

# **The Density Matrix**

**Reference**

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## Pure vs Mixed State Example

Suppose that we are measuring the value of the spin magnetic moment of electrons. We take the spin magnetic dipole moment operator to be

$$\vec{\mu}_e = g\mu_B \vec{\sigma}.$$

We compare the average value we measure in two different states, both of which are equal mixtures of  $x$  and  $y$  spin character for the electrons.

### Pure spin state

$$|s_p\rangle = \frac{1}{\sqrt{3}}(|s_x\rangle + |s_y\rangle).$$

Here,

$$|s_x\rangle = \frac{1}{\sqrt{2}}(|\uparrow\rangle + |\downarrow\rangle),$$

$$|s_y\rangle = \frac{1}{\sqrt{2}}(|\uparrow\rangle + i|\downarrow\rangle).$$

(a) Let us show that  $|s_p\rangle$  is normalized.

$$\langle s_p | s_p \rangle = \frac{1}{3}(\langle s_x | + \langle s_y |)(|s_x\rangle + |s_y\rangle),$$

$$\langle s_p | s_p \rangle = \frac{1}{6}(\langle \uparrow | 2 + \langle \downarrow | (1 - i))(2|\uparrow\rangle + (1 + i)|\downarrow\rangle),$$

$$\langle s_p | s_p \rangle = \frac{1}{6}(4\langle \uparrow | \uparrow \rangle + 2\langle \downarrow | \downarrow \rangle) = 1,$$

(b) Let us find the expected value of the spin magnetic dipole moment corresponding to the state  $|s_p\rangle$ .

In  $\sigma_z$  basis,

$$|s_p\rangle = \frac{1}{\sqrt{6}} \begin{bmatrix} 2 \\ 1+i \end{bmatrix}.$$

$$\vec{\sigma} = \begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix} \vec{n}_x + \begin{bmatrix} 0 & -i \\ i & 0 \end{bmatrix} \vec{n}_y + \begin{bmatrix} 1 & 0 \\ 0 & -1 \end{bmatrix} \vec{n}_z.$$

$$\vec{\mu}_e = \langle s_p | \vec{\mu}_e | s_p \rangle = \frac{1}{\sqrt{6}} [2 \quad (1-i)] g\mu_B \left( \begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix} \vec{n}_x + \begin{bmatrix} 0 & -i \\ i & 0 \end{bmatrix} \vec{n}_y + \begin{bmatrix} 1 & 0 \\ 0 & -1 \end{bmatrix} \vec{n}_z \right) \frac{1}{\sqrt{6}} \begin{bmatrix} 2 \\ 1+i \end{bmatrix},$$

$$\vec{\mu}_e = \frac{g\mu_B}{6} [2 \quad (1-i)] \left( \begin{bmatrix} 1+i \\ 2 \end{bmatrix} \vec{n}_x + \begin{bmatrix} 1-i \\ 2i \end{bmatrix} \vec{n}_y + \begin{bmatrix} 2 \\ -1-i \end{bmatrix} \vec{n}_z \right),$$

$$\vec{\mu}_e = \frac{g\mu_B}{6} \left( (2 + 2i + 2 - 2i) \vec{n}_x + (2 - 2i + 2i + 2) \vec{n}_y + (4 - 1 - 1) \vec{n}_z \right),$$

$$\vec{\mu}_e = \frac{g\mu_B}{3} (2\vec{n}_x + 2\vec{n}_y + 1\vec{n}_z).$$

(c) The magnitude of the average dipole moment is,

$$\bar{\mu}_e = \frac{g\mu_B}{3} \sqrt{4 + 4 + 1} = g\mu_B.$$

## Mixed spin state

$|s_M\rangle$ :  $|s_x\rangle$  with probability  $P$  or  $|s_y\rangle$  with probability  $(1 - P)$ .

- (a) Let us find the expected value of the spin magnetic dipole moment corresponding to this mix state.

In  $\sigma_z$  basis,

$$|s_x\rangle = \frac{1}{\sqrt{2}} \begin{bmatrix} 1 \\ 1 \end{bmatrix},$$

$$|s_y\rangle = \frac{1}{\sqrt{2}} \begin{bmatrix} 1 \\ i \end{bmatrix}.$$

Corresponding to  $|s_x\rangle$  the average dipole moment is,

$$\langle s_x | \hat{\mu}_e | s_x \rangle = \frac{1}{\sqrt{2}} [1 \quad 1] g \mu_B \left( \begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix} \vec{n}_x + \begin{bmatrix} 0 & -i \\ i & 0 \end{bmatrix} \vec{n}_y + \begin{bmatrix} 1 & 0 \\ 0 & -1 \end{bmatrix} \vec{n}_z \right) \frac{1}{\sqrt{2}} \begin{bmatrix} 1 \\ 1 \end{bmatrix},$$

$$\langle s_x | \hat{\mu}_e | s_x \rangle = \frac{1}{2} [1 \quad 1] g \mu_B \left( \begin{bmatrix} 1 \\ 1 \end{bmatrix} \vec{n}_x + \begin{bmatrix} -i \\ i \end{bmatrix} \vec{n}_y + \begin{bmatrix} 1 \\ -1 \end{bmatrix} \vec{n}_z \right),$$

$$\langle s_x | \hat{\mu}_e | s_x \rangle = g \mu_B \vec{n}_x.$$

Corresponding to  $|s_y\rangle$  the average dipole moment is,

$$\langle s_y | \hat{\mu}_e | s_y \rangle = \frac{1}{\sqrt{2}} [1 \quad -i] g \mu_B \left( \begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix} \vec{n}_x + \begin{bmatrix} 0 & -i \\ i & 0 \end{bmatrix} \vec{n}_y + \begin{bmatrix} 1 & 0 \\ 0 & -1 \end{bmatrix} \vec{n}_z \right) \frac{1}{\sqrt{2}} \begin{bmatrix} 1 \\ i \end{bmatrix},$$

$$\langle s_y | \hat{\mu}_e | s_y \rangle = \frac{1}{2} [1 \quad -i] g \mu_B \left( \begin{bmatrix} i \\ 1 \end{bmatrix} \vec{n}_x + \begin{bmatrix} 1 \\ i \end{bmatrix} \vec{n}_y + \begin{bmatrix} 1 \\ -i \end{bmatrix} \vec{n}_z \right),$$

$$\langle s_y | \hat{\mu}_e | s_y \rangle = g \mu_B \vec{n}_y.$$

Thus the average value of the spin magnetic dipole moment corresponding to the given mixed state is,

$$\vec{\mu}_e = P \langle s_x | \hat{\mu}_e | s_x \rangle + (1 - P) \langle s_y | \hat{\mu}_e | s_y \rangle.$$

$$\vec{\mu}_e = g \mu_B (P \vec{n}_x + (1 - P) \vec{n}_y).$$

For  $P = \frac{1}{2}$ ,

$$\vec{\mu}_e = \frac{g \mu_B}{2} (\vec{n}_x + \vec{n}_y).$$

## Pure vs Mixed spin state

In case of pure spin state, the magnitude of the average magnetic dipole moment remains conserved that is  $g \mu_B$ . However, in case of mixed spin state, the magnitude of the average magnetic dipole moment is less than  $g \mu_B$ .

Note that a pure state is represented by a vector with a tip somewhere on the surface of the bloch sphere. On the other hand, a mixed state is represented by a vector with a tip somewhere on or inside the bloch sphere.

## Density Operator/Matrix

Let us represent the mixed state in the previous example in terms of the following density operator/matrix:

$$\rho = P|s_x\rangle\langle s_x| + (1 - P)|s_y\rangle\langle s_y|.$$

In  $\sigma_z$  basis,

$$\begin{aligned}\rho &= P\frac{1}{2}\begin{bmatrix} 1 & 1 \\ 1 & 1 \end{bmatrix} + (1 - P)\frac{1}{2}\begin{bmatrix} 1 & -i \\ i & 1 \end{bmatrix}, \\ \rho &= P\frac{1}{2}\begin{bmatrix} 1 & 1 \\ 1 & 1 \end{bmatrix} + (1 - P)\frac{1}{2}\begin{bmatrix} 1 & -i \\ i & 1 \end{bmatrix}, \\ \rho &= \frac{1}{2}\begin{bmatrix} 1 & (1+i)P-i \\ (1-i)P+i & 1 \end{bmatrix},\end{aligned}$$

Note that the trace of the density matrix  $Tr[\rho]$  is 1.

Now consider the following in  $\sigma_z$  basis,

$$\begin{aligned}\hat{\rho}\vec{\mu}_e &= \frac{1}{2}\begin{bmatrix} 1 & (1+i)P-i \\ (1-i)P+i & 1 \end{bmatrix}g\mu_B\left(\begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix}\vec{n}_x + \begin{bmatrix} 0 & -i \\ i & 0 \end{bmatrix}\vec{n}_y + \begin{bmatrix} 1 & 0 \\ 0 & -1 \end{bmatrix}\vec{n}_z\right), \\ \hat{\rho}\vec{\mu}_e &= \frac{g\mu_B}{2}\left(\begin{bmatrix} (1+i)P-i & 1 \\ 1 & (1-i)P+i \end{bmatrix}\vec{n}_x + \begin{bmatrix} (1-i)P+i & -i \\ i & -(1-i)P+i \end{bmatrix}\vec{n}_y + \begin{bmatrix} 1 & -(1+i)P+i \\ (1-i)P+i & -1 \end{bmatrix}\vec{n}_z\right),\end{aligned}$$

Look at the trace of the above matrix.

$$\begin{aligned}Tr\left[\hat{\rho}\vec{\mu}_e\right] &= \frac{g\mu_B}{2}\left(2P\vec{n}_x + 2(1 - P)\vec{n}_y + 0\vec{n}_z\right). \\ Tr\left[\hat{\rho}\vec{\mu}_e\right] &= g\mu_B\left(P\vec{n}_x + (1 - P)\vec{n}_y\right) = \vec{\mu}_e.\end{aligned}$$

Let us develop the theory of density matrix in a more general way. Assume that we have an ensemble of systems. The probability that a system in this ensemble is in a certain pure state  $|\psi_j\rangle$  is  $P_j$  for  $j = 1, 2, \dots$ . We have.

$$\sum_j P_j = 1. \quad (1)$$

The states  $|\psi_j\rangle$ 's are not necessarily orthogonal but are normalized. Let us define the density operator (as hinted above),

$$\rho = \sum_j P_j |\psi_j\rangle\langle\psi_j|. \quad (2)$$

Let us express  $|\psi_j\rangle$ 's in some orthonormal basis  $|\phi_n\rangle$  as,

$$|\psi_j\rangle = \sum_n a_{jn} |\phi_n\rangle. \quad (3)$$

Since the states  $|\psi_j\rangle$  are normalized we have  $\langle\psi_j|\psi_j\rangle = 1$ . This implies the following about the coefficients  $a_{jn}$  in Eq. (3)

$$\sum_n |a_{jn}|^2 = 1. \quad (4)$$

Let us express the density operator as a matrix in this orthonormal basis.

$$\rho = \sum_j P_j \sum_n a_{jn} |\phi_n\rangle \sum_m a_{jm}^* \langle \phi_m|,$$

$$\rho = \sum_j P_j \sum_n \sum_m a_{jn} a_{jm}^* |\phi_n\rangle \langle \phi_m|. \quad (5)$$

Consider the matrix element  $\rho_{pq}$ .

$$\rho_{pq} = \sum_j P_j a_{jp} a_{jq}^*.$$

Note the following.

$$\rho_{pq}^* = \sum_j P_j a_{jp}^* a_{jq} = \sum_j P_j a_{jq} a_{jp}^* = \rho_{qp}$$

Thus we note that the density matrix and hence the corresponding operator as defined in Eq. (2) is Hermitian.

$$\rho_{pq}^* = \rho_{qp}. \quad (6)$$

Consider the trace of the density operator.

$$Tr[\rho] = \sum_k \rho_{kk} = \sum_k \sum_j P_j a_{jk} a_{jk}^* = \sum_k \sum_j P_j |a_{jk}|^2 = \sum_k P_j = 1.$$

If we measure state of the ensemble in this basis the probability of getting a state  $|\phi_n\rangle$  is,

$$\sum_j P_j |a_{jn}|^2 = \sum_j P_j a_{jn} a_{jn}^* = \rho_{nn}.$$

Thus,

$$\text{probability of finding the ensemble in state } |\phi_n\rangle = \rho_{nn}. \quad (7)$$

The Eq. (7) provides us with the utility of the density matrix. We can calculate the probability of finding the system in a certain state as simply the diagonal element of the density matrix corresponding to that state when expressed as a matrix in the orthonormal basis including that state.

Next consider a general operator  $\hat{O}$ . The average value of the operator corresponding to a state  $|\psi_j\rangle$  is  $\langle \psi_j | \hat{O} | \psi_j \rangle$ . Thus the average value of this operator for our ensemble is,

$$\bar{O} = \sum_j P_j \langle \psi_j | \hat{O} | \psi_j \rangle, \quad (8)$$

Let us try to find relationship of  $\bar{O}$  with the density matrix  $\rho$ . Consider the following.

$$Tr[\rho \hat{O}] = \sum_k \langle \phi_k | \rho \hat{O} | \phi_k \rangle.$$

Using Eq. (5),

$$Tr[\rho \hat{O}] = \sum_k \langle \phi_k | \sum_j P_j \sum_n \sum_m a_{jn} a_{jm}^* |\phi_n\rangle \langle \phi_m| \hat{O} | \phi_k \rangle,$$

$$\begin{aligned}
Tr[\rho \hat{O}] &= \sum_k \sum_j P_j \sum_m \left( \sum_n \langle \phi_k | \phi_n \rangle a_{jn} \right) a_{jm}^* \langle \phi_m | \hat{O} | \phi_k \rangle, \\
Tr[\rho \hat{O}] &= \sum_k \sum_j P_j \sum_m a_{jk}^* a_{jm} \langle \phi_m | \hat{O} | \phi_k \rangle, \\
Tr[\rho \hat{O}] &= \sum_j P_j \left( \sum_m a_{jm}^* \langle \phi_m | \right) \hat{O} \left( \sum_k a_{jk} | \phi_k \rangle \right), \\
Tr[\rho \hat{O}] &= \sum_j P_j \langle \psi_j | \hat{O} | \psi_j \rangle = \bar{O}, \\
\bar{O} &= Tr[\rho \hat{O}]. \quad (9)
\end{aligned}$$

## Time Evolution of Density Operator

Again assume that we have an ensemble of systems. The probability that a system in this ensemble is in a certain pure state  $|\psi_j\rangle$  is  $P_j$  for  $j = 1, 2, \dots$

Each possible state (pure state) in this ensemble evolves as per the time-dependent Schrodinger's equation.

$$i\hbar \frac{\partial}{\partial t} |\psi_j\rangle = \hat{H} |\psi_j\rangle. \quad (10)$$

The conjugate of the above equation is,

$$-i\hbar \frac{\partial}{\partial t} \langle \psi_j | = \langle \psi_j | \hat{H}. \quad (11)$$

Consider the evolution of the density operator.

$$\begin{aligned}
i\hbar \frac{\partial}{\partial t} \rho &= \sum_j P_j \left( i\hbar \frac{\partial}{\partial t} |\psi_j\rangle \right) \langle \psi_j | + \sum_j P_j |\psi_j\rangle i\hbar \frac{\partial}{\partial t} \langle \psi_j |, \\
i\hbar \frac{\partial}{\partial t} \rho &= \sum_j P_j \hat{H} |\psi_j\rangle \langle \psi_j | - \sum_j P_j |\psi_j\rangle \langle \psi_j | \hat{H}, \\
i\hbar \frac{\partial}{\partial t} \rho &= \hat{H} \left( \sum_j P_j |\psi_j\rangle \langle \psi_j | \right) - \left( \sum_j P_j |\psi_j\rangle \langle \psi_j | \right) \hat{H}, \\
i\hbar \frac{\partial}{\partial t} \rho &= \hat{H} \rho - \rho \hat{H}. \\
i\hbar \frac{\partial}{\partial t} \rho &= [\hat{H}, \rho]. \quad (12)
\end{aligned}$$

## Interaction of Light with a Two Level Atomic System

### Hamiltonian

Consider a two level atomic system with energy levels  $E_1$  and  $E_2$  and respective orthonormal states  $|\psi_1\rangle$  and  $|\psi_2\rangle$  corresponding to the static hamiltonian  $\hat{H}_0$ . Assuming the dipole

approximation, electric field being uniform over the interaction region, the hamiltonian in this case is given as follows.

$$\hat{H} = \hat{H}_0 + \hat{H}_p = \hat{H}_0 - E\hat{\mu} = \hat{H}_0 - E_0 \cos \omega t \hat{\mu}. \quad (13)$$

Here  $E = E_0 \cos \omega t$  is the electric field and  $\hat{\mu}$  is the dipole operator given as follows assuming the electric field is polarized in z direction.

$$\hat{\mu} = -ez \quad (14)$$

In the energy eigen basis, the matrix element of an operator  $\hat{O}$  are given as  $O_{mn} = \langle \psi_m | \hat{O} | \psi_n \rangle$ .

Thus,

$$H_0 = \begin{bmatrix} H_{11} & H_{12} \\ H_{21} & H_{22} \end{bmatrix} = \begin{bmatrix} E_1 & 0 \\ 0 & E_2 \end{bmatrix}, \quad (15)$$

$$\mu = \begin{bmatrix} \mu_{11} & \mu_{12} \\ \mu_{21} & \mu_{22} \end{bmatrix}.$$

We assume that the energy eigenstates have even or odd parity with respect to the polarization direction of the electric field. Therefore,

$$\mu_{jj} = \langle \psi_j | \hat{\mu} | \psi_j \rangle = -e \langle \psi_j | z | \psi_j \rangle = 0.$$

Also,

$$\mu_{21} = \mu_{12} = \mu_d = -e \langle \psi_1 | z | \psi_2 \rangle = 0.$$

Therefore,

$$\mu = \begin{bmatrix} 0 & \mu_d \\ \mu_d & 0 \end{bmatrix}. \quad (16)$$

We have chosen the relative phase of  $|\psi_1\rangle$  and  $|\psi_2\rangle$  to be zero for convenience. The total hamiltonian matrix is given as,

$$H = H_0 - E\mu = \begin{bmatrix} E_1 & -E\mu_d \\ -E\mu_d & E_2 \end{bmatrix}. \quad (17)$$

## Mixed state and its evolution

We assume the system is in a mixed state that is for example,

$\cos \theta_j |\psi_1\rangle + e^{i\phi_j} \sin \theta_j |\psi_2\rangle$  with probability  $P_j$  for  $j = 1, 2, 3, 4, \dots$

Correspondingly, the density matrix can be written as,

$$\rho = \sum_j P_j |\psi_j\rangle \langle \psi_j| = \sum_j P_j \begin{bmatrix} (\cos \theta_j)^2 & e^{i\phi_j} \cos \theta_j \sin \theta_j \\ e^{-i\phi_j} \cos \theta_j \sin \theta_j & (\sin \theta_j)^2 \end{bmatrix} = \begin{bmatrix} \rho_{11} & \rho_{12} \\ \rho_{12}^* & \rho_{22} \end{bmatrix}. \quad (18)$$

Using Eq. (12), and noticing that we can change the partial derivative to the full derivative.

$$i\hbar \frac{d}{dt} \rho = [\hat{H}, \rho],$$

$$i\hbar \frac{d}{dt} \begin{bmatrix} \rho_{11} & \rho_{12} \\ \rho_{12}^* & \rho_{22} \end{bmatrix} = \begin{bmatrix} E_1 & -E\mu_d \\ -E\mu_d & E_2 \end{bmatrix} \begin{bmatrix} \rho_{11} & \rho_{12} \\ \rho_{12}^* & \rho_{22} \end{bmatrix} - \begin{bmatrix} \rho_{11} & \rho_{12} \\ \rho_{12}^* & \rho_{22} \end{bmatrix} \begin{bmatrix} E_1 & -E\mu_d \\ -E\mu_d & E_2 \end{bmatrix},$$

$$i\hbar \frac{d}{dt} \begin{bmatrix} \rho_{11} & \rho_{12} \\ \rho_{12}^* & \rho_{22} \end{bmatrix} = \begin{bmatrix} \rho_{11}E_1 - \rho_{12}^*E\mu_d & \rho_{12}E_1 - \rho_{22}E\mu_d \\ \rho_{12}^*E_2 - \rho_{11}E\mu_d & \rho_{22}E_2 - \rho_{12}E\mu_d \end{bmatrix} - \begin{bmatrix} \rho_{11}E_1 - \rho_{12}E\mu_d & \rho_{12}E_2 - \rho_{11}E\mu_d \\ \rho_{12}^*E_1 - \rho_{22}E\mu_d & \rho_{22}E_2 - \rho_{12}^*E\mu_d \end{bmatrix},$$

$$i\hbar \frac{d}{dt} \begin{bmatrix} \rho_{11} & \rho_{12} \\ \rho_{12}^* & \rho_{22} \end{bmatrix} = \begin{bmatrix} (\rho_{12} - \rho_{12}^*)E\mu_d & -\rho_{12}(E_2 - E_1) + (\rho_{11} - \rho_{22})E\mu_d \\ \rho_{12}^*(E_2 - E_1) - (\rho_{11} - \rho_{22})E\mu_d & (\rho_{12}^* - \rho_{12})E\mu_d \end{bmatrix}.$$

The following two equations suffice here.

$$\frac{d}{dt}(\rho_{11} - \rho_{22}) = -\frac{2i}{\hbar}(\rho_{12} - \rho_{12}^*)E\mu_d, \quad (19a)$$

$$\frac{d}{dt}\rho_{12} = \frac{i}{\hbar}(\rho_{12}(E_2 - E_1) - (\rho_{11} - \rho_{22})E\mu_d). \quad (19b)$$

Let us define the following.

$$\rho_{\Delta} = \rho_{11} - \rho_{22}. \quad (20)$$

$$\omega_{21} = \frac{E_2 - E_1}{2\hbar}. \quad (21a)$$

$$\Omega = \frac{E_0\mu_d}{2\hbar}. \quad (21b)$$

We can now write Eq. (19) as

$$\frac{d}{dt}\rho_{\Delta} = -4i\Omega(\rho_{12} - \rho_{12}^*)\cos\omega t, \quad (22a)$$

$$\frac{d}{dt}\rho_{12} = i\omega_{21}\rho_{21} - 2i\Omega\rho_{\Delta}\cos\omega t. \quad (22b)$$

## Introduction of longitudinal and transverse relaxation

Consider Eq. (22) in the absence of the electric field - the dipole perturbation.

$$\frac{d}{dt}\rho_{\Delta} = 0, \quad (23a)$$

$$\frac{d}{dt}\rho_{12} = i\omega_{21}\rho_{12}. \quad (23b)$$

This implies that if the system is left to itself then  $\rho_{\Delta} = \rho_{11} - \rho_{22}$  is constant. We already have  $Tr(\rho) = \rho_{11} + \rho_{22} = 1$ . Thus, the probability of being in either state 1 or state 2 doesn't change.

Moreover, the diagonal terms just oscillate in phase at frequency given by the difference of energy between the two levels.

However, it is a common observation that, since no system is perfectly isolated, if a system is left to itself then the probabilities exponentially decay to some steady state values. Let us model this into Eq. (22). We are about to reap the real advantage of the density matrix formulation.

$$\frac{d}{dt}\rho_{\Delta} = -4i\Omega(\rho_{12} - \rho_{12}^*)\cos\omega t - \frac{\rho_{\Delta} - \rho_{\Delta 0}}{T_1}, \quad (24a)$$

$$\frac{d}{dt}\rho_{12} = i\omega_{21}\rho_{21} - 2i\Omega\rho_{\Delta}\cos\omega t - \frac{\rho_{12}}{T_2}. \quad (24b)$$

Here, we have assumed that the decay rate of  $\rho_{\Delta}$ , in the absence of external perturbation, is proportional to its difference from a thermodynamically determined steady state value of  $\rho_{\Delta 0}$ .

Here  $T_1$  is called longitudinal relaxation time and  $T_2 < T_1$  is called the transverse relaxation time.



## Rotating wave approximation

Let,

$$\rho_{12} = \beta_{12} e^{i\omega t} = \beta_{12R} e^{i\omega t} + i\beta_{12I} e^{i\omega t}.$$

Here  $\beta_{12R}$  and  $\beta_{12I}$  are real and imaginary parts of  $\beta_{12}$ . Then,

$$\frac{d}{dt}\rho_{\Delta} = -2i\Omega(\beta_{12} e^{i\omega t} - \beta_{12}^* e^{-i\omega t})(e^{i\omega t} + e^{-i\omega t}) - \frac{\rho_{\Delta} - \rho_{\Delta 0}}{T_1}, \quad (23a)$$

$$e^{i\omega t} \frac{d}{dt}\beta_{12} + i\omega e^{i\omega t} \beta_{12} = i\omega_{21} \beta_{12} e^{i\omega t} - i\Omega\rho_{\Delta}(e^{i\omega t} + e^{-i\omega t}) - \frac{\beta_{12} e^{i\omega t}}{T_2}. \quad (23b)$$

Dropping the “high-speed” terms,  $e^{\pm 2i\omega t}$  as they average to zero over a comparatively larger time scale.

$$\frac{d}{dt}\rho_{\Delta} = -2i\Omega(\beta_{12} - \beta_{12}^*) - \frac{\rho_{\Delta} - \rho_{\Delta 0}}{T_1}, \quad (24a)$$

$$\frac{d}{dt}\beta_{12} = i(\omega_{21} - \omega)\beta_{12} - \Omega\rho_{\Delta} - \frac{\beta_{12}}{T_2}. \quad (24b)$$

From Eq. (24a)

$$\frac{d}{dt}\rho_{\Delta} = 4\Omega\beta_{12I} - \frac{\rho_{\Delta} - \rho_{\Delta 0}}{T_1} \quad (25)$$

From Eq. (24b)

$$\frac{d}{dt}\beta_{12R} = -(\omega_{21} - \omega)\beta_{12I} - \frac{\beta_{12R}}{T_2}. \quad (26)$$

$$\frac{d}{dt}\beta_{12I} = (\omega_{21} - \omega)\beta_{12R} - \Omega\rho_{\Delta} - \frac{\beta_{12I}}{T_2}. \quad (27)$$

## Steady State Solution

At steady state, Eqs. (25 - 27) become,

$$0 = 4\Omega\beta_{12I} - \frac{\rho_{\Delta} - \rho_{\Delta 0}}{T_1} \quad (25)$$

$$0 = -(\omega_{21} - \omega)\beta_{12I} - \frac{\beta_{12R}}{T_2}. \quad (26)$$

$$0 = (\omega_{21} - \omega)\beta_{12R} - \Omega\rho_{\Delta} - \frac{\beta_{12I}}{T_2}. \quad (27)$$

From (25),

$$\beta_{12I} = \frac{\rho_{\Delta} - \rho_{\Delta 0}}{4\Omega T_1} \quad (28)$$

From (26),

$$\beta_{12R} = -(\omega_{21} - \omega)T_2\beta_{12I} = -\frac{(\omega_{21} - \omega)T_2}{4\Omega T_1}(\rho_{\Delta} - \rho_{\Delta 0}) \quad (29)$$

Substituting Eqs. (28) and (29) in Eq. (27).

$$(\omega_{21} - \omega)T_2 \frac{(\omega_{21} - \omega)T_2}{4\Omega T_1}(\rho_{\Delta} - \rho_{\Delta 0}) + \Omega T_2 \rho_{\Delta} + \frac{1}{4\Omega T_1}(\rho_{\Delta} - \rho_{\Delta 0}) = 0,$$

$$\left( \frac{(\omega_{21}-\omega)^2 T_2^2}{4\Omega T_1} + \Omega T_2 + \frac{1}{4\Omega T_1} \right) \rho_{\Delta} = \rho_{\Delta 0} \left( \frac{1}{4\Omega T_1} + \frac{(\omega_{21}-\omega)^2 T_2^2}{4\Omega T_1} \right),$$

$$\left( (\omega_{21} - \omega)^2 T_2^2 + 4\Omega^2 T_1 T_2 + 1 \right) \rho_{\Delta} = \rho_{\Delta 0} \left( 1 + (\omega_{21} - \omega)^2 T_2^2 \right),$$

$$\rho_{\Delta} = \rho_{\Delta 0} \frac{1 + (\omega - \omega_{21})^2 T_2^2}{1 + (\omega - \omega_{21})^2 T_2^2 + 4\Omega^2 T_1 T_2}, \quad (29a)$$

$$\beta_{12I} = \rho_{\Delta 0} \frac{-\Omega T_2}{1 + (\omega - \omega_{21})^2 T_2^2 + 4\Omega^2 T_1 T_2}, \quad (29b)$$

$$\beta_{12R} = \rho_{\Delta 0} \frac{(\omega_{21} - \omega) \Omega T_2^2}{1 + (\omega - \omega_{21})^2 T_2^2 + 4\Omega^2 T_1 T_2}. \quad (29c)$$

## Real and Imaginary parts of polarization susceptibility

Let  $N$  be the number of “two-state” atoms per volume in the medium that is being exposed to the electric field  $E = E_0 \cos \omega t$ . Correspondingly, the static polarization  $P$  and the dimensionless real and imaginary susceptibilities,  $\chi'$  and  $\chi''$ , are defined as follows,

$$P = \epsilon_0 E_0 (\chi' \cos \omega t + \chi'' \sin \omega t). \quad (30)$$

The polarization per unit volume can also be given as follows,

$$P = N \bar{\mu} = N \text{Tr}(\rho \mu) = N \text{Tr} \left( \begin{bmatrix} \rho_{11} & \rho_{12} \\ \rho_{12}^* & \rho_{22} \end{bmatrix} \begin{bmatrix} 0 & \mu_d \\ \mu_d & 0 \end{bmatrix} \right) = N \mu_d (\rho_{12} + \rho_{12}^*),$$

$$P = N \mu_d (\beta_{12R} e^{i\omega t} + i \beta_{12I} e^{i\omega t} + \beta_{12R} e^{-i\omega t} - i \beta_{12I} e^{-i\omega t}),$$

$$P = 2N \mu_d (\beta_{12R} \cos \omega t - \beta_{12I} \sin \omega t). \quad (31)$$

Defining  $\Delta N = N \rho_{\Delta 0}$ , consider,

$$\frac{N \mu_d \rho_{\Delta 0} \Omega T_2}{\epsilon_0 E_0} = \frac{\Delta N \mu_d^2 T_2}{\epsilon_0 E_0}$$

Comparing Eq. (30) and (31),

$$\chi' = \frac{\Delta N \mu_d^2 T_2}{\epsilon_0 E_0} \frac{(\omega_{21} - \omega) T_2}{1 + (\omega - \omega_{21})^2 T_2^2 + 4\Omega^2 T_1 T_2}, \quad (32)$$

$$\chi'' = \frac{\Delta N \mu_d^2 T_2}{\epsilon_0 E_0} \frac{1}{1 + (\omega - \omega_{21})^2 T_2^2 + 4\Omega^2 T_1 T_2}. \quad (33)$$

Note that  $\Omega$  is proportional to the electric field strength. Hence, the term  $4\Omega^2 T_1 T_2$  is negligible for weak fields. Equation (32), exhibits the normal “linear” refraction variation while Eq. (33) depicts the Lorentzian absorption line.

## General Equation for Density Matrix Evolution

We can generalize Eq. (12) as follows. A certain element  $\rho_{mn}$  evolves as follows where  $\gamma_{mn}$  is the relaxation rate for that element.

$$i\hbar \frac{\partial}{\partial t} \rho_{mn} = [\hat{H}, \rho]_{mn} - i\hbar \gamma_{mn} (\rho_{mn} - \rho_{mn0}). \quad (34)$$