Polaritons and Plasmons

by Dr. Colton, Physics 581 (last updated: Fall 2020)

Introduction

When photons enter a material, if their frequencies match up against other excitations which can occur (e.g. the UV resonance of bound electrons, the IR resonance of ions, and the plasma resonance of conductors), then this coupling causes the usual photon dispersion curves to be modified. At these frequencies the photons not only *can* but necessarily *must* excite these other types of oscillations as the electromagnetic wave enters the material.

The regular photon dispersion, ω vs k, is simply a straight line: $\omega = vk$, where v is the speed of light in the material, $v = c/n$. (Here n is the index of refraction.) It can be summarized by this important equation:

$$\frac{\omega}{k} = \frac{c}{n}$$

However, as was discussed in the *Lorentz oscillator model of the dielectric function* handout, near these resonances n itself is a function of ω. Therefore that simple looking equation, which appears to describe a linear relationship between ω and k actually turns into a much more complicated dispersion relation.

We can make that $\omega(k)$ dependence more explicit by squaring both sides and using $n^2 = \epsilon_r(\omega)$.

$$\frac{\omega^2}{k^2} = \frac{c^2}{\epsilon_r(\omega)}$$

$$\frac{c^2k^2}{\omega^2} = \epsilon_r(\omega) \tag{1}$$

The functional dependence of $\epsilon_r(\omega)$ was given for several situations in the *Lorentz oscillator* handout. We will use that dependence to obtain the actual, complicated dispersion relations for the IR resonance of ions and the plasma resonance of conductors.

Phonon Polaritons

Polaritons (or more specifically, *phonon polaritons*) are quasiparticles which result from the coupling of photons to the optical phonon modes in the material. From the Lorentz model handout we had the following equation for the IR resonance due to oscillating ions (assuming no damping, for simplicity):

$$\epsilon_r(\omega) = \epsilon_r(\infty) + \frac{\epsilon_r(0) - \epsilon_r(\infty)}{1 - \frac{\omega^2}{\omega_T^2}} \tag{2}$$

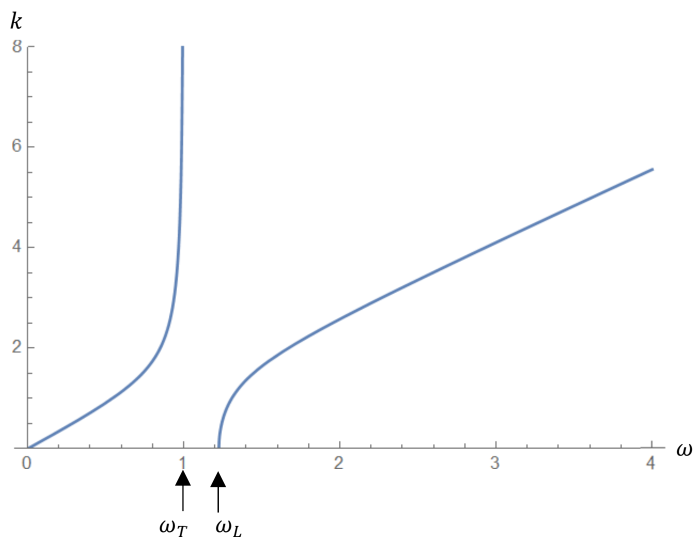
Plugging Eq. 2 into Eq. 1 yields the following polariton dispersion relation:

$$\frac{c^2k^2}{\omega^2} = \epsilon_r(\infty) + \frac{\epsilon_r(0) - \epsilon_r(\infty)}{1 - \frac{\omega^2}{\omega_T^2}} \tag{3}$$

Eq. 3 cannot easily be solved for $\omega(k)$… but using a trick that we've used before, we *can* solve it for $k(\omega)$:

$$k = \frac{\omega}{c}\sqrt{\epsilon_r(\infty) + \frac{\epsilon_r(0) - \epsilon_r(\infty)}{1 - \frac{\omega^2}{\omega_T^2}}} \tag{4}$$

I'll plot that equation for $c = 1$, $\epsilon_r(\infty) = 2$, $\epsilon_r(0) = 3$, and $\omega_T = 1$. Incidentally, those are the same $\epsilon_r(\infty)$ and $\epsilon_r(0)$ values used in Kittel, Fig. 14.13a which plot was given in the *Lorentz oscillator* handout. It will also become important to recognize that for those values, the LST relation predicts $\omega_L = 1.22474$.

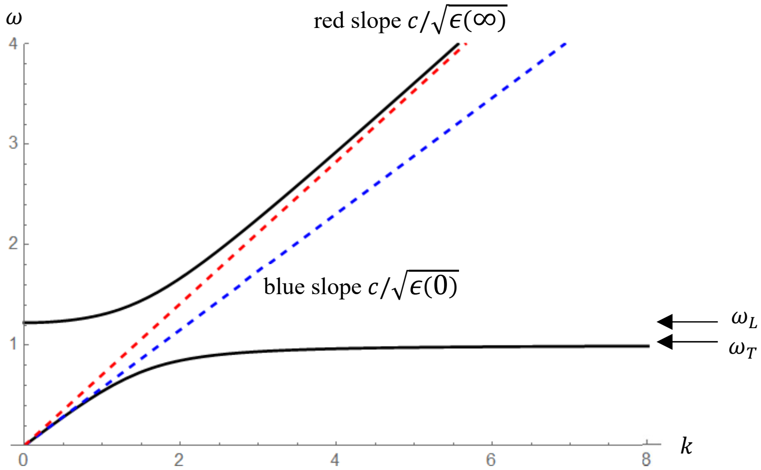


Colton plot 1. Plot of polariton dispersion, namely $k(\omega)$ for the coupling between EM waves and the TO phonon modes, with $c = 1$, $\epsilon_r(\infty) = 2$, $\epsilon_r(0) = 3$, and $\omega_T = 1$.

At very low frequencies the wave vector starts out obeying the normal photon behavior of $\omega = ck/n$, with n equal to the low frequency value $\sqrt{\epsilon(0)}$. At very high frequencies the wave vector again obeys the normal photon behavior of $\omega = ck/n$, now with n equal to the high frequency value $\sqrt{\epsilon(\infty)}$.

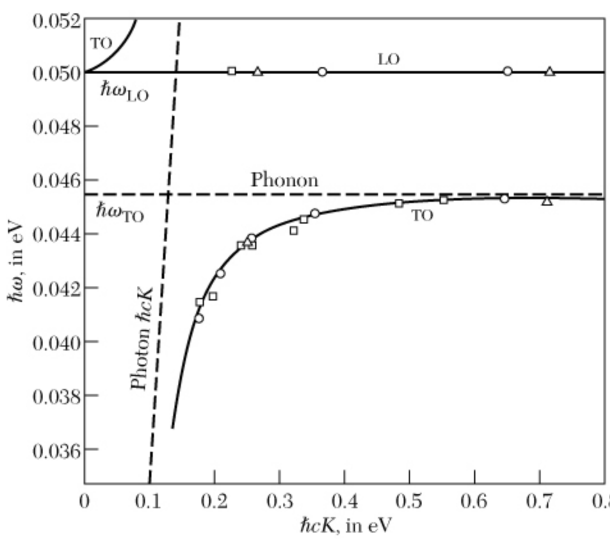
At frequencies between those two cases, namely around the TO phonon frequency $\omega = \omega_T$, the regular photon dispersion goes wacky as photons start automatically creating TO phonons! In fact, in this region, you can't even talk about photons and TO phonons as being separate entities. They are intimately connected, and therefore in this region we must only consider the joint polariton quasiparticle. Also, note that for frequencies between ω_T and ω_L no polaritons can propogate, giving rise to perfect reflectivity as depicted in Yu & Cardona Fig. 6.31(b) in the *Lorentz model* handout.

I'll use some Mathematica trickery (not shown) to reverse the axes so I can plot $\omega(k)$.



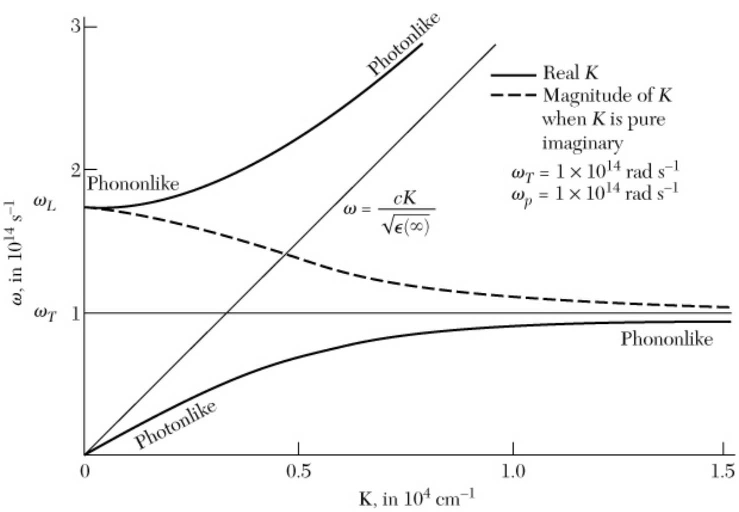
Colton plot 2. Plot of polariton dispersion, $\omega(k)$ for the coupling between EM waves and the TO phonon modes, with = 1, $\epsilon_r(\infty) = 2$, $\epsilon_r(0) = 3$, and $\omega_T = 1$.

Compare that plot to Kittel Fig. 14.11, shown next, which additionally makes the point that these polaritons are the coupling between photons and the *TO phonons*; the LO phonons don't couple to the EM wave and therefore exist undisturbed.



Kittel Fig. 14.11. A plot of the observed energies and wavevectors of the polaritons and of the LO phonons in GaP. The theoretical dispersion curves are shown by the solid lines. The dispersion curves for the uncoupled phonons and photons are shown by the short, dashed lines. (Apologies for the strange x-axis; it's essentially the wave vector k, but written in units of eV.)

Kittel Fig. 14.12 gives another plot of $\omega(k)$ for the polariton mode, but additionally plots the imaginary component of k in the region of no propagation (i.e. between ω_T and ω_L) and makes some interesting comments about absorption in the figure caption.



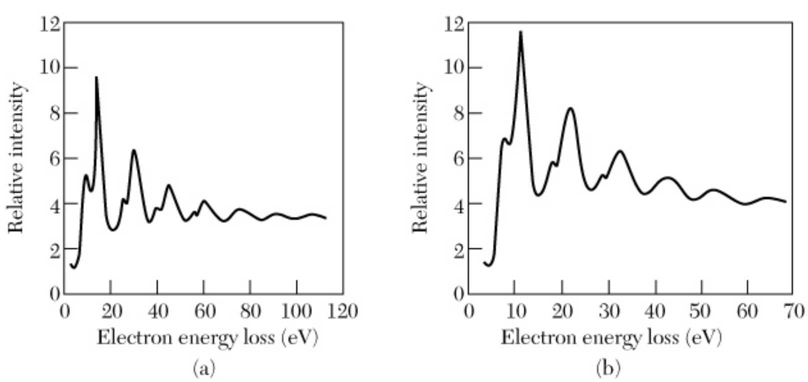
Kittel Fig. 14.12. Coupled modes of photons and TO phonons in an ionic crystal. The fine horizontal line represents oscillators of frequency ω_T in the absence of coupling to the electromagnetic field and the diagonal fine line corresponds to electromagnetic waves in the crystal, uncoupled to the lattice oscillators ω_T. The heavy lines are the dispersion relations in the presence of coupling between the lattice oscillators and the electromagnetic wave. Between the ω_T to ω_L frequency gap the wavevector is pure imaginary, with magnitude given by the broken line in the figure. In the gap the wave attenuates as $\exp(-|K|x)$, and we see from the plot that the attenuation is much stronger near ω_T than near ω_L.

Plasmon Polaritons

Plasmons are quasiparticles which result from the quantization of the plasma oscillations. To quote Wikipedia:

> The plasmon can be considered as a quasiparticle since it arises from the quantization of plasma oscillations, just like phonons are quantizations of mechanical vibrations. Thus, plasmons are collective (a discrete number) oscillations of the free electron gas density. For example, at optical frequencies, plasmons can couple with a photon to create another quasiparticle called a plasmon polariton.

Kittel Fig. 14.8 provides evidence for the creation of these plasmon quasiparticles through electron energy loss spectra. The loss of electron energy here can be considered to be due to the creation of plasmons, just like the loss of photon energy in Raman scattering can be considered to be due to the creation of phonons.



Kittel Fig. 14.8. Energy loss spectra for electrons reflected from films of (a) aluminum and (b) magnesium, for primary electron energies of 2020 eV. The 12 loss peaks observed in Al are made up of combinations of 10.3 and 15.3 eV losses, where the 10.3 eV loss is due to surface plasmons and the 15.3 eV loss is due to volume plasmons. The ten loss peaks observed in Mg are made up of combinations of 7.1 eV surface plasmons and 10.6 eV volume plasmons.

As the Wikipedia quote said, the quasiparticle created by the coupling of photons to the plasma oscillations in conductors can be called *plasmon polaritons*. From the Lorentz model handout we had the following equation for conductors (assuming no damping, for simplicity):

$$\epsilon_r(\omega) = \epsilon_r(\infty) - \frac{\omega_p^2}{\omega^2} \tag{5}$$

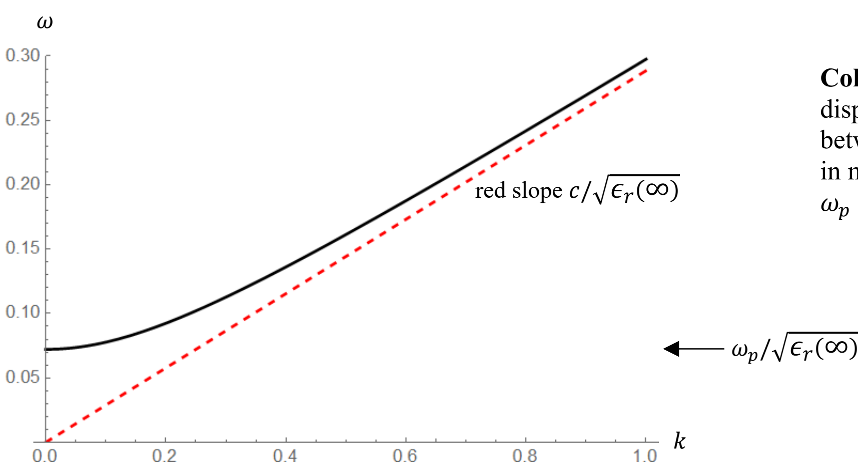
Plugging Eq. 5 into Eq. 1 yields the following plasmon polariton dispersion equation:

$$\frac{c^2k^2}{\omega^2} = \epsilon_r(\infty) - \frac{\omega_p^2}{\omega^2} \tag{6}$$

Eq. 6 can be solved analytically for $\omega(k)$ with just a couple of lines of algebra (which I'll skip):

$$\omega = \sqrt{\frac{c^2k^2 + \omega_p^2}{\epsilon_r(\infty)}} \tag{7}$$

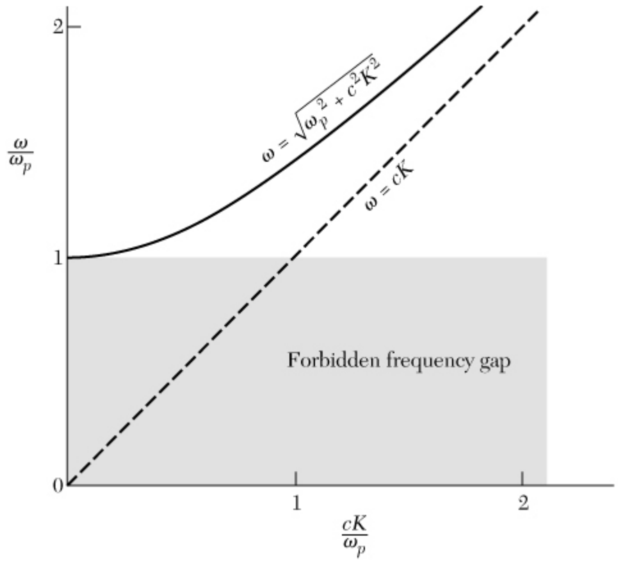
I'll plot that equation for $c = 1$, $\epsilon_r(\infty) = 12$, and $\omega_p = 0.25$, which are the same values used in "Colton plot 4" in the *Lorentz oscillator* handout.



Colton plot 3. Plot of plasmon polariton dispersion, namely $\omega(k)$ for the coupling between EM waves and the free electrons in metals, with $c = 1$, $\epsilon_r(\infty) = 12$, and $\omega_p = 0.25$.

For large frequencies the dispersion obeys the normal photon behavior of $\omega = ck/n$, with n equal to the high frequency value $\sqrt{\epsilon(\infty)}$. However, for low frequencies the photons do not follow their normal dispersion but instead they couple to the plasma oscillations of the free electrons. At frequencies sufficiently close to the modified plasma frequency, namely $\omega_p/\sqrt{\epsilon_r(\infty)}$, you can't even talk about photons and free electrons as being separate entities. The two are intimately connected, and therefore in this region we must only consider the joint plasmon polariton quasiparticle. Frequencies less than that frequency cannot couple into the material at all and give rise to perfect reflectivity as plotted in "Colton plot 4" in the *Lorentz oscillator* handout.

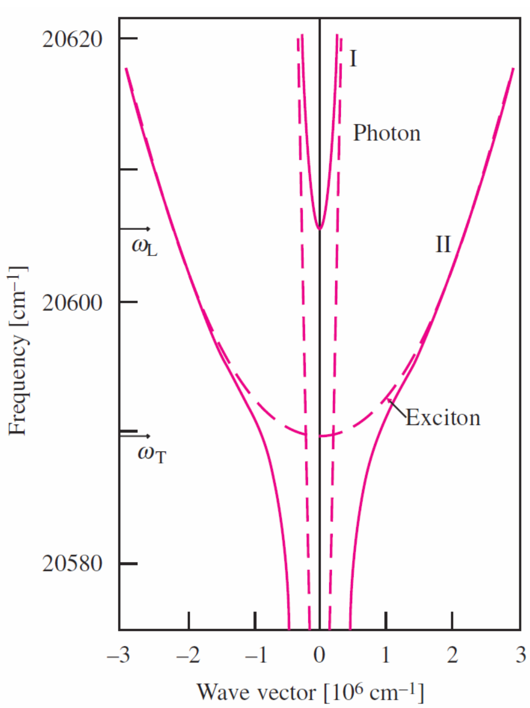
This is also depicted in Kittel Fig. 14.2, which assumes $\sqrt{\epsilon(\infty)} = 1$.



Kittel Fig. 14.2. Dispersion relation for transverse electromagnetic waves in a plasma. The group velocity $v_g = d\omega/dK$ is the slope of the dispersion curve. Although the dielectric function is between zero and one, the group velocity is less than the velocity of light in vacuum.

Exciton Polaritons

There is also such a thing as an *exciton polariton*, which is a coupling between photons and excitons which occurs when the absorption of a photon automatically leads to the creation of an exciton. Here's a plot from Yu and Cardona which demonstrates how the normal photon and excitons dispersion relations couple together in the region where frequencies and wave vectors overlap.



Yu and Cardona Fig. 6.22. Dispersion curves of a "bare" photon, a "bare" exciton (dashed curves) and an exciton-polariton (solid curves labeled I and II) for the A exciton in CdS. The curves labeled I and II are usually referred to as the "upper" and "lower" branches of the polariton.