Begin
02-20-2023

Emission and Absorption – Population Rate Equations

So far we have derived a set of Eqs. of Motion for the elements of the Density Matrix:

$$\dot{\rho}_{11} = -\Gamma_1 \rho_{11} + A_{21}\rho_{22} - \frac{i}{2}\left(\chi\rho_{12} - \chi^*\rho_{21}\right)$$

$$\dot{\rho}_{22} = -\Gamma_2 \rho_{22} - A_{21}\rho_{22} + \frac{i}{2}\left(\chi\rho_{12} - \chi^*\rho_{21}\right)$$

$$\dot{\rho}_{12} = (i\Delta - \beta)\rho_{12} + \frac{i\chi^*}{2}\left(\rho_{22} - \rho_{11}\right) = \dot{\rho}_{21}^*$$

where $\beta = \frac{1}{\tau} + \frac{A_{21}}{2} + \frac{\Gamma_1 + \Gamma_2}{2}$

(*) **These eqs. are difficult to solve in the general case. See, e. g., Allen & Eberly for discussion of some special cases and a reference to original work by Torrey et al.**

(*) **For ≥ 3 levels the Density Matrix Equations get very cumbersome and it is desirable to simplify the description when possible.**

(*) **One such simplification takes the form of Rate Equations for the populations only.**

Steady State Solutions: **(requires** $\Gamma_1 = \Gamma_2 = 0$ **)**

Let $\dot{\rho}_{12} = 0$ ⇒

$$\begin{cases} \rho_{12} = \dfrac{i\chi^*/2}{\beta - i\Delta}\left(\rho_{22} - \rho_{11}\right) \\[2ex] \rho_{21} = \dfrac{-i\chi/2}{\beta + i\Delta}\left(\rho_{22} - \rho_{11}\right) \end{cases}$$

⇓

$$\chi\rho_{12} - \chi^*\rho_{21} = \frac{i|\chi|^2\beta}{\Delta^2 + \beta^2}\left(\rho_{22} - \rho_{11}\right)$$

Plug into eqs for populations to get

$$\dot{\rho}_{11} = A_{21}\rho_{22} + \frac{|\chi|^2\beta/2}{\Delta^2 + \beta^2}\left(\rho_{22} - \rho_{11}\right) = 0$$

$$\dot{\rho}_{22} = -A_{21}\rho_{22} - \frac{|\chi|^2\beta/2}{\Delta^2 + \beta^2}\left(\rho_{22} - \rho_{11}\right) = 0$$

From these eqs. we can find <u>steady state values</u> for the populations and coherences in terms of $\chi, \Delta, A_{21}, \beta$ **when (and only when)** $\dot{\rho}_{11} = \dot{\rho}_{22} = 0$

Emission and Absorption – Population Rate Equations

Steady State Solutions: (requires $\Gamma_1 = \Gamma_2 = 0$)

Let $\dot{\rho}_{12} = 0$ ⇨

$$\begin{cases} \rho_{12} = \dfrac{i\chi^*/2}{\beta - i\Delta}(\rho_{22} - \rho_{11}) \\ \rho_{21} = \dfrac{-i\chi/2}{\beta + i\Delta}(\rho_{22} - \rho_{11}) \end{cases}$$

$$\chi\rho_{12} - \chi^*\rho_{21} = \frac{i|\chi|^2\beta}{\Delta^2 + \beta^2}(\rho_{22} - \rho_{11})$$

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$$\dot{\rho}_{11} = A_{21}\rho_{22} + \frac{|\chi|^2\beta/2}{\Delta^2 + \beta^2}(\rho_{22} - \rho_{11}) = 0$$

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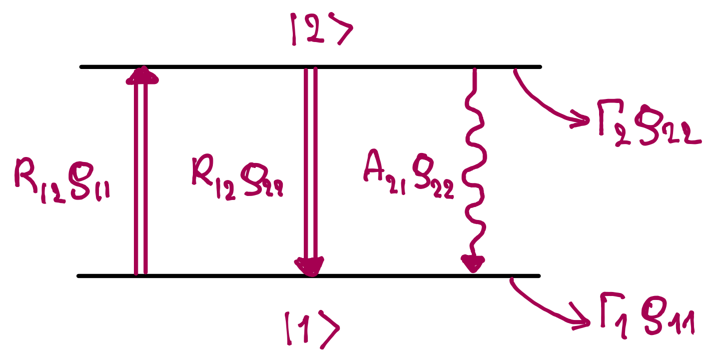
<u>Note</u>: The terms remaining after elimination of ρ_{12}, ρ_{21} are commonly identified with <u>induced</u> or <u>stimulated</u> processes. They are proportional to $|\chi|^2, |E_0|^2$ and thus the <u>intensity</u> of the light field.

<u>Def</u>: Absorption Rate = Stimulated Emission Rate

$$R_{12} = \frac{|\chi|^2\beta/2}{\Delta^2 + \beta^2} = \frac{|\vec{r}_{12}\cdot\vec{\varepsilon}E_0/\hbar|^2\beta/2}{(\omega_{21} - \omega)^2 + \beta^2}$$

Schematic:



Emission and Absorption – Population Rate Equations

<u>Note</u>: The terms remaining after elimination of ρ_{12}, ρ_{21} are commonly identified with <u>induced</u> or <u>stimulated</u> processes. They are proportional to $|X|^2, |E_0|^2$ and thus the <u>intensity</u> of the light field.

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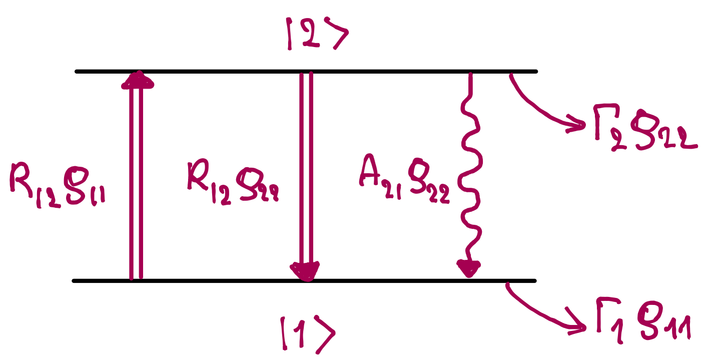

<u>Elastic Collision Broadening</u>

In hot and dense gases the dominant source of relaxation is often elastic collisions between atoms

Let $\beta \gg \Gamma_1, \Gamma_2, A_{21}$ ⇨ ρ_{12} reaches steady state much faster than ρ_{11}, ρ_{22}

We can solve the eq. for $\dot{\rho}_{12}$ assuming it is in steady state for given values of ρ_{11}, ρ_{22}

This yields Rate Equations for the populations only, valid in the <u>collision broadened</u> regime

$$\dot{\rho}_{11} = -\Gamma_1\rho_{11} + A_{21}\rho_{22} + R_{12}(\rho_{22}-\rho_{11}) \neq 0$$

$$\dot{\rho}_{22} = -\Gamma_2\rho_{22} - A_{21}\rho_{22} - R_{12}(\rho_{22}-\rho_{11}) \neq 0$$

(*) This is another example of <u>adiabatic elimination</u> of a fast variable (the coherence), leaving us with simpler equations for the slower variables.

(*) From these we can find the <u>transient</u> behavior of the coherences ρ_{11}, ρ_{22}

Emission and Absorption – Population Rate Equations

Elastic Collision Broadening

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(∗) This is another example of adiabatic elimination of a fast variable (the coherence), leaving us with simpler equations for the slower variables.

(∗) From these we can find the transient behavior of the coherences ρ_{11}, ρ_{22}

Note: When collisions are very frequent the dipole $\langle \hat{\vec{\eta}} \rangle$ is oriented at random relative to the driving field. In that case

$$\langle |\vec{\eta}_{12} \cdot \vec{\varepsilon} E_0|^2 \rangle_{angles} = \frac{1}{3} \eta_{12}^2 |E_0|^2 ⇨$$

$$R_{12} = \frac{\langle |\vec{\eta}_{12} \cdot \vec{\varepsilon} E_0/\hbar|^2 \rangle_{angles} \beta/2}{\Delta^2 + \beta^2} = \frac{1}{3} \frac{|\chi|^2 \beta/2}{\Delta^2 + \beta^2}$$

Photon Flux and Cross Section

Let $R_{12} \equiv \sigma(\Delta)\phi$ where $\hbar\omega \underset{\text{“photon flux”}}{\phi} = \underbrace{\frac{1}{2} c \varepsilon_0 |E_0|^2}_{\text{intensity}}$

This allows us to recast the Rate Eqs

$$\boxed{\begin{aligned} \dot{\rho}_{11} &= -\Gamma_1 \rho_{11} + A_{21}\rho_{22} + \sigma(\Delta)\phi(\rho_{22}-\rho_{11}) \\ \dot{\rho}_{22} &= -\Gamma_2 \rho_{22} - A_{21}\rho_{22} - \sigma(\Delta)\phi(\rho_{22}-\rho_{11}) \end{aligned}}$$

Emission and Absorption – Population Rate Equations

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"photon flux"

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$$\boxed{\begin{aligned}\dot{\rho}_{11} &= -\Gamma_1\rho_{11} + A_{21}\rho_{22} + \sigma(\Delta)\phi(\rho_{22}-\rho_{11})\\ \dot{\rho}_{22} &= -\Gamma_2\rho_{22} - A_{21}\rho_{22} - \sigma(\Delta)\phi(\rho_{22}-\rho_{11})\end{aligned}}$$

We see that per atom, per unit time

# of absorption events	=	$\sigma(\Delta)\phi\rho_{11}$
# of stim. emission events	=	$\sigma(\Delta)\phi\rho_{22}$

Note: Given N atoms, the total # of events are $N\rho_{11}, N\rho_{22}$. This is useful when we care about the total power in the light field, e. g., in the context of laser theory

Solution of the Rate Equations

Let $\Gamma_1 = \Gamma_2 = 0$ and plug in $\rho_{11} = 1-\rho_{22}$

$$\boxed{\begin{aligned}\dot{\rho}_{22} &= -A_{21}\rho_{22} - \sigma(\Delta)\phi\,(2\rho_{22}-1)\\ &= -\underbrace{(A_{21}+2\sigma(\Delta)\phi)}_{\gamma}\rho_{22} + \sigma(\Delta)\phi\end{aligned}}$$

This solution is a damped approach to Steady State!

Emission and Absorption – Population Rate Equations

We see that per atom, per unit time

$$\boxed{\begin{aligned} \text{\# of absorption events} &= \sigma(\Delta)\phi\rho_{11} \\ \text{\# of stim. emission events} &= \sigma(\Delta)\phi\rho_{22} \end{aligned}}$$

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Solution of the Rate Equations

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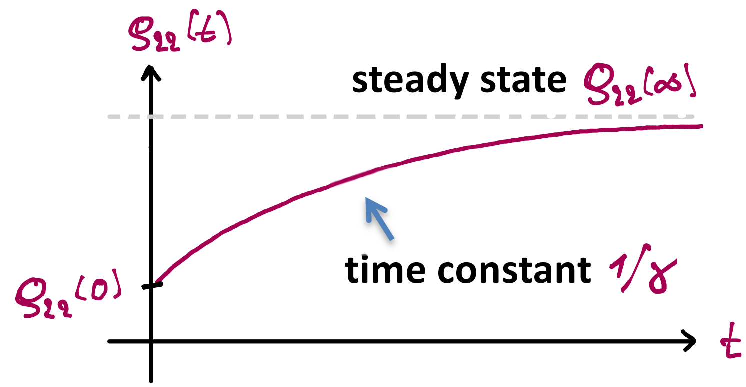
$$\begin{aligned} \dot{\rho}_{22} &= -A_{21}\rho_{22} - \sigma(\Delta)\phi\,(2\rho_{22} - 1) \\ &= -\underbrace{(A_{21} + 2\sigma(\Delta)\phi)}_{\gamma}\,\rho_{22} + \sigma(\Delta)\phi \end{aligned}$$

The solution is a damped approach to Steady State!

$$\rho_{22}(t) = [\rho_{22}(0) - \rho_{22}(\infty)]\,e^{-\gamma t} + \rho_{22}(\infty)$$

where

$$\gamma = (A_{21} + 2\sigma(\Delta)\phi)\,, \quad \rho_{22}(\infty) = \frac{\sigma(\Delta)\phi}{A_{21} + 2\sigma(\Delta)\phi}$$



(*) This transient behavior is valid in the collision broadened regime.

(*) Without collisions the transient regime Is one of damped Rabi oscillations.

(*) The steady state value $\rho_{22}(\infty)$ is good regardless

Emission and Absorption – Population Rate Equations

$$\rho_{22}(t) = \left[\rho_{22}(0) - \rho_{22}(\infty)\right] e^{-\gamma t} + \rho_{22}(\infty)$$

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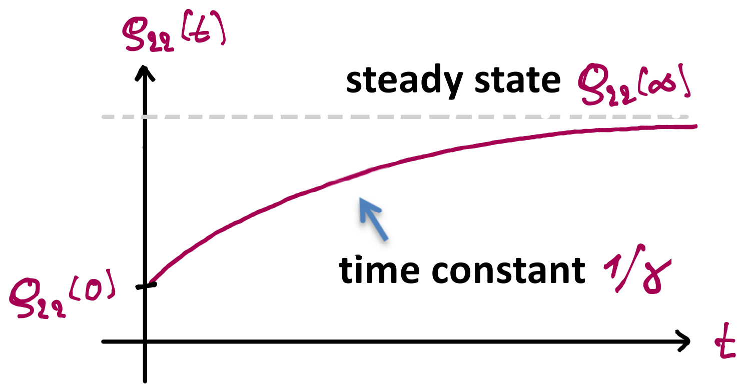

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Numerical simulation of Density Matrix Eqs (Optical Bloch Picture).

Figure from Milloni & Eberly

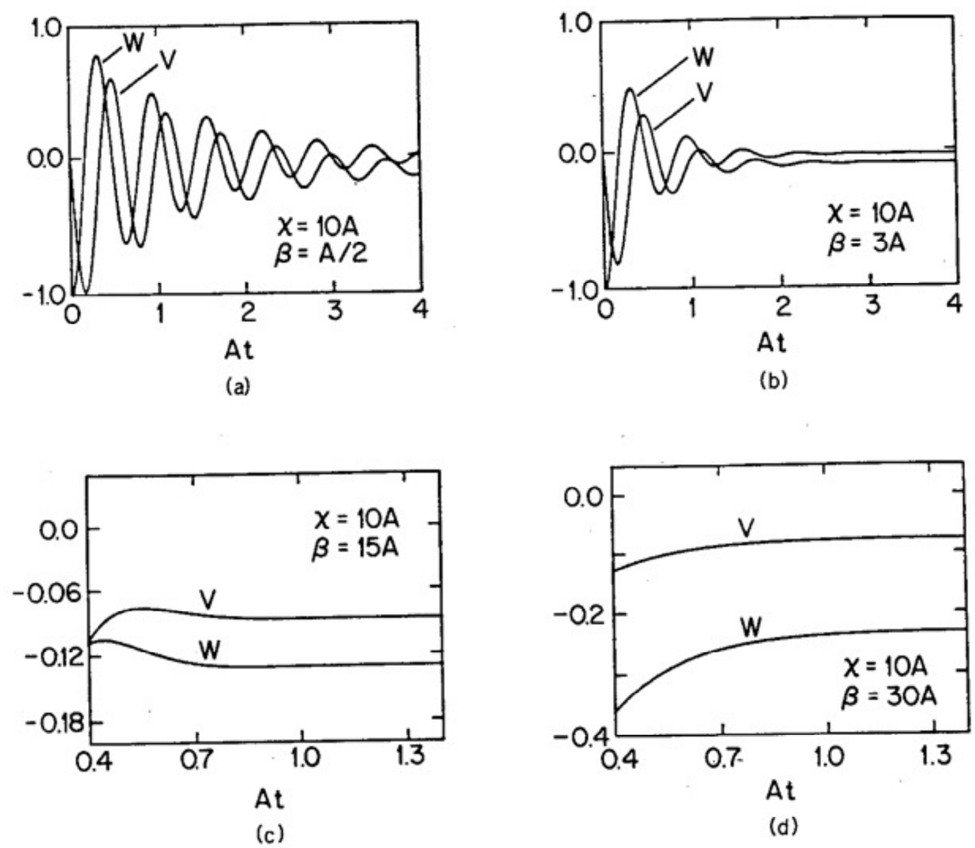

Figure 6.6 Numerical solutions of the v, w equations (6.5.21) for a range of collisional damping rates. Note scale changes.

Emission and Absorption – Population Rate Equations

$$\rho_{22}(t) = \left[\rho_{22}(0) - \rho_{22}(\infty)\right] e^{-\gamma t} + \rho_{22}(\infty)$$

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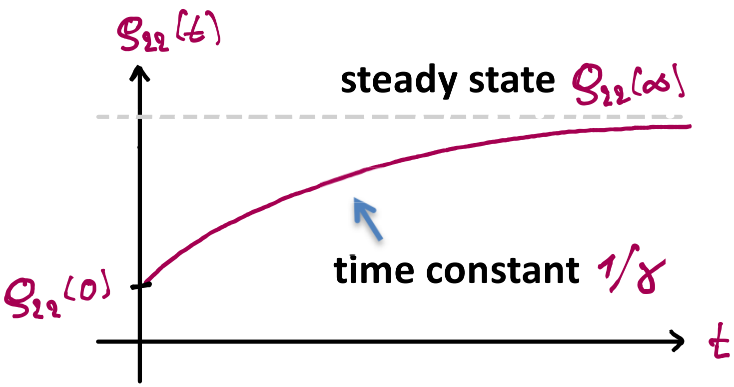

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Limiting cases:

$\sigma(\Delta)\phi = 0 \Rightarrow \rho_{22}(\infty) = 0$

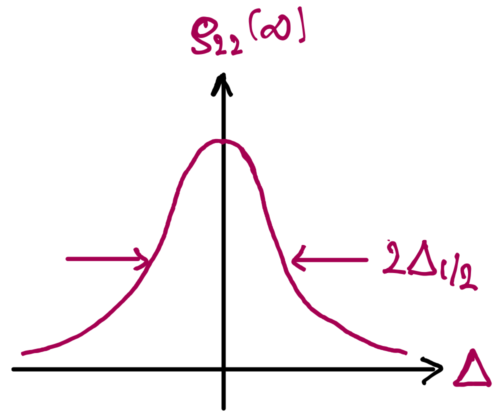
$\sigma(\Delta)\phi \ll A_{21} \Rightarrow \rho_{22}(\infty) = \dfrac{\sigma(\Delta)\phi}{A_{21}}$

$\sigma(\Delta)\phi \gg A_{21} \Rightarrow \rho_{22}(\infty) = 1/2 \leftarrow$ Saturation!

Rewrite $\rho_{22}(\infty)$ using $R_{12} = \sigma(\Delta)\phi = \dfrac{|\chi|^2\beta/2}{\Delta^2 + \beta^2}$

$$\Rightarrow \rho_{22}(\infty) = \frac{|\chi|^2\beta/2A_{21}}{\Delta^2 + \beta^2 + |\chi|^2\beta/A_{21}}$$

Plot $\rho_{22}(\infty)$ vs Δ:



HWHM line width:

$$\Delta_{1/2} = \sqrt{\beta^2 + |\chi|^2\beta/A_{21}}$$

$$= \beta\sqrt{1 + \frac{2\sigma(0)\phi}{A_{21}}}$$

$\left(\text{used } \sigma(0)\phi = \dfrac{|\chi|^2}{2\beta}\right)$

Emission and Absorption – Population Rate Equations

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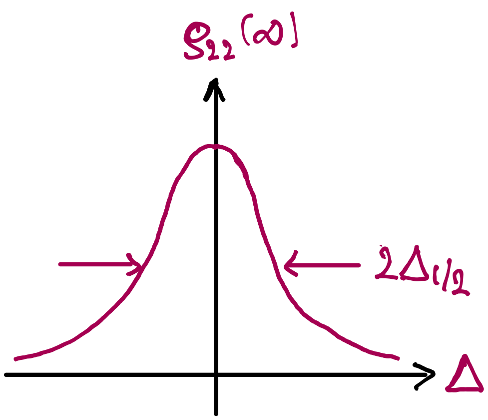


HWHM line width:

$$\Delta_{1/2} = \sqrt{\beta^2+|\chi|^2\beta/A_{21}}$$

$$= \beta\sqrt{1+\frac{2\sigma(0)\phi}{A_{21}}}$$

$$\left(\text{used } \sigma(0)\phi = \frac{|\chi|^2}{2\beta}\right)$$

Power Broadening: Rewrite

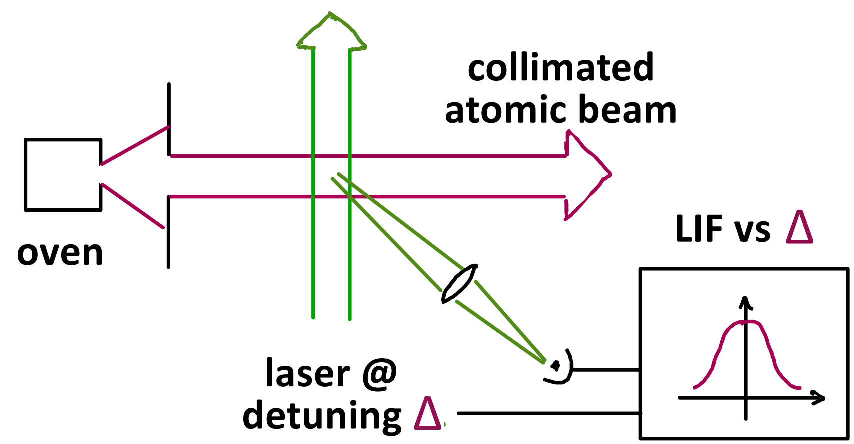
$$\Delta_{1/2} = \beta\sqrt{1+\phi/\phi_{SAT}} = \beta\sqrt{1+I/I_{SAT}}$$

where

$$\phi_{SAT} \equiv \frac{A_{21}}{2\sigma(0)}, \quad I_{SAT} \equiv \frac{\hbar\omega A_{21}}{2\sigma(0)}$$

β : natural linewidth

Power Broadening in molecular beam spectroscopy:



Keep $I \ll I_{SAT}$ for best spectroscopic resolution

Emission and Absorption – Population Rate Equations

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Power Broadening: Rewrite

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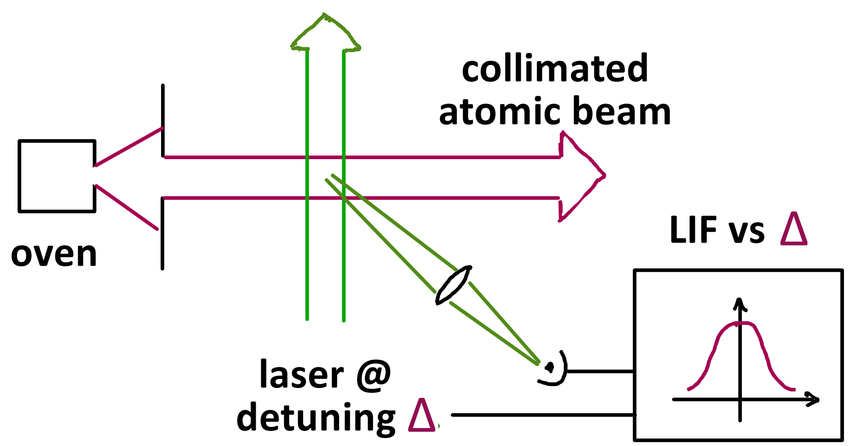

Keep $I \ll I_{SAT}$ for best spectroscopic resolution

End
02-20-2023

More about the Photon Scattering Cross Section

By Definition

$$R_{12} = \frac{|\chi|^2\beta/2}{\Delta^2+\beta^2} = \sigma(\Delta)\phi = \sigma(\Delta)\frac{\frac{1}{2}c\varepsilon_0|E_0|^2}{\hbar\omega}$$

where $|\chi|^2 = |\vec{\mu}_{12}\cdot\vec{\varepsilon}E_0/\hbar|^2 = f\,\dfrac{|\vec{\mu}_{12}|^2|E_0|^2}{\hbar^2}$

and Collision free $\rightarrow$ $\boxed{1 \geq f \geq 1/3}$ $\leftarrow$ Collision broadened

This gives us

$$\boxed{\sigma(\Delta) = f\,\frac{\omega\mu_{12}^2}{\hbar c\varepsilon_0\beta}\,\frac{\beta^2}{\Delta^2+\beta^2} \equiv \sigma(0)\,\frac{\beta^2}{\Delta^2+\beta^2}}$$

Let $\beta = A_{21}/2$, $\Gamma_1=\Gamma_2=0$ (collision free)

$$\boxed{\sigma(0) = f\,\frac{2\omega\mu_{12}^2}{\hbar c\varepsilon_0 A_{21}} = f\,\frac{4\pi}{\hbar\varepsilon_0\lambda}\,\frac{\mu_{21}^2}{A_{21}}} \quad *$$

The connection between A_{21} and μ_{21}^2 is intuitive, derived rigorously during the QED part of OPTI 544

Emission and Absorption – Population Rate Equations

Begin
02-22-2023

More about the Photon Scattering Cross Section

By Definition

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This gives us

$$\boxed{\sigma(\Delta) = f\frac{\omega\,\mathfrak{r}_{12}^2}{\hbar c\varepsilon_0\beta}\frac{\beta^2}{\Delta^2+\beta^2} \equiv \sigma(0)\frac{\beta^2}{\Delta^2+\beta^2}}$$

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The connection between A_{21} and $\mathfrak{r}_{21}^2$ is intuitive, derived rigorously during the QED part of OPTI 544

End
02-20-2023

Here we simply note the result :

$$\boxed{A_{21} = \frac{\mathfrak{r}_{12}^2\,\omega^3}{3\pi\varepsilon_0\hbar c^3}}$$

Substituting in $*$ we get

$$\sigma(0) = f\frac{2\omega}{\hbar c\varepsilon_0}\frac{3\pi\varepsilon_0\hbar c^3}{\omega^3} = f\frac{3\lambda^2}{2\pi}$$

$$\frac{3\lambda^2}{2\pi} \geq \sigma(0) \geq \frac{\lambda^2}{2\pi}$$

Collision free, polarized light

Collision broadened or un-polarized light

– Remarkably simple result – easy to remember

Why ?

Emission and Absorption – Population Rate Equations

Here we simply note the result :

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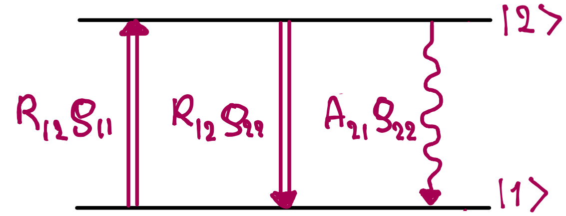
Rate Eq. Model of Absorption

Remember our Physical Picture of absorption/ stim. emission

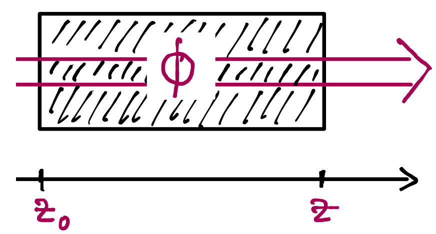


The loss over distance dz is $d\phi = \sigma(\Delta)\phi N(\rho_{22} - \rho_{11}) dz$

If $\phi \ll \phi_{sat}$ then $\rho_{11} \approx 1$ and $\rho_{22} \approx 0$ and we get

$$d\phi = -N\sigma(\Delta)\phi\, dz \;\Rightarrow\; \frac{d}{dz}\phi = -N\sigma(\Delta)\phi = -a\phi$$

Absorption:



$$\boxed{\begin{aligned} \phi(z) &= e^{-a(z-z_0)}\phi(z_0) \\ I(z) &= e^{-a(z-z_0)} I(z_0) \end{aligned}}$$

Typical $\lambda = 500\,nm$, $a = N \times 10^{-9}\,cm^2 \dfrac{\beta^2}{\Delta^2 + \beta^2}$

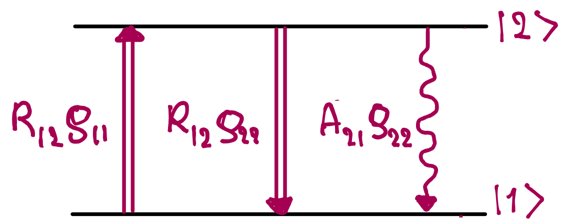
Solid: $N \sim 10^{21}/cm^3$ ⇨ totally opaque @ resonance

Gas: $N \sim 10^{8}/cm^3$ ⇨ transparent @ resonance

Emission and Absorption – Population Rate Equations

Rate Eq. Model of Absorption

Remember our Physical Picture of absorption/ stim. emission



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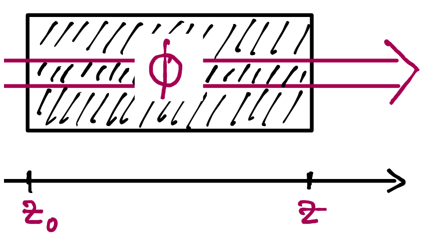

$$\boxed{\begin{aligned}\phi(z) &= e^{-a(z-z_0)}\,\phi(z_0)\\ I(z) &= e^{-a(z-z_0)}\,I(z_0)\end{aligned}}$$

Typical $\lambda = 500\,nm$, $a = N \times 10^{-9}\,cm^2 \frac{\beta^2}{\Delta^2+\beta^2}$

Solid: $N \sim 10^{21}/cm^3$ ⇒ totally opaque @ resonance

Gas: $N \sim 10^{8}/cm^3$ ⇒ transparent @ resonance

Note: This is a low saturation result !

High flux ⇒ photon scattering per atom saturates

If $\phi \gg \phi_{sat}$ then $\rho_{22}(\infty) = 1/2$ ⇒ $d\phi = N\frac{A_{21}}{2}dz$

$$\phi(z) = \phi(z_0) - \frac{NA_{21}}{2}(z-z_0)$$

At very high flux the loss is relatively insignificant

$$\boxed{\phi(z), \phi(z_0) \gg \frac{NA_{21}}{2}(z-z_0)}$$

– This is referred to as <u>bleaching</u> or <u>hole burning</u>

Emission and Absorption – Population Rate Equations

Blackbody Radiation

This is a standard problem in Optical Physics, which we review only briefly in OPTI 544. See almost any textbook for details, including Milloni & Eberly.

Step (1): We define a Lineshape Function:

$$\boxed{\sigma(\Delta) = \frac{\wp_{12}^2 \omega_{21}}{2\varepsilon_0 \hbar c} \underbrace{\frac{\delta\nu_{21}/\pi}{(\nu_{21}-\nu)^2 + \delta\nu_{21}^2}}_{S(\nu)} , \quad \delta\nu_{21} = \beta/2\pi}$$

$S(\nu)$ is sharply peaked compared to the energy density $\rho(\nu)$ of Blackbody Radiation.

⇒ we can approximate $S(\nu) = \delta(\nu_{21} - \nu)$

Step (2): Sub in the expression for transition rates and integrate over all frequencies

$$R_{12} = \int_0^\infty \sigma(\nu)\phi(\nu)\,d\nu = \frac{\wp_{21}^2 \omega_{21}}{2\varepsilon_0 \hbar} \int_0^\infty S(\nu) \frac{\rho(\nu)}{\hbar\nu}\, d\nu$$

$$\Rightarrow \quad R_{12} = \boxed{\frac{\wp_{12}^2}{2\varepsilon_0 \hbar^2}} \rho(\nu_{21}) = B\,\rho(\nu_{21})$$

← Einstein B coefficient

Step (3): 2-level atom in thermal equilibrium with Blackbody Radiation field

$$\left.\begin{aligned} \frac{\rho_{22}}{\rho_{11}} &= e^{-\hbar\nu_{21}/k_B T} \\ (A_{21}+R_{21})\rho_{22} &= R_{12}\rho_{11} \end{aligned}\right\} \Rightarrow \boxed{\rho(\nu_{21}) = \frac{A_{21}/B}{e^{\hbar\nu_{21}/k_B T} - 1}}$$

(Detailed Balance)

Step (4): Use prior result for A_{21} to find A_{21}/B which is independent of $\wp_{12}^2$

$$\boxed{\rho(\nu) = \frac{8\pi\hbar\nu^3/c^3}{e^{\hbar\nu/k_B T} - 1}}$$

Energy Density of the Blackbody Radiation field

Step (5): We can extend this to find the

- Total electromagnetic energy density
- Total radiated intensity from a black body

Emission and Absorption – Population Rate Equations

Step (3): 2-level atom in thermal equilibrium with Blackbody Radiation field

$$\left.\begin{array}{l} \dfrac{\mathcal{N}_{22}}{\mathcal{N}_{11}} = e^{-h\nu_{21}/k_B T} \\ (A_{21}+R_{21})\mathcal{N}_{22} = R_{12}\mathcal{N}_{11} \end{array}\right\} \Rightarrow \boxed{\rho(\nu_{21}) = \frac{A_{21}/B}{e^{h\nu_{21}/k_B T}-1}}$$

(Detailed Balance)

Step (4): Use prior result for A_{21} to find A_{21}/B, which is independent of r_{12}^2

$$\boxed{\rho(\nu) = \frac{8\pi h\nu^3/c^3}{e^{h\nu/k_B T}-1}}$$

Energy Density of the Blackbody Radiation field

Step (5): We can extend this to find the

- Total electromagnetic energy density
- Total radiated intensity from a black body

Step (6): Relative importance of spontaneous and stimulated emission

We have $\dfrac{A_{21}}{R_{12}} = \dfrac{A_{21}}{B\rho(\nu)} = e^{h\nu/k_B T}-1 \gg 1$

for $T <$ several $\times$ 1000K

Take the surface of the Sun, $T = 5800\text{K}$

$$\boxed{\frac{A_{21}}{R_{12}} \sim 140 \text{ @ } \lambda_{max} = 500\,\text{nm}}$$

End 02-22-2023