Classical Electron Oscillator Model of Absorption

We present a simple classical model for the response of a medium subject to a time-dependent electromagnetic field. Under the influence of the field charges are perturbed from their equilibrium positions. For applied field strengths which are small compared to those produced by the nucleus, we can treat bound charges as simple harmonic oscillators perturbed by the applied field. The charges will respond not to the macrospoic field but to the local field. For a dilute medium the distinction beweeen macrospoic and the local field is negligible. Since our main interest is in the frequency response of the medium we will take this to be the case. For dense media we can use Lorentz-Lorenz equation to express the microscopic field in terms of the macroscopic field.

Consider now the response of a bound electron to the applied field. Let $\vec{r}_0$ be the mean position and $\delta\vec{r}$ be the complex[1] displacement of the electron from its mean position. Then for small displacements, the equation of motion for the electron can

[1] The real displacment is given by the real part of $\delta\vec{r}$.

be written as

$$\frac{d^2\delta\vec{r}}{dt^2} + 2\gamma_0\frac{d\delta\vec{r}}{dt} + \omega_0^2\delta\vec{r} = -(e/m)\left[\vec{\mathcal{E}}(\vec{r}_0,t) + \frac{d\delta\vec{r}}{dt}\times\vec{\mathcal{B}}(\vec{r}_0,t)\right]. \tag{5.1}$$

This is the equation of a charged charged harmonic oscillator driven by an electromagnetic field. Here 2 γ_0 is the radiative decay rate (the rate at which the oscillator loses energy due to radiation), ω_0 is the natural frequency (one of the transition frequencies of the atom or molecule). $\vec{\mathcal{E}}$ and $\vec{\mathcal{B}}$ are the (complex) electric and magnetic fields of the incident wave. We have assumed the displacement $\delta\vec{r}$ from equilibrium to be small compared with the wavelength of light (dipole approximation), the variation of the field over dimensions of the order $\delta\vec{r}$ can be ignored. This allows us to use fields evaluated at the equilibrium position $\vec{r}_0$ of the electron in the equation of motion. Now the magnitudes of the electric and magnetic fields are related by $|\vec{\mathcal{B}}| = |\vec{\mathcal{E}}|/c$. This relation holds for a plane wave and for more general waves in transparent media in the geometrical optics limit. Then a comparison of the electric and magnetic force terms in the equation of motion shows that for nonrelativistic motion of the electron the magnetic force is smaller by the factor $|\dot{\delta\vec{r}}|/c << 1$ compared to the electric force. For the nonrelativistic electron motion considered here we can therefore drop the magnetic term. The equation of motion for the electron then reduces to

$$\frac{d^2\delta\vec{r}}{dt^2} + 2\gamma_0\frac{d\delta\vec{r}}{dt} + \omega_0^2\delta\vec{r} = -\frac{e}{m}\vec{\mathcal{E}}(\vec{r}_0, t) \tag{5.2}$$

To study the response of the oscillator to the field, we consider a monochromatic driving field [2], $\vec{\mathcal{E}}(\vec{r}, t) = \vec{\mathcal{E}}(\vec{r})e^{-i\omega t}$. Recalling that the forced oscillations are at the driving frequency, we write the steady-state displacement from the equilibrium position as

$$\delta\vec{r}(t) = \delta\vec{r}_0 e^{-i\omega t} \tag{5.3}$$

where the steady-state amplitude $\vec{r}_0$ is to b e determined. Substituting this in Eq. (5.2) and solving for $\delta\vec{r}_0$ we find the steady-state electronic dsiplacement is given by

$$\delta\vec{r}(t) = \frac{-e\vec{\mathcal{E}}(\vec{r}_0, t)/m}{\omega_0^2 - \omega^2 - 2i\gamma_0\omega}. \tag{5.4}$$

This will give rise to an induced dipole moment $-e\delta\vec{r}(t)$. Different electrons in an atom or molecule will have different natural frequencies and damping. Let a fraction f_j of electrons ($\sum_j f_j = 1$) have natural frequency ω_j and damping constant γ_j. Then their displacement $\delta\vec{r}_j$ from equilibrium position will be

$$\delta\vec{r}_j = -\frac{e\vec{\mathcal{E}}(\vec{r}_j, t)/m}{\omega_j^2 - \omega^2 - 2i\gamma_j\omega}, \tag{5.5}$$

[2] If the field is not monochromatic, we can Fourier analyze the field and consider one of the Fourier components.

Assuming that all the electron oscillators respond in identical fashion (homogeneous medium) we can add the contribution from all such groups of electrons. By carrying out a spatial averaging by introducing a density of electrons $\mathcal{N}$ (number of electrons per unit volume), we find the macroscopic polarization is given by

$$\vec{\mathcal{P}}(\vec{r},t) = \frac{\mathcal{N}e^2}{m}\sum_j \frac{f_j}{\omega_j^2 - \omega^2 - 2i\gamma_j\omega}\vec{\mathcal{E}}(\vec{r})e^{-i\omega t} \equiv \epsilon_0\chi(\omega)\vec{\mathcal{E}}(\vec{r})e^{-i\omega t}\,. \tag{5.6}$$

Here $\vec{\mathcal{E}}(\vec{r})$ is the spatially averaged electric field. Using the definition of dielectric permittivity $\epsilon(\omega) = \epsilon_0[1+\chi(\omega)]$ we obtain

$$\epsilon(\omega) = \epsilon_0\left[1 + \frac{\mathcal{N}e^2}{m\epsilon_0}\sum_j \frac{f_j}{\omega_j^2 - \omega^2 - 2i\gamma_j\omega}\right]. \tag{5.7}$$

In terms of the plasma dispersion frequency

$$\omega_p = \frac{\mathcal{N}e^2}{m\epsilon_0} \tag{5.8}$$

we can write the real and imaginary parts of $\epsilon(\omega)$ as

$$\epsilon'(\omega) = \epsilon_0\left[1 + \omega_p^2\sum_j \frac{f_j(\omega_j^2-\omega^2)}{(\omega_j^2-\omega^2)^2 + 4\gamma_j^2\omega^2}\right] \tag{5.9}$$

$$\epsilon''(\omega) = 2\epsilon_0\omega_p^2\sum_j \frac{f_j\gamma_j\omega}{(\omega_j^2-\omega^2)^2 + 4\gamma_j^2\omega^2} \tag{5.10}$$

Note that $\epsilon'(\omega)$ is an even and $\epsilon''(\omega)$ is an odd function of ω.

For field frequencies comparable to the eigenfrequencies, the dielectric permittivity $\epsilon(\omega)$ is complex. In this range, it is convenient to introduce the complex refractive index $\tilde{n}$ by writing

$$\sqrt{\epsilon(\omega)/\epsilon_0} = \tilde{n}(\omega) \equiv n(\omega) + i\kappa(\omega)\,, \tag{5.11}$$

where n and κ are the real and imaginary parts of $\epsilon(\omega)$. The real part n is called the refractive index and the imaginary part κ is called the attenuation index of the medium. Both are frequency dependent quantities. Equating the real and imaginary parts from the two sides of Eq. (5.11), we can express n and κ in terms of the real and imaginary parts of $\epsilon(\omega)$ as

$$n(\omega) = \sqrt{\frac{1}{2\epsilon_0}\left[\epsilon'(\omega) + \sqrt{\epsilon'^2(\omega) + \epsilon''^2(\omega)}\right]} \tag{5.12}$$

$$\kappa(\omega) = \sqrt{\frac{1}{2\epsilon_0}\left[-\epsilon'(\omega) + \sqrt{\epsilon'^2(\omega) + \epsilon''^2(\omega)}\right]} \tag{5.13}$$

The intermediate frequency behavior will now be discussed in terms of the refractive index and the attenuation index.

Nonresonant behavior: $|\omega - \omega_j| >> \gamma_j$

For field frequencies far removed from any of the resonance frequencies such that the inequality $|\omega_j - \omega| \gg \gamma_j$ holds, the dielectric permittivity can be written as

$$\epsilon(\omega) \approx \epsilon_0 \left[1 + \omega_p^2 \sum_j \frac{f_j}{\omega_j^2 - \omega^2} + i\omega_p^2 \sum_j \frac{2 f_j \gamma_j \omega}{(\omega_j^2 - \omega^2)^2} \right] \equiv \epsilon'(\omega) + i\epsilon''(\omega) \tag{5.14}$$

On comparing the magnitudes of the real and imaginary parts of $\epsilon(\omega)$ in the nonresonant limit, we see that the imaginary part is small compared to the real part $\epsilon''(\omega) \ll \epsilon'(\omega)$ at least by the ratio $\gamma/\omega \ll 1$. This implies that in this frequency range dissipation is negligble. The medium is said to be transparent in this frequency range. It is important to bear in mind that transparency (or opacity) of a medium is frequency dependent phenomenon.

In the transparency frequency range, since the imaginary part of the permittivity is small compared to its real part ($\epsilon'' \ll \epsilon'$), Eqs. (5.12) and (5.13) lead to the following expressions for the refractive index and the attenuation index

$$n^2(\omega) = \frac{\epsilon'(\omega)}{\epsilon_0} = 1 + \omega_p^2 \sum_j \frac{f_j}{\omega_j^2 - \omega^2} \tag{5.15}$$

$$\kappa(\omega) = \frac{\epsilon''(\omega)}{2\epsilon_0 n(\omega)} = \frac{\omega_p^2}{2n(\omega)} \sum_j \frac{f_j \gamma_j \omega}{(\omega_j^2 - \omega^2)^2} \tag{5.16}$$

For many common materials that are transparent at optical frequencies, the eigenfrequencies ω_j lie in the ultraviolet or the infrared part of the spectrum. For such materials the refractive index n is an increasing function of frequency at optical frequencies. This behavior of the refractive index is known as normal dispersion. The expression for the refractive index can be written in terms of the wavelength $\lambda = 2\pi c/\omega$ as

$$n^2 - 1 = \frac{\omega_p^2}{4\pi^2 c^2} \sum_j \frac{f_j \lambda_j^2 \lambda^2}{\lambda^2 - \lambda_j^2} \equiv \sum_j \frac{b_j \lambda^2}{\lambda^2 - \lambda_j^2} \,. \tag{5.17}$$

This formula is known as Sellmeir's dispersion formula. Its form is used to fit the refractive index of materials in their transparency range. For example, the following formula gives the refractive index of fused silica at 20^o C (λ in microns) accurate to $\pm 3 \times 10^{-5}$ in the wavelength range 0.2 μ − 2.1 μ

$$n^2 - 1 = \frac{0.6961663\lambda^2}{\lambda^2 - (0.0684043)^2} + \frac{0.4079426\lambda^2}{\lambda^2 - (0.1162414)^2} + \frac{0.8974794\lambda^2}{\lambda^2 - (9.896161)^2} \tag{5.18}$$

Resonant Behavior : $|\omega_0 - \omega| \sim \gamma_0$

When the field frequency ω lies within a few γ_0 of an eigen frequency, say ω_0, we can use the approximation $\omega_0^2 - \omega^2 \approx 2\omega(\omega_0 - \omega)$ in the resonant term. For example, for atomic electrons with $\omega_j \approx 2\pi \times 10^{14}$ Hz, $\gamma_j \approx 2\pi \times 10^9$ Hz, even when ω differs from ω_j by $10\gamma_j$, $\omega + \omega_j \approx 2\omega$ to one part in 10^4. In all the other terms we can use the off-resonance approximation. By separating the nonresonant (background) and resonant contributions we can write the permittivity as

$$\epsilon'(\omega) = \epsilon_0 \left[1 + \omega_p^2 \sum_j{}' \frac{f_j}{\omega_j^2 - \omega^2} + \frac{\omega_p^2 f_0}{2\omega} \frac{(\omega_0 - \omega)}{(\omega_0 - \omega)^2 + \gamma_0^2}\right]$$

$$\equiv \epsilon_b' + \frac{\omega_p^2 \epsilon_0 f_0}{2\omega} \frac{(\omega_0 - \omega)}{(\omega_0 - \omega)^2 + \gamma_0^2} \tag{5.19}$$

$$\epsilon''(\omega) = 2\epsilon_0 \omega_p^2 \sum_j{}' \frac{f_j \gamma_j \omega}{(\omega_j^2 - \omega^2)^2} + \frac{\omega_p^2 \epsilon_0 f_0}{2\omega} \frac{\gamma_0}{(\omega_0 - \omega)^2 + \gamma_0^2}$$

$$\equiv \epsilon_b'' + \frac{\omega_p^2 \epsilon_0 f_0}{2\omega} \frac{\gamma_0}{(\omega_0 - \omega)^2 + \gamma_0^2} \tag{5.20}$$

As noted earlier, for the nonresonant background the inequality $\epsilon_b'' << \epsilon_b'$ holds. On introducing a background refractive index $n_b \approx \sqrt{\epsilon_b'/\epsilon_0}$ and an attenuation index $\kappa_b = \epsilon_b''/2\epsilon_o n_b$ and writing $\sqrt{\epsilon/\epsilon_0} = n + i\kappa$, we obtain

$$n \approx n_b + \frac{\omega_p^2 f_0}{4 n_b \omega} \frac{(\omega_0 - \omega)}{(\omega_0 - \omega)^2 + \gamma_0^2} \tag{5.21}$$

$$\kappa \approx \kappa_b + \frac{\omega_p^2 f_0}{4 n_b \omega} \frac{\gamma_0}{(\omega_0 - \omega)^2 + \gamma_0^2} \tag{5.22}$$

The resonant terms vary rapidly with frequency ω in the vicinity of ω_0, whereas the background terms n_b and κ_b vary slowly. Dissipation characterized by κ is largest on resonance. A plot of n and κ near a resonance is shown in Fig. (5.1). Note that near a resonance, n can be a decreasing function of frequency. This

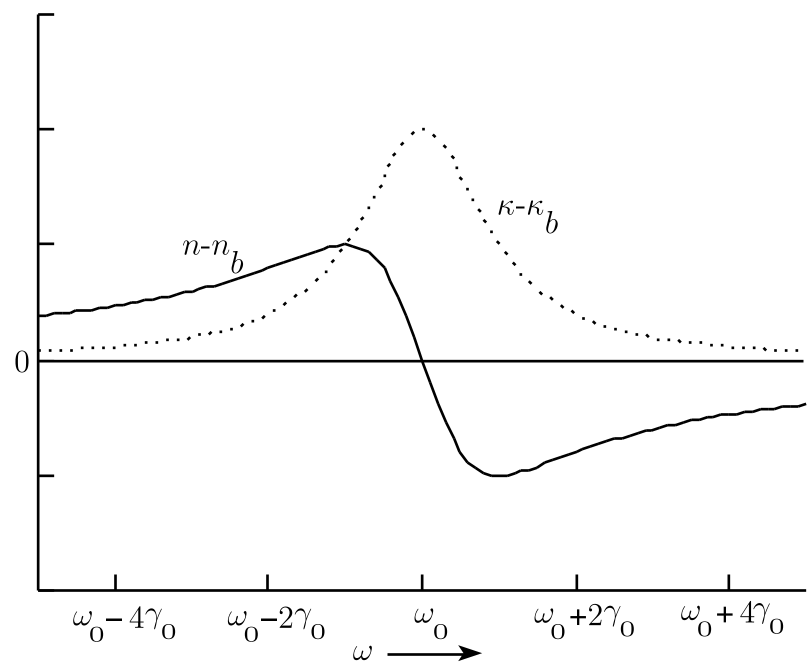

Figure 5.1: Beahvior of n and κ as a function of frequency near a resonance for a homogeneous collection of oscillators.

behavior of n, in contrast to the normal behavior where n is an increasing function of frequency, is termed anomalous. Away from a resonance, n reverts to its normal behavior, viz, that it increases with frequency.

We have considered a simple model of dispersion for an isolated resonance. Clearly, if several resonances overlap, more complicated behavior can result. Other refinements such as the modifications due to thermal motion of atoms are possible and are discussed in the literature [See for example *Lasers* by A. Siegman (University Science Books, Sausalito, CA, 1986)]

5.1 Near Resonant Susceptibility

The atomic sucsepetibility can be written as

$$\chi_{at} = \frac{Nf_0e^2}{m\epsilon_0}\left[\frac{(\omega_0^2-\omega^2)+i2\omega\gamma_0}{(\omega_0^2-\omega^2)^2+4\omega^2\gamma_0^2}\right] \tag{5.23a}$$

For driving field frequencies close to ω_0 such that $|\omega_0-\omega| \ll \omega_0\,,\omega$ and $|\omega_0-\omega| \sim 10\gamma_0$ we can use the approximation $\omega_0^2-\omega^2 = (\omega_0+\omega)(\omega_0-\omega)s \approx 2\omega(\omega_0-\omega)$ and write the real and imaginary part of the suceptibility $\chi_{res} = \chi'_{at} + i\chi''_{at}$ as

$$\begin{aligned}
\chi''_{at} &= \frac{Nf_0e^2}{m\epsilon_0}\left[\frac{2\omega\gamma_0}{(\omega_0^2-\omega^2)^2+4\omega^2\gamma_0^2}\right] \\
&\approx \frac{Nf_0e^2}{2m\epsilon_0\omega}\left[\frac{\gamma_0}{(\omega_0-\omega)^2+\gamma_0^2}\right] \\
&= \frac{Nf_0e^2}{4m\epsilon_0\omega}\left[\frac{1}{\pi}\frac{\gamma_0/2\pi}{(\nu_0-\nu)^2+(\gamma_0/2\pi)^2}\right] \equiv \frac{Nf_0e^2}{4m\epsilon_0\omega}S(\nu) \\
\chi'_{at} &= \frac{Nf_0e^2}{m\epsilon_0}\left[\frac{(\omega_0^2-\omega^2)}{(\omega_0^2-\omega^2)^2+4\omega^2\gamma_0^2}\right] \\
&\approx \frac{Nf_0e^2}{2m\epsilon_0\omega}\left[\frac{(\omega_0-\omega)}{(\omega_0-\omega)^2+\gamma_0^2}\right] && (5.23b) \\
&= \frac{Nf_0e^2}{4m\epsilon_0\omega}\left[\frac{1}{\pi}\frac{(\nu_0-\nu)}{(\nu_0-\nu)^2+(\gamma_0/2\pi)^2}\right] && (5.23c)
\end{aligned}$$

The function $S(\nu)$ is called the line shape function. For a homogeneous medium considered here it is a Lorentzian

$$S(\nu) = \frac{1}{\pi}\frac{\gamma_0/2\pi}{(\nu_0-\nu)^2+(\gamma_0/2\pi)^2} \tag{5.23d}$$

This has a width (FWHM) given by

$$\Delta\nu_o = \frac{\gamma_0}{\pi} \tag{5.24}$$

So that we can write the Lorentzian in the standard form

$$S(\nu) = \frac{1}{\pi}\frac{\Delta\nu_o/2}{(\nu_0-\nu)^2+(\Delta\nu_o/2)^2} \tag{5.25}$$

Then we can write

$$\chi''_{at} = \frac{Nf_0e^2}{4m\epsilon_0\omega}S(\nu) \tag{5.26}$$

$$\chi''_{at} = \frac{2(\nu_0-\nu)}{\Delta\nu_o}\chi''_{at} \tag{5.27}$$

Using the classical expression for the radiative decay rate $2\ \gamma_0 = A_{rad}$ in a medium of refractive index $n = \sqrt{\epsilon/\epsilon_0}$,

$$2\gamma_0 \equiv A_{rad} = \frac{2}{3}\frac{e^2}{4\pi\epsilon}\frac{\omega^2}{mv^3} = \frac{2}{3}\frac{e^2}{4\pi\epsilon_0 n^2}\frac{\omega^2}{m(c/n)^3} = \frac{e^2n\omega^2}{6\pi\epsilon_0 mc^3} \tag{5.28}$$

we can write the equation of suceptibility as

$$\chi''_{at} = \frac{Nf_0A_{rad}6\pi c^3}{4n\omega^3}S(\nu) = \frac{Nf_0A_{rad}3\lambda^3}{16n\pi^2}S(\nu) \tag{5.29a}$$

$$\chi'_{at} = \frac{2(\nu_0-\nu)}{\Delta\nu_o}\chi''_{at} \tag{5.29b}$$

The absorption coefficient is

$$\alpha = \frac{\omega}{nc}\chi''_{at} = \frac{3\lambda^2}{8\pi n^2} N f_0 A_{rad} \tag{5.29c}$$

Quantum mechanically we replace $N f_0$ by the population difference $N_1 - N_2$.

In our model of sucseptibility we have assumed isotropic response of the medium and the electric polarization to match medium polarization. In real atoms, the dielctric resposne has, in general, a tensorial character so that the induced polarization has the form

$$\mathcal{P}_i = \chi_{ij}\mathcal{E}_j \, . \tag{5.30}$$

Then the power absorbed per unit volume by the atoms is

$$\frac{1}{u}\frac{du}{dt} = \frac{1}{2}\omega\chi''_{ij}\underset{\sim}{E}_i^*\underset{\sim}{E}_j \tag{5.31}$$

The scalar product can be evaluated given the atomic response and the polarization of the field. The net result is that the factor of "3" in the expression for the coefficient of absorption can be any number from 0 to 3 depending on the polarization of the field and atomic response tensor [Siegman Sec. 3.5]. Siegman handles this by replacing 3 in Eqs. (5.29b)-(5.29c) For example, if the fields are linearly polarized and the resoponse is random. In this case $3^* = 1$. On the other hand if the field and the induced polarization line up $3^* = 3$.