

Classical Light-Matter Interaction

Absorption and Dispersion in Gases

Approximations:

$$\begin{aligned} |\omega_0 - \omega| &\ll \omega_0, \omega && \text{near resonance} \\ |n(\omega)| &\sim 1 && \text{weakly polarizable} \end{aligned}$$

Let $\omega_0^2 - \omega^2 = (\omega_0 + \omega)(\omega_0 - \omega) \approx 2\omega(\omega_0 - \omega)$



$$\begin{aligned} \alpha(\omega) &= \frac{e^2/m}{\omega_0^2 - \omega^2 - 2i\beta\omega} = \frac{e^2/2m\omega}{\omega_0 - \omega - i\beta} \\ &= \frac{e^2}{2m\omega} \frac{\omega_0 - \omega + i\beta}{(\omega_0 - \omega)^2 + \beta^2} \end{aligned}$$

Furthermore

$$n(\omega)^2 = 1 + \frac{N\alpha(\omega)}{\epsilon_0} = 1 + \epsilon, \epsilon \ll 1$$

Expand to 1st order $(1 + \epsilon)^{1/2} \approx 1 + \epsilon/2$

Putting it together

$$n_R(\omega) = 1 + \frac{Ne^2}{4\epsilon_0 m \omega} \frac{\omega_0 - \omega}{(\omega_0 - \omega)^2 + \beta^2}$$

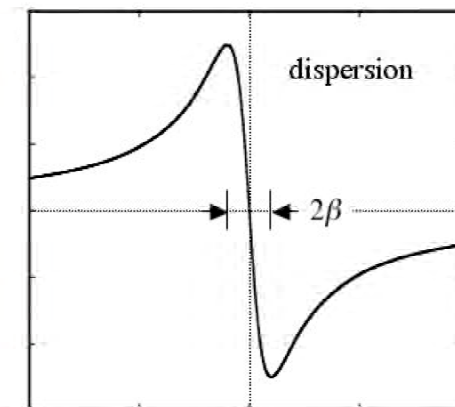
dispersive line shape

$$n_I(\omega) = \frac{Ne^2}{4\epsilon_0 m \omega} \frac{\beta}{(\omega_0 - \omega)^2 + \beta^2}$$

Lorentzian line shape

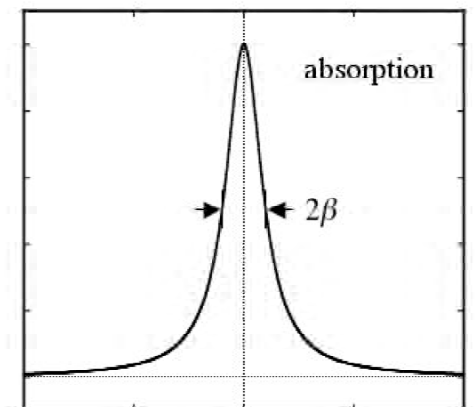
General behavior:

$n_R(\omega)$



$(\omega - \omega_0)/\beta$

$n_I(\omega)$



$(\omega - \omega_0)/\beta$

Classical Light-Matter Interaction

Putting it together

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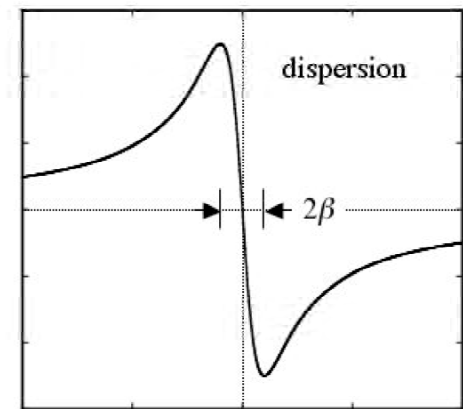
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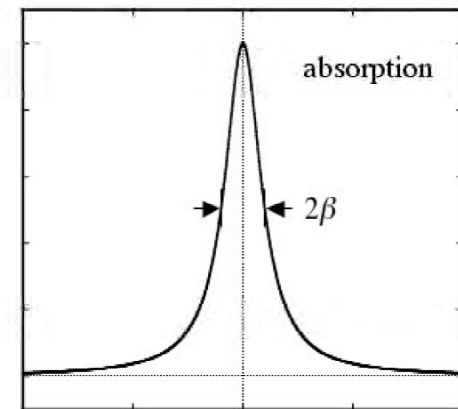
General behavior:

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$(\omega - \omega_0)/\beta$

$n_I(\omega)$



$(\omega - \omega_0)/\beta$

Note:

for

$$|\omega_0 - \omega| \gg \beta$$

$$\text{dispersion} \propto \frac{1}{(\omega_0 - \omega)}$$

$$\text{absorption} \propto \frac{1}{(\omega_0 - \omega)^2}$$



We can have loss-less dispersive media

Note: If we introduce the detuning $\Delta = (\omega_0 - \omega)$ we can rewrite $n_R(\omega)$, $n_I(\omega)$ as

$$n_R(\Delta) = 1 + \frac{Ne^2}{4\epsilon_0 m \omega} \frac{\Delta}{\Delta^2 + \beta^2}$$

$$n_I(\Delta) = \frac{Ne^2}{4\epsilon_0 m \omega} \frac{\beta}{\Delta^2 + \beta^2}$$

From the above we see that

$$n_R(\omega) < 1 \text{ for } \omega > \omega_0 \Rightarrow \frac{c}{n_R(\omega)} \gg c$$

Superluminal propagation?

Classical Light-Matter Interaction

dispersion $\propto \frac{1}{(\omega_0 - \omega)}$ for $|\omega_0 - \omega| \gg \beta$

Note: absorption $\propto \frac{1}{(\omega_0 - \omega)^2}$

➡ We can have loss-less dispersive media

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$n_R(\omega) < 1$ for $\omega > \omega_0$ ➡ $\frac{c}{n_R(\omega)} \gg c$

Superluminal propagation?

Free Electrons

Consider the limit $\omega \gg \omega_0$

➡ effectively unbound electrons

This is a reasonable model of plasmas & metals

In this limit we have

$$\alpha(\omega) = \frac{e^2/m}{\omega_0^2 - \omega^2 - 2i\beta\omega} \approx -\frac{e^2}{m\omega} \quad \rightarrow$$

$$n(\omega) = \sqrt{1 + \frac{N(\alpha)}{\epsilon_0}} \approx \sqrt{1 - \frac{Ne^2}{\epsilon_0 m \omega^2}} \equiv \sqrt{1 - \frac{\omega_p^2}{\omega^2}}$$

We introduce the Plasma Frequency :

$$\omega_p = \sqrt{\frac{Ne^2}{\epsilon_0 m}}$$

Classical Light-Matter Interaction

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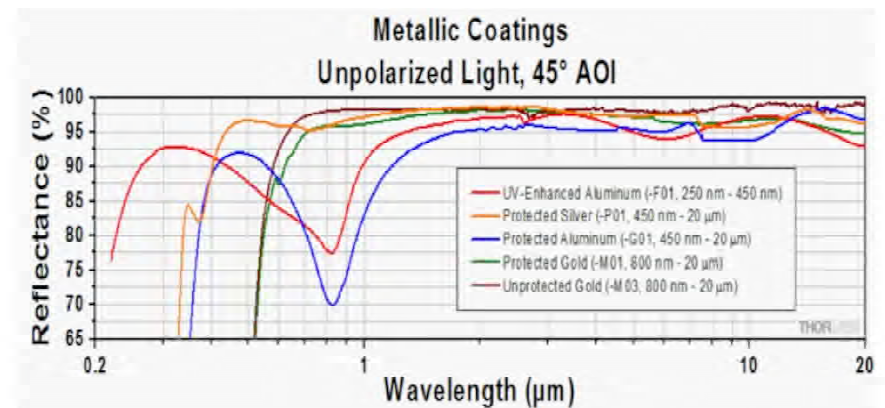
Let $\left. \begin{array}{l} \omega_0 \ll \omega \ll \omega_p \\ |\omega_0 - \omega| \gg \beta \end{array} \right\} \Rightarrow n(\omega) \text{ purely imaginary} \\ \text{- but no loss !}$

We now have

$$\begin{aligned} \vec{E}(z,t) &= \vec{E}_0 e^{-i\omega[t - n(\omega)z/c]} \\ &= \vec{E}_0 e^{-i\omega t} e^{i(z/c)\sqrt{\omega^2 - \omega_p^2}} \\ &= \vec{E}_0 e^{-i\omega t} e^{-b(\omega)z} \end{aligned}$$

where $b(\omega) = -\frac{i}{c} \sqrt{\omega^2 - \omega_p^2}$

Reflection at surface, penetration depth $\sim 1/b(\omega)$



Quantum Theory of Light-Matter Interaction

Quantum Theory of Light-Matter Interaction

Completed:

- Fully classical description of fields & Atoms

Next Step:

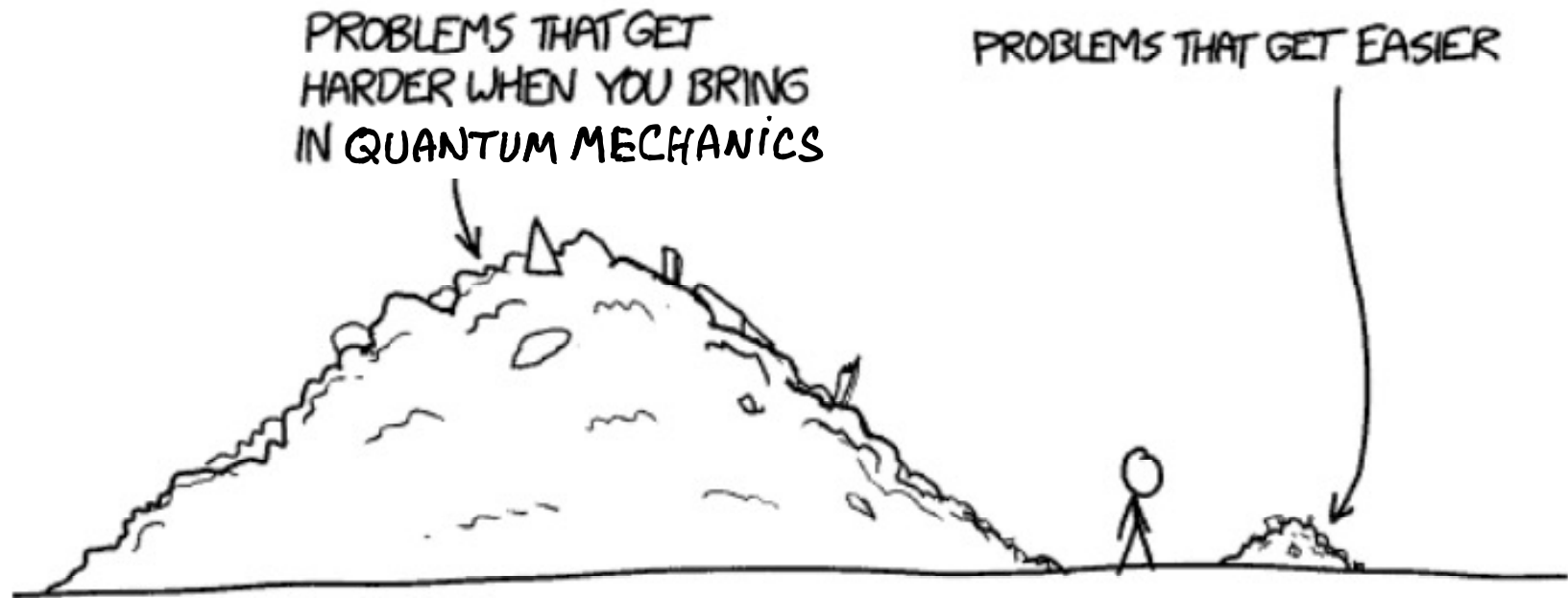
- Semiclassical description
 - { Classical field
 - { Quantum atoms

Self-Consistent Description

Electromagnetic Field → Atom/Molecule/Solid



Quantum Theory of Light-Matter Interaction



Source: xkcd.com

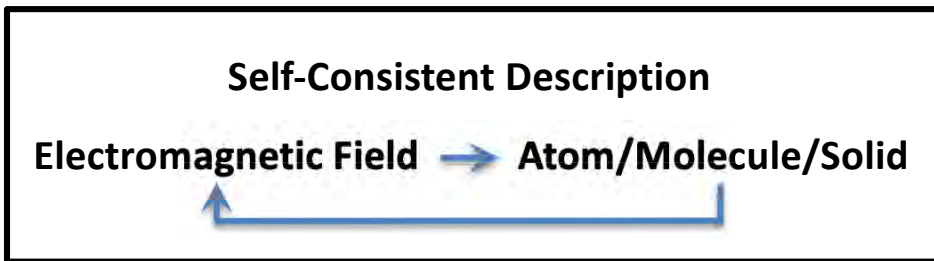
Quantum Theory of Light-Matter Interaction

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Needed: Quantum theory of atomic response
analogous to classical $\vec{p} = \alpha(\omega) \vec{E}$

Note: In QM the dipole is an Observable
Observable = Hermitian operator \hat{p}
Classical Field = C-valued vector \vec{E}

Cannot plug into Wave Eq. for classical field!

Wave Equation w/classical field & atoms

$$\left(\nabla^2 - \frac{1}{c^2} \frac{\partial^2}{\partial t^2}\right) \vec{E} = \frac{1}{\epsilon_0 c^2} \frac{\partial^2}{\partial t^2} \vec{P}, \quad \vec{P} = N \vec{p}$$

How do we solve the mismatch?

Repeated measurements of $\vec{p}(t)$



Quantum fluctuations $\vec{p}(t) = \langle \vec{p}(t) \rangle + \Delta \vec{p}(t)$

where $\langle \vec{p}(t) \rangle = \langle \psi(t) | \hat{p} | \psi(t) \rangle$ mean
 $\Delta \hat{p}(t)$ fluctuations

Note: Given $|\psi(t=0)\rangle$ and \vec{E} , the mean $\langle \vec{p}(t) \rangle$
follows from the Schrödinger Eq.,
radiates coherently like classical $\vec{p}(t)$

$\langle \vec{p}(t) \rangle$ is a Real-valued vector (more later)
→ we can plug it into the Wave Eq.

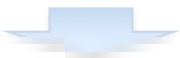
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Wave Eq. w/classical field & quantum atoms

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Note: - The Equations look very similar

- Polarizability, index of refraction, etc will be *very different* in some regimes
- Notably, the model is no longer linear in \vec{E} and will lead to phenomena like saturation and wave mixing
- $\Delta \vec{p}(t)$ represents quantum fluctuations driven by the empty modes of the EM field, a process also responsible for spontaneous decay.

Note: Do not identify $\langle \vec{p} \rangle$ and $\Delta \vec{p}$ with Stimulated and spontaneous emission. Those labels are not meaningful here.

Quantum Theory of Light-Matter Interaction

Wave Eq. w/classical field & quantum atoms

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Atom-field interaction

Hamiltonian:

$$H = H_a + V_{\text{ext}}(\vec{R}, t)$$

H_a : time-independent atomic Hamiltonian

V_{ext} : time-dependent driving term, non necessarily a perturbation

Question: Time evolution of the atomic system?
Is there a steady state?

Schrödinger Eq.:

$$i\hbar \frac{\partial}{\partial t} |\psi(t)\rangle = H |\psi(t)\rangle$$

Expand in basis $\{|\varphi_n\rangle\}$ of eigenstates of H_a

$$|\psi(t)\rangle = \sum_n a_n(t) |\varphi_n\rangle, \quad H_a |\varphi_n\rangle = E_n |\varphi_n\rangle$$

Quantum Theory of Light-Matter Interaction

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Plug into S. E. 


$$i\hbar \sum_n \dot{a}_n(t) |\varphi_n\rangle = \sum_n a_n(t) [E_n + V_{\text{ext}}] |\varphi_n\rangle$$

Take scalar product w/ $|\varphi_m\rangle$ on both sides 

$$\begin{aligned} i\hbar \sum_n \dot{a}_n(t) \langle \varphi_m | \varphi_n \rangle &= \sum_n a_n(t) [E_n \langle \varphi_m | \varphi_n \rangle + \underbrace{\langle \varphi_m | V_{\text{ext}} | \varphi_n \rangle}_{V_{mn}}] \\ &= \sum_n a_n(t) [E_n \delta_{mn} + V_{mn}] \end{aligned}$$

On vector-matrix form this can be written

$$i\hbar \dot{\underline{a}} = \underline{H}_a \underline{a} + \underline{V} \underline{a}$$

 S.E. in $\{|\varphi_n\rangle\}$ rep.
Still exact!

Quantum Theory of Light-Matter Interaction

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Note: If \underline{H}_a and $\underline{V}_{\text{ext}}$ are known we can do

- Perturbation Theory
(OK for short times or “weak” driving fields)
- Numerical integration of the S. E.
- Few-level approximations to simplify and obtain analytical solutions outside the perturbative regime

**General problem:
No analytical solution !**

Quantum Theory of Light-Matter Interaction

Atom-Light Interaction: 2-Level Approximation

Begin
01-25-2023

General observation:

- Atoms and molecules often behave as if they have a single, dominant transition frequency
- We expect this when the freq. of the driving is resonant with one transition $|q_n\rangle \rightarrow |q_m\rangle$ and far off resonance with all others.

Interaction

$$V_{ext} = -\hat{\vec{p}} \cdot \vec{E}(t)$$

State space

$$\text{Dim}(\mathcal{E}) = 2, \{ |1\rangle, |2\rangle \}$$

$\xrightarrow{\hbar\omega_{21}} |2\rangle$
 $\xleftarrow{\hbar\omega_{21}} |1\rangle$

State vector

$$|\psi(t)\rangle = a_1(t)|1\rangle + a_2(t)|2\rangle$$

Schröd. eq.

$$\begin{aligned} i\hbar\dot{a}_1 &= E_1 a_1 + V_{11} a_1 + V_{12} a_2 \\ i\hbar\dot{a}_2 &= E_2 a_2 + V_{21} a_1 + V_{22} a_2 \end{aligned}$$

Interaction

$$\begin{aligned} V_{12}(t) &= -\vec{p}_{12} \cdot \frac{1}{2} (\hat{E} E_0 e^{-i\omega t} + \text{c.c.}) \\ V_{21}(t) &= -\vec{p}_{21} \cdot \frac{1}{2} (\hat{E} E_0 e^{-i\omega t} + \text{c.c.}) \end{aligned}$$

Parity selection rule

Definition: $\vec{r} \rightarrow -\vec{r}$ is a reflection through the origin

Atomic Hamiltonian $H \propto \frac{1}{r} \Rightarrow H(\vec{r}) = H(-\vec{r})$

Eigenstates $\varphi(\vec{r}) = \varphi(-\vec{r}) = \pm \varphi(\vec{r})$

\uparrow two reflections equals the identity
 \uparrow "+" = even parity
 \uparrow "-" = odd parity

The dipole $\hat{\vec{p}}$ is a vector operator

transforms like a vector when $\vec{r} \rightarrow -\vec{r}$

Thus $\hat{\vec{p}}(\vec{r}) = e^{\vec{r}} = -\hat{\vec{p}}(-\vec{r})$ and

$$\hat{p}_{nm} = \int d^3r \varphi_n^*(\vec{r}) \hat{\vec{p}} \varphi_m(\vec{r}) \neq 0 \text{ only when}$$

φ_n and φ_m have opposite parity

Parity rule:

No dipole moment in energy eigenstate!

$$\begin{aligned} \vec{p}_{12} &= \langle 1 | \hat{\vec{p}} | 2 \rangle, \quad \vec{p}_{21} = \vec{p}_{12}^* \\ \vec{p}_{11} &= \vec{p}_{22} = 0 \Rightarrow V_{11} = V_{22} = 0 \end{aligned}$$

Atom-Light Interaction: 2-Level Approximation

We define

$$\omega_{21} = \frac{E_2 - E_1}{\hbar}, \quad E_1 = 0$$

$$\left. \begin{aligned} \chi_{12} &= \vec{p}_{12} \cdot \hat{E} E_0 / \hbar \\ \chi_{21} &= \vec{p}_{21} \cdot \hat{E} E_0 / \hbar \end{aligned} \right\} \begin{array}{l} \text{interaction energy is } \hbar \chi \\ \chi \text{ Rabi frequency} \end{array}$$

Note:
$$\left\{ \begin{aligned} \chi_{12}^* &= \vec{p}_{21} \cdot (\hat{E} E_0 / \hbar)^* \neq \chi_{21} \\ \chi_{21}^* &= \vec{p}_{12} \cdot (\hat{E} E_0 / \hbar)^* \neq \chi_{12} \end{aligned} \right.$$

Plug into $i\hbar \dot{\underline{a}} = \underline{H}_a \underline{a} + \underline{V} \underline{a}$ (S. E.) to get

$$\begin{aligned} i\dot{a}_1 &= -\frac{1}{2} (\chi_{12} e^{-i\omega t} + \chi_{21}^* e^{i\omega t}) a_2 \\ i\dot{a}_2 &= \omega_{21} a_2 - \frac{1}{2} (\chi_{21} e^{-i\omega t} + \chi_{12}^* e^{i\omega t}) a_1 \end{aligned}$$

Switch to rotating frame (slow variables)

$$C_1(t) = a_1(t), \quad C_2(t) = a_2(t) e^{i\omega t}$$



$$\begin{aligned} i\dot{C}_1(t) &= -\frac{1}{2} (\chi_{12} e^{-i2\omega t} + \chi_{21}^*) C_2(t) \\ i\dot{C}_2(t) &= (\omega_{21} - \omega) C_2(t) - \frac{1}{2} (\chi_{21} + \chi_{12}^* e^{i2\omega t}) C_1(t) \end{aligned}$$

Rotating Wave Approximation (RWA)

Very important, equivalent to resonant approximation

Terms $\propto e^{\pm i2\omega t}$ average to zero on time scale for variations in C_1, C_2



$$\begin{aligned} i\dot{C}_1(t) &= -\frac{1}{2} \chi_{21}^* C_2(t) \\ i\dot{C}_2(t) &= \Delta C_2(t) - \frac{1}{2} \chi_{21} C_1(t) \end{aligned} \quad \begin{array}{l} \Delta = \omega_{21} - \omega \\ \text{(detuning)} \end{array}$$

Exactly Solvable !

Atom-Light Interaction: 2-Level Approximation

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Exactly Solvable !

To simplify, make a global phase choice such that

$$\chi_{21} = \vec{p}_{21} \cdot \hat{E}_0 / \hbar = \chi \text{ is real (not required)}$$



Simplest 2-level equations

$$\begin{aligned} i\dot{C}_1(t) &= -\frac{1}{2} \chi C_2(t) \\ i\dot{C}_2(t) &= \Delta C_2(t) - \frac{1}{2} \chi C_1(t) \end{aligned}$$

Rabi Solutions for $C_1(0) = 1, C_2(0) = 0$

$$C_1(t) = \left(\cos \frac{\Omega t}{2} + i \frac{\Delta}{\Omega} \sin \frac{\Omega t}{2} \right) e^{-i\Delta t/2}$$

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χ : Rabi freq. Δ : Detuning

$\Omega \equiv \sqrt{\chi^2 + \Delta^2}$: Generalized Rabi freq.

Atom-Light Interaction: 2-Level Approximation

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Note: The Rabi Solutions give us the entire state, in the lab (a's) and rotating (c's) frames

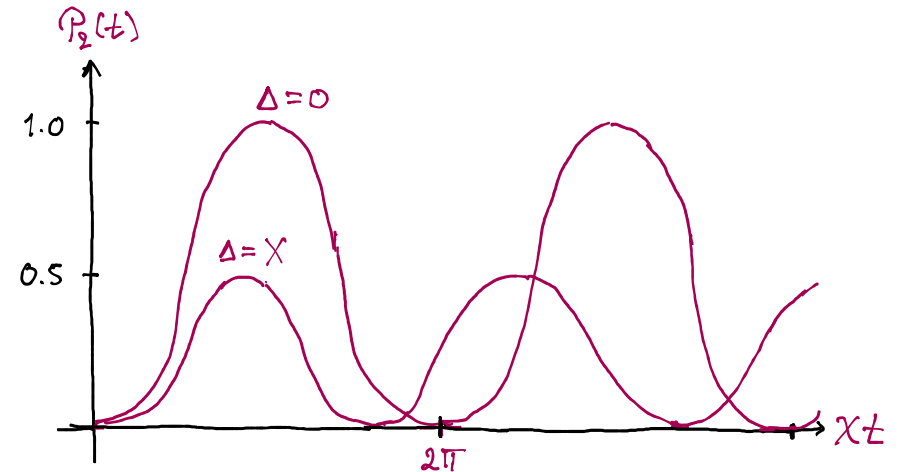


We have maximum information about the system and can make any predictions allowed by QM

Probabilities of finding the atom in $|1\rangle, |2\rangle$:

$$P_1(t) = |C_1(t)|^2 = \frac{1}{2} \left(1 + \frac{\Delta^2}{\Omega^2} \right) + \frac{1}{2} \frac{\chi^2}{\Omega^2} \cos \Omega t$$

$$P_2(t) = |C_2(t)|^2 = \frac{1}{2} \frac{\chi^2}{\Omega^2} (1 - \cos \Omega t)$$



Atom-Light Interaction: 2-Level Approximation

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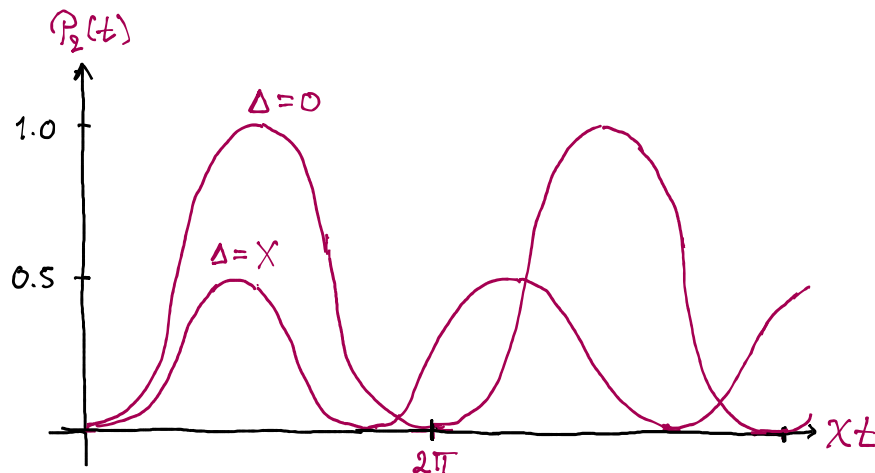


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Note: All 2-level systems are isomorphic

- Equivalent Observables
- Equivalent Phenomena
- The Rabi problem was first solved in ESR and NMR, for spin-1/2 particles with a magnetic moment $\vec{\mu}$ driven by a magnetic field \vec{B} with interaction $H = \vec{\mu} \cdot \vec{B}$
- 2-level systems are now often called qubits

Dressed States

The 2-level eqs. in the RWA look like a S.E. with

$$H_{RWA} = \hbar \begin{pmatrix} 0 & -\frac{1}{2}\chi \\ -\frac{1}{2}\chi & \Delta \end{pmatrix}$$

The eigenstates of H_{RWA} are called Dressed States

The DS are stationary only in the Rotating Frame.

In the Lab Frame (Schrödinger Picture) they are periodic, oscillating w/frequency ω