Semiclassical Theory of Light Matter Interactions

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Outline

1. Atom-Field Hamiltonian
2. Two level atom
    1. Rabi Oscillations
    2. Density matrix
3. Maxwell-Schrodinger (Optical Bloch Equations)
4. Stimulated Emission and Absorption
5. *Semiclassical theory of Laser
6. *Time Dependent Perturbation Theory

Question ("Semi" in semiclassical?)

- *Quantize atom (observables $\leftrightarrow$ operators)*
- *Treat light classically ($E = E\cos(\nu t)$ $\leftarrow$ scalar field)*

Atom-Field Hamiltonian - Obtaining a Hamiltonian

- Start with free-electron Hamiltonian, $\mathcal{H}_0 = \frac{p^2}{2m}$
- Ask for local phase invariance:

$$\psi(\mathbf{r}, t) \text{ solution} \implies \psi(\mathbf{r}, t)e^{i\theta(\mathbf{r},t)} \text{ solution}$$

- Introduce the following Hamiltonian:

$$\mathcal{H} = \frac{1}{2m}(\,\mathbf{p} - i\frac{e}{c}\mathbf{A}(\mathbf{x}, t)\,)^2 + eU(\mathbf{r}, t)$$

with $(U, \mathbf{A})$ having the following transformation rules:

$$\mathbf{A}(\mathbf{r}, t) \rightarrow \mathbf{A}(\mathbf{r}, t) + \frac{\hbar}{e}\nabla\theta(\mathbf{r}, t)$$

$$U(\mathbf{r}, t) \rightarrow U(\mathbf{r}, t) - \frac{\hbar}{e}\frac{\partial\theta}{\partial t}(\mathbf{r}, t)$$

- Gauge-invariant, hence physical, quantities:

$$\mathbf{E} = -\nabla U - \frac{\partial \mathbf{A}}{\partial t}$$

$$\mathbf{B} = \nabla \times \mathbf{A}$$

Atom-Field Hamiltonian - Obtaining a Hamiltonian

- Local phase invariance:

$$\mathcal{H} = \frac{1}{2m}(\,\mathbf{p} - i\frac{e}{c}\mathbf{A}(\mathbf{r},t)\,)^2 + eU(\mathbf{r},t) + V(\mathbf{r})$$

- Transform $\mathcal{H}$ gauge transformation:

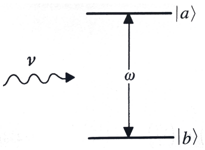
$$\psi(\mathbf{r},t) \mapsto \exp\left(-\frac{ie}{\hbar}\mathbf{A}(\mathbf{r}_0,t)\cdot\mathbf{r}\right)\psi(\mathbf{r},t)$$

$$\mathcal{H} = [\frac{\mathbf{p}^2}{2m} + V(\mathbf{r})] - e\mathbf{r}\cdot\mathbf{E}$$

Rabi Oscillations

Consider two-level atom in a monochromatic em-field:

$$|\psi(t)\rangle = C_a(t)\,|a\rangle + C_b(t)\,|b\rangle$$
$$H = H_0 - e\hat{\mathbf{r}} \cdot \mathbf{E}(\mathbf{r}_0, t)$$

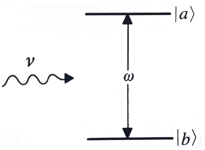


$$\begin{aligned} H_0 &= \hbar\omega_a\,|a\rangle\,\langle a| + \hbar\omega_b\,|b\rangle \\ H_1 &= -e\hat{x}\mathcal{E}\cos(\nu t) \\ &= -e\mathcal{E}\cos(\nu t)(\langle a|\hat{x}|b\rangle + \langle b|\hat{x}|a\rangle) \\ &= -\mathcal{E}\cos(\nu t)(\wp_{ab} + \wp_{ba}) \end{aligned}$$

$\mathcal{E}$ is the amplitude of the field - complex valued.
$\wp_{ab} = \langle a|e\hat{x}|b\rangle$ is the dipole moment.

Rabi Oscillations

$$\mathcal{H} = \begin{pmatrix} \hbar\omega_a & \wp_{ab}\mathcal{E}\cos(\nu t) \\ \wp_{ba}\mathcal{E}\cos(\nu t) & \hbar\omega_b \end{pmatrix}$$



Schrodinger equation becomes: $(\Omega_R = |\wp_{ba}|\mathcal{E}/\hbar, \; \wp_{ba} = |\wp_b a|e^{i\phi})$

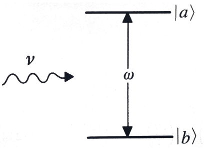
$$\dot{C}_a = -i\omega_a + i\Omega_R e^{-i\phi}\cos(\nu t) C_b$$
$$\dot{C}_b = -i\omega_b + i\Omega_R e^{i\phi}\cos(\nu t) C_a$$

- Transform $c_\mu := C_\mu e^{i\omega_\mu t}$ for $\mu = a, b$
- Ignore terms $\propto e^{\pm i(\omega+\nu)t} \leftarrow$ Rotating wave approximation (RWA)

$$\dot{c}_a = i\frac{\Omega_R}{2}e^{-i\phi}c_b\, e^{i(\omega-\nu)t}$$
$$\dot{c}_b = i\frac{\Omega_R}{2}e^{i\phi}c_a\, e^{i(\omega-\nu)t}$$

Rabi Oscillations - Solutions/Special cases

$$\mathcal{H} = \begin{pmatrix} \hbar\omega_a & \wp_{ab}\mathcal{E}\cos(\nu t) \\ \wp_{ba}\mathcal{E}\cos(\nu t) & \hbar\omega_b \end{pmatrix}$$



Solutions:

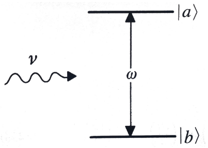
$$c_a(t) = \left(a_1 e^{i\Omega t/2} + a_2 e^{-i\Omega t/2}\right) e^{i\Delta\cdot t/2}$$

$$c_b(t) = \left(b_1 e^{i\Omega t/2} + b_2 e^{-i\Omega t/2}\right) e^{i\Delta\cdot t/2}$$

where $\Omega = \sqrt{\Omega_R^2 + (\omega - \nu)^2}$.

Rabi Oscillations - Solutions/Special cases

$$\mathcal{H} = \begin{pmatrix} \hbar\omega_a & \wp_{ab}\mathcal{E}\cos(\nu t) \\ \wp_{ba}\mathcal{E}\cos(\nu t) & \hbar\omega_b \end{pmatrix}$$

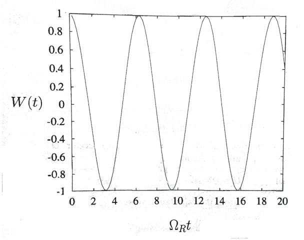


Special Case: detuning $\Delta = \omega - \nu = 0$ induce Rabi oscillations.

$$c_a(t) = \left[c_a(0)\cos\left(\frac{\Omega_R t}{2}\right) + ic_b(0)\sin\left(\frac{\Omega_R t}{2}\right)\right]$$
$$c_b(t) = \left[c_b(0)\cos\left(\frac{\Omega_R t}{2}\right) + ic_a(0)\sin\left(\frac{\Omega_R t}{2}\right)\right]$$

Inversion:

$$W(t) = |c_a(t)|^2 - |c_b(t)|^2$$



Polarization - expectation of dipole-moment operator:

$$\begin{aligned} P(t) &= e \langle \psi(t) | \hat{r} | \psi(t) \rangle \\ &= c_a^* c_b \wp_{ab} e^{i\omega t} + c.c. \end{aligned}$$

Optical Bloch Equations

Density matrices, ρ, characterized by

- $\text{Tr}(\rho) = 1$,
- $\rho = \rho^\dagger$,
- $\langle\psi|\rho|\psi\rangle \geq 0$ for all $|\psi\rangle$.

Von-Neumann equation of motion:

$$i\hbar\dot{\rho} = [\mathcal{H}, \rho]$$

In two level atom:

$$\rho = \begin{pmatrix} \rho_{aa} & \rho_{ab} \\ \rho_{ba} & \rho_{bb} \end{pmatrix}$$

- Diagonal elements correspond to populations of atoms
- Off-diagonal elements, called *coherences*, correspond to response at driving frequency

Optical Bloch Equations - with noise

**Spontaneous emission is taken into account by introducing a dissipative matrix, Γ, with matrix element:

$$\langle n|\Gamma|m\rangle = \gamma_n \delta_{nm}$$

The equation of motion becomes:

$$i\hbar\dot{\rho} = [\mathcal{H}, \rho] - \frac{i\hbar}{2}(\Gamma\rho + \rho\Gamma)$$

In component form (Optical Bloch Equations):

$$\dot{\rho}_{aa} = -\gamma_1\rho_{aa} + \frac{i}{\hbar}[\wp_{ab}E\rho_{ba} - cc]$$

$$\dot{\rho}_{bb} = -\gamma_2\rho_{bb} - \frac{i}{\hbar}[\wp_{ab}E\rho_{ba} - cc]$$

$$\dot{\rho}_{ab} = -(i\omega + \gamma_{ab})\rho_{ab} - \frac{i}{\hbar}\wp_{ab}E(\rho_{aa} - \rho_{bb})$$

Formulation allows for more effects!

Maxwell-Schrodinger

Slowly working towards a theory of the laser...

- Atoms pumped to excited state
- Interact with EM-field

Maxwell-Schrodinger – Population Matrix

- Atoms will not interact with field until pumped to excited state

$$\rho(z, t, t_0) - \text{pumped at } t_0$$

- $r_a(z, t_0)$ – pumping rate (atoms per second per volume)
- Population matrix – the density matrix of all the atoms in a cavity

$$\rho(z, t) = \int_{-\infty}^{t} dt_0 \, r_a(z, t_0) \rho(z, t, t_0)$$

$$\rho(z,t) = \int_{-\infty}^{t} dt_0\, r_a(z,t_0)\rho(z,t,t_0)$$
$$= \sum_{\alpha,\beta} \int_{-\infty}^{t} dt_0\, r_a(z,t_0)\rho_{\alpha,\beta}(z,t,t_0)\,|\alpha\rangle\,\langle\beta|$$

Macroscopic polarization - crucial to tie in with Maxwell's equations:

$$P(z,t) = \mathrm{Tr}(\,\hat{\wp}\rho(z,t)\,) = \wp_{ab}\rho_{ab}(z,t) + cc$$

Differentiating,

$$\rho(z,t) = \sum_{\alpha,\beta} \int_{-\infty}^{t} dt_0 \, r_a(z,t_0)\rho_{\alpha,\beta}(z,t,t_0) \left|\alpha\right\rangle \left\langle\beta\right|$$

We get the "Schrodinger" part of Maxwell-Schrodinger:

$$\dot{\rho}_{aa} = \lambda_a - \gamma_a \rho_{aa} + \frac{i}{\hbar}[\wp_{ab} E \rho_{ba} - cc]$$

$$\dot{\rho}_{bb} = \lambda_b - \gamma_b \rho_{bb} - \frac{i}{\hbar}[\wp_{ab} E \rho_{ba} - cc]$$

$$\dot{\rho}_{ab} = -(i\omega + \gamma)\rho_{ab} - \frac{i}{\hbar}\wp_{ab} E(\rho_{aa} - \rho_{bb})$$

where $\lambda_a = r_a \rho_{aa}^{(0)}$, $\lambda_b = r_a \rho_{bb}^{(0)}$.

Maxwell's Equations:

$$\nabla \cdot \mathbf{D} = 0 \qquad \nabla \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t}$$

$$\nabla \cdot \mathbf{B} = 0 \qquad \nabla \times \mathbf{H} = \mathbf{J} + \frac{\partial \mathbf{D}}{\partial t},$$

where

$$\mathbf{D} = \epsilon_0 \mathbf{E} + \mathbf{P}, \qquad \mathbf{B} = \mu_0 \mathbf{H}, \qquad \mathbf{J} = \sigma \mathbf{E}$$

Then:

$$\nabla \times (\nabla \times \mathbf{E}) + \mu_0 \sigma \frac{\partial E}{\partial t} + \mu_0 \epsilon_0 \frac{\partial^2 \mathbf{E}}{\partial t^2} = -\mu_0 \frac{\partial^2 \mathbf{P}}{\partial t^2}$$

Assume, unidirectional electric field inside cavity, $\mathbf{E}(\mathbf{r}, t) = E(z, t)\hat{\mathbf{x}}$

$$E(z,t) = \frac{1}{2}\mathscr{E}(z,t)e^{-i[\nu t - kz + \phi(z,t)]} + cc$$
$$P(z,t) = \frac{1}{2}\mathscr{P}(z,t)e^{-i[\nu t - kz + \phi(z,t)]} + cc$$

where $\mathscr{E}, \mathscr{P}, \phi$ are all slowly-varying functions.

$$\mathscr{P}(z,t) := 2\wp\rho_{ab}e^{i[\nu t - kz + \phi(z,t)]}$$

Now, we find equations of motion for $\mathcal{E}, \phi$.

$$-\frac{\partial^2 E}{\partial z^2} + \mu_0 \sigma \frac{\partial E}{\partial t} + \frac{1}{c^2}\frac{\partial^2 E}{\partial t^2} = -\mu_0 \frac{\partial^2 P}{\partial t^2}$$

We arrive at two equations that describe $\mathscr{E}, \phi$:

$$\frac{\partial \mathscr{E}}{\partial z} + \frac{1}{c}\frac{\partial \mathscr{E}}{\partial t} = -\kappa \mathscr{E} - \frac{1}{2\epsilon_0} k \operatorname{Im} \mathscr{P}$$
$$\frac{\partial \phi}{\partial z} + \frac{1}{c}\frac{\partial \phi}{\partial t} = k - \frac{\nu}{c} - \frac{1}{2\epsilon_0} k \mathscr{E}^{-1} \operatorname{Re} \mathscr{P}$$

Self-consistent: E driven by population-matrix elements [1]

[1] PRA 81 033833

Stimulated emission and absorption

Suppose $\Delta = \omega - \nu = 0$, $c_a(0) = 1, c_b(0) = 0$; solution becomes:

$$c_a(t) = \cos\left(\frac{\Omega_R t}{2}\right) \approx 1$$
$$c_b(t) = i\sin\left(\frac{\Omega_R t}{2}\right) \approx i\frac{\Omega_R t}{2}$$

Polarization

$$\mathscr{P} = 2\wp\rho_{ab}e^{i\nu t} \approx -i\wp\Omega_R t$$

Maxwell's Equation gives:

$$\frac{1}{c}\frac{\partial\mathscr{E}}{\partial t} = \frac{k}{2\epsilon_0}\frac{\wp^2}{\hbar}\mathscr{E}t^2 \implies \Delta\mathscr{E} \approx \frac{ck}{2\epsilon_0}\frac{\wp^2}{\hbar}\mathscr{E}t^2$$

If $c_a(0) = 0, c_b(0) = 1$ then $\Delta\mathscr{E} \approx -\frac{ck}{2\epsilon_0}\frac{\wp^2}{\hbar}\mathscr{E}t^2$

Appendix

$$\nabla \times (\nabla \times \mathbf{E}) + \mu_0 \sigma \frac{\partial E}{\partial t} + \mu_0 \epsilon_0 \frac{\partial^2 \mathbf{E}}{\partial t^2} = -\mu_0 \frac{\partial^2 \mathbf{P}}{\partial t^2}$$
$$\implies -\frac{\partial^2 E}{\partial z^2} + \mu_0 \sigma \frac{\partial E}{\partial t} + \frac{1}{c^2}\frac{\partial^2 E}{\partial t^2} = -\mu_0 \frac{\partial^2 P}{\partial t^2}$$

$$\implies \left(\frac{\partial}{\partial z} + \frac{1}{c}\frac{\partial}{\partial t}\right)\underbrace{\left(-\frac{\partial}{\partial z} + \frac{1}{c}\frac{\partial}{\partial t}\right)}_{\simeq -2ikE} E = -\mu_0 \sigma \frac{\partial E}{\partial t} - \mu_0 \frac{\partial^2 P}{\partial t^2}$$

We arrive at two equations that describe $\mathscr{E}, \phi$:

$$\frac{\partial \mathscr{E}}{\partial z} + \frac{1}{c}\frac{\partial \mathscr{E}}{\partial t} = -\kappa \mathscr{E} - \frac{1}{2\epsilon_0} k \operatorname{Im} \mathscr{P}$$
$$\frac{\partial \phi}{\partial z} + \frac{1}{c}\frac{\partial \phi}{\partial t} = k - \frac{\nu}{c} - \frac{1}{2\epsilon_0} k \mathscr{E}^{-1} \operatorname{Re} \mathscr{P}$$

1. Integrate off-diagonal Schrodinger equation to obtain $\mathscr{P}(z,t)$

$$\dot{\rho}_{ab} = -(i\omega + \gamma)\rho_{ab} - \frac{i}{\hbar}\wp E(\rho_{aa} - \rho_{bb})$$

2. Substitute into the other Schrodinger equation:

$$\dot{\rho}_{aa} = \lambda_a - \gamma_a\rho_{aa} - R(\rho_{aa} - \rho_{bb})$$
$$\dot{\rho}_{bb} = \lambda_b - \gamma_b\rho_{bb} + R(\rho_{aa} - \rho_{bb})$$

   where $R = \frac{1}{2}\left(\frac{\wp\mathscr{E}}{\hbar}\right)^2 \frac{\gamma}{\gamma^2+(\omega-\nu)^2}$.

3. Solve for $\rho_{aa} - \rho_{bb}$ in steady state, obtain $\mathscr{P}$
4. Introduce dimensionless intensity (“photon number”)

$$n = \frac{\epsilon_0\mathscr{E}^2 V}{2\hbar\nu}$$

5. Rewrite Maxwell’s equations

Time-Dependent Perturbation

Let $H = H_0 + \lambda V(t)$, so that H_0 is time-independent and $\lambda V(t) \ll 1$.

In the interaction picture, time dependence only due to interaction:

$$|\psi(t)\rangle_D = U_0 |\psi(t)\rangle$$
$$A(t)_D = U_0 A(t) U_0^\dagger$$

we have the following evolution:

$$i \frac{d}{dt} |\psi(t)\rangle_D = \lambda \hat{V}_D |\psi(t)\rangle_D$$

Time-Dependent Perturbation

$H = H_0 + \lambda V(t)$

$$|\psi(t)\rangle_D = U_0 |\psi(t)\rangle$$
$$A(t)_D = U_0 A(t) U_0^\dagger$$
$$i\frac{d}{dt}|\psi(t)\rangle_D = \lambda \hat{V}_D |\psi(t)\rangle_D$$

Take $U_D(t, t_0)$ so that $|\psi(t)\rangle_D = U_D(t, t_0)|\psi(t_0)\rangle_D$ then:

$$\frac{d}{dt}U_D(t, t_0) = -i\lambda V_D(t) U_D(t, t_0)$$

Integrating,

$$U_D(t, t_0) = \mathbb{1} - i\lambda \int_{t_0}^{t} d\tau V_D(\tau) U_D(\tau, t_0)$$

$$\begin{aligned} U_D(t, t_0) &= \mathbb{1} - i\lambda \int_{t_0}^{t} d\tau_1 V_D(\tau_1)\left(\mathbb{1} - i\lambda \int_{t_0}^{\tau_1} d\tau_2 V_D(\tau_2)(\cdots)\right) \\ &= \mathbb{1} + U_D^{(1)}(t, t_0) + U_D^{(2)}(t, t_0) + \cdots \end{aligned}$$