Lecture 13

Suppose dephasing collisions occur with a <u>rate</u> $\frac{1}{T'_2}$

Then the number of collisions dN_c occurring in a time interval dt is

$$dN_{c} = N_{c} \frac{dt}{T_{2}'} \Longrightarrow dN_{in} = -N_{in} \frac{dt}{T_{2}'}$$

$$\Rightarrow \frac{dN_{in}}{T_{2}'} = -\frac{1}{T_{2}'} N_{in} \Longrightarrow N_{in}(t) = N_{0} e^{-t/T_{2}'}$$

$$\Rightarrow \overline{P(t)} = -Ne \overline{x} e^{-(\frac{\gamma}{2} + \frac{1}{T_{2}'})^{t}} e^{i\omega_{0}t} \equiv -Ne \overline{x} e^{-t/T_{2}} e^{i\omega_{0}t}$$

Therefore the macroscopic polarization decays at a rate

$$\frac{1}{T_2} = \frac{\gamma}{2} + \frac{1}{T_2'}$$

A common notation in the literature defines

$$\frac{1}{T_1} = \gamma = \gamma_{rad} + \gamma_{nr} = \underline{\text{energy}} \text{ decay rate}$$
$$\frac{1}{T_2} = \underline{\text{polarization}} \text{ decay rate}$$

Note that, in the absence of "extra dephasing" such as pure dephasing collisions, one would have

$$T_2 = 2T_1$$

In general, $T_2 < 2T_1$

To recap: $\frac{1}{T_2}$ gives the rate at which the macroscopic polarization decays. In the absence of

dephasing collisions, the polarization decays only because of energy relaxation, at half the rate (since the energy is proportional to the <u>square</u> of the individual dipoles' polarization amplitude). Additional process such as elastic collisions or vibrational coupling may not reduce the amplitude of an individual dipole's oscillation (=> no energy decay), but may randomize its phase (or direction), thereby causing the macroscopic polarization to decay at a <u>faster</u> rate.

Typical numbers

$$\omega_0 \sim 10^{15} \, s^{-1}$$

Atoms in a gas:

$$T_1 = \gamma^{-1} = 10^{-6} to 10^{-7} s$$

$$T_2' = 10^{-8} to 10^{-10} s$$

Atoms in a solid

$$T_1 = 10^{-3} to 10^{-12} s$$
$$T_2 = 10^{-13} to 10^{-16} s$$

Materials show a wide range of possible $T_{
m l}$ and $T_{
m 2}$, but usually (although not

always) , $T_2 \ll T_1 soT_2 \simeq T_2'$

Now we can go back to the Lorentz eqn, for a simple dipole (see P.80)

$$\frac{d^2 \bar{x}}{dt^2} + \gamma \frac{d \bar{x}}{dt} + \omega_0^2 \bar{x} = -\frac{e\bar{E}}{m}$$

The equation of motion for the <u>macroscopic</u> polarization is obtained by multiplying through by -Ne (from $\vec{P} = -Ne\vec{x}$) and putting in the correct form of the damping:

$$\frac{d^{2}\vec{P}}{dt^{2}} + (\gamma + \frac{2}{T_{2}'})\frac{d\vec{P}}{dt} + \omega_{0}^{2}\vec{P} = \frac{Ne^{2}}{m}\vec{E}$$

Now we can solve to find the polarization induced in the material by an applied electromagnetic field.

Steady – State Solution

Assume a simple harmonic incident field:

$$\vec{E}(t) = \hat{\varepsilon} E_0 e^{i\theta}$$

Where

 $\hat{\mathcal{E}}$ = polarization vector

 E_0 = complex amplitude

(as usual ,the real part is understood; E_0 is complex => determines the <u>phase</u> of the field) We therefore <u>expect</u> a <u>solution</u> of the form

$$\dot{P}(t) = \hat{\varepsilon} P_0 e^{i\omega t}$$
 (P_0 also complex; may have a different phase from E_0 !)

$$\frac{d\vec{P}}{dt} = i\omega\vec{P} \qquad \frac{d^2\vec{P}}{dt^2} = -\omega^2\vec{P}$$

⇔

after dividing out $\hat{arepsilon}_0 e^{i\omega t}$ on both sides ,we get the scalar equation for the amplitudes:

$$-\omega^{2}P_{0} + i\omega(\gamma + \frac{2}{T_{2}'})P_{0} + \omega_{0}^{2}P = \frac{Ne^{2}}{m}E_{0}$$

$$P_{0}[(\omega_{0}^{2} - \omega^{2}) + i\omega(\gamma + \frac{2}{T_{2}'})] = \frac{Ne^{2}}{m}E_{0}$$
Or
$$P_{0} = [\frac{Ne^{2}}{m}\frac{1}{(\omega_{0}^{2} - \omega^{2}) + i\omega(\gamma + \frac{2}{T_{2}'})}]E_{0}$$

Note:

- The <u>amplitude</u> of the induced polarization is <u>linearly proportional</u> to the driving field amplitude
- (2) The proportionality constant is <u>complex</u>, so there is a <u>phase shift</u> between the field and polarization <u>due to the damping</u> of the polarization (i.e. finite T_2).

The usual procedure is to define

$$\vec{P} = \varepsilon_0 \chi \vec{E} \text{ or } \chi = \frac{Ne^2}{\varepsilon_0 m} \frac{1}{\left(\omega_0^2 - \omega^2\right) + i\omega(\gamma + \frac{2}{T_2})}$$

Where $\chi =$ <u>electric susceptibility</u> (dimensionless)

(Note: Guenther defines $ec{P}=\chiec{E}$, but this is quite inconvenient +

nonstandard, so we will use the above definition.)

Now we have half the story .We have determined the polarization induced by the field (atomic response to the field), but now we must find <u>self-consistently</u> the effect of that polarization back on the field – i.e. the polarization is a <u>source term</u> in the wave eqn. Thus to understand the propagation of light in matter, we must simultaneously solve the Lorentz eqn. and the wave eqn.

Wave eqn. with source:

As usual, begin with $\nabla \times \vec{E} = -\frac{\partial \vec{B}}{\partial t}$

$$\nabla \times \left(\nabla \times \vec{E} \right) = -\frac{\partial}{\partial t} \left(\nabla \times \vec{B} \right) = -\mu_0 \frac{\partial}{\partial t} \left(\nabla \times \vec{H} \right)$$
$$\nabla \left(\nabla \cdot \vec{E} \right) - \nabla^2 \vec{E} = -\mu_0 \frac{\partial}{\partial t} \left(\frac{\partial \vec{D}}{\partial t} \right) = -\mu_0 \frac{\partial^2}{\partial t^2} \left(\varepsilon_0 \vec{E} + \vec{P} \right)$$
$$\nabla^2 \vec{E} - \mu_0 \varepsilon_0 \frac{\partial^2 \vec{E}}{\partial t^2} = \mu_0 \frac{\partial^2 \vec{P}}{\partial t^2}$$

This shows that a time-dependent polarization acts as a source for electromagnetic waves.

For weak fields (linear response) $\vec{P} = \varepsilon_0 \chi \vec{E}$

For a harmonic driving field $\vec{E} = \vec{\varepsilon}(\vec{r})e^{i\omega t}$ we have the <u>Helmholtz eqn.</u>

$$\nabla^2 \vec{E}(\vec{r}) - \mu_0 \varepsilon_0 (1 + \chi) \omega^2 \vec{E}(\vec{r}) = 0 \qquad \mu_0 \varepsilon_0 = \frac{1}{c^2}$$

The form of this equation is familiar, so we can try plane wave solutions

$$\vec{E} = \hat{\varepsilon} E_0 e^{i(\omega t - \vec{k} \cdot \vec{r})}$$

Plug in =>

$$-k^2 + \frac{\omega^2}{c^2} \left(1 + \chi\right) = 0$$

Or
$$k^2 = \frac{\omega^2}{c^2} (1 + \chi)$$

As usual, one can define a complex index of refraction \tilde{n} so that

$$k^2 = \frac{\tilde{n}^2 \omega^2}{c^2}$$
 where $\boxed{\tilde{n}^2 = 1 + \chi}$

Just as we saw with metals, a complex wavevector (or complex index ,or complete dielectric constant – those are all just different ways of saying the same thing) leads to damping .We will come back to consider absorption in some detail , but first we should take a look at the traditional ,familiar <u>real index of refraction</u> .This is important when light is propagating through nonabsorbing, i.e., transparent media (e.g. glass !)

Consider light at a frequency much lower than the resonant frequency of the dipoles composing the dielectric:

$$\omega \ll \omega_0$$

This is quite reasonable to do, since the relevant resonance frequency for most transparent materials is somewhere in the near <u>ultraviolet</u> (e,g. glass $\omega \sim 1=200$ nm)

Specifically, we consider:

$$\omega_0^2 - \omega^2 \gg \omega(\gamma + \frac{2}{T_2})$$

So the frequency is far from the absorption resonance.

Then

$$\chi(\omega) \simeq \frac{Ne^2}{\varepsilon_0 m} \frac{1}{\omega_0^2 - \omega^2}$$
 which is real !

Thus the index of refraction is

$$n(\omega)^{2} = 1 + \chi(\omega) = 1 + \frac{Ne^{2}}{\varepsilon_{0}m} \frac{1}{(\omega_{0}^{2} - \omega^{2})}$$

The fact that n is a function of frequency means that waves of different frequencies

propagate with different speeds in the medium, which is the phenomenon of <u>dispersion</u>. Important features:

(1)
$$\omega_0^2 \succ \omega^2 \Rightarrow \underline{n > 1}$$

(2) n increases as $\omega(\lambda)$ increases (decreases)

This is the familiar behavior of the "<u>normal dispersion</u>" of transparent materials in the visible and near-IR regions of the spectrum.

In real materials, there will be of course be more than one resonance frequency .That means in general the index will have the form

$$n^2 = 1 + \sum_j \frac{A_j}{\left(\omega_{0j}^2 - \omega^2\right)}$$

Here the strengths A_i (proportional to the "effective number of oscillators") and the

resonance frequencies ω_{0j} are <u>fitting parameters</u>. In fact, very good fits to the index in transparent media are obtained .In fact, this was known <u>long before</u> Lorentz came up with a physical model explaining this formula. Often, it was expressed in the equivalent form

$$n^{2} = 1 + \sum_{j} \frac{B_{j} \lambda^{2}}{\left(\lambda^{2} - \lambda_{j}^{2}\right)}$$

Which is known as Sellmeier's formula

Of course, the classical model cannot predict the resonance frequencies, nor can it predict the amplitudes A (we'll see that further below). Those parameter can in principle only be obtained from quantum theory.

q	Chapter	3 Elec	tromagnet	ic Theory, Photon Dense flint glass	s and Light $(\mathcal{P}S)$ will be yow, in the infrared. Molecules such as H_2O as CO_2 will have resonances in both the infrared as ultraviolet. If water was trapped within a piece of gla
action #					during its manufacture, these molecular oscillato would be available, and an infrared absorption bar would exist. The presence of oxides will also result infrared absorption. Figure 3.28 shows the $n(\omega)$ curv
ndex of refr 				Light flint glass Crystal quartz	for a number of important optical crystals ranging from the straviolet to the infrared. Note now they rise the ultraviolet and fall in the infrared. At the even low from the ultraviolet and fall in the infrared.
				osilicate crown glass × plastic Vitreous quartz	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
1.4	- 50-	400 Wa	600 ¢length λ (n m)	800 1000	plementary color. As a final point, notice that if the driving frequencis greater than any of the ω_{0j} terms, then $n^2 < 1$ and
igure 3. or vario	.26 The way us materials.	velength de	ependence of 1	the index of refraction	n < 1. Such a situation can occur, for example, if w beam x-rays onto a glass plate. This is an intriguin south, since it leads to $v > c$, in seeming contradiction
=			2 7		to special relativity. We will consider this behavior againlater on, when we discuss the group velocity (Section 7.6).
				3 3	$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$
Inf	rared ~ 01	Visible	-02 Iltr±vioio	et X-ray	2.4 Thallium bromoiodide
figure 3	.27 Refract	ive index v	versus freque		Ag Thallium chlorobromide

Resonant Absorption

We have seen that when $\, \varpi \,$ is $\, \underline{\rm far} \,$ from the atomic resonance frequency $\varpi_{\!_0}$, the index of refraction (or susceptibility) is real, and the dispersion is "normal". Now we will consider what happens when ω is <u>near</u> ω_0 .

It will be convenient to define

$$\Delta \omega_0 = \gamma + \frac{2}{T_2'} = \gamma_{rad} + \gamma_{nr} + \frac{2}{T_2'} = \frac{2}{T_2}$$

Then the susceptibility can be written (from P.90)

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

Note that in general $\Delta \omega_0 \ll \omega_0$. (Only in a few special cases, such as some organic dyes, and some solid-state laser materials such as T_1 : sapphire, does $\Delta \omega_0$ becomes as large as ~10% * ω_0 .In dilute gases, it is 10^{+3} * smaller or even less)

<u>Resonance approximation</u>: $\omega \simeq \omega_0$

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

It is useful to split this into real and imaginary parts:

$$\chi = \chi' + i\chi'' = \frac{Ne^2}{2\varepsilon_0 m\omega_0} \frac{(\omega_0 - \omega) - i\frac{\Delta\omega_0}{2}}{(\omega_0 - \omega)^2 + (\frac{\Delta\omega_0}{2})^2} = \frac{Ne^2}{\varepsilon_0 m\omega_0 \Delta\omega_0} \left[\frac{(\omega_0 - \omega)/2\Delta\omega_0}{(\omega_0 - \omega)^2 + (\frac{\Delta\omega_0}{2})^2} - i\frac{(\frac{\Delta\omega_0}{2})^2}{(\omega_0 - \omega)^2 + (\frac{\Delta\omega_0}{2})^2}\right]$$

Define $\chi_0'' = \frac{Ne^2}{\varepsilon_0 m\omega_0 \Delta\omega_0}$

In laser physics, it is convenient to write this as (exercise)

$$\chi_0^{\prime\prime} = \frac{3}{4\pi^2} \frac{N\lambda^3 \gamma_{rad}}{\Delta \omega_0}$$

This is useful since

 $N\lambda^3$ = number of oscillators in a cubic wavelength

 $\gamma_{\it rad}$ = a measure of the strength of the interaction of light with an atomic resonance

(one atom)

 $\Delta \omega_0$ = "width" of resonance, as we'll see below

Connection to real atomic transitions: Recall (P.76)

$$\gamma_{rad} = \frac{e^2 \omega_0^2}{6\pi \varepsilon_0 m c^3}$$

Is the radiative decay rate for a classical dipole oscillator? <u>Real</u> atomic transitions show radiative decay rates which may vary over orders of magnitude .This is taken into account in the classical model by writing

$$\gamma_{rad} = \frac{e^2 f \,\omega_0^2}{6\pi\varepsilon_0 mc^3}$$

Where f is called the oscillator strength of the transition.

In the classical model, f should be considered a <u>"fitting</u> parameter" that can be used to fit the strength of a real resonance. It turns out (you'll consider this in some detail in "optical wave in crystal" course) that f can be calculated from first principles in the quantum theory.

Note: large oscillator strength \leftrightarrow fast γ_{rad}

$$\leftrightarrow$$
 large χ_0''

Recall:

 $\Delta \omega_0 = \gamma_{rad} + \gamma_{nr} + \frac{2}{T'_2}$

Suppose there is no nonradiative (collisional) damping of the polarization:

$$\gamma_{nr}=0,\frac{2}{T_2'}=0$$

 $\chi_0'' = \frac{3}{4\pi^2} N\lambda^3$

 $\Rightarrow \Delta arphi_0 = \gamma_{rad}$ (pure radiative damping)

Then

i.e the only thing that matters is the number of oscillators (atoms) in a cubic wavelength!

For convenience, define a dimensionless quantity (normalized detuning)

$$\Delta y = 2 \frac{\omega - \omega_0}{\Delta \omega_0}$$

Then the susceptibility can be written => $\Delta \omega_0$ -FWHM

$$\chi = \chi' + i\chi'' = -\chi_0'' \left[\frac{\Delta y}{1 + (\Delta y)^2} + i \frac{1}{1 + (\Delta y)^2} \right]$$

 $\frac{\Delta y}{1+\left(\Delta y\right)^2}$: "dispersive line shape" we shall see that this is responsible for

dispersion, or phase shifts .

$$irac{1}{1+\left(\Delta y
ight)^2}$$
 : "Lorentzian line shape "FWHM = $\gamma+rac{2}{T_2'}=\Delta arphi_0$.We shall see that

this part is responsible for <u>absorption</u>. --see Siegman fig.2.8 (next page)

Remember that this form of χ is an <u>approximation</u>, valid when the light frequency ω is <u>close</u> to the resonance frequency ω_0



Recall the connection to the dielectric constant :

$$\tilde{n}^2 = \varepsilon_r = 1 + \chi(\varepsilon = \varepsilon_r \varepsilon_0)$$

 χ is complex => ε is complex.

We have seen before that a <u>complex</u> dielectric constant gives rise to optical <u>absorption</u>.We could carry out exactly the same steps as in pp.44-46 to define a real index of refraction and <u>extraction</u> <u>coefficient</u> which will describe the <u>exponential decay</u> of the wave as it propagates in the medium .(this is Guenther's approach ,)

Rather than repeat that formulation of the problem, however, we will take an alternative

approach which is particularly useful in laser theory. We begin by writing the wave equation

$$\nabla^2 \tilde{E} - \frac{1}{c^2} \left(1 + \chi \right) \frac{\partial^2 \tilde{E}}{\partial t^2} = 0$$

Where we have put a tilda over the \tilde{E} to remind ourselves it is the <u>total complex field</u>, and to distinguish it from a new expression of the field below.

To simplify the problem, let's consider only a plane wave propagating in the z direction.

We know from solving the wave eqn, before that a time-harmonic wave gives an $\frac{\omega^2}{c^2}$ term and

hence the Helmholtz eqn, in this spirit, let's write the field in a new way

$$\tilde{E} = \varepsilon(z)e^{i(\omega t - \beta z)}$$
 [$\varepsilon(z)$ = "envelope fn" \rightarrow can be complex]

$$\frac{\partial^2 E}{\partial t^2} = -\omega^2 \tilde{E} \text{ as usual}$$
$$\frac{\partial^2 \tilde{E}}{\partial t^2} = \left(\frac{\partial^2 \varepsilon}{\partial z^2} - 2i\beta \frac{\partial \varepsilon}{\partial z} - \beta^2 \varepsilon\right) e^{i(\omega t - \beta z)}$$

Now let us <u>assume</u> the z-dependence is slow:

$$\frac{\partial^2 \varepsilon}{\partial z^2} \ll 2\beta \left| \frac{\partial \varepsilon}{\partial z} \right|$$

If we do so, the wave eqn, becomes

$$-2i\beta\frac{\partial\varepsilon}{\partial z} - \beta^{2}\varepsilon + \frac{\omega^{2}}{c^{2}}(1+\chi)\varepsilon = 0$$

Now we make the assignment $\beta = \frac{\omega}{c}$. (In other words, it's the real wavevector; we didn't use k

since that would be inconsistent with our previous notation, in which k is allowed to be complex . Also, $\beta = \frac{\omega}{c}$ is a common notation in optical physics – e.g. as in Siegman ,)

Then
$$\frac{\partial \varepsilon}{\partial z} = -\frac{i\beta}{2}\chi\varepsilon$$

Which describes the propagation of the "slowly varying envelope " $arepsilon \left(z
ight)$