

### 3. Spectral line shapes and how to model them

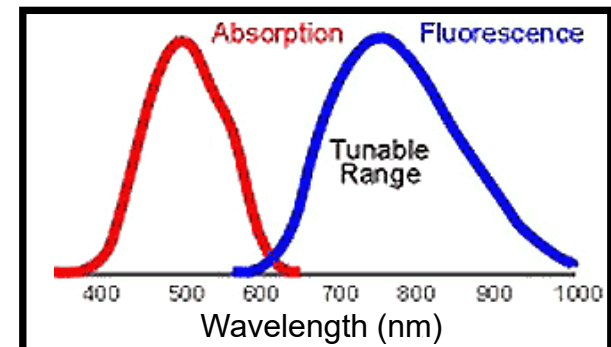
Some properties of our favorite gain medium: Ti:sapphire  
excited state lifetime (radiative, non-radiative)  
fluorescence line width

Classical electron oscillator model for spectral line shapes

The electromagnetic constitutive relation and  $\epsilon(\omega)$

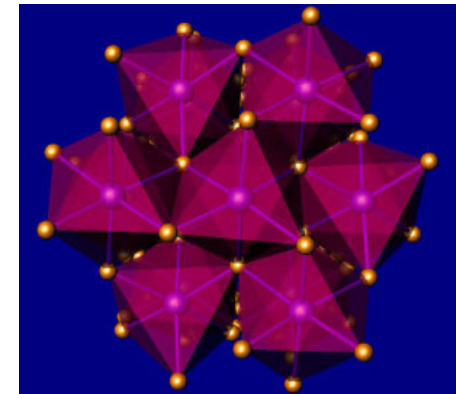
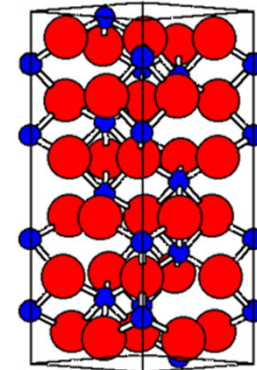
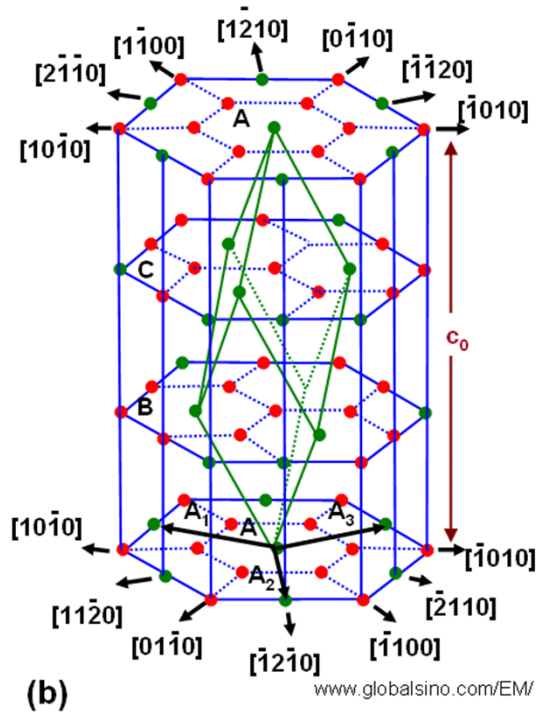
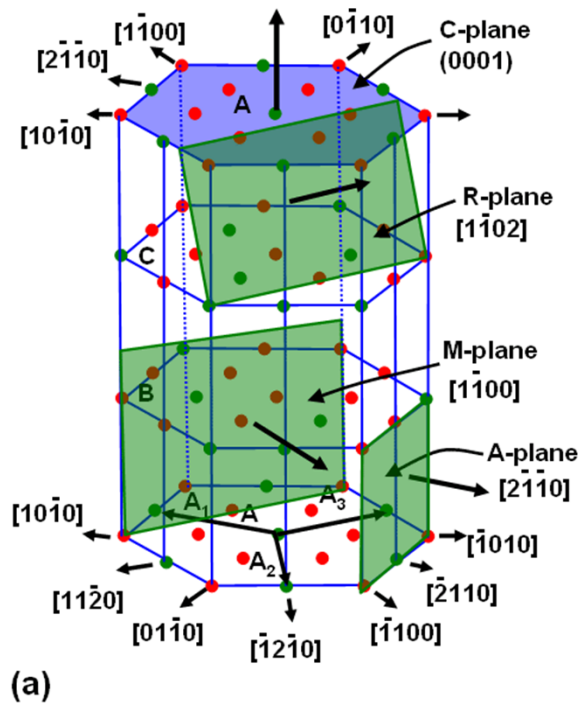
The connection between  $\epsilon(\omega)$  and experimentally  
measurable parameters

Absorption (or gain) cross-section



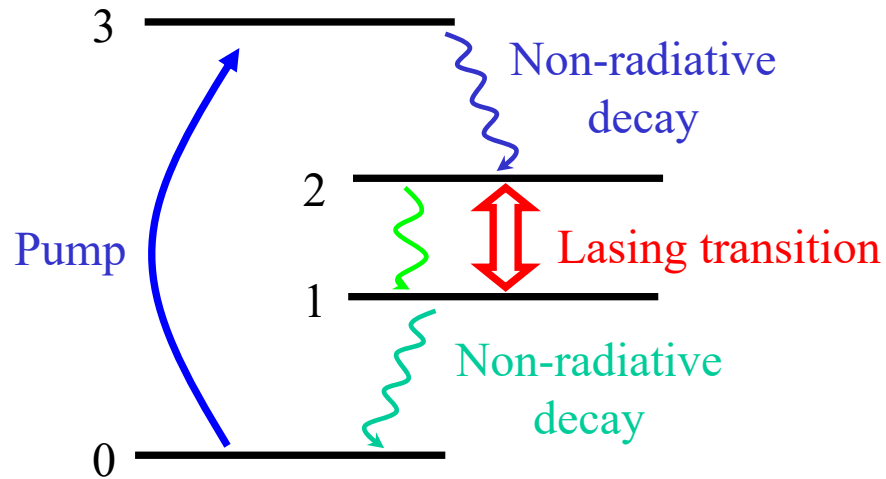
# Ti:sapphire - the femtosecond workhorse

crystal structure of sapphire,  $\text{Al}_2\text{O}_3$   
Hexagonal system, rhomboidal class  $3m$

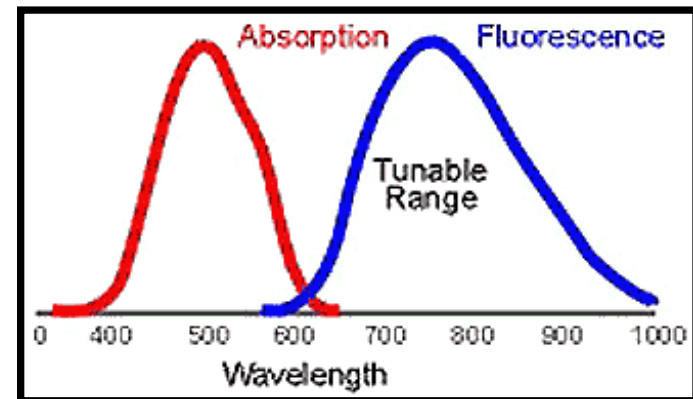


To make Ti:sapphire: replace a fraction of the Al atoms by Ti.

# 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><math>\tau_{21}</math> (<math>\mu\text{s}</math>)</u>
Ti:sapphire	3.2
Alexandrite (Cr:BeAl <sub>2</sub> O <sub>4</sub> )	260
Nd:YAG	230
Nd:YLF	450
Nd:glass	300
Cr:LiSAF	67

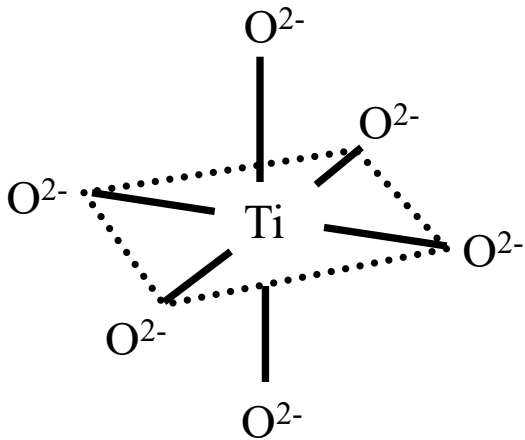
Recall: a larger  $\tau_{21}$  = easier to make a laser

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

# Lasing transition of Ti:sapphire

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

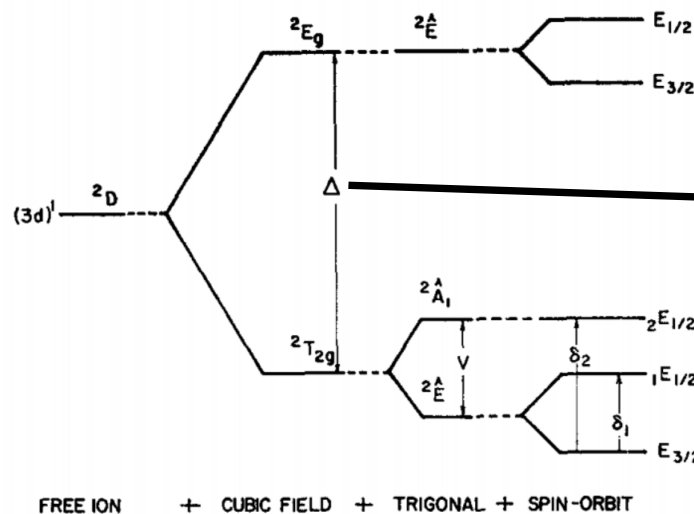
→  $\text{Ti}^{3+}$  ions sit at the center of an octahedral site, with six  $\text{O}^{2-}$  atoms around, one at each apex.



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

→ Effectively a one-electron system

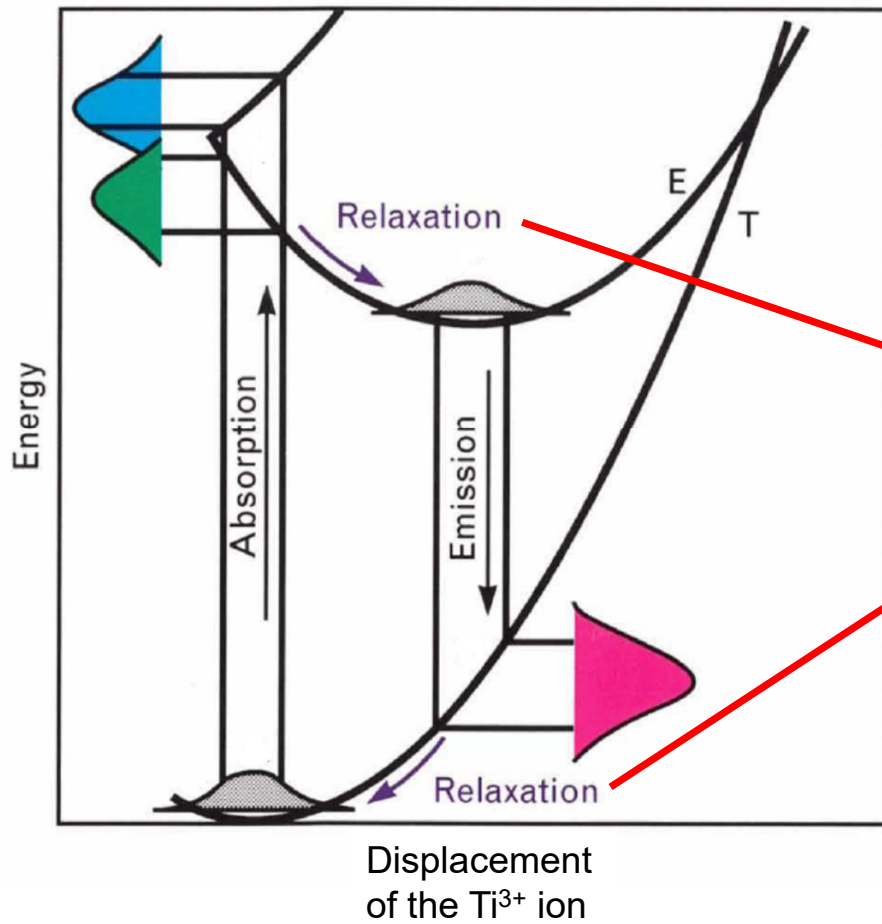
Crystal field of the oxygen atoms splits the 10 degenerate 3d states:



This energy splitting is about 1.5 eV ( $\sim 800$  nm)

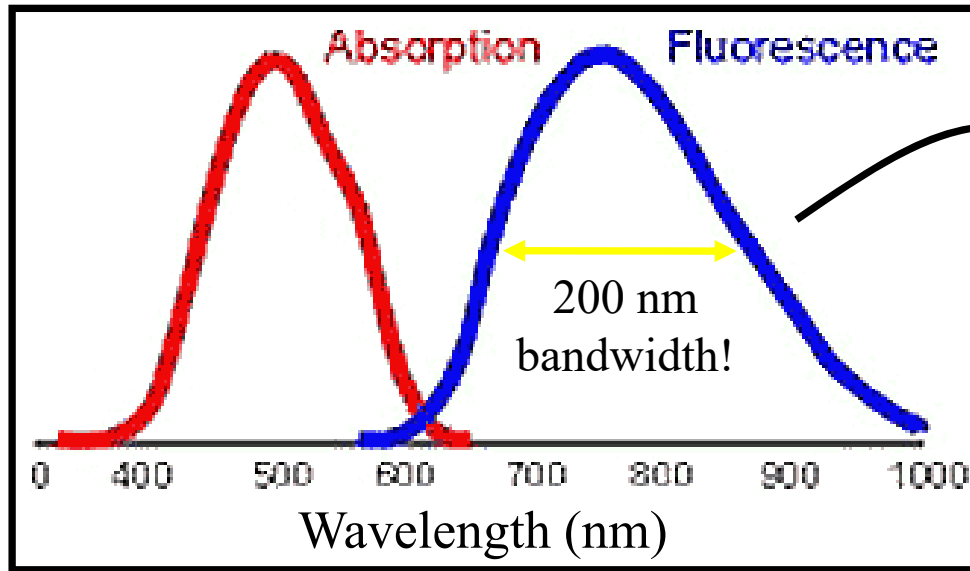
# Ti:sapphire is a 'vibronic gain medium'

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

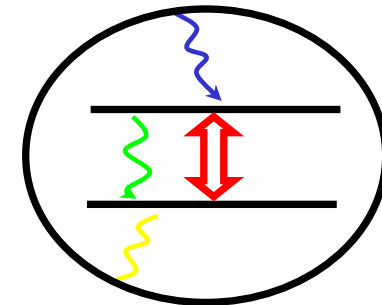


These are **fast**  
(~100 fsec)  
processes.

# What about this fluorescence line?

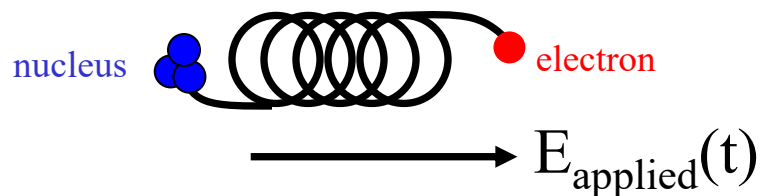


Single-frequency emission?

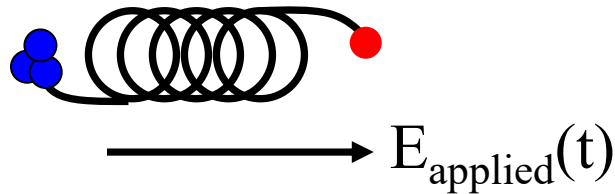


How do we describe frequency-dependent effects?

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



# Classical electron oscillator model



$$F = ma = m \frac{d^2 z(t)}{dt^2}$$

Forces are:

1. Hooke's law (the spring)
2. externally applied electric field

$$m \frac{d^2 z(t)}{dt^2} = -k \cdot z(t) + eE(t)$$

Also add in damping (a term proportional to  $\frac{dz(t)}{dt}$ )

$$\frac{d^2 z(t)}{dt^2} + \gamma \cdot \frac{dz(t)}{dt} + \omega_0^2 \cdot z(t) = \frac{eE(t)}{m} \quad \text{where } \omega_0^2 = \frac{k}{m}$$

This will tell us the position of the electron as a function of time  $z(t)$ , and therefore the induced dipole  $p(t) = e \cdot z(t)$



# Classical electron oscillator model

$$\frac{d^2 z(t)}{dt^2} + \gamma \cdot \frac{dz(t)}{dt} + \omega_0^2 \cdot z(t) = \frac{eE(t)}{m}$$

To solve:

Assume  $E(t) = E_0 \cos \omega t = \text{Re} \{ E_0 e^{i\omega t} \}$

and  $z(t) = z_0 \cos \omega t = \text{Re} \{ z_0 e^{i\omega t} \}$

Plug in:  $-\omega^2 \cdot z_0 + i\gamma\omega \cdot z_0 + \omega_0^2 \cdot z_0 = \frac{eE_0}{m}$

$$z_0 = \frac{eE_0/m}{\omega_0^2 - \omega^2 + i\gamma\omega}$$

Polarization density  $P = (\text{dipole moment per atom}) \cdot (\text{number of atoms / cm}^3)$

$$P = \frac{Ne^2}{m} \frac{E_0}{\omega_0^2 - \omega^2 + i\gamma\omega}$$

# Electromagnetic constitutive relations

How does electromagnetism deal with an induced polarization?

$$\vec{\nabla} \times \vec{B} = \mu\epsilon \frac{\partial \vec{E}}{\partial t} = \mu \frac{\partial \vec{D}}{\partial t}$$

where the 'displacement field' D is:  $\mathbf{D} = \epsilon\mathbf{E} = \mathbf{E} + 4\pi\mathbf{P}$

Classical electron oscillator (CEO) model: P is proportional to E

$$\mathbf{P} = \chi \mathbf{E}$$

*electronic susceptibility*

It tells us how much polarization an external field induces in a medium.

# Electromagnetic constitutive relations

$$\text{If } P = \chi E$$

$$\text{then: } D = \epsilon E = E + 4\pi \chi E \longrightarrow \epsilon = 1 + 4\pi \chi$$

$$\text{Thus our CEO result: } P = \frac{Ne^2}{m} \frac{E_0}{\omega_0^2 - \omega^2 + i\gamma\omega}$$

immediately tells us the dielectric permittivity:

$$\epsilon(\omega) = 1 + \frac{4\pi Ne^2}{m} \frac{1}{\omega_0^2 - \omega^2 + i\gamma\omega}$$

## Simplifying the expression for $\varepsilon(\omega)$

$$\varepsilon(\omega) = 1 + \frac{4\pi N e^2}{m} \frac{1}{\omega_0^2 - \omega^2 + i\gamma\omega}$$

Assume that for optical fields,  $\omega$  and  $\omega_0$  are similar:

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

Then we have:  $\varepsilon(\omega) = 1 - i \frac{4\pi N e^2}{m\gamma\omega_0} \cdot \frac{1}{1 + i\zeta}$

$\swarrow$  'plasma frequency'  $\omega_p^2$ 
 $\rightarrow$   $\zeta = \frac{2(\omega - \omega_0)}{\gamma}$   
(contains all of the frequency dependence)

This is the complex Lorentzian line shape function.

$$\operatorname{Re}\{\varepsilon(\omega)\} = 1 - \frac{\omega_p^2}{\gamma\omega_0} \cdot \frac{\zeta}{1 + \zeta^2} \quad \operatorname{Im}\{\varepsilon(\omega)\} = -\frac{\omega_p^2}{\gamma\omega_0} \cdot \frac{1}{1 + \zeta^2}$$

# Why did we need to find $\epsilon(\omega)$ ?

Electromagnetic wave equation: 
$$\nabla^2 E = \frac{\epsilon\mu}{c^2} \frac{\partial^2 E}{\partial t^2}$$

Assume that  $E(z,t)$  is harmonic in time:  $E(z,t) = E(z) \exp(i\omega t)$

$$\left[ \nabla^2 + \frac{\epsilon\mu\omega^2}{c^2} \right] E(z) = 0$$

Define the 'wave vector'  $k^2(\omega) = \epsilon\mu\omega^2/c^2$

Helmholtz equation: 
$$\left[ \nabla^2 + k^2 \right] E(z) = 0$$

(usually  $\mu$  doesn't do anything interesting for optical waves:  $\mu = 1$  in CGS units)

Solutions are plane waves:  $E(z_2) = E(z_1) \cdot e^{ikz}$

Propagation is governed by  $k(\omega)$ , which is proportional to  $[\epsilon(\omega)]^{1/2}$

'electromagnetic propagator'

Velocity is  $\frac{\omega}{k} = \frac{c}{\sqrt{\epsilon}}$  ← refractive index! 13

# From $\epsilon(\omega)$ to experimental parameters

The connection between  $\epsilon(\omega)$  and  $\alpha(\omega)$  and  $n(\omega)$ :

$$n(\omega) = n_R + i \cdot n_I = \sqrt{\epsilon(\omega)}$$

ordinary refractive index

power absorption coefficient is:  $\alpha = \frac{2\omega}{c} \cdot n_I$

In terms of  $\epsilon$ :

$$n(\omega) = \sqrt{\frac{|\epsilon| + \text{Re}(\epsilon)}{2}}$$
$$\alpha(\omega) = \frac{2\omega}{c} \sqrt{\frac{|\epsilon| - \text{Re}(\epsilon)}{2}}$$

NOTE: all of the results on this page are true for any  $\epsilon(\omega)$ , not just the one from our simple CEO model.

# Classical electron oscillator, again

From  $\epsilon(\omega)$ , we can derive  $\alpha(\omega)$  and  $n(\omega)$  for an oscillator.

Lorentzian line shape:  
(approximate)

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

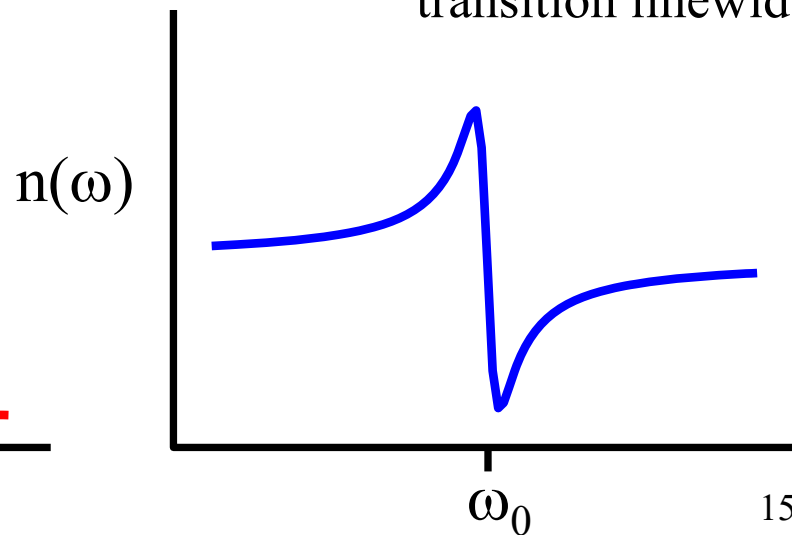
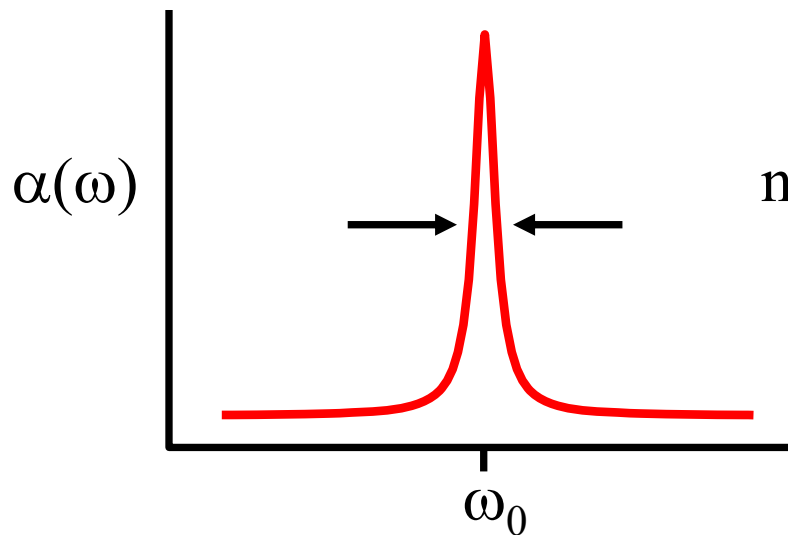
$$n(\omega) \propto \Delta N \frac{\zeta}{1 + \zeta^2}$$

absorption becomes *gain* when  
the sign of  $\Delta N$  changes!

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

$\gamma$

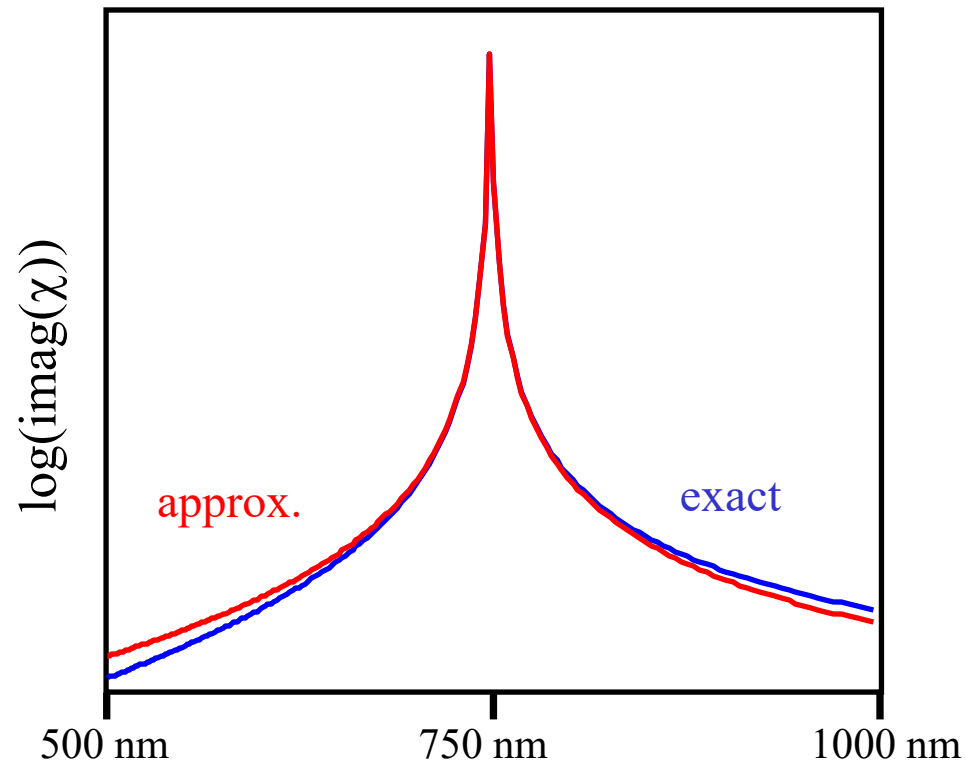
transition linewidth



# Classical electron oscillator, again

How good is the approximation?

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





# Classical electron oscillator, again

The proportionality constant:

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

units of atoms/cm<sup>3</sup>                      units of cm<sup>2</sup>/atom

$\sigma(\omega)$  = “gain (or absorption) cross-section”

Some laser materials:

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