Begin 01-23-2025

Light-Matter Interaction

Classical Theory of Light-Matter Interaction

Self-consistent, fully classical description

Electromagnetic Field $\longrightarrow$ Atom/Molecule/Solid

Motivation: We will

- Develop Concepts $\alpha(\omega),\ n,\ \chi$
- Develop Intuition
- Classical is often adequate, sometimes accurate
- A Quantum Theory has classical limits $\Rightarrow$ Identify/understand regime of validity
- The Classical description is a useful starting point for Nonlinear Optics

The Electromagnetic Field: Basic Eqs. in SI Units

Maxwell's eqs.

(no free charges, currents $\Rightarrow$ dielectrics)

(i) $\nabla\cdot\vec{D} = \rho = 0$ $\quad$ $\vec{D}$: Dielectric displacement

(ii) $\nabla\cdot\vec{B} = 0$ $\quad$ $\vec{B}$: Magnetic induction

(iii) $\nabla\times\vec{E} = -\frac{\partial\vec{B}}{\partial t}$ $\quad$ $\vec{E}$: Electric field

(iv) $\nabla\times\vec{H} = \frac{\partial\vec{D}}{\partial t} + \vec{J}$ $\quad$ $\vec{H}$: Magnetic field

Material Response:

(v) $\vec{B} = \mu_0\vec{H} + \vec{M}$ $\quad\leftarrow$ Non-magnetic $\Rightarrow$ $\vec{M} = 0$

(vi) $\vec{D} = \varepsilon_0\vec{E} + \vec{P}$ $\quad\leftarrow$ Info about response in dipole moment density (polarization density)

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We need equations that describe:

- the behavior of $\vec{E}$ for given $\vec{P}$
- the medium response $\vec{P}$ for given $\vec{E}$

Wave Equation:

Take curl of (iii), then use (iv)

$$\nabla \times (\nabla \times \vec{E}) = -\nabla \times \frac{\partial \vec{B}}{\partial t} = -\frac{\partial}{\partial t}(\nabla \times \vec{B}) = -\mu_0 \frac{\partial^2 \vec{D}}{\partial t^2}$$

Next, use the identity

$$\nabla \times (\nabla \times \vec{E}) = \nabla(\nabla \cdot \vec{E}) - \nabla^2 \vec{E}$$

to obtain $\vec{D} = \nabla(\nabla \cdot \vec{E}) = -\mu_0 \frac{\partial^2 \vec{D}}{\partial t^2}$

Finally, let $\vec{D} = \varepsilon_0 \vec{E} + \vec{P}$ and use $\varepsilon_0 \mu_0 = \frac{1}{c^2}$

to obtain

$$\boxed{-\nabla(\nabla \cdot \vec{E}) + \nabla^2 \vec{E} = \frac{1}{c^2} \frac{\partial^2 \vec{E}}{\partial t^2} + \frac{1}{\varepsilon_0 c^2} \frac{\partial^2 \vec{P}}{\partial t^2}}$$

This is the Wave Equation, still exact in this form

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Transverse Fields

Definition: a field for which $\nabla\cdot\vec{E} \equiv 0$ is **Transverse**

Example: a plane wave, $\vec{E}(\vec{r},t) = \vec{E}(t)\,e^{i\vec{k}\cdot\vec{r}}$,

where $\vec{E}(t)\perp\vec{k}$, is transverse.

The physical field is $Re[\vec{E}(\vec{r},t)]$

For transverse fields the wave equation simplifies to

$$\boxed{\nabla^2\vec{E} - \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\vec{E} = \frac{1}{\varepsilon_0 c^2}\frac{\partial^2}{\partial t^2}\vec{P}}$$

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Isotropic Media

Absent a preferred direction, the induced $\vec{P}$ must be parallel to the driving field $\vec{E}$

Linear response, most general case:

$$\vec{D}(t) = \varepsilon_0 \vec{E}(t) + \vec{P}(t)$$
$$= \varepsilon_0 \vec{E}(t) + \varepsilon_0 \int_{-\infty}^{t} dt' R(t-t') \vec{E}(t')$$

where the *response function* $R(t-t')$ is a scalar and we have $R(\tau) = 0$ for $\tau < 0$

Take $\nabla\cdot$ on both sides, divide by ε_0 & use M.E. (i)

$$\nabla\cdot\vec{D}(t) = \varepsilon_0 \nabla\cdot\vec{E}(t) + \varepsilon_0 \int_{-\infty}^{t} dt' R(t-t') \nabla\cdot\vec{E}(t') = 0$$

$$\Rightarrow \nabla\cdot\vec{E}(t) = -\int_{-\infty}^{t} dt' R(t-t') \nabla\cdot\vec{E}(t') \quad \text{for all } t$$

It follows that $\nabla\cdot\vec{E}(t) = 0$ (transverse) for all t

<u>OR</u> $\qquad R(\tau) = -2\delta(\tau)$

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<u>OR</u> $R(\tau) = -2\delta(\tau)$

Note: if $R(\tau) \propto \delta(\tau)$ (instantaneous response) then

$$\varepsilon_0 \int_{-\infty}^{t} dt' R(t-t') \vec{E}(t') = \varepsilon_0 \chi \vec{E}(t)$$

susceptibility

The case $R(\tau) = -2\delta(\tau)$ is an example of negative susceptibility, $\chi<0$, which only occurs in certain engineered metamaterials.

Electric fields are transverse in linear, isotropic dielectric media

(including the vacuum)

Wave Equation in free space

$$\boxed{\nabla^2\vec{E} - \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\vec{E} = 0}$$

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Monochromatic trial solution $\vec{E}(\vec{r},t) = \vec{E}_0(\vec{r})e^{-i\omega t}$

$$\nabla^2 \vec{E}_0(\vec{r})e^{-i\omega t} + \frac{\omega^2}{c^2}\vec{E}_0(\vec{r})e^{-i\omega t} = 0$$

Equation for the spatial component alone:

$$\nabla^2 \vec{E}_0(\vec{r}) + |\vec{k}|^2 \vec{E}_0(\vec{r}) = 0\ , \ |\vec{k}| = \omega/c$$

Plane wave solutions

$$\boxed{\vec{E}_0(\vec{r}) = \vec{\varepsilon} E_0 e^{i\vec{k}\cdot\vec{r}}\ , \ |\vec{k}| = \omega/c}$$

<u>Optical Cavities</u>: Here we need to solve the wave equation subject to boundary conditions. See, e. g., Millony & Eberly for examples such as rectangular cavities, Fabryt-Perot etalons, and spherical mirror resonators.

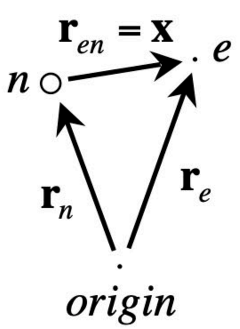
Light-Matter Interaction

Theory of Atomic Response

So far, we have a model for the field. Next, we need a model of how the constituents of the medium responds to the field.

This will allow us to find the polarization density $\vec{P}$ as function of the field $\vec{E}$

Classical "atom"



Simple model:
nucleus + electron

Lorentz Force

$$\boxed{\vec{F} = q(\vec{E} + \vec{v} \times \vec{B})}$$

~ 0 **if non-relativistic**

Newton:

$$\text{(i)} \quad m_n \frac{d^2}{dt^2}\vec{r}_n(t) = -e\vec{E}(\vec{r}_n, t) - \vec{F}_{en}(\vec{r}_{en}, t)$$

$$\text{(ii)} \quad m_e \frac{d^2}{dt^2}\vec{r}_e(t) = e\vec{E}(\vec{r}_e, t) + \vec{F}_{en}(\vec{r}_{en}, t)$$

This is a standard 2-body problem which we can re-cast as in terms of relative and COM motion.

We define:

$$\vec{x} = \vec{r}_{en} = \vec{r}_e - \vec{r}_n \qquad m = \frac{m_e m_n}{m_e + m_n} \sim m_e$$

$$\vec{R} = \frac{m_e \vec{r}_e + m_n \vec{r}_n}{M} \qquad M = m_e + m_n \sim m_n$$

$\vec{x}$ **Relative coord.** $\qquad$ m **Reduced mass**

$\vec{R}$ **Center-of-mass** $\qquad$ M **Total mass**

Light-Matter Interaction

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Sub into (i), (ii) and rewrite:

$$M\frac{d^2}{dt^2}\vec{R} = e\left[\vec{E}\left(\vec{R} + \frac{m_n}{M}\vec{x}, t\right) - \vec{E}\left(\vec{R} - \frac{m_e}{M}\vec{x}, t\right)\right]$$

$$m\frac{d^2}{dt^2}\vec{x} = \frac{e}{2}\left[\vec{E}\left(\vec{R} + \frac{m_n}{M}\vec{x}, t\right) + \vec{E}\left(\vec{R} - \frac{m_e}{M}\vec{x}, t\right)\right] + \vec{F}_{en}(\vec{x}) + \frac{1}{2}(m_n - m_e)\frac{d^2}{dt^2}\vec{R}$$

Basic result, no approximations !

Milloni & Eberly, main text ➡ **Set** $\vec{R} \approx \vec{r}_n$, $\vec{x} \approx \vec{r}_{en}$

Throw away eq. for $\vec{R}$

Electric Dipole approximation

Atomic dimensions		**Optical Wavelength**
$\lvert\vec{x}\rvert \sim 1\text{Å}$	$\ll$	$\lambda \sim 10^4\text{Å}$

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<u>EDA</u>: the field is <u>nearly constant</u> on the scale of an atom

Good approximation: 1st order expansion in $\vec{x}$

$$\vec{E}\left(\vec{R}-\frac{m_e}{M}\vec{x},t\right)\approx\vec{E}(\vec{R},t)-\frac{m_e}{M}(\vec{x}\cdot\nabla)\vec{E}(\vec{R}(t))$$

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Light-Matter Interaction

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Physical Interpretation:

$\vec{p} = e\vec{x}$: **electric dipole moment of the atom**

The Eqs. Of motion can then be recast as

$$M\frac{d^2}{dt^2}\vec{R} \approx (\vec{p}\cdot\nabla)\vec{E}(\vec{R},t) = \vec{F} = -\nabla_R V(\vec{x},\vec{R},t)$$

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where $V(\vec{x},\vec{R},t) = -\vec{p}\cdot\vec{E}(\vec{R},t)$

electric-dipole interaction

Note: The COM Eq. is the foundation for a range of laser Atom Traps and Optical Tweezers. We will not explore this further in OPTI 544 lectures, but good review articles can be found in the published literature.

Light-Matter Interaction

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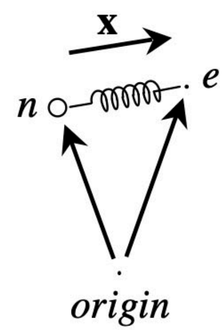
The Electron Oscillator/Lorentz Oscillator

Simple model w/a harmonically bound electron:

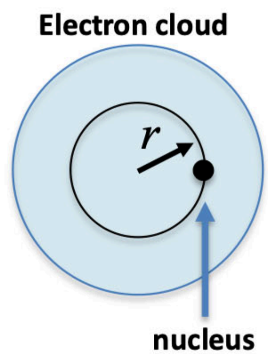


$\vec{F}_{en} = -m\omega_0^2\vec{x}$

resonance frequency

This is meant as a model of the atomic response, not a model of the atom itself.

Nevertheless: QM suggest the atom consists of a point-like nucleus and a spherical electron cloud



Force from charge inside r as if entire charge was at the center

Force from charge outside r is zero

Force $F \propto \frac{r^3}{r^2} \propto r$

harmonic restoring force

Light-Matter Interaction

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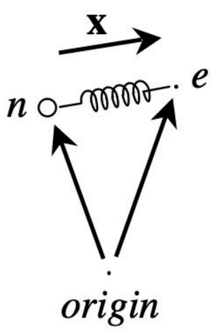

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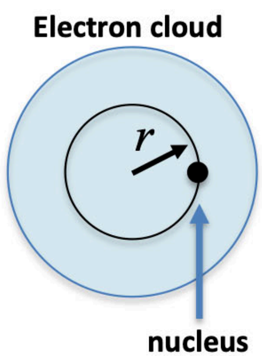

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Force $F \propto \frac{r^3}{r^2} \propto r$

harmonic restoring force

Now substitute $\vec{F}_{en} = -m\omega_0^2 \vec{x}$ into eq. for $\vec{x}$

$$\boxed{\frac{d^2}{dt^2}\vec{x} + \omega_0^2 \vec{x} = \frac{e}{m}\vec{E}(\vec{R},t)}$$

Combine with $\vec{P} = N\vec{p}$, $\vec{p} = e\vec{x}$ where N is the number density of atoms. This relates the macroscopic $\vec{P}$ to the microscopic $\vec{x}$

We now have

Maxwell's Equations
The Lorentz model

Maxwell-Lorentz Equations
We can seek self-consistent solutions to wave propagation

Light-Matter Interaction

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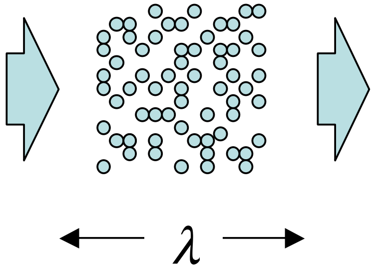
Classical Model of Absorption & Dispersion

Maxwell's Eqs: Oscillating dipole loses energy

Must include damping in Eq, of Motion

<u>Note</u>: In perfectly homogeneous media the coherently scattered light from a collection of Lorentz oscillators interferes constructively only in the forward direction



No energy loss for a propagating fields
(See note set "Classical Light-Matter")

QM to the rescue: Part of the radiation from quantum mechanical atoms is incoherent.

For now we add damping "by hand"

Light-Matter Interaction

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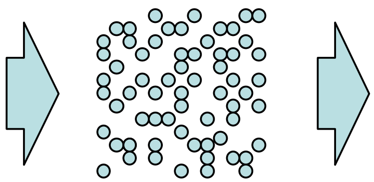

$\longleftarrow \; \lambda \; \longrightarrow$

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The Lorentz Model with Damping

We add an ad hoc friction term w/ $\beta \ll \omega_0$

damping rate

This gives us our basic equation for the atomic response:

$$\boxed{\frac{d^2}{dt^2}\vec{x} + 2\beta\frac{d}{dt}\vec{x} + \omega_0^2\vec{x} = \frac{e}{m}\vec{E}(\vec{R},t)}$$

This type of differential equation generally has both oscillating and decaying terms. Solutions without source terms generally decay as $e^{-\beta t}$

➡ We adopt a trial solution

Driving Field $\quad E(\vec{R},t) = \vec{\varepsilon} E_0 e^{-i(\omega t - kz)}$

Response $\quad \vec{x}(\vec{R},t) = \vec{a}\, e^{-i(\omega t - kz)}$

complex amplitude

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complex amplitude

Solution for $\vec{a}$: $\boxed{\vec{a} = -\vec{\varepsilon}\frac{(e/m)E_0}{\omega^2 - \omega_0^2 + 2i\beta\omega}}$

Physical Quantities:

Field

$$Re[\vec{E}(\vec{R},t)] = \vec{\varepsilon}E_0\cos(\omega t)$$

Dipole ($\vec{\varepsilon}$ real)

$$Re[\vec{p}(\vec{R},t)] = Re[e\vec{x}(\vec{R},t)]$$

$$= \vec{\varepsilon}E_0\frac{e^2}{m}\frac{(\omega_0^2-\omega^2)\cos(\omega t - kz) + 2\beta\omega\sin(\omega t - kz)}{(\omega_0^2-\omega^2) + 4\beta^2\omega^2}$$

Note: $\vec{p}$ and $\vec{E}$ generally oscillate out of phase

$\omega \ll \omega_0$ ➡ $\vec{p}$ & $\vec{E}$ in-phase

$\omega = \omega_0$ ➡ $\vec{p}$ lags $\vec{E}$ by $\pi/2$

$\omega \gg \omega_0$ ➡ $\vec{p}$ Lags $\vec{E}$ by π

Best to stick with complex notation !

Light-Matter Interaction

Solution for $\vec{a}$:

$$\boxed{\vec{a} = -\vec{\varepsilon}\,\frac{(e/m)\,E_0}{\omega^2-\omega_0^2+2i\beta\omega}}$$

Physical Quantities:

Field

$$Re\left[\vec{E}(\vec{R},t)\right] = \vec{\varepsilon}E_0\cos(\omega t)$$

Dipole ($\vec{\varepsilon}$ real)

$$Re\left[\vec{p}(\vec{R},t)\right] = Re\left[e\vec{x}(\vec{R},t)\right]$$

$$= \vec{\varepsilon}E_0\frac{e^2}{m}\frac{(\omega_0^2-\omega^2)\cos(\omega t-kz)+2\beta\omega\sin(\omega t-kz)}{(\omega_0^2-\omega^2)+4\beta^2\omega^2}$$

Note: $\vec{p}$ and $\vec{E}$ generally oscillate out of phase

$\omega \ll \omega_0$ ➡ $\vec{p}$ & $\vec{E}$ in-phase

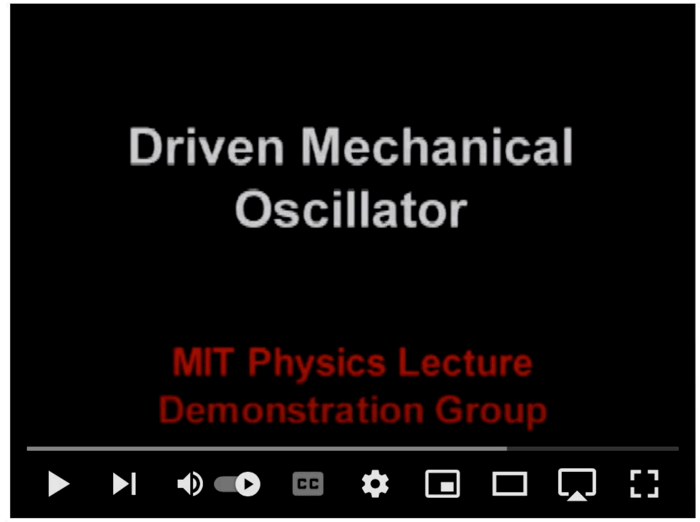
$\omega = \omega_0$ ➡ $\vec{p}$ lags $\vec{E}$ by $\pi/2$

$\omega \gg \omega_0$ ➡ $\vec{p}$ Lags $\vec{E}$ by π

Best to stick with complex notation !

Video of driven – damped harmonic oscillator

https://www.youtube.com/watch?v=aZNnwQ8HJHU



Complex polarizability:

$$\vec{p} = e\vec{x} = e\vec{a}e^{-i(\omega t-kz)} \equiv \alpha(\omega)\vec{\varepsilon}E_0e^{-i(\omega t-kz)}$$

$$\alpha(\omega) = \frac{e^2/m}{\omega_0^2-\omega^2-2i\beta\omega} = \frac{e^2}{m}\frac{\omega_0^2-\omega^2+2i\beta\omega}{(\omega_0^2-\omega^2)^2+4\beta^2\omega^2}$$

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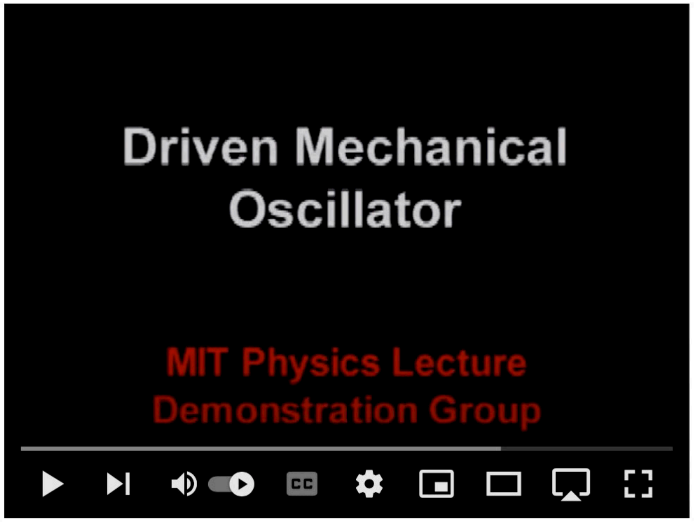

Complex polarizability:

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Easy to show that if $\vec{E}(\vec{R},t) = \vec{\varepsilon}E_0 e^{-i(\omega t-kz)}$

and $\vec{P} = N\vec{p}$

then the wave equation reduces to

$$\left(-k^2+\frac{\omega^2}{c^2}\right)\vec{\varepsilon}E_0 e^{-i(\omega t-kz)} = -\frac{\omega^2}{c^2}\frac{N\alpha(\omega)}{\varepsilon_0}\vec{\varepsilon}E_0 e^{-i(\omega t-kz)}$$

➡ **plane wave solutions with** $k = n(\omega)\omega/c$ **where**

$$\boxed{k^2 = \frac{\omega^2}{c^2}\left[1+\frac{N\alpha(\omega)}{\varepsilon_0}\right] \equiv \frac{\omega^2}{c^2}n(\omega)}$$

Complex index of refraction

Light-Matter Interaction

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Complex index of refraction

Complex <u>Index of Refraction</u> – Physical discussion

Let $\boxed{n(\omega) = n_R(\omega) + i n_i(\omega)}$

Plane wave propagation $k = n(\omega)\omega/c$

$$\vec{E}(z,t) = \vec{\varepsilon} E_0 e^{-i(\omega t - kz)}$$
$$= \vec{\varepsilon} E_0 e^{-i(\omega t - [n(\omega)\omega/c]z)}$$
$$= \vec{\varepsilon} E_0 e^{-n_i(\omega)\omega z/c} e^{-i\omega(t - n_R(\omega)z/c)}$$

We can now identify

$\frac{c}{\omega n_i(\omega)}$ ←	attenuation length
$\frac{c}{n_R(\omega)}$ ←	phase velocity

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Absorption

The intensity of a plane wave field E is

$$I_\omega(z) = \frac{1}{2} n_R(\omega) c \varepsilon_0 |E(c,z)|^2 = I_0(0) e^{-2n_i(\omega)\omega z/c}$$
$$\equiv I_0 e^{-a(\omega) z}$$

where the absorption coefficient is

$$a(\omega) \equiv 2 n_I(\omega) \omega/c = \frac{2\omega}{c} \mathrm{Im}\left[\left(1 + \frac{N\alpha(\omega)}{\varepsilon_0}\right)^{1/2}\right]$$

Possibility of gain ?

No – there is no energy source !

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Absorption and Dispersion in Gases

Approximations:

$\lvert\omega_0-\omega\rvert \ll \omega_0, \omega$	near resonance
$\lvert n(\omega)\rvert \sim 1$	weakly polarizable

Let $\omega_0^2-\omega^2=(\omega_0+\omega)(\omega_0-\omega)\approx 2\omega(\omega_0-\omega)$

$$\alpha(\omega)=\frac{e^2/m}{\omega_0^2-\omega^2-2i\beta\omega}=\frac{e^2/2m\omega}{\omega_0-\omega-i\beta}$$

$$=\frac{e^2}{2m\omega}\,\frac{\omega_0-\omega+i\beta}{(\omega_0-\omega)^2+\beta^2}$$

Furthermore

$$n(\omega)^2=1+\frac{N\alpha(\omega)}{\varepsilon_0}=1+\varepsilon,\ \varepsilon\ll 1$$

Expand to 1st order $(1+\varepsilon)^{1/2}\approx 1+\varepsilon/2$

Putting it together

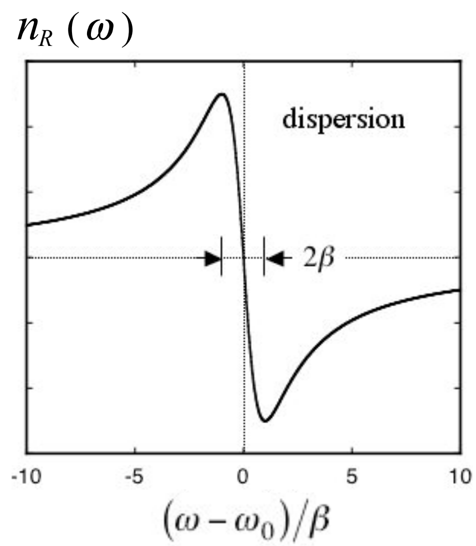
$$n_R(\omega)=1+\frac{Ne^2}{4\varepsilon_0 m\omega}\,\frac{\omega_0-\omega}{(\omega_0-\omega)^2+\beta^2}$$

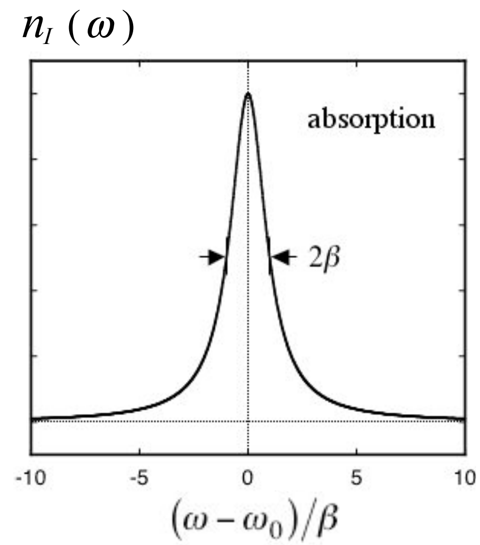
dispersive line shape

$$n_I(\omega)=\frac{Ne^2}{4\varepsilon_0 m\omega}\,\frac{\beta}{(\omega_0-\omega)^2+\beta^2}$$

Lorentzian line shape

General behavior:





Light-Matter Interaction

Putting it together

$$n_R(\omega) = 1 + \frac{Ne^2}{4\varepsilon_0 m\omega} \frac{\omega_0-\omega}{(\omega_0-\omega)^2+\beta^2}$$

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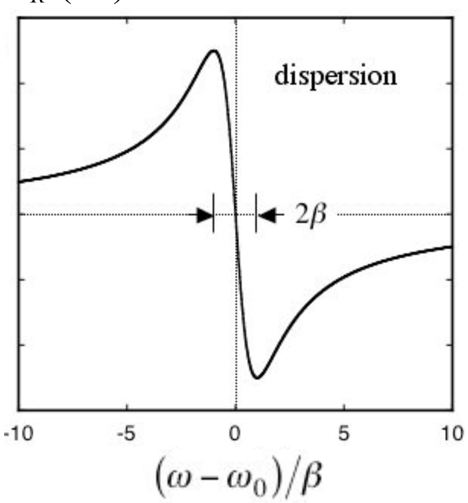

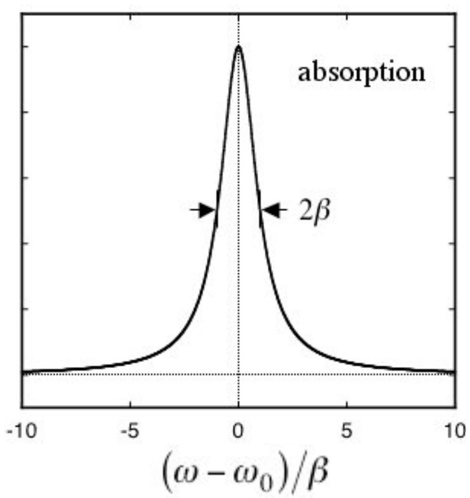

Note: for $|\omega_0-\omega| \gg \beta$

dispersion $\propto \dfrac{1}{(\omega_0-\omega)}$

absorption $\propto \dfrac{1}{(\omega_0-\omega)^2}$

➡ We can have loss-less dispersive media

Note: If we introduce the detuning $\Delta = (\omega_0-\omega)$ we can rewrite $n_R(\omega)$, $n_I(\omega)$ as

$$n_R(\Delta) = 1 + \frac{Ne^2}{4\varepsilon_0 m\omega} \frac{\Delta}{\Delta^2+\beta^2}$$

$$n_I(\Delta) = \frac{Ne^2}{4\varepsilon_0 m\omega} \frac{\beta}{\Delta^2+\beta^2}$$

From the above we see that

$n_R(\omega) < 1$ for $\omega > \omega_0$ ➡ $\dfrac{c}{n_R(\omega)} \gg c$

Superluminal propagation ?

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Superluminal propagation?

Free Electrons

Consider the limit $\omega \gg \omega_0$

➡ effectively unbound electrons

This is a reasonable model of plasmas & metals

In this limit we have

$$\alpha(\omega) = \frac{e^2/m}{\omega_0^2-\omega^2-2i\beta\omega} \approx -\frac{e^2}{m\omega} \Rightarrow$$

$$n(\omega) = \sqrt{1+\frac{N\langle\alpha\rangle}{\varepsilon_0}} \approx \sqrt{1-\frac{Ne^2}{\varepsilon_0 m\omega^2}} \equiv \sqrt{1-\frac{\omega_p^2}{\omega^2}}$$

We introduce the <u>Plasma Frequency</u>:

$$\boxed{\omega_p = \sqrt{\frac{Ne^2}{\varepsilon_0 m}}}$$

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$$n(\omega) = \sqrt{1 + \frac{N\alpha}{\varepsilon_0}} \approx \sqrt{1 - \frac{Ne^2}{\varepsilon_0 m \omega^2}} \equiv \sqrt{1 - \frac{\omega_p^2}{\omega^2}}$$

We introduce the **Plasma Frequency**:

$$\boxed{\omega_p = \sqrt{\frac{Ne^2}{\varepsilon_0 m}}}$$

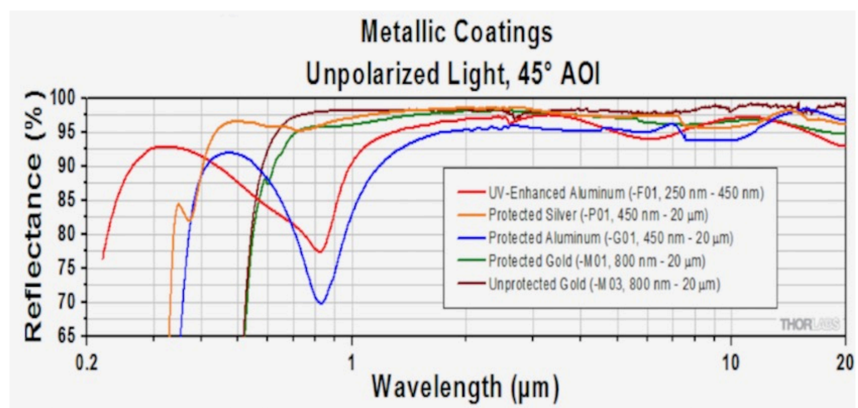
Let $\left.\begin{array}{l}\omega_0 \ll \omega \ll \omega_p \\ |\omega_0 - \omega| \gg \beta\end{array}\right\}$ ⇒ $n(\omega)$ purely imaginary - but no loss!

We now have

$$\vec{E}(z,t) = \vec{\varepsilon} E_0 e^{-i\omega[t - n(\omega)z/c]}$$
$$= \vec{\varepsilon} E_0 e^{-i\omega t} e^{i(z/c)\sqrt{\omega^2 - \omega_p^2}}$$
$$= \vec{\varepsilon} E_0 e^{-i\omega t} e^{-b(\omega)z}$$

where $b(\omega) = -\frac{i}{c}\sqrt{\omega^2 - \omega_p^2}$

Reflection at surface, penetration depth $\sim 1/b(\omega)$



Examples of this kind of medium includes plasmas, and metals such as aluminum, silver and gold which are known to be excellent mirrors for visible and IR radiation.