# Atoms and photons <br> Chapter 2 

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## Outline

(1) Interaction Hamiltonian

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(2) Non-resonant interaction: perturbative approach

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(3) Classical field and free atom

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4 Atomic relaxation

## Outline

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(3) Classical field and free atom
(4) Atomic relaxation
(5) Optical Bloch equations

## Outline

(1) Interaction Hamiltonian
(2) Non-resonant interaction: perturbative approach
(3) Classical field and free atom
(4) Atomic relaxation
(5) Optical Bloch equations
(6) Applications

## Interaction Hamiltonian

We consider a single electron atom (Hydrogen). The free Hamiltonian is:

$$
\begin{equation*}
H_{0}=\frac{P^{2}}{2 m}+q U(\mathbf{R}) \tag{1}
\end{equation*}
$$

$\mathbf{P}$ and $\mathbf{R}$ : momentum and position operators. Eigenstates $H_{0}|i\rangle=E_{i}|i\rangle$, ground state $|g\rangle$
Atom in a radiation field (potential vector $\mathbf{A}(\mathbf{r}, t)$, scalar potential $\mathbf{V}(\mathbf{r}, t)$ ):

$$
\begin{equation*}
H=\frac{1}{2 m}(\mathbf{P}-q \mathbf{A}(\mathbf{R}, t))^{2}+q U(\mathbf{R})+q V(\mathbf{R}) \tag{2}
\end{equation*}
$$

Note that $\mathbf{A}(\mathbf{R}, t)$ is an operator in the electron's Hilbert space.

## Interaction Hamiltonian

Gauge choice

Gauge transformation

$$
\begin{align*}
& \mathbf{A}^{\prime} \rightarrow \mathbf{A}=\mathbf{A}^{\prime}+\nabla \chi(\mathbf{r}, t) \\
& V^{\prime} \rightarrow V=V^{\prime}-\frac{\partial \chi}{\partial t} \tag{3}
\end{align*}
$$

where $\chi$ is an arbitrary function of space and time.

Coulomb gauge

$$
\begin{equation*}
\boldsymbol{\nabla} \cdot \mathbf{A}=0 \tag{4}
\end{equation*}
$$

## Interaction Hamiltonian

## Fourier space

## Space-time Fourier transform

$$
\begin{equation*}
\mathbf{A}(\mathbf{r}, t)=\frac{1}{4 \pi^{2}} \int \mathcal{A}(\mathbf{k}, \omega) e^{i(\mathbf{k} \cdot \mathbf{r}-\omega t)} d \mathbf{k} d \omega \tag{5}
\end{equation*}
$$

Longitudinal and transverse potentials w.r.t. k:

$$
\begin{equation*}
\mathcal{A}(\mathbf{k}, \omega)=\mathcal{A}_{\|}+\mathcal{A}_{\perp} \tag{6}
\end{equation*}
$$

Hence:

$$
\begin{equation*}
\mathbf{A}(\mathbf{k}, \omega)=\mathbf{A}_{\|}+\mathbf{A}_{\perp} \tag{7}
\end{equation*}
$$

Space-time Fourier transform of $\boldsymbol{\nabla} \cdot \mathbf{A}: i \mathbf{k} \cdot \mathcal{A}$. Coulomb:

$$
\begin{equation*}
\mathcal{A}_{\|}=\mathbf{A}_{\|}=0 \tag{8}
\end{equation*}
$$

## Interaction Hamiltonian

## Fourier space

Same decomposition for fields. Transverse electric field, since divergence-free as $\mathbf{A}$ in the Coulomb gauge. With

$$
\begin{equation*}
\mathbf{E}=-\frac{\partial \mathbf{A}}{\partial t}-\nabla V \tag{9}
\end{equation*}
$$

and the fact that $\nabla V$ is longitudinal (proportional to $\mathbf{k}$ in Fourier space)

$$
\begin{equation*}
\nabla V=0 \tag{10}
\end{equation*}
$$

and (no physical effect of a constant potential)

$$
\begin{equation*}
V=0 \tag{11}
\end{equation*}
$$

## Interaction Hamiltonian

A. P interaction

Expansion of $(\mathbf{P}-q \mathbf{A}(\mathbf{R}, t))^{2}$ taking care of the commutation of $\mathbf{P}$ with A. Noting:

$$
\begin{gather*}
{\left[P_{i}, f(\mathbf{R})\right]=-i \hbar \frac{\partial f}{\partial R_{i}} \quad i \in\{x, y, z\}}  \tag{12}\\
\sum_{i}\left[P_{i}, A_{i}\right]=-i \hbar \sum_{i} \frac{\partial A_{i}}{\partial R_{i}}=-i \hbar \boldsymbol{\nabla} \cdot \mathbf{A}=0  \tag{13}\\
\mathbf{P} \cdot \mathbf{A}=\sum_{i} P_{i} A_{i}=\sum_{i} A_{i} P_{i}=\mathbf{A} \cdot \mathbf{P} \tag{14}
\end{gather*}
$$

And finally

$$
\begin{equation*}
H=\frac{P^{2}}{2 m}+q U(\mathbf{R})-\frac{q}{m} \mathbf{P} \cdot \mathbf{A}+\frac{q^{2}}{2 m} \mathbf{A} \cdot \mathbf{A} \tag{15}
\end{equation*}
$$

Weak fields (much lower than atomic field unit, $10^{11} \mathrm{~V} / \mathrm{m}$ ), A.A quadratic term negligible compared to first order contribution.

$$
\begin{equation*}
H=H_{0}-\frac{q}{m} \mathbf{P} \cdot \mathbf{A}(\mathbf{R}, t) \tag{16}
\end{equation*}
$$

## Interaction Hamiltonian

$\mathbf{A} \cdot \mathbf{P}$ interaction: dipole approximation

- Radiation wavelength: about $1 \mu \mathrm{~m}$
- Atomic size: about 100 pm
- Neglect spatial variation of the vector potential across atomic orbit: $\mathbf{A}(\mathbf{R}, t)=\mathbf{A}(0, t)$

$$
\begin{equation*}
H=H_{0}-\frac{q}{m} \mathbf{P} \cdot \mathbf{A}(0, t), \tag{17}
\end{equation*}
$$

Useful, but not the intuitive form for the interaction of a dipole with a field.

## Interaction Hamiltonian

D.E interaction

Cast the interaction Hamiltonian in the more familiar form -d.E (interaction energy of a dipole with a field, manifestly independent of the gauge choice). Two possible (and equivalent) approaches
(1) The Göppert-Mayer transformation
(2) Unitary transformation on the Hilbert space

## Interaction Hamiltonian

## The Göppert-Mayer transformation

Restart from full Hamiltonian

$$
\begin{equation*}
H=\frac{1}{2 m}(\mathbf{P}-q \mathbf{A}(\mathbf{R}, t))^{2}+q U(\mathbf{R})+q V(\mathbf{R}) \tag{18}
\end{equation*}
$$

and perform dipole approximation first. For the vector potential

$$
\begin{equation*}
\mathbf{A}(\mathbf{R}, t)=\mathbf{A}(0, t) \tag{19}
\end{equation*}
$$

and (keeping first order)

$$
\begin{equation*}
V=V(0, t)+\mathbf{R} \cdot \nabla V(0, t) \tag{20}
\end{equation*}
$$

The space-independent term in $V$ has no effect

$$
\begin{equation*}
H=H_{0}-\frac{q}{m} \mathbf{P} \cdot \mathbf{A}(0, t)+\mathbf{D} \cdot \nabla V \tag{21}
\end{equation*}
$$

with

$$
\begin{equation*}
\mathbf{D}=q \mathbf{R} \tag{22}
\end{equation*}
$$

## Interaction Hamiltonian

The Göppert-Mayer transformation
Perform a gauge transformation:

$$
\begin{align*}
& \mathbf{A} \rightarrow \mathbf{A}^{\prime}=\mathbf{A}+\nabla \chi(\mathbf{r}, t) \\
& V \rightarrow V^{\prime}=V-\frac{\partial \chi}{\partial t} \tag{23}
\end{align*}
$$

and choose

$$
\begin{equation*}
\chi(\mathbf{r}, t)=-\mathbf{r} \cdot \mathbf{A}(0, t) \tag{24}
\end{equation*}
$$

so that $\mathbf{A}^{\prime}(0, t)=0$. Then

$$
\begin{gather*}
V^{\prime}=V+\mathbf{r} \cdot \frac{\partial \mathbf{A}(0, t)}{\partial t}  \tag{25}\\
\nabla V^{\prime}(0)=\nabla V(0)+\frac{\partial \mathbf{A}(0, t)}{\partial t}=-\mathbf{E}(0)  \tag{26}\\
H=H_{0}-\mathbf{D} \cdot \mathbf{E}(0) \tag{27}
\end{gather*}
$$

## Interaction Hamiltonian

Unitary transform approach
Restart from full Hamiltonian

$$
\begin{equation*}
H=\frac{1}{2 m}(\mathbf{P}-q \mathbf{A}(\mathbf{R}, t))^{2}+q U(\mathbf{R})+q V(\mathbf{R}) \tag{28}
\end{equation*}
$$

Switch to Coulomb gauge (no $V$ contribution left) and perform dipole approximation $\mathbf{A}(\mathbf{r}, t)=\mathbf{A}(0, t)$

$$
\begin{equation*}
H=\frac{1}{2 m}(\mathbf{P}-q \mathbf{A}(0, t))^{2}+q U(\mathbf{R}) \tag{29}
\end{equation*}
$$

Unitary transform $|\Psi\rangle \rightarrow|\widetilde{\Psi}\rangle=T|\Psi\rangle\left(T^{\dagger} T=\mathbb{1}\right)$. Transformed Hamiltonian

$$
\begin{equation*}
\widetilde{H}=T H T^{\dagger}+i \hbar \frac{d T}{d t} T^{\dagger} \tag{30}
\end{equation*}
$$

## Interaction Hamiltonian

Unitary transform approach

Choose $T$ as a time-dependent translation of the momentum:

$$
\begin{gather*}
T \mathbf{P} T^{\dagger}=\mathbf{P}+q \mathbf{A}(0, t)  \tag{31}\\
T=e^{-\frac{i}{\hbar} q \mathbf{R} \cdot \mathbf{A}(0, t)}=e^{-\frac{i}{\hbar} \mathbf{D} \cdot \mathbf{A}(0, t)} \tag{32}
\end{gather*}
$$

Hence

$$
\begin{equation*}
T(\mathbf{P}-q \mathbf{A}(0, t))^{2} T^{\dagger}=\mathbf{P}^{2} \tag{33}
\end{equation*}
$$

and

$$
\begin{equation*}
i \hbar \frac{d T}{d t} T^{\dagger}=\mathbf{D} \cdot \frac{d \mathbf{A}(0, T)}{d t}=-\mathbf{D} \cdot \mathbf{E}(0, t) \tag{34}
\end{equation*}
$$

Finally,

$$
\begin{equation*}
\widetilde{H}=H_{0}-\mathbf{D} \cdot \mathbf{E}(0) \tag{35}
\end{equation*}
$$

## Interaction Hamiltonian

Unitary transform approach

We get a transformed Hamiltonian in the D • E form, with a linear atom-field coupling.
We have not performed the weak field approximation to remove the $\mathbf{A} \cdot \mathbf{A}$ term in the Hamiltonian. Where is the magic?
The observables of the electron should be changed

$$
\begin{equation*}
O \rightarrow T^{\dagger} \tag{36}
\end{equation*}
$$

and this change contains non linear terms in $\mathbf{A}$. It is only for weak fields that these terms can be neglected.

## Non-resonant interaction

A simple situation:

- An atom initially in the ground state
- A weak non-resonant field so that the atom is always nearly in its ground state
- A perturbative solution to the Schrödinger equation Recover, mutatis mutantis, all the results of the previous chapter with the harmonically bound electron model


## Non-resonant interaction

Model

Incoming plane wave:

$$
\begin{equation*}
\mathbf{E}(0, t)=E_{0} \mathbf{u}_{z} \cos \omega t \tag{37}
\end{equation*}
$$

Hamiltonian

$$
\begin{equation*}
H=H_{0}+H_{1} \tag{38}
\end{equation*}
$$

with

$$
\begin{equation*}
H_{1}=-q Z E_{0} \cos \omega t \tag{39}
\end{equation*}
$$

Interaction representation w.r.t. $H_{0}$

$$
\begin{equation*}
\widetilde{H}=U_{0}^{\dagger} H_{1} U_{0} \quad \text { with } \quad U_{0}=\exp \left(-i H_{0} t / \hbar\right) \tag{40}
\end{equation*}
$$

## Non-resonant interaction

Model
Expansion of the wave function over the eigenstates of $H_{0}$ :

$$
\begin{equation*}
|\widetilde{\Psi}\rangle=\sum_{j} \beta_{j}|j\rangle \tag{41}
\end{equation*}
$$

Injection in the Schrödinger equation and scalar product with $\langle k|$

$$
\begin{equation*}
i \hbar \frac{d \beta_{k}}{d t}=\sum_{j}\langle k| U_{0}^{\dagger} H_{1} U_{0}|j\rangle \beta_{j} \tag{42}
\end{equation*}
$$

With $U_{0}|j\rangle=\exp \left(-i \omega_{j} t\right)|j\rangle, \omega_{j}=E_{j} / \hbar$ and $\omega_{k j}=\omega_{k}-\omega_{j}$ (Bohr frequency)

$$
\begin{equation*}
\frac{d \beta_{k}}{d t}=-\frac{q E_{0}}{i \hbar} \sum_{j} e^{i \omega_{k j} t}\langle k| Z|j\rangle \beta_{j} \cos \omega t \tag{43}
\end{equation*}
$$

Set of coupled first-order differential equations

## Non-resonant interaction

## Perturbative solution

Weak, non-resonant field. The atom is nearly in its ground state. Replace $\beta_{g}$ by one (and all others by zero) in the r.h.s of the system

$$
\begin{equation*}
\frac{d \beta_{k}}{d t} \approx-\frac{q E_{0}}{i \hbar} e^{i \omega_{k g} t}\langle k| Z|g\rangle \cos \omega t \tag{44}
\end{equation*}
$$

with the explicit solution

$$
\begin{equation*}
\beta_{k}(t)=\frac{q E_{0}}{2 \hbar}\langle k| Z|g\rangle\left[\frac{e^{i\left(\omega_{k g}+\omega\right) t}-1}{\omega_{k g}+\omega}+\frac{e^{i\left(\omega_{k g}-\omega\right) t}-1}{\omega_{k g}-\omega}\right] \tag{45}
\end{equation*}
$$

Resonances (and divergences) as expected at $\omega= \pm \omega_{k g}$ when $\langle k| Z|g\rangle$ does not vanish (selection rules). To compute the dipole, we return to the initial representation

$$
\begin{equation*}
|\Psi\rangle=\sum_{k} \beta_{k} e^{-i \omega_{k} t}|k\rangle \tag{46}
\end{equation*}
$$

## Non-resonant interaction

Comparison with classical model

Average dipole $\mathbf{D}=q Z \mathbf{u}_{z}=D \mathbf{u}_{z}$ (to be compared with the classical dipole)

$$
\begin{equation*}
\langle D\rangle=\sum_{\ell, k} \beta_{\ell}^{*} \beta_{k} e^{-i \omega_{k \ell} t}\langle\ell| q Z|k\rangle \tag{47}
\end{equation*}
$$

Keeping only the first-order terms in the small $\beta_{k}, k \neq g$, amplitudes

$$
\begin{gather*}
\langle D\rangle=\sum_{k} \beta_{k} e^{-i \omega_{k g} t}\langle g| q Z|k\rangle+\text { c.c. } \\
\left.\langle D\rangle=\frac{q^{2} E_{0}}{2 \hbar} \sum_{k}|\langle g| Z| k\right\rangle\left.\right|^{2}\left[\frac{e^{i \omega t}-e^{-i \omega_{k g} t}}{\omega_{k g}+\omega}+\frac{e^{-i \omega t}-e^{-i \omega_{k g} t}}{\omega_{k g}-\omega}+\text { c.c. }\right] \tag{49}
\end{gather*}
$$

## Non-resonant interaction

Comparison with classical model
Dipole contains terms oscillating permanently at the Bohr frequencies. They are an artifact of the model (transients damped in a more realistic model)

$$
\begin{equation*}
\left.\langle D\rangle=\frac{q^{2} E_{0}}{2 \hbar} \sum_{k}|\langle g| Z| k\right\rangle\left.\right|^{2}\left[\frac{e^{i \omega t}}{\omega_{k g}+\omega}+\frac{e^{-i \omega t}}{\omega_{k g}-\omega}+\text { c.c. }\right] \tag{50}
\end{equation*}
$$

Real quantum polarizability:

$$
\begin{gather*}
\langle D\rangle=\epsilon_{0} \alpha_{Q}(\omega) E_{0} \cos \omega t  \tag{51}\\
\left.\alpha_{Q}(\omega)=\frac{2 q^{2}}{\hbar \epsilon_{0}} \sum_{k}|\langle g| Z| k\right\rangle\left.\right|^{2} \frac{\omega_{k g}}{\omega_{k g}^{2}-\omega^{2}} \tag{52}
\end{gather*}
$$

## Non-resonant interaction

Comparison with classical model

Classical polarizability ( $\omega_{0}$ : resonance frequency)

$$
\begin{equation*}
\alpha_{c}\left(\omega, \omega_{0}\right)=\frac{q^{2}}{m \epsilon_{0}} \frac{1}{\omega_{0}^{2}-\omega^{2}} \tag{53}
\end{equation*}
$$

Hence

$$
\begin{equation*}
\alpha_{Q}(\omega)=\sum_{k} f_{k g} \alpha_{c}\left(\omega, \omega_{k g}\right) \tag{54}
\end{equation*}
$$

with

$$
\begin{equation*}
\left.f_{k g}=\frac{2 m \omega_{k g}}{\hbar}|\langle g| Z| k\right\rangle\left.\right|^{2} \tag{55}
\end{equation*}
$$

being the (real) oscillator strength

## Non-resonant interaction

Oscillator strength sum rule

Rewrite

$$
\begin{equation*}
f_{k g}=\frac{2 m \omega_{k g}}{\hbar}\langle g| Z|k\rangle\langle k| Z|g\rangle \tag{56}
\end{equation*}
$$

Noting

$$
\begin{gather*}
{\left[Z, H_{0}\right]=\frac{i \hbar}{m} P_{z}}  \tag{57}\\
\langle k| P_{z}|g\rangle=\frac{m}{i \hbar}\langle k| Z H_{0}-H_{0} Z|g\rangle=-\frac{m \omega_{k g}}{i}\langle k| Z|g\rangle \tag{58}
\end{gather*}
$$

Hence

$$
\begin{equation*}
f_{k g}=\frac{2}{i \hbar}\langle g| Z|k\rangle\langle k| P_{z}|g\rangle \tag{59}
\end{equation*}
$$

## Non-resonant interaction

Oscillator strength sum rule

Summing over $k$ introduces a closure relation

$$
\begin{equation*}
\sum_{k} f_{k g}=\frac{2}{i \hbar}\langle g| Z P_{z}|g\rangle \tag{60}
\end{equation*}
$$

$f_{k g}$ being real, the r.h.s is equal to the half sum with its conjugate

$$
\begin{equation*}
\sum_{k} f_{k g}=\frac{1}{i \hbar}\langle g| Z P_{z}-P_{z} Z|g\rangle=1 \tag{61}
\end{equation*}
$$

A simple sum rule for the oscillator strengths

## Non-resonant interaction

Comparison with classical model

In this picture, an atomic medium of numeric density $N$ appears a a mixture of classical harmonically bound electrons with resonance frequencies $\omega_{k g}$ and densities $N f_{k g}$. All our conclusions on the propagation of light in the classical medium thus retain their validity in this perturbative semi-classical model. This property explains why the naive harmonically bound electron leads to realistic predictions.

## A two-level system

Consider now the case of a radiation resonant on the transition between between the two levels $|g\rangle$ (lower, possibly ground level) and $|e\rangle$ i.e.

$$
\omega_{0}=\omega_{e g}
$$

All other levels can be neglected. Boils down to the interaction of a classical field with a spin $1 / 2$ system.

## Classical field and free atom

## Atomic system

Two states $|e\rangle$ and $|g\rangle$ or $|+\rangle$ and $|-\rangle$ or $|0\rangle$ and $|1\rangle$ in quantum information science.
Operator basis set: Pauli operators

$$
\begin{gather*}
\sigma_{x}=\left(\begin{array}{ll}
0 & 1 \\
1 & 0
\end{array}\right) \quad \sigma_{y}=\left(\begin{array}{cc}
0 & -i \\
i & 0
\end{array}\right) \quad \sigma_{z}=\left(\begin{array}{cc}
1 & 0 \\
0 & -1
\end{array}\right)  \tag{62}\\
{\left[\sigma_{x}, \sigma_{y}\right]=2 i \sigma_{z}} \tag{63}
\end{gather*}
$$

Spin lowering and raising operators

$$
\begin{gather*}
\sigma_{+}=|+\rangle\langle-|=\frac{\sigma_{x}+i \sigma_{y}}{2}=\left(\begin{array}{ll}
0 & 1 \\
0 & 0
\end{array}\right)  \tag{64}\\
\sigma_{-}=|-\rangle\langle+|=\sigma_{+}^{\dagger}=\frac{\sigma_{x}-i \sigma_{y}}{2}=\left(\begin{array}{ll}
0 & 0 \\
1 & 0
\end{array}\right)  \tag{65}\\
{\left[\sigma_{z}, \sigma_{ \pm}\right]= \pm 2 \sigma_{ \pm}} \tag{66}
\end{gather*}
$$

## Classical field and free atom

Atomic system

Most general observable $\sigma_{\mathbf{u}}$ with $\mathbf{u}=(\sin \theta \cos \phi, \sin \theta \sin \phi, \cos \theta)$

$$
\sigma_{\mathbf{u}}=\left(\begin{array}{cc}
\cos \theta & \sin \theta e^{-i \phi}  \tag{67}\\
\sin \theta e^{i \phi} & -\cos \theta
\end{array}\right)
$$

Eigenvectors

$$
\begin{align*}
& \left|+_{\mathbf{u}}\right\rangle=\left|0_{\mathbf{u}}\right\rangle=\cos \frac{\theta}{2}|+\rangle+\sin \frac{\theta}{2} e^{i \phi}|-\rangle  \tag{68}\\
& \left|-_{\mathbf{u}}\right\rangle=\left|1_{\mathbf{u}}\right\rangle=-\sin \frac{\theta}{2} e^{-i \phi}|+\rangle+\cos \frac{\theta}{2}|-\rangle \tag{69}
\end{align*}
$$

## Classical field and free atom

Atomic system
Bloch sphere


## Classical field and free atom

Atomic system

Rotation on the Bloch sphere by an angle $\theta$ around the axis defined by $\mathbf{v}$

$$
\begin{equation*}
R_{\mathbf{v}}(\theta)=e^{-i(\theta / 2) \sigma_{\mathbf{v}}}=\cos \frac{\theta}{2} \mathbb{1}-i \sin \frac{\theta}{2} \sigma_{\mathbf{v}} \tag{70}
\end{equation*}
$$

e.g. angle $\theta$ around $\mathbf{u}_{z}$

$$
R_{z}(\theta)=\left(\begin{array}{cc}
e^{-i \theta / 2} & 0  \tag{71}\\
0 & e^{i \theta / 2}
\end{array}\right)
$$

with $R_{z}(\pi / 2)\left|+_{x}\right\rangle=\left|+_{y}\right\rangle$ and $R_{v}(2 \pi)=-\mathbb{1}$

## Classical field and free atom

## Atomic Hamiltonian and observables

- Hamiltonian:

$$
\begin{equation*}
H_{0}=\frac{\hbar \omega_{e g}}{2} \sigma_{z} \tag{72}
\end{equation*}
$$

Generates a rotation of the Bloch vector at angular frequency $\omega_{\text {eg }}$ around Oz (Larmor precession in the NMR context).

- Dipole operator:

$$
\mathbf{D}=\left(\begin{array}{cc}
0 & \mathbf{d}  \tag{73}\\
\mathbf{d}^{*} & 0
\end{array}\right)=\mathbf{d} \sigma_{x}=\mathbf{d}\left(\sigma_{+}+\sigma_{-}\right)
$$

where $\mathbf{d}$ describes the polarization of the atomic transition. A priori complex, but taken as real for the sake of simplicity.

- Incoming field $\mathbf{E}(0, t)=\mathbf{E}_{0} \cos (\omega t+\varphi)$. We note

$$
\begin{equation*}
E_{1}=E_{0} e^{-i \varphi} \tag{74}
\end{equation*}
$$

## Classical field and free atom

Atomic Hamiltonian and observables

Atom-field Hamiltonian:

$$
\begin{align*}
H_{1} & =-\mathbf{d} \cdot \mathbf{E}_{0} \cos (\omega t+\varphi) \sigma_{x}  \tag{75}\\
H_{1} & =-\hbar \Omega \cos (\omega t+\varphi) \sigma_{x} \tag{76}
\end{align*}
$$

with definition of the 'Rabi frequency'

$$
\begin{equation*}
\Omega=\frac{\mathbf{d} \cdot \mathbf{E}_{0}}{\hbar} \tag{77}
\end{equation*}
$$

Remove time dependence?

## Classical field and free atom

## Rabi precession

Introduce $H_{0}^{\prime}=\hbar \omega \sigma_{z} / 2$ (inducing a spin precession at the field frequency) so that

$$
\begin{equation*}
H=H_{0}^{\prime}+\frac{\hbar \Delta}{2} \sigma_{z}+H_{1} \tag{78}
\end{equation*}
$$

with

$$
\begin{equation*}
\Delta=\omega_{e g}-\omega \tag{79}
\end{equation*}
$$

Interaction representation w.r.t. $H_{0}^{\prime}$, defined by $U_{0}^{\prime}=\exp \left(-i H^{\prime}{ }_{0} t / \hbar\right)$.

$$
\begin{equation*}
\widetilde{H}=U_{0}^{\prime \dagger} H_{1} U_{0}^{\prime} \tag{80}
\end{equation*}
$$

$\sigma_{z}$ part of $H_{1}$ unchanged (commutes with the evolution operator) but

$$
\begin{equation*}
\widetilde{\sigma}_{ \pm}=U_{0}^{\prime \dagger} \sigma_{ \pm} U_{0}^{\prime} \tag{81}
\end{equation*}
$$

## Classical field and free atom

## Rabi precession

Using the Baker-Hausdorff lemma:

$$
\begin{equation*}
e^{B} A e^{-B}=A+[B, A]+\frac{1}{2!}[B,[B, A]]+\ldots \tag{82}
\end{equation*}
$$

with $B \propto \sigma_{z}$ and $\sigma_{+}=A$

$$
\begin{equation*}
\tilde{\sigma}_{+}=\sigma_{+}+i \omega t \sigma_{+}+(i \omega t)^{2} \sigma_{+}+\cdots=e^{i \omega t} \sigma_{+} \tag{83}
\end{equation*}
$$

and, by hermitic conjugation

$$
\begin{gather*}
\widetilde{\sigma}_{-}=e^{-i \omega t} \sigma_{-}  \tag{84}\\
\widetilde{H}=\frac{\hbar \Delta}{2} \sigma_{z}-\frac{\hbar \Omega}{2}\left(e^{i(\omega t+\varphi)}+e^{-i(\omega t+\varphi)}\right)\left(e^{i \omega t} \sigma_{+}+e^{-i \omega t} \sigma_{-}\right) \tag{85}
\end{gather*}
$$

Two rapidly oscillating terms, and two constant ones.

## Classical field and free atom

Rabi precession
Rotating wave approximation (RWA): neglect terms oscillating rapidly in $\widetilde{H}$

$$
\begin{equation*}
\widetilde{H}=\frac{\hbar \Delta}{2} \sigma_{z}-\frac{\hbar \Omega}{2}\left(\sigma_{+} e^{-i \varphi}+\sigma_{-} e^{i \varphi}\right)=\frac{\hbar \Delta}{2} \sigma_{z}-\frac{\hbar \Omega}{2}\left(\sigma_{x} \cos \varphi+\sigma_{y} \sin \varphi\right) \tag{86}
\end{equation*}
$$

$$
\begin{equation*}
H=\frac{\hbar \Omega^{\prime}}{2} \sigma_{\mathbf{n}} \tag{87}
\end{equation*}
$$

with

$$
\begin{equation*}
\mathbf{n}=\frac{\Delta \mathbf{u}_{z}-\Omega \cos \varphi \mathbf{u}_{x}-\Omega \sin \varphi \mathbf{u}_{y}}{\Omega^{\prime}} \tag{88}
\end{equation*}
$$

and

$$
\begin{equation*}
\Omega^{\prime}=\sqrt{\Omega^{2}+\Delta^{2}} \tag{89}
\end{equation*}
$$

Hence,

$$
\begin{equation*}
U(t)=e^{-i\left(\Omega^{\prime} t / 2\right) \sigma_{\mathbf{n}}}=R_{\mathbf{n}}(\theta) \tag{90}
\end{equation*}
$$

with

$$
\begin{equation*}
\theta=\Omega^{\prime} t \tag{91}
\end{equation*}
$$

## Classical field and free atom

## Rabi precession

Resonant case: rotation around an axis in the equatorial plane $\mathbf{n}=-\cos \varphi \mathbf{u}_{x}-\sin \varphi \mathbf{u}_{y}$. Choosing $g$ as the initial state

$$
\begin{equation*}
p_{e}(t)=\frac{1-\cos (\Omega t)}{2} \tag{92}
\end{equation*}
$$

Rabi oscillation. Some particular pulses:

- ' $\pi / 2$ pulse', i.e. $t=\pi / 2 \Omega$. Evolution operator

$$
\begin{align*}
R_{\mathbf{n}}(\pi / 2)= & \frac{1}{\sqrt{2}}\left(\mathbb{1}-i \sigma_{\mathbf{n}}\right)=\frac{1}{\sqrt{2}}\left(\begin{array}{cc}
1 & i e^{-i \varphi} \\
i e^{i \varphi} & 1
\end{array}\right)  \tag{93}\\
|g\rangle & \longrightarrow \frac{1}{\sqrt{2}}\left(|g\rangle+i e^{-i \varphi}|e\rangle\right) \\
|e\rangle & \longrightarrow \frac{1}{\sqrt{2}}\left(|e\rangle+i e^{i \varphi}|g\rangle\right) \tag{94}
\end{align*}
$$

## Classical field and free atom

## Rabi precession

- $\Omega t=\pi$ ( $\pi$-pulse) exchange of levels
- $\Omega t=2 \pi$ ( $2 \pi$ pulse) global - sign associated to a $2 \pi$ rotation of a spin-1/2.

General case: rotation is around an axis making a non-trivial angle $\alpha$ (given by $\tan \alpha=\Omega^{\prime} / \Delta$ ) with the downwards $z$ axis. When starting from $|g\rangle$ the maximum excitation probability is

$$
\begin{equation*}
p_{e, m}=\frac{\Omega^{2}}{\Omega^{2}+\Delta^{2}} \tag{95}
\end{equation*}
$$

- Lorentzian resonance
- Width of order of $\pi / \tau$ for a given interrogation time $\tau$
(no limit to the spectroscopic resolution since relaxation processes are not taken into account)


## Classical field and free atom

Ramsey separated oscillatory fields method

Two short $\pi / 2$ quasi-resonant pulses separated by a long time interval $T$. Assume $\varphi=-\pi / 2$. The pulses induce the transformations:

$$
\begin{align*}
|e\rangle & \longrightarrow \frac{1}{\sqrt{2}}(|e\rangle+|g\rangle)  \tag{96}\\
|g\rangle & \longrightarrow \frac{1}{\sqrt{2}}(-|e\rangle+|g\rangle) \tag{97}
\end{align*}
$$

Starting from $|g\rangle$, after pulse 1 , atom is in state $|\Psi(\tau)\rangle=(1 / \sqrt{2})(-|e\rangle+|g\rangle)$.

## Classical field and free atom

## Ramsey separated oscillatory fields method

During time $T$, the atom evolves under the Hamiltonian $(\hbar \Delta / 2) \sigma_{z}$ and hence $|e\rangle \rightarrow \exp (-i \Phi / 2)|e\rangle$ and $|g\rangle \rightarrow \exp (i \Phi / 2)|g\rangle$, with $\Phi=\Delta t$. State immediately before pulse 2 , within an irrelevant global phase:

$$
\begin{equation*}
|\Psi(T)\rangle=\frac{1}{\sqrt{2}}\left(-|e\rangle+e^{i \Phi}|g\rangle\right) \tag{98}
\end{equation*}
$$

Final state

$$
\begin{gather*}
\left|\Psi_{f}\right\rangle=-\frac{1}{2}\left[\left(1+e^{i \phi}\right)|e\rangle+\left(1-e^{i \phi}\right)|g\rangle\right)  \tag{99}\\
p_{e}=\frac{1}{4}\left(1+e^{i \phi}\right)^{2}=\frac{1}{2}(1+\cos \Delta T) \tag{100}
\end{gather*}
$$

(note that $p_{e}=1$ for $\Delta=0$ : addition of two in-phase $\pi / 2$ pulses). Measurement of $p_{e}$ provides a spectroscopic resolution of the order of $1 / T$.

## Classical field and free atom

Ramsey separated oscillatory fields method
Signal to noise discussion: $N$ independent atoms undergoing the same Ramsey sequence

$$
\begin{equation*}
\left\langle N_{e}\right\rangle=\frac{N}{2}(1+\cos \Phi) \tag{101}
\end{equation*}
$$

with $\Phi=\Delta T$. Variance

$$
\begin{equation*}
\Delta^{2} N_{e}=N p_{e}\left(1-p_{e}\right)=\frac{N}{4} \sin ^{2} \Phi \tag{102}
\end{equation*}
$$

and hence

$$
\begin{equation*}
\Delta N_{e}=\frac{\sqrt{N}}{2} \sin \Phi \tag{103}
\end{equation*}
$$

Two measurements for $\Delta$ and $\Delta+\delta$, with $\delta \ll 1 / T$.

$$
\begin{equation*}
\left\langle N_{e}(\Delta+\delta)\right\rangle=\left\langle N_{e}(\Delta)\right\rangle-\frac{N T}{2} \delta \sin \Delta T \tag{104}
\end{equation*}
$$

## Classical field and free atom

Ramsey separated oscillatory fields method

Resolve the small detuning increment $\delta$ if

$$
\begin{equation*}
\frac{N T}{2} \delta \sin \Delta T>\sqrt{2} \frac{\sin \Delta T}{2} \sqrt{N} \tag{105}
\end{equation*}
$$

or

$$
\begin{equation*}
\delta>\frac{\sqrt{2}}{T \sqrt{N}} \tag{106}
\end{equation*}
$$

A more precise estimate of the spectroscopic sensitivity of the method. Amusingly independent of the interferometer phase. Ranges as $\sqrt{N}$ as expected for independent measurements.

## Atomic relaxation

- Take into account spontaneous emission
- Take into account all other sources of damping
- Take into account fluctuating fields acting on the atom
- An opportunity to introduce the formal treatment of relaxation in quantum mechanics in a rather general frame: the Kraus operators and the Lindblad master equation


## Atomic relaxation

## System and environment

- Quantum system $\mathcal{S}$ (the atom here) coupled to an environment $\mathcal{E}$. Jointly in a pure state $\left|\Psi_{\mathcal{S E}}\right\rangle$.
- We are interested only in $\rho_{\mathcal{S}}$, obtained by tracing the projector $\left|\Psi_{\mathcal{S E}}\right\rangle\left\langle\Psi_{\mathcal{S E}}\right|$ over the environment (the state of the environment is forever inaccessible).
- We seek an evolution equation for $\rho_{\mathcal{S}}$ alone.


## Atomic relaxation

## Kraus operators

- Transformation of the system's density matrix during a short time interval

$$
\begin{equation*}
\rho(t) \longrightarrow \rho(t+\tau) \tag{107}
\end{equation*}
$$

- $\tau \gg \tau_{c}$, correlation time of the reservoir observables, so that there are no coherent effects in the system-reservoir interaction
- This transformation is a 'quantum map'

$$
\begin{equation*}
\mathcal{L}(\rho(t))=\rho(T+\tau) \tag{108}
\end{equation*}
$$

## Atomic relaxation

## Kraus operators

Mathematical properties of a proper quantum map:

- Linear operation, i.e. a super-operator in a space of dimension $N_{\mathcal{S}}^{2}$ ( $N_{\mathcal{S}}$ system's Hilbert space dimension).
- Preserve unit trace and positivity (a density operator does not have any negative eigenvalue).
- "Completely positive". If, at a time $t, \mathcal{S}$ entangled with $\mathcal{S}^{\prime}, \mathcal{L}$ acting on $\mathcal{S}$ alone leads to a completely positive density operator for the joint state of $\mathcal{S}$ and $\mathcal{S}^{\prime}$ (not all maps are completely positive e.g. partial transpose).


## Atomic relaxation

## Kraus operators

Any completely positive map can be written as

$$
\begin{equation*}
\mathcal{L}(\rho)=\sum_{\mu} M_{\mu} \rho M_{\mu}^{\dagger} \tag{109}
\end{equation*}
$$

with the normalization condition

$$
\begin{equation*}
\sum_{\mu} M_{\mu}^{\dagger} M_{\mu}=\mathbb{1} \tag{110}
\end{equation*}
$$

There are at most $N_{\mathcal{S}}^{2}$ 'Kraus' operators $M_{\mu}$, which are not uniquely defined (same map when mixing the $M_{\mu}$ by a linear unitary matrix $V$ : $\left.M^{\prime} \mu=V_{\mu \nu} M_{\nu}\right)$.

## Atomic relaxation

## Kraus operators

Fit also in this representation:

- Hamiltonian evolution

$$
\begin{equation*}
\rho(t+\tau)=U(\tau) \rho U^{\dagger}(\tau) \tag{111}
\end{equation*}
$$

- 'unread' generalized measurement

$$
\begin{equation*}
\rho \longrightarrow \sum_{\mu} O_{\mu} \rho O_{\mu}^{\dagger} \tag{112}
\end{equation*}
$$

but not a measurement whose result $\mu$ is known

$$
\begin{equation*}
\rho \longrightarrow \frac{O_{\mu} \rho O_{\mu}^{\dagger}}{\operatorname{Tr}\left(O_{\mu} \rho O_{\mu}^{\dagger}\right)} \tag{113}
\end{equation*}
$$

(non-linear normalization term in the denominator)

## Atomic relaxation

## Lindblad equation

Kraus representation and differential representation of the map

$$
\begin{equation*}
\rho(t+\tau)=\sum_{\mu} M_{\mu} \rho M_{\mu}^{\dagger} \approx \rho(t)+\frac{d \rho}{d t} \tau \tag{114}
\end{equation*}
$$

- Environment unaffected by the system: the $M_{\mu} \mathrm{s}$ do not depend upon time $t$.
- They, however, depend clearly upon the tiny time interval $\tau$.
- One and only one of the $M_{\mu} \mathrm{s}$ is thus of the order of unity and all others must then be of order $\sqrt{\tau}$.

$$
\begin{align*}
& M_{0}=\mathbb{1}-i K_{\tau}  \tag{115}\\
& M_{\mu}=\sqrt{\tau} L_{\mu} \quad \text { for } \mu \neq 0 \tag{116}
\end{align*}
$$

## Atomic relaxation

Lindblad equation
$K$, having no particular properties, can be split in hermitian and anti-hermitian parts:

$$
\begin{equation*}
K=\frac{H}{\hbar}-i J \tag{117}
\end{equation*}
$$

where

$$
\begin{align*}
H & =\frac{\hbar}{2}\left(K+K^{\dagger}\right)  \tag{118}\\
J & =\frac{i}{2}\left(K-K^{\dagger}\right) \tag{119}
\end{align*}
$$

are both hermitian.

$$
\begin{equation*}
M_{0}=\mathbb{1}-\frac{i \tau}{\hbar} H-J \tau \tag{120}
\end{equation*}
$$

## Atomic relaxation

Lindblad equation
Thus

$$
\begin{equation*}
M_{0} \rho M_{0}^{\dagger}=\rho-\frac{i \tau}{\hbar}[H, \rho]-\tau[J, \rho]_{+} \tag{121}
\end{equation*}
$$

where $[J, \rho]_{+}=J \rho+\rho J$ is an anti-commutator.
$M_{0}^{\dagger} M_{0}=\mathbb{1}-2 J \tau$ and thus, by normalization since $\sum_{\mu} M_{\mu}^{\dagger} M_{\mu}=\mathbb{1}$

$$
\begin{equation*}
J=\frac{1}{2} \sum_{\mu \neq 0} L_{\mu}^{\dagger} L_{\mu} \tag{123}
\end{equation*}
$$

"Lindblad form" of the master equation

$$
\begin{equation*}
\frac{d \rho}{d t}=-\frac{i}{\hbar}[H, \rho]+\sum_{\mu \neq 0}\left(L_{\mu} \rho L_{\mu}^{\dagger}-\frac{1}{2} L_{\mu}^{\dagger} L_{\mu} \rho-\frac{1}{2} \rho L_{\mu}^{\dagger} L_{\mu}\right) \tag{124}
\end{equation*}
$$

## Atomic relaxation

## Quantum jumps

Consider a single time interval $\tau$ in the simple situation where the initial state is pure $\rho(0)=|\Psi\rangle\langle\Psi|$, with no Hamiltonian evolution. Then

$$
\begin{equation*}
\rho(\tau)=|\Psi\rangle\langle\Psi|+\tau \sum_{\mu}\left(L_{\mu}|\Psi\rangle\right)\left(\langle\Psi| L_{\mu}^{\dagger}\right) \tag{125}
\end{equation*}
$$

- Density matrix at time $\tau$ is a statistical mixture of the initial pure state (with a large probability of order 1) and of projectors on the states $L_{\mu}|\Psi\rangle$.
- The $L_{\mu} \mathrm{s}$ are 'jump operators' which describe a random (possibly large) evolution of the system which suddenly (at the time scale of the evolution) changes under the influence of the environment.
- Intuitive picture of quantum jumps for an atom emitting a single photon


## Atomic relaxation

## Quantum jumps

- The quantum jump operators are not defined unambiguously. Again, the same master equation can be recovered from different sets of $M_{\mu} \mathrm{s}$ (or $L_{\mu} \mathrm{s}$ ) linked together by a unitary transformation matrix. Different choices correspond to the so-called 'unravelings' of the master equation.
- In some situations, the quantum jumps have a direct physical meaning. e.g. emitting atom completely surrounded by a photo-detector array. The quantum jump then corresponds to a click of one detector. Different unravelings may then correspond to different ways of monitoring the environment, in this case to different detectors (photon counters, homodyne recievers...)
- In other situations, the quantum jumps are an abstract representation of the system+environment evolution.


## Atomic relaxation

## Quantum trajectories

- Even when the environment is not explicitly monitored, one may imagine that it is done. We then imagine we have full information about which quantum jump occurs when.
- The system is thus, at any time, in a pure state, which undergoes a stochastic trajectory in the Hilbert space, made up of continuous Hamiltonian evolutions interleaved with sudden quantum jumps.
- However, since we only imagine the information is available, we should describe the evolution of the density operator by averaging the system evolution over all possible trajectories.
- The 'environment simulator' concept provides a simple recipe to perform this averaging.


## Atomic relaxation

## Environment simulator

$\mathcal{B}$ coupled to $\mathcal{S}$ so that the reduced dynamics for $\mathcal{S}$ is the same as when coupled to $\mathcal{E}$.

- $\mathcal{B}$ prepared in the same reference state $|0\rangle$ at the start of each time interval $\tau$
- Hamiltonian evolution of $\mathcal{S}+\mathcal{B}$ during the time interval $\tau$

$$
\begin{equation*}
U_{\mathcal{S B}}|\Psi\rangle \otimes|0\rangle=M_{0}|\Psi\rangle \otimes|0\rangle+\sqrt{\tau} \sum_{\mu}\left(L_{\mu}|\Psi\rangle\right) \otimes|\mu\rangle \tag{126}
\end{equation*}
$$

- Unread measurement of $O_{\mathcal{B}}$ having the $|\mu\rangle \mathrm{s}$ as non degenerate eigenstates, with $\mu$ as the eigenvalue. This measurement tells which jump has happened if any.


## Atomic relaxation

## Environment simulator

At the end of the time interval $\tau$ :

- With a probability $p_{0}=\langle\Psi| M_{0}^{\dagger} M_{0}|\Psi\rangle=\operatorname{Tr}\left(\rho M_{0}^{\dagger} M_{0}\right)=$ $1-\tau \sum_{\mu \neq 0} \operatorname{Tr}\left(\rho L_{\mu}^{\dagger} L_{\mu}\right)=1-\sum_{\mu \neq 0} p_{\mu}$, the result is 0 , no jump and

$$
\begin{equation*}
\frac{M_{0}|\Psi\rangle}{\sqrt{p}_{0}}=\frac{1-i H \tau / \hbar-J \tau}{\sqrt{p}_{0}}|\Psi\rangle \tag{127}
\end{equation*}
$$

Evolution can be interpreted as resulting from evolution in the non-hermitian Hamiltonian

$$
\begin{equation*}
H_{e f f}=H-i \hbar J \tag{128}
\end{equation*}
$$

- With a probability $p_{\mu}=\tau \operatorname{Tr}\left(\rho L_{\mu}^{\dagger} L_{\mu}\right)$, the result is $\mu$ and the system's state is accordingly projected onto $M_{\mu}|\Psi\rangle / \sqrt{p_{\mu}}=L_{\mu}|\Psi\rangle / \sqrt{p_{\mu} / \tau}$.
The quantum trajectory is defined by the repetition of such steps.


## Atomic relaxation

## Environment simulator

We have no access to the environment state in most real cases.

- Recovers the right evolution during $\tau$ by averaging all projectors on all possible final pure states (with proper measurement probability weights).
- Recovers the full density operator evolution by averaging the projectors on all possible quantum trajectory states.
- Full mathematical equivalence between this average and the solution of the Lindblad equation.
- Leads to an efficient numerical method for solving Lindblad equations.


## Atomic relaxation

## Quantum Monte Carlo trajectories

- Initialize the state (randomly chosen eigenstate $|\Psi\rangle$ of $\rho$ )
- For each time interval $\tau$, evolve $|\Psi\rangle$ according to:
- Compute $p_{\mu}=\tau\langle\Psi| L_{\mu}^{\dagger} L_{\mu}|\Psi\rangle$ and $p_{0}=1-\sum_{\mu \neq 0} p_{\mu}$.
- Use a (good) random number generator to decide upon the result of the measurement of $\mathcal{B}$.
- If the result of the measurement is zero, evolve $|\Psi\rangle$ with

$$
\begin{equation*}
|\Psi\rangle \longrightarrow \frac{1-i H \tau / \hbar-J \tau}{\sqrt{p_{0}}}|\Psi\rangle \tag{129}
\end{equation*}
$$

- If the result of the measurement is $\mu \neq 0$, evolve $|\Psi\rangle$ by:

$$
\begin{equation*}
|\Psi\rangle \longrightarrow \frac{L_{\mu}}{\sqrt{\langle\Psi| L_{\mu}^{\dagger} l_{\mu}|\Psi\rangle}}|\Psi\rangle=\frac{L_{\mu}}{\sqrt{p_{\mu} / \tau}}|\Psi\rangle \tag{130}
\end{equation*}
$$

- Repeat the procedure for many trajectories
- Average the projectors $\rho(t)=\overline{|\Psi(t)\rangle\langle\Psi(t)|}$


## Atomic relaxation

## Quantum Monte Carlo trajectories

Interest of the Monte Carlo method:

- For each trajectory computes only a state vector with $N_{\mathcal{S}}$ dimensions i.e. $N_{\mathcal{S}}$ coupled differential equations, instead of $N_{\mathcal{S}}^{2}$ equations for the full density operator.
- Neeeds a statistical sample of trajectories. A few hundreds is enough to get a qualitative solution. Method more efficient than the direct integration when $N_{\mathcal{S}}$ is larger than a few hundreds.
- Gives a physical picture of the relaxation process (see below).

An extremely useful method, with thousands of applications.

## Atomic relaxation

## Spontaneous emission

A practical (and important) example. Optical transition: Zero temperature model.
A single jump operator (describing photon emission in a downwards transition)

$$
\begin{equation*}
L=\sqrt{\Gamma} \sigma_{-} \tag{131}
\end{equation*}
$$

with $\Gamma=1 / T_{1}$ ('longitudinal relaxation time'). Lindblad equation

$$
\begin{equation*}
\frac{d \rho}{d t}=\Gamma\left(\sigma_{-} \rho \sigma_{+}-\frac{1}{2} \sigma_{+} \sigma_{-} \rho-\frac{1}{2} \rho \sigma_{+} \sigma_{-}\right) \tag{132}
\end{equation*}
$$

## Atomic relaxation

## Spontaneous emission

With

$$
\rho=\left(\begin{array}{ll}
\rho_{e e} & \rho_{e g}  \tag{133}\\
\rho_{g e} & \rho_{g g}
\end{array}\right)
$$

the solution of the Lindblad equation is

$$
\begin{align*}
\frac{d \rho_{e e}}{d t} & =-\Gamma \rho_{e e}  \tag{134}\\
\frac{d \rho_{e g}}{d t} & =-\frac{\Gamma}{2} \rho_{e g} \tag{135}
\end{align*}
$$

- Relaxation of excited state population with a rate Г.
- Relaxation of coherence with a rate $\Gamma / 2$ (compatible with $\left.\rho_{e g} \leq \sqrt{\rho_{e e} \rho_{g g}}\right)$


## Atomic relaxation

## Phase damping

Model atomic relaxation due to random fields altering the atomic frequency and scrambling the coherence phase.

- Jump operator $\sqrt{\gamma / 2} \sigma_{z}$ with $\gamma=1 / T_{2}$ the 'transverse' relaxation rate and $T_{2}$ the transverse relaxation time. Models sudden phase shifts of coherences.
- No damping of the populations, but coherences damped at rate $\gamma$.
- Complete Lindblad equation with spontaneous emission

$$
\begin{align*}
\frac{d \rho_{e e}}{d t} & =-\Gamma \rho_{e e}  \tag{136}\\
\frac{d \rho_{e g}}{d t} & =-\frac{\Gamma}{2} \rho_{e g}-\gamma \rho_{e g}=-\gamma^{\prime} \rho_{e g} \tag{137}
\end{align*}
$$

where we define the total relaxation rate of the coherence by:

$$
\begin{equation*}
\gamma^{\prime}=\gamma+\frac{\Gamma}{2} \tag{138}
\end{equation*}
$$

## Atomic relaxation

## Spontaneous emission

Case of an initial superposition state $\left|\Psi_{0}\right\rangle=(1 / \sqrt{2})(|e\rangle+|g\rangle)$. Analysis in terms of the Monte Carlo trajectories.

- No jump evolution. With $|\Psi(t)\rangle=c_{e}|e\rangle+c_{g}|g\rangle$ and use effective Hamiltonian

$$
\begin{gather*}
H=-i \hbar J=-\frac{i \hbar}{2} \Gamma \sigma_{+} \sigma_{-}=-\frac{i \hbar}{2} \Gamma|e\rangle\langle e|  \tag{139}\\
i \hbar \frac{d c_{e}}{d t}=-\frac{i \hbar}{2} \Gamma c_{e} \quad c_{e}(t)=c_{e}(0) e^{-\Gamma t / 2} \quad \frac{d c_{g}}{d t}=0  \tag{140}\\
|\Psi(t)\rangle=\frac{1}{\left|c_{e}(0)\right|^{2} e^{-\Gamma T}+\left|c_{g}(0)\right|^{2}}\left(c_{e}(0) e^{-\Gamma t / 2}|e\rangle+c_{g}(0)|g\rangle\right) \tag{141}
\end{gather*}
$$

A negative detection (no photon emitted) changes the system's state.

- Jump: state becomes $|g\rangle$. No further evolution.


## Optical Bloch equations

- Merge the atom-field interaction and the relaxation (phase damping and/or spontaneous emission) in a single set of equations.
- Analyse the immediate consequences of these equations.


## Optical Bloch equations

The equations

Hamiltonian in interaction representation w.r.t. the field frequency:

$$
\begin{equation*}
H=\frac{\hbar \Delta}{2} \sigma_{z}-\frac{\hbar \Omega}{2}\left(\sigma_{+} e^{-i \varphi}+\sigma_{-} e^{i \varphi}\right) \tag{142}
\end{equation*}
$$

with $\Omega=d E_{0} / \hbar$ and $\Delta=\omega_{e g}-\omega$ and

$$
\begin{equation*}
E_{1}=E_{0} e^{-i \varphi} \tag{143}
\end{equation*}
$$

## Optical Bloch equations

The equations

Coherent evolution of $\rho$ ruled by the Schrödinger equation:

$$
\begin{align*}
\frac{d \rho_{e e}}{d t} & =\Omega \operatorname{lm}\left(\rho_{e g} e^{i \varphi}\right) \\
& =\frac{d}{\hbar} \operatorname{lm}\left(\rho_{e g} E_{1}^{*}\right) \tag{144}
\end{align*}
$$

and

$$
\begin{align*}
\frac{d \rho_{e g}}{d t} & =-i \Delta \rho_{e g}+i \frac{\Omega}{2} e^{-i \varphi}\left(\rho_{g g}-\rho_{e e}\right) \\
& =-i \Delta \rho_{e g}-i \frac{d}{2 \hbar} E_{1}\left(\rho_{e e}-\rho_{g g}\right) \tag{145}
\end{align*}
$$

## Optical Bloch equations

The equations

Add relaxation (assume mere addition of evolution terms and note that Lindblad equation terms are not changed in interaction representation)

$$
\begin{align*}
\frac{d \rho_{e e}}{d t} & =\frac{d}{\hbar} \operatorname{Im}\left(\rho_{e g} E_{1}^{*}\right)-\Gamma \rho_{e e}  \tag{146}\\
\frac{d \rho_{e g}}{d t} & =-i \Delta \rho_{e g}-i \frac{d}{2 \hbar} E_{1}\left(\rho_{e e}-\rho_{g g}\right)-\gamma^{\prime} \rho_{e g} \tag{147}
\end{align*}
$$

with $\Gamma=1 / T_{1}$ and $\gamma^{\prime}=\left(1 / 2 T_{1}\right)+1 / T_{2}$

## Optical Bloch equations

## Equivalent forms

Introducing

- The populations $N_{e}=\rho_{e e}$ and $N_{g}=\rho_{g g}$
- The complex dipole amplitude

$$
\begin{equation*}
\mathcal{D}=2 d \rho_{e g} \tag{148}
\end{equation*}
$$

so that the average value of the dipole in state $\rho$ is $\operatorname{Re} \mathcal{D}$
We get:

$$
\begin{align*}
\frac{d N_{e}}{d t} & =\frac{1}{2 \hbar} \operatorname{lm}\left(\mathcal{D} E_{1}^{*}\right)-\Gamma N_{e}  \tag{149}\\
\frac{d \mathcal{D}}{d t} & =-i \Delta \mathcal{D}-\gamma^{\prime} \mathcal{D}-i \frac{d^{2} E_{1}}{\hbar}\left(N_{e}-N_{g}\right) \tag{150}
\end{align*}
$$

## Optical Bloch equations

## Equivalent forms

Introducing the Bloch vector $\mathbf{r}=(x, y, z)$ so that

$$
\begin{equation*}
\rho=\frac{1+\mathbf{r} \cdot \boldsymbol{\sigma}}{2} \tag{151}
\end{equation*}
$$

or

$$
\begin{align*}
\rho=\frac{1}{2}\left(\begin{array}{cc}
1+z & x-i y \\
x+i y & 1-z
\end{array}\right)  \tag{152}\\
x=2 \operatorname{Re} \rho_{e g} \quad y=-2 \operatorname{lm} \rho_{e g} \quad z=2 \rho_{e e}-1 \tag{153}
\end{align*}
$$

With $E_{1}=E_{x}+i E_{y}$

$$
\begin{align*}
\frac{d z}{d t} & =-\frac{d}{\hbar}\left(x E_{y}+y E_{x}\right)-\Gamma(1+z)  \tag{154}\\
\frac{d x}{d t} & =-\Delta y+\frac{d}{\hbar} z E_{y}-\gamma^{\prime} x  \tag{155}\\
\frac{d y}{d t} & =+\Delta x+\frac{d}{\hbar} z E_{x}-\gamma^{\prime} y \tag{156}
\end{align*}
$$

## Optical Bloch equations

Rabi oscillations revisited
Rabi oscillations with relaxation. Simplifying hypotheses:

- Initial state $|g\rangle$ corresponding to $z=-1$ and $x=y=0$.
- The field is purely real: $E_{y}=0, E_{x}=+E_{0}$
- Atom and field are at resonance: $\Delta=0$.

$$
\begin{align*}
\frac{d z}{d t} & =-\Omega y-\Gamma(1+z)  \tag{157}\\
\frac{d y}{d t} & =\Omega z-\gamma^{\prime} y \tag{158}
\end{align*}
$$

$x=0$ at any time.

$$
\begin{equation*}
\frac{d^{2} z}{d t^{2}}+\left(\Gamma+\gamma^{\prime}\right) \frac{d z}{d t}+\left(\Omega^{2}+\gamma^{\prime} \Gamma\right) z=-\gamma^{\prime} \Gamma \tag{159}
\end{equation*}
$$

## Optical Bloch equations

Rabi oscillations revisited

Steady state:

$$
\begin{gather*}
z_{s}=-\frac{\gamma^{\prime} \Gamma}{\Omega^{2}+\gamma^{\prime} \Gamma}  \tag{160}\\
y_{s}=\frac{\Omega}{\gamma^{\prime}} z=-\frac{\Omega \Gamma}{\Omega^{2}+\gamma^{\prime} \Gamma} \tag{161}
\end{gather*}
$$

- For $\Omega \rightarrow 0, y_{s}=0$ and $z_{s}=-1$
- For $\Omega \rightarrow \infty, z_{s}=y_{s}=0$


## Optical Bloch equations

Rabi oscillations revisited

Transient regime. Simplifying hypotheses:

- $\gamma^{\prime}=\Gamma / 2$ : no transverse relaxation
- $\Omega \gg \Gamma$ : Strong drive

$$
\begin{equation*}
\frac{d^{2} z}{d t^{2}}+\frac{3 \Gamma}{2} \frac{d z}{d t}+\Omega^{2} z=0 \tag{162}
\end{equation*}
$$

Solution:

$$
\begin{equation*}
z(t)=-\cos (\Omega t) e^{-3 \Gamma t / 2} \tag{163}
\end{equation*}
$$

an exponentially damped Rabi oscillation at the frequency $\Omega$.

## Optical Bloch equations

Rabi oscillations revisited

A simple interpretation in terms of quantum trajectories (only spontaneous emission relaxation)

- Before the first jump, an uninterrupted Rabi oscillation
- The first jump projects the atom in $|g\rangle$ and restarts the Rabi oscillation
- The occurrence of random jumps thus dephase the oscillations corresponding to different trajectories
- Hence an exponential damping of the Rabi oscillation amplitude.


## Optical Bloch equations

## Oscillator strength revisited

Return to the hypotheses of first paragraph

- Atom initially in $|g\rangle$
- Detuned field $\Delta \gg \Gamma, \gamma^{\prime}$, hence $N_{g} \approx 1$

We determine the steady state complex dipole $\mathcal{D}=2 d \rho_{\text {ge }}$ from

$$
\begin{gather*}
\frac{d \mathcal{D}}{d t}=-i \Delta \mathcal{D}-\gamma^{\prime} \mathcal{D}-i \frac{d^{2} E_{1}}{\hbar}\left(N_{e}-N_{g}\right)  \tag{164}\\
\mathcal{D}_{s}=\frac{d^{2}}{\hbar \Delta} E_{1}=\frac{\left.q^{2}|\langle e| z| g\right\rangle\left.\right|^{2}}{\hbar\left(\omega_{e g}-\omega\right)} E_{1} \tag{165}
\end{gather*}
$$

and define the quantum polarizability as

$$
\begin{equation*}
\left.\alpha_{Q}=\frac{q^{2}}{\hbar\left(\omega_{e g}-\omega\right)}|\langle e| z| g\right\rangle\left.\right|^{2} \tag{166}
\end{equation*}
$$

## Optical Bloch equations

## Oscillator strength revisited

Comparing the quantum and the classical polarizability:

$$
\begin{equation*}
\alpha_{c}=\frac{q^{2}}{2 m \epsilon_{0} \omega_{e g}} \frac{1}{\omega_{e g}-\omega} \tag{167}
\end{equation*}
$$

we get back the 'oscillator strength' (a mere consistency check)

$$
\begin{equation*}
\left.f=\frac{2 m \omega_{e g}}{\hbar}|\langle e| z| g\right\rangle\left.\right|^{2} \tag{168}
\end{equation*}
$$

## Two limit cases

Back to Einstein coefficients

Recover the Einstein coefficients as a limit case of the Optical Bloch Equations in two limit cases

- Strong transverse damping $\gamma^{\prime} \approx \gamma$
- Stochastic, noisy driving field

In both cases, stochasticity turns the coherent Rabi oscillation into transfer rates à la Einstein

## Two limit cases

## Strong transverse relaxation

Assume $\gamma^{\prime} \approx \gamma$ and $\Gamma \ll \gamma^{\prime}$. Use again

$$
\begin{equation*}
\frac{d \mathcal{D}}{d t}=-i \Delta \mathcal{D}-\gamma^{\prime} \mathcal{D}-i \frac{d^{2} E_{1}}{\hbar}\left(N_{e}-N_{g}\right) \tag{169}
\end{equation*}
$$

Fast relaxation allows to neglect $d \mathcal{D} / d t$. Assume thus that the dipole is at any time in the steady state value:

$$
\begin{equation*}
\mathcal{D}=\frac{i}{\gamma^{\prime}+i \Delta} \frac{d^{2} E_{1}}{\hbar}\left(N_{g}-N_{e}\right) \tag{170}
\end{equation*}
$$

Inject in the equation of motion for $N_{e}$ :

$$
\begin{align*}
\frac{d N_{e}}{d t} & =-\Gamma N_{e}+\frac{1}{2 \hbar} \operatorname{lm}\left[\frac{i}{\gamma^{\prime}+i \Delta} \frac{d^{2} E_{1}}{\hbar}\left(N_{g}-N_{e}\right) E_{1}^{*}\right] \\
& =-\Gamma N_{e}+\frac{d^{2} E_{0}^{2}}{2 \hbar^{2}}\left(N_{g}-N_{e}\right) \frac{\gamma^{\prime}}{\gamma^{\prime 2}+\Delta^{2}} \tag{171}
\end{align*}
$$

## Two limit cases

## Strong transverse relaxation

Assume a small but finite frequency bandwidth for the electric field: $E_{0}^{2} \propto u_{\nu_{0}}$. Make field resonant $(\Delta=0)$.

$$
\begin{gather*}
\frac{d N_{e}}{d t}=-\Gamma N_{e}+\frac{d^{2} E_{0}^{2}}{2 \hbar^{2} \gamma^{\prime}}\left(N_{g}-N_{e}\right)=-\Gamma N_{e}+\frac{\Omega^{2}}{2 \gamma^{\prime}}\left(N_{g}-N_{e}\right)  \tag{172}\\
\frac{d N_{e}}{d t}=A_{e g} N_{e}+\left(B_{g e} u_{\nu} N_{g}-B_{e g} u_{\nu} N_{e}\right) \tag{173}
\end{gather*}
$$

with the evident correspondence $A_{\text {eg }}=\Gamma$

## Two limit cases

## Stochastic fields

Described in terms of a slowly variable complex amplitude $E_{1}(t)$ modulating an oscillation at the average frequency $\bar{\omega}$ :

$$
\begin{equation*}
E(t)=E_{1}(t) e^{-i \bar{\omega} t} \tag{174}
\end{equation*}
$$

Stochastic properties encoded in the autocorrelation function:

$$
\begin{equation*}
\Gamma_{E}(\tau)=\lim _{T \rightarrow \infty} \frac{1}{T} \int_{t}^{t+T} E_{1}^{*}\left(t^{\prime}\right) E_{1}\left(t^{\prime}-\tau\right) d t^{\prime} \tag{175}
\end{equation*}
$$

or, within an ergodic hypothesis

$$
\begin{equation*}
\Gamma_{E}(\tau)=\overline{E_{1}^{*}(t) E_{1}(t-\tau)} \tag{176}
\end{equation*}
$$

where the overline denotes an average over very many realizations of the source. $\Gamma_{E}$ has a width $\tau_{c}$ (defining the source correlation time). Note

$$
\begin{equation*}
\Gamma_{E}(-\tau)=\overline{E_{1}^{*}(t) E_{1}(t+\tau)}=\overline{E_{1}^{*}\left(t^{\prime}-\tau\right) E_{1}\left(t^{\prime}\right)}=\Gamma_{E}^{*}(\tau) \tag{177}
\end{equation*}
$$

## Two limit cases

Stochastic fields

Spectral density of radiation $S_{E}(\omega)$ :

$$
\begin{equation*}
S_{E}(\omega)=\frac{1}{2 \pi} \int_{-\infty}^{\infty} d \tau \Gamma_{E}(\tau) e^{-i \omega \tau} \tag{178}
\end{equation*}
$$

real due to (177). Spectrum of the source: spectral density translated by $\bar{\omega}$. Width of the order of $1 / \tau_{c}$.

## Two limit cases

## Stochastic fields

Since $E_{1}(t)$ varies slowly at the time scale of the optical frequency:

$$
\begin{equation*}
\frac{d \rho_{e g}}{d t}=-i \Delta \rho_{e g}-\gamma^{\prime} \rho_{e g}-\frac{i d}{2 \hbar} E_{1}(t)\left(\rho_{e e}-\rho_{g g}\right) \tag{179}
\end{equation*}
$$

where $\Delta$ is now $\omega_{e g}-\bar{\omega}$. Defining

$$
\begin{equation*}
\widetilde{\rho_{e g}}=\rho_{e g} e^{\left(i \Delta+\gamma^{\prime}\right) t} \tag{180}
\end{equation*}
$$

we get

$$
\begin{equation*}
\widetilde{\rho_{e g}}(t)=-\frac{i d}{2 \hbar} \int_{0}^{t} E_{1}\left(t^{\prime}\right)\left(\rho_{e e}-\rho_{g g}\right)\left(t^{\prime}\right) e^{\left(i \Delta+\gamma^{\prime}\right) t^{\prime}} d t^{\prime} \tag{181}
\end{equation*}
$$

With $\rho_{\text {eg }}=\widetilde{\rho_{\text {eg }}} \exp \left[-\left(i \Delta+\gamma^{\prime}\right) t\right]$ :

$$
\begin{equation*}
\rho_{e g}(t)=-\frac{i d}{2 \hbar} \int_{0}^{t} E_{1}\left(t^{\prime}\right)\left(\rho_{e e}-\rho_{g g}\right)\left(t^{\prime}\right) e^{\left(-i \Delta-\gamma^{\prime}\right)\left(t-t^{\prime}\right)} d t^{\prime} \tag{182}
\end{equation*}
$$

## Two limit cases

## Stochastic fields

Plug the expression of $\rho_{\text {eg }}(t)$ in the equation of the populations:

$$
\begin{equation*}
\frac{d \rho_{e e}}{d t}=-\Gamma \rho_{e e}-\frac{d^{2}}{2 \hbar^{2}} \operatorname{Re} \int_{0}^{t} E_{1}^{*}(t) E_{1}\left(t^{\prime}\right)\left(\rho_{e e}-\rho_{g g}\right)\left(t^{\prime}\right) e^{\left(-i \Delta-\gamma^{\prime}\right)\left(t-t^{\prime}\right)} d t^{\prime} \tag{183}
\end{equation*}
$$

Settting $t-t^{\prime}=\tau$, or $t^{\prime}=t-\tau(0 \leq \tau \leq t)$

$$
\begin{equation*}
\frac{d \rho_{e e}}{d t}=-\Gamma \rho_{e e}-\frac{d^{2}}{2 \hbar^{2}} \operatorname{Re} \int_{0}^{t} E_{1}(t-\tau) E_{1}^{*}(t)\left(\rho_{e e}-\rho_{g g}\right)(t-\tau) e^{\left(-i \Delta-\gamma^{\prime}\right) \tau} d \tau \tag{184}
\end{equation*}
$$

## Two limit cases

## Stochastic fields

Perform an ensemble average of the evolution equations (leaving $\rho$ invariant)

$$
\frac{d \rho_{e e}}{d t}=-\Gamma \rho_{e e}-\frac{d^{2}}{2 \hbar^{2}} \operatorname{Re} \int_{0}^{t} \Gamma_{E}(\tau)\left(\rho_{e e}-\rho_{g g}\right)(t-\tau) e^{\left(-i \Delta-\gamma^{\prime}\right) \tau} d \tau \text { (185) }
$$

Short source correlation time $\tau_{c}$.

- Replace $\left(\rho_{e e}-\rho_{g g}\right)(t-\tau)$ by $\left(\rho_{e e}-\rho_{g g}\right)(t)$
- Extend upper integral bound to infinity


## Two limit cases

Stochastic fields

Final equation of motion:

$$
\begin{equation*}
\frac{d \rho_{e e}}{d t}=-\Gamma \rho_{e e}-C(\Delta)\left(\rho_{e e}-\rho_{g g}\right) \tag{186}
\end{equation*}
$$

where

$$
\begin{equation*}
C(\Delta)=\frac{d^{2}}{2 \hbar^{2}} \operatorname{Re} \int_{0}^{\infty} \Gamma_{E}(\tau) e^{\left(-i \Delta-\gamma^{\prime}\right) \tau} d \tau \tag{187}
\end{equation*}
$$

Neglect the transverse relaxation rate $\gamma^{\prime}$ compared to the field frequency width.

$$
\begin{equation*}
C(\Delta)=\frac{d^{2}}{2 \hbar^{2}} \operatorname{Re} \int_{0}^{\infty} \Gamma_{E}(\tau) e^{-i \Delta \tau} d \tau \tag{188}
\end{equation*}
$$

## Two limit cases

## Stochastic fields

Link with spectral density:

$$
\begin{equation*}
2 \pi S_{E}(\Delta)=\int_{-\infty}^{0} \Gamma_{E}(\tau) e^{-i \Delta \tau} d \tau+\int_{0}^{\infty} \Gamma_{E}(\tau) e^{-i \Delta \tau} d \tau \tag{189}
\end{equation*}
$$

With $\Gamma_{E}(-\tau)=\Gamma_{E}^{*}(\tau)$ :

$$
\begin{equation*}
\int_{-\infty}^{0} \Gamma_{E}(\tau) e^{-i \Delta \tau} d \tau=\int_{0}^{\infty} \Gamma_{E}(-\tau) e^{i \Delta \tau} d \tau=\left(\int_{0}^{\infty} \Gamma_{E}(\tau) e^{-i \Delta \tau} d \tau\right)^{*} \tag{190}
\end{equation*}
$$

Hence,

$$
\begin{equation*}
2 \pi S_{E}(\Delta)=2 \operatorname{Re} \int_{0}^{\infty} \Gamma_{E}(\tau) e^{-i \Delta \tau} d \tau \tag{191}
\end{equation*}
$$

and, finally

$$
\begin{equation*}
C(\Delta)=\frac{\pi d^{2}}{2 \hbar^{2}} S_{E}(\Delta) \tag{192}
\end{equation*}
$$

## Two limit cases

## Einstein at last

Assuming finally the resonance condition $(\Delta=0)$ and noting that

$$
\begin{equation*}
u_{\nu}=2 \pi^{2} \epsilon_{0} S_{E}(0) \tag{193}
\end{equation*}
$$

we get

$$
\begin{equation*}
C(0)=C=B_{e g} u_{\nu} \tag{194}
\end{equation*}
$$

and

$$
\begin{equation*}
B_{e g}=\frac{d^{2}}{4 \pi \epsilon_{0} \hbar^{2}} \tag{195}
\end{equation*}
$$

and finally

$$
\begin{equation*}
\frac{d N_{e}}{d t}=-A_{e g} N_{e}+B_{e g} u_{\nu}\left(N_{g}-N_{e}\right) \tag{196}
\end{equation*}
$$

## Two limit cases

## Einstein at last

The value of $B_{\text {eg }}$ obtained here differs by a factor $3 / 2$ from that obtained from

$$
\begin{equation*}
A_{e g}=\frac{d^{2} \omega^{3}}{3 \pi \epsilon_{0} \hbar c^{3}} \tag{197}
\end{equation*}
$$

which is

$$
\begin{equation*}
B_{e g}=\frac{d^{2}}{6 \pi \epsilon_{0} \hbar^{2}} \tag{198}
\end{equation*}
$$

Reason: no averaging over polarizations in our calculation.

## Two limit cases

## Spectrum of a lamp

An exercise on autocorrelation functions. Spontaneous emission by a large ensemble of atoms. Train of exponentially damped pulses ( $N_{p}$ per unit time) with random relative phases:

$$
\begin{gather*}
E_{1}(t)=\sum_{i=-\infty}^{\infty} E_{0} e^{i \phi_{i}} e^{-\left(t-t_{i}\right) / \tau_{e}} \Theta\left(t-t_{i}\right)  \tag{199}\\
\Gamma_{E}=N_{p} T \gamma_{e} \tag{200}
\end{gather*}
$$

with

$$
\begin{align*}
\gamma_{E}(\tau)=\frac{1}{T} & E_{0}^{2} \int_{0}^{T} e^{-t / \tau_{e}} e^{-(t-\tau) / \tau_{e}} \Theta(t-\tau) d t  \tag{201}\\
\gamma_{E}(\tau) & =\frac{1}{T} E_{0}^{2}\left[\int_{\tau}^{\infty} e^{-2 t / \tau_{e}} d t\right] e^{\tau / \tau_{e}} \\
& =\frac{1}{T} E_{0}^{2} \frac{\tau_{e}}{2} e^{-|\tau| / \tau_{e}} \tag{202}
\end{align*}
$$

## Two limit cases

Spectrum of a lamp

Finally

$$
\begin{equation*}
\Gamma_{E}(\tau)=N_{p} E_{0}^{2} \frac{\tau_{e}}{2} e^{-|\tau| / \tau_{e}} \tag{203}
\end{equation*}
$$

and

$$
\begin{equation*}
S_{E}(\omega)=\frac{N_{p} E_{0}^{2}}{\pi} \frac{1}{\omega^{2}+\left(1 / \tau_{e}\right)^{2}} \tag{204}
\end{equation*}
$$

a Lorentzian spectrum with a width $1 / \tau_{e}$.

## Applications

Explore direct applications of the Optical Bloch equations:

- Steady-state and Saturation
- Optical pumping
- Dark resonance and EIT
- Light shifts and Autler Townes splitting
- Maxwell Bloch equations


## Steady-state and Saturation

Classical model (chapter 1): power given to the matter by the field

$$
\begin{equation*}
\mathcal{E}=\frac{1}{2} \epsilon_{0} \omega \chi^{\prime \prime}\left|E_{1}\right|^{2}=\frac{1}{2} \epsilon_{0} \omega \mathcal{N} \alpha^{\prime \prime}\left|E_{1}\right|^{2} \tag{205}
\end{equation*}
$$

where $\mathcal{N}$ is the number of atoms in the medium (the populations in the Bloch equations sum to one so that the number of atoms in $|e\rangle$ is $\mathcal{N} N_{z}$ ) The complex dipole amplitude $D$ is $D=\epsilon_{0} \alpha E_{1}$ and thus

$$
\begin{equation*}
\mathcal{E}=\mathcal{N} \frac{\omega E_{1}}{2} \operatorname{Im} D \tag{206}
\end{equation*}
$$

Linear function of the incoming power. An unrealistic model: an atom cannot diffuse a MW laser field. What is the prediction of the OBEs?

## Steady-state and Saturation

## Steady state power

Replace in the classical expression of the energy exchange the dipole by $\mathcal{D}$. Recall the OBEs and assume $E_{1}$ real without loss of generality

$$
\begin{align*}
\frac{d N_{e}}{d t} & =\frac{1}{2 \hbar} \operatorname{lm}\left(\mathcal{D} E_{1}\right)-\Gamma N_{e}  \tag{207}\\
\frac{d \mathcal{D}}{d t} & =-i \Delta \mathcal{D}-\gamma^{\prime} \mathcal{D}-i \frac{d^{2} E_{1}}{\hbar}\left(N_{e}-N_{g}\right) \tag{208}
\end{align*}
$$

In the steady state:

$$
\begin{equation*}
\mathcal{D}=\frac{\Delta+i \gamma^{\prime}}{\Delta^{2}+\gamma^{\prime 2}} \frac{d^{2} E_{1}}{\hbar}\left(N_{g}-N_{e}\right) \tag{209}
\end{equation*}
$$

## Steady-state and Saturation

## Steady state power

Similarly, the steady state value of $N_{e}$ is

$$
\begin{equation*}
N_{e}=\frac{d^{2} E_{1}^{2}}{2 \hbar^{2} \Gamma}\left(N_{g}-N_{e}\right) \frac{\gamma^{\prime}}{\Delta^{2}+\gamma^{\prime 2}} \tag{210}
\end{equation*}
$$

Introducing the Rabi frequency $\Omega=d E_{1} / \hbar$ and defining the saturation parameter:

$$
\begin{equation*}
s=\frac{\Omega^{2}}{\Gamma \gamma^{\prime}} \frac{1}{1+\Delta^{2} / \gamma^{\prime 2}}, \tag{211}
\end{equation*}
$$

which has a Lorentzian variation with the atom-field detuning $\Delta$, we arrive at

$$
\begin{align*}
N_{e} & =\frac{s / 2}{1+s}  \tag{212}\\
N_{g}-N_{e} & =\frac{1}{1+s} \tag{213}
\end{align*}
$$

## Steady-state and Saturation

Steady state power

We get also $\mathcal{D}$ such that

$$
\begin{equation*}
|\mathcal{D}|^{2}=d^{2} \frac{\Gamma}{\gamma^{\prime}} \frac{s}{(1+s)^{2}} \tag{214}
\end{equation*}
$$

and finally

$$
\begin{equation*}
\mathcal{E}=\frac{\mathcal{N} \hbar \omega \Gamma}{2} \frac{s}{1+s} \tag{215}
\end{equation*}
$$

always positive, since there can be no population inversion. The absorbed energy has a Lorentzian shape for a small saturation parameter $(s \ll 1$; small Rabi frequency).

## Steady-state and Saturation

## Saturation intensity

At resonance $(\Delta=0)$ the 'saturation parameter' $s=s_{0}$ is:

$$
\begin{equation*}
s_{0}=\frac{\Omega^{2}}{\Gamma \gamma^{\prime}} \tag{216}
\end{equation*}
$$

and

$$
\begin{equation*}
\mathcal{E}=\mathcal{N} \frac{\hbar \omega}{2} \Gamma \frac{s_{0}}{1+s_{0}} \tag{217}
\end{equation*}
$$

- At low power, $\mathcal{E}$ is proportional to $s_{0}$ i.e. to the incoming field intensity. Recover classical model result
- At infinite input power,

$$
\begin{equation*}
\mathcal{E}_{s}=\mathcal{N} \hbar \omega \frac{\Gamma}{2} \tag{218}
\end{equation*}
$$

photons scattered at a rate $\Gamma / 2$.

- Onset of the saturation for $s_{0} \approx 1$


## Steady-state and Saturation

## Saturation intensity

With $s_{0}=d^{2} E_{1}^{2} / \hbar^{2} \Gamma \gamma^{\prime}$ and an incident power per unit surface $I=\epsilon_{0} c E_{1}^{2} / 2$ then

$$
\begin{equation*}
s_{0}=\frac{d^{2} E_{1}^{2}}{\hbar^{2} \Gamma \gamma^{\prime}}=\frac{l}{l_{s}} \tag{219}
\end{equation*}
$$

where the saturation intensity $I_{s}$ is

$$
\begin{equation*}
I_{s}=\frac{\Gamma \gamma^{\prime}}{d^{2}} \frac{\epsilon_{0} c}{2} \hbar^{2} \tag{220}
\end{equation*}
$$

Consider the simple case $\gamma^{\prime}=\Gamma / 2$ (no additional transverse damping) then

$$
\begin{equation*}
I_{s}=\frac{\Gamma^{2}}{4} \frac{\epsilon_{0} c}{d^{2}} \hbar^{2} \tag{221}
\end{equation*}
$$

## Steady-state and Saturation

## Saturation intensity

Using (anticipating again on Chapter 4)

$$
\begin{gather*}
\Gamma=\frac{\omega^{3} d^{2}}{3 \pi \epsilon_{0} \hbar c^{3}}  \tag{222}\\
I_{s}=\frac{\pi}{3} \hbar \omega \Gamma \frac{1}{\lambda^{2}}=\hbar \omega \frac{\Gamma}{2} \frac{1}{\sigma_{c}} \tag{223}
\end{gather*}
$$

- Saturation: one photon incident in the resonant cross section of the classical model, $\sigma_{c}=3 \lambda^{2} / 2 \pi$, at the maximum rate of diffusion $\Gamma / 2$.
- Order of magnitude: with $\Gamma=3.10^{7} \mathrm{~s}^{-1}, \lambda=1 \mu \mathrm{~m}$ we get $I_{s}=0.6 \mathrm{~mW} / \mathrm{cm}^{2}$


## Steady-state and Saturation

## Saturation spectroscopy

A useful method to get rid of the Doppler broadening of atomic transitions.
(a)



## Steady-state and Saturation

## Saturation spectroscopy

Resonance conditions for the two beams

- Direct beam: $\Delta=\omega_{e g}-\omega=-k v_{z}$
- Reflected beam: $\Delta=k v_{z}^{\prime}$
- Out of resonance ( $\Delta$ much larger than $\Omega$ and $\Gamma$ ), the two counterpropagating beams interact with different velocity classes due to the Doppler effect. The absorptions are independent and equivalent to one path in a medium with a double atom number $2 \mathcal{N}^{1}$. The absorbed energy is

$$
\begin{equation*}
\mathcal{E}=2 \times \frac{\mathcal{N} \hbar \Omega \Gamma}{2} \frac{s_{0}}{1+s_{0}} \tag{224}
\end{equation*}
$$

- At resonance $(\Delta=0)$, the two beams interact with the $v_{z}=0$ class. The saturation parameter is doubled (twice the intensity) but the atom number is twice lower (only one class). The absorbed energy is them

$$
\begin{equation*}
\mathcal{E}_{0}=\frac{\mathcal{N} \hbar \Omega \Gamma}{2} \frac{2 s_{0}}{1+2 s_{0}} \tag{225}
\end{equation*}
$$

[^0] distribution and the width of the velocity class.

## Steady-state and Saturation

## Saturation spectroscopy

Hence, the 'dip depth' is

$$
\begin{equation*}
\frac{\mathcal{E}_{0}}{\mathcal{E}}=\frac{1+s_{0}}{1+2 s_{0}} \tag{226}
\end{equation*}
$$

and its width is $\gamma^{\prime} \sqrt{1+s_{0}}$. The best compromise corresponds to $s_{0} \approx 1$, with a depth of $1 / 3$ and a width of $2 \gamma^{\prime}$ ).

## Steady-state and Saturation

## Saturation spectroscopy

Case of a multilevel atom: two nearly degenerate ground states, $|g\rangle$ and $|f\rangle$, and an excited state $|e\rangle$ ( $\Lambda$ system). Saturation resonances at:

- $\omega_{g e}$
- $\omega_{f e}$
- Crossover resonance dip: direct beam resonant at $\omega_{f e}$ for $k v_{z}=\omega-\omega_{f e}$, saturating the $f \rightarrow e$ transition, and reflected beam probing this saturation when resonant on $|g\rangle \rightarrow|e\rangle$ if $k v_{z}=-\left(\omega-\omega_{g e}\right)$ i.e.

$$
\begin{equation*}
\omega=\frac{\omega_{f e}+\omega_{g e}}{2} \tag{227}
\end{equation*}
$$

## Optical pumping

## Principle

$\Lambda$ system again. $|e\rangle$ decays towards both $|g\rangle$ and $|f\rangle$ with rates $\Gamma_{e g}$ and $\Gamma_{e f}$. Goal: populate only $|f\rangle$.
Method: drive selectively $|g\rangle \rightarrow|e\rangle$.
After a few fluorescence cycles, $|g\rangle$ is depopulated.
More quantitative approach based on Einstein's coefficients.

$$
\begin{align*}
\frac{d N_{e}}{d t} & =-\left(\Gamma_{e g}+\Gamma_{e f}\right) N_{e}+\frac{\Omega^{2}}{2 \gamma^{\prime}}\left(N_{g}-N_{e}\right)  \tag{228}\\
\frac{d N_{g}}{d t} & =\Gamma_{e g} N_{e}-\frac{\Omega^{2}}{2 \gamma^{\prime}}\left(N_{g}-N_{e}\right)  \tag{229}\\
\frac{d N_{f}}{d t} & =\Gamma_{e f} N_{e} \tag{230}
\end{align*}
$$

with $N=N_{e}+N_{f}+N_{g}$

## Optical pumping

## Dynamics

- Steady state: $N_{e}=0$ and hence $N_{g}=0$ and $N_{f}=N$.
- Puming dynamics. In the weak pump limit:

$$
\begin{equation*}
\frac{\Omega^{2}}{\gamma^{\prime}} \ll \Gamma_{e g}, \Gamma_{e f} \tag{231}
\end{equation*}
$$

$N_{e}$ is low and at any time at a steady state value

$$
\begin{gather*}
N_{e}=\frac{\Omega^{2} / 2 \gamma^{\prime}}{\Gamma_{e g}+\Gamma_{e f}+\Omega^{2} / 2 \gamma^{\prime}} N_{g} \approx \frac{\Omega^{2} / 2 \gamma^{\prime}}{\Gamma_{e g}+\Gamma_{e f}} N_{g}  \tag{232}\\
\frac{d N_{g}}{d t}=-\Gamma_{p} N_{g} \tag{233}
\end{gather*}
$$

where we define the optical pumping rate $\Gamma_{p}$ by:

$$
\begin{equation*}
\Gamma_{p}=\frac{\Gamma_{e f}}{\Gamma_{e g}+\Gamma_{e f}} \frac{\Omega^{2}}{2 \gamma^{\prime}} \tag{234}
\end{equation*}
$$

An exponential approach to the steady state.

## Dark resonances and EIT

## Dark states

Again $\Lambda$ system with two resonant laser fields $\mathbf{E}_{1}$ and $\mathbf{E}_{2}$ separately coupled to the $|g\rangle \rightarrow|e\rangle$ and $|f\rangle \rightarrow|e\rangle$ transitions (selection rules make $|g\rangle$ impervious to $E_{2}$ and $|f\rangle$ to $E_{1}$. The total interaction Hamiltonian is $H_{i}=-\mathbf{d} \cdot\left(\mathbf{E}_{1}+\mathbf{E}_{2}\right)$, where $\mathbf{d}$ is the dipole operator (back to the basics).
We set

$$
\begin{equation*}
d_{e g}=\langle e| \mathbf{d} \cdot \mathbf{E}_{1}|g\rangle ; \quad d_{e f}=\langle e| \mathbf{d} \cdot \mathbf{E}_{2}|f\rangle \tag{235}
\end{equation*}
$$

A state $\left|\Psi_{-}\right\rangle=c_{g}|g\rangle+c_{f}|f\rangle$ is decoupled from the lasers $\left(\langle e| H_{i}\left|\Psi_{-}\right\rangle=0\right)$ if $c_{g} d_{e g}+c_{f} d_{e f}=0$ i.e.

$$
\begin{equation*}
c_{g}=\frac{d_{\mathrm{ef}}}{d} \quad \text { and } \quad c_{f}=-\frac{d_{e g}}{d} \tag{236}
\end{equation*}
$$

or

$$
\begin{equation*}
\left|\Psi_{-}\right\rangle=\frac{d_{e f}}{d}|g\rangle-\frac{d_{e g}}{d}|f\rangle \tag{237}
\end{equation*}
$$

with

$$
\begin{equation*}
d=\sqrt{\left|d_{e g}\right|^{2}+\left|d_{e f}\right|^{2}} \tag{238}
\end{equation*}
$$

## Dark resonances and EIT

## Dark states

We have here written the states and the fields at a given time. This condition remains valid at all times if $|g\rangle$ and $|f\rangle$ are degenerate with lasers at resonance.
When $|g\rangle$ and $|f\rangle$ have different energies the dark state condition is maintained at any time if

$$
\begin{equation*}
\left|\Psi_{-}\right\rangle(t)=\frac{d_{e f}}{d} e^{-i \omega_{1} t} e^{-i \omega_{g} t}|g\rangle-\frac{d_{e g}}{d} e^{-i \omega_{2} t} e^{-i \omega_{f} t}|f\rangle \tag{239}
\end{equation*}
$$

is, within a global phase, independent of time i.e; if $\omega_{1}+\omega_{g}=\omega_{2}+\omega_{f}$ or

$$
\begin{equation*}
\omega_{1}-\omega_{2}=\omega_{f g} \tag{240}
\end{equation*}
$$

if the difference of the fields frequencies is equal to the Bohr frequency between the two ground states. This is nothing but a Raman resonance condition.
The first evidence of dark resonances has been obtained in 1976 by Gozzini and his group in Pisa

## Dark resonances and EIT

## Dark states

The orthogonal state $\left|\Psi_{+}\right\rangle$is maximally coupled to lasers:

$$
\begin{equation*}
\left|\Psi_{+}\right\rangle=\frac{d_{e g}^{*}}{d}|g\rangle+\frac{d_{e f}^{*}}{d}|f\rangle \tag{241}
\end{equation*}
$$

In presence of relaxation (spontaneous emission), we have one ground state coupled to the laser and another uncoupled. This is again an optical pumping situation. After a few emission cycles, we unconditionnaly end up in the dark state $\left|\Psi_{-}\right\rangle$. Fluorescence stops.

## Dark resonances and EIT

Electromagnetically induced transparency
$\Lambda$ system, strong drive of $|f\rangle \rightarrow|e\rangle$ ( $E$ at frequency $\omega$, Rabi frequency $\Omega$ ), weak probe of $|g\rangle \rightarrow|e\rangle\left(E^{\prime}, \omega^{\prime}, \Omega^{\prime}\right)$. Generalize OBES as:

$$
\begin{align*}
\frac{d \rho_{e e}}{d t} & =-\left(\Gamma_{e f}+\Gamma_{e g}\right) \rho_{e e}-\Omega \operatorname{lm} \rho_{e f}-\Omega^{\prime} \operatorname{Im} \rho_{e g}  \tag{242}\\
\frac{d \rho_{f f}}{d t} & =\Gamma_{e f} \rho_{e e}+\Omega \operatorname{lm} \rho_{e f}  \tag{243}\\
\frac{d \rho_{g g}}{d t} & =\Gamma_{e g} \rho_{e e}+\Omega^{\prime} \operatorname{lm} \rho_{e g}  \tag{244}\\
\frac{d \rho_{e f}}{d t} & =-\frac{\Gamma_{e f}}{2} \rho_{e f}+i \frac{\Omega}{2}\left(\rho_{e e}-\rho_{f f}\right)+i \frac{\Omega^{\prime}}{2} \rho_{g f}  \tag{245}\\
\frac{d \rho_{e g}}{d t} & =-\left(\frac{\Gamma_{e g}}{2}+i \Delta^{\prime}\right) \rho_{e g}+i \frac{\Omega^{\prime}}{2}\left(\rho_{g g}-\rho_{e e}\right)+i \frac{\Omega}{2} \rho_{f g}  \tag{246}\\
\frac{d \rho_{g f}}{d t} & =-i \Delta^{\prime} \rho_{g f}+i \frac{\Omega}{2} \rho_{g e}-i \frac{\Omega^{\prime}}{2} \rho_{e f} \tag{247}
\end{align*}
$$

## Dark resonances and EIT

Electromagnetically induced transparency

Simplify with $\Gamma_{e f}=\Gamma_{e g}=\Gamma$ and extract absorption of weak probe (absorbed energy $\mathcal{E}^{\prime}$, proportional to $\operatorname{Im} \rho_{\text {eg }}$ in the steady state).

$$
\begin{equation*}
\mathcal{E}^{\prime} \propto \frac{\Gamma^{2} \Delta^{\prime 2}}{\Gamma^{2} \Delta^{\prime 2}+4\left(\Delta^{\prime 2}-\Omega^{2} / 4\right)^{2}} \tag{248}
\end{equation*}
$$

## Dark resonances and EIT

Electromagnetically induced transparency



Absorption of the probe field as a function of the detuning $\Delta$ for $\Omega=10 \Gamma$ (left) or $\Omega=0.1 \Gamma$ (right). The unit of the horizontal axis is $\Gamma$.

## Dark resonances and EIT

Electromagnetically induced transparency

Slow light. In the small $\Omega$ case, the absorption has a narrow dip at resonance, whose width is only limited by the Rabi frequency. In actual experiments, the width can be only a few kHz . Hence, the (at resonance) transparent medium has an index of refraction $n$ for the weak probe field varying rapidly with the detuning $\Delta$ i.e. with $\omega^{\prime}$. Since

$$
\begin{equation*}
v_{g}=\frac{c}{n+\omega^{\prime} d n / d \omega^{\prime}} \tag{249}
\end{equation*}
$$

the light group velocity can be made very small, $17 \mathrm{~m} / \mathrm{s}$ in the original paper by Lene Hau and her group (Nature, 397, 594).

## Maxwell Bloch equations

Treat propagation in an atomic medium. Maxwell:

$$
\begin{align*}
\boldsymbol{\nabla} \times \mathbf{E} & =-\frac{\partial \mathbf{B}}{\partial t}  \tag{250}\\
\boldsymbol{\nabla} \cdot \mathbf{D} & =0  \tag{251}\\
\boldsymbol{\nabla} \cdot \mathbf{B} & =0  \tag{252}\\
\boldsymbol{\nabla} \times \mathbf{B} & =\mu_{0} \epsilon_{0} \frac{\partial \mathbf{E}}{\partial t}+\mu_{0} \frac{\partial \mathbf{P}}{\partial t} \tag{253}
\end{align*}
$$

Hence, for a transverse wave

$$
\begin{equation*}
\Delta \mathbf{E}-\frac{1}{c^{2}} \frac{\partial^{2} \mathbf{E}}{\partial t^{2}}=\mu_{0} \frac{\partial^{2} \mathbf{P}}{\partial t^{2}} \tag{254}
\end{equation*}
$$

(note that we assume a low density and treat the macroscopic fields as being the local ones)

## Maxwell-Bloch equations

Monochromatic plane wave in an isotropic medium

$$
\begin{equation*}
\mathbf{P}=P_{0}(z, t) e^{i(k z-\omega t)} \mathbf{u}_{x} \quad \text { and } \quad \mathbf{E}=E_{0}(z, t) e^{i(k z-\omega t)} \mathbf{u}_{x} \tag{255}
\end{equation*}
$$

Hence, noting

$$
\begin{equation*}
\frac{\partial P_{0}}{\partial t} \ll \omega P_{0} \tag{256}
\end{equation*}
$$

and neglecting the proper time derivatives

$$
\begin{equation*}
\frac{\partial E_{0}}{\partial z}+\frac{1}{c} \frac{\partial E_{0}}{\partial t}=i \frac{\mu_{0} \omega^{2}}{2 k} P_{0} \tag{257}
\end{equation*}
$$

with

$$
\begin{gather*}
\mathbf{P}_{0}=N \mathcal{D}=2 N d \rho_{e g}  \tag{258}\\
\frac{\partial E_{0}}{\partial z}+\frac{1}{c} \frac{\partial E_{0}}{\partial t}=i \frac{\omega N d}{\epsilon_{0} c} \rho_{e g} \tag{259}
\end{gather*}
$$

## Maxwell-Bloch equations

## Pulse propagation

A simple application: propagation in a relaxation-free medium. Atoms described by the angle $\phi(z, t)$ of the Bloch vector with the vertical axis. Assuming real quantities

$$
\begin{gather*}
\frac{d \phi(z, t)}{d t}=\frac{d E_{0}(z, t)}{\hbar}=\Omega(z, t)  \tag{260}\\
\rho_{\text {eg }}=-i \sin \frac{\phi}{2} \tag{261}
\end{gather*}
$$

Hence

$$
\begin{equation*}
\frac{\partial^{2} \phi}{\partial z \partial t}+\frac{1}{c} \frac{\partial^{2} \phi}{\partial t^{2}}=-\mu \sin \phi \tag{262}
\end{equation*}
$$

where

$$
\begin{equation*}
\mu=\frac{\omega N d^{2}}{2 \epsilon_{0} \hbar c} \tag{263}
\end{equation*}
$$

## Maxwell-Bloch equations

Pulse propagation

Using as independent variables $z$ and the 'retarded time' $\tau=t-z / c$ :

$$
\begin{equation*}
\frac{\partial^{2} \phi}{\partial z \partial \tau}-\mu \sin \phi \tag{264}
\end{equation*}
$$

Sine-Gordon equation


[^0]:    ${ }^{1}$ This quantity should be defined more carefully in terms of the Maxwell velocity

