Begin 01-23-2024

Light-Matter Interaction

Free Electrons

Consider the limit $\omega \gg \omega_0$

$\Rightarrow$ effectively unbound electrons

This is a reasonable model of plasmas & metals

In this limit we have

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

$$n(\omega) = \sqrt{1 + \frac{N\alpha(\omega)}{\varepsilon_0}} \approx \sqrt{1 - \frac{Ne^2}{\varepsilon_0 m\omega^2}} \equiv \sqrt{1 - \frac{\omega_p^2}{\omega^2}}$$

We introduce the **Plasma Frequency**

$$\boxed{\omega_p = \sqrt{\frac{Ne^2}{\varepsilon_0 m}}}$$

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

We now have

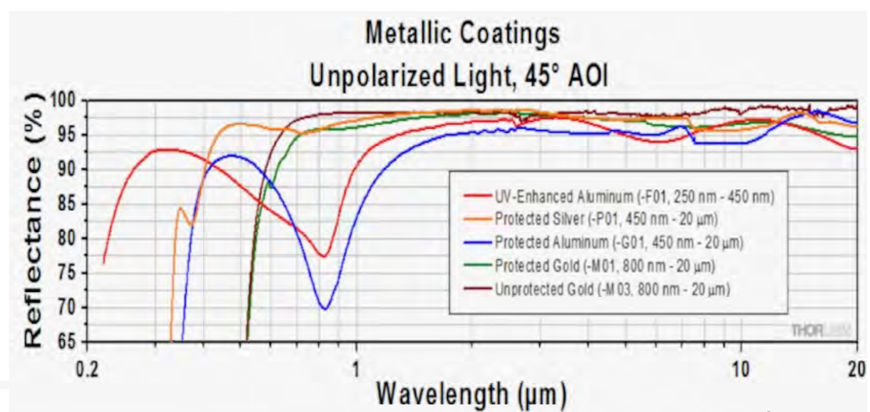
$$\vec{E}(z,t) = \vec{\varepsilon} E_0 e^{-i\omega[t - n(\omega)z/c]}$$

$$= \vec{\varepsilon} E_0 e^{-i\omega t} e^{i(z/c)\sqrt{\omega^2 - \omega_p^2}}$$

$$= \vec{\varepsilon} E_0 e^{-i\omega t} e^{-b(\omega)z}$$

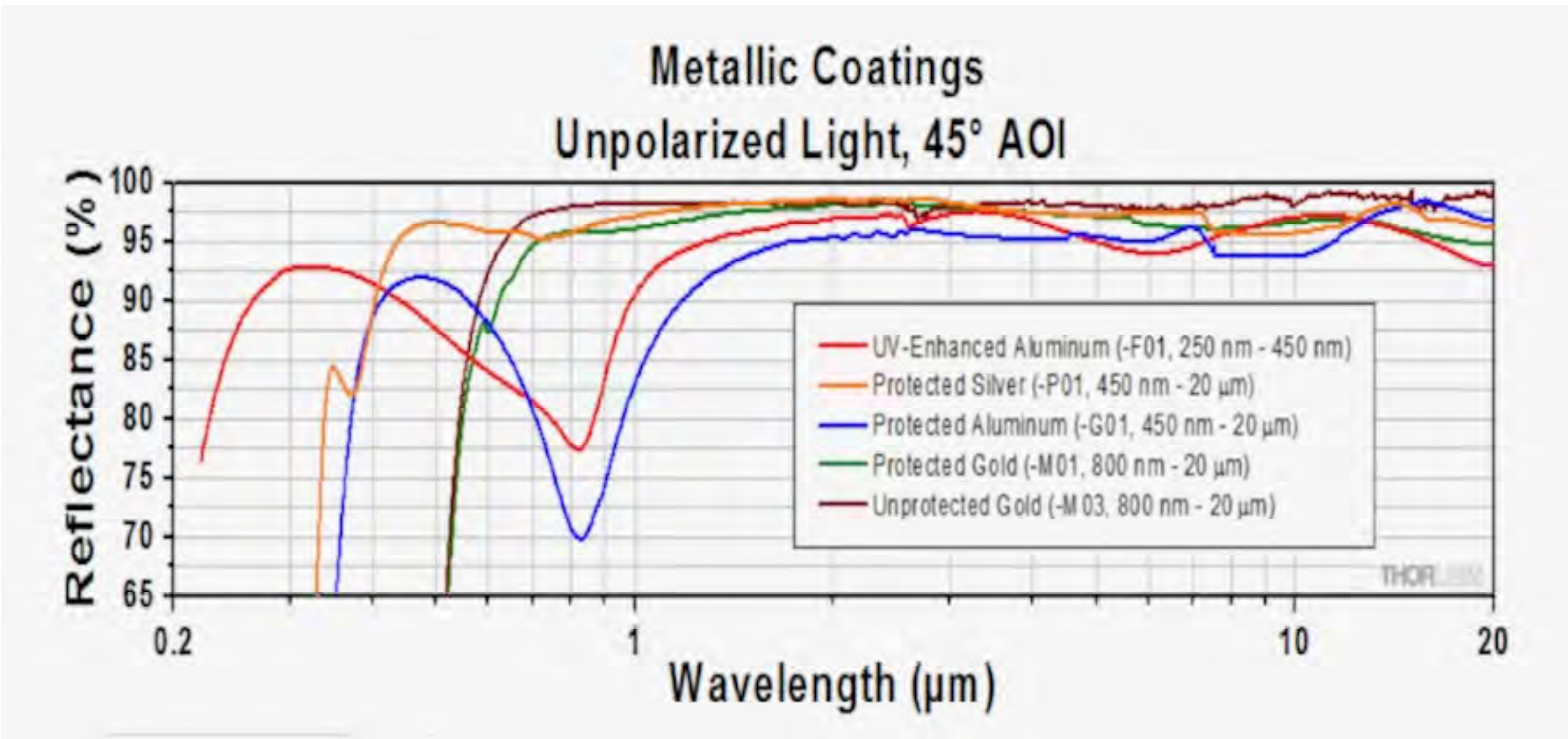
where $b(\omega) = -\frac{i}{c}\sqrt{\omega^2 - \omega_p^2}$

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



Light-Matter Interaction, Free Electrons

Examples of this kind of medium includes plasmas, and metals such as aluminum, silver and gold which are known to be excellent mirrors for visible and IR radiation.



Quantum Theory of Light-Matter Interaction

Completed:

- Fully classical description of fields & Atoms

Next Step:

- Semiclassical description $\left\{ \begin{array}{l} \text{Classical field} \\ \text{Quantum atoms} \end{array} \right.$

Self-Consistent Description

Electromagnetic Field → Atom/Molecule/Solid

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 $\hat{\vec{p}}$

Classical Field = C-valued vector $\vec{E}$

Cannot plug into Wave Eq. for classical field !

Wave Equation w/classical field & atoms

$$\boxed{\left(\nabla^2 - \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right)\vec{E} = \frac{1}{\varepsilon_0 c^2}\frac{\partial^2}{\partial t^2}\vec{P}, \qquad \vec{P} = N\vec{p}}$$

How do we deal with this mis-match?

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

⇩

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

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

$\Delta\hat{\vec{p}}(t)$ fluctuations

Note: Given $|\psi(t=0)\rangle$ and $\vec{E}$ the mean $\langle\vec{p}(t)\rangle$

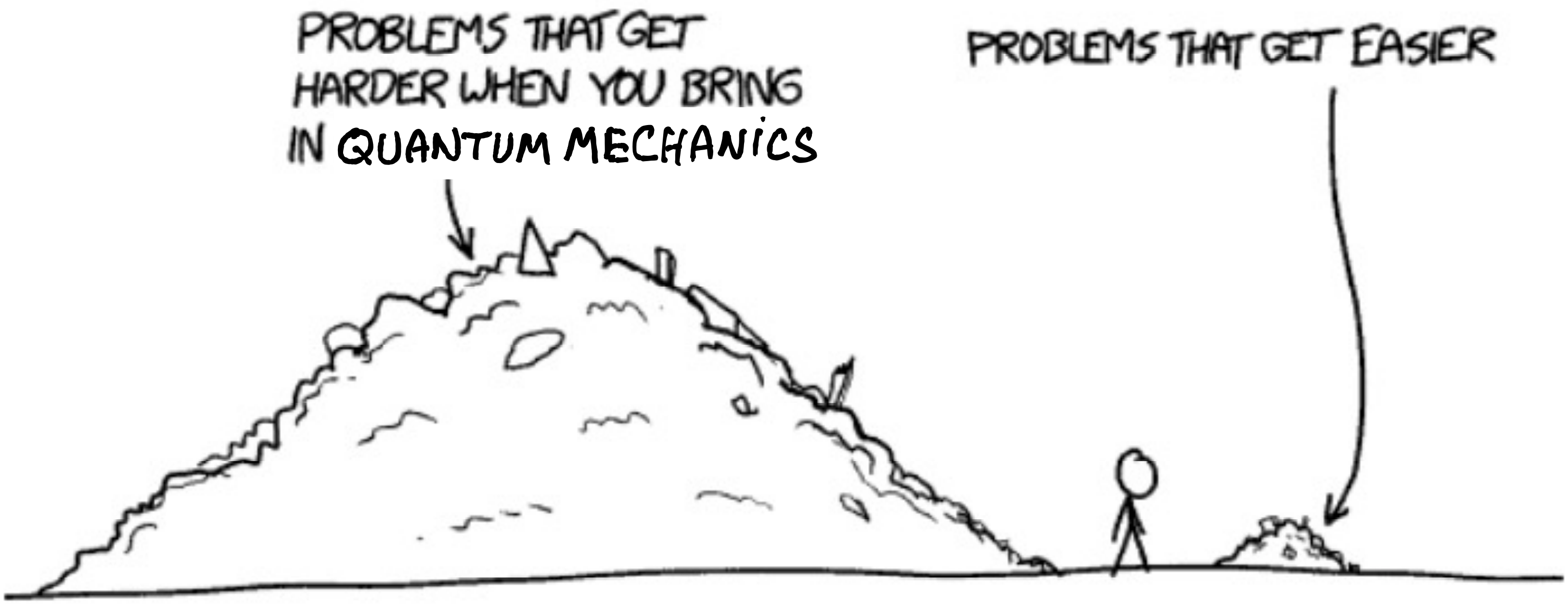
follows from the Schrödinger Eq.,

radiates <u>coherently</u> 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.

Quantum Theory of Light-Matter Interaction



Source: xkcd.com

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

$\Downarrow$

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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
- $\Delta\hat{\vec{p}}(t)$ 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 Equation w/classical field & atoms

$$\boxed{\left(\nabla^2 - \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right)\vec{E} = \frac{1}{\varepsilon_0 c^2}\frac{\partial^2}{\partial t^2}\vec{P}, \quad \vec{P} = N\langle\vec{\hat{\mu}}\rangle}$$

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Atom-field interaction

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

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

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.

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

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

$$i\hbar \sum_n \dot{a}_n(t) \langle \varphi_m | \varphi_n \rangle = \sum_n a_n(t) \left[E_n \langle \varphi_m | \varphi_n \rangle \right] + \overbrace{\langle \varphi_m | V_{ext} | \varphi_n \rangle}^{V_{mn}}$$

On vector-matrix form this can be written

$\boxed{i\hbar \dot{\underline{a}} = \underline{\underline{H}}_a \underline{a} + \underline{\underline{V}} \underline{a}}$ **S.E. in $\{|\varphi_n\rangle\}$ rep.**

Still exact!

Quantum Theory of Light-Matter Interaction

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Still exact!

Note: If $\underline{\underline{H}}_a$ and $\underline{\underline{V}}_{ext}$ 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!

Begin 01-25-2024

Atom-Light Interaction: 2-Level Approximation

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 $|\varphi_n\rangle \to |\varphi_m\rangle$ and far off resonance with all others.

State space $\quad Dim(\mathcal{E}) = 2, \; \{|1\rangle, |2\rangle\}$

Interaction $\quad \boxed{V_{ext} = -\hat{\vec{\mu}}\cdot\vec{E}(t)}$

$|2\rangle$

$\hbar\omega_{21}$

$|1\rangle$

State vector $\quad |\psi(t)\rangle = a_1(t)\,|1\rangle + a_2(t)\,|2\rangle$

Schröd. eq.

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

Observables $\quad \hat{\vec{\mu}} = \begin{pmatrix} 0 & \vec{\mu}_{12} \\ \vec{\mu}_{21} & 0 \end{pmatrix} \Rightarrow \hat{V} = -\hat{\vec{\mu}}\cdot\vec{E} = \begin{pmatrix} 0 & V_{12} \\ V_{21} & 0 \end{pmatrix}$

Interaction

$$V_{12}(t) = -\vec{\mu}_{12}\cdot\tfrac{1}{2}(\hat{\varepsilon}E_0 e^{-i\omega t} + c.c.)$$
$$V_{21}(t) = -\vec{\mu}_{21}\cdot\tfrac{1}{2}(\hat{\varepsilon}E_0 e^{-i\omega t} + c.c.)$$

Parity selection rule

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

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

$\Rightarrow$ Eigenstates $\quad \varphi(\vec{r}) = \pm\,\varphi(-\vec{r}) = \varphi(-[-\vec{r}])$

"+" for even parity
"-" for odd parity

two reflections equals the identity

The dipole $\hat{\vec{\mu}}$ is a vector operator $\Rightarrow$ transforms like a vector when $\vec{r} \to -\vec{r}$

Thus $\quad \hat{\vec{\mu}}(\vec{r}) = e\hat{\vec{r}} = -\hat{\vec{\mu}}(-\hat{\vec{r}})$ and

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

φ_n and φ_m have <u>opposite parity</u>

Parity rule: $\quad$ No dipole moment in energy eigenstate!

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

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We define

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

$$\left.\begin{aligned} \chi_{12} &= \vec{\mu}_{12}\cdot\hat{\varepsilon}E_0/\hbar \\ \chi_{21} &= \vec{\mu}_{21}\cdot\hat{\varepsilon}E_0/\hbar \end{aligned}\right\}$$

interaction energy is $\hbar\chi$

χ Rabi frequency

Note:
$$\begin{cases} \chi_{12}^* = \vec{\mu}_{21}\cdot(\hat{\varepsilon}E_0/\hbar)^* \neq \chi_{21} \\ \chi_{21}^* = \vec{\mu}_{12}\cdot(\hat{\varepsilon}E_0/\hbar)^* \neq \chi_{12} \end{cases}$$

$$H = \hbar\begin{pmatrix} 0 & -\chi_{12}e^{-i\omega t} - \chi_{21}^* e^{i\omega t} \\ -\chi_{21}e^{i\omega t} - \chi_{12}^* e^{-i\omega t} & \omega_{21} \end{pmatrix}$$

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

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

Atom-Light Interaction: 2-Level Approximation

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Switch to rotating frame (slow variables)

$$c_1(t) = a_1(t), \quad c_2(t) = a_2(t) e^{i\omega t}$$

$$\begin{aligned} i\dot{c}_1(t) &= -\frac{1}{2}\left(\chi_{12} e^{-i2\omega t} + \chi_{21}^*\right) c_2(t) \\ i\dot{c}_2(t) &= (\omega_{21} - \omega) c_2(t) - \frac{1}{2}\left(\chi_{21} + \chi_{12}^* e^{i2\omega t}\right) c_1(t) \end{aligned}$$

Rotating Wave Approximation (RWA)

Very important, equivalent to resonant approximation

Terms $\propto e^{\pm i 2\omega t}$ **average to zero on time scale for variations in** c_1, c_2

$$\begin{aligned} i\dot{c}_1(t) &= -\frac{1}{2} \chi_{21}^* c_2(t) \\ i\dot{c}_2(t) &= \Delta c_2(t) - \frac{1}{2} \chi_{21} c_1(t) \end{aligned}$$

$\Delta = \omega_{21} - \omega$

(detuning)

Exactly Solvable !

Atom-Light Interaction: 2-Level Approximation

Switch to rotating frame (slow variables)

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

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To simplify, make a global phase choice such that

$\chi_{21} = \vec{\mu}_{21}\cdot\hat{\varepsilon} E_0/\hbar = \chi$ **is real (not required)**

Simplest 2-level equations

$$i\dot{C}_1(t) = -\frac{1}{2}\chi C_2(t)$$

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

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

$$C_2(t) = \left(i\frac{\chi}{\Omega}\sin\frac{\Omega t}{2}\right) e^{-i\Delta t/2}$$

χ : **Rabi freq.** $\quad \Delta$: **Detuning**

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

Atom-Light Interaction: 2-Level Approximation

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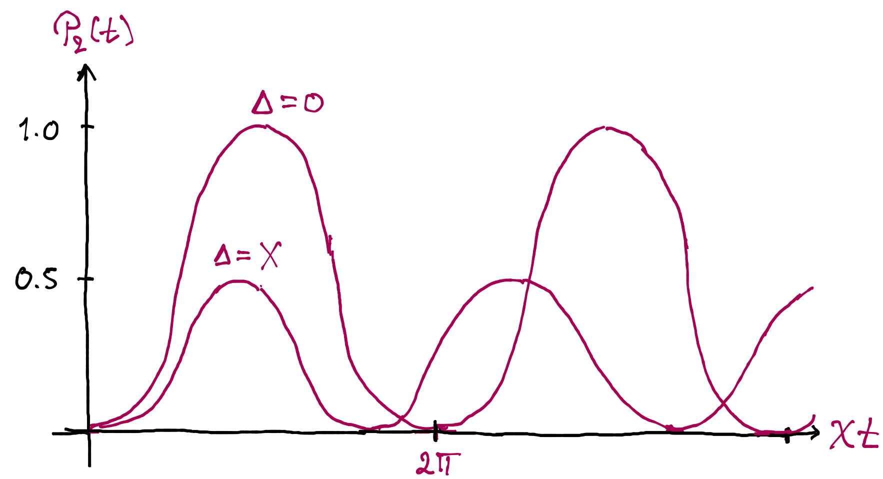
Note: The Rabi Solutions give us the entire state, in the lab (a's) and rotating (c's) frames

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

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

$$P_1(t) = |c_1(t)|^2 = \frac{1}{2}\left(1 + \frac{\Delta^2}{\Omega^2}\right) + \frac{1}{2}\frac{\chi^2}{\Omega^2}\cos\Omega t$$

$$P_2(t) = |c_2(t)|^2 = \frac{1}{2}\frac{\chi^2}{\Omega^2}\left(1 - \cos\Omega t\right)$$



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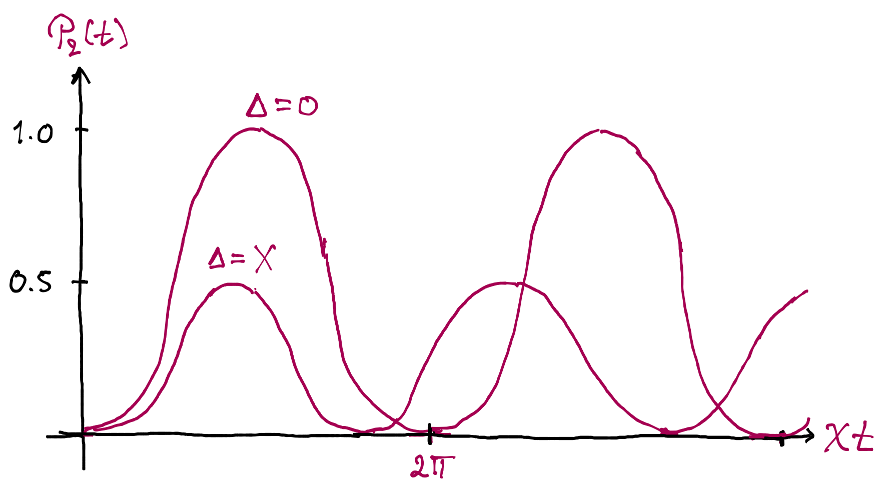

Note: All 2-level systems are isomorphic

- 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

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

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

Atom-Light Interaction: 2-Level Approximation

Comparison to the Classical Lorentz atom

Goal: To understand why the Lorentz model works so well, and its limits of validity

Classical Equation of Motion:

$$\boxed{\left(\frac{d^2}{dt^2}+\omega_0^2\right)\vec{r}=\frac{e}{m}\vec{E}}$$

will derive similar equation for $\langle\hat{\vec{r}}\rangle$

Equation of Motion for $\langle\hat{\vec{r}}\rangle$. First step:

$$\langle\hat{\vec{r}}\rangle \underset{(*)}{=} \langle\psi|\hat{\vec{r}}|\psi\rangle = (a_1^*, a_2^*)\begin{pmatrix}0 & \vec{r}_{12}\\ \vec{r}_{21} & 0\end{pmatrix}\begin{pmatrix}a_1\\ a_2\end{pmatrix}$$

$$= a_1^* a_2 \vec{r}_{12} + a_2^* a_1 \vec{r}_{21} = \vec{r}_{12}(a_1^* a_2 + a_1 a_2^*)$$

(choose phase to make $\vec{r}_{12}$ real)

We need an expression for $\frac{d^2}{dt^2}\langle\vec{r}\rangle$

We can find it from the S. E., i. e., the eqs for the *a* 's back when we first set up the Rabi problem.

We pick linear polarization so $\vec{\varepsilon}E_0$ is real-valued and $V_{12}=V_{21}=V$ ⇨ The eqs for the *a* 's are

$$\hbar\dot{a}_1^* = i(E_1 a_1^* + V a_2^*)$$

$$\hbar\dot{a}_2 = -i(E_2 a_2 + V a_1)$$

With this we have

$$\frac{d}{dt}a_1^* a_2 = (\dot{a}_1^* a_2 + a_1^*\dot{a}_2)$$

$$= -i\underbrace{\frac{E_2-E_1}{\hbar}}_{\omega_0} a_1^* a_2 - i\frac{V}{\hbar}\left(|a_1|^2-|a_2|^2\right)$$

Differentiating again gives us

$$\frac{d^2}{dt^2}(a_1^* a_2) = -\omega_0^2 a_1^* a_2 - \frac{\omega_0 V}{\hbar}\left(|a_1|^2-|a_2|^2\right) - i\hbar\frac{d}{dt}\left[\frac{V}{\hbar}\left(|a_1|^2-|a_2|^2\right)\right]$$

Looking at the eq. $(*)$ for $\langle\hat{\vec{r}}\rangle$ suggests we should add the complex conjugate and multiply w $\vec{r}_{12}$

Atom-Light Interaction: 2-Level Approximation

We pick linear polarization so $\vec{\mathcal{E}}E_0$ is real-valued and $V_{12} = V_{21} = V$ ⇨ The eqs for the *a* 's are

$$\hbar \dot{a}_1^* = i(E_1 a_1^* + V a_2^*)$$

$$\hbar \dot{a}_2 = -i(E_2 a_2 + V a_1)$$

With this we have

$$\frac{d}{dt} a_1^* a_2 = (\dot{a}_1^* a_2 + a_1^* \dot{a}_2)$$

$$= -i \underbrace{\frac{E_2 - E_1}{\hbar}}_{\omega_0} a_1^* a_2 - i \frac{V}{\hbar}(|a_1|^2 - |a_2|^2)$$

Differentiating again gives us

$$\frac{d^2}{dt^2}(a_1^* a_2) = -\omega_0^2 a_1^* a_2 - \frac{\omega_0 V}{\hbar}(|a_1|^2 - |a_2|^2)$$

$$-i\hbar \frac{d}{dt}\left[\frac{V}{\hbar}(|a_1|^2 - |a_2|^2)\right]$$

Looking at the eq. for $\langle \hat{\vec{p}} \rangle$ suggests we should add the complex conjugate and multiply w $\vec{p}_{12}$

This gives us

$$\boxed{\begin{aligned} \left(\frac{d^2}{dt^2} + \omega_0^2\right)\langle \hat{\vec{p}} \rangle &= \frac{2\omega_0 \vec{p}_{12} V}{\hbar}(|a_1|^2 - |a_2|^2) \\ &= \frac{2\omega_0}{\hbar} \vec{p}_{12}(\vec{p}_{12} \cdot \vec{E})(|a_1|^2 - |a_2|^2) \end{aligned}}$$

$\vec{p}_{12} = \langle 1 | \hat{\vec{p}} | 2 \rangle$: dipole matrix element

To wrap up, we need to know a bit about real, multilevel atoms. (We will revisit this soon)

Pick linear polarization so $\vec{\mathcal{E}}$ is real-valued.
Pick quantization axis along $\vec{\mathcal{E}}$ ⇨ $\vec{p}_{12} = p_{12} \vec{\mathcal{E}}$

⇩

$$\boxed{\left(\frac{d^2}{dt^2} + \omega_0^2\right)\langle \hat{\vec{p}} \rangle = \frac{2\omega_0 p_{12}^2}{\hbar} \vec{E}\,(|a_1|^2 - |a_2|^2)}$$

Atom-Light Interaction: 2-Level Approximation

This gives us

$$\left(\frac{d^2}{dt^2}+\omega_0^2\right)\langle\hat{\vec{r}}\rangle = \frac{2\omega_0 \vec{r}_{12} V}{\hbar}\left(|a_1|^2-|a_2|^2\right)$$
$$= \frac{2\omega_0}{\hbar}\vec{r}_{12}\left(\vec{r}_{12}\cdot\vec{E}\right)\left(|a_1|^2-|a_2|^2\right)$$

$\vec{r}_{12} = \langle 1|\hat{\vec{r}}|2\rangle$: dipole matrix element

To wrap up, we need to know a bit about real, multilevel atoms. (We will revisit this soon)

Pick linear polarization so $\vec{\mathcal{E}}$ is real-valued.
Pick quantization axis along $\vec{\mathcal{E}}$ ⇨ $\vec{r}_{12} = r_{12}\vec{\mathcal{E}}$

$$\left(\frac{d^2}{dt^2}+\omega_0^2\right)\langle\hat{\vec{r}}\rangle = \frac{2\omega_0 r_{12}^2}{\hbar}\vec{E}\left(|a_1|^2-|a_2|^2\right)$$

Compare to Classical Equation of Motion

$$\left(\frac{d^2}{dt^2}+\omega_0^2\right)\vec{r} = \frac{e}{m}\vec{E}$$

The two eqs. have the same form if $|a_1|^2 \sim 1$, $|a_2|^2 \sim 0$

This is the case for $\Delta \gg \chi$
Or when $\chi \ll \Gamma$

Limit of weak Excitation !

Decay rate of $|2\rangle$

Oscillator Strength $f = \dfrac{2m\omega_0}{\hbar e} r_{12}^2$

$$\left(\frac{d^2}{dt^2}+\omega_0^2\right)\langle\hat{\vec{r}}\rangle = \frac{e}{m}f\vec{E}$$

Like the classical equation,
but with modified polarizability !

End 01-25-2024