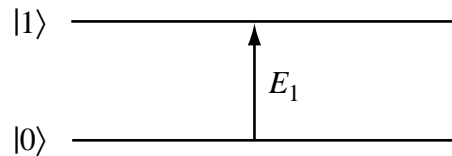


# SCHRÖDINGER EQUATION FOR 2-LEVEL SYSTEMS (1)



- A 2-level model is appropriate whenever only 2 energy levels participate significantly in an interaction with an external electromagnetic field
  - ▷ Also applies to unusual systems that have only two bound states
- State vector in the Schrödinger picture ( $c_0, c_1 =$  probability amplitudes):

$$|\psi(t')\rangle = c_0(t') |0\rangle + c_1(t') |1\rangle$$

where  $t' = t - nz/c$  for a propagation problem

- Interpretation:  $|c_k(t')|^2$  is the probability of finding the system in state  $k$  at time  $t'$  (where  $k \in (0 : 1)$ )
- Time-dependent Schrödinger equation:

$$i\hbar \frac{\partial}{\partial t'} |\psi(t')\rangle = \mathbf{H} |\psi(t')\rangle$$

## SCHRÖDINGER EQUATION FOR 2-LEVEL SYSTEMS (2)

- State vectors:

$$|0\rangle = \begin{pmatrix} 1 \\ 0 \end{pmatrix}, \quad |1\rangle = \begin{pmatrix} 0 \\ 1 \end{pmatrix}$$

- Hamiltonian for an isolated 2-level system:

$$\begin{aligned} H_0 &= \begin{matrix} & \langle 0 | & \langle 1 | \\ \begin{matrix} |0\rangle \\ |1\rangle \end{matrix} & \begin{pmatrix} 0 & 0 \\ 0 & E_1 \end{pmatrix} \end{matrix} = E_1 |1\rangle\langle 1| \\ &= \frac{E_1}{2} \mathbf{1} - \frac{E_1}{2} \boldsymbol{\sigma}_3 \quad \text{where} \quad \boldsymbol{\sigma}_3 = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} \end{aligned}$$

- Transition operators (also known as raising or lowering operators):

$$\boldsymbol{\sigma}_+ = \begin{matrix} & \langle 0 | & \langle 1 | \\ \begin{matrix} |0\rangle \\ |1\rangle \end{matrix} & \begin{pmatrix} 0 & 0 \\ 1 & 0 \end{pmatrix} \end{matrix} = |1\rangle\langle 0|, \quad \boldsymbol{\sigma}_- = \begin{matrix} & \langle 0 | & \langle 1 | \\ \begin{matrix} |0\rangle \\ |1\rangle \end{matrix} & \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix} \end{matrix} = |0\rangle\langle 1|$$

## SCHRÖDINGER EQUATION FOR 2-LEVEL SYSTEMS (3)

- Hermitian operators that have only off-diagonal matrix elements:

$$\sigma_1 = \begin{array}{c} \langle 0 | \quad \langle 1 | \\ |0\rangle \\ |1\rangle \end{array} \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} = |0\rangle\langle 1| + |1\rangle\langle 0|$$

$$\sigma_2 = \begin{array}{c} \langle 0 | \quad \langle 1 | \\ |0\rangle \\ |1\rangle \end{array} \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix} = i |1\rangle\langle 0| - i |0\rangle\langle 1|$$

- ▷ The matrices  $\sigma_1$ ,  $\sigma_2$ ,  $\sigma_3$  are called the **Pauli spin matrices**
- ▷ Every  $2 \times 2$  Hermitian matrix can be expressed as a linear combination of Pauli spin matrices and the unit matrix:

$$\begin{pmatrix} a & b \\ b^* & d \end{pmatrix} = \frac{1}{2}(a + d)\mathbf{1} + \frac{1}{2}(b + b^*)\sigma_1 + \frac{1}{2}i(b - b^*)\sigma_2 + \frac{1}{2}(a - d)\sigma_3$$

- ▷ Every physically observable quantity (such as the dipole moment) corresponds to a Hermitian operator and can be expanded in Pauli matrices

## SCHRÖDINGER EQUATION FOR 2-LEVEL SYSTEMS (4)

- For a 2-level system perturbed by an electric-dipole interaction, the Hamiltonian is

$$H = H_0 - \boldsymbol{\mu}(\hat{\boldsymbol{\mu}} \cdot \mathbf{E}(t'))$$

where  $\mathbf{E}(t')$  is the time-dependent electric field,  $\boldsymbol{\mu}$  is the dipole operator, and  $\hat{\boldsymbol{\mu}}$  is a unit vector that points in the direction of the molecular or atomic dipole moment

- ▷ If  $\langle \boldsymbol{\mu} \rangle \neq 0$  when  $\mathbf{E} = 0$ , then there is a permanent dipole moment
  - We assume that there is *only* an induced dipole moment
  - Then  $\hat{\boldsymbol{\mu}}$  points in the direction of  $\mathbf{E}$ :

$$\hat{\boldsymbol{\mu}} \cdot \mathbf{E}(t') = E(t') \cos(-\omega t' + \phi(t'))$$

where  $\omega$  is the field frequency,  $E(t')$  is the electric field envelope, and  $\phi(t')$  is a time-varying phase

- ▷ If there is no permanent dipole moment, then  $\boldsymbol{\mu}$  is completely off-diagonal:

$$\boldsymbol{\mu} = \mu_1 \boldsymbol{\sigma}_1 + \mu_2 \boldsymbol{\sigma}_2$$

## SCHRÖDINGER EQUATION FOR 2-LEVEL SYSTEMS (5)

- Time-dependent Schrödinger equation (TDSE) for the probability amplitudes if the driving field is quasimonochromatic:

$$i\hbar \frac{dc_0}{dt'} = -\mu E(t') \cos(-\omega t' + \phi(t')) c_1$$
$$i\hbar \frac{dc_1}{dt'} = E_1 c_1 - \mu E(t') \cos(-\omega t' + \phi(t')) c_0$$

where  $\mu$  is the dipole transition moment

- ▷ It can be shown that we need only  $\mu\sigma_1$  to represent the dipole operator
- The nature of the solution depends strongly on the time dependence of  $E$ 
  - ▷  $E$  switched on or off suddenly
  - ▷  $E$  switched on or off very slowly (adiabatically)

## SCHRÖDINGER EQUATION FOR 2-LEVEL SYSTEMS (6)

- To simplify the TDSE, make the unitary transformation

$$\tilde{c}_0 = c_0, \quad \tilde{c}_1 = e^{i\omega t'} c_1$$

- Resulting equation for the transformed probability amplitude  $\tilde{c}_0$ :

$$\begin{aligned} i\hbar \frac{d\tilde{c}_0}{dt'} &= -\mu E(t') \cos(-\omega t' + \phi(t')) e^{-i\omega t'} \tilde{c}_1 \\ \frac{d\tilde{c}_0}{dt'} &= \frac{i\mu E(t')}{2\hbar} \left( e^{-i\omega t' + i\phi} + e^{i\omega t' - i\phi(t')} \right) e^{-i\omega t'} \tilde{c}_1 \\ &= \frac{i\mu E(t')}{2\hbar} \left( e^{-2i\omega t' + i\phi(t')} + e^{-i\phi(t')} \right) \tilde{c}_1 \end{aligned}$$

- **Rotating-wave approximation (RWA):**

▷ If  $E$  and  $\phi$  vary slowly on the time scale of an optical period ( $2\pi/\omega$ ), then the rapidly oscillating term  $e^{-2i\omega t' + i\phi(t')}$  makes a very small net contribution, and can be neglected

## SCHRÖDINGER EQUATION FOR 2-LEVEL SYSTEMS (7)

- TDSE in the RWA:

$$\begin{aligned} i\hbar \frac{d\tilde{c}_0}{dt'} &= i\Omega^* \tilde{c}_1 \\ i\hbar \frac{d\tilde{c}_1}{dt'} &= i\Delta \tilde{c}_1 + i\Omega \tilde{c}_0 \end{aligned}$$

- The RWA TDSE can be written in the same form as the full TDSE:

$$\frac{d}{dt'} \begin{pmatrix} \tilde{c}_0 \\ \tilde{c}_1 \end{pmatrix} = -i\mathbf{H}^{\text{eff}} \begin{pmatrix} \tilde{c}_0 \\ \tilde{c}_1 \end{pmatrix}$$

where

$$-\mathbf{H}^{\text{eff}} = \begin{pmatrix} 0 & \Omega^* \\ \Omega & \Delta \end{pmatrix}$$

$$\Omega = \frac{\mu\mathcal{E}(t')}{2\hbar}, \quad \mathcal{E}(t') = E(t')e^{i\phi(t')}, \quad \Delta = \omega - \frac{E_1}{\hbar}$$

- ▷  $\mathcal{E}$  is the slowly varying complex field envelope
- ▷  $\Delta$  is the detuning (of the laser frequency) from resonance with the  $|0\rangle \leftrightarrow |1\rangle$  transition frequency

## EXACT SOLUTION FOR 2-LEVEL SYSTEMS (1)

- Goal: Obtain an exact solution to the TDSE when  $\Omega$  and  $\Delta$  are constants
- Write  $H^{\text{eff}}$  as a linear combination of Pauli matrices and the unit matrix:

$$-H^{\text{eff}} = \begin{pmatrix} 0 & \Omega^* \\ \Omega & \Delta \end{pmatrix} = \frac{\Delta}{2} \mathbf{1} + \text{Re} [\Omega] \boldsymbol{\sigma}_1 + \text{Im} [\Omega] \boldsymbol{\sigma}_2 - \frac{\Delta}{2} \boldsymbol{\sigma}_3 = \frac{\Delta}{2} \mathbf{1} + \omega_R (\hat{\mathbf{n}} \cdot \boldsymbol{\sigma})$$

where

$$\hat{\mathbf{n}} = \frac{1}{\omega_R} \begin{pmatrix} \text{Re} [\Omega] \\ \text{Im} [\Omega] \\ -\frac{\Delta}{2} \end{pmatrix},$$

$$\omega_R = \left[ \left( \frac{\Delta}{2} \right)^2 + |\Omega|^2 \right]^{1/2},$$

and

$$\hat{\mathbf{n}} \cdot \boldsymbol{\sigma} = \frac{1}{\omega_R} \begin{pmatrix} -\frac{\Delta}{2} & \Omega^* \\ \Omega & \frac{\Delta}{2} \end{pmatrix}$$

## EXACT SOLUTION FOR 2-LEVEL SYSTEMS (2)

- The solution of the RWA TDSE,

$$\frac{d}{dt'} \begin{pmatrix} \tilde{c}_0 \\ \tilde{c}_1 \end{pmatrix} = -iH^{\text{eff}} \begin{pmatrix} \tilde{c}_0 \\ \tilde{c}_1 \end{pmatrix},$$

is

$$\begin{pmatrix} \tilde{c}_0(t) \\ \tilde{c}_1(t) \end{pmatrix} = \exp \left\{ i \frac{\Delta}{2} t \right\} \exp \{ i \omega_R t (\hat{\mathbf{n}} \cdot \boldsymbol{\sigma}) \} \begin{pmatrix} \tilde{c}_0(0) \\ \tilde{c}_1(0) \end{pmatrix}$$

where

$$\exp \{ i \omega_R t (\hat{\mathbf{n}} \cdot \boldsymbol{\sigma}) \} = \cos(\omega_R t) \mathbf{1} + i \sin(\omega_R t) (\hat{\mathbf{n}} \cdot \boldsymbol{\sigma})$$

$$= \begin{pmatrix} \cos(\omega_R t) - i \frac{\Delta}{2\omega_R} \sin(\omega_R t) & i \frac{\Omega^*}{\omega_R} \sin(\omega_R t) \\ i \frac{\Omega}{\omega_R} \sin(\omega_R t) & \cos(\omega_R t) + i \frac{\Delta}{2\omega_R} \sin(\omega_R t) \end{pmatrix}$$

**EXACT SOLUTION FOR 2-LEVEL SYSTEMS (3)**

- The solution of the RWA TDSE for the initial conditions  $\tilde{c}_0(0) = 1$ ,  $\tilde{c}_1(0) = 0$  is

$$\tilde{c}_0(t) = \exp\left\{i\frac{\Delta}{2}t\right\} \left[ \cos(\omega_R t) - i\frac{\Delta}{2\omega_R} \sin(\omega_R t) \right],$$

$$\tilde{c}_1(t) = i \exp\left\{i\frac{\Delta}{2}t\right\} \frac{\Omega}{\omega_R} \sin(\omega_R t)$$

- The populations for these initial conditions are

$$|\tilde{c}_0(t)|^2 = \cos^2(\omega_R t) + \frac{\Delta^2}{4\omega_R^2} \sin^2(\omega_R t) \quad (\text{lower state})$$

$$|\tilde{c}_1(t)|^2 = \frac{|\Omega|^2}{\omega_R^2} \sin^2(\omega_R t) \quad (\text{upper state})$$

▷ Since  $\sin^2(\omega_R t) = (1 - \cos(2\omega_R t))/2$ , the populations oscillate at the frequency  $2\omega_R$  (the **Rabi frequency**)

## DRESSED STATES FOR 2-LEVEL SYSTEMS (1)

- If  $E$  and  $\phi$  are constant (or extremely slowly varying) in time, then it makes sense to look for the eigenvalues and eigenvectors of the effective Hamiltonian in the RWA
  - ▷ The eigenvectors of  $\mathbf{H}^{\text{eff}}$  are called **dressed states**, because they are the states of the 2-level system after it has been “dressed” by the driving field
  - ▷ The eigenvalues of  $\mathbf{H}^{\text{eff}}$  are called “dressed-state energies”
  - ▷ The eigenvalue equation

$$\det [-\mathbf{H}^{\text{eff}} - \lambda \mathbf{1}] = 0$$

leads to the eigenvalues

$$\lambda_0 = \frac{\Delta}{2} - \frac{1}{2} \frac{\Delta}{|\Delta|} (\Delta^2 + 4|\Omega|^2)^{1/2} \xrightarrow{\Delta \rightarrow 0} -(\text{energy of } |0\rangle) / \hbar$$
$$\lambda_1 = \frac{\Delta}{2} + \frac{1}{2} \frac{\Delta}{|\Delta|} (\Delta^2 + 4|\Omega|^2)^{1/2} \xrightarrow{\Delta \rightarrow 0} -(\text{energy of } |1\rangle) / \hbar$$

**DRESSED STATES FOR 2-LEVEL SYSTEMS (2)**

- Dressed states:

▷ The eigenvector equations

$$-H^{\text{eff}} |\lambda_0\rangle = \lambda_0 |\lambda_0\rangle, \quad -H^{\text{eff}} |\lambda_1\rangle = \lambda_1 |\lambda_1\rangle$$

lead to the normalized eigenvectors

$$|\lambda_0\rangle = \begin{pmatrix} \frac{|\Omega|}{(\lambda_0^2 + |\Omega|^2)^{1/2}} \\ \frac{|\Omega| \lambda_0}{\Omega^* (\lambda_0^2 + |\Omega|^2)^{1/2}} \end{pmatrix}$$

$$|\lambda_1\rangle = \begin{pmatrix} \frac{|\Omega|}{(\lambda_1^2 + |\Omega|^2)^{1/2}} \\ \frac{|\Omega| \lambda_1}{\Omega^* (\lambda_1^2 + |\Omega|^2)^{1/2}} \end{pmatrix}$$

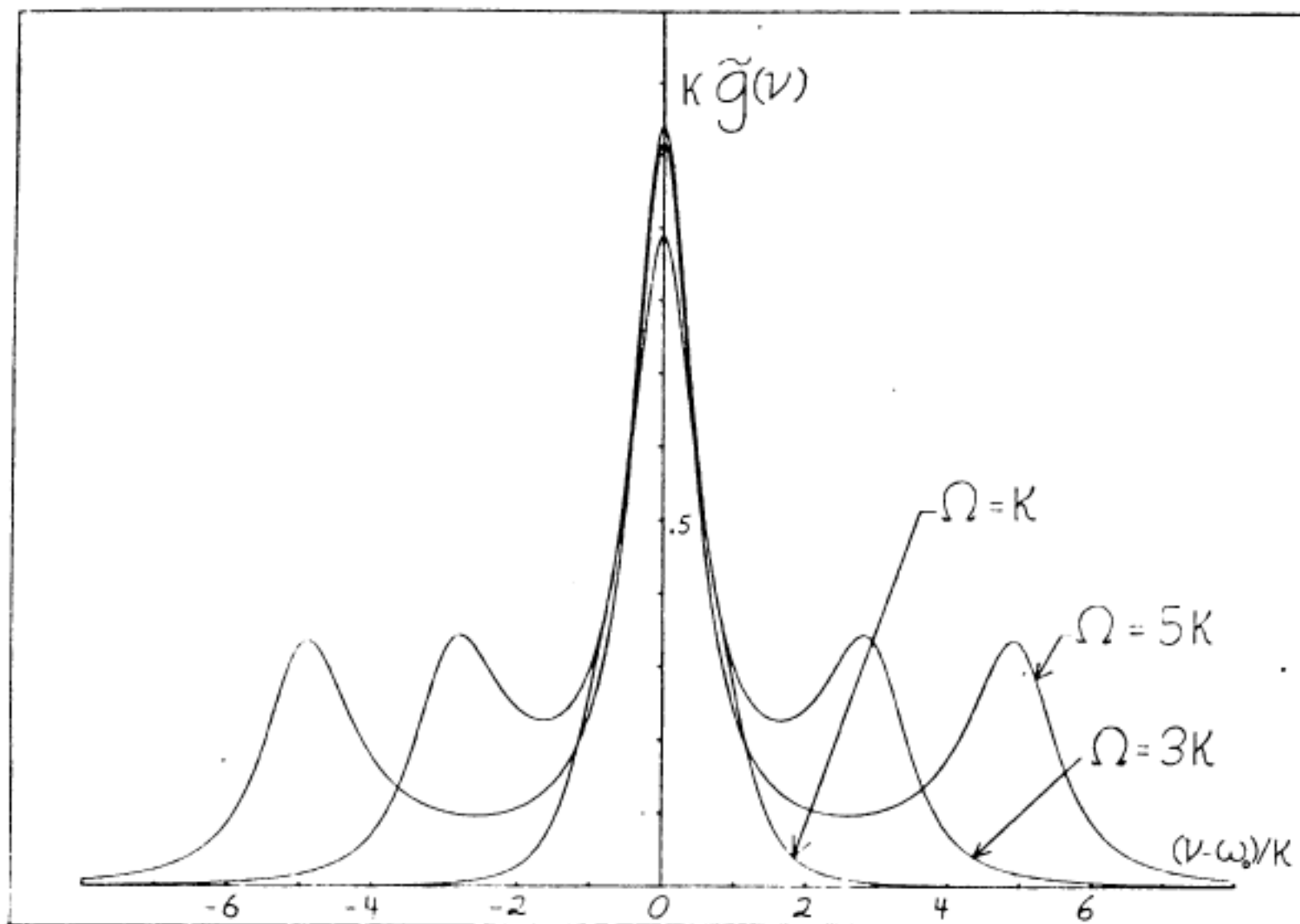


FIG. 1. Spectral density  $\tilde{g}(\nu)$  for a two-level atom driven exactly on resonance.

**SUDDEN APPROXIMATION FOR A 2-LEVEL SYSTEM (1)**

- Model a laser pulse as a field with an envelope  $E$  that is switched on suddenly at  $t' = 0$  and is constant afterwards
  - ▷ At times  $t' < 0$ , the 2-level system is in the ground state,  $|0\rangle$
  - ▷ At time  $t' = 0$ , the system is still in  $|0\rangle$ , which can be expressed as

$$|\psi(0)\rangle = \langle\lambda_0|0\rangle |\lambda_0\rangle + \langle\lambda_1|0\rangle |\lambda_1\rangle$$

in terms of the dressed states for times  $t' > 0$

- ▷ At times  $t' > 0$ , the dressed states are eigenvectors of the Hamiltonian, and each dressed state evolves in time at its own eigenfrequency:

$$|\psi(t')\rangle = \langle\lambda_0|0\rangle |\lambda_0\rangle e^{i\lambda_0 t'} + \langle\lambda_1|0\rangle |\lambda_1\rangle e^{i\lambda_1 t'}$$

- Probability that the system is in the upper bare state,  $|1\rangle$ , is

$$|\langle 1|\psi(t')\rangle|^2 = |\langle\lambda_0|0\rangle\langle 1|\lambda_0\rangle + \langle\lambda_1|0\rangle\langle 1|\lambda_1\rangle e^{i(\lambda_1-\lambda_0)t'}|^2$$

# SCHRÖDINGER EQUATION FOR MULTILEVEL SYSTEMS

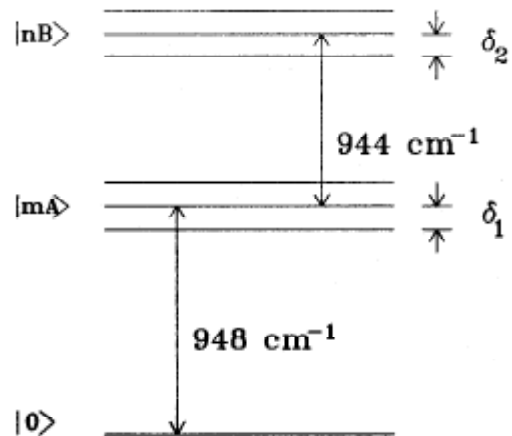
- Notation for states  $|mA\rangle$  and energy eigenvalues  $E_{mA}$  :

$m$  = band number

$A$  = all other quantum numbers

$c_{mA}$  = amplitude of  $|mA\rangle$

⋮



## SCHRÖDINGER EQUATION FOR MULTILEVEL SYSTEMS

- Schrödinger equation neglecting all far-off-resonance terms (i.e., in the rotating-wave approximation):

$$\frac{d\tilde{c}_{mA}}{dt'} = i \left( m\omega - \frac{E_{mA}}{\hbar} \right) \tilde{c}_{mA} + \frac{i}{\hbar} \sum_B \mu_{mA;m+1,B} \mathcal{E}(t')^* \tilde{c}_{m+1,B} + \mu_{mA;m-1,B} \mathcal{E}(t') \tilde{c}_{m-1,B}$$

where

$$\tilde{c}_{mA} = c_{mA} e^{im\omega t'} \quad \text{varies slowly}$$

$\omega$  = frequency of optical field

$\mathcal{E}(t')$  = envelope of optical field

## DRESSED STATES FOR MULTILEVEL SYSTEMS

- Effective Hamiltonian (constant if field envelope  $\mathcal{E}$  is constant):

$$H_{mA; mA}^{\text{eff}} = m\omega - \frac{E_{mA}}{\hbar},$$

$$H_{mA; m+1, B}^{\text{eff}} = \frac{\mu_{mA; m+1, B} \mathcal{E}(t')^*}{2\hbar}, \quad H_{mA; m-1, B}^{\text{eff}} = \frac{\mu_{mA; m-1, B} \mathcal{E}(t')}{2\hbar}$$

- The eigenvectors of  $\mathbf{H}^{\text{eff}}$  are the states “dressed” by the laser field:

$$-\mathbf{H}^{\text{eff}} |\lambda; \mathcal{E}(t')\rangle = \epsilon_\lambda(t') |\lambda; \mathcal{E}(t')\rangle$$

- Adiabatic time development when  $|\psi(t'_0)\rangle$  is the dressed state  $|\lambda; \mathcal{E}(t'_0)\rangle$ :

$$|\psi(t')\rangle = \exp \left[ -i \int_{t'_0}^{t'} \epsilon_\lambda(t'') dt'' \right] |\lambda; \mathcal{E}(t')\rangle$$

## THE SUDDEN APPROXIMATION (1)

- The laser field envelope  $\mathcal{E}(t')$  is switched instantaneously from zero to the constant value  $\mathcal{E}_0$  at  $t' = t'_0$
- Immediately after the pulse is switched on, the system is in the superposition of dressed states that gives the state  $|\psi_0\rangle$  at time  $t'_0$ :

$$|\psi(t'_0 + \epsilon)\rangle = \sum_{\lambda} \langle \lambda; \mathcal{E}_0 | \psi_0 \rangle |\lambda; \mathcal{E}_0\rangle$$

- The time development of the probability amplitudes is

$$\tilde{c}_{mA}(t') = \sum_{\lambda} \langle mA | \lambda; \mathcal{E}_0 \rangle e^{i\lambda t'} \langle \lambda; \mathcal{E}_0 | \psi_0 \rangle$$

- The polarization is

$$\mathcal{P}(\mathbf{r}_T, z', t') = 2iN \langle \psi(t') | \boldsymbol{\mu} | \psi(t') \rangle = \sum_{\lambda, \lambda'} e^{i(\lambda' - \lambda)t'} \mathcal{P}_{\lambda, \lambda'}$$

- **Conclusion:** Sidebands are generated at the generalized Rabi frequency  $\lambda - \lambda'$  corresponding to each pair of dressed states  $|\lambda; \mathcal{E}_0\rangle, |\lambda'; \mathcal{E}_0\rangle$

## THE SUDDEN APPROXIMATION (2)

- The polarization matrix element  $\mathcal{P}_{\lambda,\lambda'}$  in the state that evolves from  $|\psi_0\rangle$  is

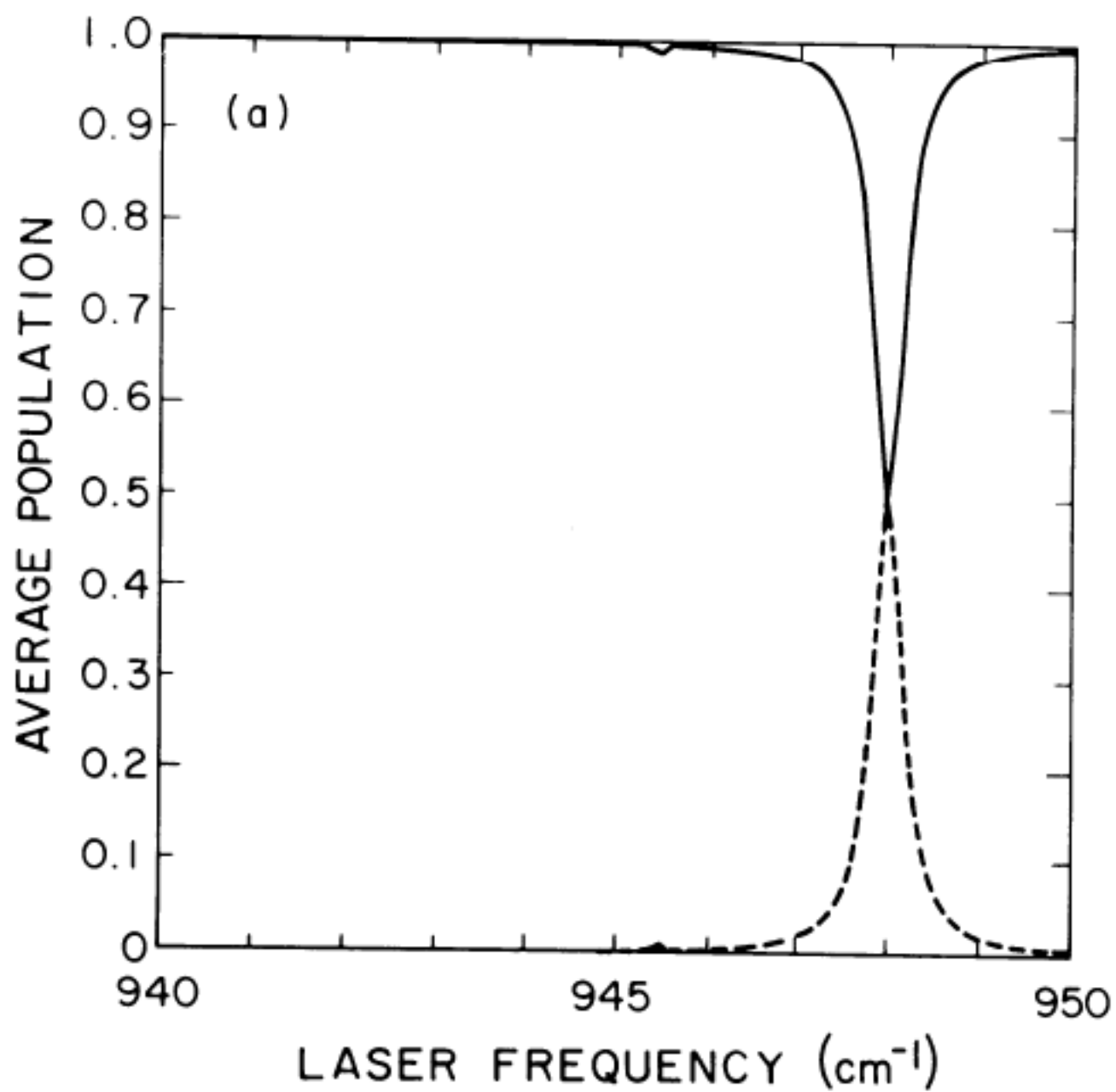
$$\mathcal{P}_{\lambda,\lambda'} = \langle\psi_0 | \lambda; \mathcal{E}_0\rangle \sum_{mA} \sum_{m'A'} \langle\lambda; \mathcal{E}_0 | mA\rangle \mu_{mA;m'A'} \langle m'A' | \lambda'; \mathcal{E}_0\rangle \langle\lambda'; \mathcal{E}_0 | \psi_0\rangle$$

- Model for illustrating multiphoton resonances:

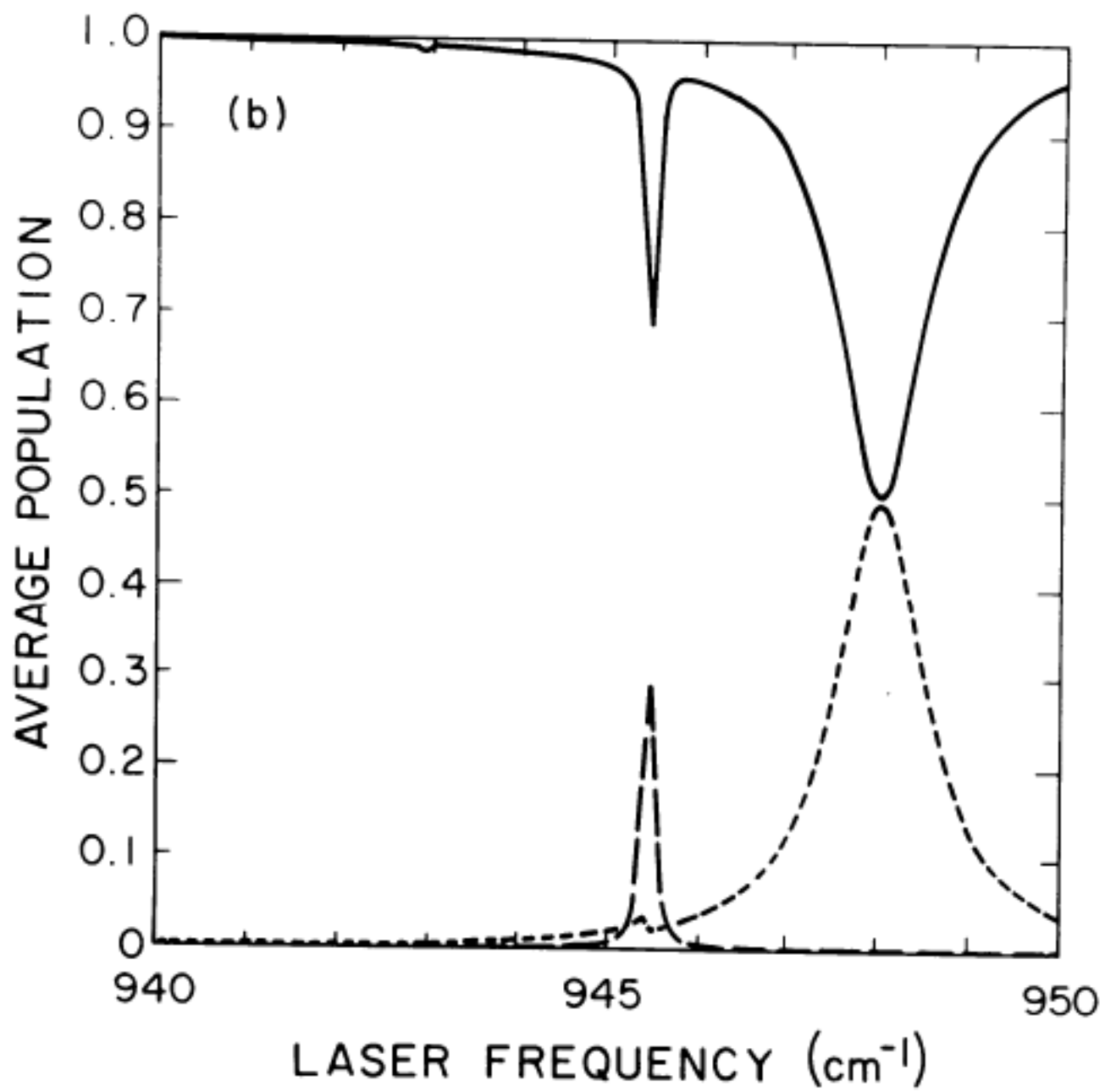
$$\frac{E_{mA}}{\hbar} = m\omega_0 + m(m-1)X$$

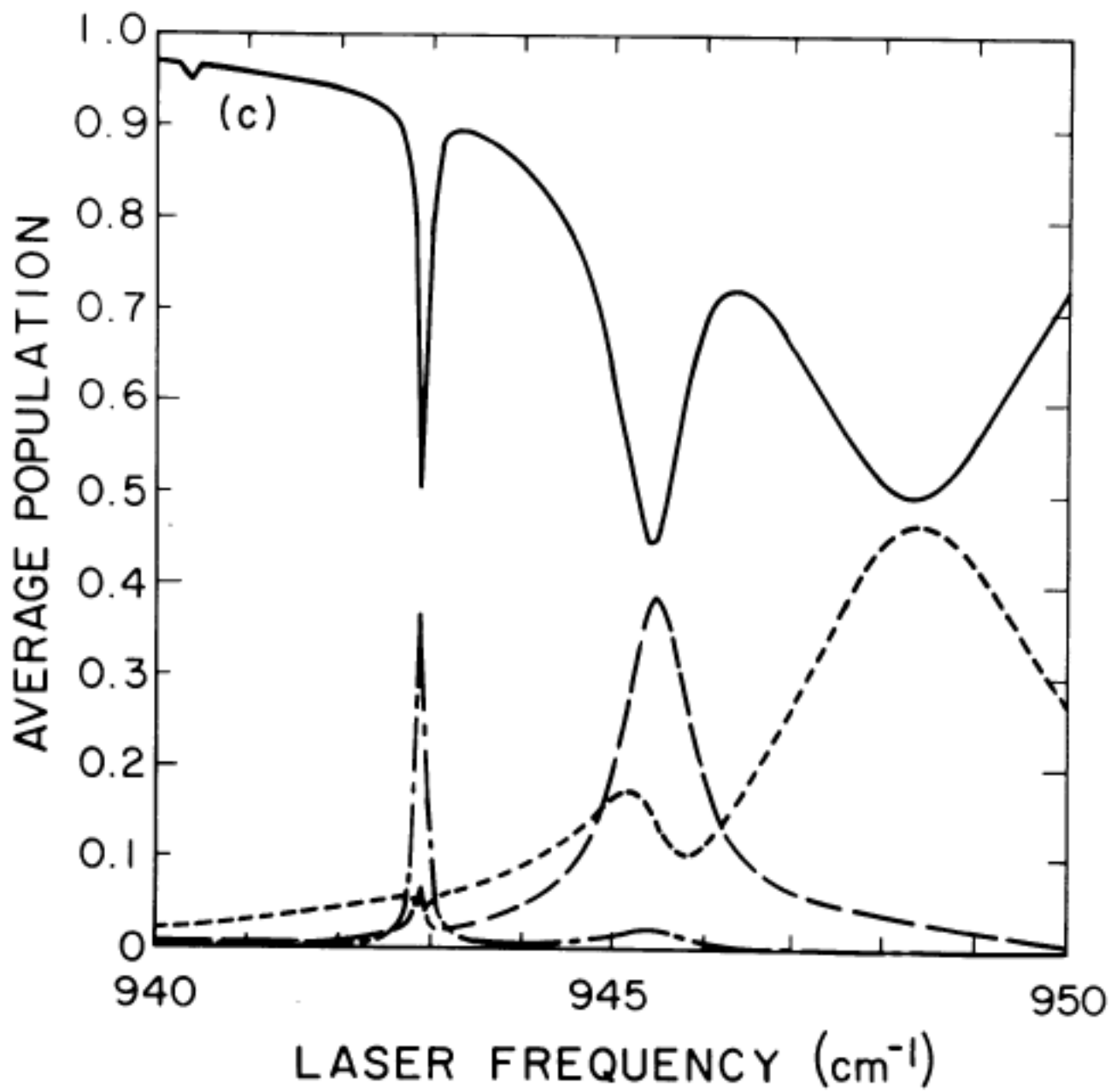
- The frequency of an  $N$ -photon resonance from  $m = 0$  to  $m = N$  is

$$\omega^{(N)} = \omega_0 + (N-1)X$$

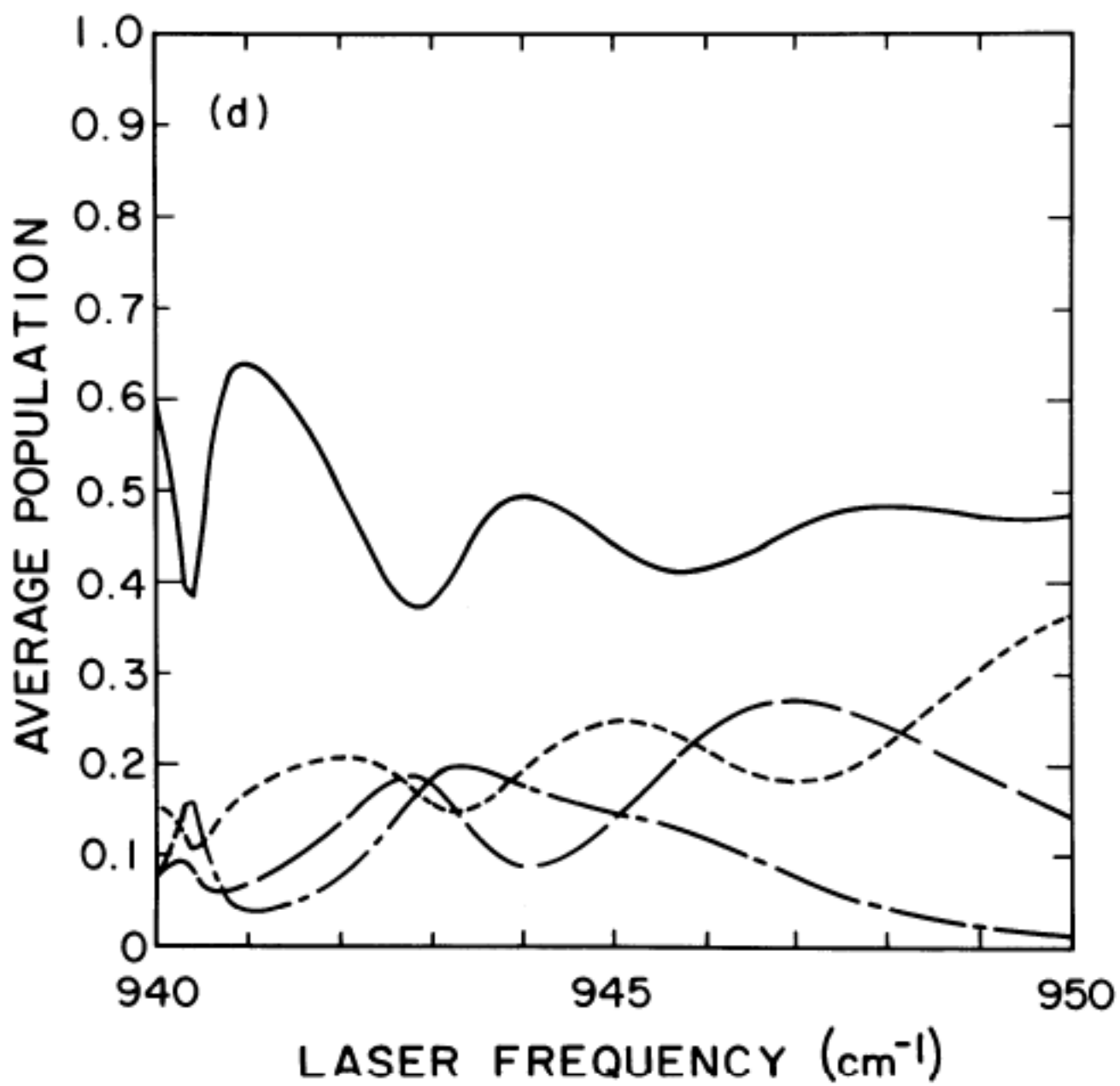


NSA

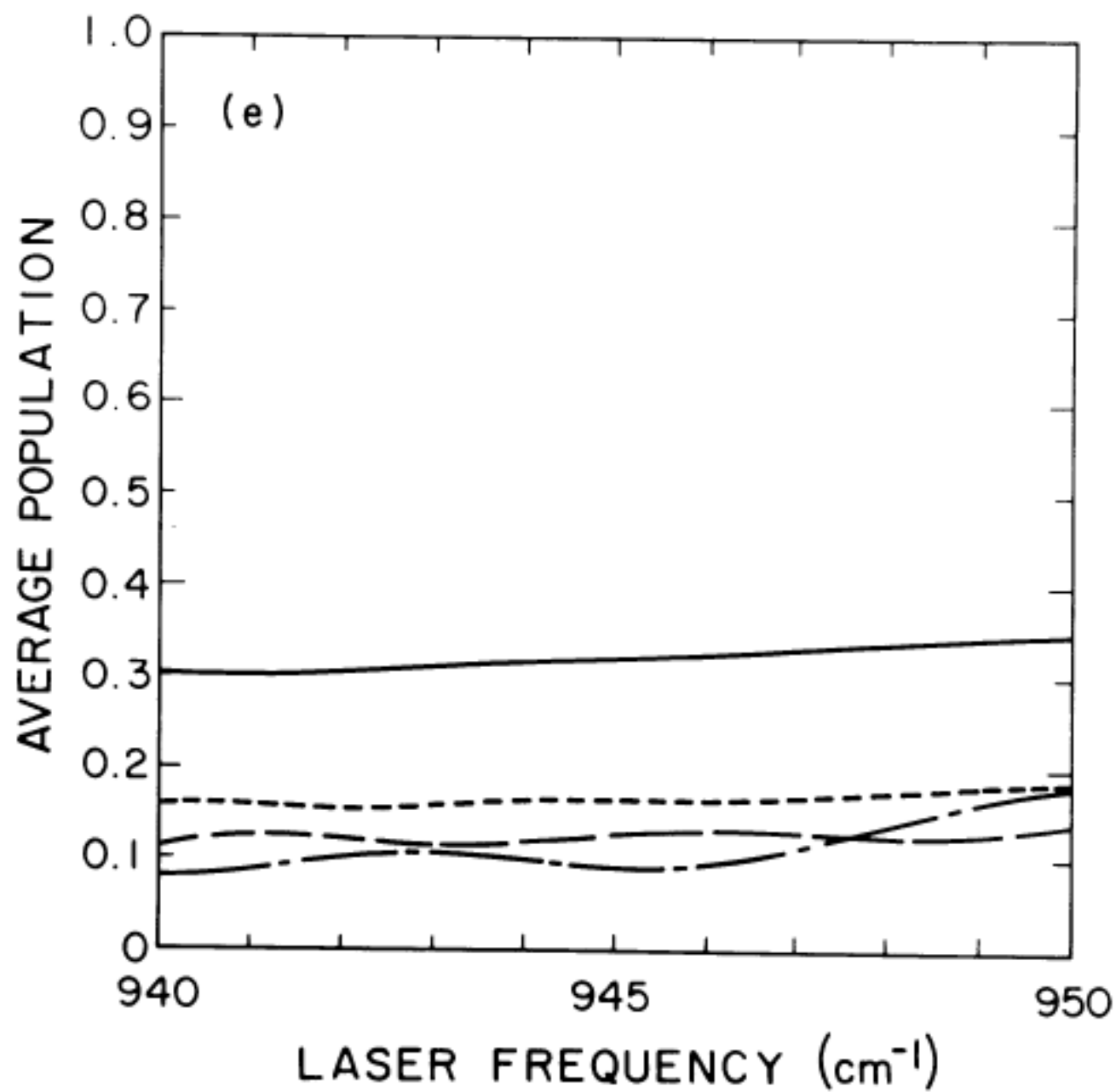




LSA



LSA



LSA

## THE ADIABATIC APPROXIMATION

- The system remains in the dressed state  $|\lambda; \mathcal{E}(t')\rangle$  that evolves continuously from the initial state  $|\lambda; \mathcal{E}(t'_0)\rangle$
- The time dependence of the probability amplitudes is

$$\tilde{c}_{mA}(t') = \langle mA | \lambda; \mathcal{E}(t') \rangle \exp \left[ i \int_{t'_0}^{t'} \epsilon_\lambda(t'') dt'' \right]$$

- The polarization is

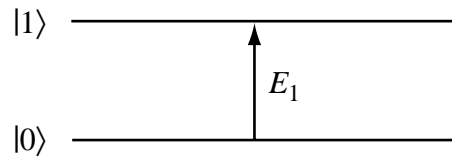
$$\mathcal{P}(\mathbf{r}_T, z', t') = 2iN \sum_{m,A,B} \langle \lambda; \mathcal{E}(t') | m-1, B \rangle \mu_{m-1,B; mA} \langle mA | \lambda; \mathcal{E}(t') \rangle,$$

and is imaginary. Hence there is a real susceptibility:

$$\chi(\mathbf{r}_T, z', t') = \frac{\mathcal{P}(\mathbf{r}_T, z', t')}{i\mathcal{E}(\mathbf{r}_T, z, t')}$$

- New frequencies are generated only through the dependence of  $\chi$  on  $t'$

## ADIABATIC APPROXIMATION FOR A 2-LEVEL SYSTEM (1)



- Schrödinger equation for the probability amplitudes in the **RWA**:

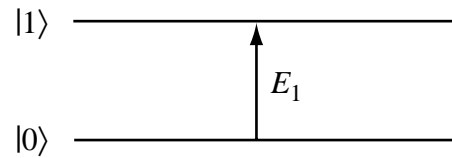
$$i\hbar \frac{d\tilde{c}_0}{dt'} = i\Omega^* \tilde{c}_1$$

$$i\hbar \frac{d\tilde{c}_1}{dt'} = i\Delta \tilde{c}_1 + i\Omega \tilde{c}_0$$

where  $\Delta = \omega - \frac{E_1}{\hbar}$  is the detuning,  $\Omega = \frac{\mu \mathcal{E}(t')}{2\hbar}$ ,  $\mathcal{E}$  is the field envelope,  $\mu$  is the dipole transition moment,  $t' = t - nz/c$ , and  $\omega$  is the field frequency

- In the adiabatic approximation, the system stays in the dressed state that evolves from the initial state at time  $t' = -\infty$ 
  - ▷ If the initial state is  $|0\rangle$ , then the dressed state that evolves from  $|0\rangle$  is  $|\lambda_0\rangle$

## ADIABATIC APPROXIMATION FOR A 2-LEVEL SYSTEM (2)



- Polarization envelope:

$$\mathcal{P} = 2iN\mu\tilde{c}_0^*\tilde{c}_1 = -2iN\mu \operatorname{sign}(\Delta) \frac{\Omega}{[\Delta^2 + 4|\Omega|^2]^{1/2}}$$

## VALIDITY OF THE ADIABATIC APPROXIMATION (1)

- **Adiabatic approximation:**

$$|\psi(t'_0)\rangle = |\lambda; E(t'_0)\rangle \Rightarrow |\psi(t')\rangle = |\lambda; E(t')\rangle$$

- ▷ Breakdown of the adiabatic approximation occurs if  $|\psi(t')\rangle = |\lambda; E(t')\rangle$  and

$$\langle \lambda'; E(t') | \psi(t') \rangle \neq 0$$

- ▷ First-order perturbation calculation (A. Messiah, *Quantum Mechanics*, Vol. 2, pp. 753-755):

$$\langle \lambda'; E(t') | \psi(t') \rangle \approx \left| \frac{\alpha_{\lambda'\lambda}}{\omega_{\lambda'\lambda}} \right|$$

where

$$\omega_{\lambda'\lambda} = \epsilon_{\lambda'} - \epsilon_{\lambda} \quad \text{and} \quad \alpha_{\lambda'\lambda} = \frac{\langle \lambda' | \dot{H}^{\text{eff}} | \lambda \rangle}{\omega_{\lambda'\lambda}}$$

- Condition for negligible “leakage” from  $|\lambda; E(t')\rangle$  to other dressed states:

$$\sum_{\lambda'} \left| \frac{\alpha_{\lambda'\lambda}}{\omega_{\lambda'\lambda}} \right|^2 \ll 1$$

## VALIDITY OF THE ADIABATIC APPROXIMATION (2)

- Matrix element of  $\dot{H}^{\text{eff}}$  between dressed states:

$$\begin{aligned} \alpha_{\lambda'\lambda}\omega_{\lambda'\lambda} &= \langle \lambda' | \dot{H}^{\text{eff}} | \lambda \rangle = \langle \lambda' | \frac{d}{dt'} \left( \Delta + \frac{E}{2\hbar}\mu \right) | \lambda \rangle \\ &= \langle \lambda' | \frac{\dot{E}}{2\hbar}\mu | \lambda \rangle = \dot{E} \langle \lambda' | \frac{H^{\text{eff}} - \Delta}{E} | \lambda \rangle \\ &= -\frac{\dot{E}}{E} \langle \lambda' | \Delta | \lambda \rangle \end{aligned}$$

- From the **adiabaticity criterion**:

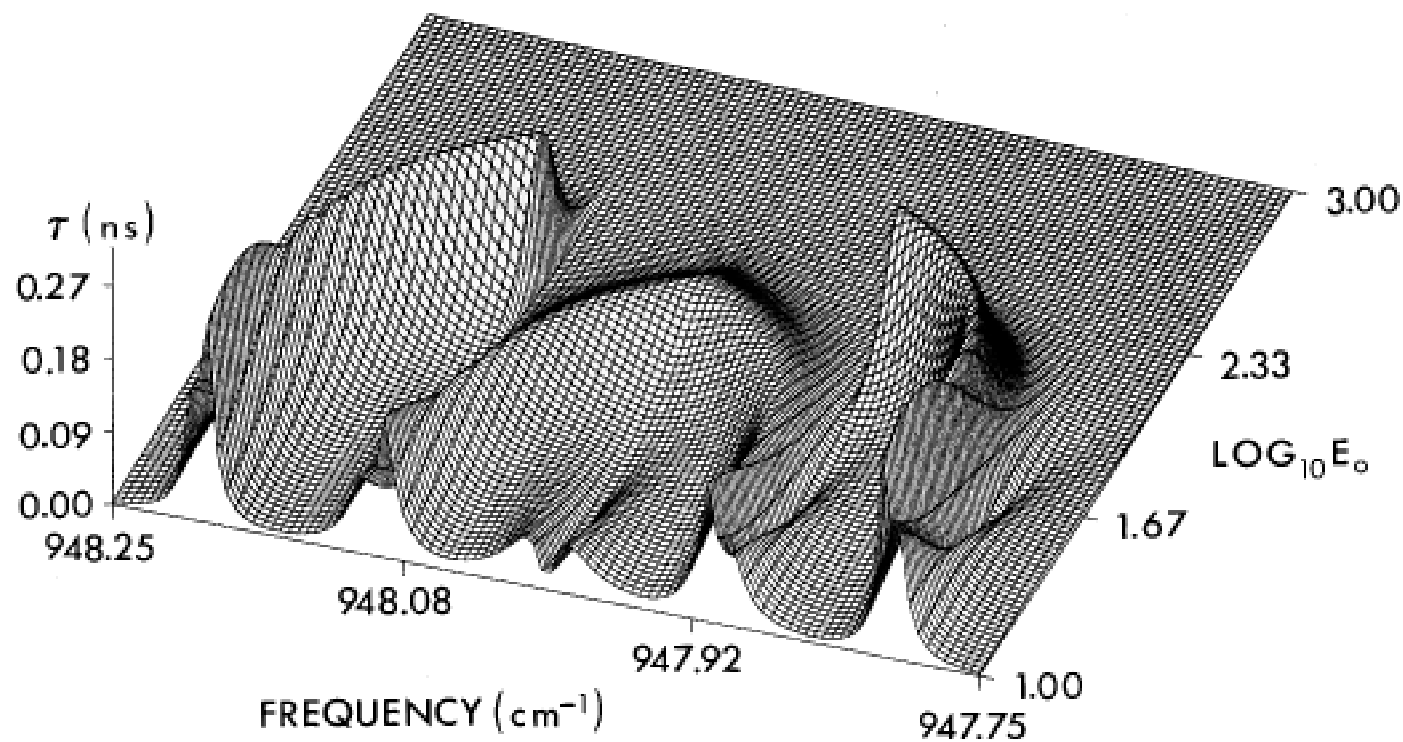
▷ The adiabatic approximation is valid when the characteristic time of the pulse,  $\tau_p$ , is long compared to an intrinsic dynamical time  $\tau$ :

$$\tau_p \approx \left| \frac{E}{\dot{E}} \right| \gg \tau$$

where

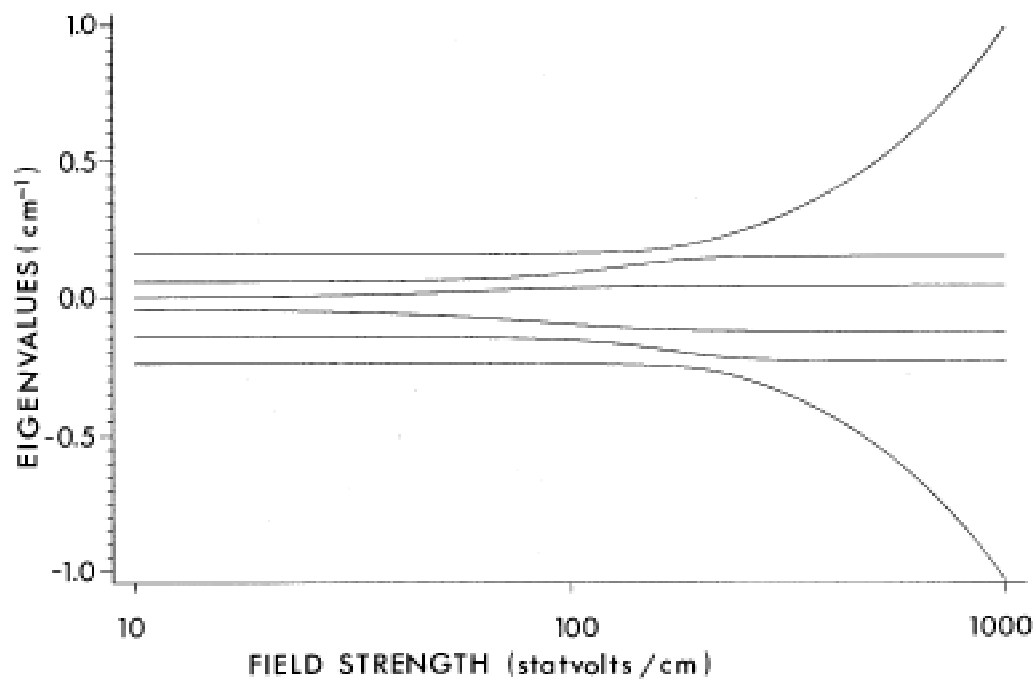
$$\tau = \left[ \sum_{\lambda'} \left| \frac{\langle \lambda' | \Delta | \lambda \rangle}{\omega_{\lambda'\lambda}^2} \right|^2 \right]^{1/2}$$

# INTERNAL DYNAMICAL TIME FOR A (1,5) SYSTEM



## EIGENVALUES OF A (1,5) SYSTEM

EIGENVALUES OF A (1,5) SYSTEM  
 $\nu = 947.96 \text{ cm}^{-1}$



## THE DENSITY MATRIX

- The density matrix must be employed instead of a wave function to describe a quantum system in either of two cases:
  - ▷ When the initial conditions are not completely specified
  - ▷ When the system is not isolated (as in the case of collisional damping)

- The elements of the density matrix  $\rho$  (Greek “rho”) are

$$\rho_{mn} = \overline{c_m c_n^*}$$

where the bar denotes averaging over the initial conditions and  $c_m$  is the probability amplitude of the state  $|m\rangle$

- Physically:
  - ▷ The diagonal element  $\rho_{mm}$  is the probability of occurrence of state  $|m\rangle$
  - ▷ An off-diagonal element  $\rho_{mn}$  is of order unity only when there is a coherent superposition of  $|m\rangle$  and  $|n\rangle$
  - ▷ One often calls  $\rho_{mm}$  a “population”, and  $\rho_{mn}$  ( $m \neq n$ ) a “coherence”

## PROPERTIES OF THE DENSITY MATRIX

- The normalization of the state vector implies that the trace of  $\rho$  is 1:

$$\text{trace}[\rho] = 1$$

- $\rho$  is Hermitian:  $\rho^\dagger = \rho$
- In a **pure case** (e.g. initial conditions that uniquely specify the state as  $|\psi\rangle$ ), the density matrix is a projection operator and is idempotent:

$$\rho = |\psi\rangle\langle\psi| \quad \text{and} \quad \rho^2 = |\psi\rangle\langle\psi|\psi\rangle\langle\psi| = \rho$$

▷ Example: If, for a 2-level system,  $|\psi\rangle = \frac{1}{\sqrt{2}}(|0\rangle - |1\rangle)$ , then

$$\rho = \left[ \frac{1}{\sqrt{2}} \begin{pmatrix} 1 \\ -1 \end{pmatrix} \right] \left[ \frac{1}{\sqrt{2}} (1, -1) \right] = \frac{1}{2} \begin{pmatrix} 1 & -1 \\ -1 & 1 \end{pmatrix} \quad (\text{verify that } \rho^2 = \rho!)$$

- In a **mixture**, the trace of the density matrix obeys the relations

$$\text{trace}[\rho^2] < \text{trace}[\rho] = 1$$

▷ Example:

$$\text{If } \rho = \begin{pmatrix} \frac{1}{2} & \frac{1}{4} \\ \frac{1}{4} & \frac{1}{2} \end{pmatrix}, \text{ then } \text{trace}[\rho^2] = \frac{5}{8} < \text{trace}[\rho] = 1$$

**TIME EVOLUTION OF THE DENSITY MATRIX**

- For an isolated system, the equation

$$i\hbar \frac{\partial \rho}{\partial t} = [\mathbf{H}, \rho]$$

is equivalent to the Schrödinger equation

- For an open system, the density matrix elements obey the master equation:

$$i\hbar \frac{\partial \rho_{mn}}{\partial t} = [\mathbf{H}, \rho]_{mn} - i\hbar \gamma_{mn}$$

for off-diagonal elements of  $\rho$  (coherences), and

$$i\hbar \frac{\partial \rho_{mm}}{\partial t} = [\mathbf{H}, \rho]_{mm} - i\hbar \rho_{mm} \sum_n w_{mn} + i\hbar \sum_{n \neq m} \rho_{nn} w_{nm}$$

for diagonal elements (populations)

- ▷ In the master equation,  $\gamma_{mn}$  is the coherence damping rate arising from phase-changing collisions, and  $w_{mn}$  is the rate of transfer of probability (population) from level  $m$  to level  $n$

## ROTATING-WAVE APPROXIMATION

- For a ladder system driven by a nearly resonant laser, it is convenient to introduce the slowly varying density-matrix elements

$$\tilde{\rho}_{mn} = e^{i(m-n)\omega t} \rho_{mn}$$

- ▷ In the rotating-wave approximation the equation of motion for the coherences is

$$\frac{\partial \tilde{\rho}_{mn}}{\partial t} = i \sum_l [H_{ml}^{\text{eff}} \tilde{\rho}_{ln} - H_{ln}^{\text{eff}} \tilde{\rho}_{ml}] - \gamma_{mn} \tilde{\rho}_{mn}$$

- ▷ The equation for the populations is

$$\frac{\partial \tilde{\rho}_{mm}}{\partial t} = -\frac{i}{\hbar} [\mathbf{H}, \boldsymbol{\rho}]_{mm} - \tilde{\rho}_{mm} \sum_n w_{mn} + \sum_{n \neq m} \tilde{\rho}_{nn} w_{nm}$$

## THE STEADY-STATE APPROXIMATION (1)

- If a system that is subject to collisions is driven by a laser pulse with a sufficiently slowly varying envelope, the coherent driving by the laser is in dynamic equilibrium with the collisional damping. For a system with  $N$  energy levels, the steady-state density matrix is determined by the equation

$$\frac{\partial \tilde{\rho}^{SS}}{\partial t} \approx 0$$

- Write the equation of motion for the  $N \times N$  matrix  $\tilde{\rho}$  as an equation for the  $N^2$ -dimensional vector  $\tilde{\rho}$ :

$$\frac{\partial \tilde{\rho}}{\partial t} = \mathbf{A} \tilde{\rho}$$

where the  $N^2 \times N^2$  matrix  $\mathbf{A}$  has the matrix elements

$$A_{mn;pq} = \delta_{nq} [-iH_{mp}^{\text{eff}} - \gamma_{mp}] + \delta_{mp} [iH_{nq}^{\text{eff}} - \gamma_{nq}] - \delta_{mn} \delta_{pq} \left[ -\delta_{np} \sum_{k \neq m} w_{mk} + w_{pm} \right]$$

## THE STEADY-STATE APPROXIMATION (2)

- The steady-state density matrix  $\tilde{\rho}^{SS}$  may be calculated from the system of linear equations

$$\mathbf{A}\tilde{\rho}^{SS} = 0$$

and

$$\text{trace}[\tilde{\rho}^{SS}] = 1$$

- References:
  1. D. R. Adams, C. D. Cantrell and W. H. Louisell, “Multilevel, Multifrequency Laser Pulse Propagation”, *Optics Communications* **43**, 292–296 (1982)
  2. G. L. Peterson and C. D. Cantrell, “Adiabatic Excitation of Multilevel Systems”, *Physical Review A* **31**, 807–822 (1985), Section V

## THE STEADY-STATE APPROXIMATION (3)

- In the zero-damping limit,

$$A_{mn;pq}^{(0)} = -i\delta_{nq}H_{mp}^{\text{eff}} + i\delta_{mp}H_{nq}^{\text{eff}}$$

- The limit of the steady-state density matrix as the damping goes to zero,

$$\tilde{\rho}_0^{SS} = \lim_{\gamma \rightarrow 0} \lim_{w \rightarrow 0} \tilde{\rho}^{SS},$$

obeys the equation of motion

$$\frac{\partial \tilde{\rho}_0^{SS}}{\partial t} = \mathbf{A}^{(0)} \rho_0^{SS} = \mathbf{0} \quad \Rightarrow \quad [\mathbf{H}^{\text{eff}}, \tilde{\rho}_0^{SS}] = \mathbf{0}$$

- It follows that  $\tilde{\rho}_0^{SS}$  is a mixture of the dressed states  $|\lambda\rangle$ :

$$\tilde{\rho}_0^{SS} = \sum_{\lambda} \tilde{\rho}_{\lambda\lambda} |\lambda\rangle\langle\lambda|$$

- ▷ This shows how the adiabatic approximation is related to the steady-state approximation