Ultrafast Optical Physics II (SoSe 2017) Lecture 5, May 8

Laser dynamics: semi-classical laser theory

(1) Two-level system and Bloch equations

(2) Rabi oscillation: coherent light-matter interaction

(3) Steady-state solution of Bloch equations: linear
   susceptibility

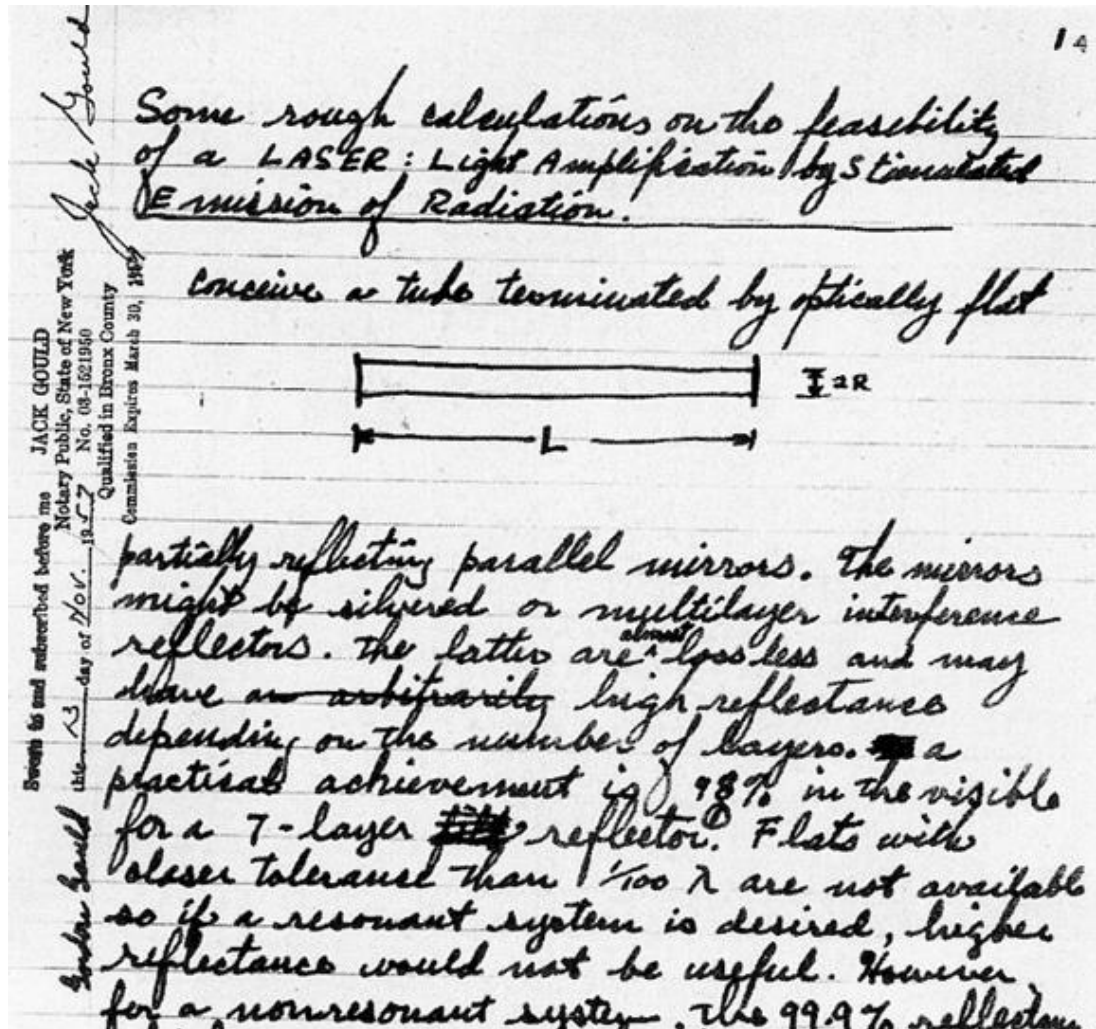(4) Adiabatic solution of Bloch equations: laser rate
   equation

Early history of lasers

- 1917: *on the quantum theory of radiation* – Einstein's paper
- 1954: MASER by Charles Townes (1915—2015) *et al.*
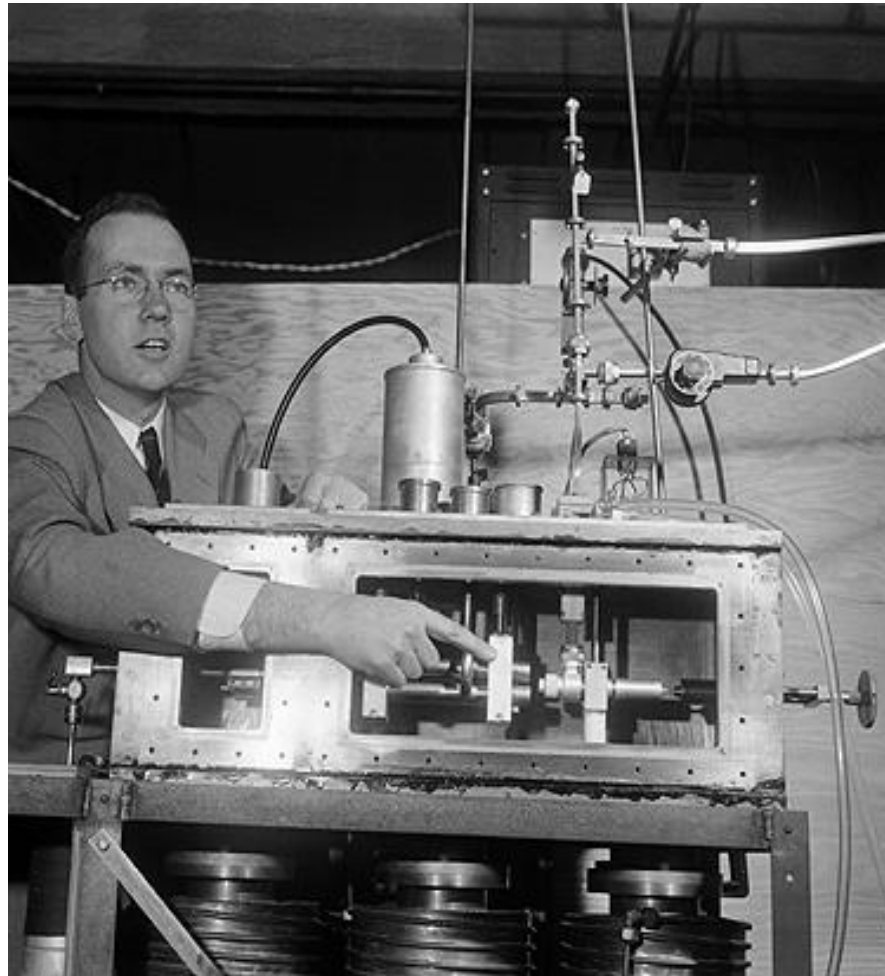
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PHYSICAL REVIEW   VOLUME 112, NUMBER 6   DECEMBER 15, 1958

Infrared and Optical Masers

A. L. SCHAWLOW AND C. H. TOWNES*
Bell Telephone Laboratories, Murray Hill, New Jersey
(Received August 26, 1958)

The extension of maser techniques to the infrared and optical region is considered. It is shown that by using a resonant cavity of centimeter dimensions, having many resonant modes, maser oscillation at these wavelengths can be achieved by pumping with reasonable amounts of incoherent light. For wavelengths much shorter than those of the ultraviolet region, maser-type amplification appears to be quite impractical. Although use of a multimode cavity is suggested, a single mode may be selected by making only the end walls highly reflecting, and defining a suitably small angular aperture. Then extremely monochromatic and coherent light is produced. The design principles are illustrated by reference to a system using potassium vapor.

LASER: Light Amplification by Stimulated Emission of Radiation

LOSER: Light Oscillation by Stimulated Emission of Radiation

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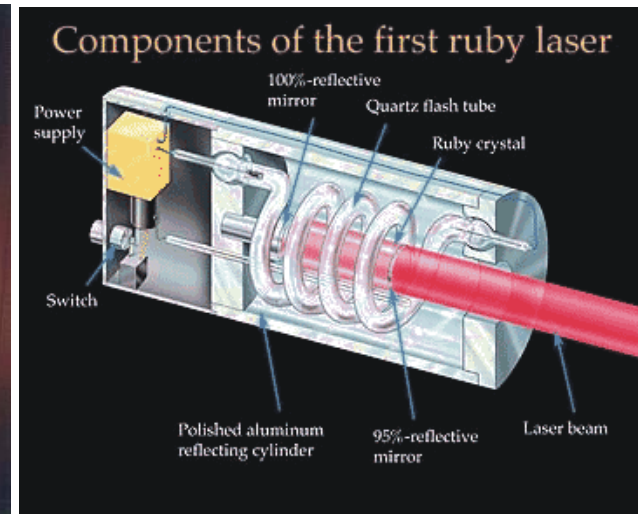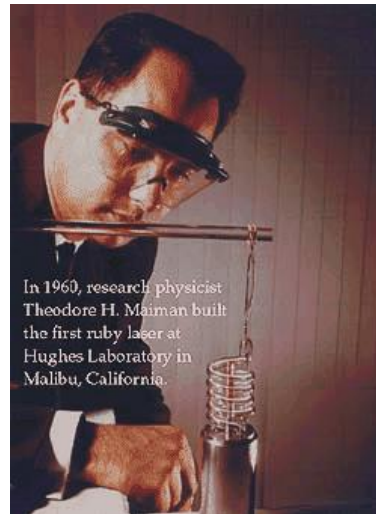
- 1960: First laser (Ruby) by Maiman

August 6, 1960   NATURE   No. 4736   493

Stimulated Optical Radiation in Ruby

Schawlow and Townes[1] have proposed a technique for the generation of very monochromatic radiation in the infra-red optical region of the spectrum using an alkali vapour as the active medium. Javan[2] and Sanders[3] have discussed proposals involving electron-excited gaseous systems. In this laboratory an optical pumping technique has been successfully applied to a fluorescent solid resulting in the attainment of negative temperatures and stimulated optical emission at a wave-length of 6943 Å.; the active

T. H. MAIMAN

Hughes Research Laboratories,
A Division of Hughes Aircraft Co.,
Malibu, California.

In 1960, research physicist Theodore H. Maiman built the first ruby laser at Hughes Laboratory in Malibu, California.

Components of the first ruby laser

Power supply
100%-reflective mirror
Quartz flash tube
Ruby crystal
Switch
Polished aluminum reflecting cylinder
95%-reflective mirror
Laser beam

Early history of lasers

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- 1958: Charles Townes (Nobel Prize in 1964) and Schawlow (Nobel Prize in 1981) conceive basic ideas for a laser.

- 1960: First laser (Ruby) by Maiman

- 1960: First HeNe laser by Ali Javan at Bell Labs

- 1961: Lasers (Ruby) first became commercially available

- 1962: First Semiconductor injection laser by Robert Hall at GE

- 1977: Gordon Gould awarded the patent for the laser.

Laser basics: three key elements

Continuous wave (CW) laser
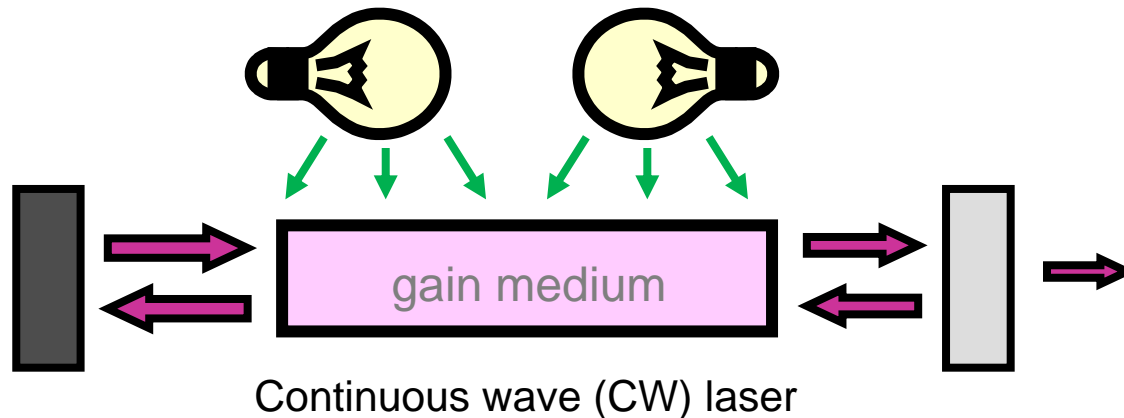
- **Gain medium**
  - Enable stimulated emission to produce identical copies of photons
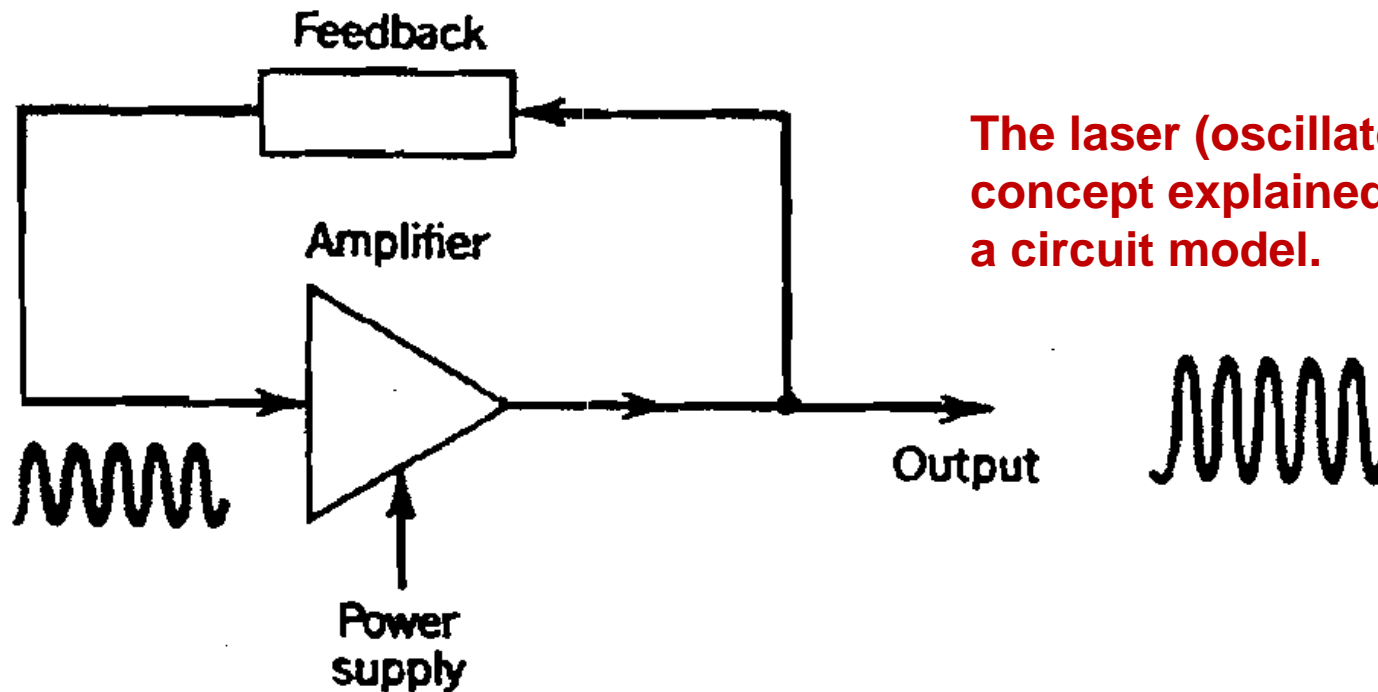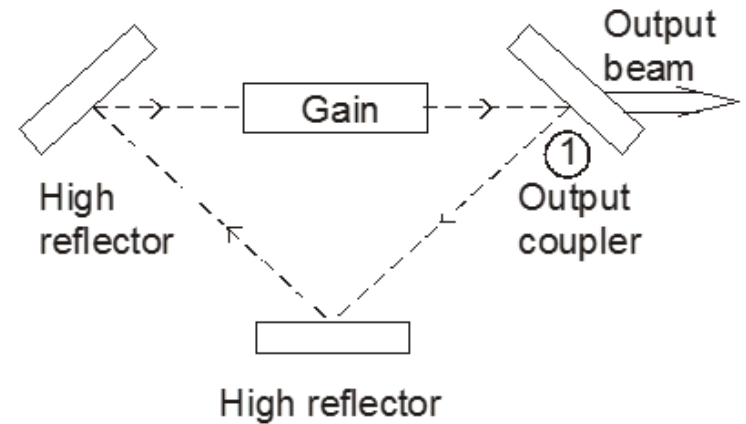 - Determine the light wavelength

- **Pump**
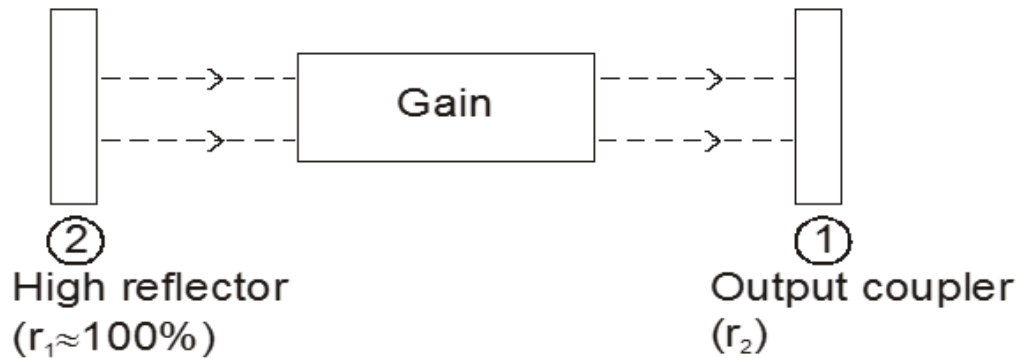 - Inject power into the gain medium
 - Achieve population inversion

- **Resonator cavity**
  - make light wave oscillating to efficiently extract energy stored in the gain medium
  - Improve directionality and color purity of the light

Possible laser cavity configurations

High reflector ($r_1 \approx 100\%$)

Output coupler (r_2)

The laser (oscillator) concept explained using a circuit model.

Three phenomena related to light-matter interaction in laser

$$\frac{dN_2}{dt} = -AN_2 = -\frac{N_2}{\tau_{21}}$$

Spontaneous emission

$$\frac{dN_1}{dt} = -BN_1 I$$

Absorption
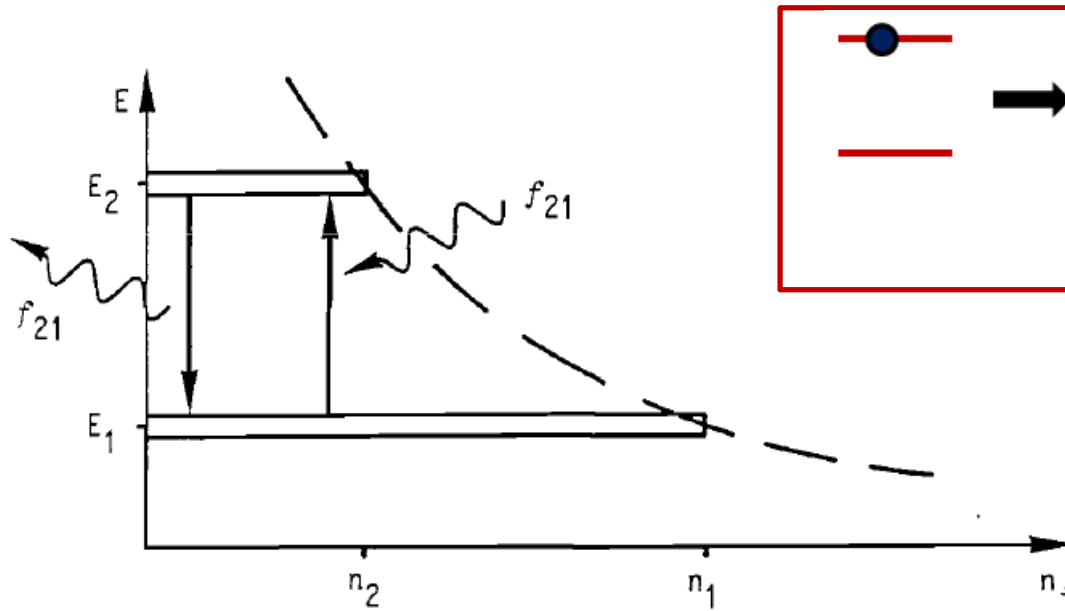
N_1 (N_2) is number of atoms in the lower (higher) energy level.
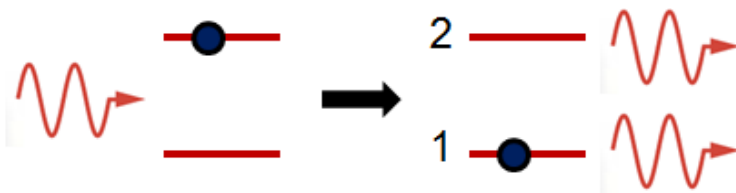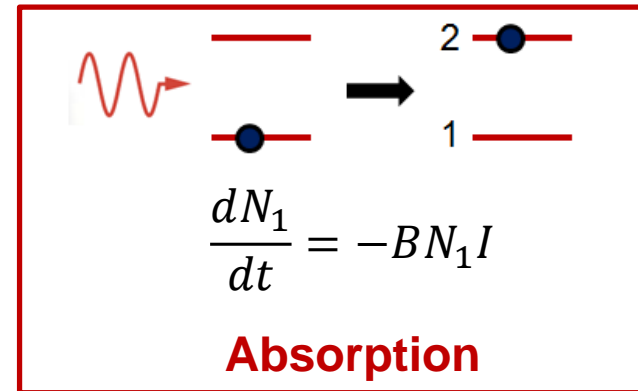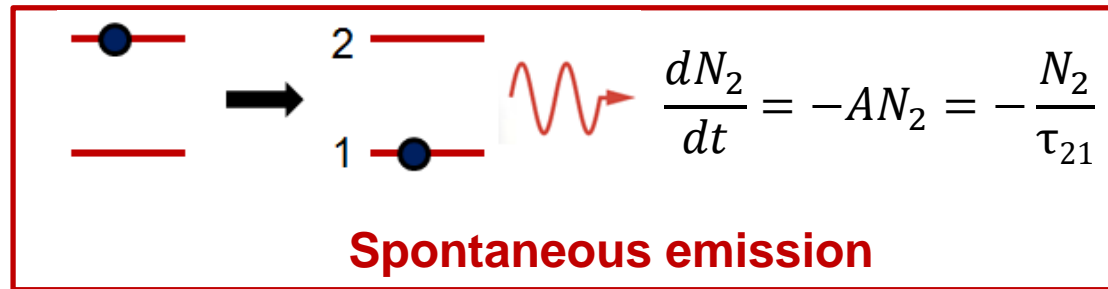
$$\frac{dN_2}{dt} = -BN_2 I$$

An incident photon triggers an excited atom to transit to a lower state by generating a second photon, which is a "clone" of the incident one—same freq., polarization, and traveling direction etc.

Stimulated emission (Laser: Light Amplification by Stimulated Emission of Radiation)

Population inversion for amplification

Amplification requirement: $N_2 > N_1$

E_m

$$\frac{N_2}{N_1} = \frac{g_2}{g_1} \exp\left(\frac{-(E_2 - E_1)}{kT}\right)$$

?

N_m
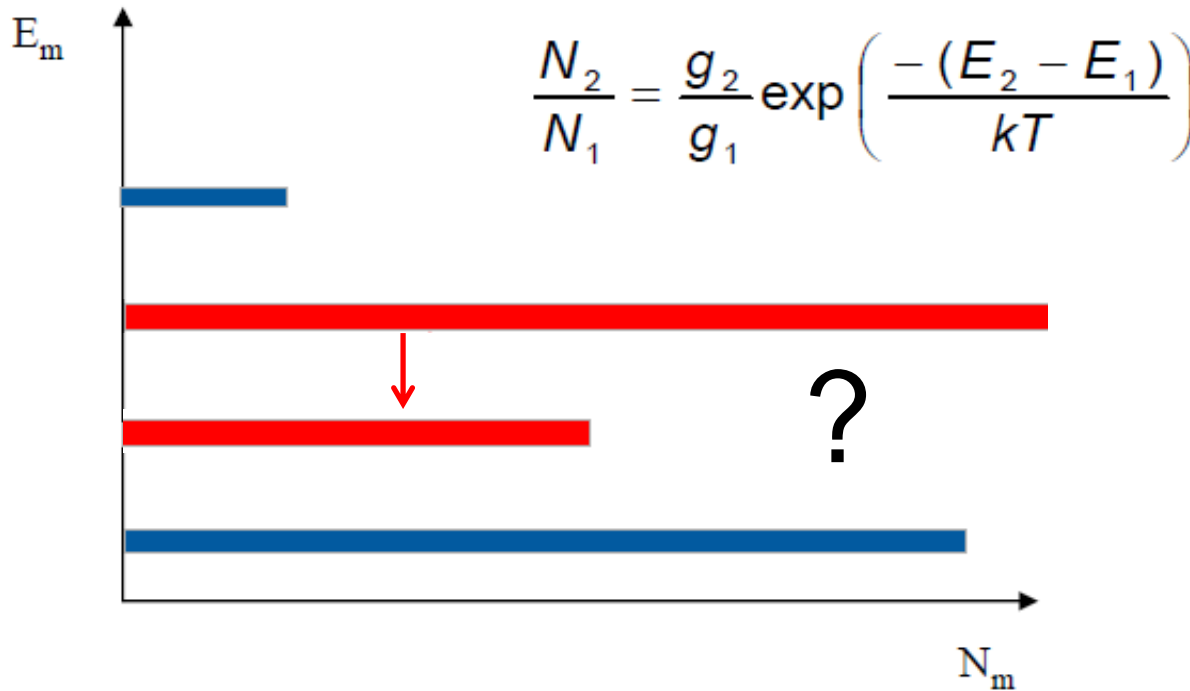
N_i is the number density (also known as the population density) of molecules in state i (i.e., the number of molecules per cm^3).

T is the temperature, and k is Boltzmann's constant = 1.3806503 × 10^{-23} J/°K

Under thermal equilibrium conditions, the lower energy levels are populated first, and are always more populated than the higher levels.

Four-level system with optical pumping

Typical lifetime of energy level 2:

Nd:YAG	~230 µs
Nd:YLF	~520 µs

3

2

808 nm
pump

1064 nm
Nd:YAG
laser

1

0
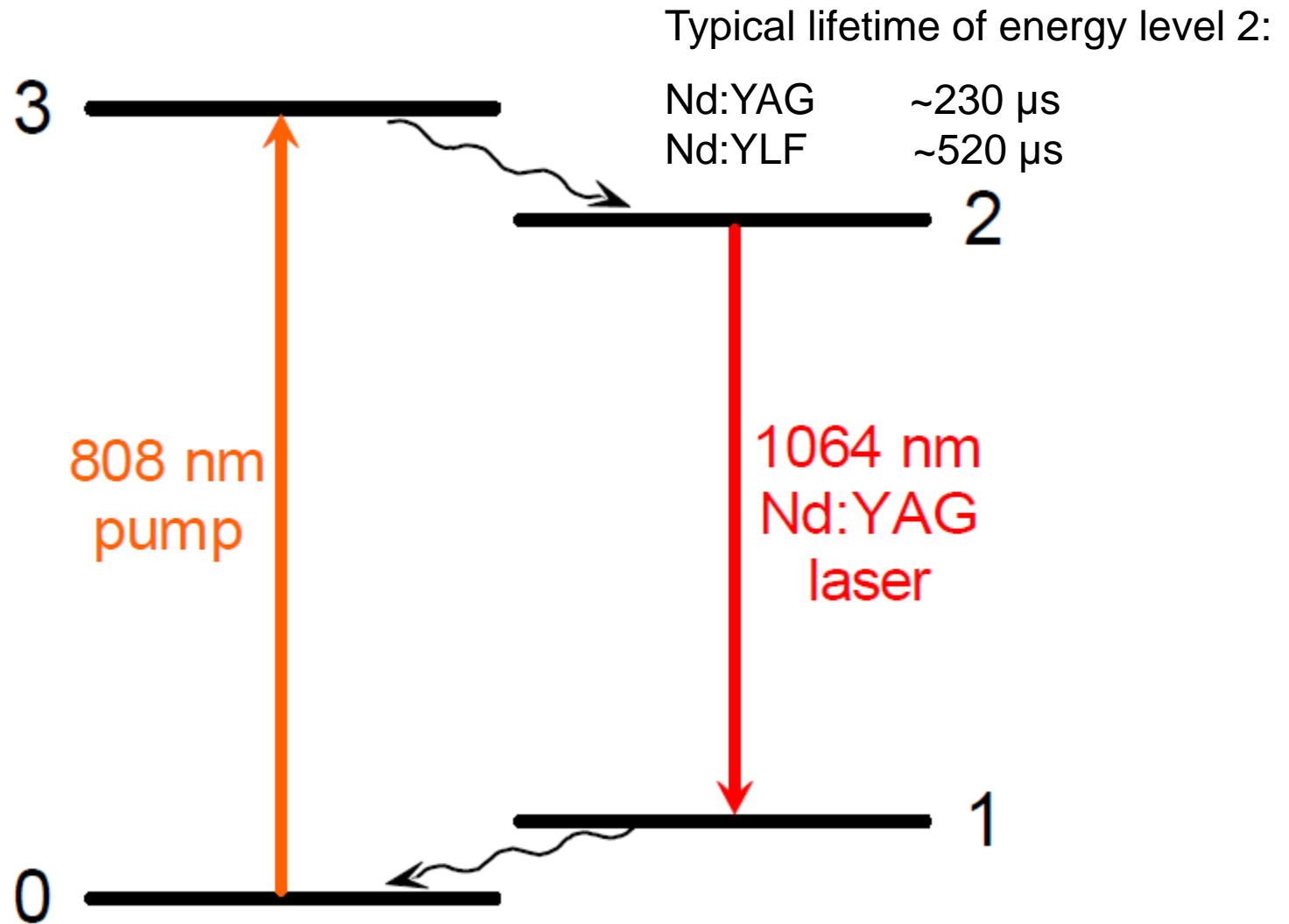
Light-matter interaction: classical harmonic oscillator model

1) Light is modeled by Maxwell eqns.

2) Mater is modeled as harmonic oscillator.

3) Two sets of equations: Maxwell equations + refractive index (i.e. dipole equation).

4) Explains linear optics (dispersion, absorption)

Pos. Charge   Neg. Charge
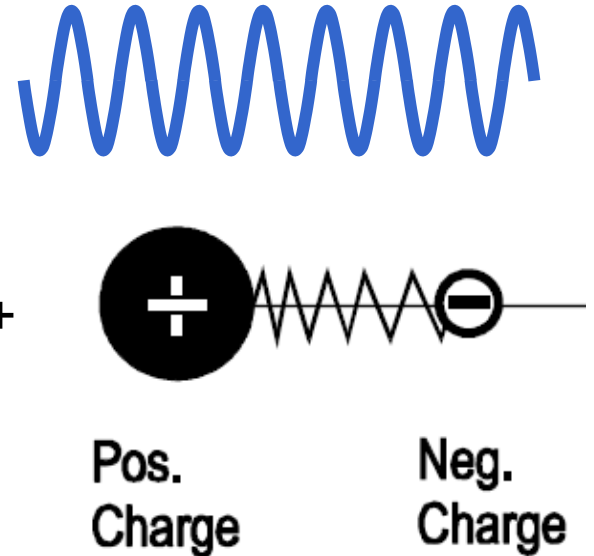
$$\widetilde{\underline{\chi}}(\Omega) = \frac{\omega_p^2}{(\Omega_0^2 - \Omega^2) + 2j\Omega\dfrac{\Omega_0}{Q}}$$

$$\left.\frac{\partial \underline{A}(z,t')}{\partial z}\right|_{(gain)} = g\left(1 + \frac{1}{\Omega_g^2}\frac{\partial^2}{\partial t^2}\right)\underline{A}(z,t') \quad + \text{dispersion}$$

HWHM – gain bandwidth

Light-matter interaction: semi-classical model

1) Light is treated as a non-quantized, classical electromagnetic field (modeled by Maxwell equations).

2) Mater is quantized (resulting energy levels) by a non-relativistic quantum-mechanical approach.

3) Three sets of Eqns (Maxwell-Bloch Eqns):

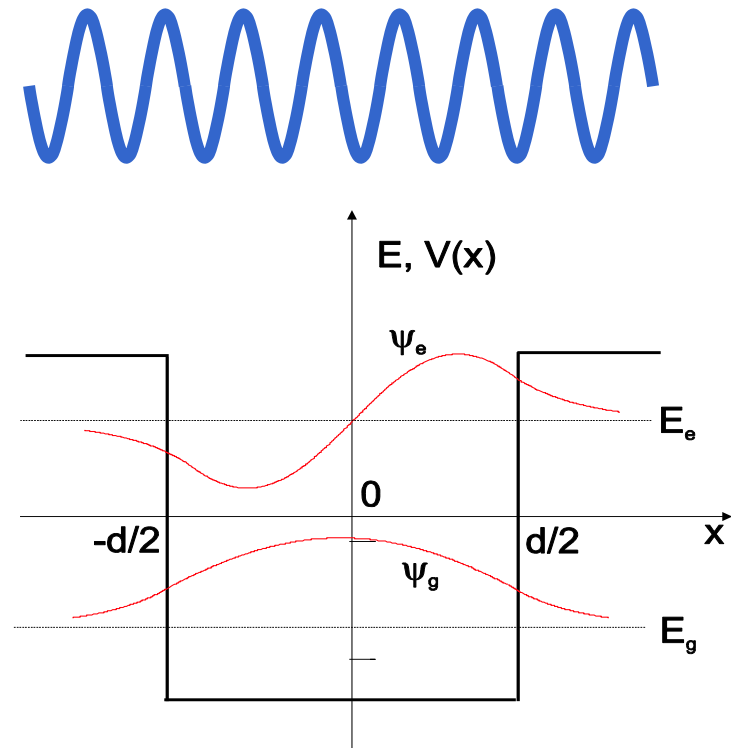   E: Maxwell equations (pulse duration T)

   w: population inversion (decay with time constant T_1)

   d: dipole moment (decay with time constant T_2 dephasing time)

4) Explains stimulated emission, absorption, Rabi oscillation etc.

E, V(x)

ψ_e

E_e

0

-d/2

d/2

x

ψ_g

E_g

Superposition states and radiative transitions

For energy eigen state:

$$\langle \vec{r} \rangle = \int_{-\infty}^{\infty} \vec{r} \, |\Psi(\vec{r}, t)|^2 \, d^3\vec{r} = 0$$

$$\vec{p} = -e \langle \vec{r} \rangle \longrightarrow \quad \bullet$$

Average dipole moment vanishes. Therefore the atom does not radiate in a stationary state as postulated in the Bohr model.

For superposition state: **1s + 2p (m=0)**

$$\frac{1}{\sqrt{2}} \left(\psi_{100}(\vec{r}, t) + \psi_{210}(\vec{r}, t) \right) = \frac{1}{\sqrt{2\pi}\sqrt{r_1^3}} \left(e^{-r/r_1} e^{-jE_1 t/\hbar} + \frac{1}{4\sqrt{2}} \frac{r}{r_1} e^{-r/2r_1} \cos\vartheta \, e^{-jE_2 t/\hbar} \right)$$

In the probability density (i.e. the magnitude square of the wave function), the contributions between the ground state and excited state interfer positively or negatively depending on the relative phase between the two wave functions, which depends on the phase angle $\Delta E t/\hbar$, with $\Delta E = E_2 - E_1$.

$$t=0 \qquad t = \frac{1}{4} \, h/\Delta E \qquad t = \frac{1}{2} \, h/\Delta E \qquad t = \frac{3}{4} \, h/\Delta E \qquad t = h/\Delta E$$
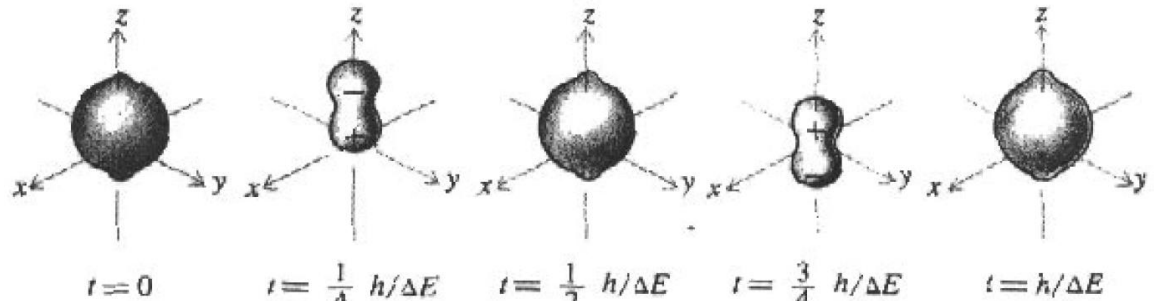
Oscillating dipole moment emits new EM wave at the oscillating frequency
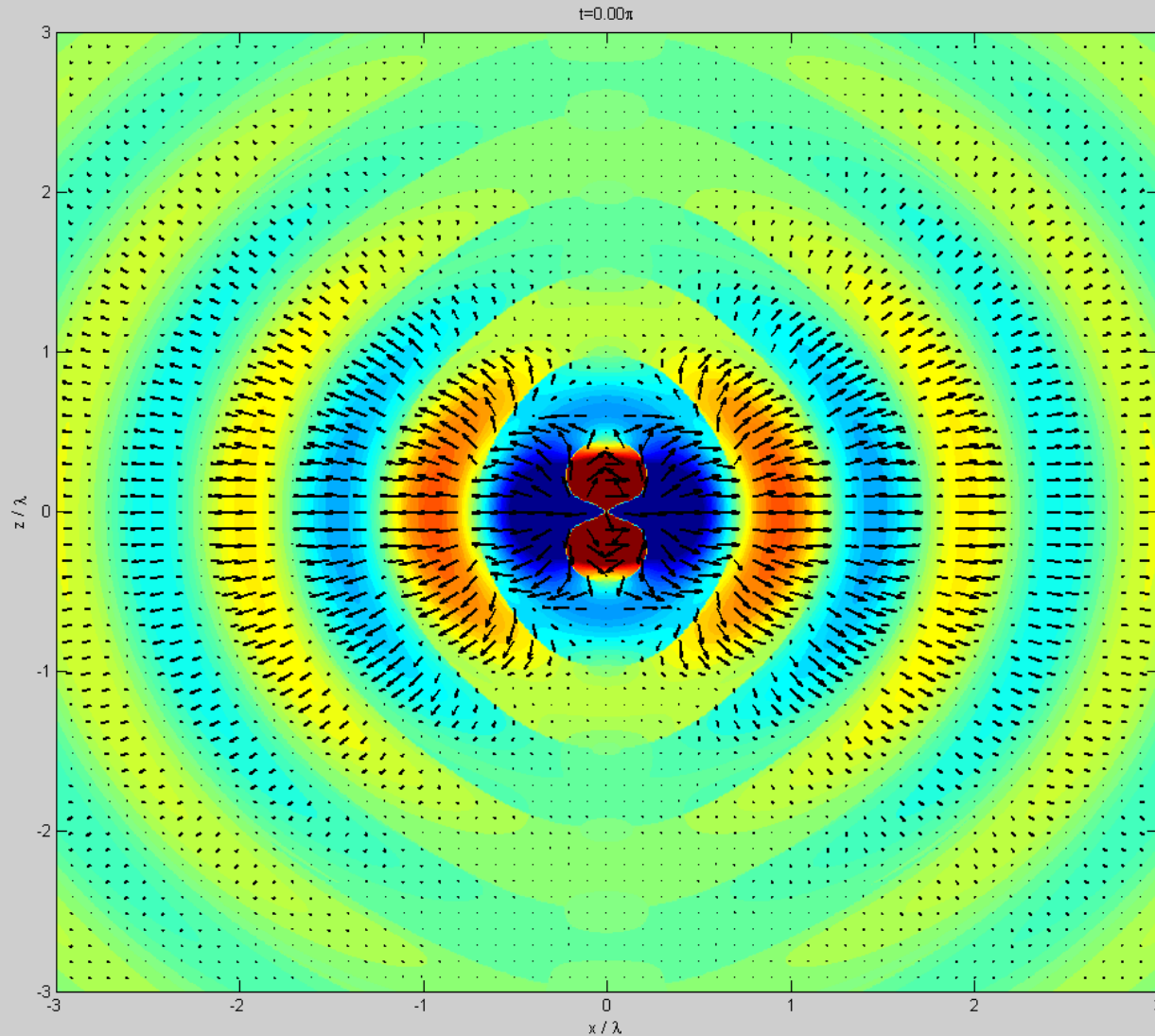
It does not emit in the oscillating direction!   *From wiki*

The two-level model for light-matter interaction
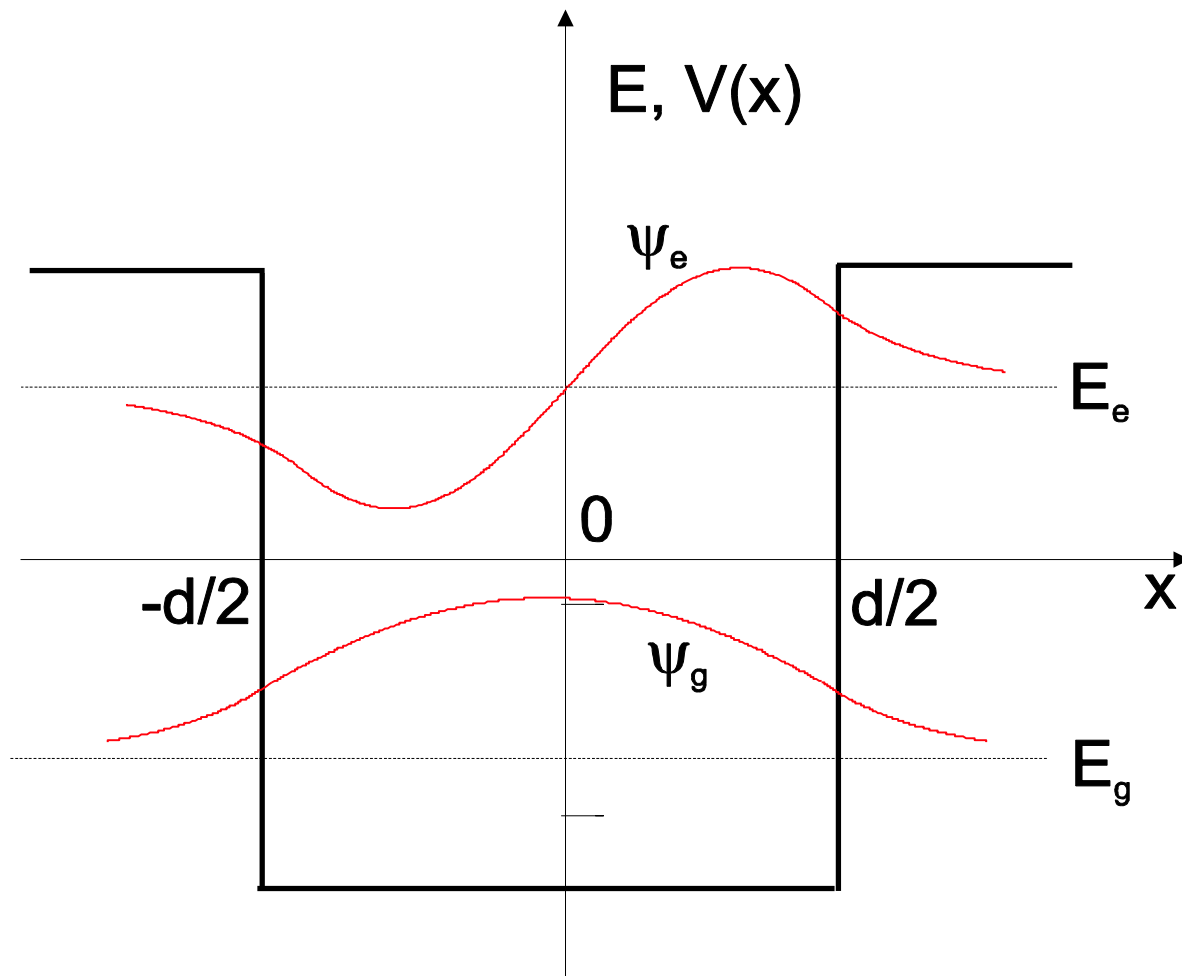
1D-model for a two-level atom.

The two-level model for light-matter interaction

Hamiltonian of the atom: H_A

$$\mathbf{H}_A \, \psi_e(\vec{r}) \;=\; E_e \, \psi_e(\vec{r})$$

$$\mathbf{H}_A \, \psi_g(\vec{r}) \;=\; E_g \, \psi_g(\vec{r})$$

General state of this two dimensional quantum mechanical system is:

$$\Psi(\vec{r}, t) = c_g(t) \, \psi_g(\vec{r}) + c_e(t) \, \psi_e(\vec{r})$$

$|c_g|^2$: propability to find the atom in the ground state

$|c_e|^2$: propability to find the atom in the excited state

The time dependence of these coefficients follows from the Schrödinger Equation:

$$j\,\hbar\frac{\partial}{\partial t}\Psi\left(\vec{r}, t\right) \;=\; \mathbf{H}_A \, \Psi\left(\vec{r}, t\right)$$

$$j\,\hbar\left(\dot{c}_g(t)\,\psi_g(\vec{r}) + \dot{c}_e(t)\,\psi_e(\vec{r})\right) = \left(E_g\,c_g(t)\,\psi_g(\vec{r}) + E_e\,c_e(t)\,\psi_e(\vec{r})\right)$$

The two-level model for light-matter interaction

By multiplication of this equation from the left with the complex conjugate ground state or the excited state and integration over space using the orthogonality relations for the energy eigenstates, we obtain two separate equations for the time dependence of the coefficients:

$$\dot{c}_e = -j\omega_e c_e, \text{ with } \omega_e = E_e / \hbar,$$
$$\dot{c}_g = -j\omega_g c_g, \text{ with } \omega_g = E_g / \hbar$$

The solution is the general time dependent solution of the two level system

$$\Psi(\vec{r}, t) = c_g(0) e^{-j\omega_g t} \psi_g(\vec{r}) + c_e(0) e^{-j\omega_e t} \psi_e(\vec{r})$$

How does the atomic dynamics change in the presence of an external electro-magnetic field and environmental perturbations?

Light-matter interaction in dipole approximation

Dipole moment: $\vec{d} = -e_0 \vec{r}.$

Dipole energy in E-field: $H_d = -\vec{d} \cdot \vec{E}(\vec{r}, t)$

Position of electron

Dipole approximation:

The spatial extension of the electron cloud in an atom is a few Angstrom, while the light wavelength typically a few hundred nanometers.

$$H_d = -\vec{d} \cdot \vec{E}(\vec{r}, t) = -\vec{d} \cdot \vec{E}(\vec{r}_A, t) = \boxed{-\vec{d} \cdot \vec{E}(t)} \longrightarrow$$

Spatially uniform vector potential

Position of electron   Position of atom

Hamiltonian for Atom in E-field: $\mathbf{H}_{AF} = \mathbf{H}_A - \vec{d} \cdot \vec{E}(t)$

The time-dependent solution of the two level system under a EM field can be written as

$$\Psi(\vec{r}, t) = c_g(t)\, \psi_g(\vec{r}) + c_e(t)\, \psi_e(\vec{r})$$

Expected value of the dipole moment

$$\left\langle \vec{d} \right\rangle = \int_{-\infty}^{\infty} \Psi^*(\vec{r},t)\, \vec{d}\, \Psi(\vec{r},t)\, d\vec{r} \qquad \Psi(\vec{r},t) = c_g(t)\, \psi_g(\vec{r}) + c_e(t)\, \psi_e(\vec{r})$$

$$\left\langle \vec{d} \right\rangle = |c_e|^2 \vec{M}_{ee} + |c_g|^2 \vec{M}_{gg} + c_e^* c_g \vec{M}_{eg} + c_g^* c_e \vec{M}_{ge}$$

Matrix elements of the dipole moment:

$$\vec{M}_{ee} = \int \psi_e^*(\vec{r})\, \vec{d}\, \psi_e(\vec{r})\, d\vec{r} = -e_0 \int \psi_e^*(\vec{r})\, \vec{r}\, \psi_e(\vec{r}) = 0 \quad \textbf{By symmetry}$$

$$\vec{M}_{eg} = \int \psi_e^*(\vec{r})\, \vec{d}\, \psi_g(\vec{r})\, d\vec{r} = -e_0 \int \psi_e^*(\vec{r})\, \vec{r}\, \psi_g(\vec{r})\, d\vec{r}$$

$$\vec{M}_{ge} = \int \psi_g^*(\vec{r})\, \vec{d}\, \psi_e(\vec{r})\, d\vec{r} = \vec{M}_{eg}^*$$

$$\vec{M}_{gg} = \int \psi_g^*(\vec{r})\, \vec{d}\, \psi_g(\vec{r})\, d\vec{r} = -e_0 \int \psi_g^*(\vec{r})\, \vec{r}\, \psi_g(\vec{r}) = 0 \quad \textbf{By symmetry}$$

$$\left\langle \vec{d} \right\rangle = c_e^* c_g \vec{M}_{eg} + c.c.$$

Equations of motion for the probability amplitudes

Hamiltonian for Atom in E-field: $\mathbf{H}_{AF} = \mathbf{H}_A - \vec{d} \cdot \vec{E}(t)$

The time-dependent solution of the two level system under a EM field can be written as

$$\Psi(\vec{r}, t) = c_g(t)\, \psi_g(\vec{r}) + c_e(t)\, \psi_e(\vec{r}) \qquad \mathrm{j}\hbar\frac{\partial}{\partial t}\Psi(\vec{r}, t) = \mathbf{H}_{AF}\Psi(\vec{r}, t)$$

New equations of motion for the probability amplitudes:

$$\dot{c}_e = -\mathrm{j}\omega_e c_e + \mathrm{j}c_g\frac{1}{\hbar}\left(\int \psi_e^*(\vec{r})\, \vec{d}\, \psi_g(\vec{r})\, d\vec{r}\right) \cdot \vec{E}(t),$$

$$\dot{c}_g = -\mathrm{j}\omega_g c_g + \mathrm{j}c_e\frac{1}{\hbar}\left(\int \psi_g^*(\vec{r})\, \vec{d}\, \psi_e(\vec{r})\, d\vec{r}\right) \cdot \vec{E}(t).$$

E-field as amplitude and polarization: $\vec{E}(t) = E(t)\, \vec{e},$

$$\dot{c}_e = -\mathrm{j}\omega_e c_e + \mathrm{j}c_g\frac{\vec{M}_{eg} \cdot \vec{e}}{\hbar}E(t), \quad \dot{c}_g = -\mathrm{j}\omega_g c_g + \mathrm{j}c_e\frac{\vec{M}_{eg}^* \cdot \vec{e}}{\hbar}E(t).$$

Monochromatic field:
$$E(t) = \frac{1}{2}\left(\underline{E}_0 e^{j\omega t} + \underline{E}_0^* e^{-j\omega t}\right)$$

Expect strong interaction between atom and E-field if: $\omega_{eg} = \omega_e - \omega_g \sim \boldsymbol{\omega}$

Introduce new amplitudes:
$$C_e = c_e\, e^{j\left(\frac{\omega_e + \omega_g + \omega}{2}t\right)} \qquad C_g = c_g\, e^{j\left(\frac{\omega_e + \omega_g - \omega}{2}t\right)}$$

Leads to:
$$\dot{C}_e = \left[j\left(\frac{\omega_e + \omega_g + \omega}{2}\right) - j\omega_e\right] c_e e^{j\left(\frac{\omega_e + \omega_g + \omega}{2}t\right)} + j c_g \frac{\vec{M}_{eg} \cdot \vec{e}}{\hbar}\vec{E}(t)\, e^{j\left(\frac{\omega_e + \omega_g + \omega}{2}t\right)}$$

$$\dot{C}_g = \left[j\left(\frac{\omega_e + \omega_g - \omega}{2}\right) - j\omega_g\right] c_g e^{j\left(\frac{\omega_e + \omega_g - \omega}{2}t\right)} + j c_e \frac{\vec{M}_{eg}^* \cdot \vec{e}}{\hbar}\vec{E}(t)\, e^{j\left(\frac{\omega_e + \omega_g - \omega}{2}t\right)}$$

Frequency detuning between atomic transition and electric field frequency:
$$\Delta = \frac{\omega_{eg} - \omega}{2}$$

Rabi Frequency:
$$\Omega_r = \frac{\vec{M}_{eg}^* \cdot \vec{e}}{\hbar}\left(\underline{E}_0 + \underline{E}_0^* e^{-j2\omega t}\right)$$

Coupled mode equations:
$$\frac{d}{dt}C_e = -j\Delta C_e + j\frac{\Omega_r^*}{2}C_g$$
$$\frac{d}{dt}C_g = +j\Delta C_g + j\frac{\Omega_r}{2}C_e$$

If Rabi frequency is small:
$$|\Omega_r| << \omega_{eg} \approx \omega$$

Rotating wave approximation (RWA) applies:
$$\Omega_r \approx \frac{\vec{M}_{eg}^* \cdot \vec{e}}{\hbar}\underline{E}_0 = const.$$

22

Rabi Oscillation

Zero detuning:
$$\Delta = \frac{\omega_{eg} - \omega}{2} = 0$$

$$\frac{d}{dt}C_e = -\mathrm{j}\Delta C_e + \mathrm{j}\frac{\Omega_r^*}{2}C_g$$

$$\frac{d}{dt}C_g = +\mathrm{j}\Delta C_g + \mathrm{j}\frac{\Omega_r}{2}C_e$$

$$\frac{d}{dt}C_e = \mathrm{j}\frac{\Omega_r^*}{2}C_g$$

$$\frac{d}{dt}C_g = \mathrm{j}\frac{\Omega_r}{2}C_e$$

$$\frac{d^2}{dt^2}C_e = -\frac{|\Omega_r|^2}{4}C_e$$

$$\frac{d^2}{dt^2}C_g = -\frac{|\Omega_r|^2}{4}C_g$$

If the atom is at time t = 0 in the ground-state

$$C_g(0) = 1 \text{ and } C_e(0) = 0$$

Oscillation solution:

$$C_g(t) = \cos\left(\frac{|\Omega_r|}{2}t\right)$$

$$C_e(t) = -\mathrm{j}\sin\left(\frac{|\Omega_r|}{2}t\right)$$

Probabilities for finding the atom in the ground or excited state are:

$$|c_g(t)|^2 = \cos^2\left(\frac{|\Omega_r|}{2}t\right)$$

$$|c_e(t)|^2 = \sin^2\left(\frac{|\Omega_r|}{2}t\right)$$

Rabi Oscillation

Expectation value of dipole moment:

$$\left\langle \vec{d} \right\rangle = \vec{M}_{eg} c_e c_g^* + c.c. = -\vec{M}_{eg} \sin\left(|\Omega_r| t\right) \sin\left(\omega_{eg} t\right)$$

The coherent external field drives the population of the atomic system between the two available states with a period: $T_r = 2\pi/\Omega_r$

Dipole moment oscillates with frequencies $\omega_{\pm} = \omega_{eg} \pm \Omega_r$

Atoms do not radiate at the same frequency as the incoming light. The emitted light rather shows sidebands offset by the Rabi-frequency called Mollow-sidebands. This is a nonlinear process.

<u>Where is our first order linear susceptibility χ(ω)?</u>

If this coherence is destroyed fast enough, Rabi-oscillation cannot happen and it is then impossible to generate inversion in a two-level system by interaction with light.
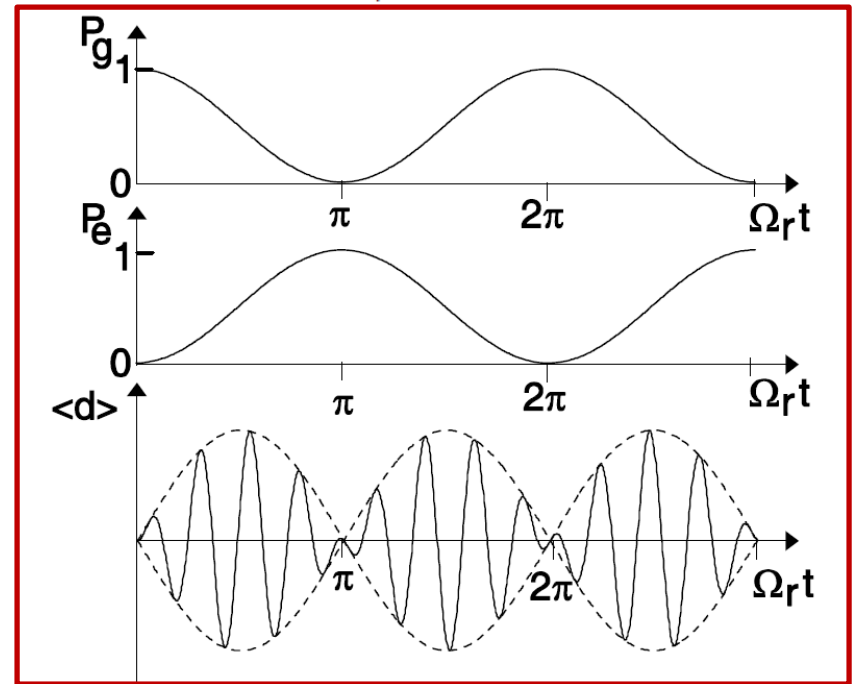
Motion eqns for dipole moment and population inversion

- Additional interactions with the environment cause the loss of coherence in the atomic system.

- These energy non-preserving processes cannot be easily included in the Schrödinger Equation.

- We can treat these processes phenomenologically in the equations of motion for the expectation values of the dipole moment and the population inversion.

Population inversion is defined as $\quad w = P_e - P_g = |c_e|^2 - |c_g|^2$

Complex slowly varying dipole moment is defined as

$$\langle \vec{d} \rangle = c_e^* c_g \vec{M}_{eg} + c.c. \quad \longrightarrow \quad \underline{d} = c_e^* c_g e^{-j\omega t} = C_e^* C_g$$

$$C_e = c_e \, e^{j\left(\frac{\omega_e + \omega_g + \omega}{2} t\right)}$$

$$C_g = c_g \, e^{j\left(\frac{\omega_e + \omega_g - \omega}{2} t\right)}$$

$$C_g(t) = \cos\left(\frac{|\Omega_r|}{2} t\right)$$

$$C_e(t) = -j \sin\left(\frac{|\Omega_r|}{2} t\right)$$

$$|\Omega_r| << \omega_{eg} \approx \omega$$

$\underline{d}$ is complex and slowly varying compared with the external EM wave's oscillation.

$$\langle \vec{d} \rangle = \vec{M}_{eg} \underline{d} \, e^{j\omega t} + c.c.$$

Motion eqns for dipole moment and population inversion

$$\frac{d}{dt}C_e \;=\; -\mathrm{j}\Delta C_e + \mathrm{j}\frac{\Omega_r^*}{2}C_g \qquad \frac{d}{dt}C_g \;=\; +\mathrm{j}\Delta C_g + \mathrm{j}\frac{\Omega_r}{2}C_e$$

Applying the product rule we find

$$\frac{d}{dt}\underline{d} \;=\; \left(\frac{d}{dt}C_e^*\right)C_g + C_e^*\left(\frac{d}{dt}C_g\right)$$

$$=\; \mathrm{j}\Delta C_e^* C_g - \mathrm{j}\frac{\Omega_r}{2}C_g^* C_g + \mathrm{j}\Delta C_e^* C_g + \mathrm{j}\frac{\Omega_r}{2}C_e^* C_e$$

$$=\; \mathrm{j}2\Delta\underline{d} + \mathrm{j}\frac{\Omega_r}{2}\cdot w$$

And for inversion:

$$\frac{d}{dt}w \;=\; \left(\frac{d}{dt}C_e\right)C_e^{*}. - \left(\frac{d}{dt}C_g\right)C_g^* + c.c.$$

$$=\; \left(-\mathrm{j}\Delta C_e C_e^* + \mathrm{j}\frac{\Omega_r^*}{2}C_g C_e^* - \mathrm{j}\Delta C_g C_g^* - \mathrm{j}\frac{\Omega_r}{2}C_e C_g^*\right) + c.c.$$

$$=\; +\mathrm{j}\Omega_r^*\underline{d} + c.c$$

Decay of population inversion

$$\frac{d}{dt}w = +\mathrm{j}\Omega_r^* \underline{d} + c.c \quad \longrightarrow \quad \dot{w} = \mathrm{j}\Omega_r^* \, \underline{d} - \mathrm{j}\Omega_r \, \underline{d}^*$$

Three incoherent processes reduce or increase the upper-level population:

- Spontaneous emission
- Interaction with the host material (collisions, lattice vibrations)
- Increase of the population by pumping

The three processes are described by the following equation:

$$\dot{w} = -\frac{w - \boxed{w_0}}{\boxed{T_1}}$$

$w_0 \longrightarrow$ Steady-state population

$T_1 \longrightarrow$ Energy relaxation time

Steady-state population:
      negative at thermal equilibrium without pumping
      positive with pumping

Include both external EM field and energy decay:

$$\dot{w} \quad = \quad -\frac{w - w_0}{T_1} + \mathrm{j}\Omega_r^* \, \underline{d} - \mathrm{j}\Omega_r \, \underline{d}^*$$

27

Decay of polarization (dipole moment)

$$\Delta = \frac{\omega_{eg} - \omega}{2}$$

$$\frac{d}{dt}\underline{d} = \mathrm{j}2\Delta\underline{d} + \mathrm{j}\frac{\Omega_r}{2} \cdot w \quad \longrightarrow \quad \underline{\dot{d}} = \mathrm{j}\left(\omega_{eg} - \omega\right)\underline{d} + \mathrm{j}\frac{\Omega_r}{2}\, w$$

An external EM field induces dipoles, which generate the macroscopic polarization. If the field is switched off, the polarization will disappear.

- Energy decay of the two-level system

- Collisions with the host material disoriented the direction of dipoles, causing dephasing. The resulting polarization becomes zero, although the single dipole still exists.

- Dephasing can happen much faster than energy relaxation and is characterized by a time constant T_2.

Include both external EM field and polarization decay:

$$\underline{\dot{d}} = -\left(\frac{1}{T_2} - \mathrm{j}\left(\omega_{eg} - \omega\right)\right)\underline{d} + \mathrm{j}\frac{\Omega_r}{2}\, w$$

dephasing time

28

Bloch equations

$$\dot{w} = -\frac{w - w_0}{T_1} + j\Omega_r^* \, \underline{d} - j\Omega_r \, \underline{d}^*$$

$$\dot{\underline{d}} = -(\frac{1}{T_2} - j\,(\omega_{eg} - \omega))\underline{d} + j\frac{\Omega_r}{2} \, w$$

$$\Omega_r = \frac{\vec{M}_{eg}^* \cdot \vec{e}}{\hbar} \left(\underline{E}_0 + \underline{E}_0^* e^{-j2\omega t} \right)$$

- **Bloch equations describe the dynamics of a statistical ensemble of two-level atoms interacting with a classical electric field.**

- **Polarization of the medium is related to the expectation value of the dipole moment of the atomic ensemble feeds into Maxwell equations, resulting in the Maxwell-Bloch Equations.**

Steady-state solution

For moderate field strength E_0, the magnitude of the Rabi-frequency is much smaller than the optical frequency, $|\Omega_r| \ll \omega$, the inversion and dipole moment do not change much within an optical cycle of the field.
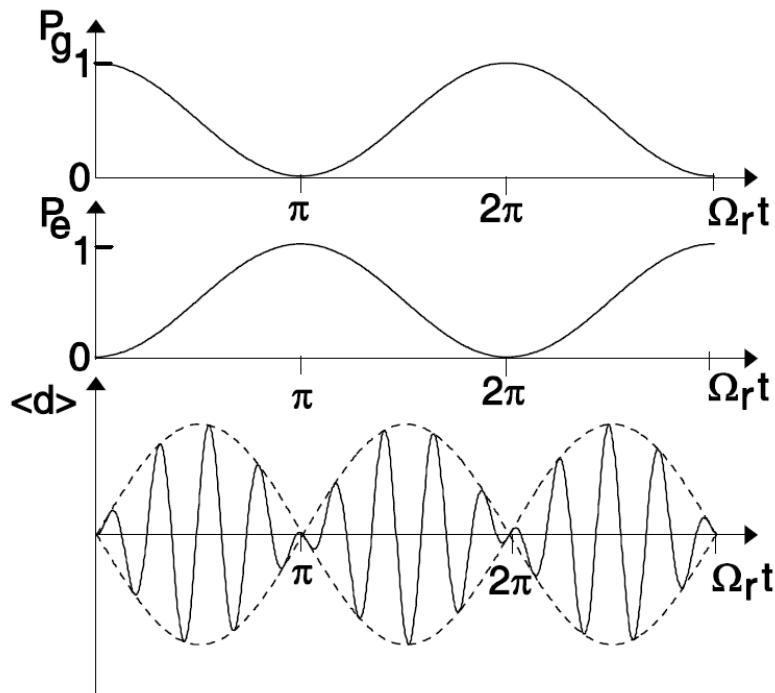
If the optical pulse duration is longer than energy relaxation time constant T_1, implying that the temporal variation of the EM field is slow than the energy decay, we can assume that population inversion and dipole moment are always at the steady-state though the steady state value adjust following the amplitude variation of the EM field.

$$\dot{\underline{d}} = 0 \qquad \dot{w} = 0$$

$$\underline{d}_s = \frac{\mathrm{j}}{2\hbar} \frac{\left(\vec{M}_{eg}^* \cdot \vec{e}\right) w_s}{1/T_2 + \mathrm{j}(\omega - \omega_{eg})} \underline{E}_0$$

$$w_s = \frac{w_0}{1 + \frac{T_1}{\hbar^2} \frac{1/T_2 \, |\vec{M}_{eg}^* \cdot \vec{e}|^2}{(1/T_2)^2 + (\omega_{eg} - \omega)^2} |\underline{E}_0|^2}$$

Inversion saturation

We introduce the normalized lineshape function, which is in this case a Lorentzian:

$$L(\omega) = \frac{(1/T_2)^2}{(1/T_2)^2 + (\omega_{eg} - \omega)^2}$$

Intensity: $\quad I = \frac{1}{2Z_F}|\underline{E}_0|^2$

Unsaturated inversion

Steady state inversion: $\quad w_s = \dfrac{w_0}{1 + \frac{I}{I_s}L(\omega)}$

Stationary inversion depends on the intensity of the incident light

Saturated inversion

Saturation intensity:

$$I_s = \left[\frac{2T_1 T_2 Z_F}{\hbar^2}|\vec{M}_{eg}^* \cdot \vec{e}|^2\right]^{-1}$$

Dielectric Susceptibility

Expectation value of the dipole moment

$$\left\langle \vec{d} \right\rangle = \vec{M}_{eg}\underline{d}\ e^{j\omega t} + c.c.$$

Multiplication with the number of atoms per unit volume, N, relates the dipole moment of the atom to the macroscopic polarization P

$$\vec{P}(t) = \frac{1}{2}\left(\underline{\vec{P}}_0 e^{j\omega t} + \underline{\vec{P}}_0^* e^{-j\omega t}\right) = N\vec{M}_{eg}\underline{d}_s\ e^{j\omega t} + c.c.$$

$$\underline{\vec{P}}_0 = 2N\vec{M}_{eg}\underline{d}_s$$

Definition of the complex susceptibility

$$\underline{\vec{P}}_0 = \epsilon_0 \chi(\omega)\vec{e}\underline{E}_0$$

Linear susceptibility of the medium

$$\chi(\omega) = \vec{M}_{eg}\vec{M}_{eg}^*\frac{jN}{\hbar\epsilon_0}\frac{w_s}{1/T_2 + j(\omega - \omega_{eg})}$$

Linear susceptibility of the medium is a 2nd-rank tensor

$$\chi(\omega) = \vec{M}_{eg}\vec{M}_{eg}^{*}\frac{jN}{\hbar\epsilon_0}\frac{w_s}{1/T_2 + j(\omega - \omega_{eg})}$$

Assume that the direction of the atom is random, i.e. the alignment of the atomic dipole moment, and the electric field is random. We have to average over the angle enclosed between the electric field of the wave and the atomic dipole moment, which results in

$$\overline{\begin{pmatrix} M_{egx}M_{egx}^* & M_{egx}M_{egy}^* & M_{egx}M_{egz}^* \\ M_{egy}M_{egx}^* & M_{egy}M_{egy}^* & M_{egy}M_{egz}^* \\ M_{egz}M_{egx}^* & M_{egz}M_{egy}^* & M_{egz}M_{egz}^* \end{pmatrix}} = \begin{pmatrix} \overline{M_{egx}^2} & 0 & 0 \\ 0 & \overline{M_{egy}^2} & 0 \\ 0 & 0 & \overline{M_{egz}^2} \end{pmatrix} = \frac{1}{3}|\vec{M}_{eg}|^2\,\mathbf{1}$$

For homogeneous and isotropic media the susceptibility tensor shrinks to a scalar

$$\chi(\omega) = \frac{1}{3}|\vec{M}_{eg}|^2\frac{jN}{\hbar\epsilon_0}\frac{w_s}{1/T_2 + j(\omega - \omega_{eg})}$$

Linear susceptibility

If the incident EM field is weak $\quad \dfrac{I}{I_s} L(\omega) \ll 1 \longrightarrow w_s \approx w_0$

Linear susceptibility derived using semi-classical model

$$\chi(\omega) = \frac{1}{3}|\vec{M}_{eg}|^2 \frac{jN}{\hbar\epsilon_0} \frac{w_0}{1/T_2 + j(\omega - \omega_{eg})}$$

Linear susceptibility derived using classical harmonic oscillator model

$$\tilde{\chi}(\omega) = \frac{N\frac{e_0^2}{m}\frac{1}{\epsilon_0}}{(\Omega_0^2 - \omega^2) + 2j\omega\frac{\Omega_0}{Q}} \xrightarrow{\ \omega \approx \Omega_0\ } \tilde{\chi}(\omega) = \frac{-jN\frac{e_0^2}{m}\frac{1}{\epsilon_0}/(2\Omega_0)}{j(\omega - \Omega_0) + \frac{\Omega_0}{Q}}$$

As the EM field has a frequency close to the oscillator's intrinsic frequency and define $Q = T_2\omega_{eg}$ **, the shape of the susceptibility computed quantum mechanically agrees well with the classical susceptibility derived from the harmonic oscillator model.**
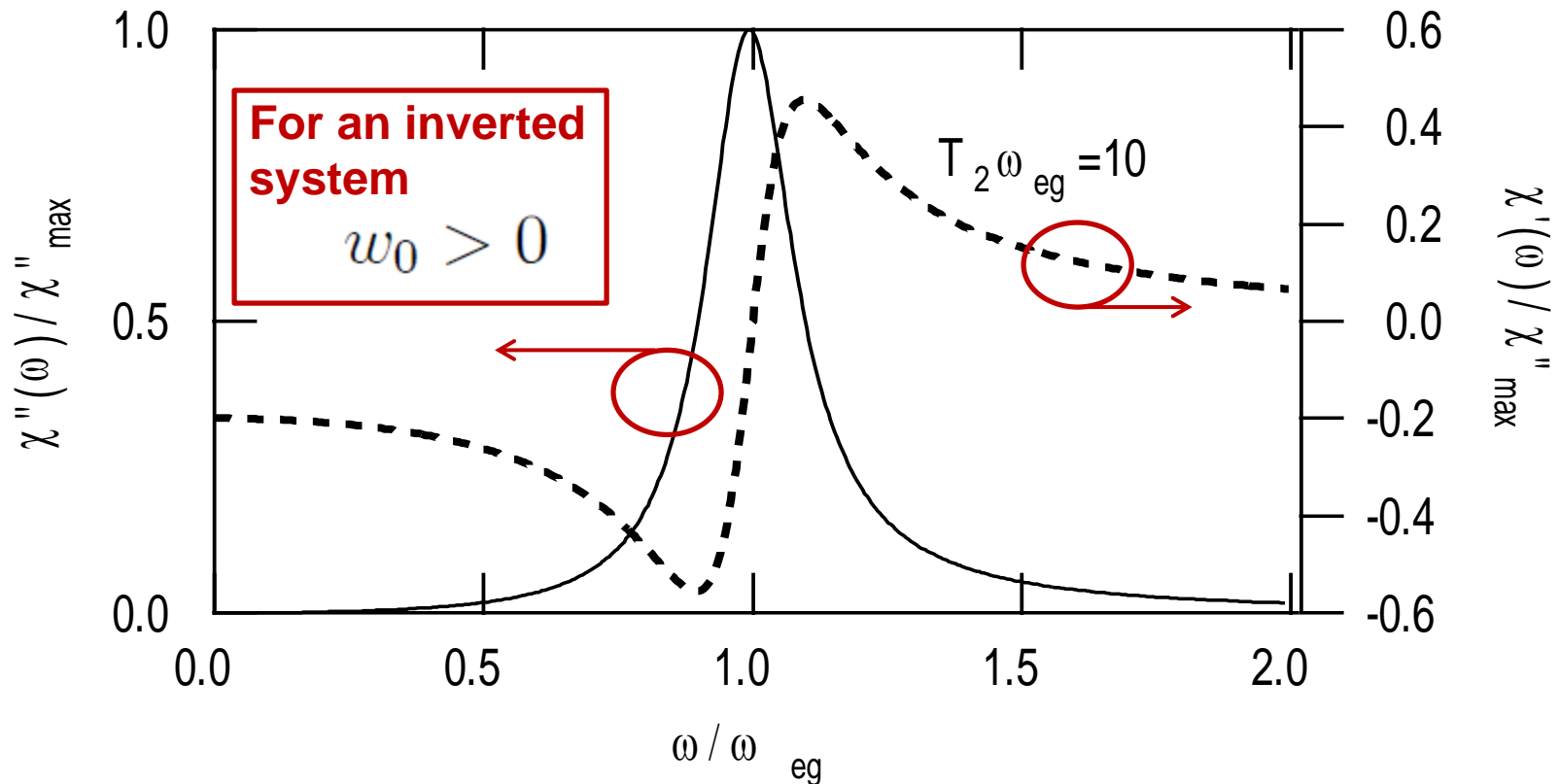
Linear susceptibility

Real and imaginary part of the susceptibility $\chi(\omega) = \chi'(\omega) + j\chi''(\omega)$

$$\chi'(\omega) = -\frac{|\vec{M}_{eg}|^2 N w_s T_2^2 (\omega_{eg} - \omega)}{3\hbar\epsilon_0} L(\omega)$$

$$\chi''(\omega) = \frac{|\vec{M}_{eg}|^2 N w_s T_2}{3\hbar\epsilon_0} L(\omega).$$

Positive imaginary susceptibility indicates exponential growth of an EM wave traveling in the medium.

For an inverted system $w_0 > 0$

$T_2 \omega_{eg} = 10$

35

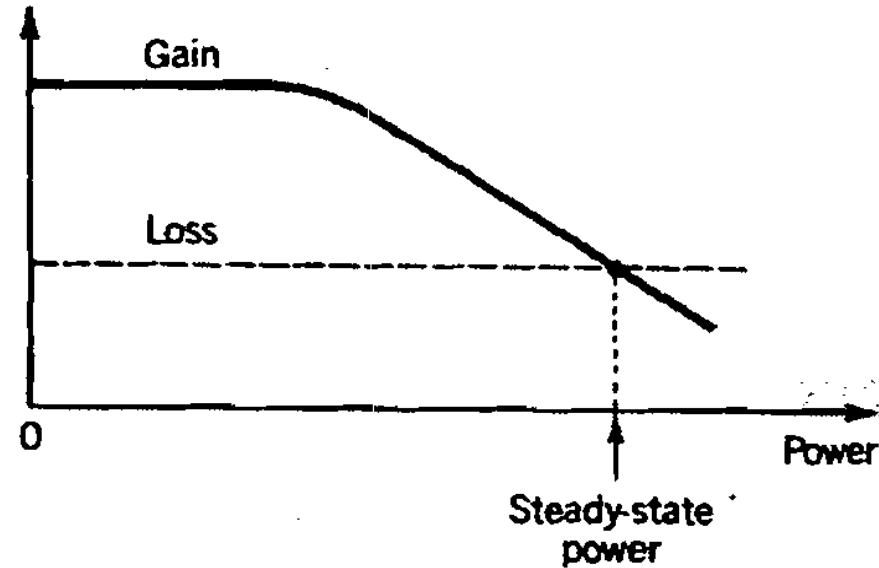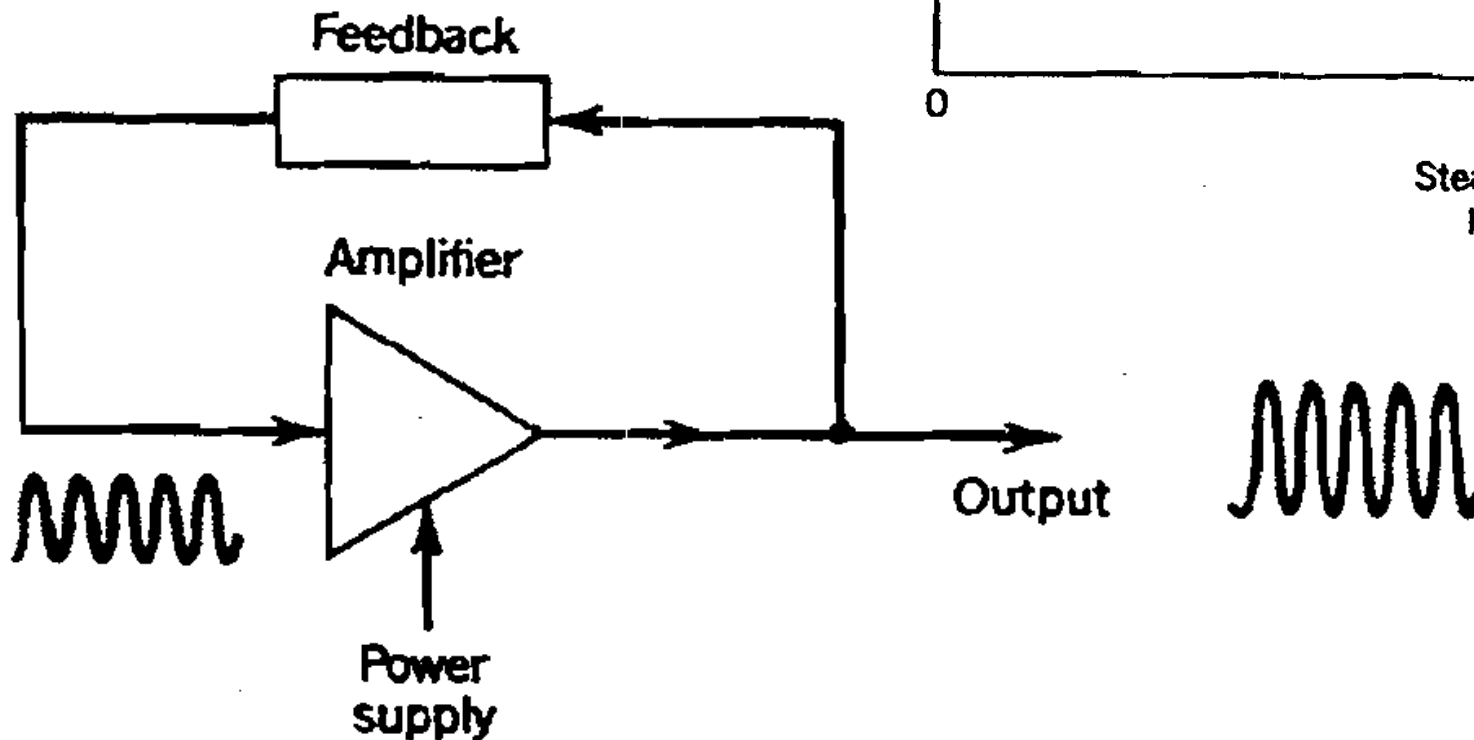# Linear susceptibility: semi-classical versus classical

The phase relaxation rate $1/T_2$ of the dipole moment determines the width of the absorption line or the bandwidth of the amplifier.

The amplification can not occur forever, because the amplifier saturates when the intensity reaches the saturation intensity. This is a strong deviation from the linear susceptibility derived from the classical oscillator model.

- **Light can not extract more energy from the atoms than the energy stored in them, i.e., energy conservation holds.**

- **Induced dipole moment in a two-level atom is limited by the maximum value of the matrix element.**

- **In contrast, the induced dipole moment in a classical oscillator grows proportionally to the applied field without limits.**
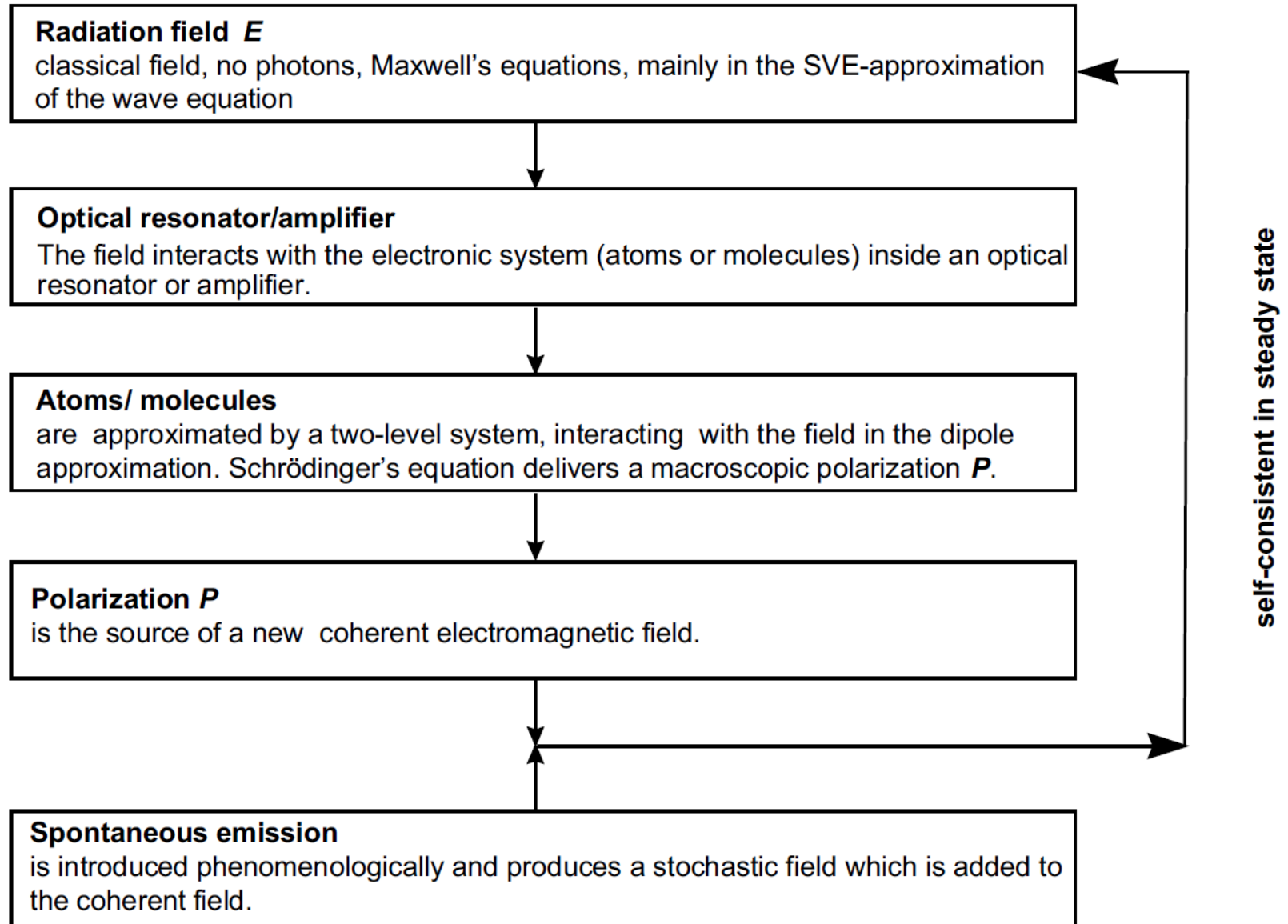
Gain saturation is critical in laser operation

Initially, unstable feedback loop. Oscillation builds up until amplifier saturates such that there is zero net roundtrip gain.

The Laser (Oscillator) Concept

Self-consistent in steady state

Radiation field *E*
classical field, no photons, Maxwell's equations, mainly in the SVE-approximation of the wave equation

↓

Optical resonator/amplifier
The field interacts with the electronic system (atoms or molecules) inside an optical resonator or amplifier.

↓

Atoms/ molecules
are approximated by a two-level system, interacting with the field in the dipole approximation. Schrödinger's equation delivers a macroscopic polarization *P*.

↓

Polarization *P*
is the source of a new coherent electromagnetic field.

↓

Spontaneous emission
is introduced phenomenologically and produces a stochastic field which is added to the coherent field.

self-consistent in steady state

V.A. Lopota and H. Weber, "fundamentals of the semiclassical laser theory"

Three regimes of solving Bloch equations

$$\dot{w} = -\frac{w - w_0}{T_1} + j\Omega_r^* \, \underline{d} - j\Omega_r \, \underline{d}^*$$

$$\dot{\underline{d}} = -\left(\frac{1}{T_2} - j\left(\omega_{eg} - \omega\right)\right)\underline{d} + j\frac{\Omega_r}{2} \, w$$

$$\Omega_r = \frac{\vec{M}_{eg}^* \cdot \vec{e}}{\hbar}\left(\underline{E}_0 + \underline{E}_0^* e^{-j2\omega t}\right)$$

Coherent equations:
Rabi oscillation

$$\dot{w} = j\Omega_r^* \, \underline{d} - j\Omega_r \, \underline{d}^*$$

$$\dot{\underline{d}} = j\left(\omega_{eg} - \omega\right)\underline{d} + j\frac{\Omega_r}{2} \, w$$

Steady state equations:
Optical pulse duration >> T_1, T_2

$$\dot{\underline{d}} = 0 \qquad \dot{w} = 0$$

Adiabatic equations:
$T_2 \ll T_1$, polarization is in equilibrium with the applied field. No transient oscillations of the electronic system.

$$\dot{\underline{d}} = 0 \qquad \dot{w} \neq 0$$

e.g. semiconductors: $T_2 \sim 50$ fs

Adiabatic equations: induced transitions

$$\dot{w} = -\frac{w - w_0}{T_1} + j\Omega_r^* \, \underline{d} - j\Omega_r \, \underline{d}^*$$

$$\underline{\dot{d}} = -\left(\frac{1}{T_2} - j\,(\omega_{eg} - \omega)\right)\underline{d} + j\frac{\Omega_r}{2}\, w$$

Adiabatic equations: $T_2 \ll T_1$

$$\underline{\dot{d}} = 0 \qquad \dot{w} \neq 0$$

$$\dot{w} = -\frac{w(t) - w_0}{T_1} - \frac{w(t)}{T_1 I_s}L(\omega)I(t)$$

Light intensity:

$$I(t) = |E_0(t)|^2 / (2Z_F)$$

energy relaxation (e.g., spontaneous emission)

Induced transitions (absorption, stimulated emission)

Resonant interaction between atom and EM field: $\omega = \omega_{eg}$ $L(\omega) = 1$

$$\dot{w}\big|_{induced} = -\frac{w}{T_1 I_s}I = -\sigma w I_{ph}$$

Interaction cross section

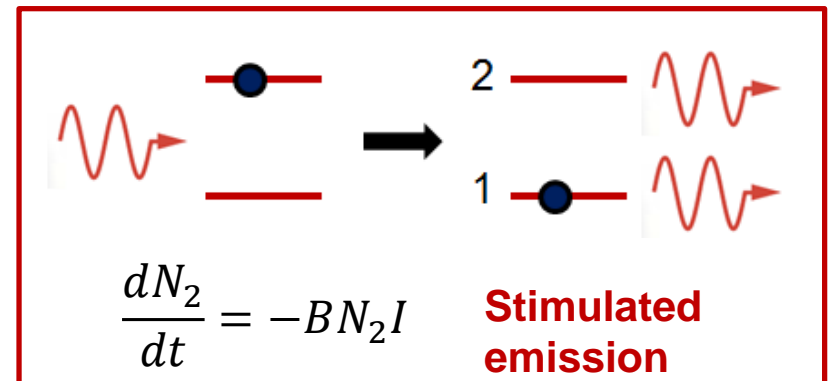**Photon flux density** $I_{ph} = I/\hbar\omega_{eg}$
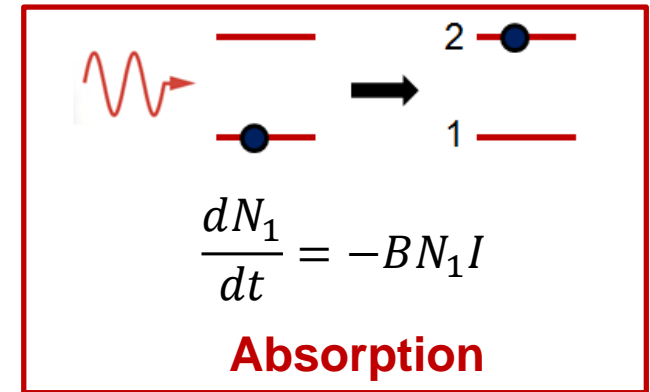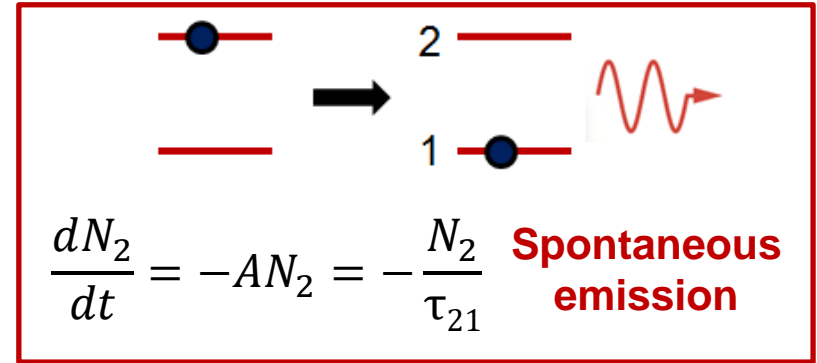
Laser rate equations

Interaction cross section: [Unit: cm^2]

$$\sigma = \frac{\hbar\omega_{eg}}{T_1 I_s} = \frac{2\omega_{eg}T_2 Z_F}{\hbar}|\vec{M}^*_{eg}\cdot\vec{e}|^2$$
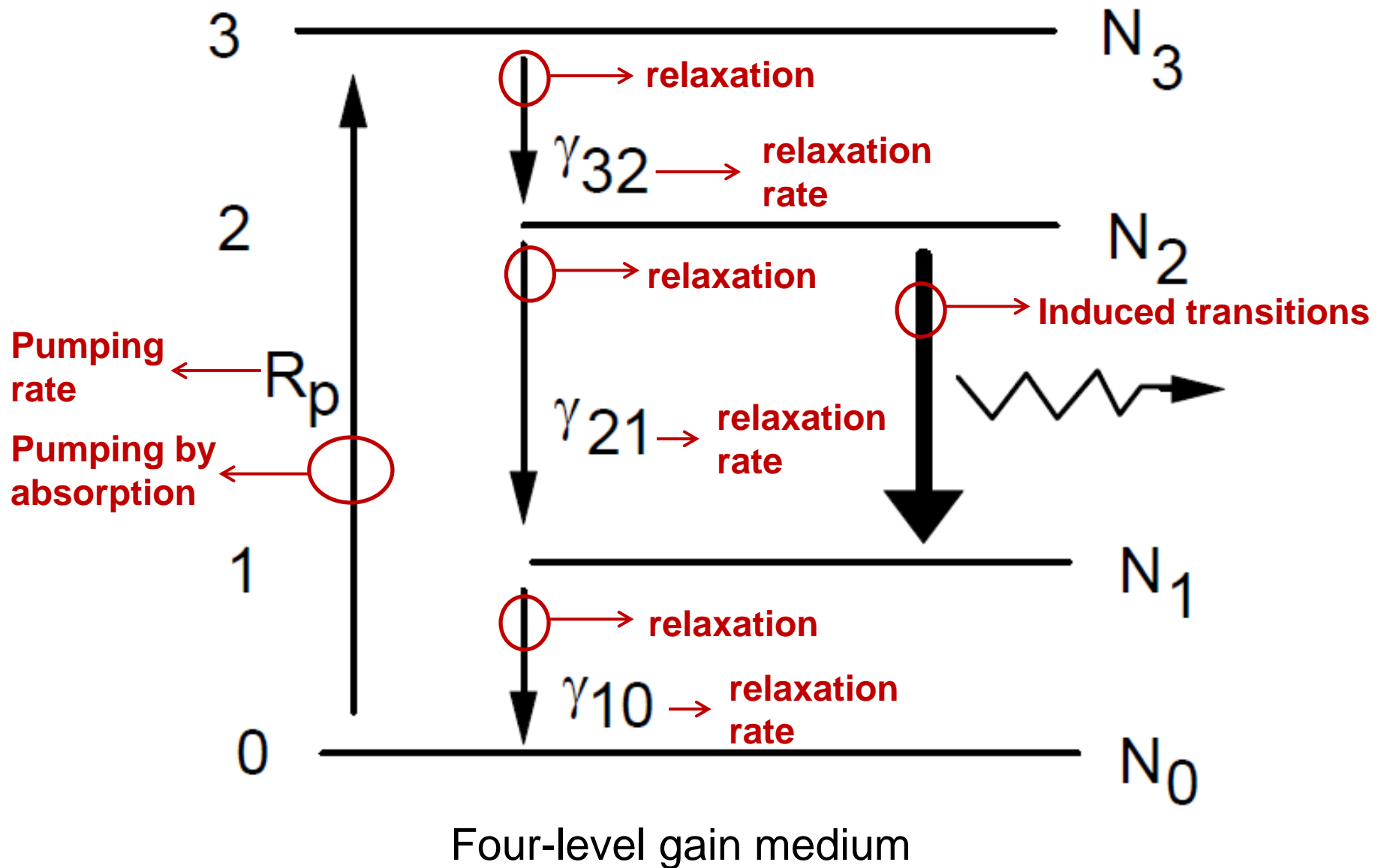
$$\dot{w}\big|_{induced} = -\frac{w}{T_1 I_s}I = -\sigma w I_{ph}$$

- Interaction cross section is the probability that an interaction will occur between EM field and the atomic system.

- Interaction cross section only depends on the dipole matrix element and the linewidth of the transition

$$\dot{w} = -\frac{w(t) - w_0}{T_1} - \sigma w I_{ph}$$

$$\frac{dN_2}{dt} = -AN_2 = -\frac{N_2}{\tau_{21}}$$ **Spontaneous emission**

$$\frac{dN_1}{dt} = -BN_1 I$$

Absorption

$$\frac{dN_2}{dt} = -BN_2 I$$ **Stimulated emission**

How to achieve population inversion?

Four-level gain medium

Laser rate equations for three-level laser medium
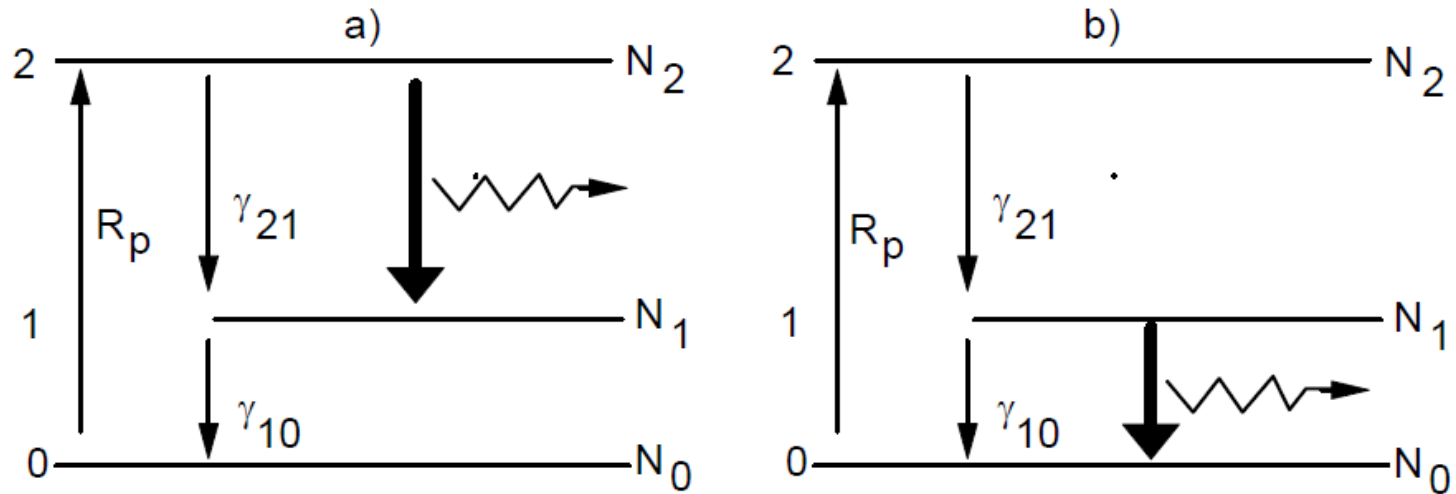
For (a):

$$\frac{d}{dt}N_2 = -\gamma_{21}N_2 - \sigma_{21}\left(N_2 - N_1\right)I_{ph} + R_p$$

$$\frac{d}{dt}N_1 = -\gamma_{10}N_1 + \gamma_{21}N_2 + \sigma_{21}\left(N_2 - N_1\right)I_{ph}$$

$$\frac{d}{dt}N_0 = \gamma_{10}N_1 - R_p$$

Many atoms are available in the ground state such that optical pumping can never deplete N_0. That is why we can assume a constant pump rate R_p.

σ_{21} **is the cross section for stimulated emission between the levels 2 and 1.** I_{ph} **is the photon flux.**

Laser rate equations for three-level laser medium

If the relaxation rate γ_{10} is much faster than γ_{21} and the number of possible stimulated emission events that can occur $\sigma_{21}\left(N_2 - N_1\right)I_{ph}$, we can set $N_1 = 0$ and obtain only a rate equation for the upper laser level:

a)
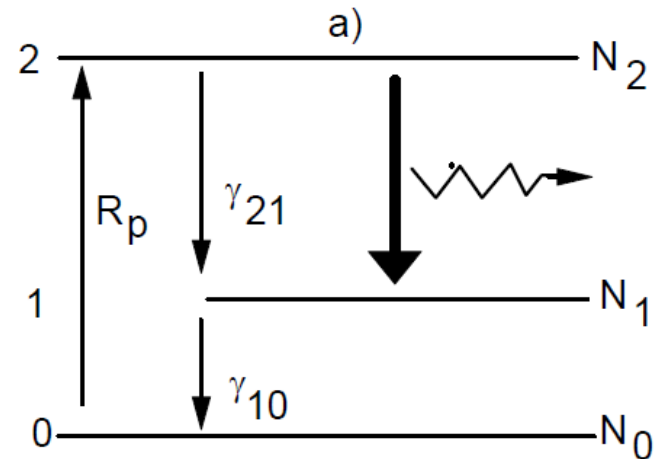
$$\frac{d}{dt}N_2 = -\gamma_{21}\left(N_2 - \frac{R_p}{\gamma_{21}}\right) - \sigma_{21}N_2 \cdot I_{ph}$$

This equation is identical to the equation for the inversion of the two-level system:

$$\dot{w} = -\frac{w(t) - w_0}{T_1} - \sigma w I_{ph}$$

$$\frac{R_p}{\gamma_{21}} \longrightarrow$$ **equilibrium upper state population w/o photons present**

$$\gamma_{21} = \frac{1}{\tau_L} \longrightarrow$$ **upper level lifetime due to radiative and non-radiative processes**

44

Spectroscopic parameters of selected laser materials

Laser Medium	Wave-length $\lambda_0 (nm)$	Cross Section σ (cm^2)	Upper-St. Lifetime τ_L (μs)	Linewidth Δf_{FWHM} $\frac{2}{T_2}$ (THz)	Typ	Refr. index n
Nd^{3+}:YAG	1,064	$4.1 \cdot 10^{-19}$	1,200	0.210	H	1.82
Nd^{3+}:LSB	1,062	$1.3 \cdot 10^{-19}$	87	1.2	H	1.47
Nd^{3+}:YLF	1,047	$1.8 \cdot 10^{-19}$	450	0.390	H	1.82
Nd^{3+}:YVO$_4$	1,064	$2.5 \cdot 10^{-19}$	50	0.300	H	2.19
Nd^{3+}:glass	1,054	$4 \cdot 10^{-20}$	350	3	H/I	1.5
Er^{3+}:glass	1,55	$6 \cdot 10^{-21}$	10,000	4	H/I	1.46
Ruby	694.3	$2 \cdot 10^{-20}$	1,000	0.06	H	1.76
Ti^{3+}:Al$_2$O$_3$	660-1180	$3 \cdot 10^{-19}$	3	100	H	1.76
Cr^{3+}:LiSAF	760-960	$4.8 \cdot 10^{-20}$	67	80	H	1.4
Cr^{3+}:LiCAF	710-840	$1.3 \cdot 10^{-20}$	170	65	H	1.4
Cr^{3+}:LiSGAF	740-930	$3.3 \cdot 10^{-20}$	88	80	H	1.4
He-Ne	632.8	$1 \cdot 10^{-13}$	0.7	0.0015	I	~ 1
Ar$^+$	515	$3 \cdot 10^{-12}$	0.07	0.0035	I	~ 1
CO$_2$	10,600	$3 \cdot 10^{-18}$	2,900,000	0.000060	H	~ 1
Rhodamin-6G	560-640	$3 \cdot 10^{-16}$	0.0033	5	H	1.33
semiconductors	450-30,000	$\sim 10^{-14}$	~ 0.002	25	H/I	3 - 4