Lecture 14

This is an easy equation to solve by

$$E = E_0 e^{\Gamma z}$$

⇨ $\Gamma\varepsilon = -i\frac{\beta\gamma}{2}\varepsilon \Rightarrow \Gamma = -i\frac{\beta}{2}\left(\chi' + i\chi''\right)$

Or $\Gamma = \frac{\beta}{2}\chi'' - i\frac{\beta}{2}\chi' \Rightarrow \varepsilon\left(z\right) = \varepsilon_0 e^{\alpha_m z - i\Delta\beta_m z}$

Def.

$$\alpha_m\left(\omega\right) = \frac{\beta}{2}\chi''$$ = absorption coefficient

- Remember $\chi'' \propto -\chi_{0''} < 0 \rightarrow \alpha_m < 0$

$\Rightarrow \left|\varepsilon\left(z\right)\right| = E_0 e^{-\left|\alpha_m\right| z}$

Exponential attenuation with propagation*(*sometimes called “Beer’s Law”)

- Note that the spectrum of the absorption coefficient is the same as that of the imaginary part of the susceptibility , i.e. a Lorentzian
- This formulation is especially useful in laser theory , where you will find that $\alpha_m > 0$ in a medium with a population inversion => then get exponential gain ,or amplification

$\boxed{\varepsilon\left(z\right) = \varepsilon_0 e^{\alpha_m z - i\Delta\beta_m z}}$ $\tilde{E}\left(\alpha, z\right) = E_0 e^{\alpha_m z - i\Delta\beta_m z} e^{i\omega t z}$

We can also define:

$$\Delta\beta_m = \frac{\beta}{2}\chi'\left(\omega\right)$$ = phase shift factor

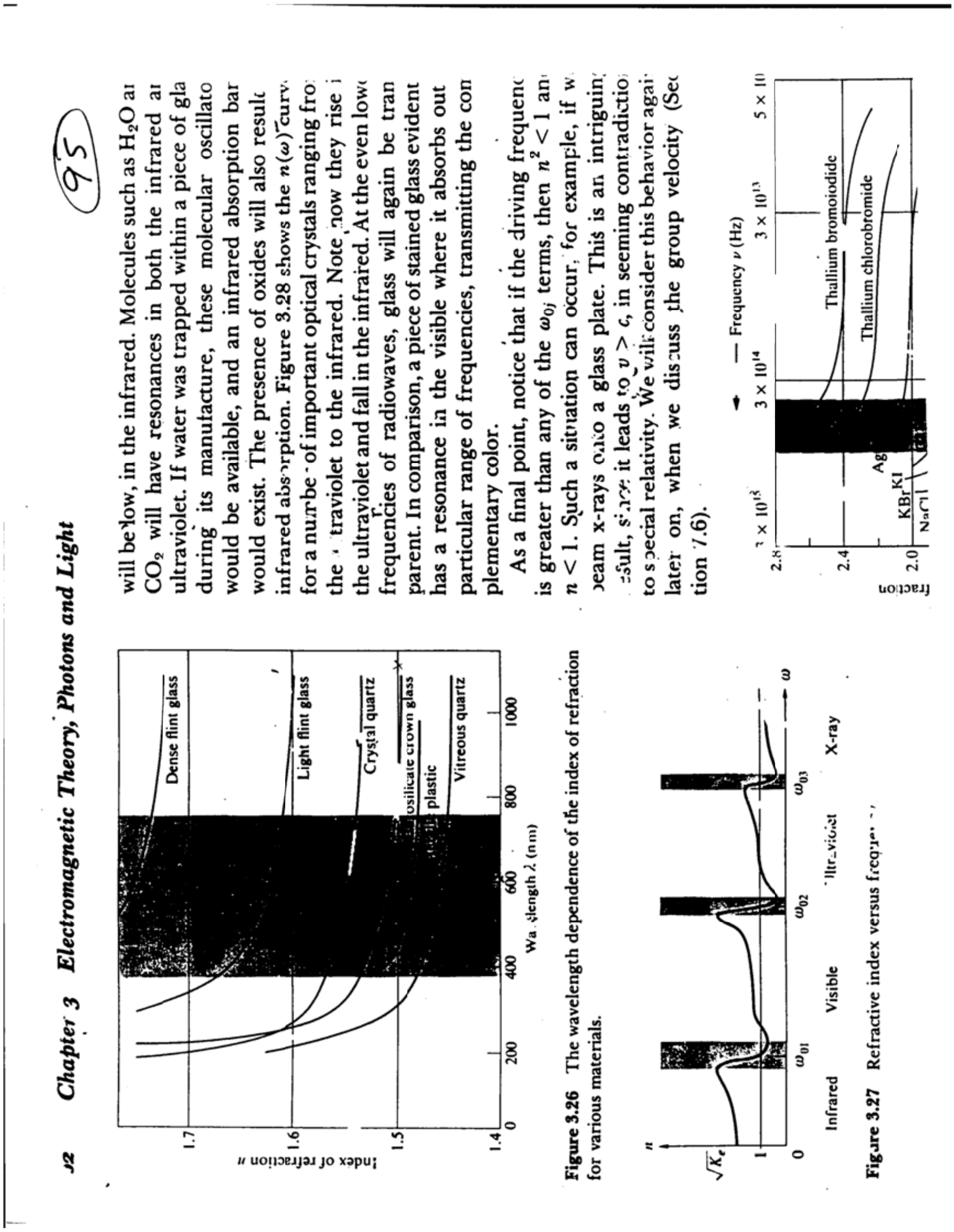
($\Delta\beta_m z$ = phase shift accumulated by the wave on propagating a distance z), relative to vacuum phase shift dispersive lineshape.

Now we can go back to the plot on P.95 + understand the frequency-dependence of the index of refraction. At each resonance, the index shows a dispersive line shape, following the real part of the susceptibility.

Note the “anomalous dispersion” near the center of an absorption resonance!

Final note: the above calculation is valid for a dilute gas, in which case the amplitude change and phase shift occur on a scale much longer than a wavelength. (It is also adaptable in a very straightforward way to resonant absorption and dispersion of atoms incorporated at not-too-high a density into a host dielectric, which is very useful for laser theory .Example include Nd ions in glass , or titanium in sapphire .)

However, if the atoms are very close, i.e. the atoms form a dense dielectric, then some modifications are required to the model.



92 **Chapter 3 Electromagnetic Theory, Photons and Light**

95

Figure 3.26 The wavelength dependence of the index of refraction for various materials.

Figure 3.27 Refractive index versus freque

will below, in the infrared. Molecules such as H_2O a
CO_2 will have resonances in both the infrared a
ultraviolet. If water was trapped within a piece of gla
during its manufacture, these molecular oscillato
would be available, and an infrared absorption bar
would exist. The presence of oxides will also resul
infrared absorption. Figure 3.28 shows the $n(\omega)$ curv
for a number of important optical crystals ranging fro
the ultraviolet to the infrared. Note how they rise i
the ultraviolet and fall in the infrared. At the even low
frequencies of radiowaves, glass will again be tran
parent. In comparison, a piece of stained glass evident
has a resonance in the visible where it absorbs out
particular range of frequencies, transmitting the con
plementary color.

As a final point, notice that if the driving frequen
is greater than any of the ω_{0j} terms, then $n^2 < 1$ an
$n < 1$. Such a situation can occur, for example, if w
beam x-rays onto a glass plate. This is an intriguin
result, since it leads to $v > c$, in seeming contradictio
to special relativity. We will consider this behavior agai
later on, when we discuss the group velocity (Se
tion 7.6).

Dense dielectric

When light interacts with individual atoms, the polarization induced in the atom is proportional to the applied electric field. Often one writes for a single atom:

$$\vec{\mu} = -e\vec{x} = \alpha\vec{E} \qquad (\alpha = \frac{x}{N})$$

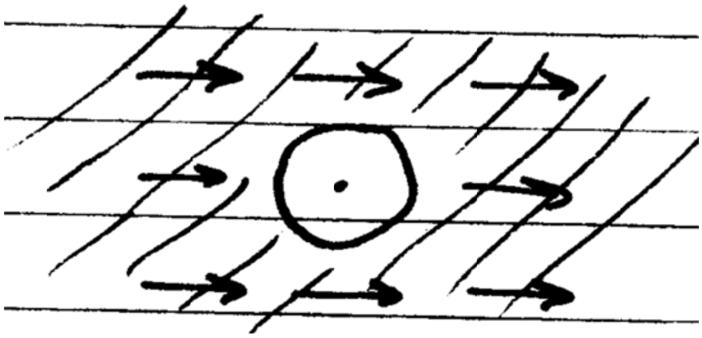
When α is the "polarizability" of the atom or molecule.

Our assumption in the treatment above has been that the atoms are sufficiently far apart (i.e. the system is "sufficiently dilute") that they do not interact. However, if the atoms are close, then a dipole moment on one atom can induce a dipole moment on another.
Therefore, the dipole moment induced in a green molecule is the sum of the moment induced by the external applied field and the moment induced by the other dipoles surrounding the molecule.

The useful way of expressing this is to say we still have the form $\vec{\mu} = \alpha\vec{E}$ but the appropriate field is not the external applied field but the "local field", where

$\vec{E}_{loc}$ = external field + field due to surrounding dipoles.

The full calculation of the field would take us too far afield; most texts on electromagnetic theory carry out the derivation (at least for d.c. fields), and I have induced a copy of a section from Reitz, Milford, and Christy's Foundations of Electromagnetic Theory. The basic idea is to consider a molecule in a void inside a uniform dielectric .The dielectric has a macroscopic polarization $\vec{P}$.



The local field inside the void is shown to be

$$\vec{E}_{loc} = \vec{E} + \frac{1}{3\varepsilon_0}\vec{P}$$

Where $\vec{E}$ = external applied field.

It is the local field which induces a polarization on our molecule:

$$\vec{\mu} = \alpha\vec{E}_{loc}$$

As before, we can make the connection $\vec{P} = N\vec{\mu}$, provided our equation of motion for $\vec{P}$ includes dephasing as well as energy decay. Thus we have (since the polarization is driven by local fields)

$$\frac{d^2\vec{P}}{dt^2}+\left(\gamma+\frac{2}{T_2{}'}\right)\frac{d\vec{P}}{dt}+\omega_0^2\vec{P}=\frac{Ne^2}{m}\vec{E}_{loc}=\frac{Ne^2}{m}\left(\vec{E}+\frac{1}{3\varepsilon_0}\vec{P}\right)$$

$$\frac{d^2\vec{P}}{dt^2}+\left(\gamma+\frac{2}{T_2{}'}\right)\frac{d\vec{P}}{dt}+\left(\omega_0^2-\frac{Ne^2}{3\varepsilon_0 m}\right)\vec{P}=\frac{Ne^2}{m}\vec{E}$$

The quantity $\sqrt{\frac{Ne^2}{\varepsilon_0 m}}$ is often called the plasma frequency (see Guenther), even though we are considering a model with bound electrons, not free electrons. Thus we have

$$\frac{d^2\vec{P}}{dt^2}+\left(\gamma+\frac{2}{T_2{}'}\right)\frac{d\vec{P}}{dt}+\omega_0'^2\vec{P}=\frac{Ne^2}{m}\vec{E}$$

Where $\omega_0'^2=\omega_0^2-\frac{\omega_P^2}{3}$

Thus the effect of the local field is to red-shift the resonance frequency. From a classical point of view, this is not all that important, since we cannot predict the resonance frequencies ω_0, which we consider to be an experimentally determined parameter.

The main significance of local fields are:

(i) In some sense , they provide a connection between a microscopic or atomic theory and a <u>macroscopic</u> quantity , the dielectric constant .This is usually done via the Lorentz-lorenz equation:

$$\frac{\varepsilon_r-1}{\varepsilon_r+2}=\frac{N\alpha}{3\varepsilon_0} \quad \text{where} \quad \varepsilon=\varepsilon_r\varepsilon_0 \text{ as usual,}$$

which you will derive in the homework (Born+Wolf 2.3.3)

(ii) Local fields are sometimes important in nonlinear optics, particularly in calculations of the nonlinear susceptibility.

Surprisingly, the Lorentz-lorenz shift was predicted in the late 19^{th} century, but was not unambiguously experimentally until 1991!

Ref.: J.J. Mark, etal., PRL 67,972(1991)

CHAPTER 5 Microscopic Theory of Dielectrics

In the preceding chapter we were concerned with the macroscopic aspects of dielectric polarization, and it was shown how in many cases the polarization could be taken into account through the introduction of a dielectric constant. In this way the electric field could be computed directly from a consideration of the external charge distribution. Although reference was made to the molecules of the dielectric several times in Chapter 4, a microscopic treatment of the material was not carried through in detail, and the over-all picture that was presented was certainly from the macroscopic point of view. We should now like to examine the molecular nature of the dielectric, and see how the electric field responsible for polarizing the molecule is related to the macroscopic electric field. Furthermore, on the basis of a simple molecular model it is possible to understand the linear behavior that is characteristic of a large class of dielectric materials.

5–1 MOLECULAR FIELD IN A DIELECTRIC

The electric field that is responsible for polarizing a molecule of the dielectric is called the molecular field, $\mathbf{E}_m$. This is the electric field at a molecular position in the dielectric; it is produced by all external sources and by all polarized molecules in the dielectric *with the exception* of the one molecule at the point under consideration. It is evident that $\mathbf{E}_m$ need not be the same as the macroscopic electric field because, as was discussed in Section 4–3, the latter quantity is related to the force on a test charge that is large in comparison with molecular dimensions.

The molecular field may be calculated in the following way. Let us cut out a small piece of the dielectric, leaving a spherical cavity surrounding the point at which the molecular field is to be computed. The dielectric that is left will be treated as a continuum, i.e., from the macroscopic point of view. Now we put the dielectric back into the cavity, molecule by molecule, except for the molecule at the center of the cavity where we wish to compute the molecular field. The molecules that have just been replaced are to be treated, not as a continuum, but as individual dipoles. The procedure just outlined can be justified only if the result of the calculation is independent of the size of the cavity; we shall see that under certain conditions this is indeed the case.

Let us suppose that the thin dielectric sample has been polarized by placing it in the uniform electric field between two parallel plates which are oppositely charged, as shown in Fig. 5–1(a). It will be assumed that the polarization is uniform on a macroscopic scale (i.e., $\nabla \cdot \mathbf{P} = 0$), and that $\mathbf{P}$ is parallel to the field producing it. The part of the dielectric outside the cavity may be replaced by a system of polarization charges as shown in Fig. 5–1(b), whence the electric field at the center of the cavity may be written as

$$\mathbf{E}_m = \mathbf{E}_x + \mathbf{E}_d + \mathbf{E}_s + \mathbf{E}'. \tag{5–1}$$

Here $\mathbf{E}_x$ is the primary electric field due to the charged parallel plates, $\mathbf{E}_d$ is the depolarizing field due to polarization charge on the outside surfaces of the dielectric, $\mathbf{E}_s$ is due to polarization charge on the cavity surface S, and $\mathbf{E}'$ is due to all of the dipoles inside of S. Although we are not concerned with the explicit form of $\mathbf{E}_x$, it is evident that if the dimensions of the plates are large compared with their separation, $E_x = (1/\epsilon_0)\sigma$, where σ is the surface charge density. The depolarizing field is also produced by two parallel planes of charge, this time with the density σ_P. Since $\sigma_P = P_n = \pm P$,

$$\mathbf{E}_d = -\frac{1}{\epsilon_0}\,\mathbf{P}. \tag{5–2}$$

Let us write the macroscopic electric field *in the dielectric* without a subscript, that is, $\mathbf{E}$. Since the normal component of the electric displacement $\mathbf{D}$ is continuous across the vacuum-dielectric interface, and since $\mathbf{D} = \epsilon_0 \mathbf{E}_x$ in the vacuum just outside the dielectric slab,

$$\epsilon_0 \mathbf{E}_x = \epsilon_0 \mathbf{E} + \mathbf{P}. \tag{5–3}$$

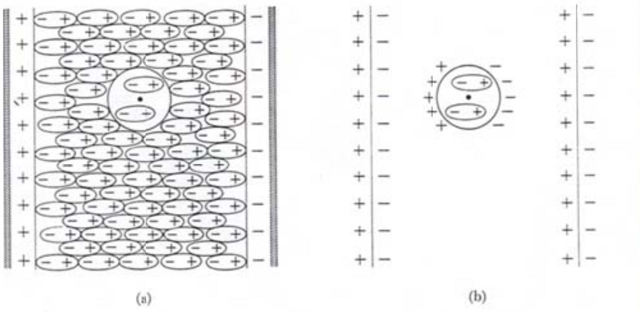

(a) (b)

Figure 5–1 Replacement of the dielectric outside the "cavity" by a system of polarization charges.

Combining Eqs. (5–1), (5–2), and (5–3) yields

$$\mathbf{E}_m = \mathbf{E} + \mathbf{E}_s + \mathbf{E}', \tag{5–4}$$

which is an equation relating the molecular field to the macroscopic electric field in the dielectric material. This result is quite general, and not restricted to the geometry of Fig. 5–1; nevertheless, the above derivation is instructive and will be useful to the subject discussed in Section 5–4.

The field $\mathbf{E}_s$ arises from polarization charge density, $\sigma_P = P_n$, on the spherical surface S. Using spherical coordinates, and taking the polar direction along the direction of $\mathbf{P}$, as in Fig. 5–2, we obtain

$$d\mathbf{E}_s = \frac{(-P\cos\theta)}{4\pi\epsilon_0 r^3}\,\mathbf{r}\,da, \tag{5–5}$$

where $\mathbf{r}$ is the vector from the surface to the center of the sphere. From symmetry, it is evident that only the component of $d\mathbf{E}_s$ along the direction of $\mathbf{P}$ will contribute to the integral of Eq. (5–5) over the complete surface. Since $da = r^2 \sin\theta\, d\theta\, d\phi$,

$$\begin{aligned} \mathbf{E}_s &= \frac{1}{4\pi\epsilon_0}\,\mathbf{P}\int_0^{2\pi} d\phi \int_0^{\pi} \cos^2\theta \sin\theta\, d\theta \\ &= \frac{1}{3\epsilon_0}\,\mathbf{P}. \end{aligned} \tag{5–6}$$

Finally, we come to the last term in Eq. (5–4), that due to the electric dipoles inside S. There is a number of important cases for which this term vanishes. If there are a great many dipoles in the cavity, if they are oriented parallel but randomly distributed in position, and if there are no correlations between the positions of the dipoles, then $\mathbf{E}' = 0$. This is the situation that might prevail in a gas or a liquid. Similarly, if the dipoles in the cavity are located at the regular atomic positions of a cubic crystal,[*] then again $\mathbf{E}' = 0$. In this connection, the reader is referred to Problem 5–2.

In the general case, $\mathbf{E}'$ is not zero, and if the material contains several species of molecule, $\mathbf{E}'$ may differ at the various molecular positions. It is this term that

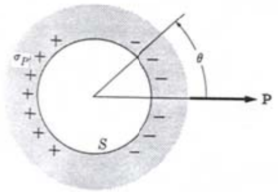


Figure 5–2 Calculation of the "cavity" surface contribution to $\mathbf{E}_m$.

---

[*] Crystals with the highest symmetry belong to the cubic system.

gives rise to the anisotropic electrical behavior of calcite, for example. It is not our purpose, however, to develop a theory of anisotropic materials; hence we restrict further discussion to the rather large class of materials in which $\mathbf{E}' = 0$. Thus, Eq. (5–4) reduces to

$$\mathbf{E}_m = \mathbf{E} + \frac{1}{3\epsilon_0}\,\mathbf{P}. \tag{5–7}$$

It is interesting to note that this result would be obtained directly by the above method if the spherical cavity were created by removing just one molecule. But under these conditions the cavity would be so small that the replacement of the rest of the dielectric by a system of polarization charges could not be justified.

The dipole moment of a molecule per unit polarizing field is called its *polarizability*, α. In other words,

$$\mathbf{p}_m = \alpha \mathbf{E}_m. \tag{5–8}$$

If there are N molecules per unit volume, then the polarization $\mathbf{P} = N\mathbf{p}_m$, and combining this result with Eqs. (5–7) and (5–8), we obtain

$$\mathbf{P} = N\alpha\left(\mathbf{E} + \frac{1}{3\epsilon_0}\,\mathbf{P}\right). \tag{5–9}$$

This equation may be rewritten in terms of the dielectric constant, K, since $\mathbf{P} = (K-1)\epsilon_0\mathbf{E}$. In this way, Eq. (5–9) becomes

$$\alpha = \frac{3\epsilon_0}{N}\,\frac{(K-1)}{(K+2)}, \tag{5–10}$$

which is known as the Clausius-Mossotti equation. It is evident that Eq. (5–10) defines a molecular property, namely, the molecular polarizability, in terms of quantities that can be determined on a macroscopic basis.

5–2 INDUCED DIPOLES. A SIMPLE MODEL

The molecules of a dielectric may be classified as *polar* or *nonpolar*. A polar molecule is one that has a permanent dipole moment, even in the absence of a polarizing field $\mathbf{E}_m$. In the next section the response of a polar dielectric to an external electric field will be studied, but here we deal with the somewhat simpler problem involving nonpolar molecules, in which the "centers of gravity" of the positive and negative charge distributions normally coincide. Symmetrical molecules such as H_2, N_2, and O_2, or monatomic molecules such as He, Ne, and Ar, fall into this category.

The application of an electric field causes a relative displacement of the positive and negative charges in nonpolar molecules, and the molecular dipoles so created are called *induced dipoles*. The simplest type of molecule that can be envisaged is that composed of a single neutral atom. It is possible to construct a simple classical model for the atom and from this model derive an expression for

Metal and Plasmas

Start with the general susceptibility

$$\chi(\omega)=\frac{Ne^2}{\varepsilon_0 m}\frac{1}{\omega_0^2-\omega^2+i\omega\gamma}$$

Fine electrons: no restoring force

$$F=-\mathrm{K}x=-m\omega_0^2x=0\Rightarrow \underline{\underline{\omega_0=0}}$$

Damping γ due to collisions (in a solid, have electron-electron and election-phonon collisions, with characteristic collision time $\tau = {}^{1}\!/_{\gamma}$).

$$\chi(\omega)=-\frac{Ne^2}{\varepsilon_0 m}\frac{1}{\omega^2-i\omega\gamma}$$

Dielectric constant $\varepsilon=\varepsilon_{\mathrm{r}}\varepsilon_0$ with

$$\varepsilon_{\mathrm{r}}(\omega)=1+\chi(\omega)$$

Let's separate out the dielectric constant due to the lattice (I.e. describes the polarization due to all other mechanism other than the free charge) by introducing a background dielectric constant ε_{L} (L for lattice)

$$\varepsilon_{\mathrm{r}}=\varepsilon_{\mathrm{L}}\underbrace{\left[1+\chi(\omega)\right]}_{\text{free electron contribution}}$$

$$\varepsilon_{\mathrm{r}}=\varepsilon_{\mathrm{L}}\left(1-\frac{\omega_P^2}{\omega^2-i\omega\gamma}\right)$$

Where we have defined

$$\boxed{\omega_P^2=\frac{Ne^2}{\varepsilon_0 m}}\quad \underline{\text{ plasma frequency }}$$

N=# free electrons/vol.

- in a solid m $\longrightarrow m^*$ = effective mass
- get real and imaginary parts

$$\begin{aligned}\frac{\varepsilon_{\mathrm{r}}}{\varepsilon_{\mathrm{L}}}&=1-\frac{\omega_P^2\left(\omega^2+i\omega\gamma\right)}{\omega^4+\omega^2\gamma^2}\\&=1-\frac{\omega_P^2\omega^2}{\omega^4+\omega^2\gamma^2}-i\frac{\omega_P^2\omega\gamma}{\omega^4+\omega^2\gamma^2}\end{aligned}$$

So $\underline{\varepsilon_{\mathrm{r}}=\varepsilon_1+i\varepsilon_2}$ with

$$\varepsilon_1 = \varepsilon_{\mathrm{L}}\left(1-\frac{\omega_P^2}{\omega^2+\gamma^2}\right)$$ real part of dielectric constant

$$\varepsilon_2 = -\varepsilon_{\mathrm{L}}\frac{\gamma}{\omega}\frac{\omega_P^2}{\omega^2+\gamma^2}$$ Imaginary part

(This is called the "Drude model")

Most good metals have $\hbar\omega_P = 3-20eV$

or $\omega_P = 4.5\times10^{15} - 3\times10^{16}s^{-1}$

which is out in the UV.

For a doped semiconductor, e.g. N~$10^{18}cm^{-3}$,

$$\omega_P \simeq 6\times10^{13}s^{-1}\left(\leftrightarrow \lambda_\rho = 33\mu m\right)$$

Which is in the mid-IR

If the damping is not too strong $\omega_P^2 \gg \frac{\alpha_\rho^{\ 2}}{4}$ then the Plasmon resonance is well defined

⇨ Neglecting the damping

$$\varepsilon_{\mathrm{r}} \simeq \varepsilon_1 \simeq \varepsilon_{\mathrm{L}}\left(1-\frac{\omega_P^2}{\omega^2}\right)$$

For light at optical frequencies incident on a metal

$$\omega \ll \omega_P$$

⇨ $\varepsilon_{\mathrm{r}} < 0$!

A negative dielectric constant gives rise to an evanescent wave, as seen from the Helmholtz eqn.

$$\left(\nabla^2 + \frac{\omega^2}{c^2}\varepsilon_{\mathrm{r}}\right)E = 0$$

$$k^2 = \frac{\omega^2}{c^2}\varepsilon_{\mathrm{r}} < 0 \Rightarrow k = i\kappa$$ imaginary

⇨ The incident wave is perfectly reflected by the metal

For xuv $\longrightarrow$ x-rays , $\omega > \omega_p$,so

$$\varepsilon_r = \varepsilon_L\left(1-\frac{\omega_P^2}{\omega^2}\right) > 0$$

⇨ Waves can propagate through the metal

⇨ Metal becomes transparent

"ultraviolet transparency"

Al is an example of a good Drude metal

Cu, Au, and even Ag require contributions from <u>bond</u> electrons to get a good description of their dielectric constants and hence reflectivity

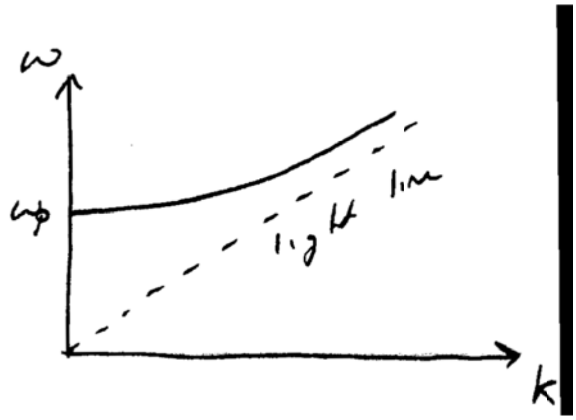
Dispersion relation for EM waves in bulk metal:

$$k^2 = \frac{\omega^2}{c^2}\varepsilon_r(\omega) = \frac{\omega^2}{c^2}\varepsilon_L\left(1 - \frac{\omega_P^2}{\omega^2}\right) = \frac{\varepsilon_L}{c^2}\left(\omega^2 - \omega_P^2\right)$$

Or $\omega^2 = \frac{k^2c^2}{\varepsilon_L} + \omega_P^2$

Propagating modes are only for

$\omega > \omega_P$ (UV transparency)



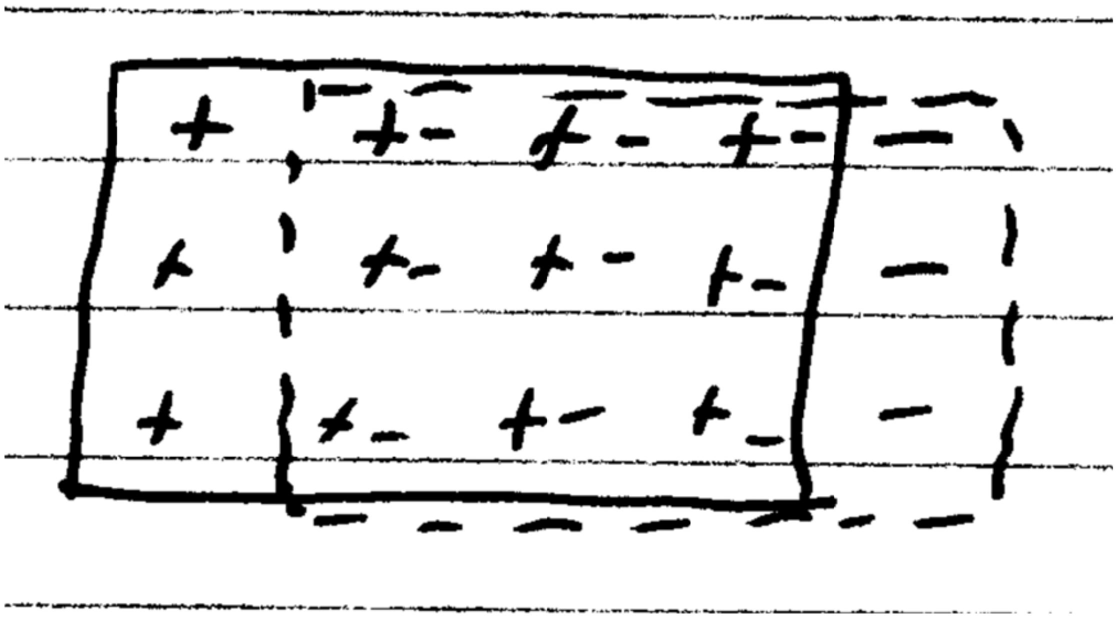
Note that k=0 corresponds to "infinite wavelength"; we see on the next page that corresponds to

$\varepsilon_r = 0$ at $\omega = \omega_P$, which are the longitudinal oscillations of the electron plasma

(Generally excited only by changed particle beams, not by EM waves .)

What is a Plasmon? (bulk, i.e. 3-D material）

- Assume ions are fixed
- Some excitation displaces the electrons



- Coulomb restoring force =>oscillations at the Plasmon frequency $\omega_P = \sqrt{\frac{Ne^2}{\varepsilon_0 m}}$
- Note that at $\omega = \omega_P$

$$\varepsilon_1 = \varepsilon_0 \left(1 - \frac{\omega_P^2}{\omega^2}\right) = 0$$

⇨ $\vec{D} = \varepsilon_0 \vec{E} + \vec{P} = \varepsilon_r \varepsilon_0 \vec{E} = 0$

So $\vec{P} = -\varepsilon_0 \vec{E}$

And $\nabla \times \vec{H} = -\frac{\partial \vec{D}}{\partial \mathrm{t}} = 0$

- $\varepsilon = 0$ corresponds to longitudinal excitations of the electron plasma, which do not radiate (are not coupled to propagating transverse EM fields ）
- This is a general result：the zeroes of the dielectric function yield the longitudinal excitations of the system.