Begin 02-20-2024

Density Matrix Description of 2-Level Atoms

Next: Non-Hamiltonian evolution

Types of events

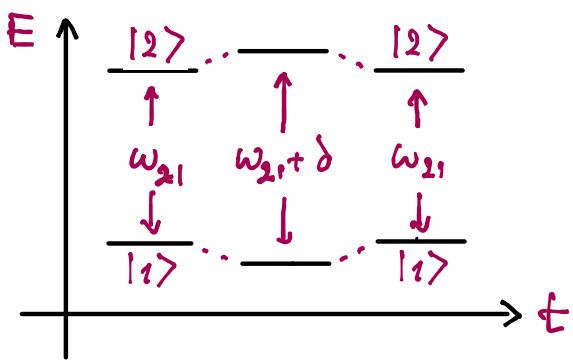
(i) Elastic collisions: No change in energy

(ii) Inelastic collisions: Atom loss

(iii) Spontaneous decay: Transition $|2\rangle \to |1\rangle$

Simple Model of Elastic Collisions

Two atoms near each other ⇒ energy levels shift, free evol. of ρ_{12} changed



(Paradigm for perturbations that do not lead to net change in energy)

Evolution of coherence (fast variables)

$$\dot{\rho}_{12} = -i\left[\omega_{21} + \delta\omega(t)\right]\rho_{12}$$

collisional history

$$\Rightarrow \rho_{12}(t) = \rho_{12}(0)\, e^{-i\omega_{21}t}\, e^{-i\int_0^t dt'\,\delta\omega(t')}$$

We need the <u>ensemble average of</u> $\rho_{12}(t)$

<u>Assumptions</u>:

- From atom to atom $\delta\omega(t)$ is a Gaussian Random Variable
- Averaged over the ensemble $\langle \delta\omega(t)\rangle_{\mathcal{E}} = 0$
- Collisions have no memory over time,

$$\langle \delta\omega(t)\,\delta\omega(t')\rangle_t = \frac{2}{\tau}\,\delta(t-t')$$

Can show that, averaged over time and the ensemble

$$\left\langle e^{-i\int_0^t dt'\,\delta\omega(t')}\right\rangle_{\mathcal{E}} = e^{-t/\tau}$$

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Can show that, averaged over time and the ensemble

$$\left\langle e^{-i\int_0^t dt'\,\delta\omega(t')} \right\rangle_{\mathcal{E}} = e^{-t/\tau}$$

It follows that: $\rho_{12}(t) = \rho_{12}(0)\, e^{-i\omega_{21}t}\, e^{-t/\tau}$

Add this decay to the equation of motion to get

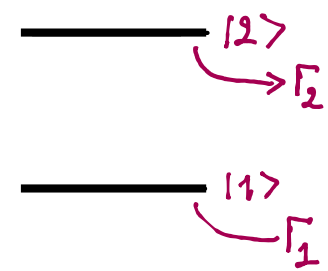
$$\boxed{\dot{\rho}_{12} = (\dot{\rho}_{12})_{S.E.} + (\dot{\rho}_{12})_{E.C.} = -(i\omega_{21} - 1/\tau)\rho_{12}}$$

Simple Model of Inelastic Collisions

As modeled by, e. g., Milloni & Eberly, this is a steady loss of atoms

$$\dot{\rho}_{11} = (\dot{\rho}_{11})_{S.E.} - \Gamma_1 \rho_{11}$$

$$\dot{\rho}_{22} = (\dot{\rho}_{22})_{S.E.} - \Gamma_2 \rho_{22}$$



This is strange because $\mathrm{Tr}\,\rho(t)$ is not preserved

Convenient when working with quantities

$$N\langle \vec{\mu} \rangle \propto N(\vec{\mu}_{12}\rho_{21} + \vec{\mu}_{21}\rho_{12})$$

Density Matrix Description of 2-Level Atoms

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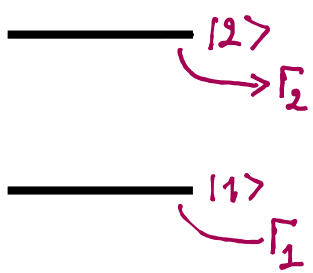

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Effect on probability amplitudes

Populations are ensemble averages of the type

$$\rho_{11}(t) = \langle |a_1(t)|^2\rangle = \langle |a_1(0)|^2\rangle e^{-\Gamma_1 t}$$

$$\rho_{22}(t) = \langle |a_2(t)|^2\rangle = \langle |a_2(0)|^2\rangle e^{-\Gamma_2 t}$$

When the populations decay, the averages of the probability amplitudes must decay accordingly,

$$\langle |a_1(t)|\rangle = \langle |a_1(0)|\rangle e^{-\Gamma_1/2\, t}$$

$$\langle |a_2(t)|\rangle = \langle |a_2(t)|\rangle e^{-\Gamma_2/2\, t}$$

Thus, for the coherences

$$\rho_{12}(t) = \langle a_1(t) a_2(t)^*\rangle = \langle a_1(0) a_2(0)^*\rangle e^{-\Gamma_1/2\, t} e^{-\Gamma_2/2\, t}$$

This gives us

elastic inelastic

$$\boxed{\dot{\rho}_{12} = (\dot{\rho}_{22})_{S.E.} - 1/\tau\, \rho_{12} - \frac{\Gamma_1 + \Gamma_2}{2}\rho_{12}}$$

Density Matrix Description of 2-Level Atoms

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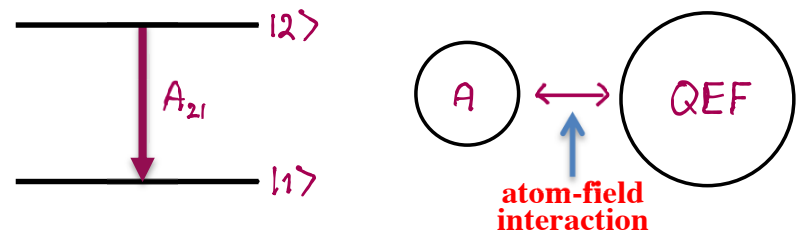
Spontaneous Decay

This process occurs due to interaction between the Atom and the Quantized Electromagnetic Field



Warm-up: A Bayesian recipe for Mixed States

Alice has 2-level atoms A & B initially in state $|2\rangle$

<u>Step (1)</u> She applies a Hamiltonian that drives the evolution

$$|2\rangle_A |2\rangle_B \rightarrow a_2 |2\rangle_A |2\rangle_B + a_1 |1\rangle_A |1\rangle_B$$

<u>Step (2)</u> She gives atom B to Bob and asks him to measure if it is in $|1\rangle_B$ or $|2\rangle_B$ and keep the result secret forever.

<u>Result</u>: Alice now has a 2-level atom in the state

$$\rho = |a_2|^2 |1\rangle_{AA}\langle 1| + |a_1|^2 |2\rangle_{AA}\langle 2|$$

Note that as long as $|a_1|^2 > 0$, this decreases the prob. that Alice's atom is in state $|2\rangle_A$. Repeating these steps will thus cause a gradual, irreversible decrease of $|a_2|^2$, i. e., a decay of the excited state population of her atom

Density Matrix Description of 2-Level Atoms

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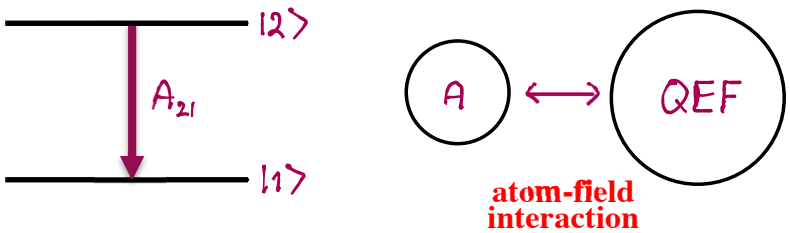

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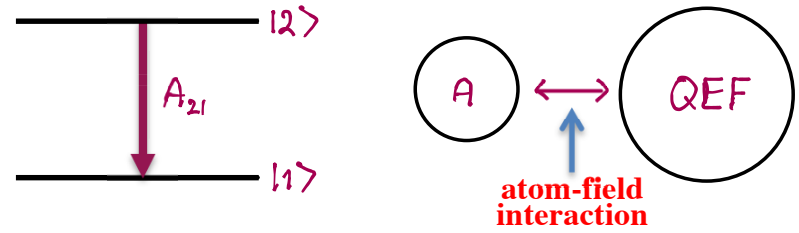


Final OPTI 544 Lectures:

$|\psi(0)\rangle = |2\rangle_A |vac\rangle_{QEF}$ ⇨ **evolution over time *t***

$|\psi(t)\rangle = C_{2,0}(t)|2\rangle_A|vac\rangle_{QEF}$ ⇨ $\sum_k C_{1,1k}(t)|1\rangle_A|n_k=1\rangle_{QEF}$

photon "in the atom" **photon in field mode *k***

Cannot recover info in continuum of field modes

Probability $|C_{2,0}(t)|^2$ of having no decay

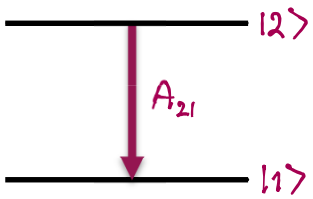
Probability $\sum_k |C_{1,1k}(t)|^2$ of having decay

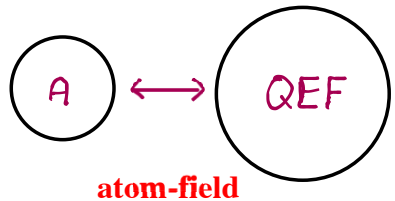
<u>No Coherence</u> established between states $|1\rangle, |2\rangle$

Density Matrix Description of 2-Level Atoms

Spontaneous Decay

This process occurs due to interaction between the Atom and the Quantized Electromagnetic Field





atom-field interaction

Final OPTI 544 Lectures:

$|\psi(0)\rangle = |2\rangle_A |Vac\rangle_{QEF}$ ⇨ evolution over time *t*

$|\psi(t)\rangle = C_{2,0}(t)|2\rangle_A |Vac\rangle_{QEF}$ ⇨ $\sum_k C_{1,1k}(t)|1\rangle_A |n_k=1\rangle_{QEF}$

photon "in the atom" — photon in field mode *k*

Cannot recover info in continuum of field modes

Probability $|C_{2,0}(t)|^2$ **of having no decay**

Probability $\sum_k |C_{1,1k}(t)|^2$ **of having decay**

No Coherence established between states $|1\rangle, |2\rangle$

Conclusion: Decay moves population $|2\rangle \to |1\rangle$ **at rate** A_{21}**, damps coherence at rate** $A_{21}/2$

$$\dot{\rho}_{11} = A_{21}\rho_{22}, \quad \dot{\rho}_{22} = -A_{21}\rho_{11}$$

$$\dot{\rho}_{12} = -\frac{A_{21}}{2}\rho_{12} = \dot{\rho}_{21}^*$$

Putting it all together:

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

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

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

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

These are our desired **Density Matrix Equations of Motion**

Emission and Absorption – Population Rate Equations

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

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

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

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

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 $\dot{\rho}_{12} = 0$ ⇒

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

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

Plug into eqs for populations to get

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

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

From these eqs. we can find <u>steady state values</u> for the populations and coherences in terms of X, Δ, A_{21}, β 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 $\dot{\rho}_{12} = 0$ ⇒

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

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

Plug into eqs for populations to get

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

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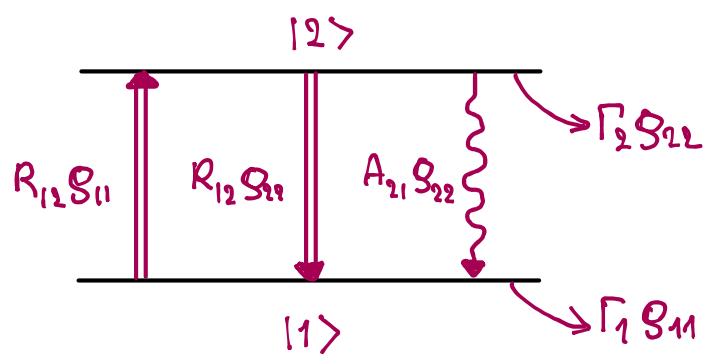
<u>Note</u>: The terms remaining after elimination of ρ_{12}, ρ_{21} **are commonly identified with <u>induced</u> or <u>stimulated</u> processes. They are proportional to** $|\chi|^2, |E_0|^2$ **and thus the <u>intensity</u> of the light field.**

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

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

Schematic:



Emission and Absorption – Population Rate Equations

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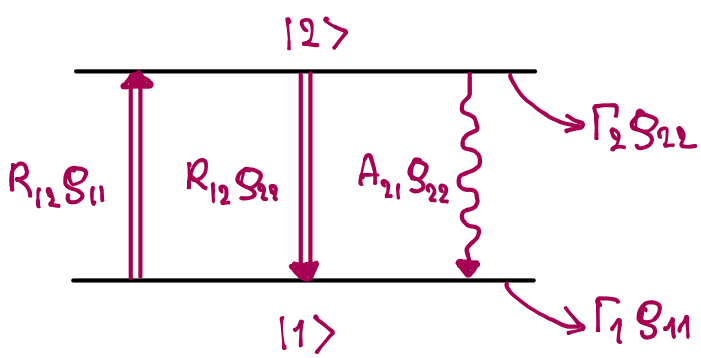

Elastic Collision Broadening

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 collision broadened 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 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}

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Emission and Absorption – Population Rate Equations

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

$$\langle |\vec{p}_{12}\cdot\vec{\varepsilon}\, E_0|^2\rangle_{angles} = \frac{1}{3} p_{12}^2 |E_0|^2 \Rightarrow$$

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

Photon Flux and Cross Section

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

"photon flux"

This allows us to recast the Rate Eqs

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

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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!

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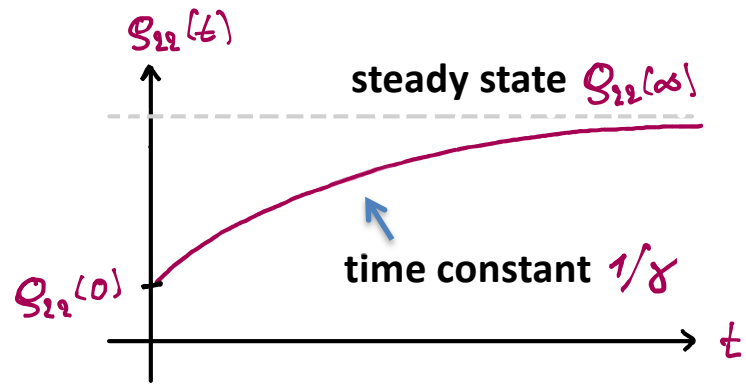
$$\rho_{22}(t) = [\rho_{22}(0) - \rho_{22}(\infty)]\,e^{-\gamma t} + \rho_{22}(\infty)$$

where

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



(*) This <u>transient</u> behavior is valid in the collision broadened regime.

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

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

Emission and Absorption – Population Rate Equations

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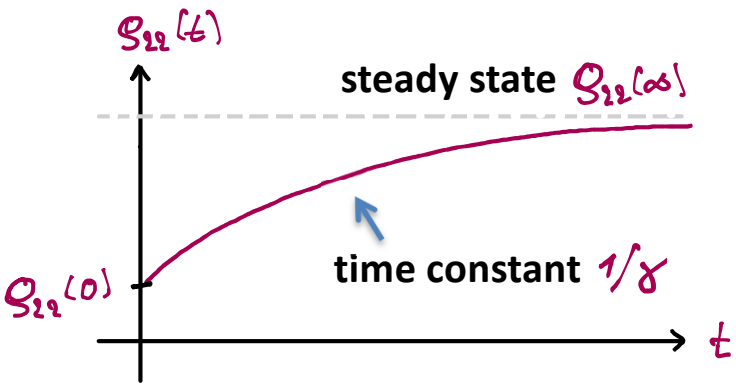
where

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



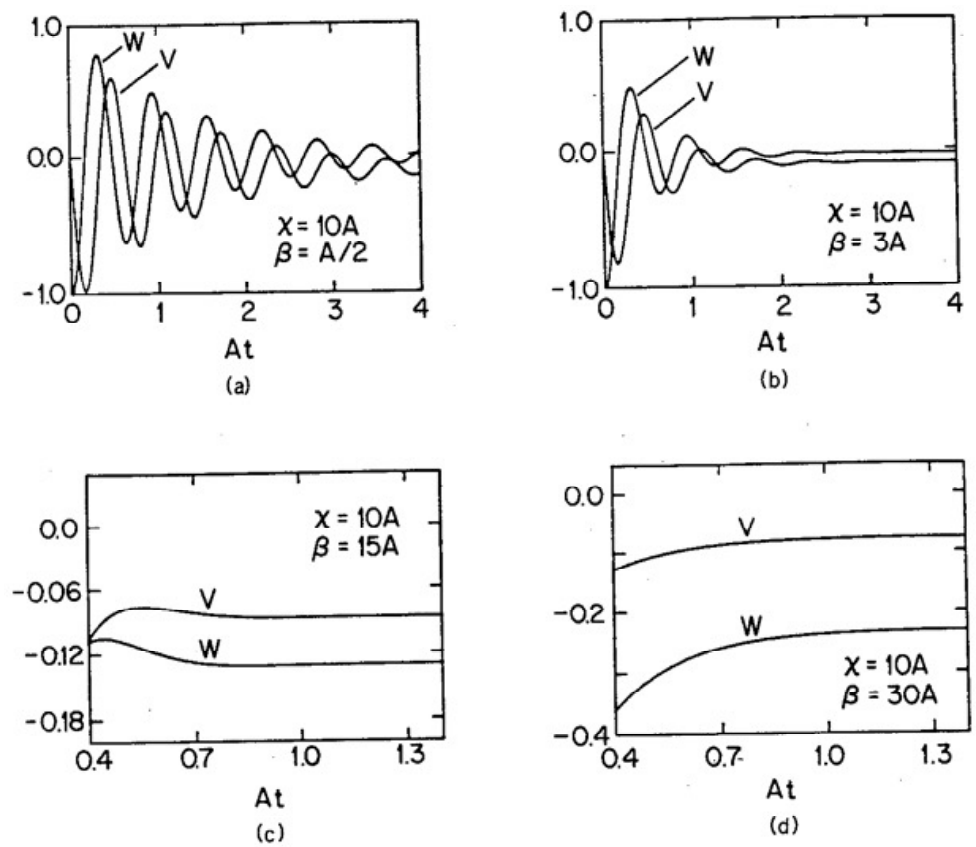
(*) This <u>transient</u> behavior is valid in the collision broadened regime.

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

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

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

Figure from Milloni & Eberly

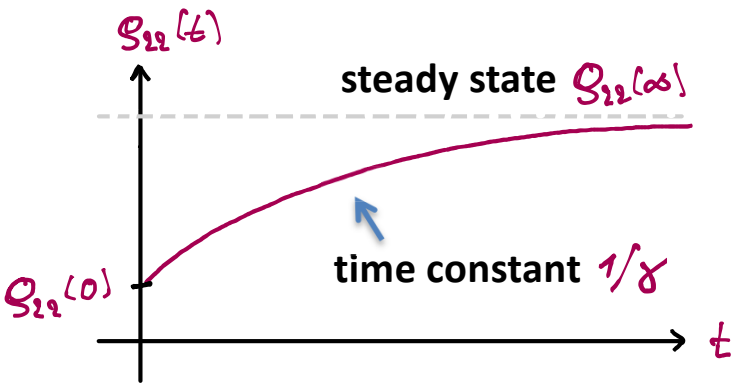


Emission and Absorption – Population Rate Equations

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

where

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



(*) This <u>transient</u> behavior is valid in the collision broadened regime.

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

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

Limiting cases:

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

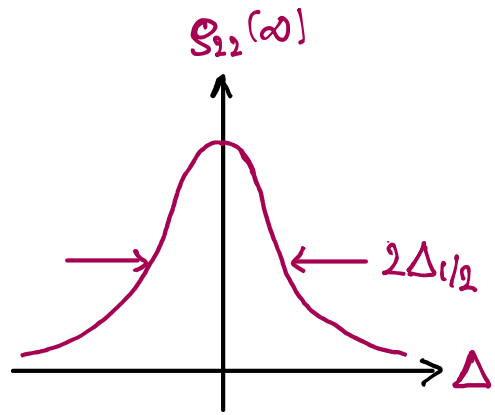
$\sigma(\Delta)\phi \ll A_{21} \Rightarrow \rho_{22}(\infty) = \dfrac{\sigma(\Delta)\phi}{A_{21}}$

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

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

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

Plot $\rho_{22}(\infty)$ vs Δ:



HWHM line width:

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

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

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

Power Broadening!

Emission and Absorption – Population Rate Equations

Limiting cases:

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

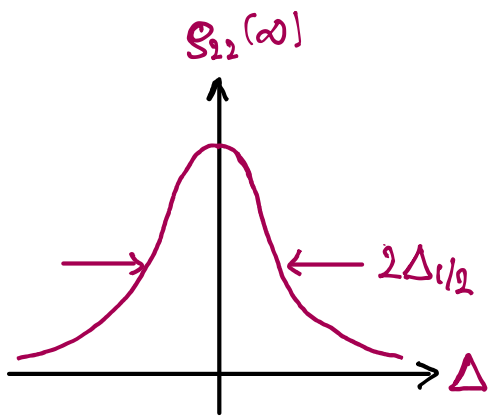
$$\sigma(\Delta)\phi \ll A_{21} \;\Rightarrow\; \rho_{22}(\infty) = \frac{\sigma(\Delta)\phi}{A_{21}}$$

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

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

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

Plot $\rho_{22}(\infty)$ **vs** Δ**:**



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

Power Broadening: Rewrite

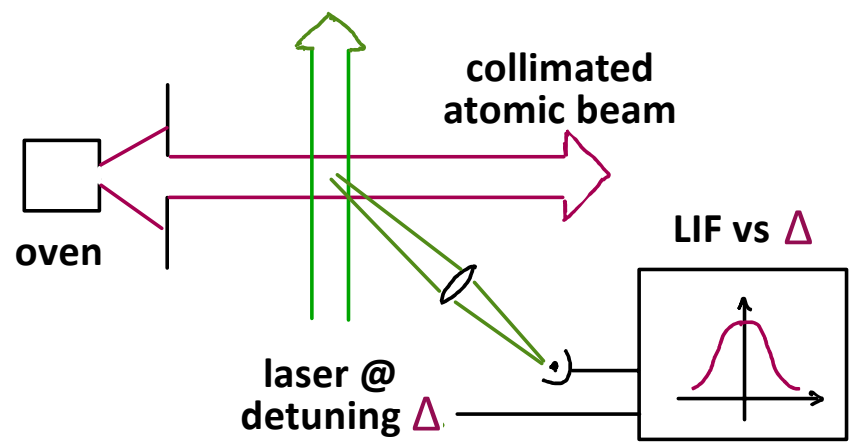
$$\Delta_{1/2} = \beta\sqrt{1+\phi/\phi_{SAT}} = \beta\sqrt{1+I/I_{SAT}}$$

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

Power Broadening: **Rewrite**

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

where

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

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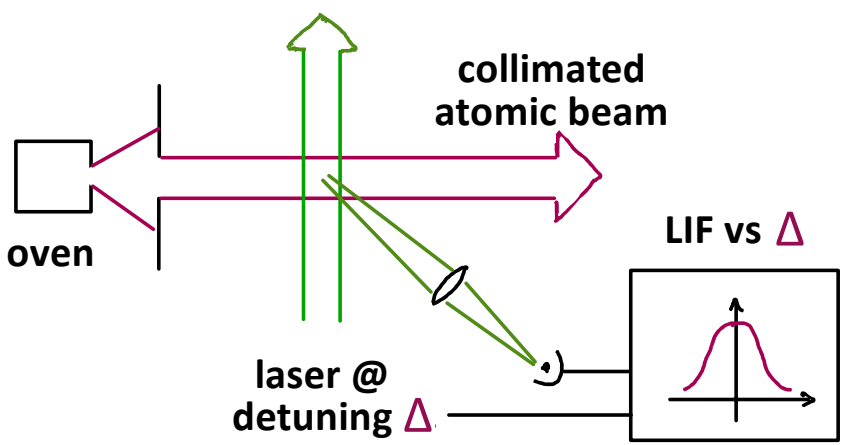
Power Broadening in molecular beam spectroscopy:



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

End 02-20-2024