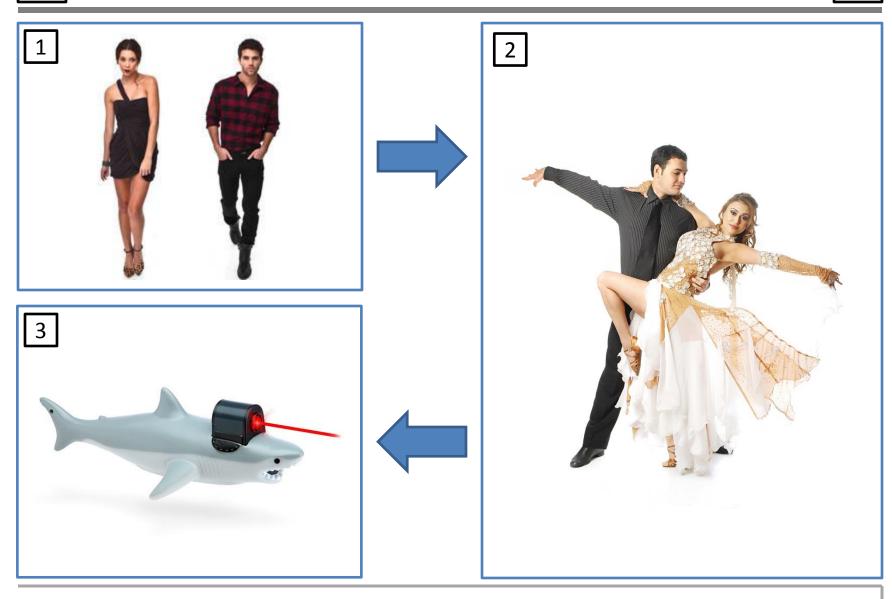
# Classical Oscillator model and electric dipole transitions in real atoms

Tomas Kristijonas Uždavinys

Laser Physics SK3410 2015-02-26 Somewhere in Alba Nova

### Outline



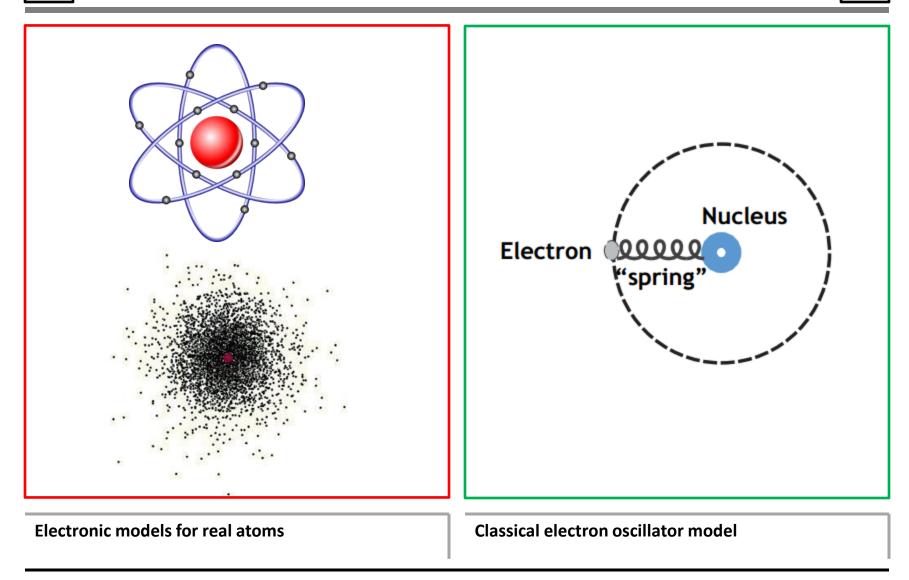
Based on "Lasers" by A. E. Siegman, 1986

# What is CEO?

### CEO – chief executive officer

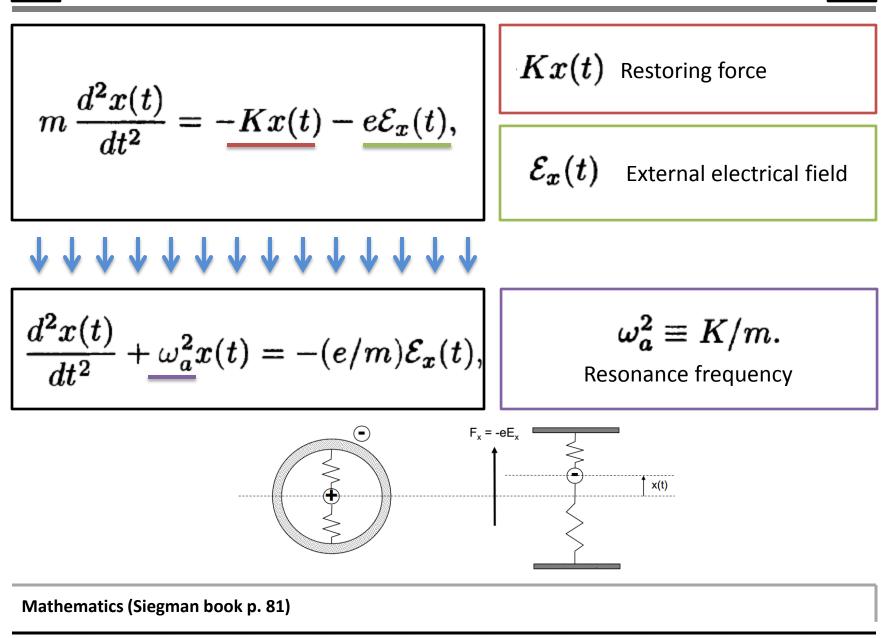


### **CEO - Classical electron oscillator**

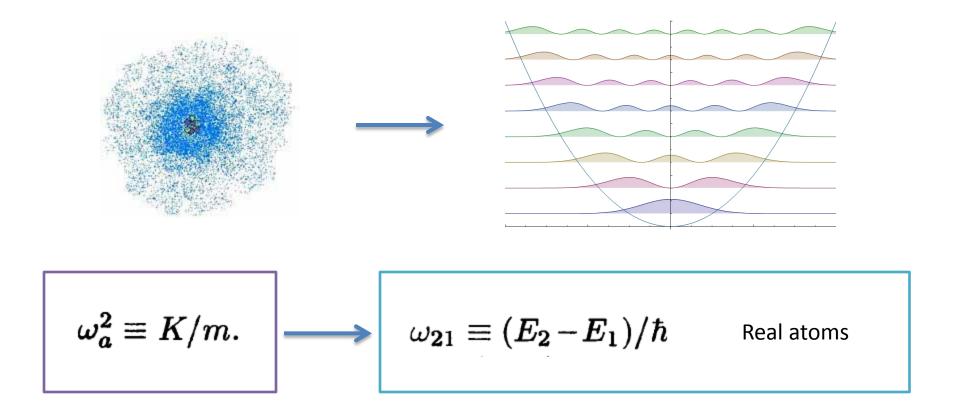


4

# Short CEO description



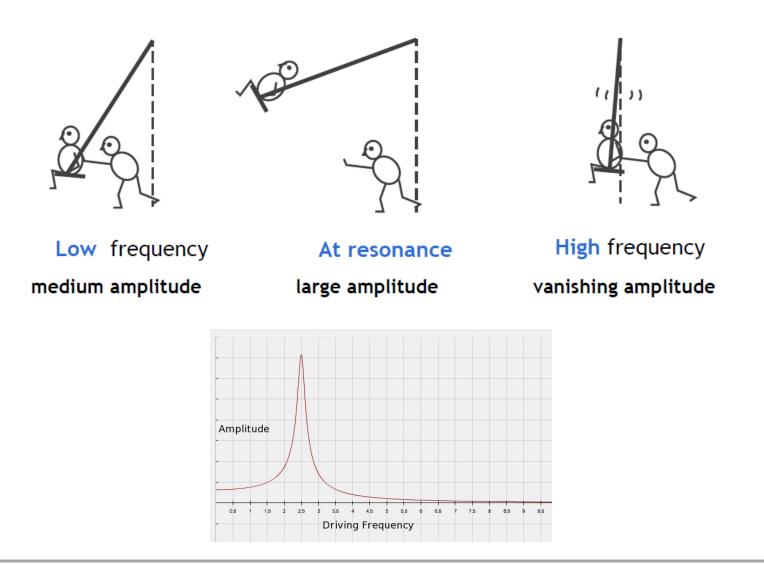
### But wait! We are in quantum world!



#### **Random iliustration**

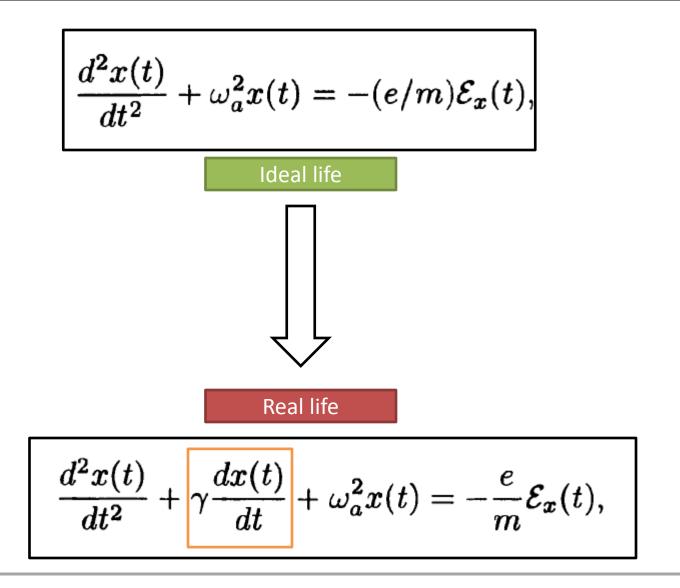
# What is resonance frequency?

# Almost real life illustration



No references- random google search pictures.

## Energy damping #1



(1) 
$$\frac{d^2x(t)}{dt^2} + \gamma \frac{dx(t)}{dt} + \omega_a^2 x(t) = -\frac{e}{h} c_x(t)$$
, No electric field

2 
$$x(t) = x(t_0) \exp[-(\gamma/2)(t-t_0) + j\omega'_a(t-t_0)],$$

**Errata** – displacement is in real part

10

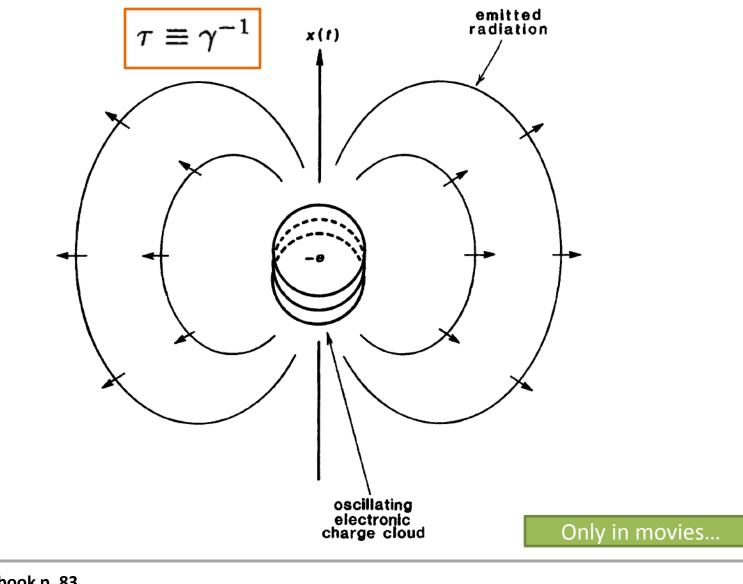
$$( \textbf{3} ) \quad \omega_a' \equiv \sqrt{\omega_a^2 - (\gamma/2)^2}.$$

exact resonance frequency

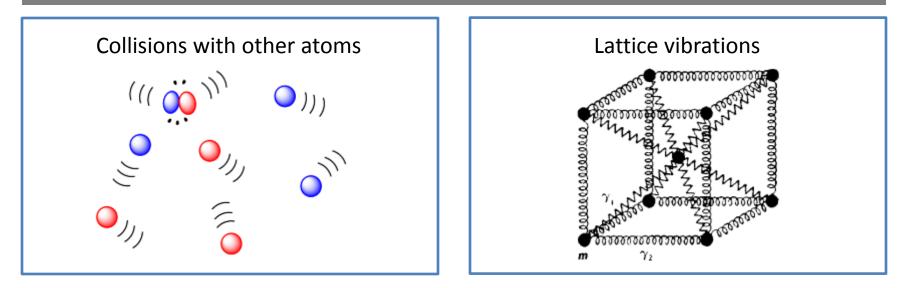
4) 
$$U_a(t) = \frac{1}{2}Kx^2(t) + \frac{1}{2}mv_x^2(t) = U_a(t_0)e^{-\gamma(t-t_0)} \equiv U_a(t_0)e^{-(t-t_0)/\tau}.$$

Siegman book p. 82-83

# Emission of electromagnetic radiation



# Real life- aka non "radiative" mechanisms



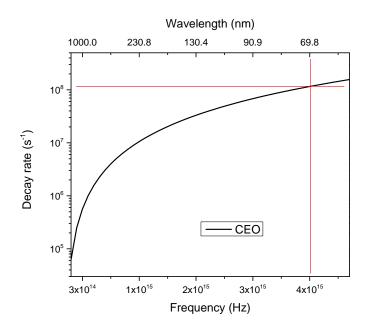
Internal charge cloud oscillation within atom

$$\gamma \equiv \left| \frac{1}{U_a} \frac{dU_a}{dt} \right| = \gamma_{\text{rad}} + \gamma_{\text{nr}}.$$
  
Errata – absolute values

Siegman book p. 83-84

#### 12

## Radiative decay rates



$$\gamma_{\rm rad,ceo} = \frac{e^2 \omega_a^2}{6\pi \epsilon m c^3}.$$

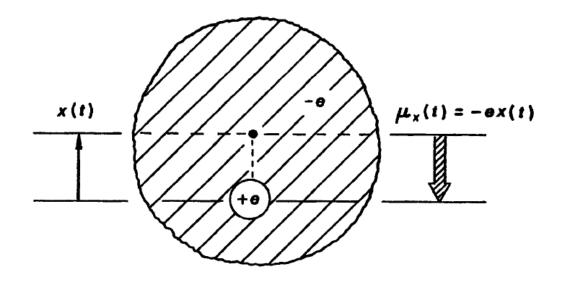
Parameter	Value
е	1.602e-19 (C)
3	8.85e-12 (F/m)
m	9.11e-31 (kg)
С	3e8 (m/s)

This classical oscillator radiative decay rate has a value  $\gamma_{\rm rad,ceo} \approx 10^8 \, {\rm sec^{-1}}$  for a visible frequency oscillator, compared to an oscillation frequency of  $\omega_a \approx 4 \times 10^{15}$   ${\rm sec^{-1}}$ .

### "UV vision"



## Microscopic dipole moments

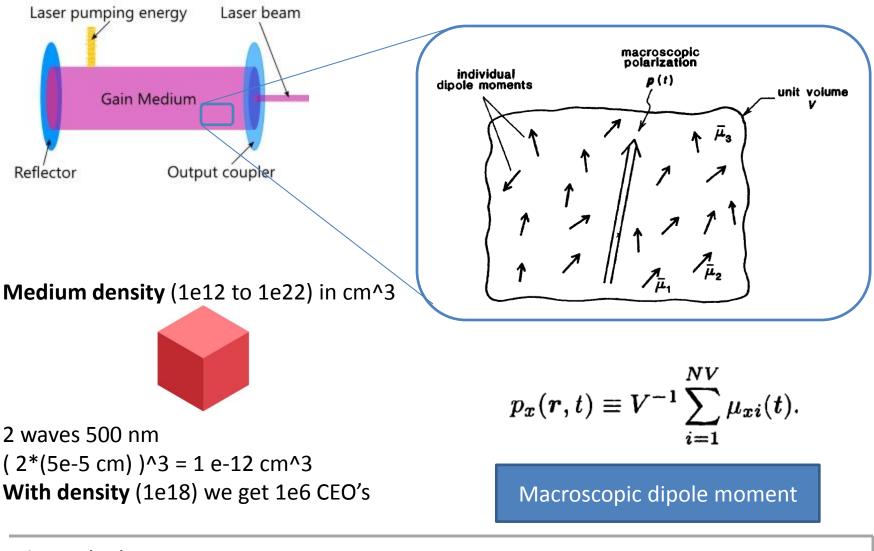


 $\mu_x(t) = [\text{charge}] \times [\text{displacement}] = -ex(t)$ 

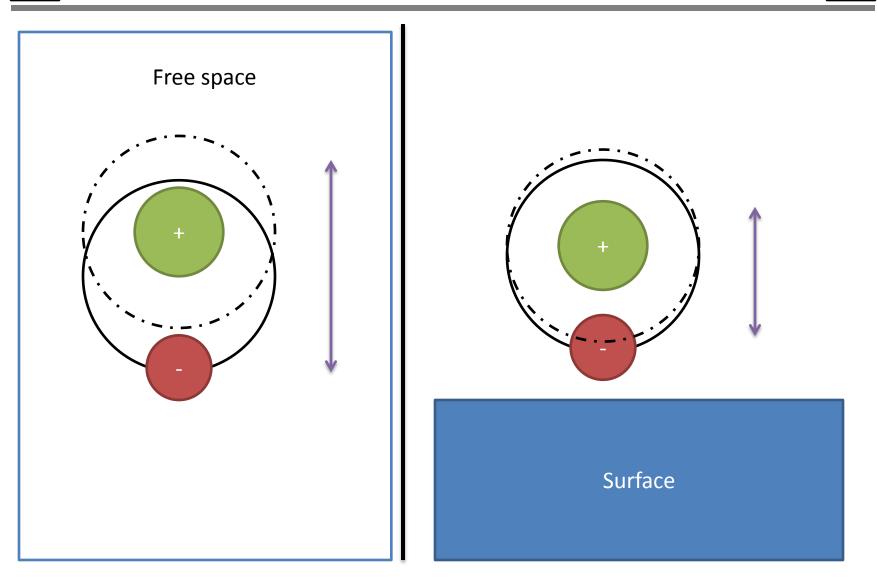
microscopic dipole moment

$$x(t) = x(t_0) \exp[-(\gamma/2)(t-t_0) + j\omega'_a(t-t_0)],$$

Siegman book p. 84-85



Siegman book p. 85-86



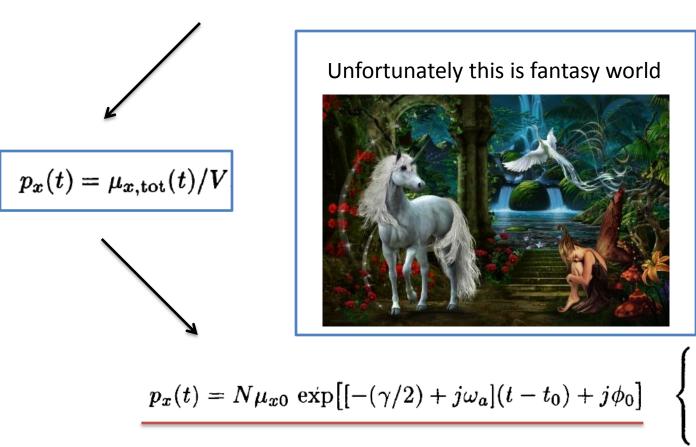
$$\frac{d^2\mu_x(t)}{dt^2} + \gamma \frac{d\mu_x(t)}{dt} + \omega_a^2 \mu_x(t) = (e^2/m)\mathcal{E}_x(t) \qquad \begin{array}{c} \text{x electron} \\ \text{chage} \end{array}$$
$$\mu_x(t) = [\text{charge}] \times [\text{displacement}] = -ex(t) \qquad \begin{array}{c} + \\ \text{microscopic} \\ \text{dipole} \end{array}$$

$$\mu_x(t) = \mu_{x0} \exp\left[-(\gamma/2)(t-t_0) + j\omega_a(t-t_0) + j\phi_0\right],$$

Siegman book p. 89-90

$$\mu_{x,\text{tot}}(t) = \sum_{i=1}^{NV} \mu_{x,i}(t) = NV \mu_x(t)$$

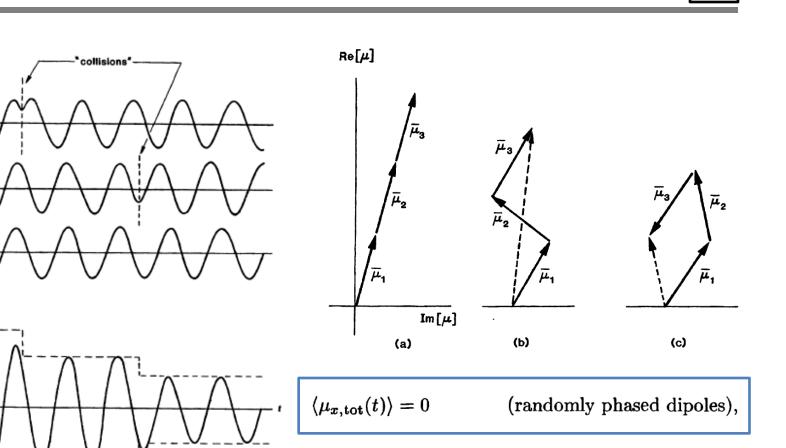
all dipoles oscillating in phase,



all dipoles oscillating in phase.

Siegman book p. 89-90

### **Random collisions**



 $\langle \mu_{x,\text{tot}}^2(t) \rangle^{1/2} = (NV)^{1/2} |\mu_x(t)|$  (randomly phased dipoles),

Siegman book p. 91-92

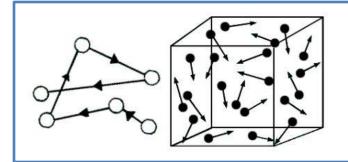
 $\mu_1(t)$ 

 $\mu_2(t)$ 

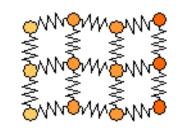
 $\mu_3(t)$ 

 $\mu_{tot}(t)$ 

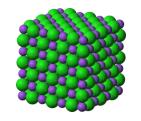
# Dephasing mechanisms



Brownian motion (atoms, or ions, or molecules). Even if coalition is elastic, no energy is transferred, electronic oscillation phase will be changed.



Solid state lasers, the quantum energy-level spacing and  $\omega$  are affected by neighboring atoms. Thermal vibrations of lattice, will modulate them.



In materials there atoms are sufficiently dense, oscillating dipole may spread out to, and be felt by, other neighboring laser atoms. Even week coupling tends to randomize overall response. (1e-13 sec)

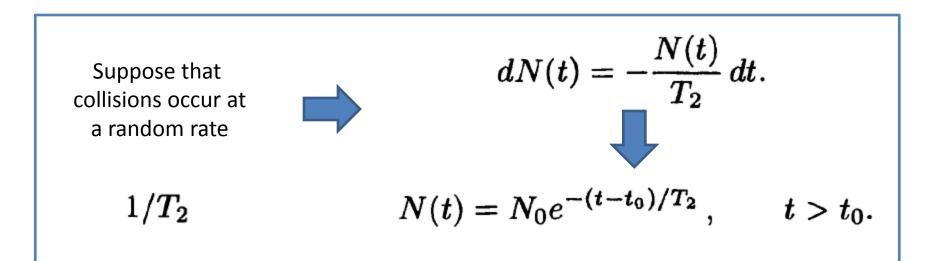
Macroscopic polarization initial state

 $p_x(t_0)=N_0\mu_{x0}.$ 

N(t)- dipoles that have not suffered at least one coalition

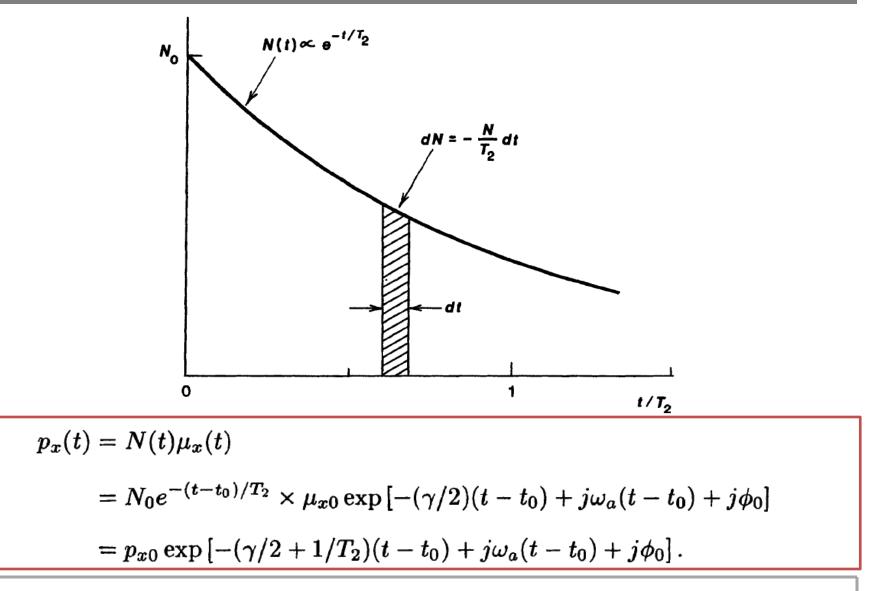
$$p_{\boldsymbol{x}}(t) = N(t)\mu_{\boldsymbol{x}}(t) = N(t)\mu_{\boldsymbol{x}\boldsymbol{0}}\cos\omega_a t,$$

Errata



Siegman book p. 94-95

### Decay of number of uncollided dipoles



$$p_{x}(t) = N(t)\mu_{x}(t)$$

$$= N_{0}e^{-(t-t_{0})/T_{2}} \times \mu_{x0} \exp\left[-(\gamma/2)(t-t_{0}) + j\omega_{a}(t-t_{0}) + j\phi_{0}\right]$$

$$= p_{x0} \exp\left[-(\gamma/2 + 1/T_{2})(t-t_{0}) + j\omega_{a}(t-t_{0}) + j\phi_{0}\right].$$

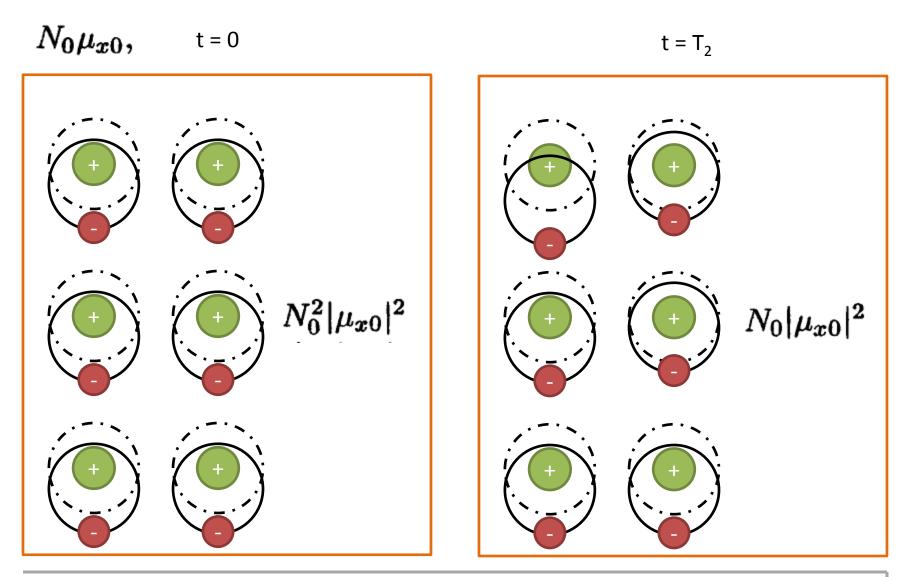
$$\begin{pmatrix} \frac{\gamma}{2} \end{pmatrix} \left( \begin{array}{c} \text{single-dipole} \\ \text{decay rate} \end{array} \right) \Rightarrow \left( \frac{\gamma}{2} + \frac{1}{T_{2}} \right) \left( \begin{array}{c} \text{macroscopic} \\ \text{polarization} \\ \text{decay rate} \end{array} \right).$$

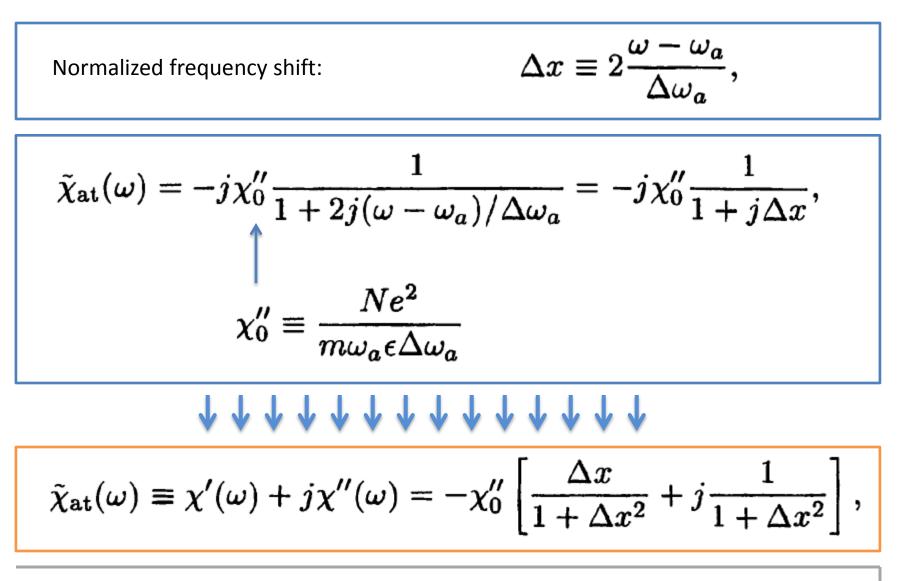
$$\frac{d^{2}\mu_{x}(t)}{dt^{2}} + \frac{\gamma}{d}\frac{d\mu_{x}(t)}{dt} + \omega_{a}^{2}\mu_{x}(t) = (e^{2}/m)\mathcal{E}_{x}(t)$$

$$\frac{d^{2}p_{x}(t)}{dt^{2}} + (\gamma + 2/T_{2})\frac{dp_{x}(t)}{dt} + \omega_{a}^{2}p_{x}(t) = (Ne^{2}/m)\mathcal{E}_{x}(t),$$

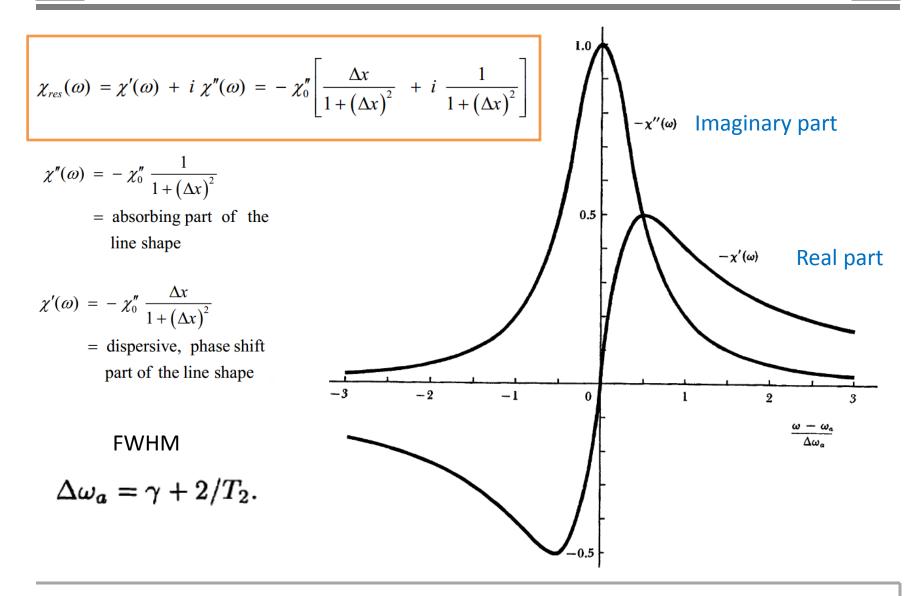
Siegman book p. 95-96

## Mental exercise

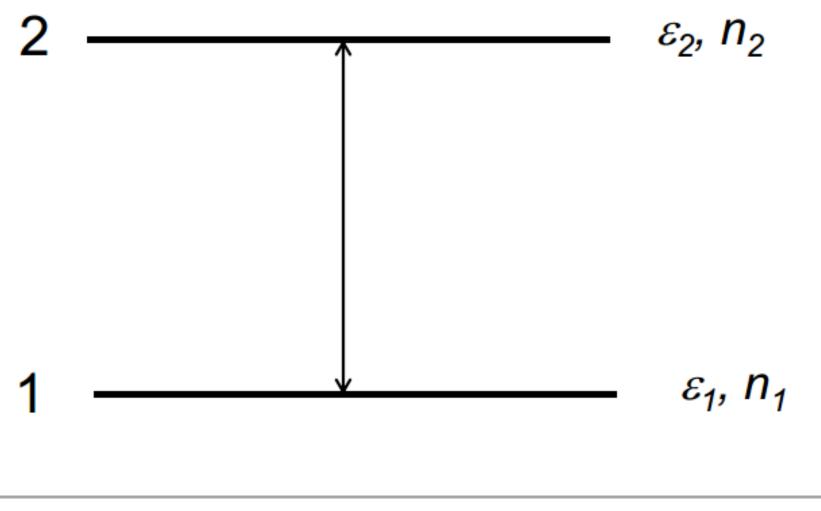




# A plot

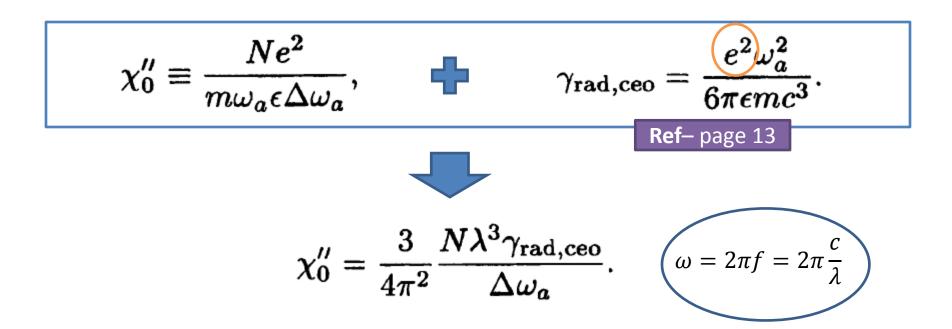


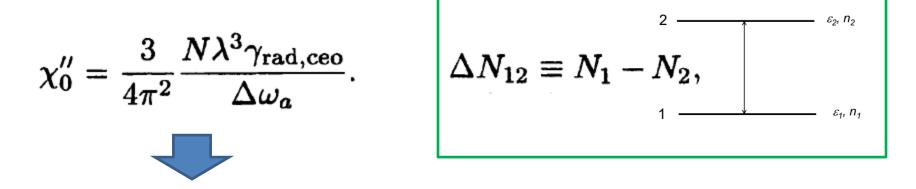
## Finally atomic levels



Random picture from somewhere

29





$$\tilde{\chi}_{\rm at}(\omega) = -j \frac{3}{4\pi^2} \frac{\Delta N \lambda^3 \gamma_{\rm rad}}{\Delta \omega_a} \frac{1}{1 + 2j(\omega - \omega_a)/\Delta \omega_a}.$$

$$\begin{split} \tilde{\chi}_{\rm at}(\omega) &= -j\chi_0'' \times \frac{1}{1+2j(\omega-\omega_a)/\Delta\omega_a} \\ &= -\chi_0'' \left[ \frac{2(\omega-\omega_a)/\Delta\omega_a}{1+\left[2(\omega-\omega_a)/\Delta\omega_a\right]^2} + j\frac{1}{1+\left[2(\omega-\omega_a)/\Delta\omega_a\right]^2} \right], \end{split}$$

Siegman book p. 110-111

$$\chi_0^{\prime\prime} = rac{3}{4\pi^2} rac{\Delta N \lambda^3 \gamma_{
m rad}}{\Delta \omega_a}.$$

Replace CEO with actual radiative decay rate

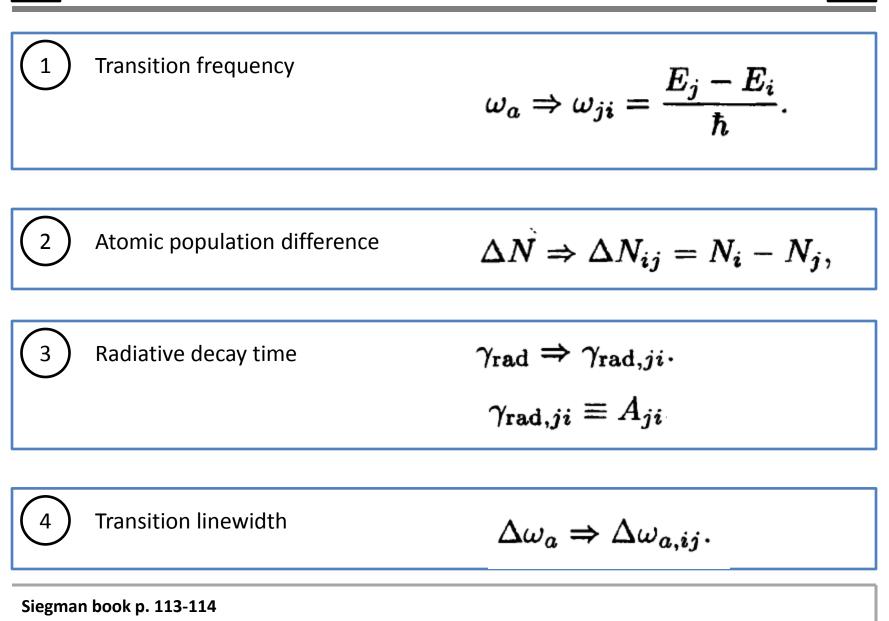
Classical 
$$N\lambda^3$$
 Quantum  $\Delta N\lambda^3$   
 $\lambda \equiv \lambda_0/n$  Spontaneous emission  
inverse linewidth  $1/\Delta \omega_a$   
is proportion to  
 $\Delta N\lambda^3\gamma_{rad}$   $\tilde{\chi}_{at}(\omega) \equiv \chi'(\omega) + j\chi''(\omega)$   
Frequency variations

Siegman book p. 111-112

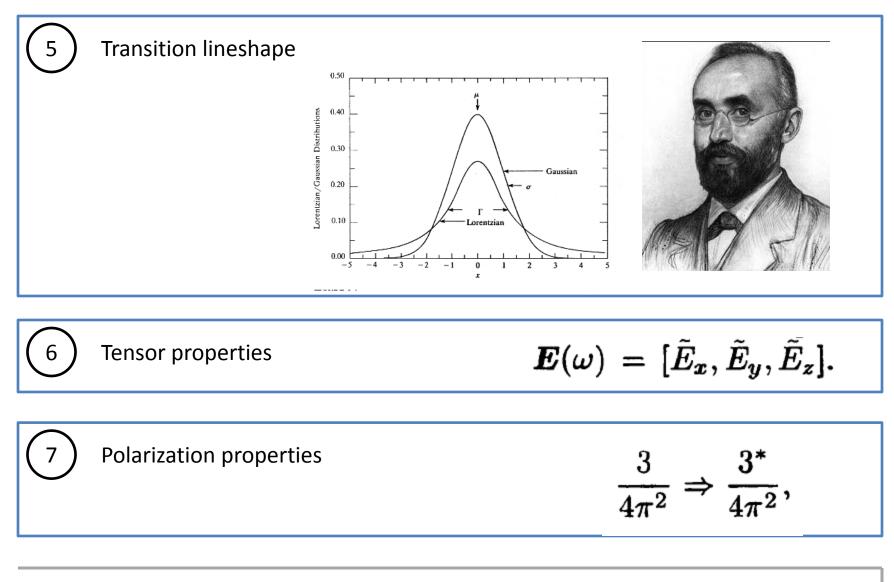
CEO  
$$\frac{d^2p_x(t)}{dt^2} + (\gamma + 2/T_2)\frac{dp_x(t)}{dt} + \omega_a^2 p_x(t) = (Ne^2/m)\mathcal{E}_x(t),$$

Quantum
$$\frac{d^2 p_x(t)}{dt^2} + \Delta \omega_a \frac{dp_x(t)}{dt} + \omega_a^2 p_x(t) = \frac{3\omega_a \epsilon \lambda^3 \gamma_{\rm rad}}{4\pi^2} \Delta N(t) \mathcal{E}_x(t),$$

### Substitutions #1

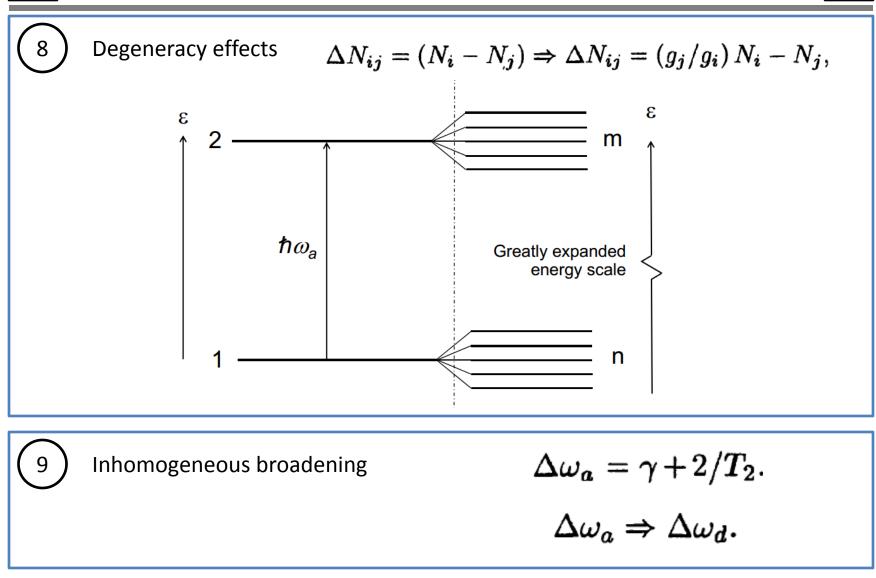


### Substitutions #2



Siegman book p. 113-114

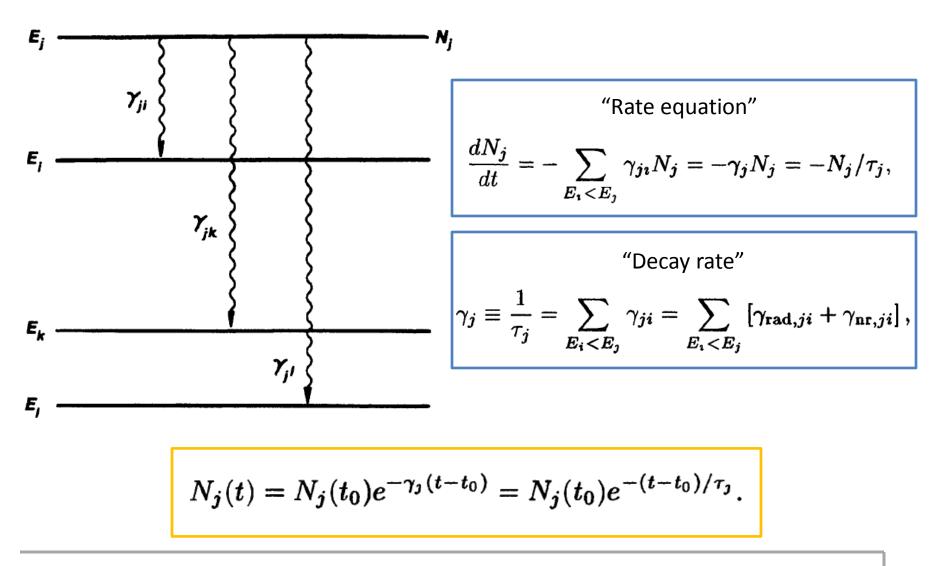




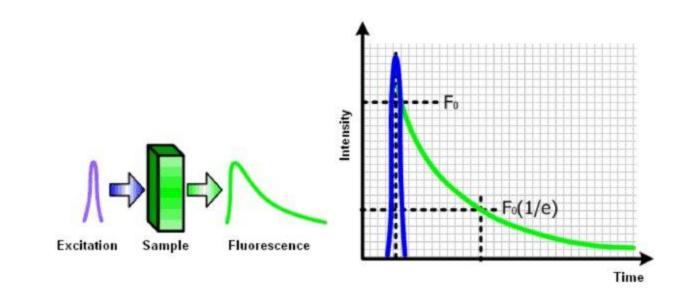
Siegman book p. 113-114

# ELECTRIC-DIPOLE TRANSITIONS IN REAL ATOMS





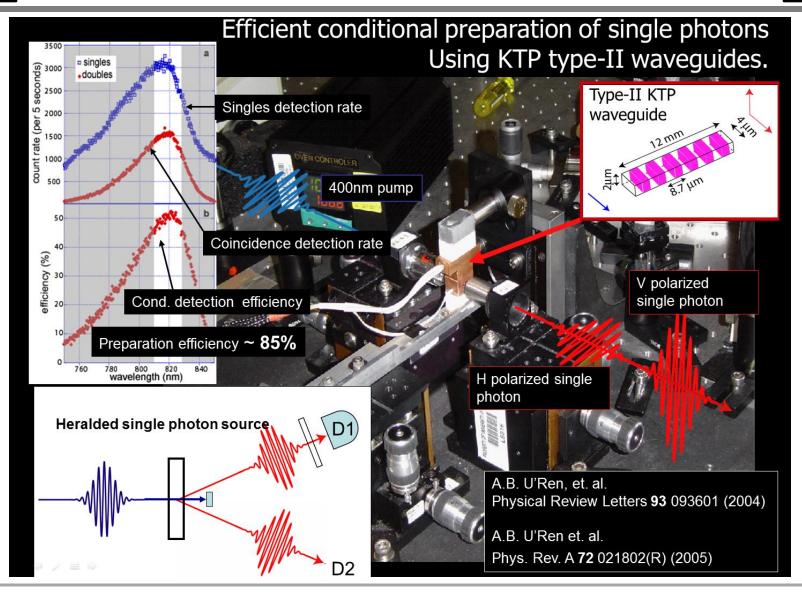
Siegman book p. 118-119



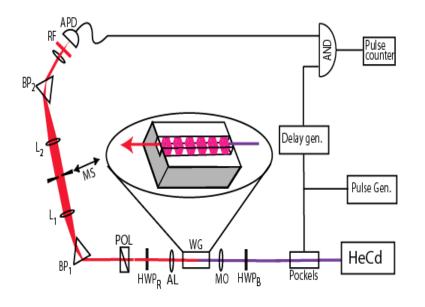
$$\begin{split} I_{\rm fl}(t) &= {\rm const} \times \gamma_{{\rm rad},j\imath} N_j(t). \\ I_{\rm fl}(t) &= {\rm const} \times N_j(t) = {\rm const} \times e^{-t/\tau_j}. \end{split}$$

Siegman book p. 119-120

## Example #1



#### Without authors permission

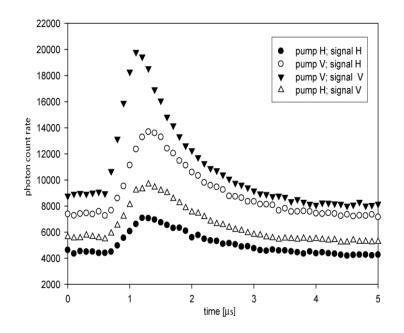


Fluorescence lifetime ~ 1-10  $\mu$ s [depending on waveguide]

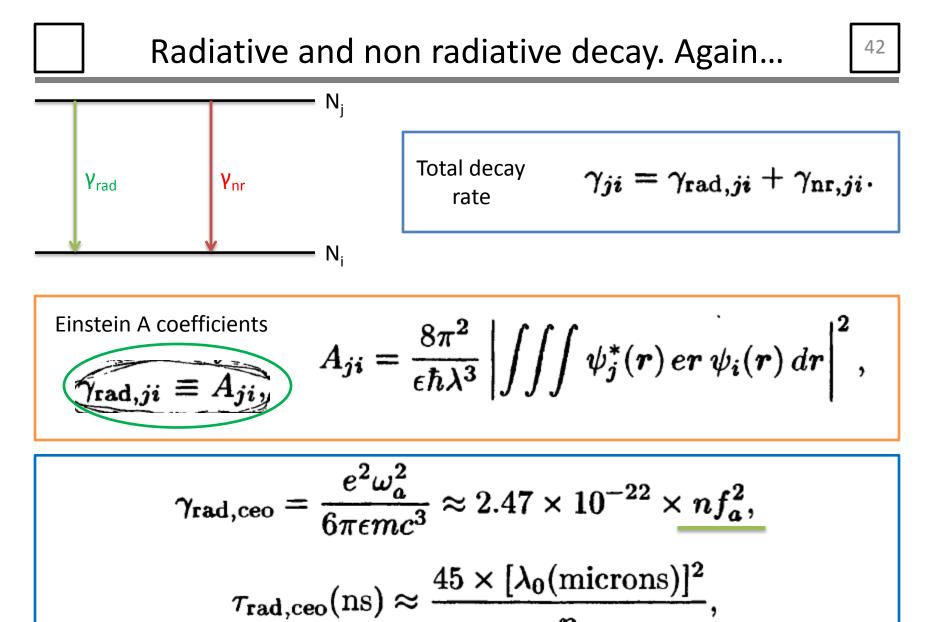
•Pulsed pump [HeCd laser + pockels cell]

•Time-gated photon counting

Can resolve fluorescence lifetime



Some unpublished data



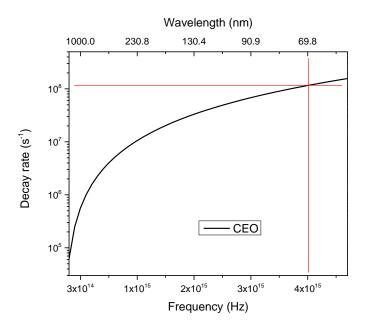
Siegman book p. 120-121

### Oscillator strength

$\mathcal{F}_{ji} \equiv rac{\gamma_{\mathrm{rad},i}}{3\gamma_{\mathrm{rad},i}}$	$rac{ji}{ceo} = rac{ au_{rad}}{3 au_{ra}}$	, <u>ceo</u> . d, <i>ji</i>	ieneral" <b>γ</b> r rule	$_{\mathrm{ad},ji} \leq \gamma_{\mathrm{rad},\mathrm{ce}}$
Examples:				
Transition	Wavelength	Radiative decay rate	Oscillator strength	Comments
Neodymium YAG l	aser transition:			
${}^4F_{3/2} \rightarrow {}^4I_{3/2}$	$1.064~\mu{ m m}$	$820 \ { m s}^{-1}$ (1.22 ms)	$pprox 8  imes 10^{-6}$	Measured $ au_2$ is 230 $\mu$ s
Ruby laser transition	on:			
$^{2}E \rightarrow {}^{4}A_{2}$	694 nm	$230 \text{ s}^{-1}$ (4.3 ms)	$\approx 10^{-6}$	Decay is almost purely radiative

Siegman book p. 121-123

### Radiative decay rates



Duplicate of slide 13

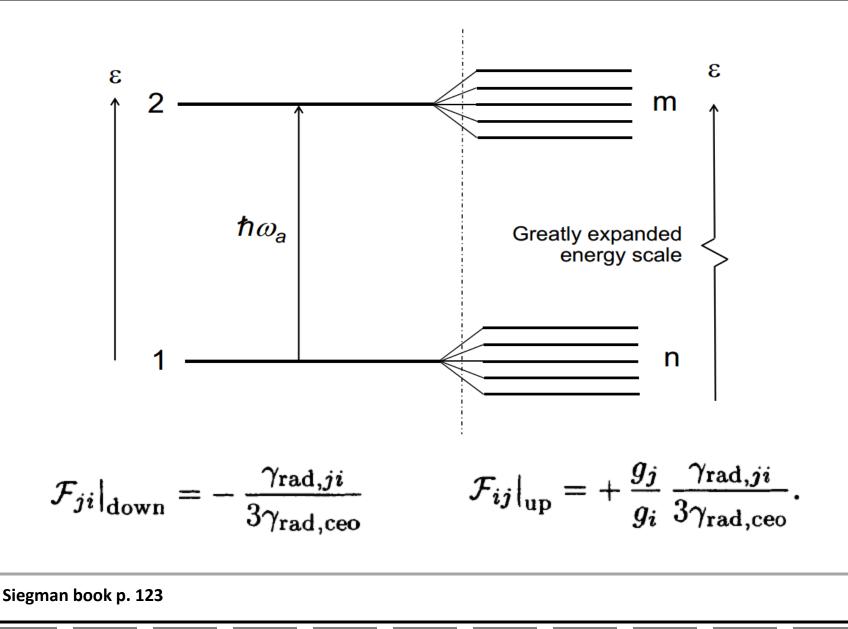
$$\gamma_{\rm rad,ceo} = \frac{e^2 \omega_a^2}{6\pi \epsilon m c^3}$$

Parameter	Value	
е	1.602e-19 (C)	
3	8.85e-12 (F/m)	
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С	3e8 (m/s)	

This classical oscillator radiative decay rate has a value  $\gamma_{\rm rad,ceo} \approx 10^8 \, {\rm sec^{-1}}$  for a visible frequency oscillator, compared to an oscillation frequency of  $\omega_a \approx 4 \times 10^{15}$   ${\rm sec^{-1}}$ .

Siegman book p. 84

### Level degeneracy #1



### Level degeneracy #2

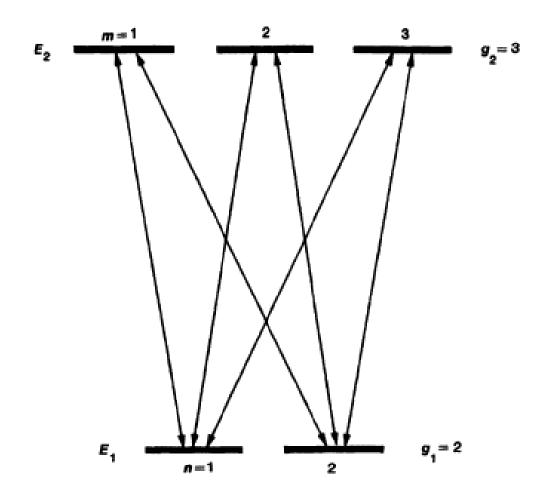
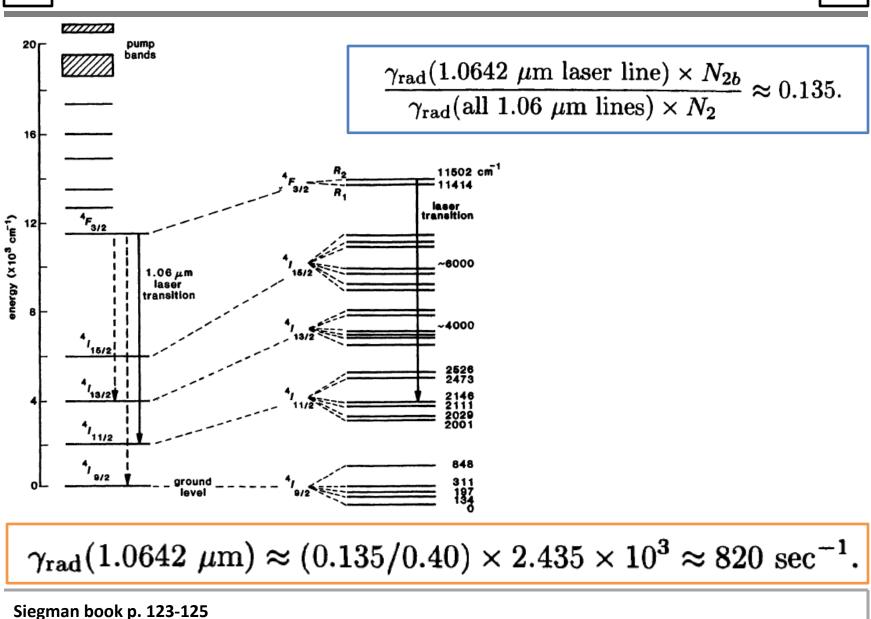
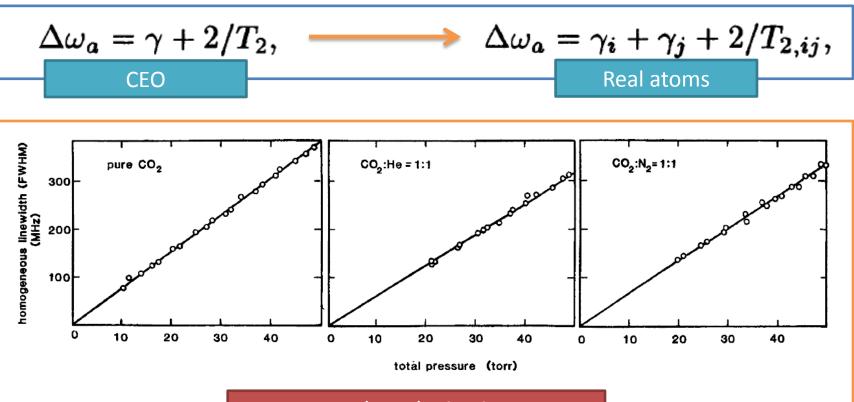


FIGURE 3.12 Degenerate sublevels of two quantum energy levels  $E_1$ and  $E_2$ . Each sublevel is a separate and distinct quantum energy eigenstate, but the degenerate sublevels all have the same energy eigenvalue.

#### Siegman book p. 155

### Nd:YAG transitions





#### Pressure broadening in gases

Homogeneous linewidth dependence:

 $\Delta \omega_a = A + BP,$ 

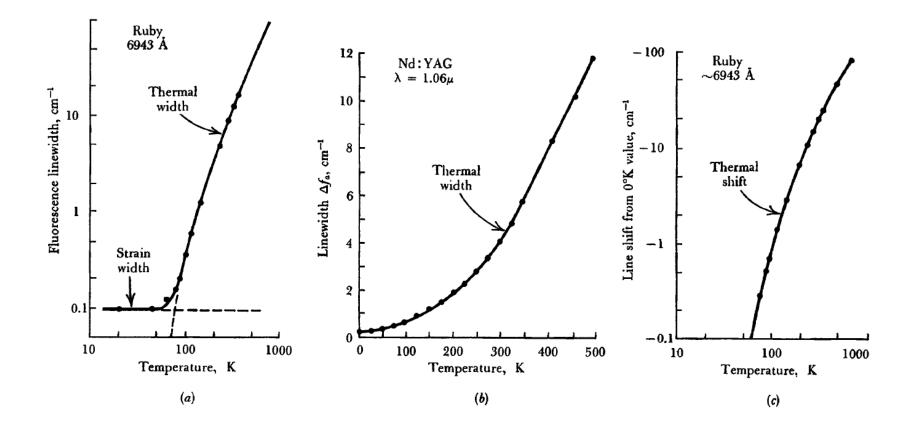
Siegman book p. 126-129

### Pressure-broadening coefficients

Wavelength	Collision partners	Pressure broadening	
Mercury resonance line:			
$2537 \text{\AA}$	$Hg + Ar, N_2, CO_2$	10–20 MHz/torr	Deletienskie ketween neutiel
Sodium resonance line:			Relationship between partial pressure and density of each
589 nm	Na + Na	$\approx 2000~{\rm MHz/torr}$	species in gas mixture.
He-Ne laser transitions:			
633 nm	He+Ne	$\approx 70~\mathrm{MHz/torr}$	$N(\text{atoms/cm}^3) = 9.65 \times 10^{18} \frac{P(\text{torr})}{T(K)}$
$3.39~\mu m$	He+Ne	50-80 MHz/torr	
CO <sub>2</sub> laser transition:			
10.6 $\mu$ m	$\rm CO_2 + \rm CO_2$	7.6 MHz/torr (5.8 GHz/atm)	
10.6 $\mu m$	$\rm CO_2$ + $\rm N_2$	5.5 MHz/torr (4.2 GHz/atm)	
10.6 $\mu m$	$CO_2 + He$	4.5 MHz/torr) (3.5 GHz/atm)	
10.6 $\mu m$	$\rm CO_2$ + H <sub>2</sub> O	2.9 MHz/torr (2.2 GHz/atm)	

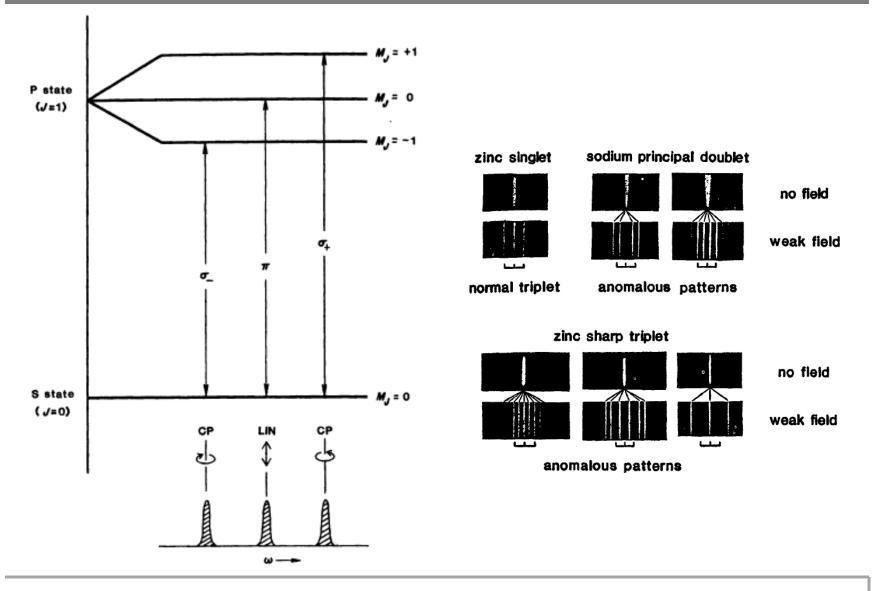
Siegman book p. 129

### Phonon broadening



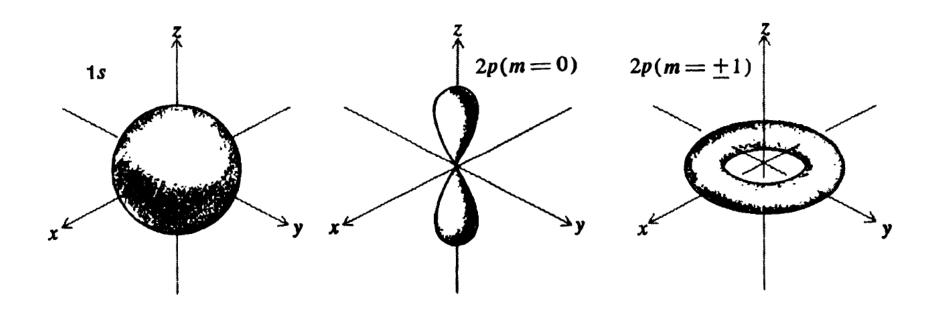
Siegman book p. 130-131

### Zeeman-split atomic transitions



Siegman book p. 136-137

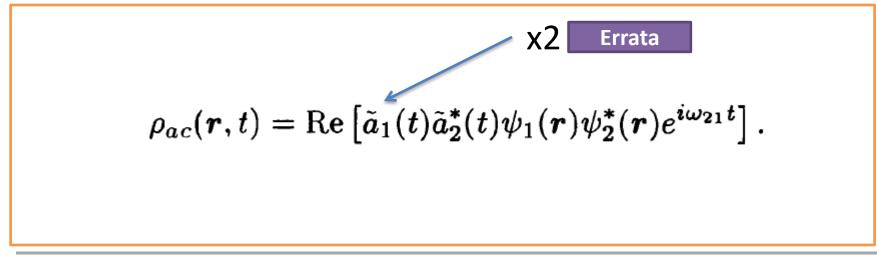
52



- The electron density is proportional to the product of the wavefunction and its complex conjugate.
- Since the electron density distribution does not change with time, the atom does not radiate in these stationary states.

Siegman book p. 139

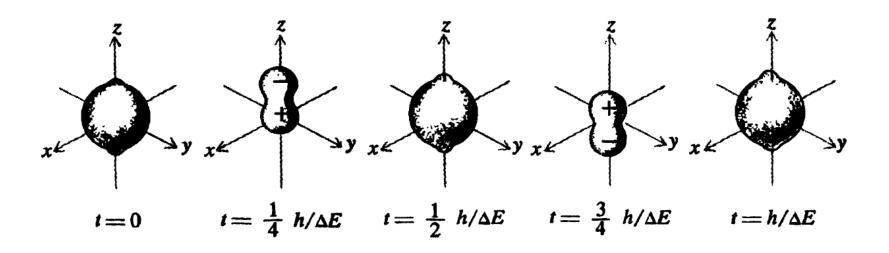
$$\begin{split} \rho(\mathbf{r},t) &= \left| \tilde{a}_1(t) e^{-iE_1 t/\hbar} \psi_1(\mathbf{r}) + + \tilde{a}_2(t) e^{-iE_2 t/\hbar} \psi_2(\mathbf{r}) \right|^2 \\ &= \left| \tilde{a}_1(t) \right|^2 \left| \psi_1(\mathbf{r}) \right|^2 + \left| \tilde{a}_2(t) \right|^2 \left| \psi_2(\mathbf{r}) \right|^2 \\ &+ \tilde{a}_1(t) \tilde{a}_2^*(t) \psi_1(\mathbf{r}) \psi_2^*(\mathbf{r}) \exp[i(E_2 - E_1) t/\hbar] \\ &+ \tilde{a}_1^*(t) \tilde{a}_2(t) \psi_1^*(\mathbf{r}) \psi_2(\mathbf{r}) \exp[-i(E_2 - E_1) t/\hbar] \\ &= \rho_{dc}(\mathbf{r}) + \rho_{ac}(\mathbf{r}, t). \end{split}$$



Siegman book p. 138

### Linear dipole

linear dipole: S(M=0) + P(M=0) states

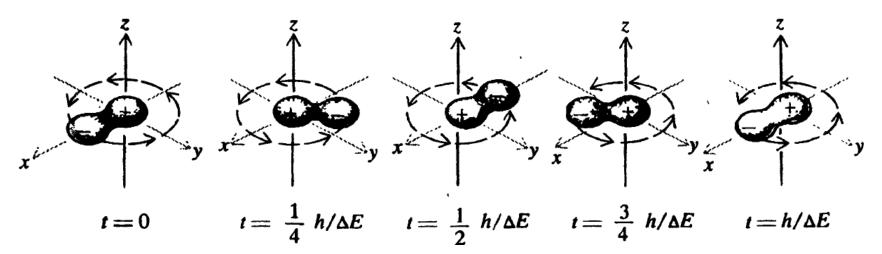


Laser radiation that is linearly polarized in the z-direction couples very efficiently with superposition states with the same M<sub>J</sub> value.

Siegman book p. 140

### Circular dipole

circular dipole:  $S(M=0) + P(M=\pm 1)$  states

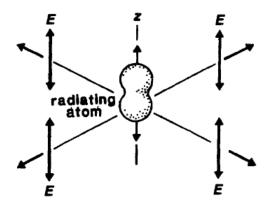


Laser radiation that is circularly polarized in the x-y plane couples very efficiently with superposition states with MJ value that differ by +1 or -1.

Siegman book p. 140

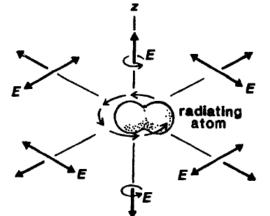
### Linear and circular micro-summary

#### $\pi$ transitions





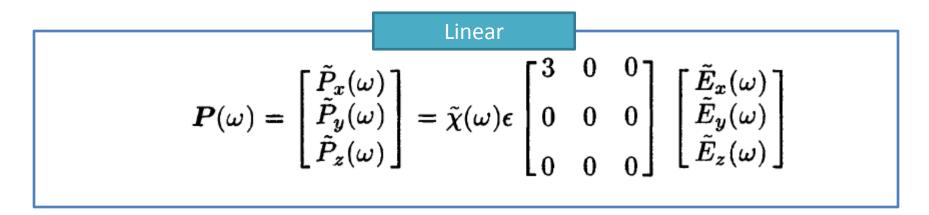
#### σ transitions



Atom whose charge distribution can oscillate only in a certain direction on a given transition will obviously respond only to applied fields that have the same direction or sense of polarization

Siegman book p. 141

$$\boldsymbol{P}(\omega) = \boldsymbol{\chi}(\omega) \boldsymbol{\epsilon} \boldsymbol{E}(\omega),$$



 $P = \begin{bmatrix} \tilde{P}_{x} \\ \tilde{P}_{y} \\ \tilde{P}_{z} \end{bmatrix} = \tilde{\chi}(\omega)\epsilon \times \frac{3}{2} \begin{bmatrix} 1 & \mp j & 0 \\ \pm j & 1 & 0 \\ 0 & 0 & 0 \end{bmatrix} \begin{bmatrix} \tilde{E}_{x} \\ \tilde{E}_{y} \\ \tilde{E}_{z} \end{bmatrix}.$ 

Siegman book p. 143-145

### The "factor of three"

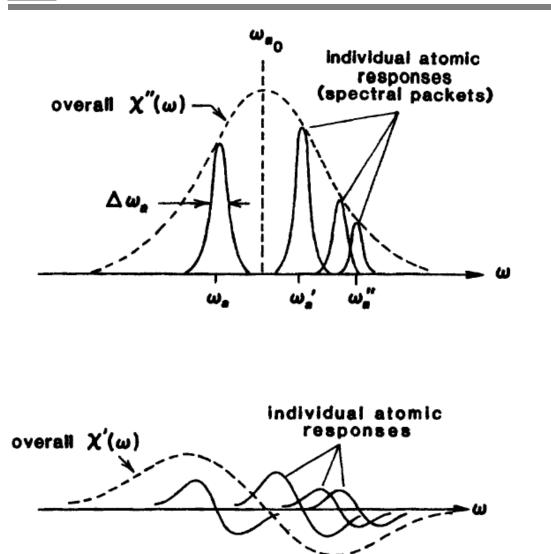
#### TABLE 3.3 Normalized tensor responses

Saturated Tensor Form	Gain Applied Field Polarization	Normalized Response	
$\overrightarrow{\text{Circular}, x \to \pm y}$	Circular, $x \rightarrow \pm y$		
Circular, $x \to \pm y$	Circular, $x \to \mp y$	0	
Circular, $x \to \pm y$	Linear $(x \text{ or } y)$	1.5	
Circular, $x \to \pm y$	Linear (x)	0	
Circular, $x \to \pm y$	Random	1	
Linear $(x)$	Linear (x)	3	
Linear $(x)$	Linear (angle $\theta$ from $x$ )	$3\cos^2\theta$	
Linear $(x)$	Circular, $x \to \pm y$	1.5	
L\inear	Random	1	
Linear $(x)$	Linear $(y \text{ or } x)$	0	
Isotı•opic	Arbitrary	1	

Siegman book p. 150-153



Siegman book p. 158



#### Gasses

Different atoms will have different kinetic velocities through space

### Solids

Atoms at different sites in a crystal may see slightly different local surroundings, or different local crystal structures, because of defects, dislocations, or lattice impurities

Siegman book p. 158-159



Average Doppler shift

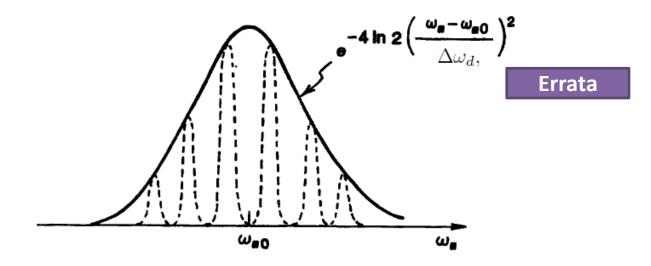
$$\frac{\omega_a - \omega_{a0}}{\omega_{a0}} \approx \sqrt{\frac{kT}{Mc^2}} \approx 10^{-6}$$

for typical atomic masses and temperatures.

Siegman book p. 160

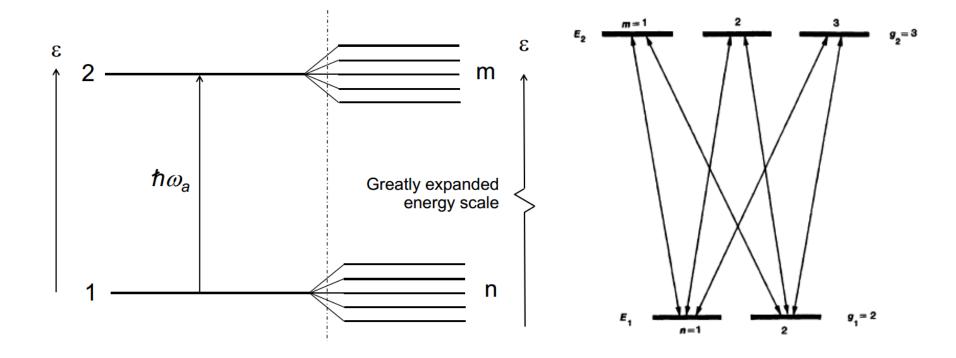
Gaussian distribution in gasses

$$g(v_z) = \left(\frac{1}{2\pi\sigma_v^2}\right)^{1/2} \exp\left(-\frac{v_z^2}{2\sigma_v^2}\right)$$

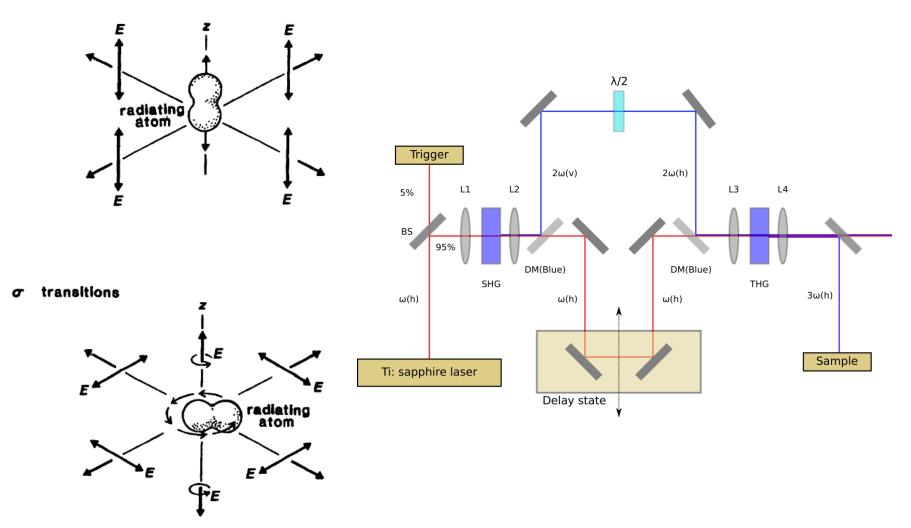


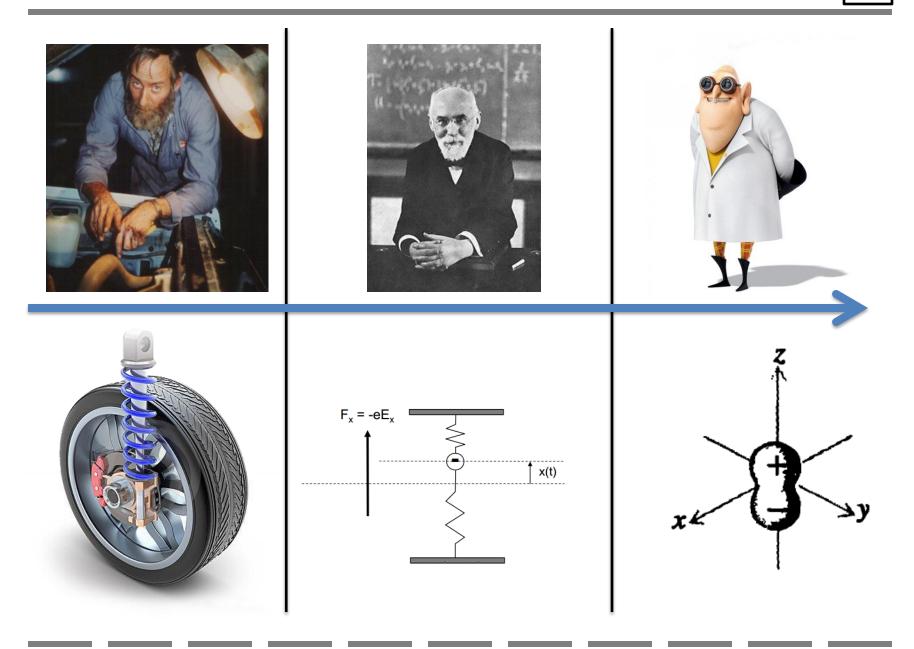
Siegman book p. 160-161

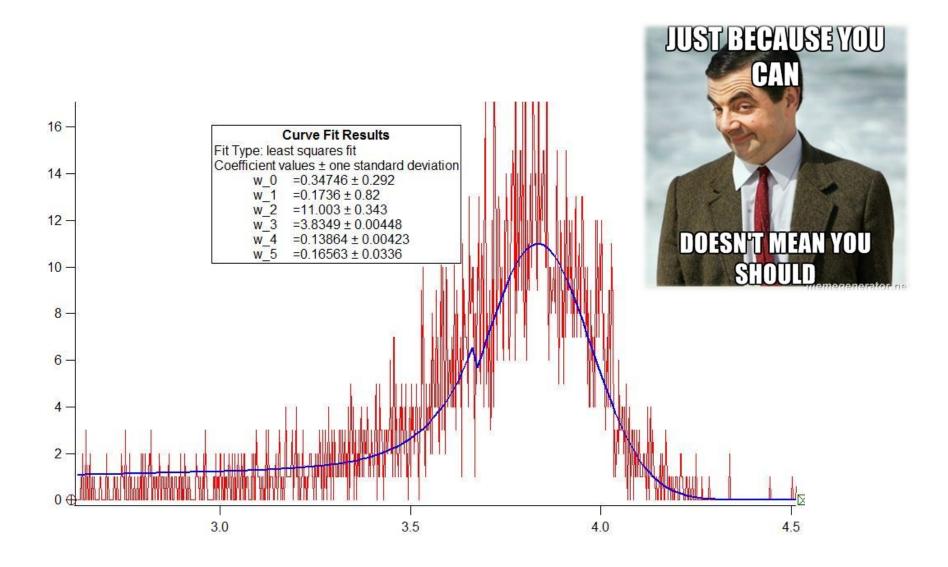


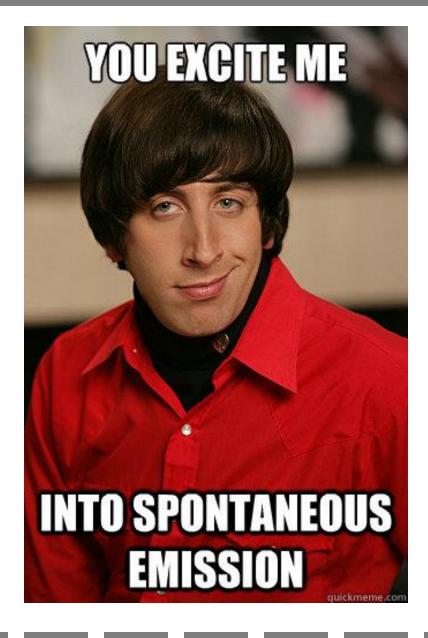


 $\pi$  transitions









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