

Basics of coherent light-matter interactions

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Outline



- Atom in an external radiation field
 - Dipole coupling and the selection rules
 - Spontaneous decay of an excited atom

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 - Eigenstates
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 - Adiabatic and non-adiabatic transitions
- Three-level atom in a (bichromatic) laser field
 - Two-photon Rabi oscillations
 - Eigenstates: Coherent population trapping (dark) state
 - Stimulated Raman adiabatic passage (STIRAP)



I. Atom in an external radiation field



H-like (alkali) atoms $\mathcal{H}^{A} = \frac{P^{2}}{2m_{e}} + V(r)$ $[V(r) \propto -\frac{e^{2}}{r}]$

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 $\mathrm{Ry} = \frac{1}{(4\pi\varepsilon_0)^2} \frac{m_e e^4}{2\hbar^2} = \frac{\alpha^2}{2} m_e c^2$

effective PQN $n^* = n - \delta_l$ (δ_l quantum defect)



*|e*₂>-----







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Wavefunctions
$$\langle \boldsymbol{r} | nlm \rangle \equiv \Psi_{nlm}(\boldsymbol{r}) = R_{nl}(r)Y_l^m(\theta,\phi)$$

OAMQN $l = 0, 1, \dots, n-1$ MQN (projection) $m = -l, -l+1, \dots, l$

[e.g.
$$R_{10}(r) = 2a_0^{-3/2}e^{-r/a_0}$$
 $a_0 = \frac{4\pi\varepsilon_0\hbar^2}{m_e e^2}$]





 $|e_1\rangle$ —

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Parity of $|nlm\rangle$:

•
$$l = 0, 2, 4, \dots$$
 (s,d,g,...) $\Rightarrow \Psi_{nlm}(\mathbf{r}) = \Psi_{nlm}(-\mathbf{r})$ even
• $l = 1, 3, 5, \dots$ (p,f,h,...) $\Rightarrow \Psi_{nlm}(\mathbf{r}) = -\Psi_{nlm}(-\mathbf{r})$ odd



$$|e_2\rangle$$

$$|e_1\rangle$$
 ——

Spin-Orbit Coupling



Fine structure: electron spin s interacts with orbital angular momentum l

- \Rightarrow *LS* interaction $\mathcal{V}_{ls} \propto \boldsymbol{l} \cdot \boldsymbol{s} \sim \alpha^2 E_n$
- \Rightarrow Total angular momentum J=l+s

$$|J,M\rangle = \sum_{\substack{m_l,m_s\\(m_l+m_s=M)}} C^j_{m_lm_s} |lm_l\rangle |sm_s\rangle$$

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Hyperfine structure: nuclear spin I interacts with l and s

- \Rightarrow *LI* interaction $V_{LI} \propto \boldsymbol{l} \cdot \boldsymbol{I} \sim \frac{m_e}{m_p} \alpha^2 E_n$
- \Rightarrow Total angular momentum $m{F} = m{J} + m{I}$

Atom-Field Coupling

$$\mathcal{H} = \frac{1}{2m_{e}} [\boldsymbol{P} - e\boldsymbol{A}(\boldsymbol{r})]^{2} + V(r) = \mathcal{H}^{A} + \mathcal{V}^{AF}$$

$$\Rightarrow \quad \mathcal{V}^{\mathrm{AF}} = - rac{e}{m_{\mathrm{e}}} \boldsymbol{P} \cdot \boldsymbol{A} + rac{e^2}{2m_{\mathrm{e}}} \boldsymbol{A}^2$$

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Dipole approximation $\mathbf{k} \cdot \mathbf{r} \lesssim ka_0 \ll 1 \Rightarrow \mathbf{A}(\mathbf{r}) \simeq \mathbf{A}(0)$ $\langle nlm_l | \mathcal{V}^{AF} | n'l'm_l' \rangle = -\frac{e}{m_e} \langle nlm_l | \mathbf{P} | n'l'm_l' \rangle \cdot \mathbf{A}$ $= i \frac{e}{\hbar} \langle nlm_l | [\mathbf{r}, \mathcal{H}^A] | n'l'm_l' \rangle \cdot \mathbf{A}$ $= -i\omega_{nn'} e \langle nlm_l | \mathbf{r} | n'l'm_l' \rangle \cdot \mathbf{A} \qquad [\omega_{nn'} = \frac{E_n - E_{n'}}{\hbar}]$

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For $\omega \sim \omega_{nn'} \Rightarrow i\omega A = E \Rightarrow$

Dipole coupling

$$\mathcal{V}^{\mathrm{AF}} = -e\boldsymbol{r}\cdot\boldsymbol{E}$$



 $\langle nlm | e\mathbf{r} | n'l'm' \rangle \equiv e \int d^3r \, \Psi^*_{nlm}(\mathbf{r}) \, \mathbf{r} \, \Psi_{n'l'm'}(\mathbf{r}) \neq 0 \quad \text{for} \quad l-l'=\pm 1$

 \Rightarrow $|nlm\rangle \& |n'l'm'\rangle$ should have different parity (*r* is an odd function)



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Linearly π **polarized field** $(\hat{z} \parallel E)$

 $\boldsymbol{E} = \hat{\boldsymbol{e}} E \quad (\hat{\boldsymbol{e}} = \hat{z}) \Rightarrow m_l - m_{l'} = 0 \quad (m_{\text{phot}} = 0)$



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Circularly σ_{\pm} **polarized** field $(\hat{z} \parallel \mathbf{k})$

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 $\langle JM | e\mathbf{r} | J'M' \rangle \neq 0$ for $J - J' = 0, \pm 1 \& M - M' = m_{\text{phot}}$ $(F \leftrightarrow J)$



Free-space EM field:
$$E = -i \sum_{k\sigma} \sqrt{\frac{\hbar\omega_k}{2\varepsilon_0 V}} \hat{e}_{k\sigma} [a_{k\sigma} - a_{k\sigma}^{\dagger}]; \quad \mathcal{V}^{AF} = -e\mathbf{r} \cdot \mathbf{E}$$



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 $|\Psi(0)\rangle = |e, \{0_{\boldsymbol{k}\sigma}\}\rangle$

 $|\Psi(t)\rangle = c_e(t) |e,0\rangle + \sum_{k\sigma} c_{k\sigma}(t) |g,1_{k\sigma}\rangle \quad \Rightarrow$





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 $G_e \equiv \sum_{\boldsymbol{k}\sigma} \frac{\omega_k}{2\varepsilon_0 \hbar V} |\boldsymbol{\wp}_{ge} \cdot \hat{\boldsymbol{e}}_{\boldsymbol{k}\sigma}|^2 \int_0^t e^{i(\omega_{eg} - \omega_k)(t - t')} dt' \qquad (\boldsymbol{\wp}_{ge} = \langle g | e\boldsymbol{r} | e \rangle)$



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$$\sum_{k\sigma} \to \frac{V}{(2\pi)^3} \int d^3k = \frac{V}{(2\pi)^3 c^3} \int d\omega_k \,\omega_k^2 \int d\Omega$$
$$\int e^{i(\omega_{eg} - \omega_k)(t - t')} dt' \to \pi \delta(\omega_{eg} - \omega_k) + i \mathrm{P} \frac{1}{\omega_{eg} - \omega_k}$$



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$$\Rightarrow \quad G_e = \frac{1}{2}\Gamma_e + iS_e$$
 with

$$\Gamma_e = \frac{1}{4\pi\varepsilon_0} \frac{4\omega_{eg}^3 |\wp_{eg}|^2}{3\hbar c^3} \qquad S_e = \frac{1}{4\pi\varepsilon_0} \frac{2|\wp_{eg}|^2}{3\pi\hbar c^3} \operatorname{P} \int d\omega_k \frac{\omega_k^3}{\omega_{eg} - \omega_k}$$



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$$\boxed{\frac{\partial}{\partial t} |c_e|^2 = -\Gamma_e |c_e|^2} \quad \Rightarrow \quad |c_e(t)|^2 = e^{-\Gamma_e t} |c_e(0)|^2$$

Problems & Questions



- Prove the commutation relation $[r, \mathcal{H}^A] = i \frac{\hbar}{m_e} P$
- Γ_{eg} is the spontaneous decay rate of an excited atomic state $|e\rangle$ to the ground state $|g\rangle$ by emitting a photon into the vacuum modes of the radiation field. If the initial state of the field was not a vacuum n = 0, but, e.g., a thermal state [with Plank distribution $\bar{n}(\omega_k) = (e^{\hbar\omega_k/k_{\rm B}T} - 1)^{-1}$], what would the decay rate Γ_{eg} be? Could an atom in the ground state $|g\rangle$ be excited to $|e\rangle$ by the thermal photons? What would the excitation rate Γ_{ge} be?



II. Two-level atom in a laser field

Atom-Field Interaction

FORTH VESL

$\mathcal{H} = \mathcal{H}^{\mathrm{A}} + \mathcal{V}^{\mathrm{AF}}(t)$



FORTH.

$\mathcal{H} = \mathcal{H}^{\mathrm{A}} + \mathcal{V}^{\mathrm{AF}}(t)$

 $\mathcal{H}^{\mathbf{A}} = \hbar \omega_g \left| g \right\rangle \langle g \right| + \hbar \omega_e \left| e \right\rangle \langle e \right|$



$$\begin{split} \mathcal{H} &= \mathcal{H}^{A} + \mathcal{V}^{AF}(t) \\ \mathcal{H}^{A} &= \hbar \omega_{g} |g\rangle \langle g| + \hbar \omega_{e} |e\rangle \langle e| \\ \\ \mathcal{V}^{AF}(t) &= -\wp \cdot \boldsymbol{E}(t) = -\wp E(t) \quad \text{with} \end{split}$$

 $\varphi \equiv \boldsymbol{\wp} \cdot \boldsymbol{e} \qquad E(t) = \mathcal{E}e^{-i\omega t} + \mathcal{E}^* e^{i\omega t} = 2|\mathcal{E}|\cos(\omega t - \varphi)|$





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$$|\Psi(t)\rangle = c_g(t) |g\rangle + c_e(t) |e\rangle \qquad \frac{\partial}{\partial t} |\Psi(t)\rangle = -\frac{i}{\hbar} \mathcal{H} |\Psi(t)\rangle \Rightarrow$$

Interaction picture $c_{g,e}(t) = \tilde{c}_{g,e}(t)e^{-i\omega_{g,e}t}$

$$\frac{\partial}{\partial t}\tilde{c}_{g} = i\tilde{c}_{e}\frac{\wp_{ge}}{\hbar}\left(\mathcal{E}e^{-i(\omega+\omega_{eg})t} + \mathcal{E}^{*}e^{i(\omega-\omega_{eg})t}\right) \simeq i\tilde{c}_{e}\frac{\wp_{ge}}{\hbar}\mathcal{E}^{*}e^{i\Delta t}$$
$$\frac{\partial}{\partial t}\tilde{c}_{e} = i\tilde{c}_{g}\frac{\wp_{eg}}{\hbar}\left(\mathcal{E}e^{-i(\omega-\omega_{eg})t} + \mathcal{E}^{*}e^{i(\omega+\omega_{eg})t}\right) \simeq i\tilde{c}_{g}\frac{\wp_{eg}}{\hbar}\mathcal{E}e^{-i\Delta t}$$

Rotating Wave Approximation (RWA): $\omega + \omega_{eg} \gg \omega - \omega_{eg} \equiv \Delta$







$$\begin{split} \frac{\partial}{\partial t} c_g &= i \Omega^* c_e \\ \frac{\partial}{\partial t} c_e &= i \Delta c_e + i \Omega c_g \end{split} \begin{array}{l} \Omega \equiv \frac{\wp_{eg}}{\hbar} \mathcal{E} \\ (c_e &= \tilde{c}_e e^{i \Delta t}) \end{split} \label{eq:gamma_state} \end{tabular} \text{Rabi frequency}$$





Time $(1/\Omega)$

Resonant field $\Delta = 0 \& \overline{\Omega} = \Omega$

$$c_g(t) = \cos(\Omega t), \ c_e(t) = i\sin(\Omega t)$$





 $\frac{\partial}{\partial t}c_g = i\Omega^*c_e$ $\frac{\partial}{\partial t}c_e = i\Delta c_e + i\Omega c_q \qquad (c_e = \tilde{c}_e e^{i\Delta t})$ **Solution** $c_q(0) = 1$, $c_e(0) = 0$ $c_q(t) = \cos(\bar{\Omega}t) - i\frac{\Delta}{2\bar{\Omega}}\sin(\bar{\Omega}t)$ $c_e(t) = i\frac{\Omega}{\overline{\Omega}}\sin(\overline{\Omega}t)$ $\bar{\Omega} \equiv \sqrt{\Omega^2 + (\Delta/2)^2}$ eff. Rabi fr. **Resonant field** $\Delta = 0 \& \overline{\Omega} = \Omega$ $c_q(t) = \cos(\Omega t), \ c_e(t) = i \sin(\Omega t)$ **Inversion** $2\Omega T = \pi$ -pulse $|c_a(T)|^2 = 0$ $|c_e(T)|^2 = 1$



$$\begin{aligned} &\frac{\partial}{\partial t}\rho_{gg} = \Gamma\rho_{ee} + i(\Omega^*\rho_{eg} - \rho_{ge}\Omega) \\ &\frac{\partial}{\partial t}\rho_{ee} = -\Gamma\rho_{ee} + i(\Omega\rho_{ge} - \rho_{eg}\Omega^*) \\ &\frac{\partial}{\partial t}\rho_{eg} = (i\Delta - \gamma_{eg})\rho_{eg} - i\Omega(\rho_{ee} - \rho_{gg}) \end{aligned}$$

$$\gamma_{eg} = \frac{1}{2}\Gamma + \dots$$



$$\begin{split} &\frac{\partial}{\partial t}\rho_{gg} = \Gamma\rho_{ee} + i(\Omega^*\rho_{eg} - \rho_{ge}\Omega) \\ &\frac{\partial}{\partial t}\rho_{ee} = -\Gamma\rho_{ee} + i(\Omega\rho_{ge} - \rho_{eg}\Omega^*) \\ &\frac{\partial}{\partial t}\rho_{eg} = (i\Delta - \gamma_{eg})\rho_{eg} - i\Omega(\rho_{ee} - \rho_{gg}) \end{split}$$

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$$\frac{\partial}{\partial t}\rho_{gg} = \Gamma\rho_{ee} + i(\Omega^*\rho_{eg} - \rho_{ge}\Omega)$$

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$$\frac{\partial}{\partial t}\rho_{eg} = (i\Delta - \gamma_{eg})\rho_{eg} - i\Omega(\rho_{ee} - \rho_{gg})$$

$$\gamma_{eg} = \frac{1}{2}\Gamma + \dots$$

Steady state $t \to \infty$ $\Delta = 0$ $\rho_{ee}(\infty) = \frac{|\Omega|^2}{\frac{\Gamma}{2\gamma_{eg}}(\Delta^2 + \gamma_{eg}^2) + 2|\Omega|^2}$ ి 0.5 $\rho_{gg} = 1 - \rho_{ee}$ 0 $w = \sqrt{2|\Omega|^2 \frac{2\gamma_{eg}}{\Gamma} + \gamma_{eg}^2}$ linewidth ౖౖ^జ 0.5 0



Time $(1/\Gamma)$



$$\begin{aligned} \frac{\partial}{\partial t}\rho_{gg} &= \Gamma\rho_{ee} + i(\Omega^*\rho_{eg} - \rho_{ge}\Omega) \\ \frac{\partial}{\partial t}\rho_{ee} &= -\Gamma\rho_{ee} + i(\Omega\rho_{ge} - \rho_{eg}\Omega^*) \\ \frac{\partial}{\partial t}\rho_{eg} &= (i\Delta - \gamma_{eg})\rho_{eg} - i\Omega(\rho_{ee} - \rho_{gg}) \end{aligned}$$

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Steady state $t \to \infty$ $\Delta = 0$ $\rho_{ee}(\infty) = \frac{|\Omega|^2}{\frac{\Gamma}{2\gamma_{eg}}(\Delta^2 + \gamma_{eg}^2) + 2|\Omega|^2}$ ి 0.5 $\rho_{gg} = 1 - \rho_{ee}$ 0 $w = \sqrt{2|\Omega|^2 \frac{2\gamma_{eg}}{\Gamma} + \gamma_{eg}^2}$ linewidth ౖౖౖ 0.5 **Resonant field** $\Delta = 0$ $\rho_{ee}(\infty) \rightarrow \frac{1}{2}$ for $\Omega^2 \gg \Gamma \gamma_{eg}$ 0 0 2



Time $(1/\Gamma)$





Interaction Hamiltonian (rotating frame ω)

 $\mathcal{H}_{\rm int} = -\hbar\Delta |e\rangle\langle e| -\hbar\Omega(|e\rangle\langle g| + |g\rangle\langle e|) = -\hbar \begin{bmatrix} 0 & \Omega\\ \Omega & \Delta \end{bmatrix} \qquad \{|g\rangle, |e\rangle\}$



Interaction Hamiltonian (rotating frame ω)

$$\mathcal{H}_{\rm int} = -\hbar\Delta \left| e \right\rangle \langle e \right| - \hbar\Omega(\left| e \right\rangle \langle g \right| + \left| g \right\rangle \langle e \right|) = -\hbar \left[\begin{array}{cc} 0 & \Omega \\ \Omega & \Delta \end{array} \right] \qquad \left\{ \left| g \right\rangle, \left| e \right\rangle \right\}$$





Interaction Hamiltonian (rotating frame ω)

 $\mathcal{H}_{\rm int} = -\hbar\Delta |e\rangle\langle e| -\hbar\Omega(|e\rangle\langle g| + |g\rangle\langle e|) = -\hbar \begin{bmatrix} 0 & \Omega\\ \Omega & \Delta \end{bmatrix} \qquad \{|g\rangle, |e\rangle\}$





Interaction Hamiltonian (rotating frame ω)

 $\mathcal{H}_{\rm int} = -\hbar\Delta |e\rangle\langle e| -\hbar\Omega(|e\rangle\langle g| + |g\rangle\langle e|) = -\hbar \begin{bmatrix} 0 & \Omega\\ \Omega & \Delta \end{bmatrix} \qquad \{|g\rangle, |e\rangle\}$





Interaction Hamiltonian (rotating frame ω)

 $\mathcal{H}_{\rm int} = -\hbar\Delta |e\rangle\langle e| -\hbar\Omega(|e\rangle\langle g| + |g\rangle\langle e|) = -\hbar \begin{bmatrix} 0 & \Omega\\ \Omega & \Delta \end{bmatrix} \qquad \{|g\rangle, |e\rangle\}$





Time-dependent detuning

$$\Delta(t) = at \ (a > 0) \quad [\omega(t) \text{ or } \omega_{eg}(t)]$$





Time-dependent detuning

 $\Delta(t) = at \ (a > 0) \quad \ \ [\omega(t) \text{ or } \omega_{eg}(t)]$

Initial state $|\Psi(-\infty)\rangle = |g\rangle \simeq |+\rangle$

Final state $|\Psi(\infty)
angle = |e
angle \simeq |+
angle$ (?)





Time-dependent detuning

 $\Delta(t) = at \ (a > 0) \quad [\omega(t) \text{ or } \omega_{eg}(t)]$

Initial state $|\Psi(-\infty)\rangle = |g\rangle \simeq |+\rangle$

Final state $|\Psi(\infty)
angle = |e
angle \simeq |+
angle$ (?)



Non-adiabatic $|+\rangle \rightarrow |-\rangle$ transition probability (Landau-Zener formula)

$$P_{\rm tr} = e^{-2\pi\Gamma}$$
 $\Gamma = \frac{\Omega^2}{\frac{\partial}{\partial t}|\lambda_+ - \lambda_i|} \sim \frac{\Omega^2}{a}$



Time-dependent detuning

 $\Delta(t) = at \ (a > 0) \quad [\omega(t) \text{ or } \omega_{eg}(t)]$

Initial state $|\Psi(-\infty)\rangle = |g\rangle \simeq |+\rangle$

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Non-adiabatic $\ket{+} \rightarrow \ket{-}$ transition probability (Landau-Zener formula)

$$P_{\rm tr} = e^{-2\pi\Gamma}$$
 $\Gamma = \frac{\Omega^2}{\frac{\partial}{\partial t}|\lambda_+ - \lambda_i|} \sim \frac{\Omega^2}{a}$

 $\Omega^2 \gg a \Rightarrow \Gamma \gg 1 \& P_{tr} \ll 1$: Adiabatic following $|\Psi(t)\rangle = |+\rangle \forall t$

Adiabatic transfer |g
angle
ightarrow |e
angle

Problems & Questions



- A resonant (Δ = 0) coherent field with Rabi frequency Ω can prepare an equally weighted superposition of atomic states |g, e⟩ for π/2-pulse
 2 ∫ dtΩ(t) = 2ΩT_{π/2} = π/2 (assuming square pulse).
 For a non-resonant field |Δ| ≤ 2Ω, what would be the pulse area/duration for preparing |c_{g,e}| = 1/√2? What would be the relative phases for the amplitudes?
- In a Landau-Zener process of some duration T = 1/v, governed by the Schrödinder eq. $iv\partial_{\tau} |\Psi(\tau)\rangle = \mathcal{H}(\tau) |\Psi(\tau)\rangle$ ($\tau \equiv vt$), show that the non-adiabatic couling between the instantanous eigenstates $|\Psi_{\pm}\rangle [\mathcal{H}(\tau) |\Psi_{\pm}\rangle = \lambda_{\pm} |\Psi_{\pm}\rangle)$ is

$$-iv\langle\Psi_{+}|\,\partial_{\tau}\,|\Psi_{-}\rangle = -i\frac{v}{\lambda_{+}-\lambda_{-}}\langle\Psi_{+}|\,\partial_{\tau}\mathcal{H}(\tau)\,|\Psi_{-}\rangle$$



III. Three-level atom

MAOP-AM, 30/08/22 - p. 18/25

Atom-Fields Interactions

 $\mathcal{H} = \mathcal{H}^{\mathrm{A}} + \mathcal{V}^{\mathrm{AF}}(t)$





Atom-Fields Interactions





$$\begin{aligned} \mathcal{H}^{A} &= \hbar \omega_{g} |g\rangle \langle g| + \hbar \omega_{e} |e\rangle \langle e| + \hbar \omega_{s} |s\rangle \langle s| \\ \mathcal{V}^{AF}(t) &= -\wp \cdot \left[\boldsymbol{E}_{1}(t) + \boldsymbol{E}_{2}(t) \right] \qquad \wp_{\mu\nu} \equiv \langle \mu | \wp \cdot \boldsymbol{e}_{j} |\nu\rangle \\ &= -\wp_{eg} |e\rangle \langle g| \mathcal{E}_{1} e^{-i\omega_{1}t} - \wp_{es(gs)} |e(g)\rangle \langle s| \mathcal{E}_{2} e^{-i\omega_{2}t} + \text{H.c.} \end{aligned}$$

Atom-Fields Interactions





$$\begin{aligned} \mathcal{H}^{A} &= \hbar \omega_{g} |g\rangle \langle g| + \hbar \omega_{e} |e\rangle \langle e| + \hbar \omega_{s} |s\rangle \langle s| \\ \mathcal{V}^{AF}(t) &= -\wp \cdot \left[\boldsymbol{E}_{1}(t) + \boldsymbol{E}_{2}(t) \right] \qquad \wp_{\mu\nu} \equiv \langle \mu | \wp \cdot \boldsymbol{e}_{j} | \nu \rangle \\ &= -\wp_{eg} |e\rangle \langle g | \mathcal{E}_{1} e^{-i\omega_{1}t} - \wp_{es(gs)} |e(g)\rangle \langle s | \mathcal{E}_{2} e^{-i\omega_{2}t} + \text{H.c.} \end{aligned}$$

Interaction Hamiltonian (rotating frame $\omega_{1,2}$)

$$\mathcal{H}_{\Xi,\Lambda} = -\hbar \begin{bmatrix} 0 & \Omega_1 & 0 \\ \Omega_1 & \Delta_1 & \Omega_2 \\ 0 & \Omega_2 & \Delta_1 \pm \Delta_2 \end{bmatrix} \qquad \mathcal{H}_V = -\hbar \begin{bmatrix} 0 & \Omega_1 & \Omega_2 \\ \Omega_1 & \Delta_1 & 0 \\ \Omega_2 & 0 & \Delta_2 \end{bmatrix}$$
$$\{ |g\rangle, |e\rangle, |s\rangle \} \qquad \Delta_{1,2} = \omega_{1,2} - \omega_{\mu\nu} \qquad \Omega_{1,2} = \frac{\wp_{\mu\nu}}{\hbar} \mathcal{E}_{1,2}$$



 $\frac{\partial}{\partial t} |\Psi(t)\rangle = -\frac{i}{\hbar} \mathcal{H}_{\Lambda} |\Psi(t)\rangle \Rightarrow$

$$\begin{split} |\Psi(t)\rangle &= c_g(t) |g\rangle + c_e(t) |e\rangle + c_s(t) |s\rangle \\ \frac{\partial}{\partial t} c_g &= i\Omega_1 c_e \\ \frac{\partial}{\partial t} c_e &= (i\Delta_1 - \gamma_e)c_e + i\Omega_1 c_g + i\Omega_2 c_s \\ \frac{\partial}{\partial t} c_s &= i(\Delta_1 - \Delta_2)c_s + i\Omega_2 c_e \end{split}$$



$$\begin{split} |\Psi(t)\rangle &= c_g(t) |g\rangle + c_e(t) |e\rangle + c_s(t) |s\rangle \qquad \frac{\partial}{\partial t} |\Psi(t)\rangle = -\frac{i}{\hbar} \mathcal{H}_{\Lambda} |\Psi(t)\rangle \Rightarrow \\ \frac{\partial}{\partial t} c_g &= i\Omega_1 c_e \\ \frac{\partial}{\partial t} c_e &= (i\Delta_1 - \gamma_e)c_e + i\Omega_1 c_g + i\Omega_2 c_s \\ \frac{\partial}{\partial t} c_s &= i(\Delta_1 - \Delta_2)c_s + i\Omega_2 c_e \end{split}$$

Resonant interaction $\Delta_1 = \Delta_2 = 0$ $\Omega_{1,2} = \sqrt{2}\Omega \gg \gamma_e \Rightarrow$

 $c_g(t) = \cos^2(\Omega t)$ $c_e(t) = i\sqrt{2}\sin(\Omega t)\cos(\Omega t)$ $c_s(t) = -\sin^2(\Omega t)$



 $\frac{\partial}{\partial t} |\Psi(t)\rangle = -\frac{i}{\hbar} \mathcal{H}_{\Lambda} |\Psi(t)\rangle \Rightarrow$

$$\begin{split} |\Psi(t)\rangle &= c_g(t) |g\rangle + c_e(t) |e\rangle + c_s(t) |s\rangle \\ \frac{\partial}{\partial t} c_g &= i\Omega_1 c_e \\ \frac{\partial}{\partial t} c_e &= (i\Delta_1 - \gamma_e)c_e + i\Omega_1 c_g + i\Omega_2 c_s \\ \frac{\partial}{\partial t} c_s &= i(\Delta_1 - \Delta_2)c_s + i\Omega_2 c_e \end{split}$$

Non-resonant interaction $\Delta_1 \gg \Omega_{1,2}, \gamma_e \Rightarrow$

$$c_e(t) = i \int_0^t e^{(i\Delta_1 - \gamma_e)(t - t')} \left[\Omega_1 c_g(t') + \Omega_2 c_s(t') \right] dt'$$

$$\simeq i \left[\Omega_1 c_g(t) + \Omega_2 c_s(t) \right] \int_0^t e^{(i\Delta_1 - \gamma_e)(t - t')} dt' = \frac{\Omega_1 c_g + \Omega_2 c_s}{\Delta_1 + i\gamma_e} \qquad (\gamma_e t \gg 1)$$



 $\frac{\partial}{\partial t} |\Psi(t)\rangle = -\frac{i}{\hbar} \mathcal{H}_{\Lambda} |\Psi(t)\rangle \Rightarrow$

$$\begin{split} |\Psi(t)\rangle &= c_g(t) |g\rangle + c_e(t) |e\rangle + c_s(t) |s\rangle \\ \frac{\partial}{\partial t} c_g &= i\Omega_1 c_e \\ \frac{\partial}{\partial t} c_e &= (i\Delta_1 - \gamma_e)c_e + i\Omega_1 c_g + i\Omega_2 c_s \\ \frac{\partial}{\partial t} c_s &= i(\Delta_1 - \Delta_2)c_s + i\Omega_2 c_e \end{split}$$

Non-resonant interaction $\Delta_1 \gg \Omega_{1,2}, \gamma_e \Rightarrow$

$$c_e(t) = i \int_0^t e^{(i\Delta_1 - \gamma_e)(t - t')} \left[\Omega_1 c_g(t') + \Omega_2 c_s(t') \right] dt'$$

$$\simeq i \left[\Omega_1 c_g(t) + \Omega_2 c_s(t) \right] \int_0^t e^{(i\Delta_1 - \gamma_e)(t - t')} dt' = \frac{\Omega_1 c_g + \Omega_2 c_s}{\Delta_1 + i\gamma_e} \qquad (\gamma_e t \gg 1)$$

 \Rightarrow Two-photon (Raman) transition $|g\rangle \rightarrow |s\rangle$

$$\frac{\partial}{\partial t}c_s = -\left[i(S_s - \Delta_1 + \Delta_2) + \gamma_s\right]c_s + i\Omega_{\text{eff}} c_g$$

Stark shifts of $|g, e\rangle$: $S_{g,s} = \frac{|\Omega_{1,2}|^2}{\Delta_1}$; decays $\gamma_{g,s} = \gamma_e \frac{S_{g,s}}{\Delta_1} \to 0$

MAOP-AM, 30/08/22 – p. 20/25



$\frac{\partial}{\partial t}c_g = i\Omega_{\text{eff}} c_s$ $\frac{\partial}{\partial t}c_s = i\Delta_{\text{eff}}c_s + i\Omega_{\text{eff}} c_g$

$$\frac{\partial}{\partial t}c_g = i\Omega_{\text{eff}} c_s$$
$$\frac{\partial}{\partial t}c_s = i\Delta_{\text{eff}}c_s + i\Omega_{\text{eff}} c_g$$

 $\begin{aligned} & \textbf{Solution} \ c_g(0) = 1, \ c_s(0) = 0 \\ & c_g(t) = \cos(\bar{\Omega}_{\text{eff}}t) - i\frac{\Delta_{\text{eff}}}{2\bar{\Omega}_{\text{eff}}}\sin(\bar{\Omega}_{\text{eff}}t) \\ & c_s(t) = i\frac{\Omega_{\text{eff}}}{\bar{\Omega}_{\text{eff}}}\sin(\bar{\Omega}_{\text{eff}}t) \\ & \bar{\Omega}_{\text{eff}} \equiv \sqrt{\Omega_{\text{eff}}^2 + (\Delta_{\text{eff}}/2)^2} \end{aligned}$



$$\frac{\partial}{\partial t}c_g = i\Omega_{\text{eff}} c_s$$
$$\frac{\partial}{\partial t}c_s = i\Delta_{\text{eff}}c_s + i\Omega_{\text{eff}} c_g$$

 $\begin{aligned} & \text{Solution } c_g(0) = 1, \ c_s(0) = 0 \\ & c_g(t) = \cos(\bar{\Omega}_{\text{eff}}t) - i\frac{\Delta_{\text{eff}}}{2\bar{\Omega}_{\text{eff}}}\sin(\bar{\Omega}_{\text{eff}}t) \\ & c_s(t) = i\frac{\Omega_{\text{eff}}}{\bar{\Omega}_{\text{eff}}}\sin(\bar{\Omega}_{\text{eff}}t) \\ & \bar{\Omega}_{\text{eff}} \equiv \sqrt{\Omega_{\text{eff}}^2 + (\Delta_{\text{eff}}/2)^2} \end{aligned}$

Two-photon resonance $\Delta_{eff} = 0 \Rightarrow$

 $c_g(t) = \cos(\Omega_{\text{eff}}t)$ $c_s(t) = i\sin(\Omega_{\text{eff}}t)$

Two-photon Rabi oscillations





Two-photon resonance $\Delta_1 = \Delta_2 \equiv \Delta$

$$\mathcal{H}_{\Lambda} = -\hbar \begin{bmatrix} 0 & \Omega_1 & 0\\ \Omega_1 & \Delta & \Omega_2\\ 0 & \Omega_2 & 0 \end{bmatrix}$$



Two-photon resonance $\Delta_1 = \Delta_2 \equiv \Delta$ $\mathcal{H}_{\Lambda} = -\hbar \begin{bmatrix} 0 & \Omega_1 & 0 \\ \Omega_1 & \Delta & \Omega_2 \\ 0 & \Omega_2 & 0 \end{bmatrix}$

Eigenvalue problem $\mathcal{H}_{\Lambda} |\Psi\rangle = \hbar\lambda |\Psi\rangle$

$$\Rightarrow \lambda_0 = 0 \qquad \lambda_{\pm} = -(\Delta/2) \pm \bar{\Omega}$$

$$\begin{aligned} |D\rangle &= \frac{1}{\sqrt{N_0}} [\Omega_2 |g\rangle - \Omega_1 |s\rangle] \\ |B_{\pm}\rangle &= \frac{1}{\sqrt{N_{\pm}}} [\Omega_1 |g\rangle - \lambda_{\pm} |e\rangle + \Omega_2 |s\rangle] \end{aligned}$$



$$\bar{\Omega} = \sqrt{\Omega_1^2 + \Omega_2^2 + (\Delta/2)^2}$$



Two-photon resonance $\Delta_1 = \Delta_2 \equiv \Delta$ $\mathcal{H}_{\Lambda} = -\hbar \begin{bmatrix} 0 & \Omega_1 & 0\\ \Omega_1 & \Delta & \Omega_2\\ 0 & \Omega_2 & 0 \end{bmatrix}$

Eigenvalue problem $\mathcal{H}_{\Lambda} \ket{\Psi} = \hbar \lambda \ket{\Psi}$

 $\Rightarrow \lambda_0 = 0 \qquad \lambda_{\pm} = -(\Delta/2) \pm \bar{\Omega} \qquad \bar{\Omega} = \sqrt{\Omega_1^2 + \Omega_2^2 + (\Delta/2)^2}$

$$\begin{aligned} |D\rangle &= \frac{1}{\sqrt{N_0}} [\Omega_2 |g\rangle - \Omega_1 |s\rangle] \\ |B_{\pm}\rangle &= \frac{1}{\sqrt{N_{\pm}}} [\Omega_1 |g\rangle - \lambda_{\pm} |e\rangle + \Omega_2 |s\rangle] \end{aligned}$$

$$|D\rangle = \cos \Theta |g\rangle - \sin \Theta |s\rangle$$
 $\tan \Theta = \frac{\Omega_1}{\Omega_2}$ mixing angle

 $\Rightarrow |c_g|^2 = \cos^2 \Theta = \frac{\Omega_2^2}{\Omega_1^2 + \Omega_2^2} |c_e|^2 = 0 |c_s|^2 = \sin^2 \Theta = \frac{\Omega_1^2}{\Omega_1^2 + \Omega_2^2}$





Stimulated Raman Adiabatic Passage



Time-dependent Rabi frequencies $(\Delta = 0)$

•
$$\Omega_2(t_i) \gg \Omega_1(t_i) \quad [\Theta = 0]$$

 $\Rightarrow |\Psi(t_i)\rangle = |D\rangle = |g\rangle$

•
$$\Omega_2(t_f) \ll \Omega_1(t_f) \quad [\Theta = \frac{\pi}{2}]$$

 $\Rightarrow |\Psi(t_f)\rangle = |D\rangle = |s\rangle$

Stimulated Raman Adiabatic Passage



Time-dependent Rabi frequencies $(\Delta = 0)$

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$$\Omega_2(t_i) \gg \Omega_1(t_i) \quad [\Theta = 0]$$

 $\Rightarrow |\Psi(t_i)\rangle = |D\rangle = |g\rangle$

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$$\Omega_2(t_f) \ll \Omega_1(t_f) \quad [\Theta = \frac{\pi}{2}]$$

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Stimulated Raman Adiabatic Passage



Time-dependent Rabi frequencies $(\Delta = 0)$

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$$\Omega_2(t_i) \gg \Omega_1(t_i) \quad [\Theta = 0]$$

 $\Rightarrow |\Psi(t_i)\rangle = |D\rangle = |g\rangle$

•
$$\Omega_2(t_f) \ll \Omega_1(t_f) \quad [\Theta = \frac{\pi}{2}]$$

 $\Rightarrow |\Psi(t_f)\rangle = |D\rangle = |s\rangle$

Adiabatic following condition

$$|\dot{\Theta}| = \left| \frac{\dot{\Omega}_1 \Omega_2 - \Omega_1 \dot{\Omega}_2}{\Omega_1^2 + \Omega_2^2} \right| \ll |\lambda_{\pm} - \lambda_0|$$

Gaussian pulses of duration \boldsymbol{T}

 $\Rightarrow \Omega_{\max}T \gtrsim 10$



Problems & Questions



- Starting with the Hamiltonian \mathcal{H}_{Ξ} for a three-level atoms with Ξ configuration of levels, including the decay of levels $|e\rangle$ and $|s\rangle$, derive the coupled differential equations for the amplitudes c_g and c_s under the conditions $|\Delta_1 + \Delta_2| \ll |\Delta_{1,2}|$ and $\sqrt{\Delta_{1,2}^2 + \gamma_e} \gg \Omega_{1,2}$.
- Under what conditions, the ac Stark shifts $S_{g,s}$ of levels $|g,s\rangle$ in a Λ and Ξ systems are equal in magnitude and sign?
- In a V-system, can we perform STIRAP between levels $|e\rangle$ and $|s\rangle$ having large decay rates $\Gamma_{e,s}$?

Further Reading



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