exciton polaritons

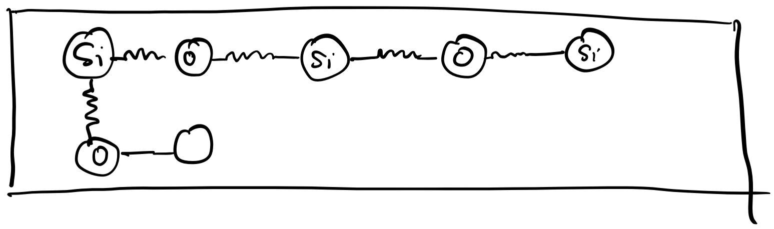
Learning objective:

1. Infrared active phonons
2. LST relationship
3. Infrared reflectivity and absorption in polar materials
4. Phonon polaritons and surface phonon polaritons

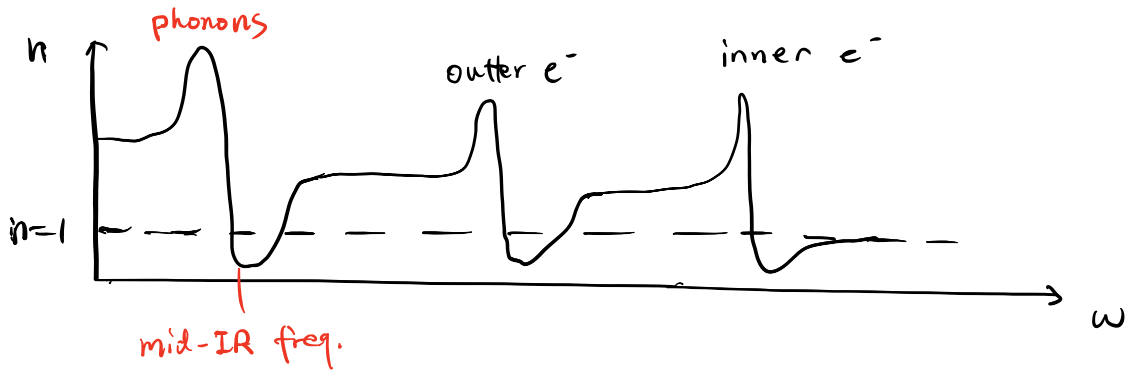
Plasmons: free electron oscillation in metals

Phonons: lattice vibration in polar dielectrics

Polar Dielectric (SiO_2)

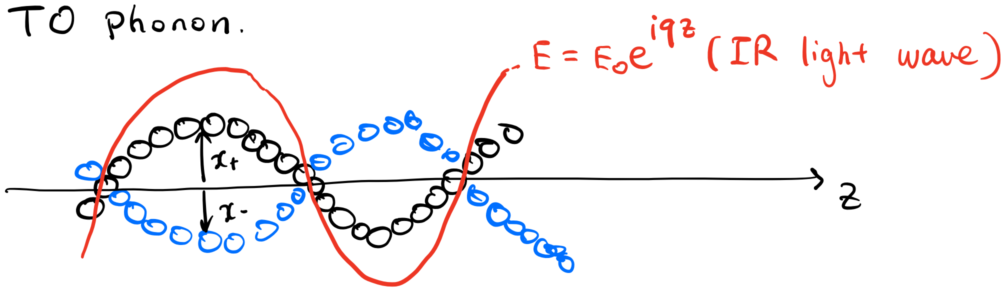


Recap: hamonic oscillator model



1. Classic oscillator model of mid-IR phonons

TO phonon.



① x_+, x_-, displacement of positive and negative ions.

② When $\omega \sim \omega_{op}$ (on resonance), wave vector of photon and phonon are same, (driving force exerted by the light on the positive and negative ions is in phase with lattice vibration)

$\Rightarrow$ Strong interaction between TO phonon and light wave when wave vector and frequencies match.

Equations of motion of positive and negative ions:

positive ion

$$m_+ \frac{d^2 x_+}{dt^2} = -k(x_+ - x_-) + qE(t) \quad ①$$

negative ion

$$m_- \frac{d^2 x_-}{dt^2} = -k(x_- - x_+) - qE(t) \quad ②$$

where m_+ (m_-) are the masses of two ions, k is the restoring constant.

Divide ① by m_+, divide ② by m_-, then substracting.

$$\frac{d}{dt^2}(x_+ - x_-) = -\frac{k}{\mu}(x_+ - x_-) + \frac{q}{\mu}E(t) \qquad ③$$

where $\frac{1}{\mu} = \frac{1}{m_+} + \frac{1}{m_-}$ is "reduced" mass

Let $x = x_+ - x_-$, and $\Omega_{TO}^2 = \frac{k}{\mu}$, ③ can be written as:

$$\frac{d^2x}{dt^2} + \Omega_{TO}^2 x = \frac{q}{\mu}E(t).$$

Introduce a phenomenological damping rate γ.

$$\boxed{\frac{d^2x}{dt^2} + \gamma\frac{dx}{dt} + \Omega_{TO}^2 x = \frac{q}{\mu}E(t)}$$

identical to harmonic oscillator model.

$$\Rightarrow \epsilon_r(\omega) = 1 + \chi + \frac{Nq^2}{\epsilon_0\mu}\frac{1}{(\Omega_{TO}^2 - \omega^2 - i\gamma\omega)}$$

Introducing static and high frequency dielectric constant.
ϵ_{st} and ϵ_∞.

$$\epsilon_{st} = \epsilon_r(0) = 1 + \chi + \frac{Nq^2}{\epsilon_0 \mu \Omega^2_{TO}}$$

$$\epsilon_\infty = \epsilon_r(\infty) = 1 + \chi$$

So $$\boxed{\epsilon_r(\omega) = \epsilon_\infty + (\epsilon_{st} - \epsilon_\infty) \frac{\Omega^2_{TO}}{(\Omega^2_{TO} - \omega^2 - i\gamma\omega)}} \quad (4)$$

2. Lyddane - Sach - Teller (LST) relationship

Assume zero damping $(\gamma = 0)$, (4) becomes

$$\epsilon_r(\omega) = \epsilon(\infty) + (\epsilon_{st} - \epsilon_\infty) \frac{\Omega^2_{TO}}{(\Omega^2_{TO} - \omega^2)}$$

At $\omega = \sqrt{\frac{\epsilon_{st}}{\epsilon_\infty}} \Omega_{TO}$, $\epsilon_r(\omega) = 0$.

In metals (free electron gas), $\epsilon_r(\omega) = 0$ means there is a longitudinal wave.

$\left(\nabla \equiv \hat{x}\frac{\partial}{\partial x} + \hat{y}\frac{\partial}{\partial y} + \hat{z}\frac{\partial}{\partial z}\right)$

$\nabla(e^{i(\vec{k}\cdot\vec{r} - \omega t)}) = i k e^{i(\vec{k}\cdot\vec{r} - \omega t)}$

why? $\nabla \cdot \vec{D} = \nabla \cdot (\epsilon_r \cdot \epsilon_0 \vec{E}) = 0$

$i\vec{k} \cdot \vec{E} = 0$

if $\epsilon_r \neq 0$. $\nabla \cdot \vec{E} = 0$, transverse.

if $\epsilon_r = 0$. $\nabla \cdot \vec{E} \neq 0$, longitudinal.

So, in polar dielectrics, longitudinal phonons (LO) phonons exist at $\omega = \sqrt{\frac{\epsilon_{st}}{\epsilon_\infty}}\,\Omega_{TO}$.

$$\Rightarrow \boxed{\frac{\Omega_{LO}^2}{\Omega_{TO}^2} = \frac{\epsilon_{st}}{\epsilon_\infty}} \quad \text{(LST Relationship)}$$

Comments:

① For non-polar dielectrics, $\Omega_{LO}^2/\Omega_{TO}^2 = 1$ (degenerate)

$\epsilon_{st} = \epsilon_\infty$, $\epsilon_r(\omega) = \epsilon_\infty$.

not $\omega = \infty$.
just ω is far away enough to the resonance.

② For polar dielectrics. $\frac{\Omega_{LO}^2}{\Omega_{TO}^2} = \frac{\epsilon_{st}}{\epsilon_\infty} > 1$

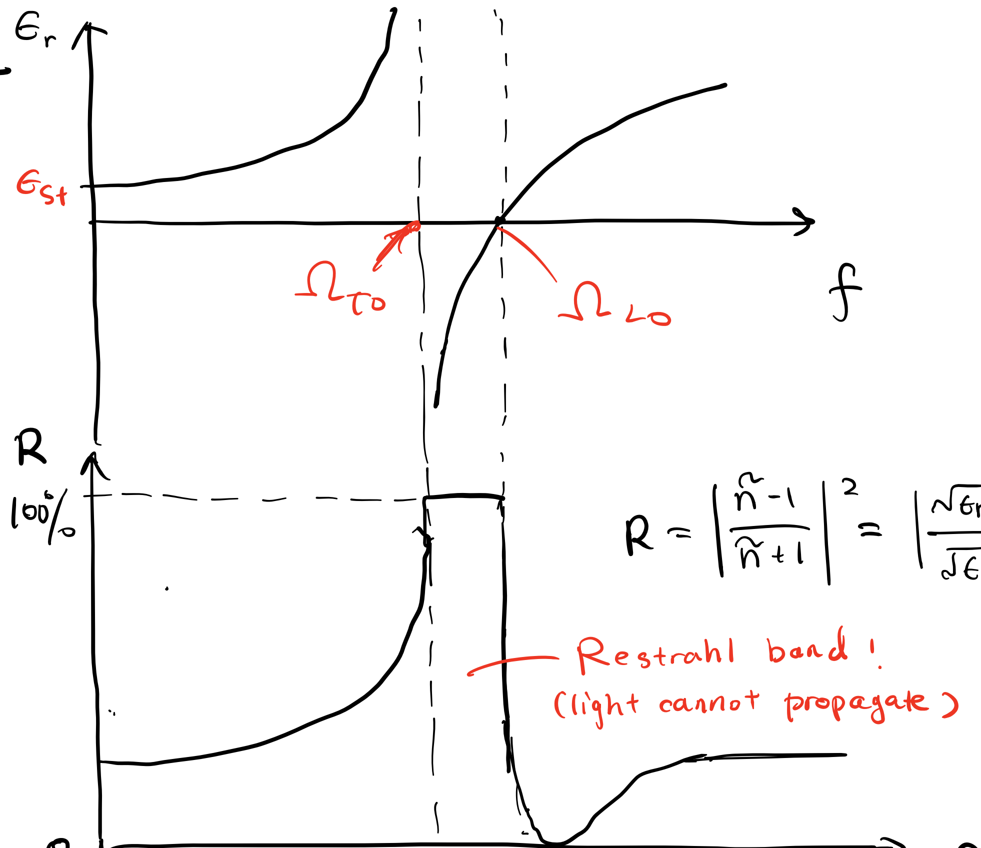
e.g. BN. $\Omega_{LO}/\Omega_{TO} = 1.24$

MgO. $\Omega_{LO}/\Omega_{TO} = 1.81$ ..

③ Ω_{TO}, Ω_{TO} are usually in mid-IR and THz

3. Infrared reflectivity and absorption in polar materials

Reflection



$$R = \left|\frac{\tilde{n}-1}{\tilde{n}+1}\right|^2 = \left|\frac{\sqrt{\epsilon_r}-1}{\sqrt{\epsilon_r}+1}\right|^2$$

(Ignoring the damping)

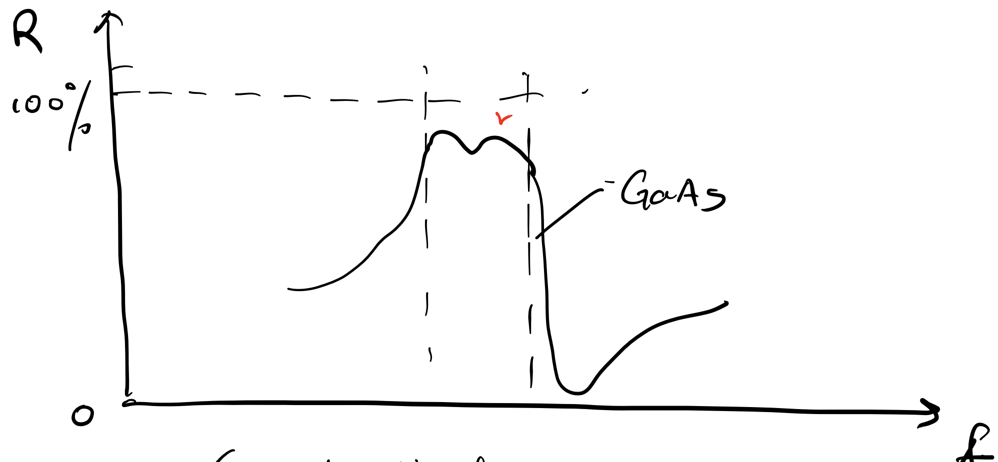
Comments:

① At low frequency, $R = \dfrac{(\sqrt{\epsilon_{st}}-1)^2}{(\sqrt{\epsilon_{st}}+1)^2}$

② $\omega \to \omega_{TO}$, $\epsilon_r \to \infty$. $R \to 1$

③ $\omega_{TO} < \omega < \omega_{LO}$, $\sqrt{\epsilon_r}$ is imaginary, $R=1$, Reststrahl band

④ $\omega > \omega_{LO}$, R drops rapidly to zero and increases gradually towards $\left(\dfrac{\sqrt{\epsilon_\infty}-1}{\sqrt{\epsilon_\infty}+1}\right)^2$



(consider the damping)

Comments

① In real polar dielectrics, when damping is considered, $R < 100\%$.

② When $\omega > \omega_{LO}$, R cannot reach zero.

③ $\gamma = 10^{11} \sim 10^{12}\ s^{-1}$, optical phonons have a lifetime about $1 \sim 10$ ps.

Absorption:

At ω_{TO}, $\epsilon_r(TO) = \epsilon_\infty + i(\epsilon_{st} - \epsilon_\infty)\dfrac{\Omega_{TO}}{\gamma} = \epsilon_1 + \epsilon_2$

$$k = \frac{1}{\sqrt{2}}\left(-\epsilon_1 + \sqrt{\epsilon_1^2 + \epsilon_2^2}\right)^{1/2}$$

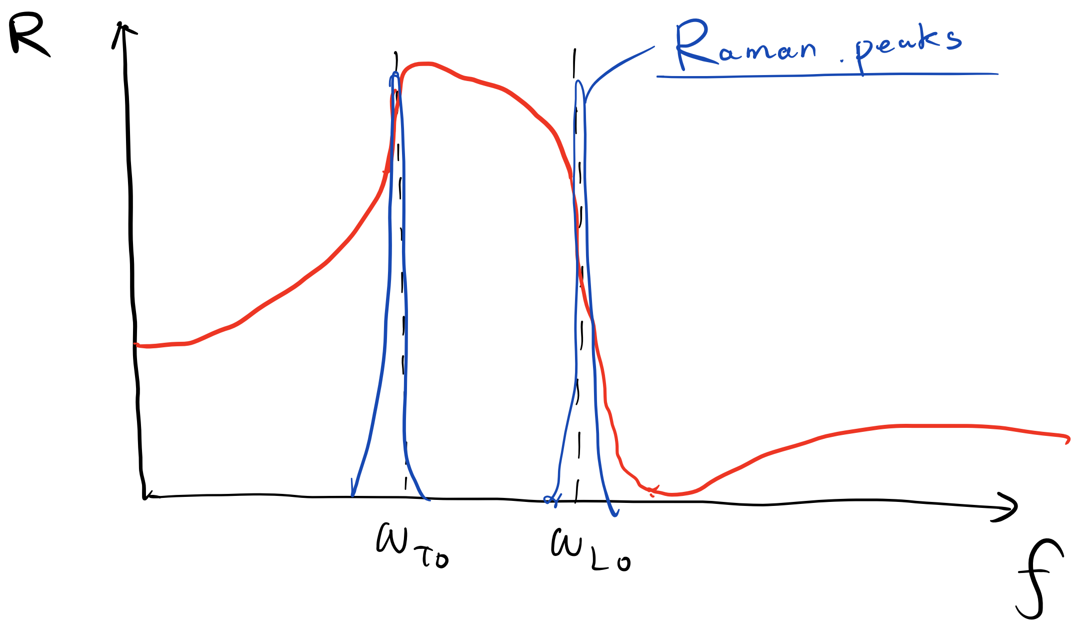
$$\alpha = \frac{4\pi k}{\lambda} \simeq 10^6 \sim 10^7\ m^{-1}$$

Comments:

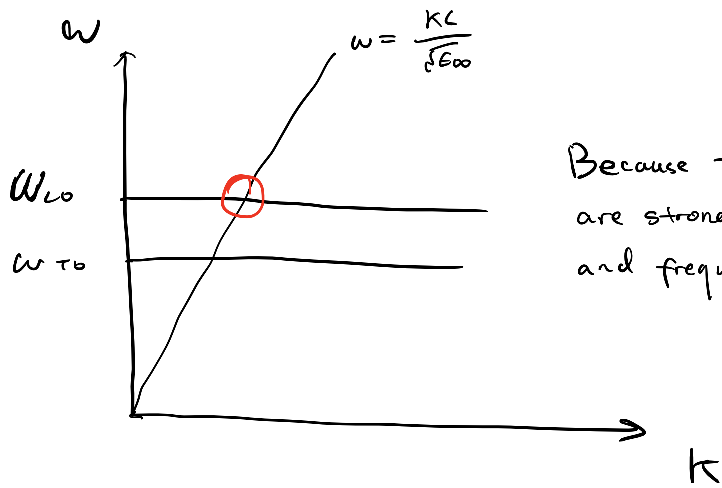
① Because of the very large α at ω_{TO}, absorption has to be measured with thin-film ($\sim 1 \mu m$ thick)

No light will be transmitted with thick samples.

② For thick samples, use Reflection spectra and Raman to determine ω_{TO}, ω_{LO}



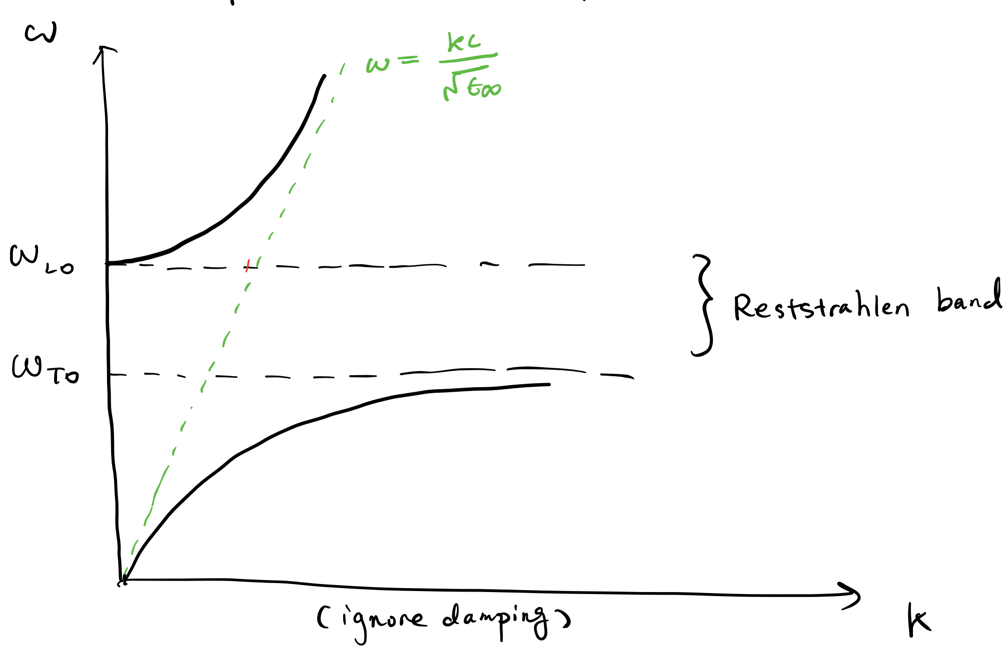
4. Phonon polaritons and surface phonon polaritons



Because TO-phonons and photons are strongly coupled when wavevector and frequency matches.

$\Rightarrow$ Anti-crossing

Dispersion of phonon-polariton



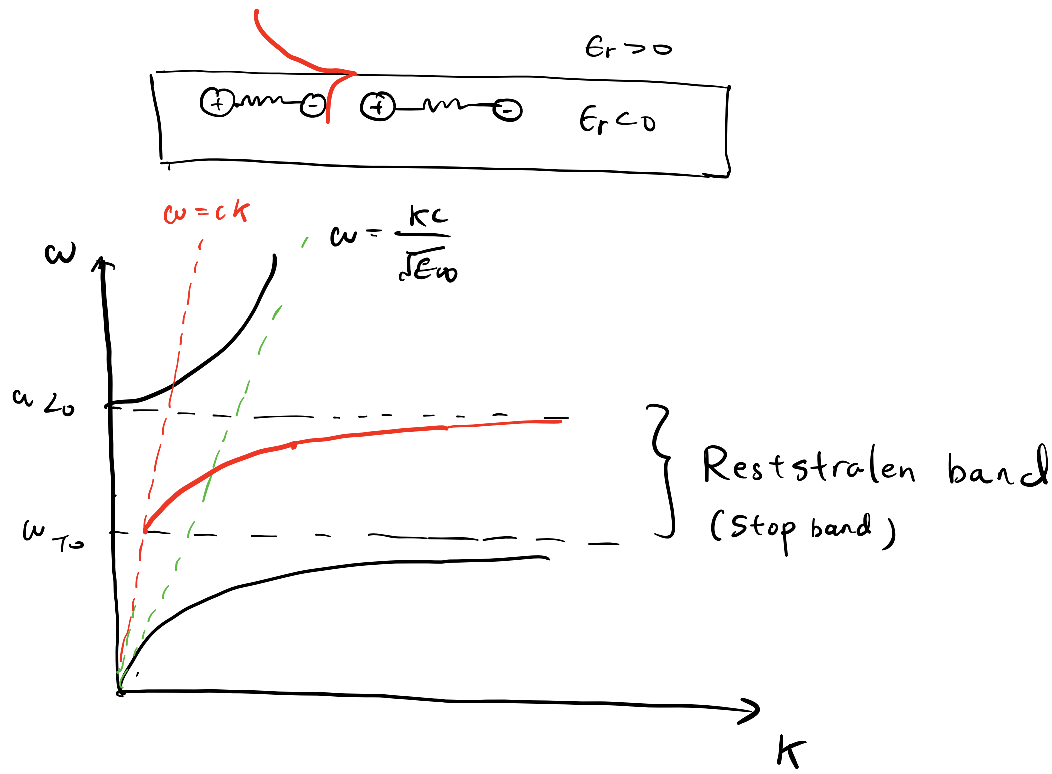
① At low frequencies, $\epsilon_r(\omega) = \epsilon_{st}$. dispersion $\omega = \frac{cq}{\sqrt{\epsilon_{st}}}$

② $\omega \to \omega_{TO}$ $\epsilon_r(\omega) \uparrow$. $v_g \to 0$ at ω_{TO}.

③ $\omega_{TO} < \omega < \omega_{LO}$, Restrahlen band. $\epsilon_r < 0$. No mode can propagate.

④ $\omega > \omega_{LO}$, propagating mode $v_g = \frac{c}{\sqrt{\epsilon_\infty}}$.

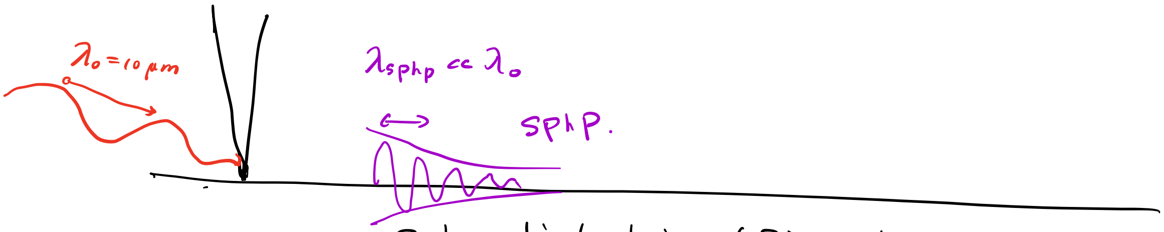
Surface phonon polaritons (SPhP)



① SphP occurs only at the interface between polar dielectric and air (not in the bulk)

② Because in polar dielectrics, γ is smaller compare to that of metal, longer propagation distance experiments:

SNOM



polar dielectric (SiC, h-BN, SiO_2...)