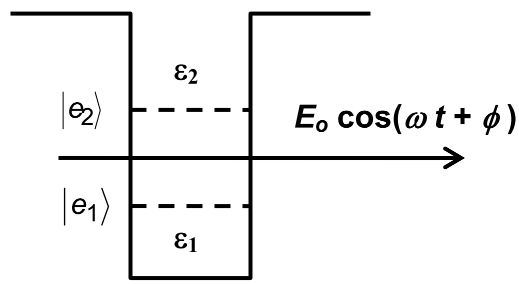
Chapter 2: Semi-Classical Light-Matter Interaction

2.1 A Two-level System Interacting with Classical Electromagnetic Field in the Absence of Decoherence

2.1.1 Hamiltonian for Interaction between Light and a Two-level System

Consider a two-level system, say an electron in a potential well or in an atom with two energy levels, interacting with electromagnetic radiation of frequency ω. The electric field at the location of the two-level system is,

$$\vec{E}(t) = \hat{n} E_o \cos(\omega t + \phi)$$



In the absence of the electric field the Hamiltonian of the electron is,

$$\hat{H}_o = \varepsilon_1 |e_1\rangle\langle e_2| \; + \; \varepsilon_2 |e_1\rangle\langle e_2|$$

The energy difference between the upper and the lower state is $\Delta\varepsilon = \varepsilon_2 - \varepsilon_1$. In the presence of the electric field, the potential energy of the electron of charge $q\ (= -e)$ is,

$$-q\,\hat{\vec{r}}.\vec{E}(t) = -q\hat{\vec{r}}.\hat{n} E_o \cos(\omega t + \phi)$$

So the Hamiltonian becomes,

$$\hat{H}(t) = \hat{H}_o - q\hat{\vec{r}}.\hat{n}\, E_o \cos(\omega t + \phi)$$

Note that the Hamiltonian is time-dependent. In the two dimensional Hilbert space consisting of only states $|e_1\rangle$ and $|e_2\rangle$, and assuming $\langle e_1|\hat{\vec{r}}|e_1\rangle = \langle e_2|\hat{\vec{r}}|e_2\rangle = 0$, the above Hamiltonian can be written as,

$$\hat{H}(t) = \varepsilon_1 |e_1\rangle\langle e_1| \; + \; \varepsilon_2 |e_2\rangle\langle e_2| \; - \; \hbar\Omega_R \left[\cos(\omega t + \phi)|e_1\rangle\langle e_2| \; + \cos(\omega t + \phi)|e_2\rangle\langle e_1|\right]$$

where the frequency Ω_R is related to the "dipole moment" of the states,

$$\hbar\Omega_R = qE_o \langle e_2|\vec{r}.\hat{n}|e_1\rangle = qE_o \;\langle e_1|\vec{r}.\hat{n}|e_2\rangle$$

In the so called *rotating wave approximation* only the important resonant term in each cosine term is retained (we will discuss this in detail later in the course) and one obtains,

$$\hat{H}(t) = \varepsilon_1 |e_1\rangle\langle e_1| \; + \; \varepsilon_2 |e_2\rangle\langle e_2| \; - \; \frac{\hbar\Omega_R}{2}\left[\exp(i\omega t + i\phi)|e_1\rangle\langle e_2| \; + \; \exp(-i\omega t - i\phi)|e_2\rangle\langle e_1|\right]$$

$$= \varepsilon_1 \hat{N}_1 \; + \; \varepsilon_2 \hat{N}_2 \; - \; \frac{\hbar\Omega_R}{2}\left[\exp(i\omega t + i\phi)\,\hat{\sigma}_- \; + \; \exp(-i\omega t - i\phi)\,\hat{\sigma}_+\right]$$

The Hamiltonian above is used to describe the interaction of a classical electromagnetic field with a two-level system. Any of the methods used for the time-independent two-level system problem can be used to solve the time-dependent problem as well.

2.1.2 Solution Using the Schrodinger Picture

We assume a time-dependent solution of the form,

$$|\psi(t)\rangle = c_1(t)\, e^{-i\frac{\varepsilon_1}{\hbar}t}\,|e_1\rangle + c_2(t)\, e^{-i\frac{\varepsilon_2}{\hbar}t}\,|e_2\rangle$$

and plug it into the Schrodinger equation,

$$i\,\hbar\,\frac{d\,|\psi(t)\rangle}{dt} = \hat{H}\,|\psi(t)\rangle$$

to get the following equations for the coefficients,

$$\frac{d\,c_1(t)}{dt} = i\,\frac{\Omega_R}{2}\,c_2(t)\,e^{-i\frac{\Delta}{\hbar}t+i\phi}$$

$$\frac{d\,c_2(t)}{dt} = i\,\frac{\Omega_R}{2}\,c_1(t)\,e^{i\frac{\Delta}{\hbar}t-i\phi}$$

The detuning Δ is defined as $\Delta = \varepsilon_2 - (\varepsilon_1 + \hbar\omega)$. The above equations can be solved using appropriate boundary conditions. It is convenient to define a frequency Ω as $\Omega = \sqrt{\Omega_R^2 + (\Delta/\hbar)^2}$. Suppose, $|\psi(t=0)\rangle = |e_1\rangle$, then the solution is,

$$c_1(t) = e^{-i\frac{\Delta}{2\hbar}t}\left[\cos\left(\frac{\Omega}{2}t\right) + i\,\frac{\Delta/\hbar}{\Omega}\sin\left(\frac{\Omega}{2}t\right)\right]$$

$$c_2(t) = e^{+i\frac{\Delta}{2\hbar}t-i\phi}\left[i\,\frac{\Omega_R}{\Omega}\sin\left(\frac{\Omega}{2}t\right)\right]$$

In the absence of detuning (i.e. $\Delta = 0$) , the probabilities of finding the electron in the upper and lower levels oscillate with a frequency equal to Ω_R. These oscillations are called Rabi oscillations, and the frequency Ω_R is called the Rabi frequency. The maximum value of the population difference, $|c_2(t)|^2 - |c_1(t)|^2$, is +1. In the presence of detuning, the populations oscillate with a frequency equal to Ω and the maximum value of the population difference, $|c_2(t)|^2 - |c_1(t)|^2$, is $\dfrac{\Omega_R^2 - (\Delta/\hbar)^2}{\Omega_R^2 + (\Delta/\hbar)^2} < 1$. There is no simple way to incoporate decoherence and/or population decay from the upper level into the lower level in the Schrodinger equation. To include these we have to use the density operator formalism.

2.1.3 Solution by Transformation to a Time-Independent Hamiltonian

Consider the time-dependent Hamiltonian describing the interaction of a two-level system with light,

$$\hat{H}(t) = \varepsilon_1|e_1\rangle\langle e_1| \;+\; \varepsilon_2|e_2\rangle\langle e_2| \;-\; \frac{\hbar\Omega_R}{2}\left[\exp(i\omega t + i\phi)\,|e_1\rangle\langle e_2| \;+\; \exp(-i\omega t - i\phi)\,|e_2\rangle\langle e_1|\right]$$

$$= \varepsilon_1\,\hat{N}_1 \;+\; \varepsilon_2\,\hat{N}_2 \;-\; \frac{\hbar\Omega_R}{2}\left[\exp(i\omega t + i\phi)\,\hat{\sigma}_- \;+\; \exp(-i\omega t - i\phi)\,\hat{\sigma}_+\right]$$

We define a unitary operator $\hat{B}(t)$ as follows,

$$\hat{B}(t) = \exp\left(-i\omega\,\hat{N}_1\,t\right)$$

To understand the effect of the unitary operator $\hat{B}(t)$ on the state $|\psi(t)\rangle$ suppose that $|\psi(t)\rangle$ is written as follows,

$$|\psi(t)\rangle = c_1(t)\, e^{-i\frac{\varepsilon_1}{\hbar}t}\,|e_1\rangle + c_2(t)\, e^{-i\frac{\varepsilon_2}{\hbar}t}\,|e_2\rangle$$

then,

$$\hat{B}(t)|\psi(t)\rangle = c_1(t)\, e^{-i\frac{\varepsilon_1+\hbar\omega}{\hbar}t}\,|e_1\rangle + c_2(t)\, e^{-i\frac{\varepsilon_2}{\hbar}t}\,|e_2\rangle$$

The operator $\hat{B}(t)$ "boosts" the energy of the lower level by $\hbar\omega$. It is easy to prove the following two identities,

$$\hat{B}(t) = \exp\left(-i\omega\,\hat{N}_1\, t\right) = 1 + \hat{N}_1\left[\exp(-i\omega\, t) - 1\right]$$

$$\hat{B}(t)\left[\hbar\omega\,\hat{N}_1 + \hat{H}(t)\right]\hat{B}^{+}(t) = \hat{H}_R$$

where,

$$\hat{H}_R = (\varepsilon_1 + \hbar\omega)\hat{N}_1 \;+\; \varepsilon_2\,\hat{N}_2 \;-\; \frac{\hbar\Omega_R}{2}\left[e^{i\phi}\,\hat{\sigma}_- \;+\; e^{-i\phi}\,\hat{\sigma}_+\right]$$

The Schrodinger equation for the state $|\psi(t)\rangle$ is,

$$i\hbar\frac{\partial|\psi(t)\rangle}{\partial t} = \hat{H}(t)\,|\psi(t)\rangle$$

Suppose we define a new $|\phi_R(t)\rangle$ state as follows,

$$|\phi_R(t)\rangle = \hat{B}(t)\,|\psi(t)\rangle$$

Then differentiating both sides with respect to time, and using the above identities, gives,

$$i\hbar\frac{\partial|\phi_R(t)\rangle}{\partial t} = \hat{H}_R\,|\phi_R(t)\rangle$$

The above equation shows that the original problem with a time-dependent Hamiltonian $\hat{H}(t)$ is equivalent to a problem with a time-independent Hamiltonian $\hat{H}_R$. One can solve the time-independent problem easily using any of the methods of the last Chapter. In particular, some insight is obtained by using the eigenstates of the Hamiltonian $\hat{H}_R$. Suppose, for simplicity, that the detuning $\Delta = \varepsilon_2 - (\varepsilon_1 + \hbar\omega)$ is zero. Then the two eigenstates, and the corresponding eigenenergies, of the Hamiltonian $\hat{H}_R$ are,

$$|v_1\rangle = \frac{1}{\sqrt{2}}\left(e^{i\phi/2}|e_1\rangle + e^{-i\phi/2}|e_2\rangle\right) \qquad \lambda_1 = \varepsilon_2 - \frac{\hbar\Omega_R}{2}$$

$$|v_2\rangle = \frac{1}{\sqrt{2}}\left(e^{i\phi/2}|e_1\rangle - e^{-i\phi/2}|e_2\rangle\right) \qquad \lambda_1 = \varepsilon_2 + \frac{\hbar\Omega_R}{2}$$

The slitting of the eigenenergies is $\hbar\Omega_R$. Suppose the initial state is,

$$|\psi(t=0)\rangle = |\phi_R(t=0)\rangle = |e_1\rangle = \frac{e^{-i\phi/2}}{\sqrt{2}}\left(|v_1\rangle + |v_2\rangle\right).$$

Then,

$$|\phi_R(t)\rangle = e^{-\frac{i}{\hbar}\hat{H}_R t}\,|\phi_R(t=0)\rangle = |\phi_R(t)\rangle = \frac{e^{-i\phi/2}}{\sqrt{2}}\left[e^{\frac{-i}{\hbar}(\varepsilon_2 - \hbar\Omega_R/2)t}\,|v_1\rangle \;+\; e^{\frac{-i}{\hbar}(\varepsilon_2 + \hbar\Omega_R/2)t}\,|v_2\rangle\right]$$

$$\Rightarrow |\phi_R(t)\rangle = e^{\frac{-i}{\hbar}\varepsilon_2 t}\cos\left(\frac{\Omega_R}{2}t\right)|e_1\rangle \;+\; i e^{\frac{-i}{\hbar}\varepsilon_2 t - i\phi}\sin\left(\frac{\Omega_R}{2}t\right)|e_2\rangle$$

Finally, the desired quantum state $|\psi(t)\rangle$ can be obtained as follows,

$$|\psi(t)\rangle = \hat{B}^{+}(t)|\phi_R(t)\rangle = e^{\frac{-i}{\hbar}\varepsilon_1 t}\cos\left(\frac{\Omega_R}{2}t\right)|e_1\rangle + ie^{\frac{-i}{\hbar}\varepsilon_2 t - i\phi}\sin\left(\frac{\Omega_R}{2}t\right)|e_2\rangle$$

The above result agrees with the one obtained in 2.1.2 when detuning is zero. The most interesting aspect of the above technique is that it shows that the time-dependent problem can be mapped onto a time-independent problem. We know that in any time-independent problem the eigenstates play a special role. Eigenstates are stationary states and whenever any arbitrary state is written as a superposition of stationary states, the probability for the state to be found in any one of the stationary states upon measurement remains time-independent. Suppose, $|\psi(t=0)\rangle = |\phi_R(t=0)\rangle = |v_1\rangle$. Then,

$$|\phi_R(t)\rangle = e^{\frac{-i}{\hbar}(\varepsilon_2 - \hbar\Omega_R/2)t}|v_1\rangle$$

$$\Rightarrow |\psi(t)\rangle = \hat{B}^{+}(t)|\phi_R(t)\rangle = \hat{B}^{+}(t)\, e^{\frac{-i}{\hbar}(\varepsilon_2 - \hbar\Omega_R/2)t}|v_1\rangle$$

$$= \frac{e^{i\frac{\Omega_R}{2}t}}{\sqrt{2}}\left[e^{\frac{-i}{\hbar}\varepsilon_1 t + i\frac{\phi}{2}}|e_1\rangle + e^{\frac{-i}{\hbar}\varepsilon_2 t - i\frac{\phi}{2}}|e_2\rangle\right]$$

And therefore,

$$\left|\langle e_1|\psi(t)\rangle\right|^2 = \left|\langle e_1|\psi(t=0)\rangle\right|^2 = \frac{1}{2}$$

$$\left|\langle e_2|\psi(t)\rangle\right|^2 = \left|\langle e_2|\psi(t=0)\rangle\right|^2 = \frac{1}{2}$$

The probabilities of finding the electron in the upper and lower levels remain time-independent when the initial state corresponds to an eigenstate of the time-independent Hamiltonian $\hat{H}_R$. This conclusion remains valid, of course, even when detuning is non-zero.

2.2 Optical Bloch Equations

2.2.1 Solution Using the Density Operator and Optical Bloch Equations

Here we will use the density operator approach in the Schrodinger picture. Starting from the density operator equation,

$$i\hbar\frac{d\,\hat{\rho}(t)}{dt} = \left[\hat{H}(t),\hat{\rho}(t)\right] = \hat{H}(t)\,\hat{\rho}(t) - \hat{\rho}(t)\,\hat{H}(t)$$

one can derive the differential equations for $\rho_{11}(t)$, $\rho_{22}(t)$, $\rho_{21}(t)$, and $\rho_{12}(t)$ by taking matrix elements of the density operator equation with respect to the states $|e_1\rangle$ and $|e_2\rangle$. The result is,

$$\frac{d\,\rho_{11}(t)}{dt} = i\frac{\Omega_R}{2}\left[\rho_{21}(t)\exp(i\omega t + i\phi) - \rho_{12}(t)\exp(-i\omega t - i\phi)\right]$$

$$\frac{d\,\rho_{22}(t)}{dt} = -i\frac{\Omega_R}{2}\left[\rho_{21}(t)\exp(i\omega t + i\phi) - \rho_{12}(t)\exp(-i\omega t - i\phi)\right]$$

$$\frac{d\,\rho_{12}(t)}{dt} = i\frac{(\varepsilon_2 - \varepsilon_1)}{\hbar}\rho_{12}(t) + i\frac{\Omega_R}{2}\exp(i\omega t + i\phi)\left[\rho_{22}(t) - \rho_{11}(t)\right]$$

$$\frac{d\,\rho_{21}(t)}{dt} = -i\frac{(\varepsilon_2 - \varepsilon_1)}{\hbar}\rho_{21}(t) - i\frac{\Omega_R}{2}\exp(-i\omega t - i\phi)\left[\rho_{22}(t) - \rho_{11}(t)\right]$$

The detuning Δ between the field frequency and the energy level separation is,

$$\Delta = \varepsilon_2 - \left(\varepsilon_1 + \hbar\omega\right)$$

The solution is most easily obtained, and insightful as well, if one uses a formalism originally developed to treat precession of nuclear spins. We define three new real quantities $V_x(t)$, $V_y(t)$, and $V_z(t)$ as follows,

$$V_x(t) = \left[\rho_{21}(t)\, e^{i\omega t + i\phi} + \rho_{12}(t)\, e^{-i\omega t - i\phi}\right]$$

$$V_y(t) = i\left[\rho_{21}(t)\, e^{i\omega t + i\phi} - \rho_{12}(t)\, e^{-i\omega t - i\phi}\right]$$

$$V_z(t) = \left[\rho_{22}(t) - \rho_{11}(t)\right]$$

The vector $\vec{V}(t)$ is,

$$\vec{V}(t) = V_x(t)\hat{x} + V_y(t)\hat{y} + V_z(t)\hat{z}$$

Note that the x- and y-components of the vector $\vec{V}(t)$ are related to the off-diagonal elements of the density operator, and the z-component of the vector $\vec{V}(t)$ is related to the occupation probability difference between the upper and the lower energy levels (also called the population difference). One can now rewrite the equations for the density matrix elements in terms of the quantities $V_x(t)$, $V_y(t)$, and $V_z(t)$,

$$\frac{dV_x(t)}{dt} = -\frac{\Delta}{\hbar} V_y(t)$$

$$\frac{dV_y(t)}{dt} = \frac{\Delta}{\hbar} V_x(t) + \Omega_R\, V_z(t)$$

$$\frac{dV_z(t)}{dt} = -\Omega_R\, V_y(t)$$

If one defines a vector frequency $\vec{\Omega}$ as $\vec{\Omega} = -\Omega_R\, \hat{x} + \frac{\Delta}{\hbar}\hat{z}$, then the equations above can be written in vector form very compactly as,

$$\frac{d\vec{V}(t)}{dt} = \vec{\Omega} \times \vec{V}(t)$$

Note that all fast time dependencies (on the scale of ω or $\left(\varepsilon_2 - \varepsilon_1\right)/\hbar$) present in the equations for the density matrix elements are absent in the equation for the vector $\vec{V}(t)$ and this is the main advantage of working with $\vec{V}(t)$. The equation for $\vec{V}(t)$ resembles the equation of the magnetic moment $\vec{M}(t)$ of a classical spin in a constant magnetic field $\vec{B}$,

$$\frac{d\vec{M}(t)}{dt} = -\gamma\, \vec{B} \times \vec{M}(t)$$

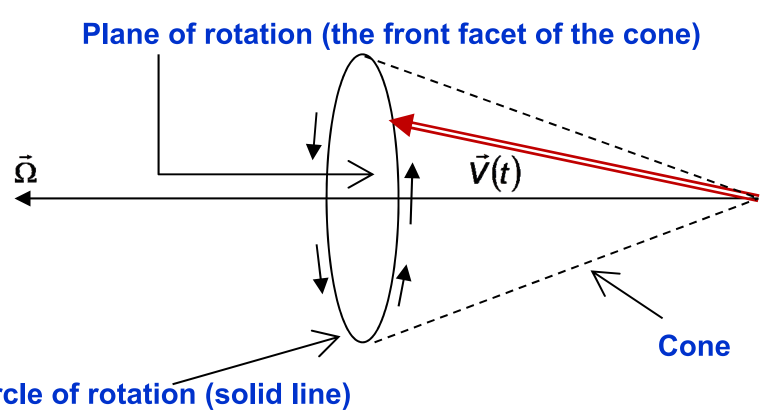
where γ is the gyromagnetic ratio (ratio between the spin magnetic moment and the spin angular momentum). The magnetic moment equation was derived by Felix Bloch in 1946. Therefore, the equation for the vector $\vec{V}(t)$ is sometimes called the **optical Bloch equation**.

The following facts are not hard to prove and follow directly from the vector equation for $\vec{V}(t)$,

i) The magnitude of the vector $\vec{V}(t)$ does not change with time.

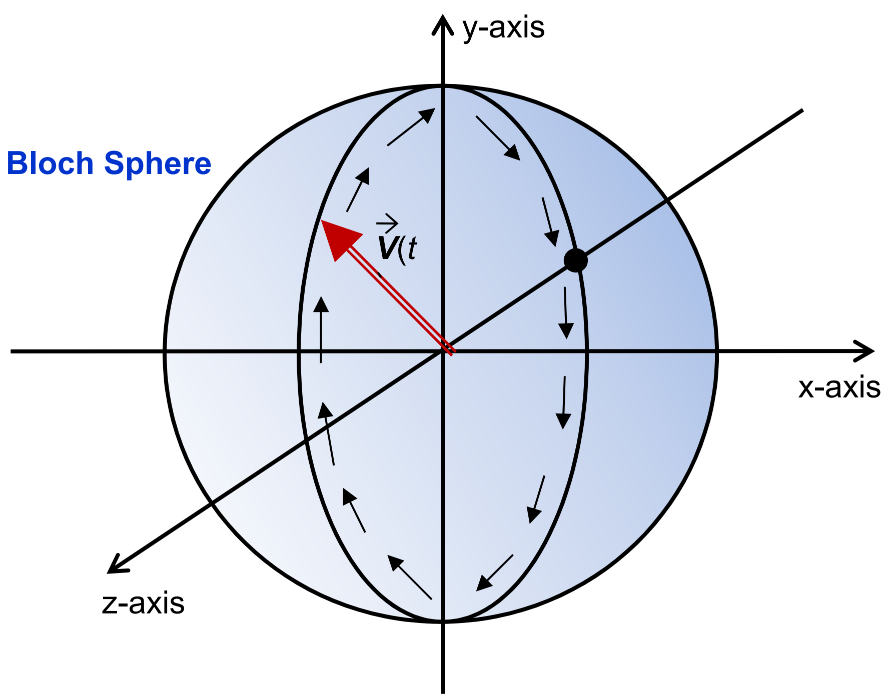
ii) The vector $\vec{V}(t)$ executes a periodic motion and the angular frequency is equal to the magnitude of the vector $\vec{\Omega}$.

iii) The "plane of rotation" is the plane in which the tip of the vector $\vec{V}(t)$ lies during rotation. The vector $\vec{\Omega}$ is always normal to the plane of rotation.



2.2.1 The Bloch Sphere

The vector $\vec{V}(t)$ completely characterizes the quantum state of an electron interacting with an electromagnetic wave.



The z-component of the vector equals $V_z(t)=[\rho_{22}(t)-\rho_{11}(t)]$ and gives the population difference between the two-levels and can have values between +1 and -1. The other components of the vector $\vec{V}(t)$ capture the coherence in the system and are related to the off-diagonal components of the density operator. Since the magnitude of the vector $\vec{V}(t)$ does not change with time, and if at time $t=0$ the quantum state of the system is $|e_1\rangle$ then $\vec{V}(t=0)=V_z(t=0)\hat{z}=-\hat{z}$, the magnitude of the vector $\vec{V}(t)$ is unity. Therefore, $\vec{V}(t)$ rotates with time but its tip always lies on a sphere of radius unity which is called the Bloch sphere, as shown in the Figure.

Case of Zero Detuning: If the initial quantum state of the electron is $|e_1\rangle$, then $\vec{V}(t=0) = V_z(t=0)\hat{z} = -\hat{z}$, and the vector tip at time $t=0$ is at the location indicated by the black dot on the sphere in the Figure. If electromagnetic field with zero detuning (i.e. $\Delta = 0$) is turned on at $t=0$, then since $\vec{\Omega} = -\Omega_R\,\hat{x}$, the tip of the vector $\vec{V}(t)$ rotates in the y-z plane (with x-axis as the axis of rotation) as shown in the Figure. The y-z plane is the plane of rotation in this case. The tip of the vector $\vec{V}(t)$ follows a circle formed where the plane of rotation intersects with the Bloch sphere. This circle is called the circle of rotation. At any time, the projection of the vector $\vec{V}(t)$ onto the z-axis (i.e. the z-component) gives the population difference $[\rho_{22}(t) - \rho_{11}(t)]$. The projection onto x-axis and y-axis gives the coherences (i.e. the off-diagonal components of the density matrix). At time when $|\vec{\Omega}|t = \Omega_R t = \pi/2$, the tip reaches the south pole, and the population difference goes to zero but the coherence is maximum. At this stage, the electron wavefunction (obtained by direct computation using the Schrodinger equation) is,

$$|\psi(t)\rangle = \frac{1}{\sqrt{2}}\left[e^{-i\frac{\varepsilon_1}{\hbar}t}|e_1\rangle + i\,e^{-i\frac{\varepsilon_2}{\hbar}t - i\phi}|e_2\rangle \right]_{t=\frac{\pi}{2\Omega_R}}$$

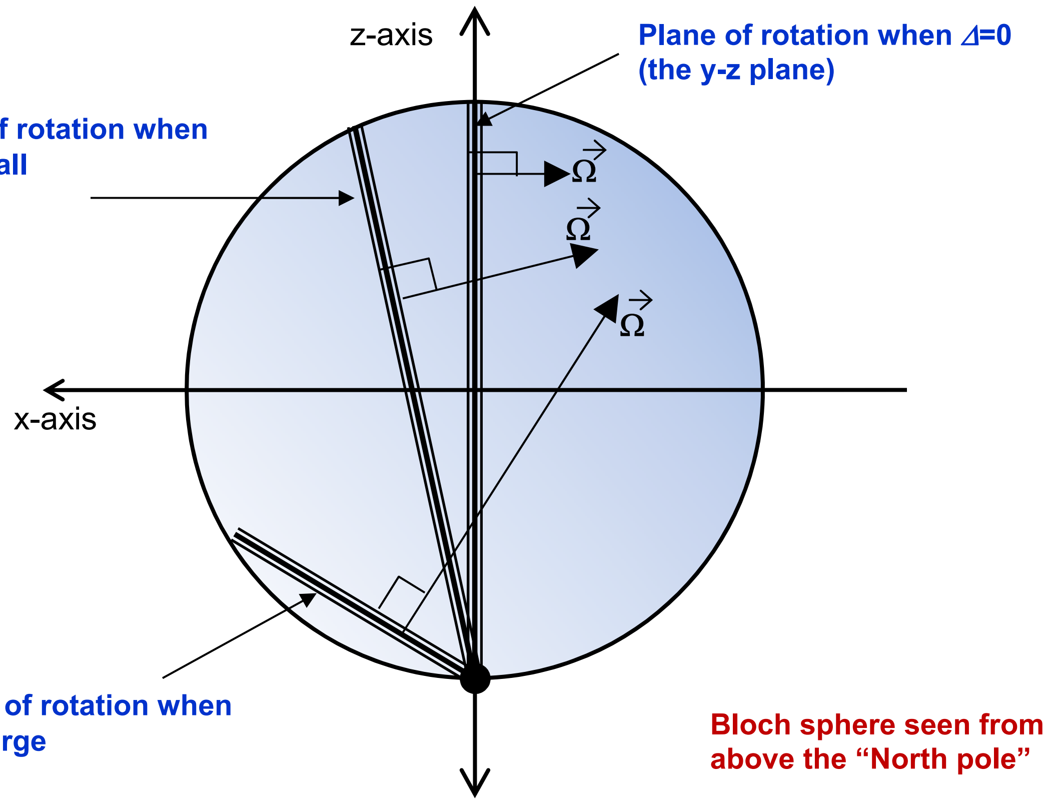
When $|\vec{\Omega}|t = \Omega_R t = \pi$, the tip reaches the positive z-axis, the electron is in the upper level, and the population difference is maximum (+1) but the coherence is minimum. The electron wavefunction at this stage (obtained by direct computation using the Schrodinger equation) is,

$$|\psi(t)\rangle = i\,e^{-i\frac{\varepsilon_2}{\hbar}t - i\phi}|e_2\rangle_{t=\frac{\pi}{\Omega_R}}$$

When $|\vec{\Omega}|t = \Omega_R t = 3\pi/2$, the population difference goes to zero again. And when $|\vec{\Omega}|t = \Omega_R t = 2\pi$, the population difference is at its minimum value of -1, the electron is in the lower state, and the tip of the vector $\vec{V}(t)$ is at the starting location shown by the black dot in the Figure. Thus, in one Rabi period (given by $2\pi/|\vec{\Omega}| = 2\pi/\Omega_R$) the population difference goes through one complete cycle. This phenomenon is called Rabi flopping or Rabi oscillation.

Case of Non-Zero Detuning: Using the Bloch sphere picture, one can **visually** solve many problems. Suppose there is some detuning so that the axis of rotation, given by the direction of $\vec{\Omega} = -\Omega_R\,\hat{x} + \frac{\Delta}{\hbar}\hat{z}$, is slightly tilted away from the negative x-axis, as shown below in the Figure. The vector $\vec{V}(t)$ starts from the location of the black dot, but now the plane of rotation is not the y-z plane but a plane that contains the black dot (since this is the initial starting point) and is also perpendicular to the direction given by $\vec{\Omega}$. The best way to see what this means is to look at the sphere from the top (i.e. from above the North pole) as shown in the Figure below. As the detuning is increased, the plane of rotation of the vector $\vec{V}(t)$ tilts. The tip of the vector $\vec{V}(t)$, of course, always remains on the Bloch sphere and in the plane of rotation. As the detuning increases, the frequency of rotation (given by $|\vec{\Omega}|$) increases and the tip of the vector $\vec{V}(t)$

rotates in circles of smaller and smaller radii (the circle of rotation formed by the intersection of the plane of rotation and the Bloch sphere). The maximum population difference, given by the z-component of the vector $\vec{V}(t)$, decreases from +1, and at large detunings it is no longer even positive at any time during the dynamics.



2.3 A Two-level System Interacting with Classical Electromagnetic Field in the Presence of Decoherence and Population Decay

2.3.1 Optical Bloch Equations with Decoherence and Population Decay

We know that electrons in materials or in atoms in higher lying energy levels usually come down to lower energy levels by giving off their energy to phonons or to any other non-radiative channel. In a two-level system, one can include relaxation of the population from the upper level to the lower level via such non-radiative mechanisms. We assume that the population relaxation time is T_1. One can also include the effects of decoherence by assuming that the off-diagonal components of the density matrix decay with a time constant T_2. In physical systems, the same scattering mechanism is often, but not always, the source of both population relaxation (i.e. T_1) and decoherence (i.e. T_2). When this is true, T_1 and T_2 are related; $T_2 \approx 2T_1$. Otherwise, $T_2 < 2T_1$. In the presence of population relaxation and decoherence, the equations for the elements of the density matrix become,

$$\frac{d\,\rho_{11}(t)}{dt} = \frac{\rho_{22}(t)}{T_1} + i\frac{\Omega_R}{2}\left[\rho_{21}(t)\exp(i\omega t + i\phi) - \rho_{12}(t)\exp(-i\omega t - i\phi)\right]$$

$$\frac{d\,\rho_{22}(t)}{dt} = -\frac{\rho_{22}(t)}{T_1} - i\frac{\Omega_R}{2}\left[\rho_{21}(t)\exp(i\omega t + i\phi) - \rho_{12}(t)\exp(-i\omega t - i\phi)\right]$$

$$\frac{d\,\rho_{12}(t)}{dt} = -\frac{\rho_{12}(t)}{T_2} + i\frac{(\varepsilon_2 - \varepsilon_1)}{\hbar}\rho_{12}(t) + i\frac{\Omega_R}{2}\exp(i\omega\,t + i\phi)\,[\rho_{22}(t) - \rho_{11}(t)]$$

$$\frac{d\,\rho_{21}(t)}{dt} = --\frac{\rho_{12}(t)}{T_2} - i\frac{(\varepsilon_2 - \varepsilon_1)}{\hbar}\rho_{21}(t) - i\frac{\Omega_R}{2}\exp(-i\omega\,t - i\phi)\,[\rho_{22}(t) - \rho_{11}(t)]$$

The equations for the components, $V_x(t)$, $V_y(t)$, and $V_z(t)$ are now as follows,

$$\frac{dV_x(t)}{dt} = -\frac{1}{T_2}V_x(t) - \frac{\Delta}{\hbar}V_y(t)$$

$$\frac{dV_y(t)}{dt} = -\frac{1}{T_2}V_y(t) + \frac{\Delta}{\hbar}V_x(t) + \Omega_R\,V_z(t)$$

$$\frac{dV_z(t)}{dt} = -\frac{(V_z(t)+1)}{T_1} - \Omega_R\,V_y(t)$$

One can no longer write the above set of equations in the compact form,

$$\frac{d\vec{V}(t)}{dt} = \vec{\Omega}\,X\,\vec{V}(t)$$

Also note that the magnitude of the vector $\vec{V}(t)$ is not conserved anymore. The above equations have a well defined steady state solution,

$$V_z(t\to\infty) = \frac{-\left[1+\left(\frac{\Delta}{\hbar}T_2\right)^2\right]}{1+\left(\frac{\Delta}{\hbar}T_2\right)^2 + \Omega_R^2 T_2 T_1}$$

$$V_y(t\to\infty) = \frac{-\Omega_R T_2}{1+\left(\frac{\Delta}{\hbar}T_2\right)^2 + \Omega_R^2 T_2 T_1}$$

$$V_x(t\to\infty) = \frac{\left(\frac{\Delta}{\hbar}T_2\right)\Omega_R T_2}{1+\left(\frac{\Delta}{\hbar}T_2\right)^2 + \Omega_R^2 T_2 T_1}$$

The above expressions for the steady state show that electromagnetic radiation, no matter how strong, cannot create population inversion in steady state (i.e. make the z-component $V_z(t)$ positive in steady state).

Optical Control by Short Pulses: Short pulses of light can be used to control and/or prepare desired quantum states of a two-level system. For example, consider a situation where **detuning is zero** and population relaxation and decoherence times are sufficiently long. Suppose, $|\psi(t=0)\rangle = |e_1\rangle$. Radiation is switched on at time $t=0$ and switched off at time given by $|\vec{\Omega}|t = \Omega_R t = \pi$. Such a radiation pulse is called a π pulse. At the end of the pulse, the value of $V_z(t)$ is approximately +1 and population inversion is achieved. The quantum state of the two-level system at the end of the π pulse is,

$$|\psi(t)\rangle = i\,e^{-i\frac{\varepsilon_2}{\hbar}t - i\phi}\,|e_2\rangle_{\;t=\frac{\pi}{\Omega_R}}$$

The population will, of course, relax into the lower state after the pulse because of T_1. Similarly, as shown earlier, a $\pi/2$ pulse (i.e. a pulse whose duration in time is $\pi/2|\vec{\Omega}| = \pi/2\Omega_R$) can be used to take an electron from an initial state $|e_1\rangle$ into a linear superposition of $|e_1\rangle$ and $|e_2\rangle$ given by,

$$|\psi(t)\rangle = \frac{1}{\sqrt{2}}\left[e^{-i\frac{\varepsilon_1}{\hbar}t} |e_1\rangle + i\, e^{-i\frac{\varepsilon_2}{\hbar}t - i\phi} |e_2\rangle \right]_{t=\frac{\pi}{2\Omega_R}}$$

2.4 Photon Echo Experiments

Photon echo is a useful experimental technique to characterize decoherence times in materials. Consider an isolated two-level system (in the absence of any radiation) in which the mean position of the electron in both states, $|e_1\rangle$ and $|e_2\rangle$, is zero, and,

$$\vec{d} = \langle e_2|\vec{r}|e_1\rangle = \langle e_1|\vec{r}|e_2\rangle$$

Now consider the superposition state,

$$|\psi(t=0)\rangle = e^{i\alpha}\left(\frac{|e_1\rangle + e^{-i\theta}|e_2\rangle}{\sqrt{2}}\right) \quad \Rightarrow \quad |\psi(t)\rangle = e^{i\alpha}\left(\frac{e^{-i\frac{\varepsilon_1}{\hbar}t}|e_1\rangle + e^{-i\theta}\, e^{-i\frac{\varepsilon_2}{\hbar}t}|e_2\rangle}{\sqrt{2}}\right)$$

The mean position of the electron in such a superposition state is,

$$\langle\psi(t)|\,\vec{r}\,|\psi(t)\rangle = \vec{d}\,\cos\left(\frac{\varepsilon_2 - \varepsilon_1}{\hbar}t + \theta\right)$$

The charge density associated with the electron wavefunction oscillates in space with a frequency related to the energy level difference. Electron in such a superposition state can therefore radiate electromagnetic energy, just like a classical dipole antenna. Radiation from one such electron is too difficult to detect. Radiation from many such electrons in a collection of two-level systems would also be difficult to detect unless all the electrons were oscillating in-phase. The trick is to get many electrons to oscillate in-phase. Electron states in many materials, such as semiconductors, can be modeled as a collection of two-level systems. In most cases, electron energies of the upper and/or levels would have a range of values. In other words, the energy difference $\Delta\varepsilon/\hbar$, or the detuning $\Delta/\hbar$ measured with respect to some fixed ω, would not be the same for all the two-level systems. For some systems, Δ would be zero, for some Δ would be positive, and for some Δ would be negative. Therefore, charge oscillations in different two-level systems would not stay in-phase for long. Note that the superposition state,

$$|\psi(t=0)\rangle = e^{i\alpha}\left(\frac{|e_1\rangle + e^{-i\theta}|e_2\rangle}{\sqrt{2}}\right) \quad \Rightarrow \quad |\psi(t)\rangle = e^{i\alpha}\left(\frac{e^{-i\frac{\varepsilon_1}{\hbar}t}|e_1\rangle + e^{-i\theta}\, e^{-i\frac{\varepsilon_2}{\hbar}t}|e_2\rangle}{\sqrt{2}}\right)$$

corresponds to a vector $\vec{V}(t)$ with the following components,

$$V_x(t) = \cos\left(\frac{\Delta}{\hbar}t + \theta - \phi\right) \qquad V_y(t) = \sin\left(\frac{\Delta}{\hbar}t + \theta - \phi\right) \quad V_z(t) = 0$$

The argument of the cosine and the sine is also the time-dependent phase of the charge oscillation relative to ωt. If for two different two-level systems the vectors $\vec{V}(t)$ were to become identical at any point in time, then at that moment the electron charge in these two two-level systems will be oscillating in-phase.

Photon echo techniques are able to achieve in-phase charge oscillations in a collection of two-level systems with even different values of $\Delta\varepsilon/\hbar$. Consider such a collection of two-level systems interacting with radiation of frequency ω. Suppose a short and strong ($\Omega_R >> \Delta/\hbar$) electromagnetic $\pi/2$ pulse is used to excite the two-level systems assumed to be all initially in the ground state. The dynamics of each two-level system are governed by the equations,

$$\frac{dV_x(t)}{dt} = -\frac{1}{T_2}V_x(t) - \frac{\Delta}{\hbar}V_y(t)$$

$$\frac{dV_y(t)}{dt} = -\frac{1}{T_2}V_y(t) + \frac{\Delta}{\hbar}V_x(t) + \Omega_R\, V_z(t)$$

$$\frac{dV_z(t)}{dt} = -\frac{(V_z(t)+1)}{T_1} - \Omega_R\, V_y(t)$$

Assume that decoherence and population relaxation times are very long. Right after the pulse, the vector $\vec{V}(t)$ equals $-\hat{y}$ for all the two-level systems, as shown in Fig.(a) below. The subsequent evolution of the two-level systems is according to the equations,

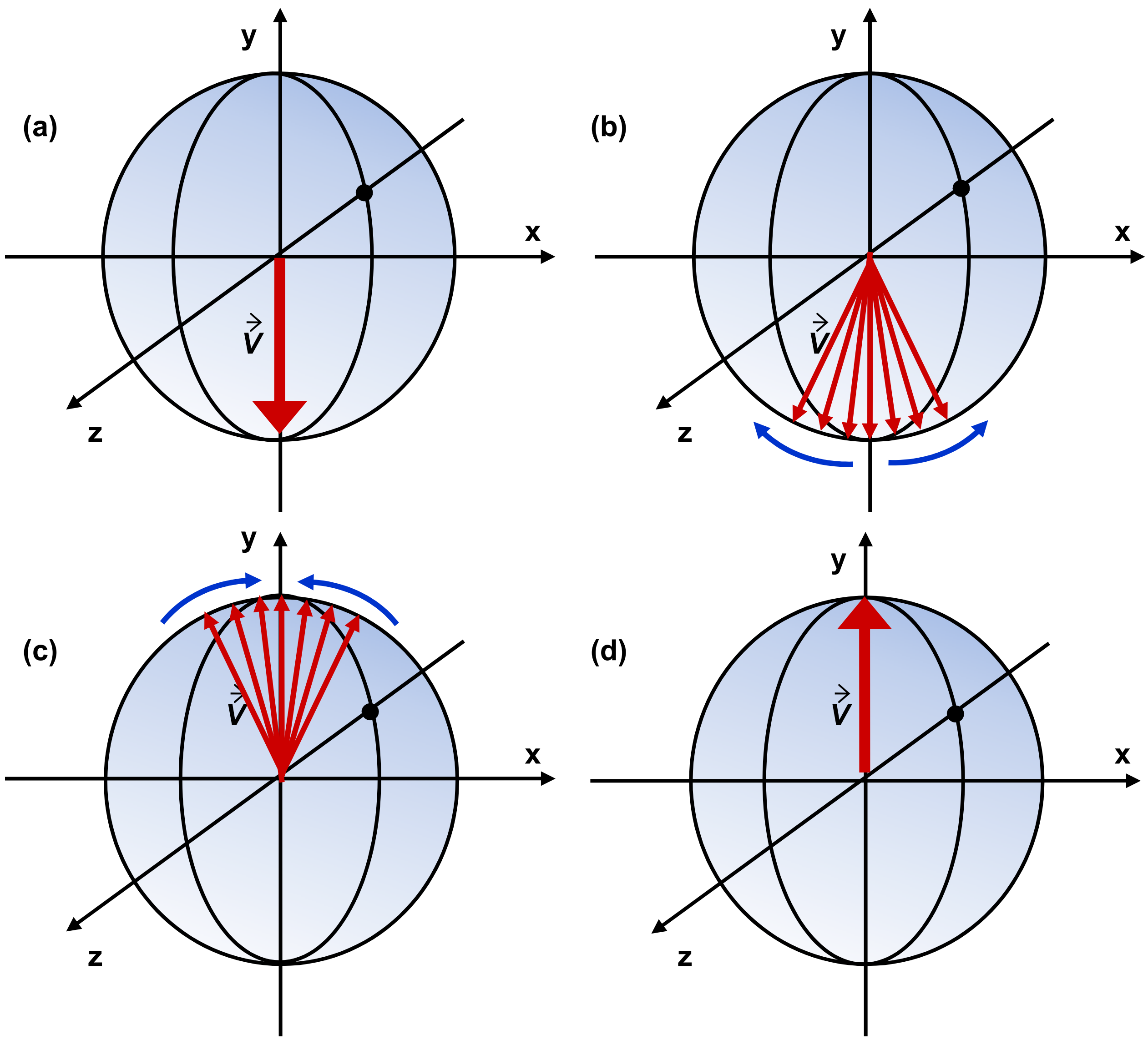
$$\frac{dV_x(t)}{dt} \approx -\frac{\Delta}{\hbar}V_y(t) \qquad \frac{dV_y(t)}{dt} \approx +\frac{\Delta}{\hbar}V_x(t) \qquad \frac{dV_z(t)}{dt} \approx 0$$

The above equations show that the vectors $\vec{V}(t)$ of two-level systems with different detunings move apart in the x-y plane as shown below in Fig.(b). After some time, say T_d, another short and strong ($\Omega_R >> \Delta/\hbar$) electromagnetic π pulse is used. The π pulse rotates the vectors $\vec{V}(t)$ of all two level systems by 180-degrees around the x-axis and the final positions of the vectors $\vec{V}(t)$ are as shown in Fig.(c) below. Right after the π pulse, the free evolution of the two-level systems is again according to the equations,

$$\frac{dV_x(t)}{dt} \approx -\frac{\Delta}{\hbar}V_y(t) \qquad \frac{dV_y(t)}{dt} \approx +\frac{\Delta}{\hbar}V_x(t) \qquad \frac{dV_z(t)}{dt} \approx 0$$

The π pulse changed the sign of the V_y component of every two-level system, but did not change the sign of the V_x component. Consequently, the vectors $\vec{V}(t)$ of two-level systems with different detunings, which were moving apart in the x-y plane before the π pulse, now start moving closer with time. At time equal to exactly T_d after the π pulse, the vectors $\vec{V}(t)$ of al two-level systems come together, as shown in Fig.(d). At this point in time, electrons in all the two-level systems are oscillating in-phase. Consequently, the radiation emitted by them can easily be detected. Because of different detunings, charge oscillations in different two-level systems will soon go out of phase with each other. In actual experiments, a weak but detectable radiation pulse is detected at the moment when the electrons in all the two-level systems are oscillating in-phase. This pulse is called the photon echo pulse.

Now suppose decoherence is present. The coherences will get reduced the longer the time delay T_d is compared to the decoherence time T_2 and, therefore, the strength of the photon echo pulse will also get reduced as T_d is made longer. Thus, if the strength of the photon echo pulse is measured as a function of the time delay T_d then this information can be used to extract the decoherence time T_2 (assuming $T_1 >> T_2$).



2.5 Ramsey Fringes and Atomic Clocks

Consider the problem of determining the frequency ω of a radiation source with high accuracy (fractional accuracy better than one part in 10^{14}). One way to do this would be to make an atom, or a two-level system, interact with the radiation from the source. Suppose the energy level separation of the two-level system is known to be $\Delta\varepsilon$ with a very high accuracy. The frequency of the source is assumed to be close to $\Delta\varepsilon/\hbar$ but the detuning $\Delta/\hbar$ is unknown and needs to be determined with high precision. Suppose, one considers the following scheme: the two-level system is made to interact with the radiation from the source for a duration T_{int}. Immediately afterwards, the upper state occupation (i.e. ρ_{22}) is measured. The question is whether this absorption experiment can be used to determine $\Delta/\hbar$. We will assume that decoherence and population relaxation times are very long. Suppose the duration T_{int} is chosen such that the radiation appears to the two-level system as a π pulse ($|\vec{\Omega}|T_{int} = \pi$). But $|\vec{\Omega}| = \sqrt{\Omega_R^2 + (\Delta/\hbar)^2}$ and

$\Delta/\hbar$ is unkown. To overcome this problem, one can deploy a sufficiently strong pulse such that $\Omega_R >> |\Delta|/\hbar$. After the pulse, the value of ρ_{22} is,

$$\rho_{22} = \frac{\Omega_R^2}{\Omega_R^2 + (\Delta/\hbar)^2}$$

If $\Omega_R >> |\Delta|/\hbar$, the above expression shows that ρ_{22} is close to unity irrespective of the value of $\Delta/\hbar$. Therefore, this simple absorption experiment will not be able to determine $\Delta/\hbar$ with high precision. Atomic coherences can be exploited to obtain much better accuracies, as we will see now.

2.5.1 Ramsey Fringes

Consider a two level system prepared in the ground state, i.e. $|\psi(t=0)\rangle = |e_1\rangle$. At time $t=0$, a strong $\pi/2$ pulse ($\Omega_R >> |\Delta|/\hbar$) of duration T_{int} and detuning $\Delta/\hbar$ excites the system. The dynamics of the two-level system are governed by the equations,

$$\frac{dV_x(t)}{dt} = -\frac{1}{T_2}V_x(t) - \frac{\Delta}{\hbar}V_y(t)$$

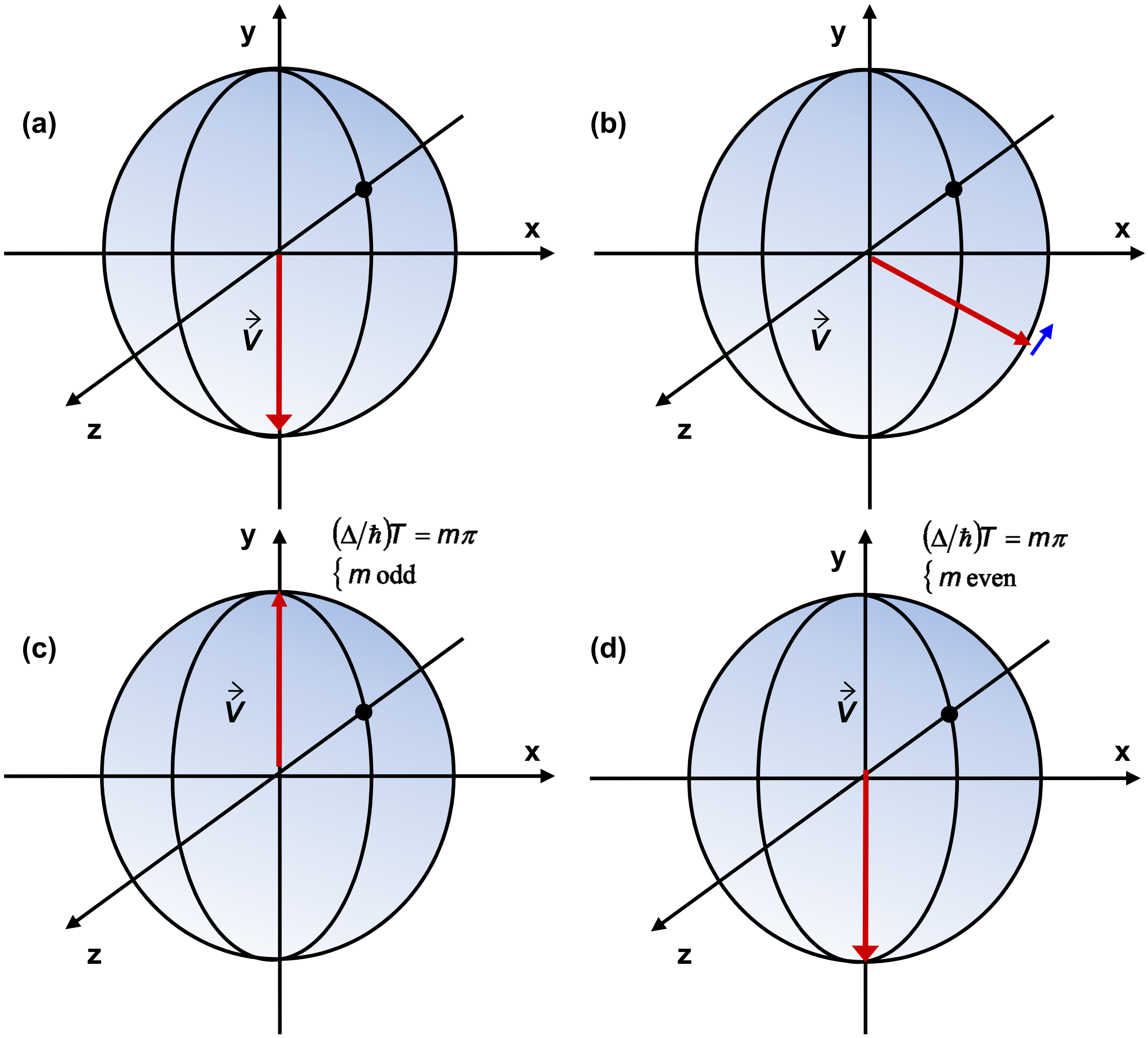
$$\frac{dV_y(t)}{dt} = -\frac{1}{T_2}V_y(t) + \frac{\Delta}{\hbar}V_x(t) + \Omega_R\, V_z(t)$$

$$\frac{dV_z(t)}{dt} = -\frac{(V_z(t)+1)}{T_1} - \Omega_R\, V_y(t)$$

Assume that decoherence and population relaxation times are very long. After the pulse, the state vector $\vec{V}(t)$ equals $-\hat{y}$ (Fig.(a) below). The subsequent free evolution of the two-level system is according to the equations (Fig.(b) below),

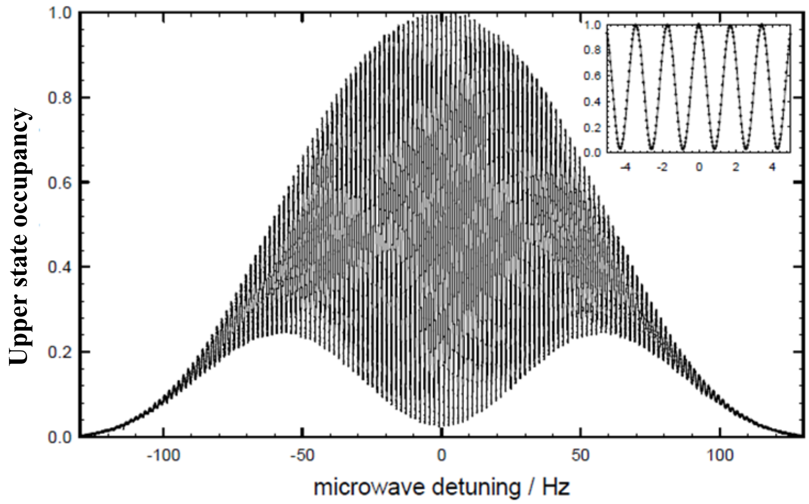
$$\frac{dV_x(t)}{dt} \approx -\frac{\Delta}{\hbar}V_y(t) \qquad \frac{dV_y(t)}{dt} \approx +\frac{\Delta}{\hbar}V_x(t) \qquad \frac{dV_z(t)}{dt} \approx 0$$

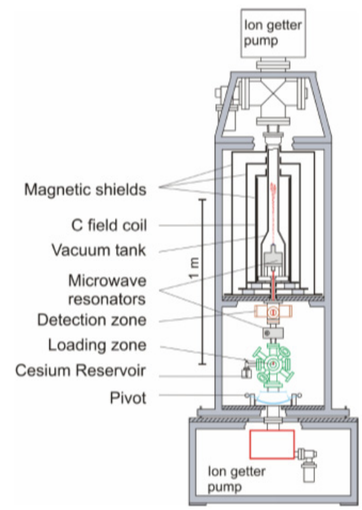
The vector $\vec{V}(t)$ rotates in the x-y plane with a frequency equal to $|\Delta|/\hbar$ for a duration T (Fig.(b) below). After this duration T, a second $\pi/2$ pulse of duration T_{int} interacts with the two-level system. If the time duration T is such that $(|\Delta|/\hbar)T = m\pi$, where m is any positive odd integer, and the state vector $\vec{V}(t)$ equals $+\hat{y}$ (Fig,(c) below) , then the second $\pi/2$ pulse will make the state vector $\vec{V}(t)$ equal to $-\hat{z}$ and the two-level system will be in the lower state $|e_1\rangle$ after the second pulse. On the other hand, if the time duration T is such that $(|\Delta|/\hbar)T = m\pi$, where m is any positive even integer, and the state vector $\vec{V}(t)$ equals $-\hat{y}$ (Fig,(d) below), then the second $\pi/2$ pulse will make the state vector $\vec{V}(t)$ equal to $+\hat{z}$ and the two-level system will be in the upper state $|e_2\rangle$ after the second pulse. It is not difficult to show that under the assumptions $\Omega_R >> |\Delta|/\hbar$ and very long decoherence and relaxation times, the value of V_z at the end of the second pulse is given by $\cos\left(\frac{\Delta}{\hbar}T\right)$, and the occupancy of the upper state is given by $0.5\left[1+\cos\left(\frac{\Delta}{\hbar}T\right)\right]$. Therefore, if one makes a measurement on the two-level system after the second pulse and determines whether the system is in the upper state or the lower state, then, knowing T, the detuning $\Delta/\hbar$ of the radiation from the energy level separation of the two-level system can be determined to a very high accuracy. Note that frequency accuracy in this case is not determined by the time of interaction T_{int} of the two-level system with the radiation but by the duration T between the two $\pi/2$ pulses which can be made very long subject to the constrains imposed by decoherence and relaxation times.



2.5.2 Cesium Atomic Clocks

The principle discussed above is used in atomic clocks which are very stable and high-precision frequency sources. In a Cesium atomic clock, radiation from a tunable RF oscillator is coupled to Cesium atoms which have an energy level separation close to ~9.192 GHz. The frequency of the RF oscillator can drift over time and needs to be locked to a stable reference with high precision. The principle of operation of the clock is as follows. Cesium atoms are prepared in the ground state and made to interact twice with $\pi/2$ pulses from the RF source. The duration between these pulses is T . At the end of the second pulse, the upper state occupancy of the Cesium atoms is determined. This procedure is repeated many times while the frequency of the RF source is varied. A typical plot of the measured upper state occupancy after the second pulse vs the detuning of the RF source is shown in the Figure below.





The observed oscillations are called Ramsey fringes (after Norman Ramsey). Note that the width of the center fringe (in Hz) is $1/T$ (see the inset). A more detailed analysis shows that if one does not make the assumption $\Omega_R >> \Delta/\hbar$ and also does not assume that decoherence and relaxation times are infinitely long, then the observed envelope of the oscillations can also be reproduced. Using feedback from these measurements, the frequency of the RF oscillator is adjusted and locked to the energy level separation of the Cesium atoms. In an actual Cesium fountain clock (see Figure above), Cesium atoms are prepared in the ground state and hurled upwards against gravity (like water in a fountain). They pass through a microwave a cavity twice; on their way up and then again on their way down. While passing through the cavity, the atoms interact with the radiation from the RF source. Finally, the state of the Cesium atoms is detected and the cycle is repeated. Recently, ultra-cold atoms with temperatures less than μK have been used to reduce decoherence and relaxation rates resulting in very stable atomic clocks. The root-mean-square fractional frequency stability obtained in typical Cesium atomic clocks is given by the expression,

$$\frac{\Delta\omega}{\omega} \sim \frac{1}{\pi\omega T}\sqrt{\frac{T_c}{\tau}}\sqrt{\frac{1}{N_a}}$$

Here, T_c is the cycle time (time needed to complete one measurement), τ is the integration time used in estimating the frequency, and N_a is the number of atoms used in one cycle. Typical values of T_c and N_a are 1 second and 6×10^5 atoms, respectively. This gives,

$$\frac{\Delta\omega}{\omega} \sim \frac{3\times 10^{-14}}{\sqrt{\tau}} \qquad \{\ \tau \text{ is in seconds}$$

Very stable RF sources, with fractional stabilities smaller than 10^{-16}, have been realized using these principles.