2. The Optical Bloch Equation

2.1 Introduction

The interaction between an atom and optical fields can be approximated as a two-level transition. Conceptually, the two-level atom can be treated as a spin-one-half particle in a magnetic field. To describe the systems, we employ a fictitious electric spin vector, say pseudospin vector. This is related to the atom's dipole moment and its inversion. The time dependent equations are called the optical Bloch equations. We also talk about the rotating wave approximation (RWA), which reduces into pseudospin dynamics.

2.2 Real Atom and the Two-Level Atom

It is difficult to predict the energy levels of multiple atoms due to its complexity. However, we can determine the states of many-electron-atom system with L-S (Russell-Saunders) coupling. In fact, the ideal atoms can be dealt with two-energy level for many purposes. In a word, it ca be approximated in terms of emission and absorption. The sodium has two adjacent spectra lines due to spin. The difference between them is 6 Å, but it takes longer in collision cases. For example, the collision time due to dilute gas of $10^{10}-10^{13}\text{atoms/cm}^3$ is of the order of $10^{7}\,\text{sec}$. Namely, the frequency line width $\delta\omega_H$ will be the order of $10^{8}\,\text{Hz}$. From the ratio between the angular frequencies and the wavelengths, we have

$$\delta\lambda_H = \lambda\frac{\delta\omega_H}{\omega} \approx 10^{-3}\ \text{Å}.$$

It is obviously small compared with the D line spectrum width. In inhomogeneous gas, the frequency is much greater than that in homogeneous gas because of the Doppler Effect. Now let us summarize the discussion above. In terms of wavelength, we have following conclusion:

$$\text{D lines width } (6\,\text{Å}) >> \delta\lambda_I \ \ (10^{-1}\,\text{Å}) >> \ \delta\lambda_H\,(10^{-3}\ \text{Å})$$

where subscript I and H denote inhomogeneous and homogeneous, respectively. As mentioned, the spectral width is very narrow compared with D lines width, which is derived from the spin. Considering a coherent quasi-monochromatic field, only sub-picosecond pulses have such a large spectral width because of the relation, $\frac{1}{T} = \delta\omega$. Now we should know the following: In order to embrace two-level absorption, only using optical pulse is not useful. For

example, the transition between $3p^2P_{3/2}(F=2)$ and $3s^2S_{1/2}(F=1)$ has 35 MHz as the natural width. It implies that the probe pulse must be narrower than the frequency and longer than $3\times10^8\,\text{sec}$ as duration time. If we use an appropriate pulse, it can be treated as a two-level system.

2.3 Tow-Level Atoms and Semiclassical Radiation Theory

We will discuss the radiation theory as a two-level system with a semiclassical way. It is an important concept for resonance and quantum optics.

The optical interaction with a material uses a driving field, which can be the electric field. In that case, a dipole moment can be used for the energy change. The Hamiltonian will be

$$\hat{H} = \hat{H}_A - \hat{d}\cdot\hat{E}(r_0). \tag{2.1}$$

The unperturbed Hamiltonian H_A can be any associated with the physical systems. We here use hydrogen atom. As mentioned, we can assume that the system can be described by two values, $|+\rangle$ and $|-\rangle$. This is of great interest for dealing with such problems. It can be expressed with matrix elements:

$$\langle +|\hat{H}_A|+\rangle = W_+, \qquad \langle +|\hat{H}_A|-\rangle = 0,$$

$$\langle -|\hat{H}_A|+\rangle = 0, \qquad \langle -|\hat{H}_A|-\rangle = W_-;$$

and

$$\langle +|\hat{d}|+\rangle = 0, \qquad \langle +|\hat{d}|-\rangle = d_{+-},$$

$$\langle -|\hat{d}|+\rangle = (d_{+-})^*, \qquad \langle -|\hat{d}|-\rangle = 0.$$

The W_+ and W_- are the assigned energies. There are no diagonal elements of $\hat{d}$ because of the vector operator having odd parity. In order to obtain the elements of $\hat{d}$, we take the states, (211) and (100). The values can be obtained because we know the perturbation method in Stark effect. Thus,

$$d_{+-} = \int \Psi^*_{211}(r) e\hat{r} \Psi_{100}(r) d^3 r . \tag{2.2}$$

Taking radial and angular integral of this, we have

$$d_{+-} = -\frac{2^7}{3^5} e a_0 (\mathbf{x} - i\mathbf{y}), \tag{2.3}$$

The value has an imprecision due to phase arbitrariness. In general, the dipole matrix elements are complex vectors:

$$d_{+-} = d_r + i d_i ; \qquad d_{-+} = d_r - i d_i ,$$

In this example,

$$d_r = -\left(\frac{2^7}{3^5}\right) e a_0 \mathbf{x} \tag{2.4}$$

and

$$d_i = \left(\frac{2^7}{3^5}\right) e a_0 \mathbf{y} . \tag{2.5}$$

As a result, we obtain the matrix elements in the two-dimensional space.

$$\hat{d} \Rightarrow \begin{bmatrix} 0 & d_r + i d_i \\ d_r - i d_i & 0 \end{bmatrix}. \tag{2.6}$$

Using Pauli matrices, we finally get

$$\hat{d} = d_r\hat{\sigma}_1 - d_i\hat{\sigma}_2 . \tag{2.7}$$

Similarly we have

$$\hat{H}_A = \tfrac{1}{2}(W_+ + W_-)\hat{\mathbf{I}} + \tfrac{1}{2}(W_+ - W_-)\hat{\sigma}_3 . \tag{2.8}$$

The total Hamiltonian is:

$$\hat{H} = \tfrac{1}{2}(W_+ + W_-)\hat{\mathbf{I}} + \tfrac{1}{2}(W_+ - W_-)\hat{\sigma}_3 - (d_r \cdot \hat{E})\hat{\sigma}_1 + (d_i \cdot \hat{E})\hat{\sigma}_2 . \tag{2.9}$$

In order to get time-dependent equations of motion, we use

$$i\hbar\dot{\hat{O}} = \left[\hat{O}, \hat{H}\right], \tag{2.10}$$

and the relation

$$[\hat{\sigma}_1, \hat{\sigma}_2] = 2i\hat{\sigma}_3 \qquad \text{et cycl.} \tag{2.11}$$

Using the above relationships, we can have the equation of motion as follows:

$$i\hbar\frac{d\hat{\sigma}_1}{dt} = \left[\hat{\sigma}_1, \hat{H}\right]$$

$$= \frac{1}{2}(W_+ - W_-)[\hat{\sigma}_1, I] - i\frac{1}{2}(W_+ - W_-)[\hat{\sigma}_1, \hat{\sigma}_3] - (\mathbf{d_r} \cdot \hat{\mathbf{E}})[\hat{\sigma}_1, \hat{\sigma}_1] + (\mathbf{d_i} \cdot \hat{\mathbf{E}})[\hat{\sigma}_1, \hat{\sigma}_2]$$

$$= 0 - i\hbar\omega_0\hat{\sigma}_2 - 0 + 2i(\mathbf{d_i} \cdot \hat{\mathbf{E}})\hat{\sigma}_3 .$$

The three operators of Pauli matrix therefore obey these equations:

$$\dot{\hat{\sigma}}_1(t) = -\omega_0\hat{\sigma}_2(t) + \frac{2}{\hbar}\left[d_i \cdot \hat{E}(t)\right]\hat{\sigma}_3(t) \tag{2.12a}$$

$$\dot{\hat{\sigma}}_2(t) = \omega_0\hat{\sigma}_1(t) + \frac{2}{\hbar}\left[d_r \cdot \hat{E}(t)\right]\hat{\sigma}_3(t) \tag{2.12b}$$

$$\dot{\hat{\sigma}}_3(t) = -\frac{2}{\hbar}\left[d_r \cdot \hat{E}(t)\right]\hat{\sigma}_2(t) - \frac{2}{\hbar}\left[d_i \cdot \hat{E}(t)\right]\hat{\sigma}_1(t). \tag{2.12c}$$

We define

$$\omega_0 \equiv \frac{W_+ - W_-}{\hbar} \tag{2.13}$$

to represent the atomic transition frequency.

We hereinafter interpret the above equations as semiclassical, so we use $s_i(t)$ which are the expectation values instead of the operator notation, $\hat{\sigma}_i(t)$. If we employ a wave function, $|\psi\rangle = a|+\rangle + b|-\rangle$, we will have

$$s_1(0) = \langle\psi|\hat{\sigma}_1|\psi\rangle = a^* b + a b^*,$$

$$s_2(0) = \langle\psi|\hat{\sigma}_2|\psi\rangle = -i\left(a^* b - a b^*\right),$$

$$s_3(0) = \langle\psi|\hat{\sigma}_3|\psi\rangle = |a|^2 - |b|^2.$$

Therefore,

$$s_1^2(0) + s_2^2(0) + s_3^2(0) = \left(|a|^2 + |b|^2\right)^2 = \text{constant}.$$

But if $|a|^2 + |b|^2 = 1$,

$$s_1^2(0) + s_2^2(0) + s_3^2(0) = 1. \tag{2.14}$$

In order to make (2.12) simple, we adjust the arbitrary phases so that $\mathbf{d}_i$ vanishes, and we use following notation:

$$\frac{2}{\hbar}\mathbf{d}_\mathbf{r} \cdot \mathbf{E} \equiv \frac{2d}{\hbar}\mathbf{u}_\mathbf{d} \cdot \mathbf{E} \equiv \kappa E.$$

In addition, we have to use $s_i(t)$ mentioned above. Consequently,

$$\dot{s}_1(t) = -\omega_0 s_2(t), \tag{2.15a}$$

$$\dot{s}_2(t) = \omega_0 s_1(t) + \kappa E(t, \mathbf{r_0}) s_3(t), \tag{2.15b}$$

$$\dot{s}_3(t) = -\kappa E(t, \mathbf{r_0}) s_2(t) \tag{2.15c}$$

As a result, they are recognized as the electric-dipole analogues of equations that express spin precession in magnetic resonance. They provide the kinetic theory for a two-level atom.

2.4 The Rotating Wave Approximation

The equations in (2.15) can be rewritten as follows:

$$\frac{d}{dt}\mathbf{s}(t) = \mathbf{\Omega}^F(t) \times \mathbf{s}(t), \tag{2.16}$$

where the vector $\mathbf{s}$ has three components, and the "torque" vector $\mathbf{\Omega}^F(t)$ has components:

$$\Omega_1^F(t) = -\kappa E, \tag{2.17a}$$

$$\Omega_2^F(t) = 0, \tag{2.17b}$$

$$\Omega_3^F(t) = \omega_0. \tag{2.17c}$$

We estimate that the magnitude of (2.17a) is roughly equal to that of (2.17b); namely, $\hbar\kappa E \approx \hbar\omega_0$. Therefore, the field can be expressed as $E \approx \frac{\omega_0}{\kappa} = \frac{\omega_0 \hbar}{2d} = \frac{\hbar\omega_0}{e a_0}$ where a_0 and the Bohr radius. The energy of an optical transition is of order 1 eV. Thus, we can obtain $E \approx 1V/a_0$ or $E \approx 10^8 V/cm$. This corresponds to power densities of the order of $10^{15} V/cm^2$, which can be attained with lasers. On the other hand, we can approximate the energy difference as $\hbar\omega_0 \approx e^2/a$ where a is the radius around a single charged central core composed of the nucleus and the inner shell electrons. However, the external field may be estimated as very strong. In fact, we should safely take the inequality, $\kappa E << \omega_0$ to be well-satisfied in every situation of interest in optical resonance. Thus, the "torque" vector, $\mathbf{\Omega}^F$, is pointed almost straight along with the **3** (z) axis. The torque vector, $\mathbf{\Omega}^F$, and

pseudo-vector $\mathbf{s}$ are oscillating at frequency ω_0. It is interesting the transition toward nearly resonant state due to the applied field, $E(t)$.

$$E(t) = \varepsilon(t)\left[e^{i\omega t} + \text{c.c.}\right]. \tag{2.18}$$

In the case that $\omega \approx \omega_0$, the vectors change rapidly. The following assumption properly explains the preceding statement. The "torque" vector is rewritten as the sum of three "torques":

$$\mathbf{\Omega}^F = \mathbf{\Omega}^+(t) + \mathbf{\Omega}^-(t) + \mathbf{\Omega}^0, \tag{2.19}$$

where

$$\Omega^0 = (0,0,\omega_0), \tag{2.20a}$$

$$\Omega^+ = (-\kappa\varepsilon\cos\omega t, -\kappa\varepsilon\sin\omega t, 0), \tag{2.20b}$$

$$\Omega^- = (-\kappa\varepsilon\cos\omega t, +\kappa\varepsilon\sin\omega t, 0). \tag{2.20c}$$

$\Omega^{\pm}$ depend on time and are related to resonant effect. Ω^0 is along with the 3 axis. Ω^+ rotates as a right-handed screw as t increases. Ω^- does in the opposite direction. However, Ω^- is not real, so the "torque" vector will be expressed in terms of $\Omega^+ + \Omega^0$, which is called the rotating wave approximation (RWA). Using (2.16), we can obtain:

$$\begin{aligned}\frac{d}{dt}\mathbf{s}(t) &= \mathbf{\Omega}^F(t)\times\mathbf{s}(t)\\ &= \left[\mathbf{\Omega}^+ + \mathbf{\Omega}^0\right]\times\mathbf{s}.\end{aligned}$$

where $\mathbf{\Omega}^+ + \mathbf{\Omega}^0 = (-\kappa\varepsilon\cos\omega t, -\kappa\varepsilon\sin\omega t, \omega_0)$.

$$\begin{aligned}\tfrac{d}{dt}\mathbf{s} = &\left(-\kappa\varepsilon\cos\omega t\cdot s_3 - \omega_0 s_2\right)_{\mathbf{1}} + \left(\kappa\varepsilon\cos\omega t\cdot s_3 + \omega_0 s_1\right)_{\mathbf{2}}\\ &+ \left(-\kappa\varepsilon\cos\omega t\cdot s_2 + \kappa\varepsilon\sin\omega t\cdot s_1\right)_{\mathbf{3}}\end{aligned}$$

In other words,

$$\frac{d}{dt}\begin{bmatrix} s_1 \\ s_2 \\ s_3 \end{bmatrix} = \begin{bmatrix} 0 & -\omega_0 & -\kappa\varepsilon\sin\omega t \\ \omega_0 & 0 & \kappa\varepsilon\cos\omega t \\ \kappa\varepsilon\sin\omega t & -\kappa\varepsilon\cos\omega t & 0 \end{bmatrix}\begin{bmatrix} s_1 \\ s_2 \\ s_3 \end{bmatrix}. \tag{2.21}$$

In order to make (2.21) simpler, we employ the following notations:

$$\vec{s} = \begin{bmatrix} s_1 \\ s_2 \\ s_3 \end{bmatrix}, \quad \vec{\rho} = \begin{bmatrix} u \\ v \\ w \end{bmatrix}, \quad \mathbf{A}^{\pm 1} = \begin{bmatrix} \cos\omega t & \pm\sin\omega t & 0 \\ \mp\sin\omega t & \cos\omega t & 0 \\ 0 & 0 & 1 \end{bmatrix}, \text{ and}$$

$$\mathbf{B} = \begin{bmatrix} 0 & -\omega_0 & -\kappa\varepsilon\sin\omega t \\ \omega_0 & 0 & \kappa\varepsilon\cos\omega t \\ \kappa\varepsilon\sin\omega t & -\kappa\varepsilon\cos\omega t & 0 \end{bmatrix}.$$

From (2.21), we can express

$$\dot{\vec{s}} = \mathbf{B}\vec{s}\,. \tag{2.22}$$

Use a rotating matrix $\mathbf{A}$ that is unitary.

$$\vec{\rho} = \mathbf{A}\vec{s} \quad \text{or} \quad \vec{s} = \mathbf{A}^{-1}\vec{\rho}\,. \tag{2.23}$$

Thus,

$$\dot{\vec{s}} = \mathbf{B}\mathbf{A}^{-1}\vec{\rho} \tag{2.24}$$

Because $\vec{\rho} = \mathbf{A}\vec{s}$, the result of the derivative with respect to time is:

$$\dot{\vec{\rho}} = \dot{\mathbf{A}}\vec{s} + \mathbf{A}\dot{\vec{s}}\,. \tag{2.25}$$

Using (2.23), and (2.24), we obtain

$$\dot{\vec{\rho}} = \dot{\mathbf{A}}\mathbf{A}^{-1}\vec{\rho} + \mathbf{A}\mathbf{B}\mathbf{A}^{-1}\vec{\rho}\,. \tag{2.26}$$

As a result,

$$\begin{bmatrix} \dot{u} \\ \dot{v} \\ \dot{w} \end{bmatrix} = \begin{bmatrix} 0 & \omega & 0 \\ -\omega & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix} \begin{bmatrix} u \\ v \\ w \end{bmatrix} + \begin{bmatrix} 0 & -\omega_0 & 0 \\ \omega_0 & 0 & \kappa\varepsilon \\ 0 & -\kappa\varepsilon & 0 \end{bmatrix} \begin{bmatrix} u \\ v \\ w \end{bmatrix}$$

$$\therefore \quad \begin{bmatrix} \dot{u} \\ \dot{v} \\ \dot{w} \end{bmatrix} = \begin{bmatrix} 0 & -(\omega_0 - \omega) & 0 \\ \omega_0 - \omega & 0 & \kappa\varepsilon \\ 0 & -\kappa\varepsilon & 0 \end{bmatrix} \begin{bmatrix} u \\ v \\ w \end{bmatrix}. \tag{2.27a}$$

We can rewrite it as

$$\frac{d}{dt}\vec{\rho} = \mathbf{\Omega} \times \vec{\rho} \tag{2.27b}$$

Provided that the rotating frame torque vector has

$$\mathbf{\Omega} \equiv (-\kappa\varepsilon, 0, \omega_0 - \omega). \tag{2.27c}$$

According to (2.27a), we can conclude that v is the absorptive component of the dipole moment, while u is the dispersive component due to the terms, $\mp(\omega_0 - \omega)$.

Summary of Chap. 2

- Using an appropriate pulse, we can treat the optical interaction with matter as two-level system.
- From the Hamiltonian composed by a hydrogenic atom with a dipole moment, we can obtain the three-dimensional equations of motion in terms of Pauli matrices to describe tow-level system.
- By using RWA, we can derive the simple resonant equations relevant to the original equations of two-level system.