

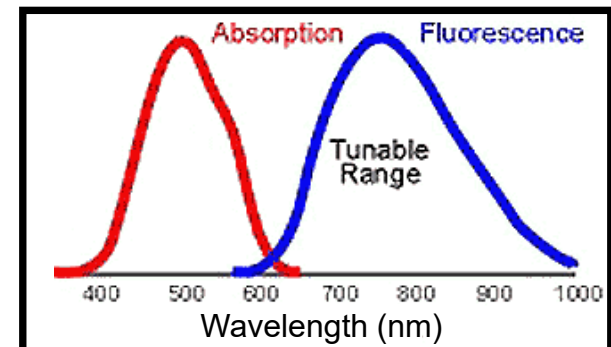
3. Spectral line shapes and how to model them

Some properties of our favorite gain medium: Ti:sapphire
excited state lifetime (radiative, non-radiative)
fluorescence line width

Classical electron oscillator model for spectral line shapes

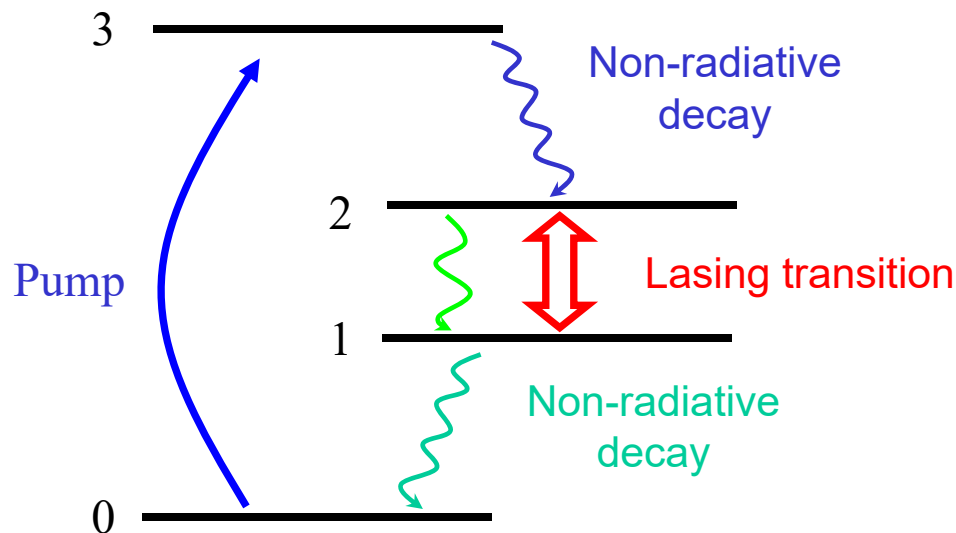
The connection between $\epsilon(\omega)$ and experimentally
measurable parameters

Absorption (or gain) cross-section



A more careful analysis of the 4 level system

Here, we include the lifetimes of the upper and lower lasing levels.



Steps 1 and 2:

Combine to give an *effective* pumping rate for level 2: R_p

Step 3:

- stimulated transitions due to I_{laser}
 - spontaneous decay rate: γ_{21}
- reminder: this is the Einstein A coefficient

Step 4:

- spontaneous decay rate: γ_{10}

$$\frac{dN_2}{dt} = R_p - \gamma_{21}N_2 + BI_{laser}(N_1 - N_2)$$

$$\frac{dN_1}{dt} = \gamma_{21}N_2 - BI_{laser}(N_1 - N_2) - \gamma_{10}N_1$$

$$\frac{dN_0}{dt} = \gamma_{10}N_1 - R_p$$

The four-level model

Recalling that $I_{sat} = A/B$, we find the steady-state solution:

$$\Delta N = N_1 - N_2 = R_p \left[\frac{\gamma_{21} - \gamma_{10}}{\gamma_{10}\gamma_{21}} \right] \left(\frac{1}{1 + I_{laser}/I_{sat}} \right)$$

Key result:

Population inversion (i.e., $\Delta N < 0$) is assured if $\gamma_{10} > \gamma_{21}$
(no matter what I_{laser} is, and even if R_p is small)

This relation between lifetimes is therefore a
necessary condition for lasing
(the lower lasing level has to empty out faster than the upper one)

Of course, there are other necessary conditions, e.g.:

- a resonant cavity - provides feedback and defines modes
- net gain per round trip > net loss per round trip (threshold)

Steady-state population inversion

This expression allows us to define ΔN_0 :

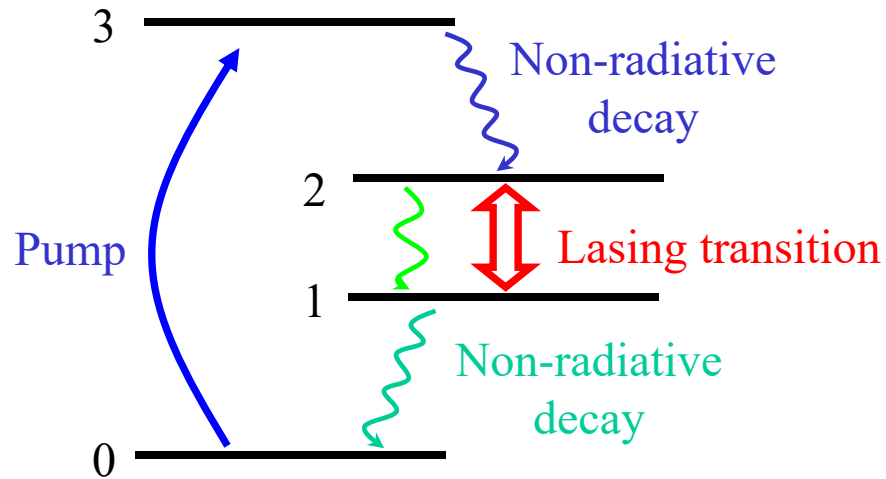
$$\begin{aligned}\Delta N = N_1 - N_2 &= R_P \left[\frac{\gamma_{21} - \gamma_{10}}{\gamma_{10}\gamma_{21}} \right] \left(\frac{1}{1 + I_{laser}/I_{sat}} \right) \\ &= \Delta N_0 \left(\frac{1}{1 + I_{laser}/I_{sat}} \right)\end{aligned}$$

which is the population inversion that is achieved in the case where the laser intensity is zero (below threshold), or is positive but much smaller than the saturation intensity (i.e., when the laser is operating in the linear regime)

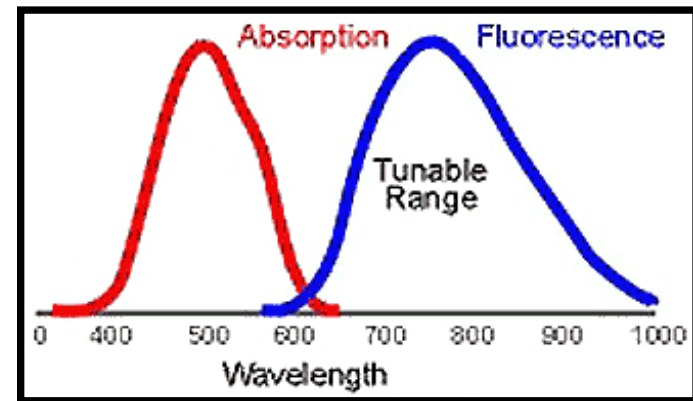
Note that it is directly proportional to the pump rate R_p .

And it is always negative if $\gamma_{10} > \gamma_{21}$.

Ti:sapphire – absorption and emission



Absorption and emission spectra of Ti:sapphire



Upper level lifetime:

$$\tau_{21} = 3.2 \mu\text{sec}$$

$$\tau_{21}^{-1} = \gamma_{21} = \gamma_{\text{rad}} + \gamma_{\text{nr}} = 3.13 \times 10^5 \text{ sec}^{-1}$$

$$2.6 \times 10^5 \text{ sec}^{-1}$$

$$5.3 \times 10^4 \text{ sec}^{-1}$$

So: radiative transitions are about 5 times more likely than non-radiative ones.

Some solid-state laser materials

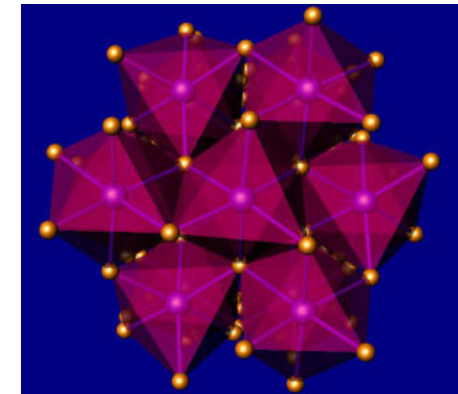
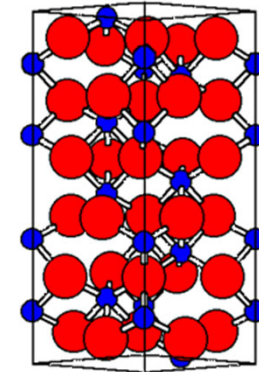
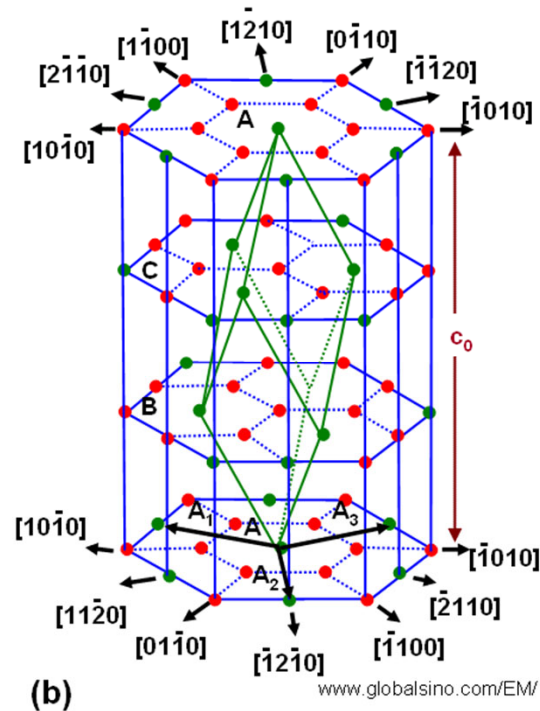
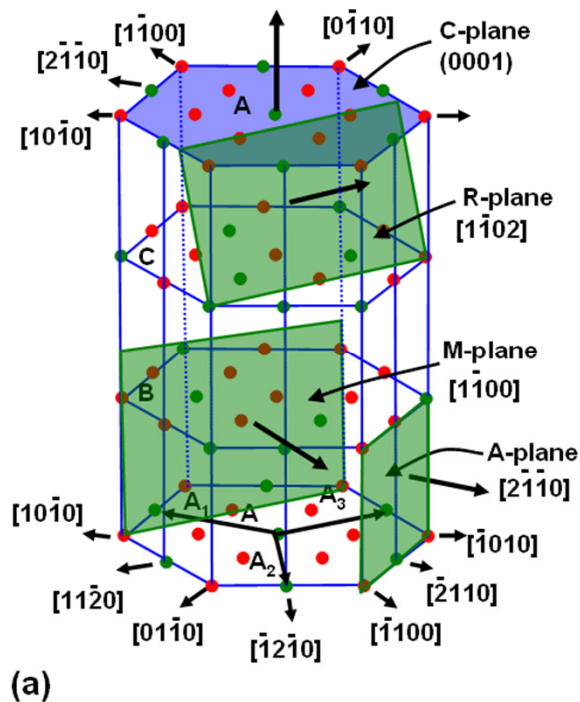
<u>Material</u>	<u>τ_{21} (μs)</u>
Ti:sapphire	3.2
Alexandrite (Cr:BeAl ₂ O ₄)	260
Nd:YAG	230
Nd:YLF	450
Nd:glass	300
Cr:LiSAF	67

Recall: a larger τ_{21} = easier to make a laser

So why is Ti:sapphire the most useful for femtosecond pulses?

Ti:sapphire - the femtosecond workhorse

crystal structure of sapphire, Al_2O_3
Hexagonal system, rhomboidal class $3m$

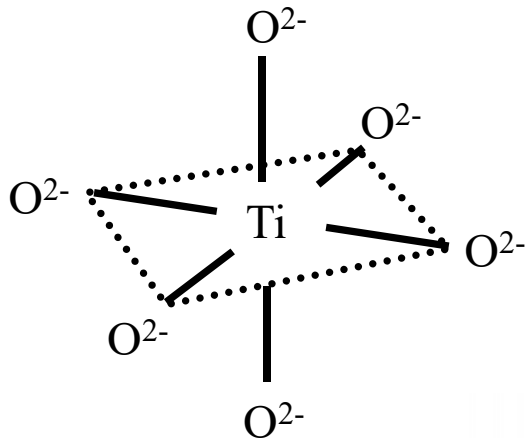


To make Ti:sapphire: replace a fraction of the Al atoms by Ti.

Lasing transition of Ti:sapphire

Fabrication: Ti^{3+} ions replace $\sim 0.1\%$ of the Al^{3+} ions in the sapphire lattice

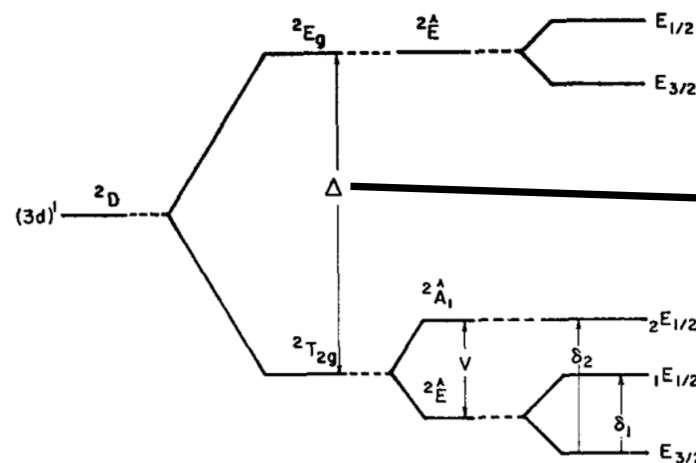
→ Ti^{3+} ions sit at the center of an octahedral site, with six O^{2-} atoms around, one at each apex.



Electronic configuration of Ti^{3+} : $[\text{Ar}]3d^1$

→ Effectively a one-electron system

Crystal field of the oxygen atoms splits the 10 degenerate 3d states:

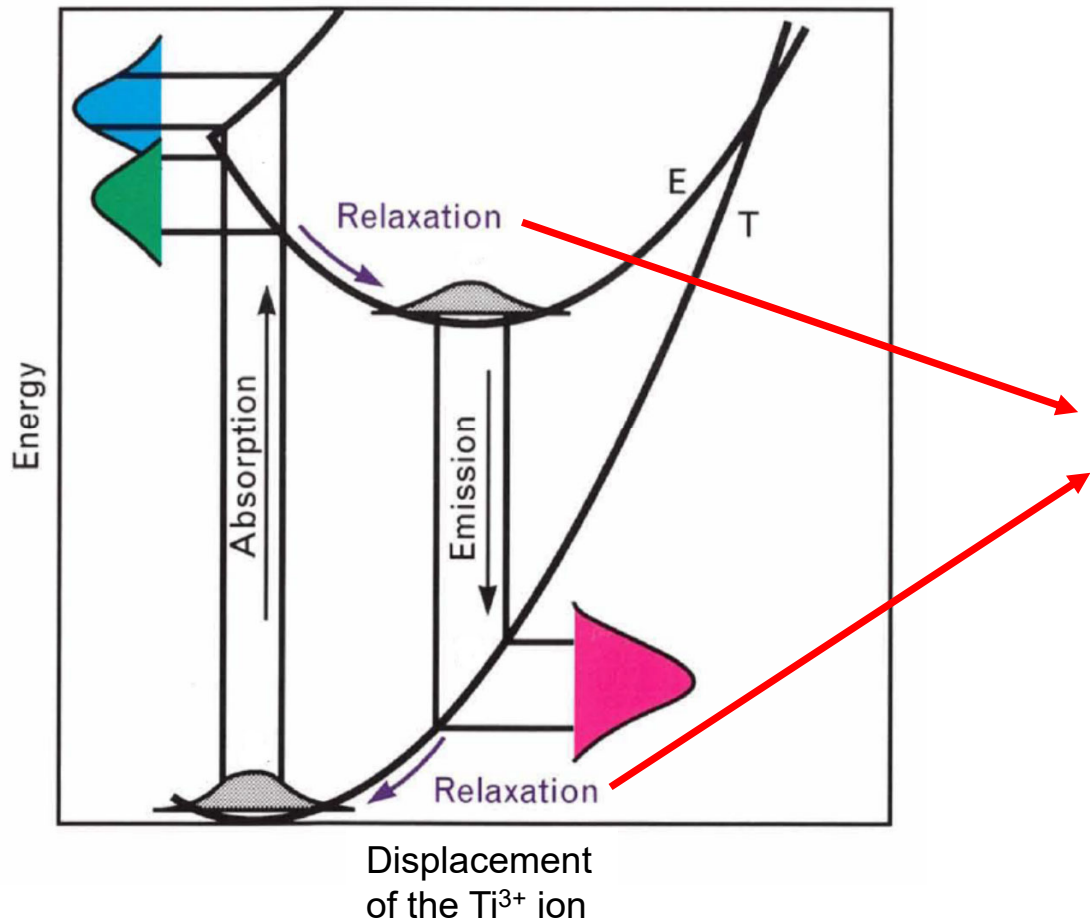


This energy splitting is about 1.5 eV (~ 800 nm)

FREE ION + CUBIC FIELD + TRIGONAL + SPIN-ORBIT

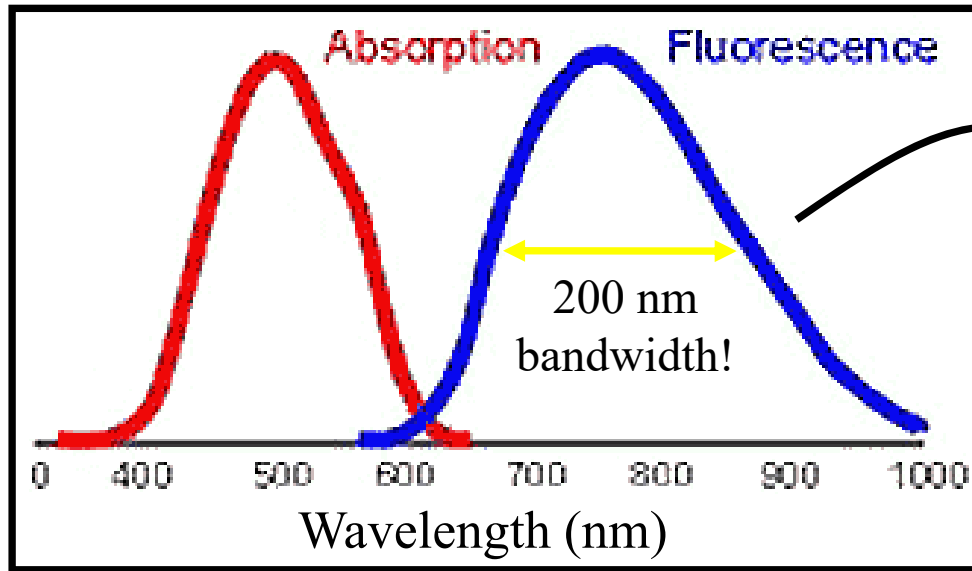
Ti:sapphire is a 'vibronic gain medium'

The energies of both the E (upper doublet) and T (lower triplet) states depend on the Ti-O bond length:

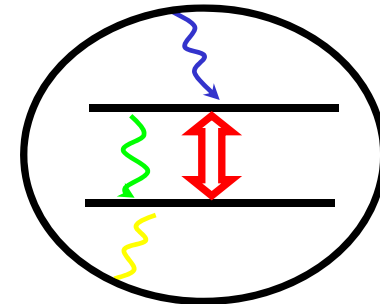


These are **fast**
(~100 fsec)
processes.

What about this fluorescence line?

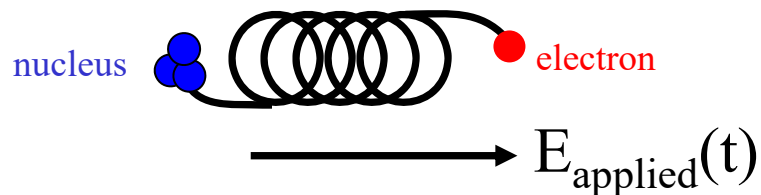


Single-frequency emission?



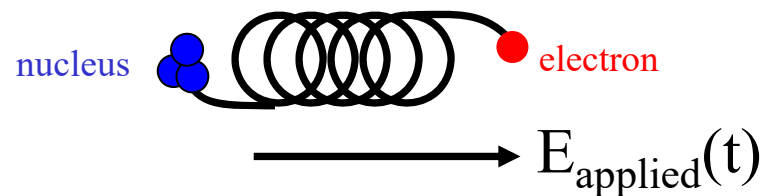
How do we describe frequency-dependent effects?

Simplest model: classical electron oscillator - treat the electronic state as a classical dipole (a charge on a spring)



The Forced Oscillator model

Consider an electron on a spring with position $x_e(t)$, and driven by an incident light wave, $E_0 \exp(-j\omega t)$:

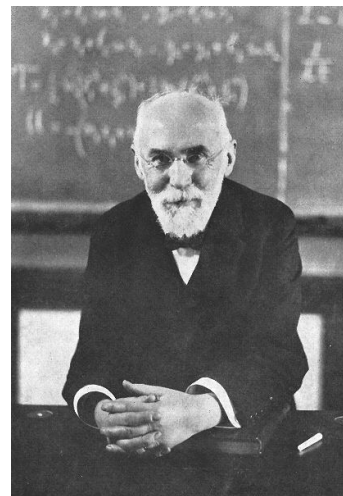


The forces on the electron are:

1. The restoring force of the spring: $-k_{\text{spring}}x_e$
2. The force exerted by the electric field: eE

This model was first proposed by Hendrik Lorentz in the 1890's. Hence it is known as the

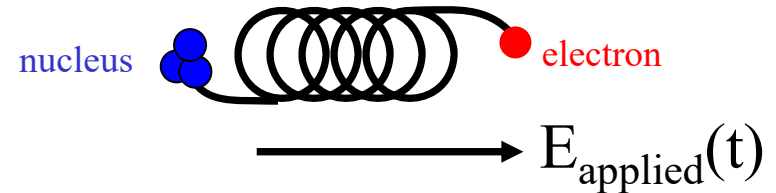
“Lorentz oscillator model”



Hendrik Lorentz
1853-1928

Lorentz Oscillator model

Use Newton's Law, $F = ma$, to write down an equation of motion for the electron:



$$m_e a(t) = m_e \frac{d^2 x_e(t)}{dt^2} = -k_{spring} x_e(t) + eE_0 e^{-j\omega t}$$

Define a new constant ω_0 to characterize the spring: $\omega_0^2 = \frac{k_{spring}}{m_e}$

the “resonance frequency” of the spring

this ω is the frequency of the light wave

→ $\frac{d^2 x_e(t)}{dt^2} = -\omega_0^2 x_e(t) + \frac{eE_0}{m_e} e^{-j\omega t}$

We generally envision that the frequency of the light wave can be varied, but the resonant frequency of the atom is a fixed constant.

The Forced Oscillator model: solution

$$\frac{d^2 x_e(t)}{dt^2} = -\omega_0^2 x_e(t) + \frac{eE_0}{m_e} e^{-j\omega t}$$

The standard way to solve a DE: guess the solution, and see if it works.

$$x_e(t) = A \exp(-j\omega t) \quad \Rightarrow \quad \frac{d^2 x_e}{dt^2} = -\omega^2 A \exp(-j\omega t)$$

Plug into the equation:

$$-A\omega^2 \cancel{e^{-j\omega t}} = -A\omega_0^2 \cancel{e^{-j\omega t}} + \frac{eE_0}{m_e} \cancel{e^{-j\omega t}}$$

—————→ Our guess is indeed a solution, but only if the factor A satisfies:

$$A = \frac{eE_0/m_e}{(\omega_0^2 - \omega^2)}$$

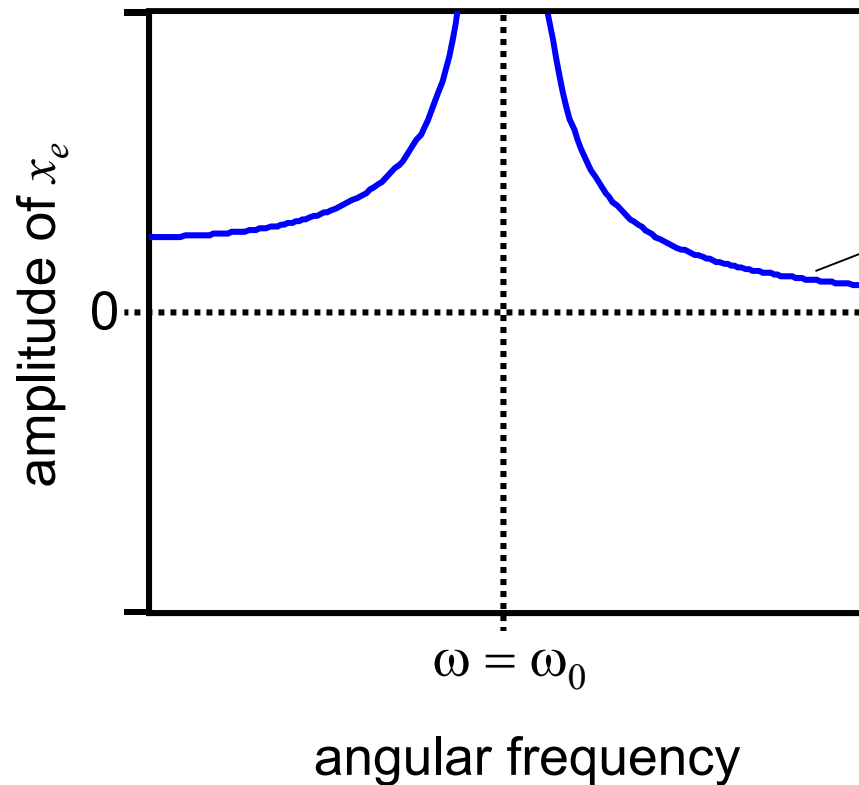
$$\text{Thus the solution is: } x_e(t) = \frac{eE_0/m_e}{(\omega_0^2 - \omega^2)} e^{-j\omega t}$$

So the electron oscillates at the frequency of the incident light wave (ω), but with an amplitude that depends on the light wave's frequency ω .

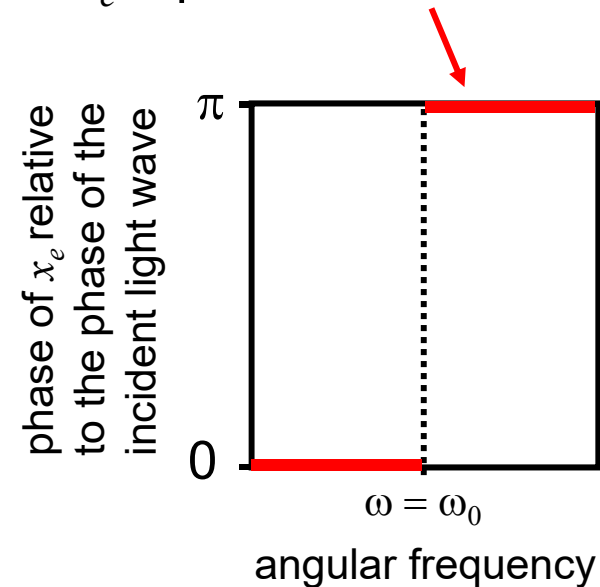
Amplitude and phase response

How does the amplitude (and phase) of the motion of the charge depend on the frequency of the incident light wave?

$$|x_e(t)| = \frac{|e|E_0}{m_e} \cdot \frac{1}{|\omega_0^2 - \omega^2|}$$



negative x_e = phase of 180°



Question: what if the light wave oscillates at frequency $\omega = \omega_0$?


The Damped Forced Oscillator

A damped forced oscillator is a harmonic oscillator experiencing a sinusoidal force and friction. It is not realistic to ignore friction.

We must add a frictional drag term,
proportional to the velocity of the electron: $-2m_e\Gamma \frac{dx_e}{dt}$

$$\frac{d^2 x_e(t)}{dt^2} = -\omega_0^2 x_e(t) + \frac{eE_0}{m_e} e^{-j\omega t} - \underbrace{2\Gamma \frac{dx_e(t)}{dt}}_{\text{new term}}$$

The solution is: $x_e(t) = \left[\frac{(eE_0 / m_e)}{(\omega_0^2 - \omega^2 - j2\omega\Gamma)} \right] e^{-j\omega t}$

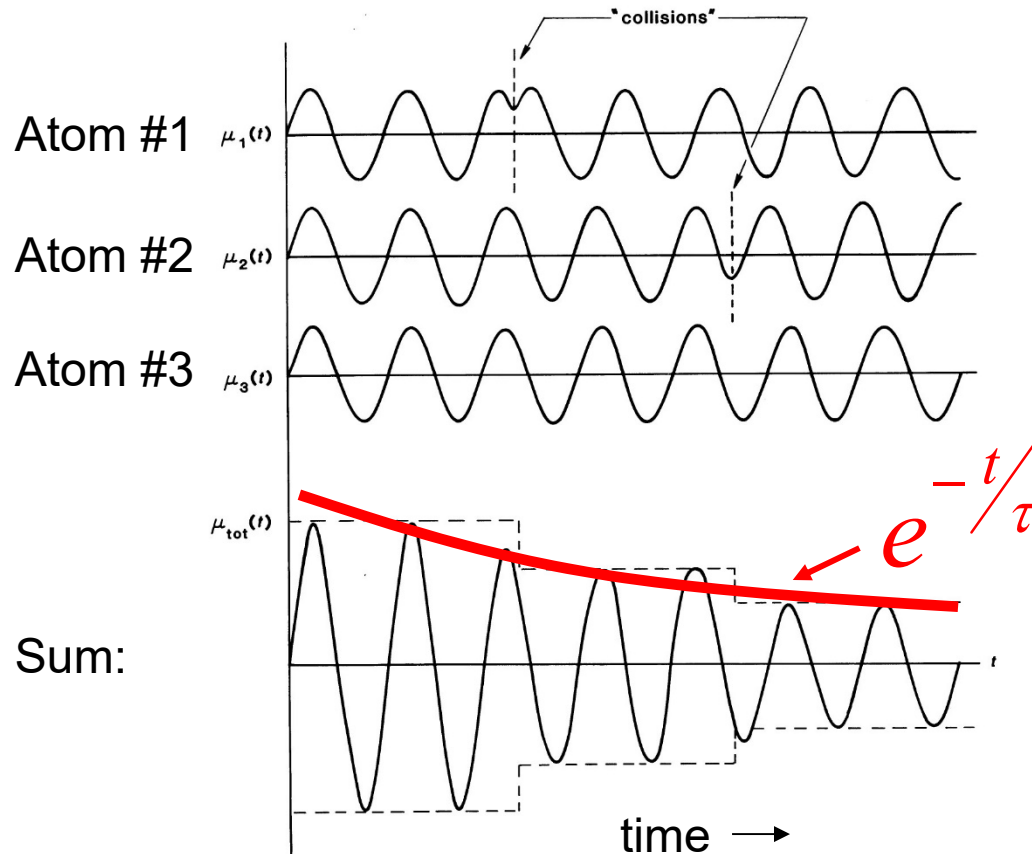
complex! 

The electron oscillates at the frequency of the incident light wave (ω), but with an amplitude **and a phase** that both depend on ω .

Why we included the damping factor, Γ

Atoms spontaneously decay to the ground state after a time.

Also, the net vibration of a medium is the sum of the vibrations of all the atoms in the medium.



Collisions “dephase” the vibrations, causing cancellation of the total medium vibration, typically exponentially.

The time constant of this exponential decay is the inverse of the damping factor:

$$\tau \propto \frac{1}{\Gamma}$$

The line shape: a complex Lorentzian

Our solution for $x_e(t)$:
$$x_e(t) = \left[\frac{(eE_0 / m_e)}{(\omega_0^2 - \omega^2 - j2\omega\Gamma)} \right] e^{-j\omega t}$$

Consider this factor:
$$\frac{1}{\omega_0^2 - \omega^2 - j2\omega\Gamma} = \frac{1}{(\omega_0 + \omega)(\omega_0 - \omega) - j2\omega\Gamma}$$

Assuming $\omega \approx \omega_0$ this becomes:
$$\approx \frac{1}{2\omega_0(\omega_0 - \omega) - j2\omega_0\Gamma}$$

$$= \frac{1}{2\omega_0} \left[\frac{1}{(\omega_0 - \omega) - j\Gamma} \right]$$

We can now rewrite $x_e(t)$ in a more compact form,
 using the following definition:

$$\delta = \omega_0 - \omega$$

This factor is called
 a “complex Lorentzian”

Our solution
 then becomes:

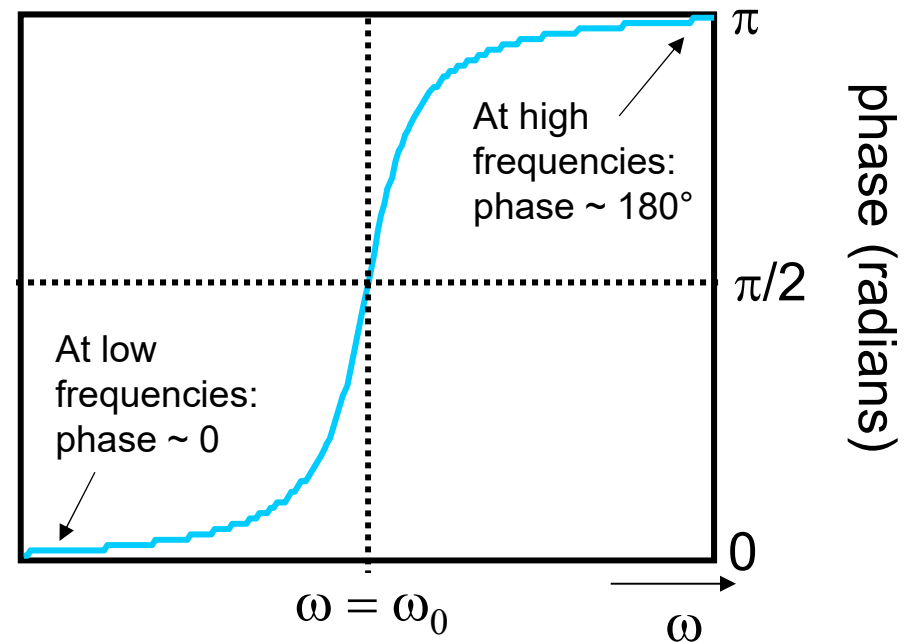
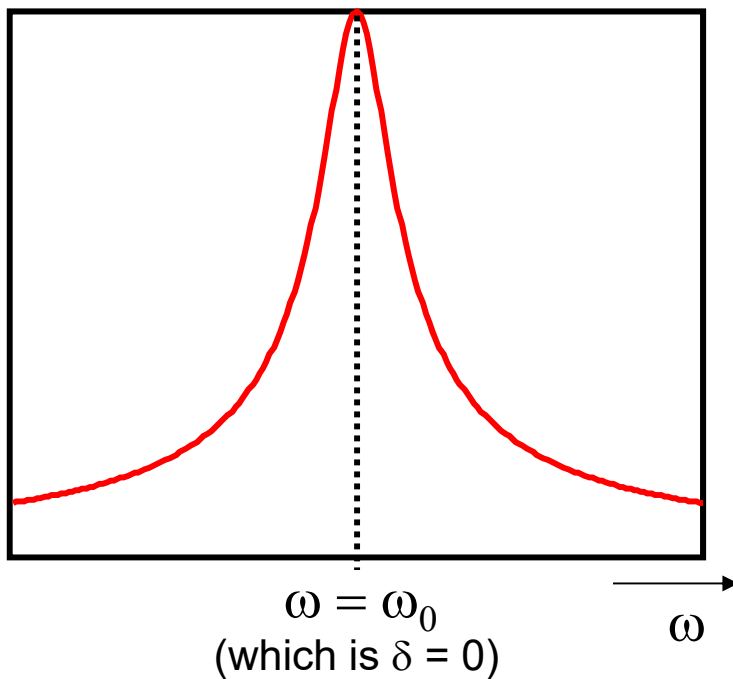
$$x_e(t) = \frac{e / m_e}{2\omega_0} \left[\frac{1}{\delta - j\Gamma} \right] E_0 e^{-j\omega t}$$

the incident
 light field

Plots of the complex Lorentzian

In terms of its amplitude and phase.

$$\frac{1}{\delta - j\Gamma} = \underbrace{\frac{1}{\sqrt{\delta^2 + \Gamma^2}}}_{\text{Amplitude}} e^{j \underbrace{\tan^{-1}(\Gamma/\delta)}_{\text{Phase}}}$$

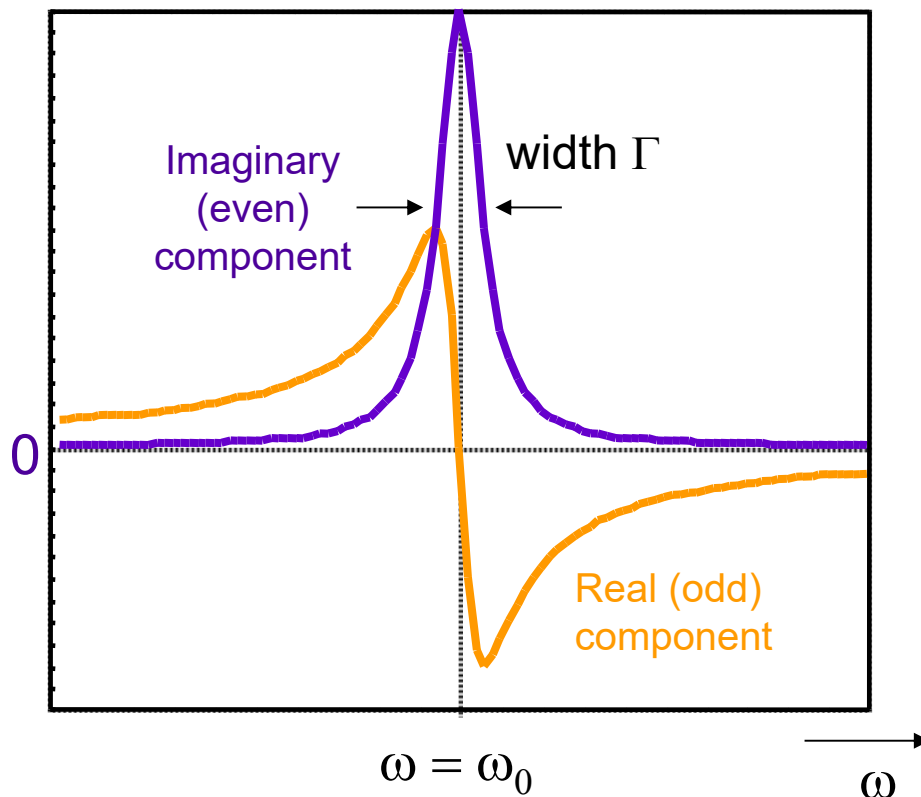


Plots of the complex Lorentzian

In terms of its real and imaginary parts.

$$\frac{1}{\delta - j\Gamma} = \frac{\delta}{\delta^2 + \Gamma^2} + j \frac{\Gamma}{\delta^2 + \Gamma^2}$$

Real Imaginary



There is a good analogy between the behavior of a mass on a spring and the behavior of an electron in an atom, even though atoms are quantum systems and the spring system is classical.

The Lorentz oscillator model is surprisingly general!

It shows up in many different places (recall RLC circuits)

Maxwell's Equations in a Medium

The induced polarization, \vec{P} , is the vector field which describes the response of the atoms to the applied field:

$$\begin{aligned}\vec{\nabla} \cdot \vec{E} &= 0 & \vec{\nabla} \times \vec{E} &= -\frac{\partial \vec{B}}{\partial t} \\ \vec{\nabla} \cdot \vec{B} &= 0 & \vec{\nabla} \times \vec{B} &= \mu_0 \epsilon_0 \frac{\partial \vec{E}}{\partial t} + \mu_0 \frac{\partial \vec{P}}{\partial t}\end{aligned}$$

where \vec{P} is given by:

$$\vec{P}(t) = Ne\vec{x}_e(t)$$

where N is the number density of charges, e is the charge per particle, and $x_e(t)$ is the displacement of each charge vs. time along the x axis.

...and we've just seen a way to compute this!

The Electric Polarization

Recall our expression for $x_e(t)$:
$$x_e(t) = \left(\frac{e}{2\omega_0 m_e} \right) \left[\frac{1}{\omega_0 - \omega - j\Gamma} \right] E(t)$$

$$P(t) = N e x_e(t) = \frac{N e^2 / m_e}{2\omega_0 (\omega_0 - \omega - j\Gamma)} E(t) \equiv \varepsilon_0 \chi(\omega) E(t)$$

which serves to define the (dimensionless) susceptibility $\chi(\omega)$.

The quantity $\varepsilon_0 \chi$ is the proportionality constant between the induced polarization and the E field that induced it.

And the dielectric permittivity of a medium is given by: $\varepsilon = \varepsilon_0 (1 + \chi)$

The wave equation with polarization

We must first include the induced polarization in Maxwell's equations, i.e., the Wave Equation:

$$\frac{\partial^2 E}{\partial x^2} - \frac{1}{c_0^2} \frac{\partial^2 E}{\partial t^2} = \mu_0 \frac{\partial^2 P}{\partial t^2}$$

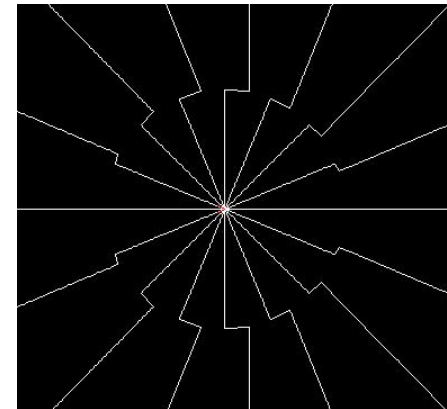
Here, we're looking at one component of the vector E and P fields, so we've dropped the vector symbol.

This equation is known as the "Inhomogeneous Wave Equation."

The polarization is a 'source term', which modifies the wave that solves Maxwell's equations

Note: Since $P(t) = N_{ex}(t)$, the 2nd derivative $\frac{\partial^2 P}{\partial t^2}$ is related to the charge acceleration.

Accelerating charges emit light!



Solving the inhomogeneous wave equation

$$\frac{\partial^2 E}{\partial x^2} - \frac{1}{c_0^2} \frac{\partial^2 E}{\partial t^2} = \mu_0 \frac{\partial^2 P}{\partial t^2}$$

We just saw that the electric susceptibility is the proportionality constant between polarization and the E field that induces it:

$$P = \varepsilon_0 \chi(\omega) E$$

So the wave equation becomes:

$$\frac{\partial^2 E}{\partial x^2} - \frac{1}{c_0^2} \frac{\partial^2 E}{\partial t^2} = \varepsilon_0 \mu_0 \chi(\omega) \frac{\partial^2 E}{\partial t^2}$$

which becomes:

$$\frac{\partial^2 E}{\partial x^2} - \frac{1 + \chi(\omega)}{c_0^2} \frac{\partial^2 E}{\partial t^2} = 0$$

This wave equation describes the propagation of waves in a medium in which the polarization is proportional to the electric field (which is almost universally true).

Solving the inhomogeneous wave equation

$$\frac{\partial^2 E}{\partial x^2} - \frac{1 + \chi(\omega)}{c_0^2} \frac{\partial^2 E}{\partial t^2} = 0$$

This looks just like the wave equation in empty space, except for a modification of the wave velocity:

$$c_{medium} = \frac{c_{empty\ space}}{\sqrt{1 + \chi}} \approx \frac{c_{empty\ space}}{1 + \frac{1}{2}\chi}$$

Therefore we can solve the equation by simply modifying the well-known solution for waves in empty space:

$$E_{empty\ space}(x, t) = E_0 \exp[j(kx - \omega t)] = E_0 \exp\left[j\left(\frac{\omega}{c_0}x - \omega t\right)\right]$$

Waves in a linear medium

The solution in empty space:

$$E_{\text{empty space}}(x, t) = E_0 \exp[j(kx - \omega t)] = E_0 \exp\left[j\left(\frac{\omega}{c_0}x - \omega t\right)\right]$$

by substituting $c_0 \rightarrow \frac{c_0}{1 + \chi/2}$, becomes the solution in a medium:

$$E_{\text{medium}}(x, t) = E_0 \exp\left[j\left(\frac{\omega}{c_0}\left(1 + \frac{\chi}{2}\right)x - \omega t\right)\right] = E_0 \exp\left[jk_0\left(1 + \frac{\chi}{2}\right)x - j\omega t\right]$$

using $k_0 = \omega/c_0$
for the value in
empty space

But χ is a complex quantity,
so write $\chi = \text{Re}\{\chi\} + j \text{Im}\{\chi\}$

$$E(x, t) = E_0 \exp\left[jk_0\left(1 + \frac{1}{2}\text{Re}\{\chi\}\right)x - j\omega t\right] \exp\left[-\frac{1}{2}k_0 \text{Im}\{\chi\}x\right]$$

Waves in a linear medium

This solution contains the two well-known material constants which together characterize the propagation of light in the medium:

$$E(x, t) = E_0 \exp \left[jk_0 \left(1 + \frac{1}{2} \operatorname{Re} \{ \chi \} \right) x - j\omega t \right] \exp \left[-\frac{1}{2} k_0 \operatorname{Im} \{ \chi \} x \right]$$

refractive index absorption coefficient

$$E(x, t) = E_0(x=0) \cdot e^{-\alpha x/2} \cdot e^{jn k_0 x - j\omega t}$$

Absorption causes
attenuation of the field
with increasing z

Refractive index
changes the k-vector

Note: if $\alpha = 0$ and $n = 1$, this reduces to the familiar result for waves in empty space:

$$E(x, t) = E_0(x=0) \cdot e^{jk_0 x - j\omega t}$$

Absorption and the susceptibility χ

We have seen that the **absorption coefficient** of a medium is related to the imaginary part of the complex susceptibility:

$$\alpha = k_0 \operatorname{Im} \{ \chi \}$$

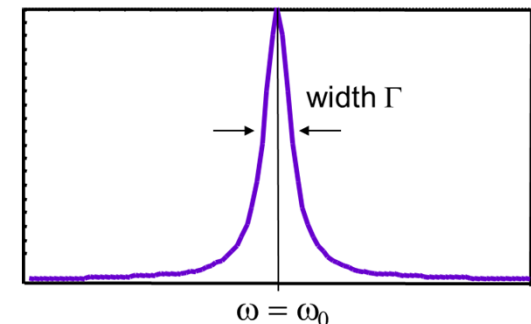
The Lorentz oscillator model gave us an expression for the susceptibility:

$$\chi(\omega) = \frac{Ne^2 / \varepsilon_0 m_e}{2\omega_0 (\omega_0 - \omega - j\Gamma)}$$

Plugging this in to the expression above for α , we find:

$$\alpha(\omega) = \frac{Ne^2}{2\varepsilon_0 c_0 m_e} \cdot \frac{\Gamma}{(\omega_0 - \omega)^2 + \Gamma^2}$$

i.e., the absorption peaks at the resonance of the two-level transition. The spectral width is set by the damping.



Refractive Index and the susceptibility χ

In the case of a time varying E-field (i.e., a light wave), the oscillating polarization can be **out of phase** with the oscillating E field.

Thus, it is not surprising that the susceptibility must be a complex quantity.

As we just saw, the **refractive index** of a medium comes from the real part of χ :

$$n(\omega) = \sqrt{1 + \text{Re}\{\chi(\omega)\}} \approx 1 + \frac{1}{2} \text{Re}\{\chi(\omega)\}$$

The Lorentz oscillator model gave us an expression for the susceptibility. Plugging it in to this, we find:

$$n(\omega) = 1 + \frac{Ne^2 / m_e}{4\omega_0 \varepsilon_0} \frac{\omega_0 - \omega}{(\omega_0 - \omega)^2 + \Gamma^2}$$

Refractive Index and the Speed of Light

The speed of light is ω/k . Since k becomes nk in a medium,

$$c = \omega / (nk) = (\omega / k) / n \longrightarrow c = c_0 / n$$

where c_0 is the speed of light in vacuum.

Note: often people don't write the subscript 0 on c , even when they mean c_0 .

- The refractive index is (almost) always > 1 .

So **LIGHT SLOWS DOWN** inside materials.

(But it can be < 1 . This appears to violate Relativity, but it doesn't.)

- The refractive index generally depends on frequency.

So **the speed of light in materials also depends on frequency.**

Refractive Index and the permittivity ϵ

We have just seen that $n = c_0/c$

But we also know from the wave equation that the speed of light is given by:

$$c_0 = \frac{1}{\sqrt{\epsilon_0 \mu_0}}$$

in empty space

$$c = \frac{1}{\sqrt{\epsilon \mu}}$$

in a medium with ϵ, μ

Thus we have:
$$n = \frac{c_0}{c} = \frac{\sqrt{\epsilon \mu}}{\sqrt{\epsilon_0 \mu_0}}$$

In optics, we rarely deal with magnetic materials, so we can take $\mu = \mu_0$. So:

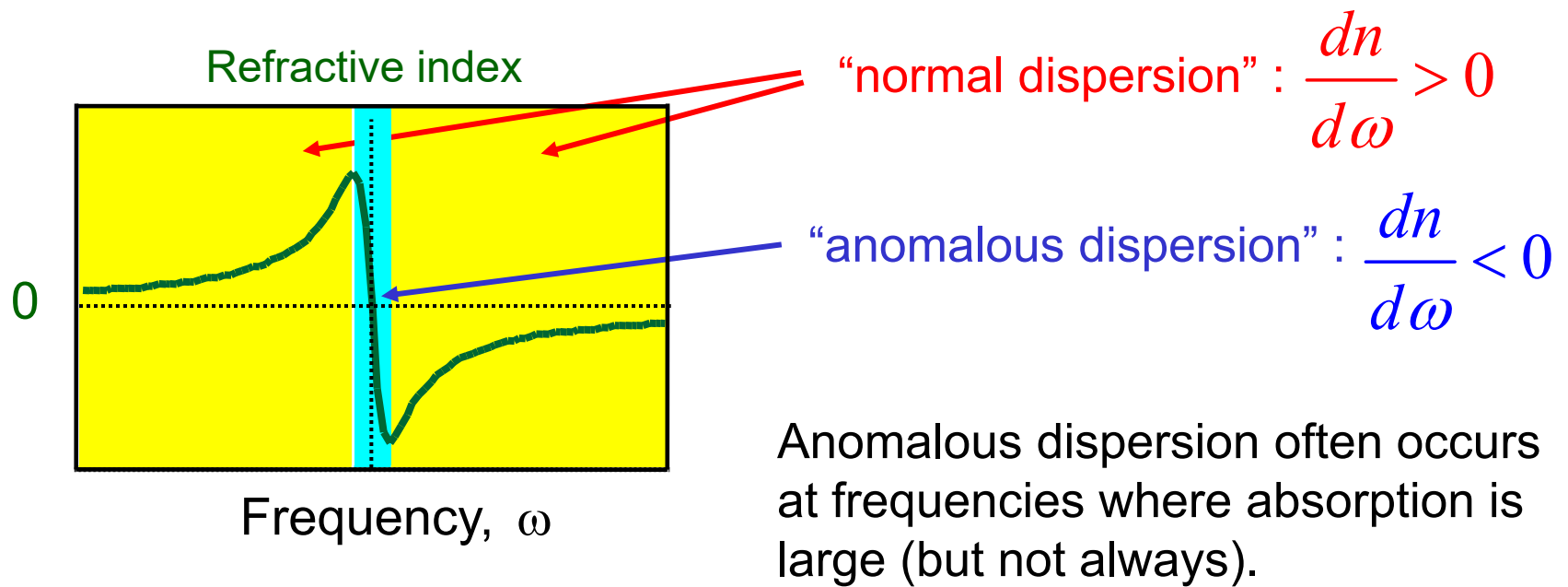
$$n = \sqrt{\frac{\epsilon}{\epsilon_0}} = \sqrt{\epsilon_r}$$

The relative permittivity is equal to the square of the refractive index.

Dispersion: n depends on ω

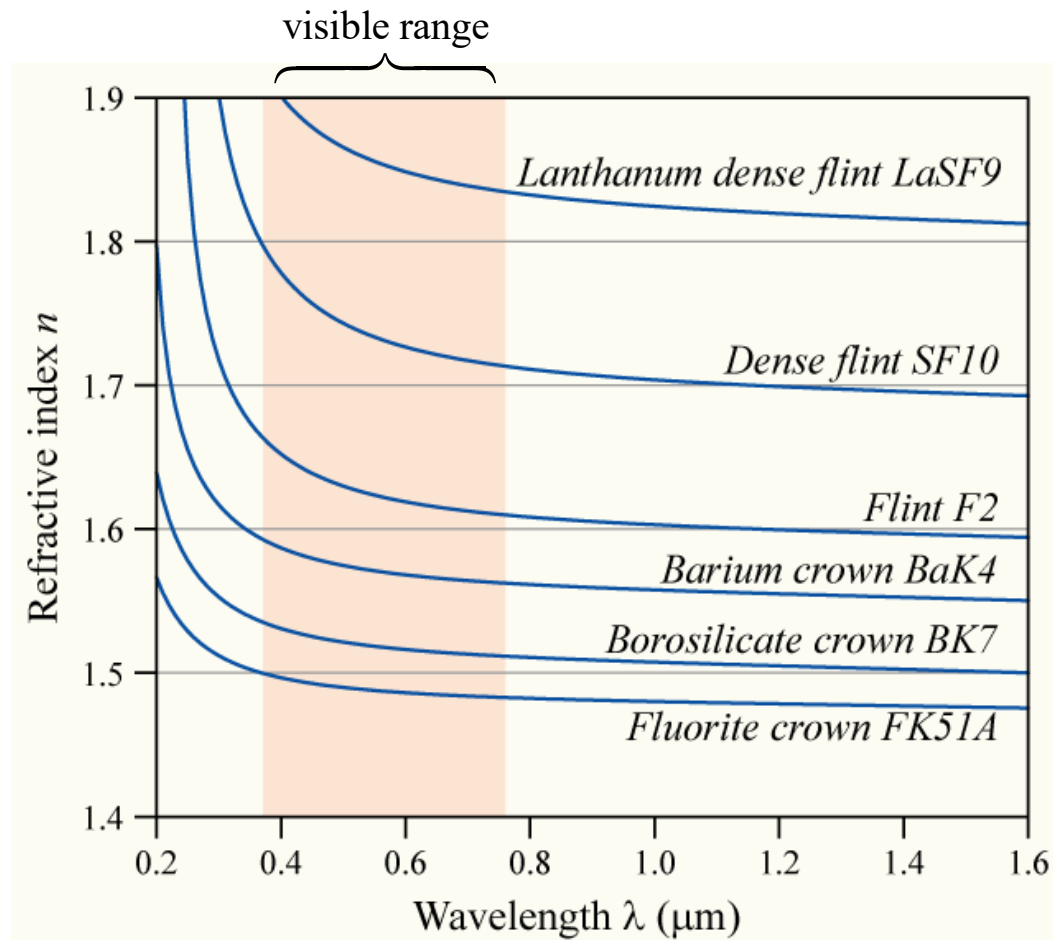
The word “dispersion” refers to the frequency-dependence of the refractive index of a medium.

We typically distinguish two particular cases.



Normal dispersion is only called “normal” because it is more commonly observed, not because there is anything more ‘normal’ about it.

Refractive indices of various types of glass



Note

As λ increases, the index decreases. Thus: $\frac{dn}{d\lambda} < 0$

But:
$$\frac{dn}{d\omega} = \frac{dn}{d\lambda} \cdot \frac{d\lambda}{d\omega}$$

(the chain rule)

and:
$$\lambda = \frac{c}{f} = \frac{2\pi c}{\omega}$$

(which implies that $d\lambda/d\omega < 0$)

Therefore:
$$\frac{dn}{d\omega} > 0$$

→ Normal dispersion

What about population inversion?

These line shape functions were derived under the assumption that all of the participating atoms start out in their lower levels. So the factor of N in these expressions is “total number of atoms per m^3 ”.

$$\alpha(\omega) = \frac{Ne^2}{2\varepsilon_0 c_0 m_e} \cdot \frac{\Gamma}{(\omega_0 - \omega)^2 + \Gamma^2}$$

$$n(\omega) = 1 + \frac{Ne^2 / m_e}{4\omega_0 \varepsilon_0} \frac{\omega_0 - \omega}{(\omega_0 - \omega)^2 + \Gamma^2}$$

But in a laser, we strive to put a significant fraction (more than half) of the atoms in the upper level (population inversion), by pumping.

So in a medium under optical pumping (e.g., the gain medium of a laser), α and n should really be written as proportional to $\Delta N = N_1 - N_2$, not just to N .

Lorentzian line shapes for both α and n

The line shape functions including population difference:

$$\alpha(\omega) \propto \frac{\Delta N}{1 + \zeta^2}$$

$$n(\omega) \propto \Delta N \frac{\zeta}{1 + \zeta^2}$$

absorption becomes *gain* when the sign of ΔN changes!

$$\zeta = \frac{2(\omega - \omega_0)}{\Gamma}$$

Γ

transition linewidth

Written in terms of cross-section:

$$\alpha(\omega) = \frac{1}{2} \Delta N \cdot \sigma(\omega) = \frac{\Delta N}{2} \cdot \frac{\sigma_0}{1 + \zeta^2}$$

units of atoms/cm³

units of cm²/atom

Some values for common laser materials:

Ti:sapphire (at a 0.1% doping level)	$\sigma_0 = 28 \cdot 10^{-20} \text{ cm}^2$
Alexandrite	$\sigma_0 = 0.8$
Cr:LiSAF	$\sigma_0 = 4.8$
Cr:LiCAF	$\sigma_0 = 1.3$
R6G laser dye	$\sigma_0 = 10,000$