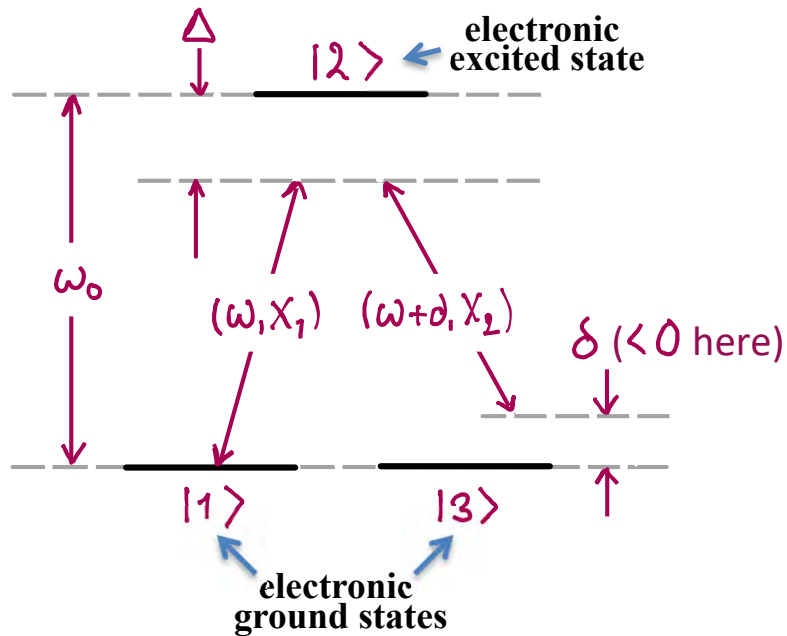


# Raman Coupling in 3-level Atoms

## Raman Coupling in 3-Level Systems

Consider an atom with this 3-level structure



For simplicity we set  $E_1 = E_3$  (no loss of generality)

Fields  $\left\{ \begin{array}{l} \text{at } \omega, \text{ coupling } |1\rangle, |2\rangle \text{ w/Rabi freq. } \chi_1 \\ \text{at } \omega + \delta, \text{ coupling } |3\rangle, |2\rangle \text{ w/Rabi freq. } \chi_2 \end{array} \right.$

The Hamiltonian for this system is ( $\chi_1, \chi_2$  real)

$$H = \frac{\hbar}{2} \begin{pmatrix} 0 & \chi_1(t) & 0 \\ \chi_1(t) & \omega_0 & \chi_2(t) \\ 0 & \chi_2(t) & 0 \end{pmatrix}$$

where

$$\chi_1(t) = \frac{\chi_1}{2} (e^{i\omega t} + e^{-i\omega t})$$

$$\chi_2(t) = \frac{\chi_2}{2} (e^{i(\omega+\delta)t} + e^{-i(\omega+\delta)t})$$

Setting  $|2(t)\rangle = a_1(t)|1\rangle + a_2(t)|2\rangle + a_3(t)|3\rangle$   
we get a S.E.

$$\dot{a}_1 = -i \frac{\chi_1}{2} (e^{i\omega t} + e^{-i\omega t}) a_2$$

$$\dot{a}_2 = -i\omega_0 a_2 - i \frac{\chi_1}{2} (e^{i\omega t} + e^{-i\omega t}) a_1 - i \frac{\chi_2}{2} (e^{i(\omega+\delta)t} + e^{-i(\omega+\delta)t}) a_3$$

$$\dot{a}_3 = -i \frac{\chi_2}{2} (e^{i(\omega+\delta)t} + e^{-i(\omega+\delta)t}) a_2$$

# Raman Coupling in 3-level Atoms

(5) Finally, the last term  $\propto \frac{e^{-i\Delta t}}{\Delta}$  can be ignored because it averages to zero on the timescale on which  $b_1, b_3$  evolve.

## Note:

The ground state amplitudes evolve slowly because  $X_1/\Delta, X_2/\Delta \ll 1$ , while the excited state amplitude evolves fast and adiabatically follows the instantaneous values of  $b_1, b_3$

Plug the solution for  $b_2(t)$  into the eqs. for  $b_1, b_3$



$$\begin{aligned} \dot{b}_1(t) &= i \frac{X_1^2}{4\Delta} b_1(t) + i \frac{X_1 X_2}{4\Delta} b_3(t) \\ \dot{b}_3(t) &= -i \left( \delta - \frac{X_2^2}{4\Delta} \right) b_3(t) + i \frac{X_1 X_2}{4\Delta} b_1(t) \end{aligned}$$

We simplify by making a final change of variables

$$C_1(t) = b_1(t) e^{-i \frac{X_1^2}{4\Delta} t}, \quad C_3(t) = b_3(t) e^{-i \frac{X_1^2}{4\Delta} t}$$



$$\begin{aligned} \dot{C}_1(t) &= i \frac{X_1 X_2}{4\Delta} C_3(t) \\ \dot{C}_3(t) &= -i \left( \delta + \frac{X_1^2 - X_2^2}{4\Delta} \right) C_3(t) + i \frac{X_1 X_2}{4\Delta} C_1(t) \end{aligned}$$

These are two-level equations!

**Physical Discussion:** We have an effective 2-level atom with effective Rabi Frequency and detuning.

$$\chi_{\text{eff}} = \frac{X_1 X_2}{2\Delta}, \quad \delta_{\text{eff}} = \delta + \frac{X_1^2 - X_2^2}{4\Delta}$$

Note that  $\chi_{\text{eff}} \sim X^2/\Delta$  while the excited state population  $P_2 \sim X^2/\Delta^2$ . This means that for large  $X, \Delta$  we can have large  $\chi_{\text{eff}}$  and no opportunity for spontaneous decay.



**Coherent Rabi oscillations and long lived superposition states**

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We simplify by making a final change of variables

$$c_1(t) = b_1(t) e^{-i \frac{\chi_1^2}{4\Delta} t}, \quad c_3(t) = b_3(t) e^{-i \frac{\chi_1^2}{4\Delta} t}$$



$$\dot{c}_1(t) = i \frac{\chi_1 \chi_2}{4\Delta} c_3(t)$$

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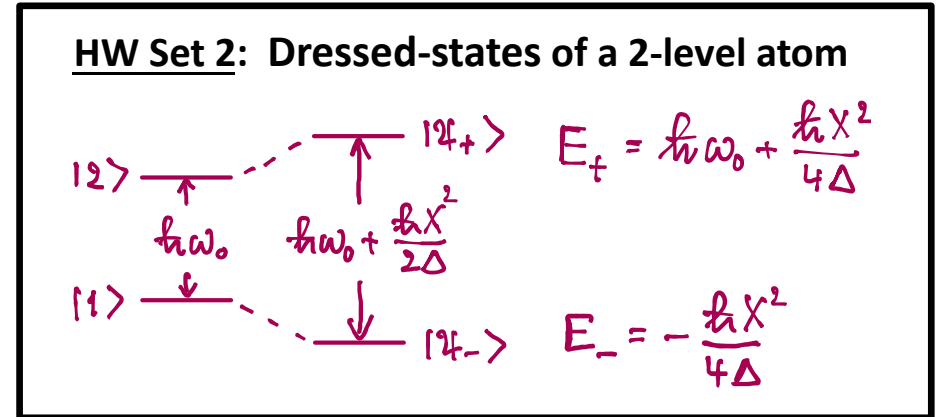
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Note also: The effective Raman detuning is shifted.



3-level system  $\Rightarrow$  ground state shifts  $\frac{\chi_1^2}{4\Delta}, \frac{\chi_2^2}{4\Delta}$

$\Rightarrow$  Differential ground state shift  $\frac{\chi_1^2 - \chi_2^2}{4\Delta}$

Final note: The atomic dipole  $\langle \hat{\mu} \rangle$  will have components that match the frequency and polarization of both driving fields, with amplitudes that depend on both fields.



**Non-Linear wave mixing, Breakdown of superposition principle**

# Raman Coupling in 3-level Atoms

Note also: The effective Raman detuning is shifted.

**HW Set 2: Dressed-states of a 2-level atom**

$|2\rangle \xrightarrow{\hbar\omega_0} |2_+\rangle$   $E_+ = \hbar\omega_0 + \frac{\hbar X^2}{4\Delta}$   
 $|1\rangle \xrightarrow{\hbar\omega_0 + \frac{\hbar X^2}{2\Delta}} |2_+\rangle$   
 $|1\rangle \xrightarrow{\hbar\omega_0} |2_-\rangle$   $E_- = -\frac{\hbar X^2}{4\Delta}$   
 $|2\rangle \xrightarrow{\hbar\omega_0 + \frac{\hbar X^2}{2\Delta}} |2_-\rangle$

3-level system  $\Rightarrow$  ground state shifts  $\frac{X_1^2}{4\Delta}, \frac{X_2^2}{4\Delta}$

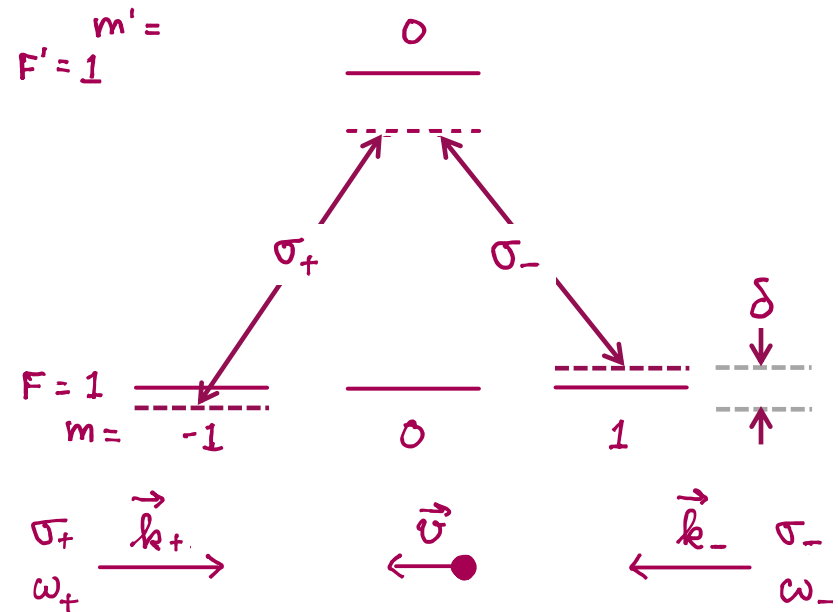
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**Final note:** The atomic dipole  $\langle \vec{d} \rangle$  will have components that match the frequency and polarization of both driving fields, with amplitudes that depend on both fields.



**Non-Linear wave mixing,  
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**Example: Velocity dependent Raman Coupling**



field freqs. in co-moving frame

velocity dependent Raman detuning

$$\left. \begin{aligned} \omega_+ &= \omega + k_+ v \\ \omega_- &= \omega - k_- v \end{aligned} \right\} \Rightarrow \delta = 2k_+ v$$

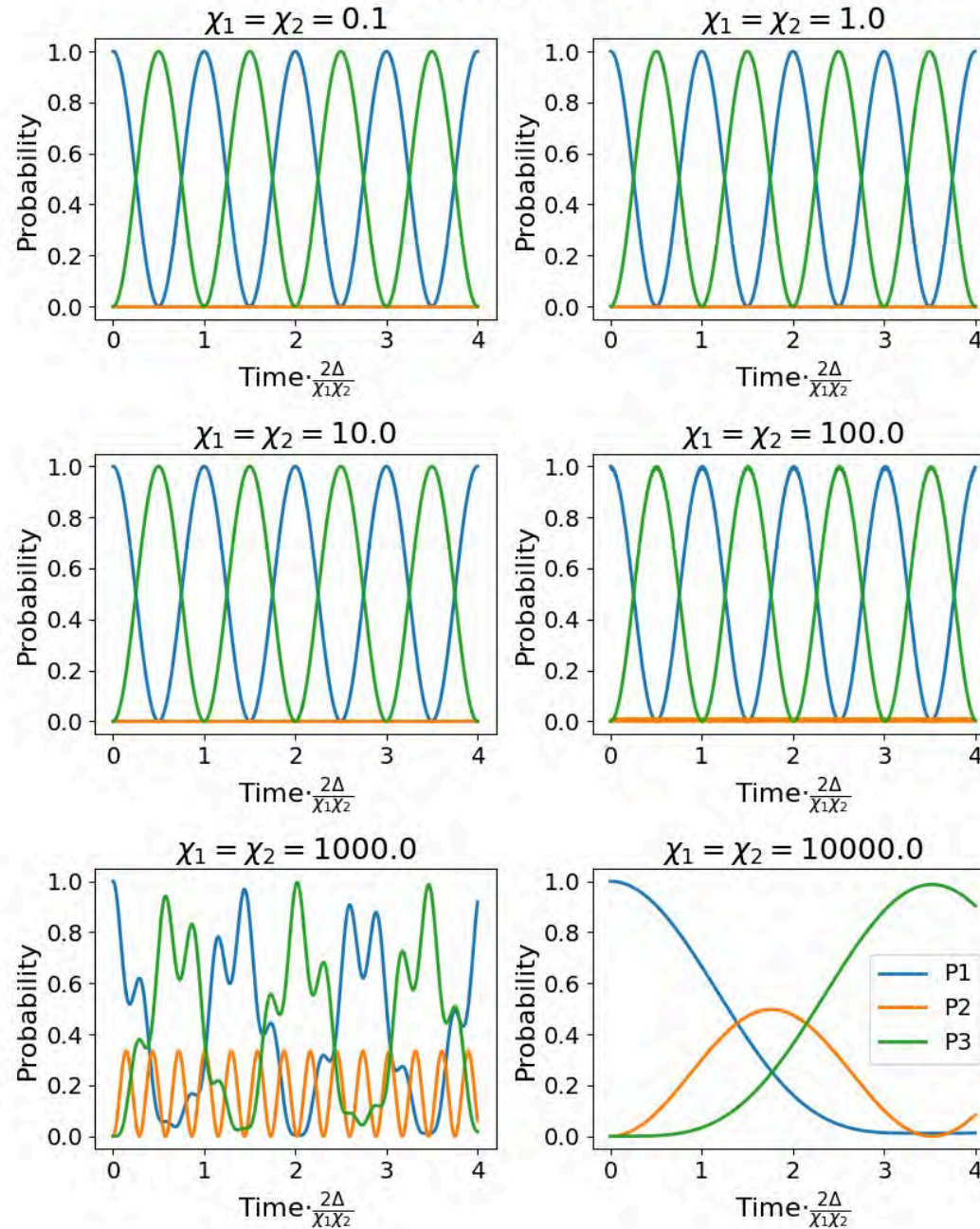
**Applications:**

- Doppler velocimetry
- Raman Cooling by velocity selective momentum transfer
- What if we apply a  $\pi/2$  Raman pulse?
- Atom Interferometry



**Numerical integration of the equations for the probability amplitudes in a 3-level Lambda system with zero Raman Detuning ( $\delta = 0$ ).**

$\Delta = 1000.0, \delta = 0.0$



# Density Matrix Description of 2-Level Atoms

Begin 02-18-2025

## Mental Warmup: What is a probability?

### (1) Example: Coin toss

- We can describe physical states by probability distributions
- Probabilities are assigned based on prior knowledge, updated when additional info becomes available
- As such, probability distributions are subjective ( states of knowledge)

### (2) Example: Quincunx

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### (4) Mixed Quantum & Classical Case

- We can easily envision a hybrid Quincunx that is part classical, part quantum.
- Physics needs an efficient description these kinds of intermediate situations



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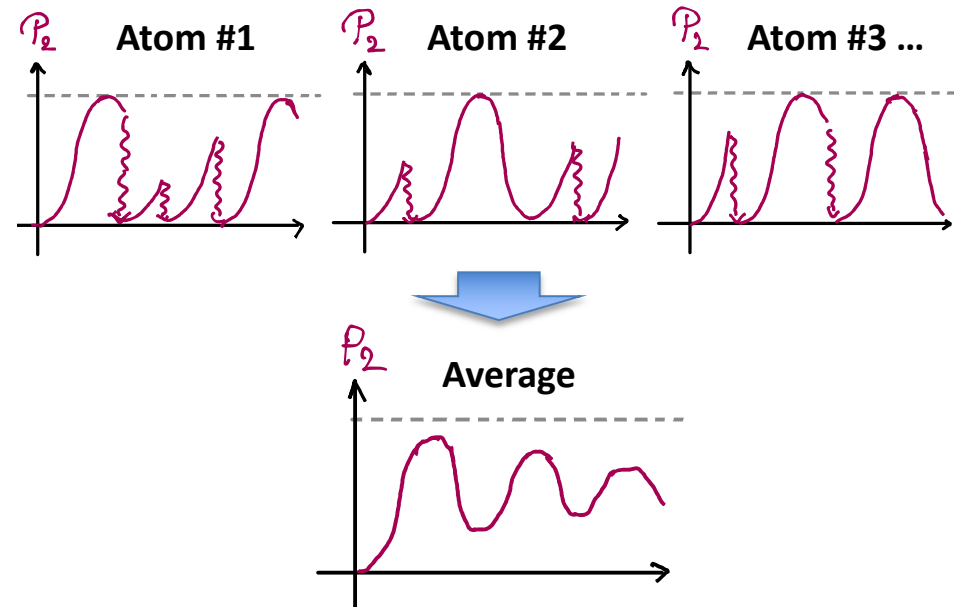
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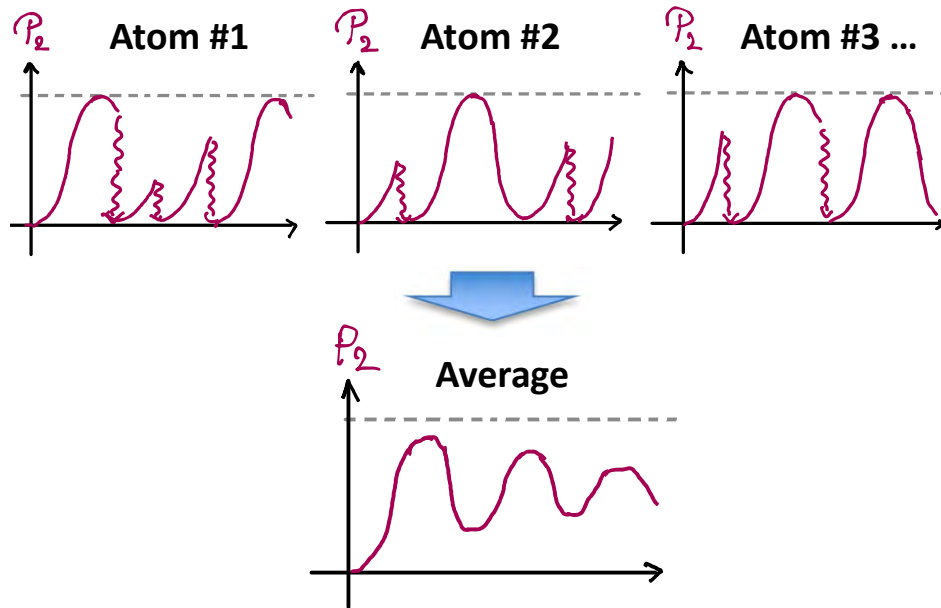
**Definition:** A system for which we know only the probabilities  $p_k$  of finding the system in state  $| \psi_k \rangle$  is said to be in a statistical mixture of states. Shorthand: mixed state.

Shorthand for non-mixed state: pure state

# Density Matrix Description of 2-Level Atoms

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Shorthand for non-mixed state: pure state

**Definition:** Density Operator for pure states

$$\rho(t) = |\psi(t)\rangle\langle\psi(t)|$$

**Definition:** Density Matrix

$$|\psi(t)\rangle = \sum_n C_n(t) |u_n\rangle \rightarrow$$

$$\rho_{pn}(t) = \langle u_p | \rho(t) | u_n \rangle = C_p(t) C_n^*(t)$$

**Definition:** Density Operator for mixed states

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**Note:** A pure state is just a mixed state for which one  $\rho_k = 1$  and the rest are zero.

The terms Density Operator and Density Matrix are used interchangeably

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Let  $A$  be an observable w/eigenvalues  $a_n$

Let  $P_n$  be the projector on the eigen-subspace of  $a_n$

For a pure state,  $\rho(t) = |\psi(t)\rangle\langle\psi(t)|$ , we have

$$(*) \quad \text{Tr} \rho(t) = \sum_n \rho_{nn}(t) = \sum_n |c_n|^2 = 1$$

$$\begin{aligned} (*) \quad \langle A \rangle &= \langle \psi(t) | A | \psi(t) \rangle = \sum_p \langle \psi(t) | A | u_p \rangle \langle u_p | \psi(t) \rangle \\ &= \sum_p \langle u_p | \psi(t) \rangle \langle \psi(t) | A | u_p \rangle = \sum_p \langle u_p | \rho(t) A | u_p \rangle \\ &= \text{Tr} [\rho(t) A] \quad (|u_p\rangle \text{ basis in } \mathcal{H}) \end{aligned}$$

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$$P(a_n) = \langle \psi(t) | P_n | \psi(t) \rangle = \text{Tr} [\rho(t) P_n]$$

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**Density Operator formalism is general !**

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Density Operator formalism is general !

## Important properties of the Density Operator

(1)  $\rho$  is Hermitian,  $\rho^\dagger = \rho \Rightarrow \rho$  is an observable

$\Rightarrow \exists$  basis in which  $\rho$  is diagonal

In this basis a pure state has one diagonal element = 1, the rest = 0

(2) Test for purity.

Pure:  $\rho^2 = \rho \Rightarrow \text{Tr} \rho^2 = 1$

Mixed:  $\rho^2 \neq \rho \Rightarrow \text{Tr} \rho^2 < 1$

(3) Schrödinger evolution does not change the  $p_k$

$\Rightarrow \left\{ \begin{array}{l} \text{Tr} \rho^2 \text{ is conserved} \\ \text{pure states stay pure} \\ \text{mixed states stay mixed} \end{array} \right.$

Changing pure  $\Rightarrow$  mixed requires non-Hamiltonian evolution – see Cohen Tannoudji D<sub>III</sub> & E<sub>III</sub>

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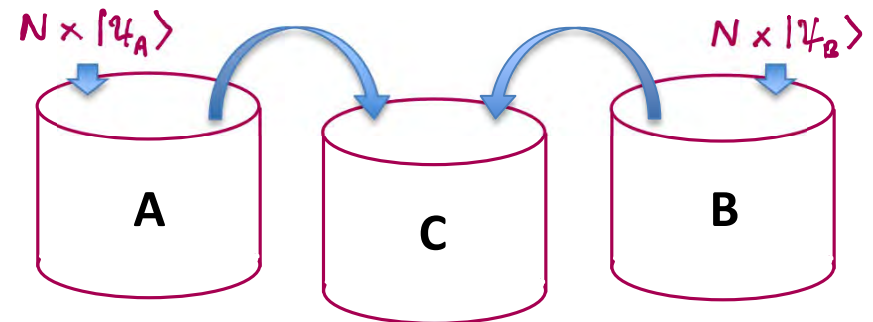
## A cooks recipe – interpretations of $\rho$

**Step 1** Add  $N$  atoms in state  $|\psi_A\rangle$  to bucket A  
Add  $N$  atoms in state  $|\psi_B\rangle$  to bucket B



We now have two ensembles, each of which consist of  $N$  atoms in a known pure state

**Step 2** Add buckets A and B to bucket C and stir.



Pick an atom from C

Which is Correct?

The atom is in a pure state but we don't know if it is in  $|\psi_A\rangle$  or  $|\psi_B\rangle$

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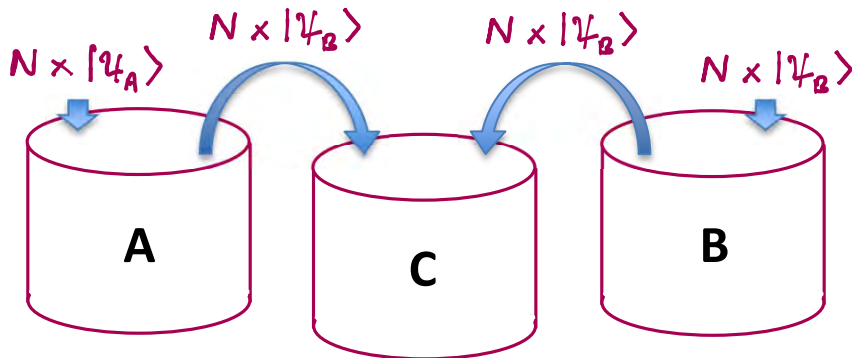
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$$\rho = \frac{1}{2} |\psi_A\rangle\langle\psi_A| + \frac{1}{2} |\psi_B\rangle\langle\psi_B|$$

There is no difference!

The two interpretations lead to identical predictions for any measurement we can do on atoms from C

Quantum Mechanics:

If two descriptions lead to identical predictions for observable outcomes then they are identical

Loosely, (i) is a *frequentist view*  
 (ii) is a *Bayesian view*

Quantum Bayesianism

Quantum States are States of Knowledge  
 (subjective)



# Density Matrix Description of 2-Level Atoms

## More about the Density Matrix

Let  $|\psi\rangle = \sum_j c_j |\mu_j\rangle$ , where  $\{|\mu_j\rangle\}$  is a basis and the index  $k$  labels members of the ensemble

### Populations:

(real-valued)

$$S_{nn} = \langle c_n^{(k)} c_n^{(k)*} \rangle_k = \langle |c_j^{(k)}|^2 \rangle_k$$

### Bayesianist:

Single system:  $S_{nn}$  is prob of observing the state  $|\mu_n\rangle$

### Frequentist:

Ensemble: The state  $|\mu_n\rangle$  occurs with frequency  $S_{nn}$

### Coherences:

(complex-valued)

$$S_{np} = \langle c_n^{(k)} c_p^{(k)*} \rangle_k, S_{pn} = S_{np}^*$$

**Note:** Defining  $c_q = |c_q| e^{i\theta_q}$  we have

$$|\langle c_n^{(k)} c_p^{(k)*} \rangle_k| = |\langle |c_n^{(k)}| |c_p^{(k)}| e^{i(\theta_n^{(k)} - \theta_p^{(k)})} \rangle_k| \leq \langle |c_n^{(k)}| |c_p^{(k)*}| \rangle_k$$

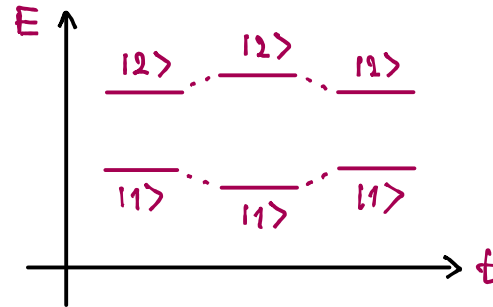
It follows that

$$S_{np} S_{pn} \leq S_{nn} S_{pp}$$

with = for pure states

$$S = \begin{pmatrix} S_{nn} & \dots & S_{np} \\ \vdots & \ddots & \vdots \\ S_{pn} & \dots & S_{pp} \end{pmatrix}$$

## Example: 2-level atom w/random perturbations



Perturbing events cause random phase shifts  $e^{i\theta}$  between states.

The ensemble average

$$S_{np} = \langle c_n c_p^* e^{i\theta} \rangle_k$$

is reduced by the randomly fluctuating phase

### Dipole Radiation:

$$\begin{aligned} \langle \hat{n} \rangle &= \text{Tr} [S \hat{n}] = \text{Tr} \left[ \begin{pmatrix} S_{11} & S_{12} \\ S_{21} & S_{22} \end{pmatrix} \begin{pmatrix} 0 & \vec{n}_{12} \\ \vec{n}_{21} & 0 \end{pmatrix} \right] \\ &= S_{12} \vec{n}_{21} + S_{21} \vec{n}_{12} = 2 \text{Re} [S_{12} \vec{n}_{21}] \end{aligned}$$

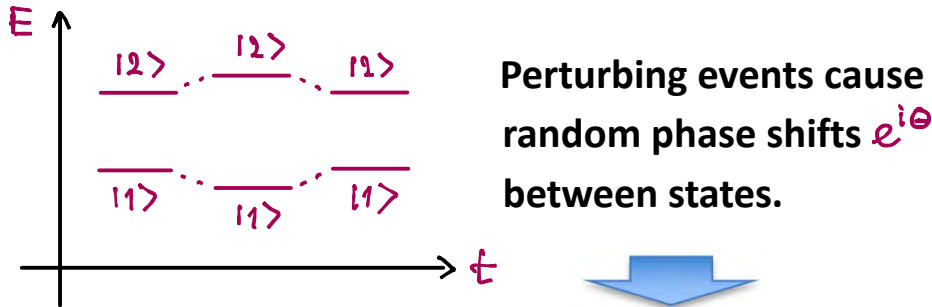
For an ensemble of pure states w/different  $\theta$

$$\langle \hat{n} \rangle = \langle \text{Re} [S_{12}^{(k)} \vec{n}_{21}] \rangle_k$$

Oscillating dipole w/phase that varies between atoms with different perturbation history

# Density Matrix Description of 2-Level Atoms

## Example: 2-level atom w/random perturbations



Perturbing events cause random phase shifts  $e^{i\theta}$  between states.

The ensemble average  $\rho_{np} = \langle c_n c_p^* e^{i\theta} \rangle_R$  is reduced by the randomly fluctuating phase

## Dipole Radiation:

$$\langle \hat{n} \rangle = \text{Tr} [\rho \hat{n}] = \text{Tr} \left[ \begin{pmatrix} \rho_{11} & \rho_{12} \\ \rho_{21} & \rho_{22} \end{pmatrix} \begin{pmatrix} 0 & \vec{n}_{12} \\ \vec{n}_{21} & 0 \end{pmatrix} \right]$$

$$= \rho_{12} \vec{n}_{21} + \rho_{21} \vec{n}_{12} = 2 \text{Re} [\rho_{12} \vec{n}_{21}]$$

For an ensemble of pure states w/different  $\theta$

$$\langle \hat{n} \rangle = \langle \text{Re} [\rho_{12}^{(k)} \vec{n}_{21}] \rangle_R$$

Oscillating dipole w/phase that varies between atoms with different perturbation history

## Time Evolution of the Density Matrix

**Challenge:** We need "equations of motion" that combine the Schrödinger Equation with the effect of processes that can change  $\text{Tr} \rho^2$  (measure of purity)

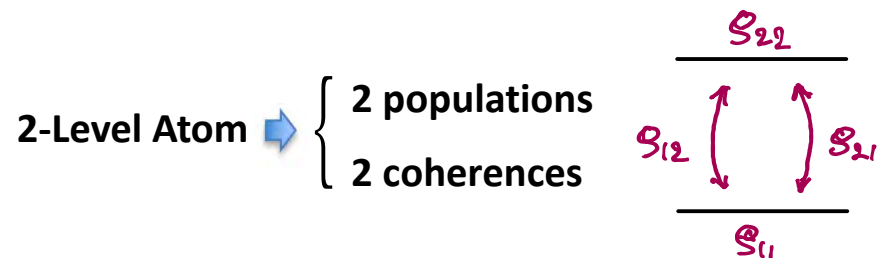
**Approach:** We do not have time for a rigorous derivation, so will rely on plausible arguments to justify the equations

**Schrödinger Evolution:** In general, we have

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

matrix elements

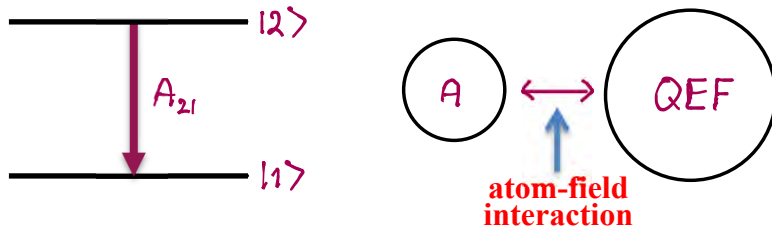
$$\dot{\rho}_{nm} = -\frac{i}{\hbar} \sum_{k=1,2} (H_{nk} \rho_{km} - \rho_{nk} H_{km})$$



# Density Matrix Description of 2-Level Atoms

## Spontaneous Decay

This process occurs due to interaction between the Atom and the Quantized Electromagnetic Field



## Final OPTI 544 Lectures:

$|\psi(0)\rangle = |2\rangle_A |vac\rangle_{QEF}$  evolution over time  $t$   
 $|\psi(t)\rangle = c_{2,0}(t) |2\rangle_A |vac\rangle_{QEF} + \sum_k c_{1,1k}(t) |1\rangle_A |n_k=1\rangle_{QEF}$   
 photon "in the atom"                      photon in field mode  $k$

Cannot recover info in continuum of field modes



Probability  $|c_{2,0}(t)|^2$  of having **no decay**

Probability  $\sum_k |c_{1,1k}(t)|^2$  of having **decay**

No Coherence established between states  $|1\rangle, |2\rangle$

Conclusion: Decay moves population  $|2\rangle \rightarrow |1\rangle$  at rate  $A_{21}$ , damps coherence at rate  $A_{21}/2$



$$\dot{\rho}_{11} = A_{21} \rho_{22}, \quad \dot{\rho}_{22} = -A_{21} \rho_{11}$$

$$\dot{\rho}_{12} = -\frac{A_{21}}{2} \rho_{12} = \dot{\rho}_{21}^*$$

## Putting it all together:

$$\dot{\rho}_{11} = -\Gamma_1 \rho_{11} + A_{21} \rho_{22} - \frac{i}{2} (X \rho_{12} - X^* \rho_{21})$$

$$\dot{\rho}_{22} = -\Gamma_2 \rho_{22} - A_{21} \rho_{11} + \frac{i}{2} (X \rho_{12} - X^* \rho_{21})$$

$$\dot{\rho}_{12} = (i\Delta - \beta) \rho_{12} + \frac{iX^*}{2} (\rho_{22} - \rho_{11}) = \dot{\rho}_{21}^*$$

where  $\beta = \frac{\Gamma_1}{2} + \frac{A_{21}}{2} + \frac{\Gamma_2}{2}$

These are our desired

**Density Matrix Equations of Motion**

# Emission and Absorption – Population Rate Equations