Interaction of light with atoms

QM estimation of dipole radiation and lifetime Summary of time-dependent perturbation theory approach

Reading: Svelto 2.3-2.4

Interaction of light with a 2-level system

• Three allowed processes:



- Note photon energy matches transition energy
- All three process are related in the quantum picture

QM atomic transitions

We'll take an approach to understanding transitions from the quantum perspective

An isolated atom in a pure energy eigenstate is in a stationary state:

$$\boldsymbol{\psi}_n(\mathbf{r},t) = \boldsymbol{u}_n(\mathbf{r})e^{-E_nt/\hbar}$$

- There is time dependence to the phase, but the amplitude remains constant. So, no transitions.
- An applied EM field of the right frequency can induce a mixture of two states:

 $\psi_1(\mathbf{r},t) = u_1(\mathbf{r})e^{-E_1t/\hbar}$ $\psi_2(\mathbf{r},t) = u_2(\mathbf{r})e^{-E_2t/\hbar}$

– Superposition:

$$\psi(\mathbf{r},t) = a_1(t)\psi_1(\mathbf{r},t) + a_2(t)\psi_2(\mathbf{r},t)$$

- w/ normalization: $|a_1(t)|^2 + |a_2(t)|^2 = 1$

QM charge distribution

- The electron is not localized in QM.
- The charge density can be calculated from ψ : $\rho(\mathbf{r},t) = -e |\psi(\mathbf{r},t)|^2$
- For a stationary state:

$$\rho(\mathbf{r},t) = -e \left| \psi_n(\mathbf{r},t) \right|^2 = -e \left| u_n(\mathbf{r}) e^{-E_n t/\hbar} \right|^2 = -e \left| u_n(\mathbf{r}) \right|^2$$

– No time dependence, charge is not moving!

• For a superposition state: $\rho(\mathbf{r},t) = -e |\psi(\mathbf{r},t)|^{2} = -e |a_{1}\psi_{1} + a_{2}\psi_{2}|^{2}$ $= -e (|a_{1}\psi_{1}|^{2} + |a_{2}\psi_{2}|^{2} + a_{1}a_{2}^{*}\psi_{1}\psi_{2}^{*} + a_{1}^{*}a_{2}\psi_{1}^{*}\psi_{2})$ Cross terms will lead to time dependence in the char

Cross terms will lead to time dependence in the charge.

QM dipole moment calculation

- The nucleus is localized, but the electron charge is distributed.
- The effective position is calculated like the center of mass, so dipole moment is:

$$\mu(t) = -e \int \mathbf{r} |\psi(\mathbf{r}, t)|^2 dV \qquad \mathbf{p} = q \mathbf{r}$$

$$= -e \left(\int \mathbf{r} |a_1 \psi_1|^2 dV + \int \mathbf{r} |a_2 \psi_2|^2 dV + \int a_1^* a_2 \mathbf{r} \psi_1^* \psi_2 dV \right)$$

- Terms in red go to zero: parity.

Time dependent dipole moment

 The cross terms (which are like interference terms in optics), lead to time dependent oscillation:

 $\mu_{osc}(t) = -e \left(a_1 a_2^* \int \mathbf{r} \psi_1 \psi_2^* dV + a_1^* a_2 \int \mathbf{r} \psi_1^* \psi_2 dV \right)$ = $-e \left(a_1 a_2^* \int \mathbf{r} u_1(\mathbf{r}) u_2^*(\mathbf{r}) e^{+i(E_2 - E_1)t/\hbar} dV + a_1^* a_2 \int u_1^*(\mathbf{r}) u_2(\mathbf{r}) e^{-i(E_2 - E_1)t/\hbar} dV \right)$

- Oscillation frequency: $\omega_{21} = (E_2 - E_1) / \hbar$

 $\mu_{osc}(t) = -e\left(a_{1}a_{2}^{*}\mu_{21}e^{i\omega_{21}t} + a_{1}^{*}a_{2}\mu_{12}e^{-i\omega_{21}t}\right) = -e\operatorname{Re}\left[2a_{1}a_{2}^{*}\mu_{21}e^{i\omega_{21}t}\right]$

 $\mu_{21} = \int u_1(\mathbf{r})(-e\mathbf{r}) u_2^*(\mathbf{r}) dV \qquad \text{Dipole "matrix element"}$

- μ₂₁ is the part that depends on the atomic structure, independent of the populations.
- This is a vector, but the direction of r corresponds to the E-field direction, relative to the atom or molecule.

QM dipole radiated power

 Use classical Larmor expression to estimate the radiated power from this oscillating dipole.

$$\langle P_{rad} \rangle = \frac{1}{4\pi\varepsilon_0} \frac{2}{3} \frac{e^2 \langle \ddot{x}^2(t) \rangle}{c^3} = \frac{1}{4\pi\varepsilon_0} \frac{2}{3} \frac{1}{c^3} \int \ddot{\mu}^2(t) dt$$
 Note: $\mu = p$
Integrate over one period

$$\mu_{osc}(t) = -e \left(a_1 a_2^* \mu_{21} e^{i\omega_{21}t} + a_1^* a_2 \mu_{12} e^{-i\omega_{21}t} \right)$$

$$\ddot{\mu}_{osc}(t) = e \omega_{21}^2 \left(a_1 a_2^* \mu_{21} e^{i\omega_{21}t} + a_1^* a_2 \mu_{12} e^{-i\omega_{21}t} \right)$$

$$\ddot{\mu}_{osc}^2(t) = e^2 \omega_{21}^4 \left(\left(a_1 a_2^* \mu_{21} e^{i\omega_{21}t} \right)^2 + \left(a_1^* a_2 \mu_{12} e^{-i\omega_{21}t} \right)^2 + 2 |a_1|^2 |a_2|^2 |\mu_{21}|^2 \right)$$

$$\langle \ddot{\mu}_{osc}^2 \rangle = e^2 \omega_{21}^4 2 |a_1|^2 |a_2|^2 |\mu_{21}|^2$$

$$\text{Let } |\mu_{21}| \rightarrow \mu_{21}$$

$$P_{rad} = \frac{1}{4\pi\varepsilon_0} \frac{2}{3} \frac{1}{c^3} e^2 \omega_{21}^4 \mu_{21}^2 2 |a_1|^2 |a_2|^2$$

QM dipole radiation: rate of decay

Simplify the cycle-averaged radiated power

$$\left\langle P_{rad} \right\rangle = \frac{e^2 \omega_{21}^{4} \mu_{21}^{2}}{3\pi \varepsilon_0 c^3} |a_1|^2 |a_2|^2 = \hbar \omega_{21} \frac{e^2 \omega_{21}^{3} \mu_{21}^{2}}{3\hbar \pi \varepsilon_0 c^3} |a_1|^2 |a_2|^2 \frac{1}{2} \frac{1}{2} \left| \frac{1}{2} \right|^2}{8} \frac{1}{2} \left| \frac{1}{2} \right|^2 \frac{1}{2} \frac{1}{2} \left| \frac{1}{2} \right|^2}{8} \frac{1}{2} \frac{1}{2} \left| \frac{1}{2} \right|^2}{8} \frac{1}{2} \frac$$

- If we assume that the excitation probability of the upper level is small, then $|a_1|^2 = 1 - |a_2|^2 \approx 1$
- We can then deduce the change in upper level population:

This connects the spontaneous emission rate to a quantum calculation of the dipole moment.

Selection rules

• In Dirac notation, the dipole matrix element is:

$$\mu_{21} = \langle 2| - e\mathbf{r}|1 \rangle = \int u_1(\mathbf{r})(-e\mathbf{r}) u_2^*(\mathbf{r}) dV$$

- Working with the symmetries of wavefunctions leads to selection rules about which transitions can take place.
 - Parity: r is odd, so u_1 must be opposite parity of u_2
 - Angular momentum: $\Delta I = \pm 1$. Photon carries 1 unit of ang. mom.
- Exceptions:
 - Transition might take place under other moments:
 - Magnetic dipole, electric quadrupole, etc.
 - Leads to longer lifetimes.
 - States might not be "pure", mixture of eigenstates
 - External or internal perturbations

HeNe laser transitions



Full QM approach

- Next level up in accuracy in QM is to approximately solve the Schrodinger equation in the presence of the incident field
 - QM representation of the electron wavefunction $\psi(\mathbf{r},t)$
 - Classical representation of the EM field as a perturbation

$$\hat{H}\psi = i\hbar\frac{\partial\psi}{\partial t}$$
 $\hat{H} = \hat{H}_{0} + \hat{H}'$

- Without external field: With external field (E-dipole): $\hat{H}_0 \psi = i\hbar \frac{\partial \psi}{\partial t} \rightarrow \hat{H}_0 \psi_n = E_n \psi_n$ $\hat{H}' = \mu \cdot \mathbf{E} = -e \mathbf{r} \cdot \mathbf{E}_0 \sin \omega t$
- Assume wavefunction with field can be written in terms of a linear combination of wavefunctions without field

$$\psi(r,t) = \sum_{n} a_{n}(t)\psi_{n}(r,t) \qquad \qquad \psi_{n}(\mathbf{r},t) = u_{n}(\mathbf{r})e^{-E_{n}t/\hbar}$$

Time-dependent perturbation theory

- Easiest to concentrate on 2 levels
- Assume close to resonance:

$$\boldsymbol{\omega} \approx \left(E_2 - E_1 \right) / \hbar = \boldsymbol{\omega}_{21}$$

• Assume weak probability of excitation:

 $a_1(t) \approx 1, \quad a_2(t) \ll 1$

- Put form of solution into time-dependent SE (with field)
- Transition rate will be

$$W_{12} = \frac{d}{dt} \left| a_2(t) \right|^2$$

Result: "Fermi's Golden Rule"

$$W_{12}(v) = \frac{\pi^2}{3h^2} |\mu_{21}|^2 E_0^2 \delta(v - v_0)$$

$$\delta(v - v_0)$$
 Dirac delta function
 $\int f(v)\delta(v - v_0)dv = f(v_0)$

Fermi's golden rule

• Express field in terms of (total) energy density:

$$\rho = \frac{1}{2}n^{2}\varepsilon_{0}E_{0}^{2}$$
For other lineshape:

$$\rightarrow W_{12}(v) = \frac{2\pi^{2}}{3n^{2}\varepsilon_{0}h^{2}}|\mu_{21}|^{2}\rho\delta(v-v_{0}) = \frac{2\pi^{2}}{3n^{2}\varepsilon_{0}h^{2}}|\mu_{21}|^{2}\rho g(v-v_{0})$$

- When EM source varies in frequency, energy density btw v' and v'+dv' is $d\rho = \rho_{v'} dv'$
- So the contribution to the rate at v' is

$$dW_{12}(v') = \frac{2\pi^2}{3n^2\varepsilon_0 h^2} |\mu_{21}|^2 \rho_{v'} g(v - v_0) dv'$$

• Total rate is:

$$W_{12} = \int \frac{2\pi^2}{3n^2 \varepsilon_0 h^2} |\mu_{21}|^2 \rho_{v'} g(v - v_0) dv'$$

Working with spectral lineshapes

• For atomic system, replace Dirac delta with transition lineshape $\int a(y-y_{-}) dy = 1$

$$\int g(v-v_0)dv = 1$$

- Lorentzian lineshape (radiative, collisional broadening) $\delta(v - v_0) \rightarrow g_L(v - v_0) = \frac{2}{\pi \Delta v_0} \frac{1}{1 + \left(\frac{2(v - v_0)}{\Delta v_0}\right)^2}$ $\Delta v_0 \quad \text{FWHM}$
- Doppler broadened (Gaussian) lineshape

$$\delta(v - v_0) \to g_G^*(v - v_0) = \frac{2}{\Delta v_0^*} \sqrt{\frac{\ln 2}{\pi}} \exp\left\{-4\ln 2\frac{(v - v_0)^2}{\Delta v_0^{*2}}\right\}$$

Lorentzian vs Gaussian lineshapes

Lorentzian is much broader in spectral wings



Natural broadening

- Radiative broadening results directly from the spontaneous emission lifetime of the state
- Fourier transforms

- Forward: FT
$$F(\omega) = \int_{-\infty}^{\infty} f(t) e^{i\omega t} dt$$

- Inverse: FT⁻¹
$$f(t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} F(\omega) e^{-i\omega t} d\omega$$

Suppose exponential, oscillating decay in time domain

 $f(t) = \left. \begin{array}{l} e^{-\gamma t} e^{-i\omega_0 t} & \text{for } t \ge 0\\ 0 & \text{for } t < 0 \end{array} \right.$ $F(\omega) = \int_0^\infty e^{-\gamma t - i\omega_0 t} e^{i\omega t} dt = \left. \frac{e^{\left(-\gamma + i\left(\omega - \omega_0\right)\right)t}}{-\gamma + i\left(\omega - \omega_0\right)} \right|_0^\infty = \frac{1}{\gamma - i\left(\omega - \omega_0\right)} \right.$

Complex Lorentzian

Lorentzian lineshape

Complex Lorentzian separated into Re and Im

$$\frac{1}{\gamma - i(\omega - \omega_0)} = \frac{\gamma}{(\omega - \omega_0)^2 + \gamma^2} + i\frac{(\omega - \omega_0)}{(\omega - \omega_0)^2 + \gamma^2}$$
- Real part corresponds to absorption effects

Normalize

$$c\int \frac{\gamma}{(\omega-\omega_0)^2+\gamma^2} d\omega = c\gamma \frac{\pi}{\gamma} = 1 \quad \rightarrow g_L(\omega-\omega_0) = \frac{\gamma/\pi}{(\omega-\omega_0)^2+\gamma^2}$$

• Convert ω to v

$$c\int \frac{\gamma}{4\pi^{2}(v-v_{0})^{2}+\gamma^{2}} dv = c\gamma \frac{1}{2\gamma} = 1$$

$$\rightarrow g_{L}(v-v_{0}) = \frac{2}{\gamma} \left[1 + \left(\frac{2(v-v_{0})}{\gamma/\pi}\right)^{2} \right]^{-1} = \frac{2}{\pi \Delta v_{0}} \left[1 + \left(\frac{2(v-v_{0})}{\Delta v_{0}}\right)^{2} \right]^{-1}$$

Collisional broadening

- Elastic collisions don't cause transition, but interrupt the phase
- Timescales:
 - Period of EM cycle much less than radiative lifetime
 - Avg time btw collisions < lifetime
 - Duration of a collision << time btw coll, lifetime
- Calculation:
 - FT over time 0 to τ_1 to get lineshape for a specific oscillation length
 - Average over probability of a given time between collisions:

$$P(\tau_1)d\tau_1 = \frac{1}{\tau_c}e^{-\tau_1/\tau_c}d\tau_1$$

Result:

Lorentzian shape with new width

 $\frac{2\pi}{-}\ll \tau$

 $\tau_c < \tau$

 $\Delta \tau_c \ll \tau_c, \tau$

 $\boldsymbol{\omega}_{0}$

 $\Delta v = \gamma / 2\pi + 1 / \pi \tau_c$

Doppler broadening

• From relative velocity of atom to input beam, Doppler shift:

$$V_0' = \frac{V_0}{1 - v_z / c}$$
 Beam propagating in z direction

Each atom in distribution is shifted according to its velocity

Boltzmann distribution

$$P(\mathbf{v}_z) \sim \exp\left[-\frac{1}{2}Mv_z^2 / k_BT\right]$$

• Average over distribution to get effective lineshape:

$$g^{*}(v - v_{0}) = \frac{1}{v_{0}} \left(\frac{Mc^{2}}{2\pi k_{B}T}\right)^{1/2} \exp\left\{\frac{Mc^{2}}{2k_{B}T} \frac{(v - v_{0})^{2}}{v_{0}^{2}}\right\}$$

FWHM: $\Delta v_{0}^{*} = 2v_{0} \left[\frac{2k_{B}T\ln 2}{Mc^{2}}\right]^{1/2}$

Doppler broadening in HeNe lasers

$$\Delta v_0^* = 2v_0 \left[\frac{2k_B T \ln 2}{Mc^2} \right]^{1/2}$$

$$\lambda_0 = 632.8 \text{ nm}$$

$$v_0 = 4.74 \times 10^{14} \text{ s}^{-1}$$

$$M = 20.12 \text{ amu} = 3.34 \times 10^{-26} \text{kg}$$
 For Neon

$$k_B T = 1/40 eV = 4 \times 10^{-21} J$$

$$\Delta v_0^* = 1.55 GHz$$

Inhomogeneous vs homogeneous broadening

- Homogeneous broadening: every atom is broadened by same shape
 - Radiative, collisional, phonon
 - All atoms participate in absorption or gain
- Inhomogeneous broadening:
 - Doppler broadening
 - Absorption or gain only by atoms in resonance
 - Leads to "spectral hole burning"



Amplifiers: pumping and small-signal gain

- Absorption $I[z] = I_0 \exp[-N_0 \sigma_{12} z] = I_0 \exp[-\alpha z]$
- Gain $I[z] = I_0 \exp[N_{inv}\sigma_{21}z] = I_0 \exp[gz]$
 - What is the inversion density?
 - How to express it in terms of the pump distribution
 - How does gain depend on λ or ω ?
 - What happens when the inversion density is depleted?





Simple gain calculation

- Assume uniform pump distribution $G_0 = \exp[N_{inv}\sigma_{21}L]$ Small-signal gain
- Available energy for extraction:

$$E_{stor} = N_{inv} A L h V_{21} \rightarrow N_{inv} = \frac{E_{stor}}{A L h V_{21}}$$

A = area of beam

$$G_0 = \exp\left[\frac{E_{stor}}{A}\frac{\sigma_{21}}{hv_{21}}\right]$$

- Energy fluence = energy per unit area
- Define:
 - "stored fluence"

- "saturation fluence"

$$\Gamma_{stor} = \frac{E_{stor}}{A}$$

 $\sigma_{_{21}}$

I_{sat}

$$_{0} = \exp\left[\frac{\Gamma_{stor}}{\Gamma_{sat}}\right]$$

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Example: Ti:sapphire amplifier

Pump laser has 10mJ per pulse, calculate spot size in crystal for G₀ = 5

Ti:sapphire:

- λ_{21} =800nm, hv = 1.55eV = 2.48x10⁻¹⁹ J
- $-\sigma_{21} = 2.8 \text{ x } 10^{-19} \text{ cm}^2$
- $-\Gamma_{sat} = 0.85 \text{ J/cm}^2$



$$\Gamma_{\text{stor}} = \Gamma_{\text{sat}} \ln[G_0] = 1.37 \text{ J/cm}^2$$

 $A = 7.3 \times 10^{-3} \text{ cm}^2$

 $w_0 = 480 \text{ um}$

For pulse duration of 10ns, pump intensity is

 $I = 1.37 \times 10^8 \text{ W/cm}^2$

Optical pumping geometries

Technique depends on properties of light source

- Diffuse:
 - flashlamp, arc lamp (CW), LED sun
 - Pump chambers, non-imaging concentrators
- Laser beams with poor divergence:
 - laser diode elements and arrays, multi-mode fibercoupled LDs
 - Longitudinal and side pumping
- High-quality laser beams
 - Longitudinal pumping



Fig. 6.47. Major pump configurations a) side pumping with flashlamps, b) side-and edge pumping with laser diodes, c) end pumped lasers, d) face pumping with flashlamps or laser diodes.

Population dynamics of absorption

• Closed 2 level system, assume $g_1 = g_2$ $\frac{dN_1}{dN_2}$

• Since system is closed, reduce to one equation for population difference: $\Delta N = N_1 - N_2$ $N_t = N_1 + N_2$

$$\frac{dN_1}{dt} - \frac{dN_2}{dt} = \frac{d}{dt}\Delta N = -2\frac{dN_2}{dt} \qquad \qquad N_t = \Delta N + 2N_2$$

$$\frac{d}{dt}\Delta N = -2\left(W\Delta N - A_{21}N_2\right)$$

$$\frac{d}{dt}\Delta N = -2W\Delta N + A_{21}\left(N_t - \Delta N\right) = -\Delta N\left(A_{21} + 2W\right) + A_{21}N_t$$

$$- \text{ Steady state:} \qquad \Delta N = \frac{N_t}{1 + 2W\tau_{21}} \qquad A_{21} = 1/\tau_{21}$$

Saturation of absorption

- The key parameter in this situation is W T_{21}
 - $W_{21} = \rho_v B_{21}$
 - Low intensity, 2W $T_{21} \ll 1$, $\Delta N \approx N_t$
 - High intensity, 2W τ_{21} >> 1, Δ N ≈ 0. Here N₁ ≈ N₂
- Energy balance:

Input power (into 4π) Absorbed by atoms

Stimulated emission (back into beam)

• Radiated power per unit volume:

$$\frac{dP}{dV} = hV_{21}W\Delta N(W) = hV_{21}\frac{N_tW}{1+2W\tau_{21}} \to hV_{21}\frac{N_t}{2\tau_{21}} \quad \text{For W } \tau_{21} >> 1$$

Power radiated in high intensity limit: half of atoms are radiating

$$\Delta N = \frac{N_t}{1 + 2W\tau_{21}}$$

$$\Delta N = N_1 - N_2$$

Saturation intensity

- Absorbed power per atom: $\sigma_{12}I$
- Absorption rate: Absorption rate: $W = \frac{1}{hv_{21}}$ • In steady state: $\frac{\Delta N}{N_t} = \frac{1}{1+2W\tau_{21}} = \frac{1}{1+2\frac{\sigma_{12}I}{hv_{21}}\tau_{21}} = \frac{1}{1+2\frac{\sigma_{12}I}{hv_{21}}\tau_{21}}$
- Saturation intensity for absorption:
 - 2: transition affects both levels at once I_{sat}
 - At I = I_{sat} , stimulated and spontaneous emission rates are equal.
- Intensity-dependent absorption coefficient:

$$\alpha(I) = \frac{\alpha_0}{1 + I/I_{sat}}$$

At high intensity, material absorbs less.

Saturable absorbers are used for pulsed lasers: Q-switching and mode-locking

Saturated CW propagation through absorbing medium

 For a given thickness for an absorbing medium, the transmission will increase with intensity



Pulsed input: saturation fluence $\Delta N = N_1 - N_2$

Rewrite equation using intensity:

$$\frac{d}{dt}\Delta N = -\Delta N \left(A_{21} + \frac{2\sigma}{hv_{21}} I(t) \right) + A_{21}N_t \equiv -\Delta N \left(A_{21} + \frac{I(t)}{\Gamma_{sat}} \right) + A_{21}N_t$$

- Scaling of equation
 - Two timescales: τ_p and τ_{21} , but pay attention to weighting $d_{AN} = AN \begin{pmatrix} 1 & \Gamma_{in} & 1 \end{pmatrix} + \begin{pmatrix} 1 & N & 2N_2 & \Gamma_{in} & 1 \\ N & N & N \end{pmatrix}$

$$\frac{d}{dt}\Delta N = -\Delta N \left(\frac{1}{\tau_{21}} + \frac{1}{\Gamma_{sat}}\frac{1}{\tau_p}\right) + \frac{1}{\tau_{21}}N_t = \frac{2N_2}{\tau_{21}} - \frac{1}{\Gamma_{sat}}\frac{1}{\tau_p}\Delta N$$

• For short pulse input: ignore stimulated emission and fluorescence

$$\frac{\Gamma_{in}}{\Gamma_{sat}} \frac{1}{\tau_p} |\Delta N| \gg \frac{2N_2}{\tau_{21}} \longrightarrow \frac{d}{dt} \Delta N \approx -\frac{\Gamma_{in}}{\Gamma_{sat}} \frac{1}{\tau_p} \Delta N \longrightarrow \ln\left[\frac{\Delta N(t)}{\Delta N(0)}\right] \approx -\frac{1}{\Gamma_{sat}} \int_0^t I(t') dt'$$
$$\longrightarrow \Delta N(t) = N_1(t) - N_2(t) \approx N_t \exp\left[-\frac{1}{\Gamma_{sat}} \int_0^t I(t') dt'\right] = N_t \exp\left[-\frac{\Gamma_{in}}{\Gamma_{sat}}\right]$$

Short pulse limit

- For short pulse input: $T_p << T_{21}$, so ignore fluorescence
 - Medium just integrates energy of pulse.
 - Example: Ti:sapphire: τ₂₁=3.2µs, τ_p=10ns or 200ns for Q-switched Nd:YAG lasers pumped with flashlamps or CW arc lamps
- Square input pulse Gaussian input pulse

 $-\tau = 3$, $I_{0/}I_{sat} = 0.1$, (no fluorescence)



Shape of transmitted pulse is affected

Long pulse limit

 For *long* pulse input: τ_p>>τ₂₁, and peak I << I_{sat}, ΔN(t) follows I(t)

 $\rightarrow \frac{d}{dt} \Delta N \ll A_{21} N_t$ $\frac{\Delta N}{N_t} = \frac{1}{1 + I(t)/I_{sat}}$

Quasi-static, quasi-CW limit N_t adiabatically follows I(t)



Gain saturation

• Consider a 4-level system:



No factor of 2 $I_{sat} = \frac{hv_{21}}{\sigma_{21}\tau_{21}} = \frac{\Gamma_{sat}}{\tau_{21}}$ $g(I) = \frac{g_0}{1 + I/I}$

Beam growth during amplification

Calculation just as with absorption

$$\int_{I_0}^{I} \left(\frac{1}{I} + \frac{1}{I_{sat}}\right) dI = + \int_{0}^{L} g_0 \, dz \rightarrow \ln\left[\frac{I(z)}{I(0)}\right] + \frac{I(z) - I(0)}{I_{sat}} = + g_0 z$$
Net gain over 1 absorption length
$$\int_{I_0}^{I_0} \frac{1}{I_{sat}} = 0.1$$

$$\int_{I_0}^{I_0} \frac{1}{I_{sat}} = 0.1$$

$$\int_{I_0}^{I_0} \frac{1}{I_{sat}} = 10$$

Even though saturated gain is low, it is efficient at extracting stored energy

Spatial dependence

- Gain follows distribution of pump intensity
- Spatial variation of gain affects beam profile
- Examples:
 - Iongitudinal pumping with Gaussian beam leads to gain narrowing of spatial profile. More gain in center, less at edges
 - Saturated absorption by a Gaussian beam: saturation in center suppresses intensity there. Leads to widening of output beam.

Pulse amplification: saturated gain algorithm


Example: Ti:sapphire multipass amp

- Seed pulse from pulsed laser oscillator: 1nJ (800nm)
- Amplify to 1mJ, use 7mJ of pump energy (532nm)
- Multipass designs: spatially separate beams

Three-mirror ring preamp:

- Up to 12 passes
- Focused beam in crystal
- 2 mirror alignment

Q-switched Nd;YLF IOW max Mask TI: Sapphire Pulses input

Bowtie power amp:

- Collimated beam
- 8 mirrors



Multipass design

- Assume uniform pumping with round beams
- Calculate stored fluence and small signal gain
- Use saturated gain expression to calculate new energy after 1st pass
- Subtract extracted energy from stored energy (over seed spot area)
- Repeat for N passes

Conditions: 1nJ seed, 7mJ pump energy, 95% absorption, 10% loss/pass Stored energy: hV_{mad}

$$E_{stor} = E_{pump} \eta_{abs} \frac{h v_{seed}}{h v_{pump}} = 4.4 \, mJ$$

Small signal gain estimate:

$$G_0 = \left(\frac{E_{\text{target}}}{E_{\text{seed}}}\right)^{1/N} \frac{1}{1-L} = 4.42$$

Estimated spot size:

$$A_{pump} = \frac{E_{stor}}{\Gamma_{sat} \ln[G_0]}, \quad w_p = 300 \,\mu m$$

Multipass: Simple calculated results

 Small signal gain estimate works as long as stored energy is not depleted



- Smaller seed size to ensure full overlap with pump
- Avoid damage thresholds for pump and seed
- Saturate at desired energy to reduce noise
- Account for size change in Brewster cut crystal



Polarization issues in pumping birefringent materials

- For Ti:sapphire, both polarizations contribute to seed gain along c-axis
- Much higher pump absorption for E along c-axis

 $- \alpha$ across c-axis is about 40% lower than along c-axis



Transverse Pumping Gain Estimates

- Seed: 2nJ
 - Cavity Losses: ~1%
 - т_{pass}: 1ns
- Pump (CW): 1kW (Total: 2X .5kW Bars)
 - η_{Abs}=63.2%
 - η_{QD}=55.6%
 - $-\eta_{Pump} = \eta_{Abs} \eta_{QD} = 35.1\%$
 - − Heat: ~560 W
 - Significant (Cylindrical) Thermal Lens Expected
 - w=30um
- Single Pass Gain (small signal)
 - Astigmatic Seed: g≈1.64
 - w_x=200um, w_y=30µm
 - Spatially Chirped Seed: g≈1.64
 - w_x=2mm, w_y=30µm

•Multi-Pass Extraction: 37 Passes

-Astigmatic Mode: ~136uJ (small extraction area) -Spatially Chirped:~.53mJ (46% extraction)

Central dip in gain: spatial gain mode *expansion*. This could be used to counter gain narrowing for spatially-chirped seed



Frequency dependence: account for lineshapes

• Absorption and gain coefficients and saturation intensity both depends on frequency

$$\alpha(I,v) = \frac{\alpha_0(v-v_0)}{1+\frac{I(v)}{I_{sat}(v-v_0)}}$$

- For broadband input, saturation changes shape of transmitted spectrum
 - Absorption: power broadening
 - Gain: spectral gain narrowing

Amplified Spontaneous Emission (ASE)

- Spontaneous emission is emitted into 4π steradians, but is amplified on the way out if there is gain.
 - D ______ >
 - ASE can be considered to be a noise source
 - ASE is more directional than fluorescence, but not as directional as a coherent laser beam
 - Some high-gain lasers are essentially ASE sources (no mirrors)
- Implications for amplifier design
 - ASE can deplete stored energy before pulse extraction
 - Use timing and good seed energy to extract energy from medium before ASE
 - Ensure that transverse gain is smaller than longitudinal to avoid parasitic depletion.

Self-absorption and "optically-thick" media

- A related phenomenon for an absorbing medium is when radiation is *absorbed* along the way out.
- More absorption near the line center, so the transmitted light is broader in spectrum.
- For an extended luminous body (e.g. the Sun), the individual spectral lines get merged together to look like the blackbody.

Interference: ray and wave pictures

 Interference results from the sum of two waves with different phase:

$$E_{tot}\left(\Delta\phi\right) = E_1 e^{ikz} + E_2 e^{ikz + \Delta\phi}$$

We measure intensity, which leads to interference

$$I_{tot} (\Delta \phi) \propto \left| E_1 e^{ikz} + E_2 e^{ikz + \Delta \phi} \right|^2 = \left| E_1 + E_2 e^{i\Delta \phi} \right|^2$$
$$= I_1 + I_2 + \sqrt{I_1 I_2} e^{i\Delta \phi} + \sqrt{I_1 I_2} e^{-i\Delta \phi}$$
$$= I_1 + I_2 + 2\sqrt{I_1 I_2} \cos(\Delta \phi)$$

- For the case where $I_1 = I_2$,

$$I_{tot}(\Delta\phi) = 2I(1 + \cos(\Delta\phi)) = 4I\cos^2(\Delta\phi/2)$$

• How to generate, calculate phase difference?

The Fizeau Wedge Interferometer

The Fizeau wedge yields a complex pattern of variable-width fringes, but it can be used to measure the wavelength of a laser beam.



Fizeau wedge calculation

Interference between reflections from internal surfaces



- Angle is very small, neglect change in direction
- Path difference: $\Delta l = 2L \sin \alpha \approx 2L \alpha$
- Phase difference: $\Delta \phi = \frac{\omega}{c} n \Delta l \approx 2\pi \frac{2L}{\lambda} \alpha$ n = 1
- Interference: $I_{tot}(\Delta \phi) = I_1 + I_2 + 2\sqrt{I_1I_2}\cos(\Delta \phi)$
- One fringe from one max to the next, so maxima are at $\Delta \phi = 2\pi m$
- In this interferometer, minimum path = 0, we can measure absolute wavelength: $\Delta \phi = 2\pi m = 2\pi \frac{2L}{\lambda} \alpha \rightarrow \lambda = \frac{2L}{m} \alpha$

Newton's Rings





Newton's Rings

Get constructive interference when an integral number of half wavelengths occur between the two surfaces (that is, when an integral number of full wavelengths occur between the path of the transmitted beam and the twice reflected beam).



Tilted window: ray propagation

- Calculate phase shift caused by the insertion of the window into an interferometer.
- Ray optics:
 - Add up optical path for each segment
 - Subtract optical path w/o window



Tilted window: wave propagation

Write expression for tilted plane wave

$$E(x,z) = E_0 \exp\left[i\left(k_x x + k_z z\right)\right] = E_0 \exp\left[i\frac{\omega}{c}n\left(x\sin\theta_2 + z\cos\theta_2\right)\right]$$

Snell's Law: phase across surfaces is conserved

 $k_{x}x = \frac{\omega}{c}n\sin\theta \qquad \text{is constant}$ $\Delta\phi = (k_{2}\cos\theta_{2})L_{w} - (k_{1}\cos\theta_{1})L_{w}$

 This approach can be used to calculate phase of prism pairs and grating pairs



Multiple-beam interference: The Fabry-Perot Interferometer or Etalon

A Fabry-Perot interferometer is a pair of **parallel** surfaces that reflect beams back and forth. An etalon is a type of Fabry-Perot etalon, and is a piece of glass with parallel sides.

The transmitted wave is an infinite series of multiply reflected beams.



Multiple-beam interference: general formulation



r, t = reflection, transmission coefficients from air to glassr', t' = ""from glass to air

δ = round-trip phase delay inside medium = k₀(2 n L cos θ_t)

Transmitted wave:

$$E_{0t} = tt' e^{-i\delta/2} E_0 \left(1 + (r')^2 e^{i\delta} + ((r')^2 e^{i\delta})^2 + ((r')^2 e^{i\delta})^3 + \dots \right)$$

Reflected wave:

$$E_{0r} = rE_0 + tt'r'e^{i\delta}E_0 + tt'r'(((r')^2 e^{i\delta})^2 E_0 + \dots$$

Stokes Relations for reflection and transmission



Notes:

- relations apply to angles connected by Snell's Law
- true for any polarization, but not TIR
- convention for which interface experiences a sign change can vary

$$\begin{aligned} \textbf{Fabry-Perot transmission} & \text{Stokes'} \quad r' = -r \\ \text{relations} \quad r'^2 = r^2 \\ \text{The transmitted wave field is:} \\ E_{0r} = tt' e^{i\delta/2} E_0 \Big(1 + (r')^2 e^{i\delta} + ((r')^2 e^{i\delta})^2 + ((r')^2 e^{i\delta})^3 + ... \Big) \\ = tt' e^{i\delta/2} E_0 \Big(1 + r^2 e^{i\delta} + (r^2 e^{i\delta})^2 + (r^2 e^{i\delta})^3 + ... \Big) \\ \Rightarrow \quad E_{0r} = tt' E_0 / (1 - r^2 e^{-i\delta}) \\ \text{Where:} \\ \text{Power transmittance:} \quad T \equiv \left| \frac{E_{0r}}{E_0} \right|^2 = \left| \frac{tt' e^{i\delta/2}}{1 - r^2 e^{i\delta}} \right|^2 = \frac{(tt')^2}{(1 - r^2 e^{+i\delta})(1 - r^2 e^{-i\delta})} \\ = \left[\frac{(tt')^2}{(1 + r^4 - 2r^2 \cos(\delta))} \right] = \left[\frac{(1 - r^2)^2}{(1 + r^4 - 2r^2 [1 - 2\sin^2(\delta/2)])} \right] = \left[\frac{(1 - r^2)^2}{(1 - 2r^2 + r^4 + 4r^2 \sin^2(\delta/2)]} \right] \\ \text{Dividing numerator and denominator by } (1 - r^2)^2 \\ \qquad \qquad T = \frac{1}{1 + F \sin^2(\delta/2)} \quad \text{where:} \quad F = \left[\frac{2r}{(1 - r^2)} \right]^2 \end{aligned}$$

Multiple-beam interference: simple limits

Reflected waves

$$T = \frac{1}{1 + F\sin^2\left(\delta / 2\right)}$$

Full transmission: $sin() = 0, d = 2 \pi m$



Minimum transmission: sin() = 1, d = 2 π (m+1/2)



Constructive interference for reflected wave

Etalon transmittance vs. thickness, wavelength, or angle π 1



- The transmittance varies significantly with thickness or wavelength.
- We can also vary the incidence angle, which also affects δ .
- As the reflectance of each surface (R=r²) approaches 1, the widths of the high-transmission regions become very narrow.

The Etalon Free Spectral Range

The Free Spectral Range is the wavelength range between transmission maxima.



1/(round trip time)

Etalon Linewidth

The Linewidth δ_{LW} is a transmittance peak's full-width-half-max (FWHM).

$$T = \frac{1}{1 + F \sin^2\left(\delta / 2\right)}$$

A maximum is where $\delta/2 \approx m\pi + \delta'/2$ and $\sin^2(\delta/2) \approx \delta'/2$

Under these conditions (near resonance),

$$T = \frac{1}{1 + F\delta'^2 / 4}$$

This is a Lorentzian profile, with FWHM at:

$$\frac{F}{4} \left(\frac{\delta_{LW}}{2} \right)^2 = 1 \quad \Rightarrow \quad \delta_{LW} \approx 4 / \sqrt{F}$$

This transmission linewidth corresponds to the minimum resolvable wavelength.

Etalon Finesse

The Finesse, \Im , is the ratio of the Free Spectral Range and the Linewidth:



Using: $F = \left[\frac{2r}{1}\right]^2$

$$\Im = \frac{\pi}{1 - r^2} \qquad \text{taking } r \approx 1$$

The Finesse is the number of wavelengths the interferometer can resolve.

Multilayer coatings

Typical laser mirrors and camera lenses use many layers.

The reflectance and transmittance can be custom designed





Quarter-wave stack

Double-quarter

Multilayer thin-films: wave/matrix treatment



- Use boundary conditions to relate fields at the boundaries
- Phase shifts connect fields just after I to fields just before II
- Express this relation as a transfer matrix
- Multiply matrices for multiple layers

High-reflector design



Reflectivity can reach > 99.99% at a specific wavelength > 99.5% for over 250nm Bandwidth and reflectivity are better for "S" polarization.

Interference filter design



A thin layer is sandwiched between two high reflector coatings -very large free spectral range, high finesse

- typically 5-10nm bandwidth, available throughout UV to IR





Raytracing

- Approches:
 - Paraxial tracing (assume small angle to optical axis)
 - Computer tracing (no approximations). Example: Zemax, Oslo,...
- Design procedure
 - Find existing design close to what could work
 - Paraxial trace with ray diagram
 - Calculate magnification, limiting apertures
 - Optimize with ABCD matrices or computer program
 - Analyze aberrations

ABCD ray matrices

- Formalism to propagate rays through optical systems
 - Keep track of ray height r and ray angle $\theta = dr/dz = r'$
 - Treat this pair as a vector: $\left(\begin{array}{c} r \\ r' \end{array} \right)$
 - Optical system will modify both the ray height and angle, e.g.

$$\begin{pmatrix} r_2 \\ r'_2 \end{pmatrix} = \begin{pmatrix} A & B \\ C & D \end{pmatrix} \begin{pmatrix} r_1 \\ r'_1 \end{pmatrix}$$

- Successive ABCD matrices multiply from the left
- Translation

 $r_{2} =$

$$\begin{array}{c} r_2 = r_1 + Lr_1' \\ r_2' = r_1' \end{array} \longrightarrow \left(\begin{array}{cc} 1 & L \\ 0 & 1 \end{array} \right)$$



Refraction in ABCD

- Translation: $\begin{pmatrix} 1 & L \\ 0 & 1 \end{pmatrix}$
- Flat interface

n₁

 n_2

• Window: calculate matrix

$$\rightarrow \begin{pmatrix} 1 & 0 \\ 0 & n \end{pmatrix} \begin{pmatrix} 1 & L \\ 0 & 1 \end{pmatrix} \begin{pmatrix} 1 & 0 \\ 0 & 1/n \end{pmatrix}$$
$$= \begin{pmatrix} 1 & 0 \\ 0 & n \end{pmatrix} \begin{pmatrix} 1 & L/n \\ 0 & 1/n \end{pmatrix} = \begin{pmatrix} 1 & L/n \\ 0 & 1 \end{pmatrix}$$
Effective thickness reduced by *n*

Curved surfaces in ABCD

 Thin lens: matrix computes transition from one side of lens to other



• Spherical interface: radius R

$$\rightarrow \left(\begin{array}{ccc} 1 & 0\\ \frac{n_1 - n_2}{n_2} \frac{1}{R} & \frac{n_1}{n_2} \end{array}\right)$$

Curved wavefronts

- Rays are directed normal to surfaces of constant phase
 - These surfaces are the wavefronts
 - Radius of curvature is approximately at the focal point



• Spherical waves are solutions to the wave equation (away from r = 0) $\nabla^{2}E + \frac{n^{2}\omega^{2}}{c^{2}}E = 0$ $E \propto \frac{1}{r}e^{i(\pm kr - \omega t)}$ $E \propto \frac{1}{r}e^{i(\pm kr - \omega t)}$
Paraxial approximations

- For **rays**, paraxial = small angle to optical axis
 - Ray slope: $\tan \theta \approx \theta$

 $e^{ikr} = \exp\left[ik\sqrt{x^2 + y^2 + z^2}\right]$

• For **spherical waves** where power is directed forward:

$$k\sqrt{x^{2} + y^{2} + z^{2}} = kz\sqrt{1 + \frac{x^{2} + y^{2}}{z^{2}}} \approx kz\left(1 + \frac{x^{2} + y^{2}}{2z^{2}}\right) \qquad \begin{array}{l} \text{Expanding to} \\ 1^{\text{st}} \text{ order} \end{array}$$
$$e^{i(kr-\omega t)} \rightarrow e^{ikz} \exp\left[i\left(k\frac{x^{2} + y^{2}}{2z} - \omega t\right)\right] \qquad z \text{ is radius of curvature} \end{array}$$

Wavefront = surface of constant phase For x, y >0, t must increase. Wave is diverging:

$$k\frac{x^2+y^2}{2z} = \omega t$$

3D wave propagation $\nabla^{2}\mathbf{E} - \frac{n_{j}^{2}}{c^{2}} \frac{\partial^{2}}{\partial t^{2}} \mathbf{E} = \frac{\partial^{2}}{\partial z^{2}} \mathbf{E} + \nabla_{\perp}^{2}\mathbf{E} - \frac{n(\mathbf{r})^{2}}{c^{2}} \frac{\partial^{2}}{\partial t^{2}} \mathbf{E} = 0$ ote: $\nabla_{\perp}^{2} = \partial_{x}^{2} + \partial_{y}^{2}$ $\nabla_{\perp}^{2} = \frac{1}{r} \partial_{r} (r \partial_{r}) + \frac{1}{r^{2}} \partial_{\phi}^{2}$

- Note:
 - All linear propagation effects are included in LHS: diffraction, interference, focusing...
 - Previously, we assumed plane waves where transverse derivatives are zero.
- More general examples:
 - Gaussian beams (including high-order)
 - Waveguides
 - Arbitrary propagation
 - Can determine discrete solutions to linear equation (e.g. Gaussian modes, waveguide modes), then express fields in terms of those solutions.

Paraxial, slowly-varying approximations

- Assume
 - waves are forward-propagating: $\mathbf{E}(\mathbf{r},t) = \mathbf{A}(\mathbf{r})e^{i(kz-\omega_0 t)} + c.c.$
 - Refractive index is isotropic

$$\frac{\partial^2}{\partial z^2}\mathbf{A} + 2ik\frac{\partial}{\partial z}\mathbf{A} - k^2\mathbf{A} + \nabla_{\perp}^2\mathbf{A} + \frac{n^2\omega_0^2}{c^2}\mathbf{A} = 0$$

- Fast oscillating carrier terms cancel (blue)
- Slowly-varying envelope: compare red terms
 - Drop 2nd order deriv if $\frac{2\pi}{\lambda} \frac{1}{L} A \gg \frac{1}{L^2} A$
 - This ignores:
 - Changes in z as fast as the wavlength
 - Counterpropagating waves

Gaussian beam solutions to wave equation

• Without any source term, paraxial equation is

$$2ik\frac{\partial}{\partial z}\mathbf{A} + \nabla_{\perp}^{2}\mathbf{A} = 0$$

• Gaussian beam solutions can be written as:

$$A(r,z) = A_0 \frac{1}{1+i\xi} e^{-\frac{r^2}{w_0^2(1+i\xi)}} \qquad \xi = \frac{z}{z_R} \qquad z_R = n \frac{\pi w_0^2}{\lambda} = \frac{k w_0^2}{2}$$

Rayleigh range

$$E(r,z,t) = A_0 \frac{W_0}{w(z)} e^{i(kz-\omega t)} e^{-\frac{r^2}{w^2(z)}} e^{i\frac{kr^2}{2R(z)}} e^{-i\eta(z)}$$

Standard form of Gaussian beam equations



Complex q form for Gaussian beam

 This combines beam size and radius of curvature into one complex parameter

This form is used for for ABCD calculations



Complex q vs standard form

$$u(r,z) = \frac{1}{q(z)}e^{-ik\frac{r^2}{2q(z)}}$$
 with $\frac{1}{q(z)} = \frac{1}{R(z)} - i\frac{\lambda}{\pi w^2(z)}$

Expand exponential:

$$\exp\left[-ik\frac{r^2}{2q(z)}\right] = \exp\left[-ik\frac{r^2}{2}\left(\frac{1}{R(z)} - i\frac{\lambda}{\pi w^2(z)}\right)\right]$$
$$= \exp\left[-ik\frac{r^2}{2}\frac{1}{R(z)} - i\frac{2\pi}{\lambda}\frac{r^2}{2}\left(-i\frac{\lambda}{\pi w^2(z)}\right)\right] = e^{-ik\frac{r^2}{2R(z)}}e^{-\frac{r^2}{w^2(z)}}$$

$$a + ib = \sqrt{a^2 + b^2} e^{i \arctan(b/a)}$$

Expand leading inverse q:

$$\frac{1}{q(z)} = -i\left(\frac{iz}{z^2 + z_R^2} + \frac{z_R}{z^2 + z_R^2}\right) = -i\left(\frac{\sqrt{z^2 + z_R^2}}{z^2 + z_R^2}\right)e^{i\arctan(z/z_R)}$$
$$= -i\left(\frac{1}{z_R\sqrt{1 + z^2/z_R^2}}\right)e^{i\arctan(z/z_R)} = \frac{w_0}{iz_Rw(z)}e^{i\eta(z)}$$

Difference between Siegman's complex q and standard form

$$u(r,z) = \frac{1}{q(z)} e^{-ik\frac{r^2}{2q(z)}} = \frac{1}{iz_R} \frac{w_0}{w(z)} e^{i\eta(z)} e^{-ik\frac{r^2}{2R(z)}} e^{-\frac{r^2}{w^2(z)}}$$
$$E(r,z,t) = A_0 \frac{w_0}{w(z)} e^{i(kz-\omega t)} e^{-\frac{r^2}{w^2(z)}} e^{i\frac{kr^2}{2R(z)}} e^{-i\eta(z)}$$

- Siegman's form for the complex q is used almost everywhere for the ABCD calculations.
- He uses the exp[+ I w t] convention, which accounts for the sign difference in the complex exponentials.
- With exp[-I w t] convention, define q as:

$$\frac{1}{q(z)} = \frac{1}{R(z)} + i \frac{\lambda}{\pi w^2(z)} = \frac{1}{z - i z_R}$$

Compare Boyd's form to standard:

 Boyd's complex form is consistent with standard Gaussian beam form

$$A(r,z) = A_0 \frac{1}{1+i\xi} e^{-\frac{r^2}{w_0^2(1+i\xi)}} = A_0 \frac{1}{1+iz/z_R} e^{-\frac{r^2}{w_0^2(1+iz/z_R)}}$$

$$\frac{1}{1+i\xi} = \frac{1}{1+iz/z_R} = \frac{z_R}{z_R+iz} = \frac{-iz_R}{z-iz_R} = \frac{-iz_R}{q(z)}$$

$$A(r,z) = A_0(-iz_R)\frac{1}{q(z)}e^{+\frac{iz_Rr^2}{w_0^2q(z)}} = -iz_RA_0\frac{1}{q(z)}e^{+\frac{ikr^2}{2q(z)}}$$