

## Module 1 : Introduction and Background Material

### Lecture 2 : Linear Optical Response Of Matter

#### Objectives

##### In this lecture we will look at

- Microscopic and macroscopic Maxwell's equations.
- Polarization in homogeneous isotropic media.
- Lorentz model for electronic polarization.

To describe optical wave in a material medium, it is important to know how light interacts with the medium. This is a complex problem in general, because the medium consists of a large number of charged particles and a complete description of their motion is difficult. Clearly many approximations need to be made.

We may start by saying that matter consists of relatively loosely bound electrons and nearby rigid ions, bound to each other by a net force that is attractive at long distance. At very short distances ( $\sim a$ , the atomic size) quantum confinement causes a net repulsive interaction between atoms.

In general, one can formulate the interaction of light with matter in terms of microscopic Maxwell's equation. This is based on the following scenario. The oscillating fields  $\vec{E}, \vec{H}$  induce currents and charge polarization represented by an induced current density  $\vec{j}(\vec{x}, t)$  and an induced charge density  $\rho(\vec{x}, t)$ . These in turn modify  $\vec{E}, \vec{H}$  and so create a self consistent field  $\vec{E}, \vec{H}$  which obey the microscopic Maxwell's equations.

$$\vec{\nabla} \cdot \vec{E} = \frac{\rho}{\epsilon_0} \quad (2.1)$$

$$\vec{\nabla} \cdot \vec{H} = 0 \quad (2.2)$$

$$\vec{\nabla} \times \vec{H} = \epsilon_0 \frac{\partial \vec{E}}{\partial t} + \vec{j} \quad (2.3)$$

$$\vec{\nabla} \times \vec{E} = -\mu_0 \frac{\partial \vec{H}}{\partial t} \quad (2.4)$$

Both the fields the charge density  $\rho$  (charge per unit volume) and current density  $\vec{j}$  (current per unit cross-sectional area) are functions of space coordinates  $\vec{x}$  and time  $t$ .

Usually, it is necessary to make many more approximation for the medium to progress further.

The simplest model of "polarization" of a medium is due to Lorentz. In this the motion of an electron relative to the ion is represented by a damped harmonic oscillator. Denoting the displacement of the electronic charge by  $r$

$$m(\ddot{r} + \gamma \dot{r} + \omega_0^2 r) = \text{Force} = -e(\vec{E} + \vec{v} \times \vec{H}) \quad (2.5)$$

in an optical field  $\vec{v} \times \vec{H}$  term is much smaller and can be neglected. Then  $\vec{r} \parallel \vec{E}$ , and only 1 component is needed.

$$\ddot{r} + \gamma \dot{r} + \omega_0^2 r = -eE/m \quad (2.6)$$

For harmonic field

$$E = E_0 e^{-ikt} + \text{Complex Conjugate (c.c.)} \quad (2.7)$$

We try a solution of the form

$$r = r_0 e^{-i\omega t} + \text{Complex Conjugate(c.c.)}$$

$$(-\omega^2 - i\omega\gamma + \omega_0^2)r = (-e/m)E_0$$

$$r = \frac{(-e/m)E}{(\omega_0^2 - \omega^2 - i\omega\gamma)} \quad (2.8)$$

Induced dipole moment given by

$$-er = \frac{(e^2/m)E}{\omega_0^2 - \omega^2 - i\omega\gamma} \quad (2.9)$$

If there are N oscillators per unit volume, we may then write, the dipole moment per unit volume as

$$\vec{P} = \frac{Ne^2/m}{\omega_0^2 - \omega^2 - i\omega\gamma} \vec{E} = \epsilon_0 \chi(\omega) \vec{E} \quad (2.10)$$

$\chi(\omega)$  is called the Susceptibility and is given by.

$$\chi(\omega) = \frac{Ne^2}{m\epsilon_0} \cdot \frac{1}{\omega_0^2 - \omega^2 - i\omega\gamma} \quad (2.11)$$

For an isotropic medium  $\chi$  is a scalar.

More generally, the susceptibility  $\vec{\chi}$  is a tensor of rank two. The general relation between  $\vec{P}$  and  $\vec{E}$  is written as

$$\vec{P}(\omega) = \vec{\chi}(\omega) \cdot \vec{E}(\omega) \quad (2.12)$$

i.e.

$$\vec{P}_i(\omega) = \sum_j \chi_{ij}(\omega) E_j(\omega) \quad (2.13)$$

where  $\vec{P}(\omega)$  the polarization that varies with frequency  $\omega$  and  $\vec{E}(\omega)$  is the corresponding electric field.

We write

$$\chi(\omega) = \chi_R(\omega) + i\chi_I(\omega) \quad (2.14)$$

with  $\chi_R, \chi_I$  as the real and imaginary parts of the susceptibility  $\chi(\omega)$ , given by

$$\chi_R(\omega) = \frac{(Ne^2/m\epsilon_0)(\omega_0^2 - \omega^2)}{(\omega_0^2 - \omega^2)^2 + \omega^2\gamma^2} \quad (2.15)$$

$$\chi_I(\omega) = (Ne^2/m\epsilon_0) \cdot \frac{\omega\gamma}{(\omega_0^2 - \omega^2)^2 + \omega^2\gamma^2} \quad (2.16)$$

In the limit

$$\gamma \rightarrow 0$$

$$\begin{aligned} \chi_R &\rightarrow \frac{Ne^2}{m\epsilon_0} \cdot \frac{1}{\omega_0^2 - \omega^2} \\ &\rightarrow \frac{Ne^2}{m\omega_0^2\epsilon_0} \text{ as } \omega \rightarrow 0 \\ &\rightarrow -\frac{Ne^2}{m\omega_0^2\epsilon_0} \text{ as } \omega \ll \omega_0 \end{aligned} \quad (2.17)$$

and

$$\chi_R \rightarrow -\frac{Ne^2}{m\omega_0^2\epsilon_0} \text{ as } \omega \rightarrow \infty \text{ or } \omega \gg \omega_0 \quad (2.18)$$

For the imaginary part we have

$$\chi_I = \frac{Ne^2}{m\epsilon_0} \pi \delta(\omega_0^2 - \omega^2) \quad (2.19)$$

Thus for narrow resonances the imaginary part of the susceptibility is large only near the resonance  $\omega \approx \omega_0$  while the real parts become negligibly small for  $\omega \approx \omega_0$ .

This has an important implication- the contribution of any materials excitation may be significant at all frequencies below or comparable to the resonance frequencies while at frequencies, well above the resonance this excitation does not contribute significantly. Of course, at any frequency  $\omega$ , there excitations contribute more which are close to it. When free electrons present as in metals and semiconductor, we let  $\omega_0 \rightarrow 0$  then Lorentz model goes to the Drude model and we get

$$\chi_r(\omega) \rightarrow -\frac{Ne^2}{m\epsilon_0} \frac{\omega^2}{\omega^4 + \omega^2\gamma^2} = -\frac{Ne^2}{m(\omega^2 + \gamma^2)} \quad (2.20)$$

and

$$\chi_I(\omega) \rightarrow \frac{Ne^2}{m\epsilon_0} \frac{\gamma}{\omega^2 + \gamma^2} \quad (2.21)$$

We can now define a displacement vector  $\vec{D}$  which is related to  $\vec{E}$  and  $\vec{P}$  by

$$\vec{D}(\omega) = \epsilon_0 \vec{E}(\omega) + \vec{P}(\omega) = \epsilon \vec{E}(\omega) \quad (2.22)$$

With the dielectric function  $\epsilon(\omega)$  is given by

$$\epsilon(\omega) = \epsilon_0 (1 + \chi(\omega)) \quad (2.23)$$

Since  $\chi(\omega)$  is in general complex, so is  $\epsilon(\omega)$ .

We note that when we use complex susceptibility and dielectric function, the response of bound and free electrons can be treated to ether and both cab be included in the polarization. So in the rest of this course, we do not write  $\vec{P}_{free}$  or  $\vec{E}_{free}$  explicitly .both then as well as the polarization of bound are included in generalized polarization  $\vec{P}$ .

Retaining only the electric dipole polarization is called the electric dipole approximation. It is equivalent to long wavelength approximation i.e., the wavelength of light is much larger than the size of a typical polarizable unit in the medium. With a more realistic model of the medium one can develop the light-matter interaction as a series expansion in the parameter  $(a/\lambda)$ . The electric approximation is the first term in that expansion. we can write  $\epsilon(\omega)$  in a particularly compact form

$$\epsilon(\omega) = \epsilon_0 + \frac{\omega_p^2}{\omega_0^2 - \omega^2 - i\omega\gamma} \quad (2.24)$$

where

$$\omega_p^2 = Ne^2/m\epsilon_0 \quad (2.25)$$

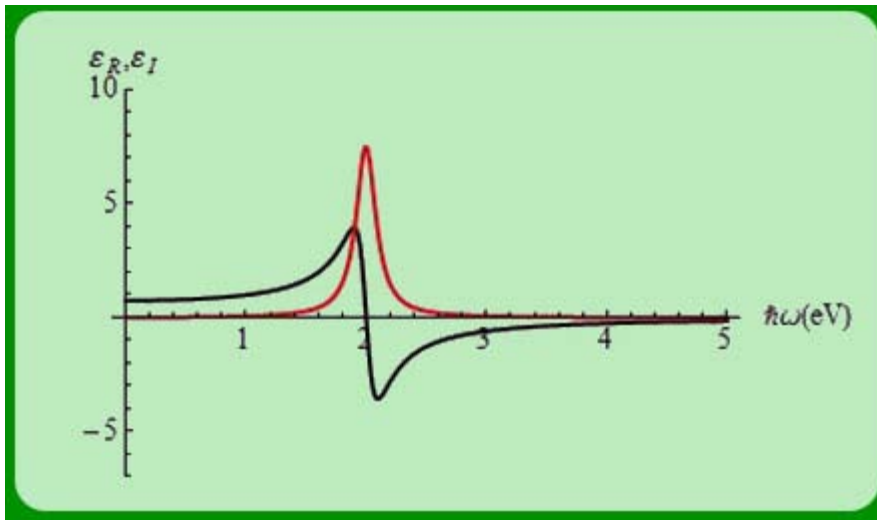


Figure 2.1 Real (Black curve) and Imaginary parts (red curve) of the susceptibility function as function of photon energy

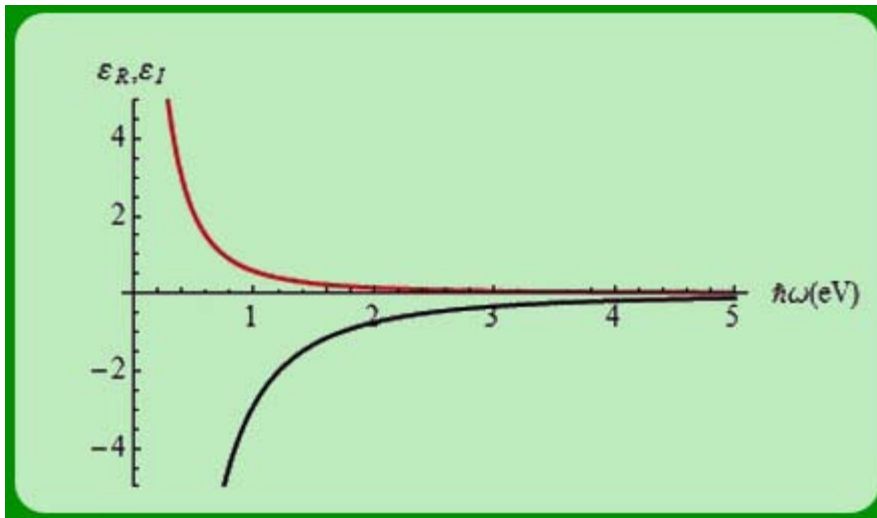


Figure 2.2 Real (Black curve) and Imaginary parts (red curve) of the susceptibility function as function of photon energy for a metal

**Exercise:** Show that  $\omega_p$  has the dimension of a frequency calculate the frequency  $\omega_p$  for silver which has a density of valence electrons =  $M^{-3}$ .

We recall that Maxwell had introduced the displacement current even without knowing the structure of medium simply to accommodate the change of capacitance caused by a dielectric medium in a capacitor.

The Lorentz model is unrealistic to the extent that it has only one resonant frequency. In general, an atomic, molecular or solid state system would have several frequencies of absorption or several resonances.

Generalization of the Lorentz formula is straight forward-one writes (Jackson p.310)

$$\epsilon(\omega) = \epsilon_0 \left( 1 + \frac{Ne^2}{m\epsilon_0} \sum_j \frac{f_j}{\omega_j^2 - \omega^2 - i\omega\gamma_j} \right) \quad (2.26)$$

where  $\omega_j$  is the  $j^{\text{th}}$  resonance frequency.  $f_j$  is called the oscillator strength. This form can be derived by using quantum mechanical perturbation theory (Lecture 3.1). The oscillator strength  $f_j$  is related to square of the matrix element of electric dipole operator between initial and final state corresponding to the  $j^{\text{th}}$  transition. It obeys the sum rule.

Also for a molecular or atomic system it obeys the sum rule

$$\sum_j f_j = Z \quad (2.27)$$

where the sum is over all electron transitions and  $Z$  is the total number of electron in the system. We also note that,

- This accounts only for polarization due to the motion of electrons relative to the nuclear skeleton molecule. The ions are assumed not to move. This is justified at optical frequencies but in the infra-red and the far infrared ionic motion also becomes important. **The motion of ions and its modulation of visible light gives rise to the all important Raman scattering.**
- While for  $\epsilon(\omega)$  we can use the generalized expression given above for  $\mu(\omega)$  we do not have a simple mode. However, for most homogeneous materials  $\mu \simeq \mu_0$  at optical frequencies. Some materials for which  $\mu(\omega)$  and  $\epsilon(\omega)$  are both negative are called left handed or meta- materials. They correspond to a negative refractive index. [See e.g., Veselago... ]
- This form for the dielectric function directly translates to the well known Sellmeier equation for the refractive index.

$$n^2(\lambda) = 1 + \frac{B_1 \lambda^2}{\lambda^2 - C_1} + \frac{B_2 \lambda^2}{\lambda^2 - C_2} + \dots \quad (2.28)$$

Where  $B_1, C_1$  are called the Sellmeier Coefficients.

## RECAP:

### In this lecture we have learned about the

- Lorentz model for electronic polarization is used to describe polarization of a medium in linear approximation.
- Real and imaginary parts of the dielectric function which describe the dispersion and absorption in a medium are obtained for dielectrics and metals.