Handout on Optical Properties of (Insulating) Solids

CHY

Ref: Kittel, Ashcroft and Mermin

Refer to Chapter 10 of Kittel. The main purpose of this handout is to help explain the origin of Fig. 11: the dispersion relation of photon-phonon-coupled polariton, which describes how light propagates in (insulating) solids. By definition, the propagating wave must be “transverse modes.” To be more specific, we call them “phonon-photon coupled polariton” modes. That is, the nature of the energy is a combination of transverse optical phonons and the transverse EM wave. In other words, the photons (TEM wave) interact with TO phonons, and the two dispersion relations couple and split into two branches near the region where interaction occurs. (Near the TO phonon energy, and the low-k region.) Although by definition a photon does not couple with LO phonons, we will show in the following that: (1) In between the LO phonon energy and TO phonon energy, the solid is actually opaque for the TEM wave. This property is clearly observable by using infrared transmission spectroscopy. (2) In other frequencies, light can pass through the solid all right. The “pass band” starts from LO phonon energy and up; and from TO phonon energy and down to zero.

We start with the Clausius-Mossotti relation (page 390, edition 7):

$$\frac{\epsilon - 1}{\epsilon + 2} = \frac{N\alpha}{3\varepsilon_o}$$  \hspace{1cm} (1)

This relation is important in that it links the “Macroscopic” property, $\epsilon$, with the “Microscopic” property, atomic polarizability $\alpha$. The total polarizability has three contributions: (1) dipole; (2) ionic (phonon); and (3) electronic. The end result is like that shown in Fig. 8, p. 391, 7th Ed. of Kittel. So, ignoring the dipole contribution for most solids,

$$\alpha = \alpha_{\text{atomic}}^\oplus + \alpha_{\text{atomic}}^\ominus + \frac{e^2}{M(\bar{\omega}^2 - \omega^2)} + \frac{e^2}{M(\bar{\omega}^2 - \omega^2)}$$  \hspace{1cm} (2)

where $\alpha_{\text{atomic}}^\oplus$ and $\alpha_{\text{atomic}}^\ominus$ are atomic polarizability from deformation of electron clouds of ions, and $\frac{e^2}{M(\bar{\omega}^2 - \omega^2)}$ is contribution from phonons, $\bar{\omega} = \sqrt{k/M}$, and $M^{-1} = M_\oplus^{-1} + M_\ominus^{-1}$, the ions’ reduced mass.

Taking the “low frequency limit,” i.e., $\omega \ll \bar{\omega}$,

$$\lim_{\omega \to 0} \frac{\epsilon - 1}{\epsilon + 2} = \frac{\epsilon_0 - 1}{\epsilon_0 + 2} = \frac{N}{3\varepsilon_o}(\alpha_{\text{atomic}} + \frac{e^2}{M\bar{\omega}^2}).$$  \hspace{1cm} (3)

Taking the “high frequency limit,” i.e., $\omega \gg \bar{\omega}$,

$$\lim_{\omega \to \infty} \frac{\epsilon - 1}{\epsilon + 2} = \frac{\epsilon_\infty - 1}{\epsilon_\infty + 2} = \frac{N}{3\varepsilon_o}(\alpha_{\text{atomic}}).$$  \hspace{1cm} (4)
So, combining these two results in equation (3) and (4), equation (1) can be expressed as:

\[
\frac{\epsilon(\omega) - 1}{\epsilon(\omega) + 2} = \frac{\epsilon_\infty - 1}{\epsilon_\infty + 2} + \frac{1}{1 - (\tilde{\omega}^2)^2} \left( \frac{\epsilon_o - 1}{\epsilon_o + 2} - \frac{\epsilon_\infty - 1}{\epsilon_\infty + 2} \right). \tag{5}
\]

Now, solve for the only variable \(\epsilon(\omega)\) in equation (5):

\[
\epsilon(\omega) = \epsilon_\infty + \frac{\epsilon_\infty - \epsilon_o}{(\tilde{\omega}^2)^2 - 1}, \tag{6}
\]

where

\[
\omega_T^2 = \tilde{\omega}^2 \left( \frac{\epsilon_\infty + 2}{\epsilon_o + 2} \right) = \tilde{\omega}^2 \left( 1 - \frac{\epsilon_o - \epsilon_\infty}{\epsilon_o + 2} \right). \tag{7}
\]

The above result already implies that \(\omega_T < \tilde{\omega}\), since \(\epsilon_o > \epsilon_\infty\). Because at lower frequencies, all external stimulations can be responded by internal excitations, the polarizability (and thus the dielectric constant) is always larger at lower frequencies.

With this functional form reached, we can plot \(\epsilon(\omega)\) as a function of \(\omega\). Next, we further argue and learn about the two frequency limits of interest: \(\omega_{LO}\) and \(\omega_{TO}\).

First, we take the long wavelength limit (k approaches zero). This is coming from that the photon wavelength at the TO phonon energy is much longer than that of the lattice constant. So, according to the dispersion relation, near the region where the phonon-photon polariton occurs, it is true that the value of wave vector k is small.

For long wavelength (IR and FIR), small wavevector k, TEM wave traveling in an ionic insulator,

\[
D = \epsilon E = E + 4\pi P. \quad \text{(CGS)} \tag{8}
\]

Since there is no free charge, \(\nabla \cdot D = 0\). Considering a propagating wave, we are looking for solutions like:

\[
D \rightarrow D_0 e^{ik \cdot r}, \\
E \rightarrow E_0 e^{ik \cdot r}, \text{ and} \\
P \rightarrow P_0 e^{ik \cdot r}. \tag{9}
\]

We further assume that \(D \parallel E\), i.e., we are dealing with an isotropic medium. So, \(\nabla \cdot D = 0\) implies that \(k \cdot D = 0\), i.e., either \(D = 0\), or \(D, E, P\) must be all perpendicular to \(k\).

On the other hand, \(\nabla \times E = 0\), i.e., \(k \times E = 0\), which requires that either \(E = 0\) or \(E, D, P\) are all parallel to \(k\).
Combining these two observations, we conclude that the meaningful solution for a propagating transverse mode \((P \perp k)\) is that \(E = 0\), and this perfect screening leads to \(\epsilon = \infty\). \((D = E + 4\pi P = 4\pi P\) and \(D, P \perp k)\).

Use \(\epsilon = \infty\),

\[
\epsilon = \infty = \epsilon_\infty + \frac{\epsilon_\infty - \epsilon_o}{(\frac{\omega}{\omega_T})^2 - 1},
\]

we solve for \(\omega = \omega_T\): by definition, this \(\omega_T\) is the frequency of \(k \to 0\) transverse optical mode, i.e., TO phonon frequency at \(k \to 0\).

For “longitudinal modes,” \(p \parallel k\), there is no propagating mode, and \(\epsilon = 0\). If considering the movement of atoms, the \(P\) is parallel to \(k\). So, \(D = 0\), and thus \(E = -4\pi P\).

When \(\epsilon = 0\), we can solve for \(\omega = \omega_L\):

\[
0 = \epsilon_\infty + \frac{\epsilon_\infty - \epsilon_o}{(\frac{\omega}{\omega_T})^2 - 1},
\]

and the solution of \(\omega\) (and we define \(\omega = \omega_L\)) gives the relation between \(\omega_T\) and \(\omega_L\): (the Lyddane-Sachs-Teller relation)

\[
\omega_L = \omega_T \sqrt{\frac{\epsilon_o}{\epsilon_\infty}}.
\]

The solved \(\omega_L\) is the frequency of \(k \to 0\) longitudinal optical mode (which doesn’t propagate).

With the dielectric function derived as a function of frequency, we can then use it in \(\omega = \frac{kc}{\sqrt{\epsilon}}\) and plot \(\omega\) versus \(k\). The result is shown in Fig. 11, chapter 10, Kittel (7th edition)).

Note that in vacuum, photon travels with \(\omega = \frac{kc}{\sqrt{\epsilon}},\) and \(\epsilon = 1\).

At the low frequency limit \(\omega \ll \omega_{TO}, \omega = \frac{kc}{\sqrt{\epsilon_o}}\).

At the high frequency limit \(\omega_{LO} \ll \omega, \omega = \frac{kc}{\sqrt{\epsilon_\infty}}\).

At the intermediate frequencies, either \(\omega \leq \omega_{TO}\) or \(\omega_{LO} \leq \omega\), the TEM wave travels in a combined ”polariton” mode.

For \(\omega_{TO} \leq \omega \leq \omega_{LO}\), light cannot propagate through the insulator. There is simply not such transverse mode allowed. As a result, the reflectivity within this frequency range becomes one.

The reflectivity \(R\) is defined to be (as can be found in many standard E&M textbooks): \(|\frac{1 - \sqrt{\epsilon}}{1 + \sqrt{\epsilon}}|^2\), and \(\epsilon\) is a function of frequency. Plugging the derived \(\epsilon\) into this formula, we can then obtain the frequency dependence of \(R\), due to polaritons.