UFS Lecture 8: Bloch Equations

Semi-classical Light Matter Interaction

- (1) Two-level system and Bloch equations
- (2) Rabi oscillation: coherent light-matter interaction
- (3) Steady-state solution of Bloch equations: linear susceptibility
- (4) Adiabatic solution of Bloch equations: laser rate equation

Population inversion for amplification

Amplification requirement:

$$N_2 > N_1$$



 N_i is the number density (also known as of molecules in state *i* (i.e., the number of molecules per cm³).

T is the temperature, and k is Boltzmann's constant = 1.3806503× 10⁻²³ J/^oK

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

Four-level system with optical pumping



Light-matter interaction: classical harmonic oscillator model

- 1) Light is modeled by Maxwell eqns.
- 2) Matter is modeled as harmonic oscillator.
- 3) Two sets of equations: Maxwell equations + dipole equation.
- 4) Explains linear optics (dispersion, absorption)



$$\begin{split} \widetilde{\underline{\chi}}(\Omega) &= \frac{\omega_p^2}{(\Omega_0^2 - \Omega^2) + 2j\Omega\frac{\Omega_0}{Q}} \\ \frac{\partial \underline{A}(z, t')}{\partial z} \Big|_{(gain)} &= g\left(1 + \frac{1}{\Omega_g^2}\frac{\partial^2}{\partial t^2}\right)\underline{A}(z, t') + \text{dispersion} \\ \mathbf{WHM} - \text{gain bandwidth} \end{split}$$

Light-matter interaction: semi-classical model

- Light is treated as a non-quantized, classical electromagnetic field (modeled by Maxwell equations).
- 2) Mater is quantized (resulting energy levels) by a non-relativistic quantum-mechanical approach.
- 3) Three sets of Eqns (Maxwell-Bloch Eqns):
 - E: Maxwell equations (pulse duration T)
 - w: population inversion (decay with time constant T_1)
 - d: dipole moment (decay with time constant T₂ dephasing time)
- 4) Explains Rabi oscillation, stimulated emission, absorption, etc.



Superposition states and radiative transitions

For energy eigen state:

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

$$\vec{p} = -e \, \langle \vec{r} \rangle \longrightarrow \mathbf{O}$$

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

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

$$\frac{1}{\sqrt{2}}\left(\psi_{100}(\vec{r},t) + \psi_{210}(\vec{r},t)\right) = \frac{1}{\sqrt{2\pi}\sqrt{r_1^3}} \left(e^{-r/r_1}e^{-jE_1t/\hbar} + \frac{1}{4\sqrt{2}}\frac{r}{r_1}e^{-r/2r_1}\cos\vartheta e^{-jE_2t/\hbar}\right)$$

In the probability density (i.e. the magnitude square of the wave function), the contributions between the ground state and excited state interfer positively or negatively depending on the relative phase between the two wave functions, which depends on the phase angle

$$\Delta E t/\hbar$$
, with $\Delta E = E_2 - E_1$.



The two-level model for light-matter interaction



1D-model for a two-level atom.

The two-level model for light-matter interaction

Hamiltonian of the atom: H_A

$$\mathbf{H}_A \ \psi_e(\vec{r}) = E_e \ \psi_e(\vec{r})$$
$$\mathbf{H}_A \ \psi_g(\vec{r}) = E_g \ \psi_g(\vec{r})$$

General state of this two dimensional quantum mechanical system is:

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

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

The time dependence of these coefficients follows from the Schrödinger Equation: $j \hbar \frac{\partial}{\partial t} \Psi(\vec{r}, t) = \mathbf{H}_A \Psi(\vec{r}, t)$

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

The two-level model for light-matter interaction

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

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

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

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

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

Light-matter interaction in dipole approximation

 $\vec{d} = -e_0 \vec{r}$. Dipole energy in $H_d = -\vec{d} \cdot \vec{E}(\vec{r}, t)$ E-field: **Dipole moment:** Position of electron

Dipole approximation:

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

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

Position of electron Position of atom

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

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

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

potential

Expected value of the dipole moment

$$\left\langle \vec{d} \right\rangle = \int_{-\infty}^{\infty} \Psi^*(\vec{r},t) \vec{d} \Psi(\vec{r},t) d\vec{r} \qquad \Psi(\vec{r},t) = c_g(t) \ \psi_g(\vec{r}) + c_e(t) \ \psi_e(\vec{r})$$
$$\left\langle \vec{d} \right\rangle = |c_e|^2 \vec{M}_{ee} + |c_g|^2 \vec{M}_{gg} + c_e^* c_g \vec{M}_{eg} + c_g^* c_e \vec{M}_{ge}$$

Matrix elements of the dipole moment:

$$\vec{M}_{ee} = \int \psi_e^*(\vec{r}) \ \vec{d} \ \psi_e(\vec{r}) \ d\vec{r} = -e_0 \int \psi_e^*(\vec{r}) \ \vec{r} \ \psi_e(\vec{r}) = \mathbf{0} \quad \underset{\text{symmetry}}{\text{symmetry}}$$
$$\vec{M}_{eg} = \int \psi_e^*(\vec{r}) \ \vec{d} \ \psi_g(\vec{r}) \ d\vec{r} = -e_0 \int \psi_e^*(\vec{r}) \ \vec{r} \ \psi_g(\vec{r}) \ d\vec{r}$$
$$\vec{M}_{ge} = \int \psi_g^*(\vec{r}) \ \vec{d} \ \psi_e(\vec{r}) \ d\vec{r} = \vec{M}_{eg}^*$$
$$\vec{M}_{gg} = \int \psi_g^*(\vec{r}) \ \vec{d} \ \psi_g(\vec{r}) \ d\vec{r} = -e_0 \int \psi_g^*(\vec{r}) \ \vec{r} \ \psi_g(\vec{r}) = \mathbf{0} \quad \underset{\text{symmetry}}{\text{symmetry}}$$
$$\vec{M}_{gg} = \int \psi_g^*(\vec{r}) \ \vec{d} \ \psi_g(\vec{r}) \ d\vec{r} = -e_0 \int \psi_g^*(\vec{r}) \ \vec{r} \ \psi_g(\vec{r}) = \mathbf{0} \quad \underset{\text{symmetry}}{\text{symmetry}}$$

Equations of motion for the probability amplitudes

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

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

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

New equations of motion for the probability amplitudes:

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

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

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

Monochromatic field:

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

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

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

Leads to:

$$\dot{C}_{e} = \left[j \left(\frac{\omega_{e} + \omega_{g} + \omega}{2} \right) - j \omega_{e} \right] c_{e} e^{j \left(\frac{\omega_{e} + \omega_{g} + \omega}{2} t \right)} + j c_{g} \frac{\vec{M}_{eg} \cdot \vec{e}}{\hbar} \vec{E}(t) e^{j \left(\frac{\omega_{e} + \omega_{g} + \omega}{2} t \right)} \\ \dot{C}_{g} = \left[j \left(\frac{\omega_{e} + \omega_{g} - \omega}{2} \right) - j \omega_{g} \right] c_{g} e^{j \left(\frac{\omega_{e} + \omega_{g} - \omega}{2} t \right)} + j c_{e} \frac{\vec{M}_{eg}^{*} \cdot \vec{e}}{\hbar} \vec{E}(t) e^{j \left(\frac{\omega_{e} + \omega_{g} - \omega}{2} t \right)}$$

Frequency detuning between atomic transition and electric field frequency:

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

Coupled mode equations:

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

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

If Rabi frequency is small:

$$egin{aligned} &|\Omega_r| << \omega_{eg} pprox \omega \ & extbf{Rotating wave approximation (RWA)} \ & extbf{applies:} & \ & \Omega_r pprox rac{ec{M}^*_{eg} \cdot ec{e}}{\hbar} \underline{E}_0 = const. \end{aligned}$$

Rabi Oscillation



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

 $C_g(0) = 1$ and $C_e(0) = 0$

Oscillation solution:

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

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

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

Rabi Oscillation

Expectation value of dipole moment:

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

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

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

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

<u>Where is our first order linear</u> susceptibility $\chi(\omega)$?



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

Motion eqns for dipole moment and population inversion

- Additional interactions with the environment cause the loss of coherence in the atomic system.
- These energy non-preserving processes cannot be easily included in the Schrödinger Equation.
- We can treat these processes phenomenologically in the equations of motion for the expectation values of the dipole moment and the population inversion.

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

Complex slowly varying dipole moment is defined as

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

$$\left\langle \vec{d} \right\rangle = c_e^* (\frac{|\Omega_r|}{2}t)$$

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

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

Motion eqns for dipole moment and population inversion

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

Applying the product rule we find

$$\frac{d}{dt}\underline{d} = \left(\frac{d}{dt}C_e^*\right)C_g + C_e^*\left(\frac{d}{dt}C_g\right)$$
$$= j\Delta C_e^*C_g - j\frac{\Omega_r}{2}C_g^*C_g + j\Delta C_e^*C_g + j\frac{\Omega_r}{2}C_e^*C_e$$
$$= j2\Delta \underline{d} + j\frac{\Omega_r}{2} \cdot w$$

And for inversion:

$$\frac{d}{dt}w = \left(\frac{d}{dt}C_e\right)C_e^* - \left(\frac{d}{dt}C_g\right)C_g^* + c.c.$$
$$= \left(-j\Delta C_e C_e^* + j\frac{\Omega_r^*}{2}C_g C_e^* - j\Delta C_g C_g^* - j\frac{\Omega_r}{2}C_e C_g^*\right) + c.c.$$
$$= +j\Omega_r^*\underline{d} + c.c$$

Decay of population inversion

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

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

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



Steady-state population:

negative at thermal equilibrium without pumping positive with pumping

Include both external EM field and energy decay:

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

Decay of polarization (dipole moment)

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

$$\frac{d}{dt}\underline{d} = j2\Delta\underline{d} + j\frac{\Omega_r}{2} \cdot w \longrightarrow \underline{d} = j(\omega_{eg} - \omega)\underline{d} + j\frac{\Omega_r}{2} w$$

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

- Energy decay of the two-level system
- Collisions with the host material disoriented the direction of dipoles, causing dephasing. The resulting polarization becomes zero, although the single dipole still exists.
- Dephasing can happen much faster than energy relaxation and is characterized by a time constant T₂.

Include both external EM field and polarization decay:

$$\underline{\dot{d}} = -(\underbrace{T_2}_{I_2} - j(\omega_{eg} - \omega))\underline{d} + j\frac{\Omega_r}{2} w$$
dephasing time

Bloch equations

$$\dot{w} = -\frac{w - w_0}{T_1} + j\Omega_r^* \underline{d} - j\Omega_r \underline{d}^*$$
$$\underline{\dot{d}} = -(\frac{1}{T_2} - j(\omega_{eg} - \omega))\underline{d} + j\frac{\Omega_r}{2} w$$
$$\Omega_r = \frac{\vec{M}_{eg}^* \cdot \vec{e}}{\hbar} \left(\underline{E}_0 + \underline{E}_0^* e^{-j2\omega t}\right)$$

- Bloch equations describe the dynamics of a statistical ensemble of two-level atoms interacting with a classical electric field.
- Polarization of the medium is related to the expectation value of the dipole moment of the atomic ensemble feeds into Maxwell equations, resulting in the Maxwell-Bloch Equations.

Steady-state solution

For moderate field strength E_0 , the magnitude of the Rabi-frequency is much smaller than the optical frequency, $|\Omega_r| << \omega$, the inversion and dipole moment do not change much within an optical cycle of the field.



If the optical pulse duration is longer than energy relaxation time constant T_1 , implying that the temporal variation of the EM field is slow than the energy decay, we can assume that population inversion and dipole moment are always at the steady-state though the steady state value adjust following the amplitude variation of the EM field.

$$\underline{\dot{d}} = \mathbf{0} \quad \dot{w} = \mathbf{0}$$

$$\underline{d}_{s} = \frac{j}{2\hbar} \frac{\left(\vec{M}_{eg}^{*} \cdot \vec{e}\right) w_{s}}{1/T_{2} + j(\omega - \omega_{eg})} \underline{E}_{0}$$

$$w_{s} = \frac{w_{0}}{1 + \frac{T_{1}}{\hbar^{2}} \frac{1/T_{2} |\vec{M}_{eg}^{*} \cdot \vec{e}|^{2}}{(1/T_{2})^{2} + (\omega_{eg} - \omega)^{2}} |\underline{E}_{0}|^{2}}$$

Inversion saturation

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

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

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

Intensity:

Unsaturated inversion

Steady state inversion:

$$\underbrace{w_s}_{s} = \frac{w_0}{1 + \frac{I}{I_s}L(\omega)}$$

Stationary inversion depends on the intensity of the incident light

Saturated inversion

Saturation intensity:

$$I_s = \left[\frac{2T_1T_2Z_F}{\hbar^2}|\vec{M}_{eg}^*\cdot\vec{e}|^2\right]^{-1}$$

Dielectric Susceptibility

Expectation value of the dipole moment $\left\langle \vec{d} \right\rangle = \vec{M}_{eg} \underline{d} \ e^{j\omega t} + c.c.$

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

$$\vec{P}(t) = \frac{1}{2} \left(\underline{\vec{P}}_{0} e^{j\omega t} + \underline{\vec{P}}_{0}^{*} e^{-j\omega t} \right) = N \vec{M}_{eg} \underline{d}_{s} e^{j\omega t} + c.c.$$
$$\vec{P}_{0} = 2N \vec{M}_{eg} \underline{d}_{s}$$

Definition of the complex susceptibility

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

Linear susceptibility of the medium

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

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

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

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

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

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

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

Linear susceptibility

If the incident EM field is weak

$$\frac{I}{I_s}L(\omega) \ll 1 \longrightarrow w_s \approx w_0$$

Linear susceptibility derived using semi-classical model

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

Linear susceptibility derived using classical harmonic oscillator model

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

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

Linear susceptibility

Real and imaginary part of the susceptibility

$$\chi(\omega) = \chi'(\omega) + j\chi''(\omega)$$

