1 Basic equations

The basic equations describe the rotation of a pseudo-polarization vector $\vec{\mathcal{P}}$ rotating around the pseudo-electric vector $\vec{\mathcal{E}}$ with an angular velocity given by the amplitude of the vector $\vec{\mathcal{E}}$ [Fig. 1(a)]. The vectorial form for the interaction equations is:

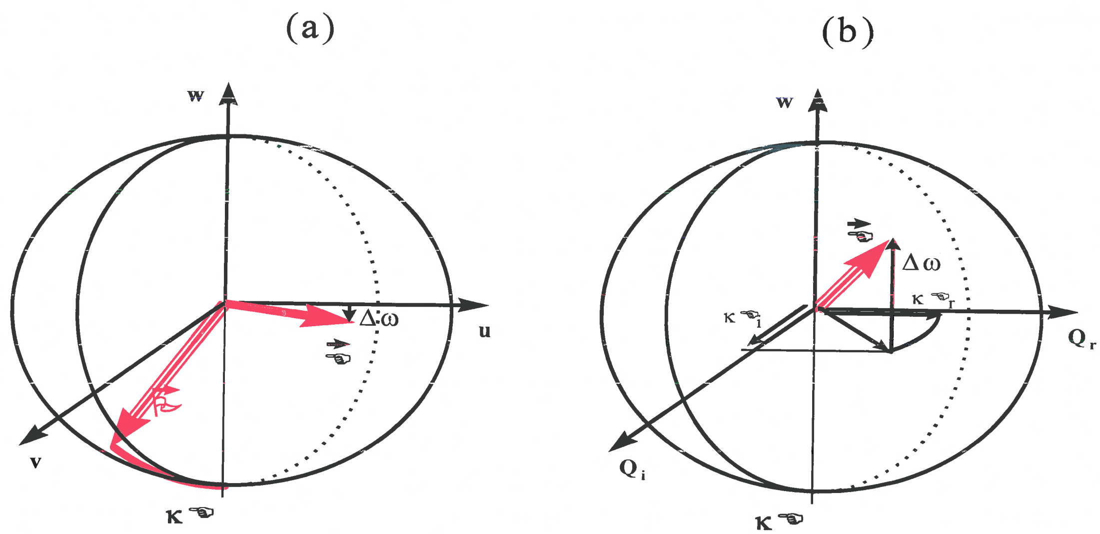

Figure 1: Vector model for Bloch's equations. (a) The motion of the pseudopolarization vector $\vec{\mathcal{P}}$ (initially pointing downwards along the w axis) is a rotation around the pseudo-electric field vector $\vec{\mathcal{E}}$ with an angular velocity proportional to the amplitude of that vector. (b) In the complex amplitude representation, the phase of the electric field determines the particular vertical plane containing the pseudo-electric field vector $\vec{\tilde{\mathcal{E}}}$.

$$\partial\vec{\mathcal{P}}/\partial t = \vec{\mathcal{E}} \times \vec{\mathcal{P}} \tag{1}$$

Depending on whether the two-level system is initially in the ground state or inverted, the pseudo-polarization vector is initially pointing down or up.

For the subensemble of two-level systems having a resonance frequency ω_0, the system of equations for the three components of vector $\vec{\mathcal{P}}$ is[1]:

$$\dot{u} = (\omega_0 - \omega_\ell - \dot{\varphi})v - \frac{u}{T_2} \tag{2}$$

$$\dot{v} = -(\omega_0 - \omega_\ell - \dot{\varphi})u - \kappa\mathcal{E}w - \frac{v}{T_2} \tag{3}$$

$$\dot{w} = \kappa\mathcal{E}v - \frac{w - w_0}{T_1} \tag{4}$$

[1]These equations are the electric-dipole analogues of equations derived by F. Bloch [1] to describe spin precession in magnetic resonance, and are therefore called Bloch's equations.

where the initial value for w at $t = -\infty$ is

$$w_0 = p\bar{N}(\rho_{11}^{(e)} - \rho_{00}^{(e)}). \tag{5}$$

The propagation equation Eq. (12), in terms of $\tilde{\mathcal{E}}$ and φ, becomes

$$\frac{\partial \mathcal{E}}{\partial z} = -\frac{\mu_0 \omega_\ell c}{2n} \int_0^\infty v(\omega_0')g_{inh}(\omega_0')d\omega_0' \tag{6}$$

$$\frac{\partial \varphi}{\partial z} = -\frac{\mu_0 \omega_\ell c}{2n} \int_0^\infty \frac{u(\omega_0')}{\mathcal{E}} g_{inh}(\omega_0')d\omega_0'. \tag{7}$$

Defining

$$\tilde{Q} = (iu + v)e^{i\varphi}, \tag{8}$$

using the complex electric field $\tilde{\mathcal{E}}$:

$$\tilde{\mathcal{E}} = \mathcal{E}e^{i\varphi} \tag{9}$$

and substituting in the above system, leads to another form of the interaction equation:

$$\dot{\tilde{Q}} = i(\omega_0 - \omega_\ell)\tilde{Q} - \kappa\tilde{\mathcal{E}}w - \frac{\tilde{Q}}{T_2} \tag{10}$$

$$\dot{w} = \frac{\kappa}{2}[\tilde{Q}^*\tilde{\mathcal{E}} + \tilde{Q}\tilde{\mathcal{E}}^*] - \frac{w - w_0}{T_1} \tag{11}$$

$$\frac{\partial \tilde{\mathcal{E}}}{\partial z} = -\frac{\mu_0 \omega_\ell c}{2n} \int_0^\infty \tilde{Q}(\omega_0')g_{inh}(\omega_0' - \omega_{ih})d\omega_0'. \tag{12}$$

The last Eq. 12 clearly shows that the quantity Q is the field due to the induced dipoles, which opposes the applied field from the laser.

The vector representation applies also — with a slightly different twist — to the system of Eqs. (10)–(11). The pseudo-polarization vector is then the vector $\vec{Q}(Q_i, Q_r, w)$ rotating around a pseudo-electric field vector $\vec{\mathcal{E}}(\kappa\tilde{\mathcal{E}}_r, \kappa\tilde{\mathcal{E}}_i, -\Delta\omega)$ [Fig. 1(b)]. Physically, the first two components of the pseudo-polarization vector $\vec{Q}$ represent the dipolar resonant field that opposes the applied external field (and is thus responsible for absorption).

2 No relaxation

If we assume no relaxation, the length of the pseudo-polarization vector is a constant of the motion, and the tip of the vector moves on a sphere. The conservation of length of the pseudo-polarization vector can be verified directly from the set of Bloch's equations. Indeed, the sum of each equation (2), (3) and (4) multiplied by u, v, and w, respectively, yields after integration:

$$u^2 + v^2 + w^2 = w_0^2 \tag{13}$$

which is satisfied for each subensemble of two-level systems. As shown in Fig. 1(a), a resonant excitation ($\Delta\omega = 0$)) will tip the pseudo-polarization vector by an angle $\theta_0 = \int_{-\infty}^{\infty} \kappa\mathcal{E}dt$ in the (v, w) plane. For a sufficiently intense pulsed excitation, it is possible to achieve complete population inversion when $\theta_0 = \pi$. The effect of relaxation (homogeneous broadening) is to shrink the pseudo-polarization vector as it moves around. To take into account inhomogeneous broadening, we have to consider an ensemble of pseudo-polarization vectors, each corresponding to a different detuning $\Delta\omega$.

3 Slow motion

If the vector $\vec{\mathcal{E}}$ evolves slowly (but still faster than any relaxation), the vector $\vec{\mathcal{P}}$ follows vector $\vec{\mathcal{E}}$. This type of dynamics —sketched in Fig. 2 — is referred to as "adiabatic following". It can be used to completely invert a two-level system.

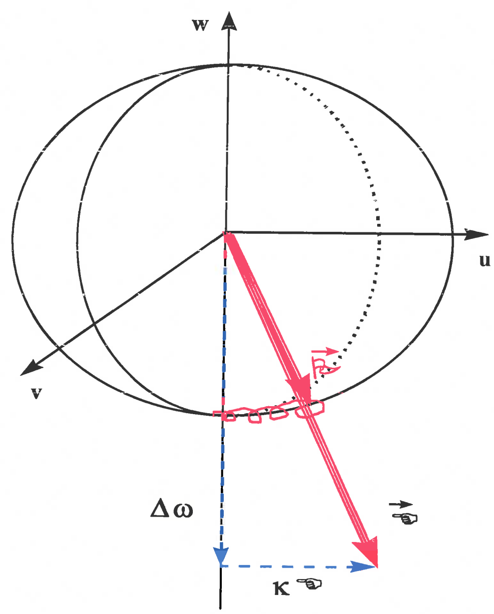

Figure 2: Adiabatic following. In this figure, the pulsed electric field starts from zero amplitude, far off resonance.

4 Steady state

Steady state solutions of the first two Bloch's equation (field variations slow compared to T_2 lead to the rate equation (after insertion of these solutions in the third Bloch equation).

$$\tilde{Q} = \frac{\kappa\tilde{\mathcal{E}}T_2 w}{1 - i\Delta\omega T_2}. \tag{14}$$

or, in terms of u and v:

$$u = -\frac{\Delta\omega T_2 \kappa\mathcal{E}T_2 w}{1 + \delta\omega^2 T_2^2} \tag{15}$$

$$v = -\frac{\kappa\mathcal{E}T_2 w}{1 + \delta\omega^2 T_2^2}. \tag{16}$$

Substituting v in the third Bloch equation (4) leads to the rate equation:

$$\dot{w} = -\frac{\kappa^2\mathcal{E}^2 T_1 T_2}{1 + \delta\omega^2 T_2^2} - \frac{w - w_0}{T_1}. \tag{17}$$

Linear optics is the steady state solution of all three equations.

$$u = -\frac{\Delta\omega T_2 \kappa \mathcal{E} T_2 w}{1 + \delta\omega^2 T_2^2 + \kappa^2 \mathcal{E}^2 T_1 T_2} \tag{18}$$

$$v = -\frac{\kappa \mathcal{E} T_2 w}{1 + \delta\omega^2 T_2^2 \kappa^2 \mathcal{E}^2 T_1 T_2} \tag{19}$$

$$w = \frac{w_0(1 + \delta\omega^2 T_2^2)}{1 + \delta\omega^2 T_2^2 \kappa^2 \mathcal{E}^2 T_1 T_2}. \tag{20}$$

5 Small motions at the bottom of the sphere

Bloch's equations can be solved analytically in the weak short pulse limit, i.e., for pulses that do not induce significant changes in population and have a duration short compared to the phase relaxation time T_2. The interaction equation (10) can be written in the integral form:

$$\tilde{Q}(t) = \int_{-\infty}^{t} \kappa \mathcal{E} w e^{-i[(\omega_0 - \omega_\ell)t' - \varphi(t')]} dt' \tag{21}$$

For weak pulses $w \approx w_0$) and the right hand side of Eq. (21) at $t = \infty$ is proportional to the Fourier transform of $\kappa\tilde{\mathcal{E}}w$. Thus we have:

$$|\tilde{Q}|^2 = u^2 + v^2 = \kappa^2 w_0^2 |\tilde{\mathcal{E}}(\omega_0 - \omega_\ell)|^2 \tag{22}$$

$$\approx -2w_0(w_\infty - w_0) \tag{23}$$

where $\tilde{\mathcal{E}}(\omega_0 - \omega_\ell)$ is the amplitude of the Fourier transform of the field envelope at the line center frequency ω_0. The last equality results from the conservation of the length of the pseudo-polarization vector ($u^2 + v^2 + w^2 = w_0^2 =$ constant). The approximation is made that the change in population is small: $w_\infty^2 = [w_0 + (w_\infty - w_0)]^2 \approx w_0^2 + 2w_0(w_\infty - w_0)$. The final expression is:

$$(w_\infty - w_0) = -\frac{\kappa^2 w_0}{2} |\tilde{\mathcal{E}}(\omega_0 - \omega_\ell)|^2. \tag{24}$$

This is a close connection to linear optics. Equation (22) tells us that the amplitude of the dipolar field that opposes the applied field is proportional to the Fourier component of the applied field at the dipole resonant frequency. The form of Eq. (24) is of equal physical importance, since it relates the energy absorbed by the two-level system to the spectral intensity of the light at the resonance frequency. The approximations made to arrive to this conclusion are more general than the steady-state approximations of the previous section.

6 Useful references

The most cited paper The original paper of the vector model: ref: [2]

Free Induction Decay Its use for short pulse generation [3, 4]. Stark cell spectroscopy for measuring dipole moments [5, 6]. Heterodyne detection is made, between the field $\tilde{Q}$, Stark shifted by an amount δ, and the applied field $\tilde{\mathcal{E}}$, which has a constant amplitude. The detected intensity is proportional to:

$$\left|\tilde{Q}e^{i\delta} + \tilde{\mathcal{E}}\right|^2 = |Q|^2 + |\mathcal{E}|^2 + Q\mathcal{E}\cos\delta t. \tag{25}$$

Free induction decay leads even to a complex rotational level spectroscopy [7, 8].

Stark cell transient spectroscopy [9]

Photon Echoes The whole field of coherent interactions startedby transposing to optics all known experiment of magnetic resonance. Photon echoes is the optical analog of "Spin Echoes". The original reference for spin echoes is a Physical Review paper by E. L. Hahn [10]. Not surprisingly, it was one of his students that transposed this technique to optics [11]. C. V. Heer is another relevant name in this field [12]. Implementation with white light is from Kobayashi [13, 14]. There has been a renewal of interest in photon echoes for the study of semiconductors.

Adiabatic Following The basic original work is by Gryschkowsky [15] Some interesting ramifications: self-defocusing of light [16] and self-adiabatic following for two-photon transitions [17].

Coherent effect in semiconductors Oscillation of a two-level systen between ground and excited states in has been studied first in a very slow time scale with magnetic resonance experiments. In optics, most experiments involved either cooled solid state systems — such as ruby at liquid He temperature — for which the dephasing time is in the nanoseconds. Vapors also provide a medium where the dephasing time can be adjusted from the nanosecond to the picosecond through the pressure. The real challenge concerns condensed matter, such as semiconductors, where the dephasing time can be of a few femtoseconds. What does not make matters easier in semiconductors, is that the dephasing time becomes even shorter at high excitation densities. With the increasing sophistication of ultrafast diagnostic methods, coherent effects in semiconductors has become a new fashion [18, 19].

7 Propagation

7.1 Propagation equations

Maxwell's propagation equations, within the slowly varying approximation, reduce to:

$$\frac{\partial\tilde{\mathcal{E}}}{\partial z} = -\frac{\mu_0\omega_\ell c}{2n}\int_0^\infty \tilde{Q}(\omega_0)g_{inh}(\omega_0 - \omega_{ih})d\omega_0, \tag{26}$$

where the subscript ℓ has been put on the light frequency ω_ℓ to prevent any confusion with another frequency. The integration is performed on all two-level systems of transition frequency

ω_0, distributed within the inhomogeneous line profile g_{inh} centered at ω_{ih}. This form of the propagation equation clearly identifies the quantity $\tilde{Q}$ as the dipolar field opposing the applied field $\mathcal{E}$.

In terms of the notations u, v and w, the propagation equations are:

$$\frac{\partial \mathcal{E}}{\partial z} = -\frac{\mu_0 \omega_\ell c}{2n} \int_0^\infty v(\omega_0') g_{inh}(\omega_0') d\omega_0' \tag{27}$$

$$\frac{\partial \varphi}{\partial z} = -\frac{\mu_0 \omega_\ell c}{2n} \int_0^\infty \frac{u(\omega_0')}{\mathcal{E}} g_{inh}(\omega_0') d\omega_0'. \tag{28}$$

7.2 Evolution laws

A pulse can be characterised by its energy W, its area $\theta = \int \kappa \mathcal{E} dt$, and its frequency $\omega + <\dot{\varphi}>$. The total energy in the resonant light–matter system should be conserved if the pulses are shorter than the energy relaxation time T_1, since no energy is dissipated into the bath. The pulse energy density is defined by:

$$W = \frac{1}{2}\epsilon_0 cn \int_{-\infty}^{\infty} \mathcal{E}^2 dt = \frac{1}{2}\sqrt{\epsilon\epsilon_0/\mu_0} \int_{-\infty}^{\infty} \mathcal{E}^2 dt. \tag{29}$$

A simple energy conservation law can be derived by integrating Eq. (27) over time, after multiplying both sides by $\mathcal{E}$ and using the third Bloch equation (4):

$$\begin{aligned}\frac{dW}{dz} &= \sqrt{\frac{\epsilon\epsilon_0}{\mu_0}} \int_{-\infty}^{\infty} \mathcal{E}\frac{\partial \mathcal{E}}{\partial z} dt \\ &= -\frac{\mu_0 \omega_\ell c}{2}\sqrt{\frac{\epsilon\epsilon_0}{\mu_0}} \int_{-\infty}^{\infty} \int_0^\infty v(\omega_0')\mathcal{E} g_{inh}(\omega_0' - \omega_{ih}) dt d\omega_0' \\ &= -\frac{\hbar\omega_\ell}{2p} \int_0^\infty \{[w_\infty(\omega_0') - w_0(\omega_0')]\} g_{inh}(\omega_0' - \omega_{ih}) d\omega_0' \end{aligned} \tag{30}$$

The population difference (per unit volume) $(w_\infty - w_0)/p$ integrated over the inhomogeneous transition is a measure of the energy stored in the medium, as a consequence of the energy lost by the pulse, dW/dz.

For inhomogeneously broadened media an "area theorem" can be derived which tells us exactly how the pulse area evolves with propagation distance. With the assumptions that the pulses are at resonance ($\omega_\ell = \omega_{ih}$), and shorter than both the energy relaxation time T_1 and the phase relaxation time T_2, a time integration of Eq. (27), taking into account Bloch's equations (2) through (4), yields the *area theorem* [20]:

$$\frac{d\theta_0}{dz} = \frac{\alpha_0}{2}\sin\theta_0. \tag{31}$$

where

$$\alpha_0 = \frac{\pi\mu_0\omega_\ell cp}{\hbar n} w_0(\omega_{ih}) = \frac{\pi\kappa^2\hbar\omega_\ell}{\epsilon_0 cnp} w_0(\omega_{ih}) \tag{32}$$

is the linear absorption coefficient (at resonance) for the inhomogeneously broadened transition. $w_0(\omega_{ih})$ is the initial inversion density at the transition center ($\omega_0' = \omega_{ih}$).

By combination and partial integration of Bloch's equations, one can derive an evolution equation for the average pulse frequency:

$$\frac{d\langle\dot{\varphi}\rangle}{dz} = \frac{\omega_\ell}{2\kappa W}\int_0^\infty g_{inh}(\omega_0' - \omega_{ih})\left[\omega_0' - \omega_\ell - \langle\dot{\varphi}\rangle\right](w_\infty - w_0)\,d\omega_0' + \frac{2\langle k\rangle}{T_2}. \tag{33}$$

In analogy to the definition of the average frequency, we have introduced the average contribution to the propagation vector due to the resonant dispersion of the two-level system:

$$\begin{aligned}\langle k\rangle &= \frac{\int_{-\infty}^{\infty}\mathcal{E}^2(\partial\varphi/\partial z)dt}{\int_{-\infty}^{\infty}\mathcal{E}^2\,dt}\\ &= \frac{\omega_\ell}{4W}\int_0^\infty d\omega_0'\int_{-\infty}^{\infty}dt\; g_{inh}(\omega_0' - \omega_{ih})u\mathcal{E}\end{aligned} \tag{34}$$

The polarization amplitude u — and hence the resonant contribution to the wave vector $\langle k\rangle$ — will shrink with time in presence of phase relaxation (finite T_2). The corresponding temporal modulation of the polarization is responsible for the second term of the right-hand side of Eq. (33). For very short pulses, however, ($\tau_p \ll T_2$), this second term can be neglected.

The frequency shift is proportional to the overlap integral of the lineshape $g_{inh}(\omega_0 - \omega_{ih})$ with the (frequency dependent) change of the inversion $(w_\infty - w_0)$ times the detuning $(\omega_0 - \omega_\ell - \langle\dot{\varphi}\rangle)$. The ratio of absorbed energy (which is proportional to $(w_\infty - w_0)$) to the pulse energy W is maximum in the weak pulse limit $(\theta \ll 1)$. Therefore, the frequency pushing as described by Eq. (33) is important in the weak pulse limit, and for narrow lines $[T_2 \to \infty; g_{inh}(\omega_0 - \omega_{ih}) \approx \delta(0)]$.

8 Density matrix derivation of Bloch's equations

We start with the equation of motion for the density matrix:

$$i\hbar\dot{\rho} = [H, \rho] = [H + H', \rho] \,, \tag{35}$$

where the perturbation of the Hamiltonian of the atom H is $H' = -p.E$, where p is the dipole moment and E the electric field. In matrix form:

$$\begin{aligned} i\hbar \begin{pmatrix} \dot{\rho}_{aa} & \dot{\rho}_{ab} \\ \dot{\rho}_{ba} & \dot{\rho}_{bb} \end{pmatrix} &= \begin{pmatrix} H_{aa} & H' \\ H'^* & H_{bb} \end{pmatrix} \cdot \begin{pmatrix} \rho_{aa} & \rho_{ab} \\ \rho_{ba} & \rho_{bb} \end{pmatrix} - \begin{pmatrix} \rho_{aa} & \rho_{ab} \\ \rho_{ba} & \rho_{bb} \end{pmatrix} \cdot \begin{pmatrix} H_{aa} & H' \\ H'^* & H_{bb} \end{pmatrix} \\ &= \begin{pmatrix} H_{aa}\rho_{aa} + H'\rho_{ba} & H_{aa}\rho_{ab} + H'\rho_{bb} \\ H_{bb}\rho_{ba} + H'^*\rho_{aa} & H_{bb}\rho_{bb} + H'^*\rho_{ab} \end{pmatrix} - \begin{pmatrix} H_{aa}\rho_{aa} + H'^*\rho_{ab} & H_{bb}\rho_{ab} + H'\rho_{aa} \\ H_{aa}\rho_{ba} + H'^*\rho_{bb} & H_{bb}\rho_{bb} + H'\rho_{ba} \end{pmatrix} \end{aligned} \tag{36}$$

where, to "lighten" the notations, we have written H' for H_{ab} and H'^* for H_{ba}. For the diagonal elements, we have:

$$\begin{aligned} i\hbar\dot{\rho}_{aa} &= H'\rho_{ba} - H'^*\rho_{ab} \\ i\hbar\dot{\rho}_{bb} &= H'^*\rho_{ab} - H'\rho_{ba} \\ i\hbar\left(\dot{\rho}_{aa} - \dot{\rho}_{bb}\right) &= 2\left(H'\rho_{ba} - H'^*\rho ab\right) = 2\left(pE^*\rho_{ab} - pE\rho_{ab}^*\right) , \end{aligned} \tag{37}$$

which leads to:

$$\dot{\rho}_{bb} - \dot{\rho}_{aa} = \frac{2p}{\hbar}\left(iE^*\rho_{ab} - iE\rho_{ab}^*\right) . \tag{38}$$

For the off diagonal element of the first row:

$$i\hbar\dot{\rho}_{ab} = \hbar(\omega_a - \omega_b)\rho_{ab} + H'(\rho_{bb} - \rho_{aa}). \tag{39}$$

Substituting the dipolar interaction:

$$\dot{\rho}_{ab} = i\omega_0\rho_{ab} + \frac{ip}{\hbar}E\left(\rho_{bb} - \rho_{aa}\right) . \tag{40}$$

Next, we introduce the complex field envelope $\tilde{\mathcal{E}}$:

$$E = \frac{1}{2}\tilde{\mathcal{E}}e^{i\omega t}, \tag{41}$$

and the complex dipolar field envelope $\tilde{Q}$:

$$ipN\rho_{ab} = \frac{1}{2}\tilde{Q}e^{i\omega t}. \tag{42}$$

The "normalized" population difference is defined as:

$$w = pN\left(\rho_{bb} - \rho_{aa}\right) . \tag{43}$$

Substitution in the diagonal density matrix equation Eq. (38) leads to:

$$\dot{w} = \frac{p}{2\hbar}\left(\tilde{\mathcal{E}}^*\tilde{Q} + \tilde{\mathcal{E}}\tilde{Q}^*\right) = \kappa Re\{\tilde{\mathcal{E}}^*\tilde{Q}\} = \kappa\mathcal{E}v, \tag{44}$$

where $\kappa = p/\hbar$ is the quantity that transforms the electric field amplitude unit into a frequency (the Rabi frequency). Multiplying the equation for the off-diagonal element Eq. (40) by ipN, we get the evolution equation for the (comples) dipolar field that opposes the applied field:

$$\dot{\tilde{Q}} = i(\omega_0 - \omega)\tilde{Q} - \kappa\tilde{\mathcal{E}}w. \tag{45}$$

8.1 Theory of cascade excitation

8.1.1 General formalism

Multiphoton transition are the result the combination of all allowed dipole transitions in the system. Each of the off-resonant dipole transition can be treated adiabatically. Thiss adiabatic approximation does not apply or the resonant multiphoton combination. Whenever there is no intermediate resonance, the problem of multiphoton (n-photon) excitation reduces to a generalization of Bloch's equation, where the driving term is the n^{th} power of the field.

One can also use a multiple wavelength source, each wavelength of the source being resonant with successive dipole transitions. If in addition the sum of the n photon frequencies is resonant with a particular level, we have a case of "cascade n-photon resonance". This problem can be solved formally in all generality from Schrödinger's equations. From the general solution, we can particularize to the case of identical fields, off-resonance intermediate levels, multiphoton resonance. For simplicity, we will limit ourselves here to a three-level system. Procedure is easily generalized to n-levels.

We consider a bichromatic laser pulse described by:

$$\begin{aligned} E(t) &= \mathcal{E}_1(t)\cos[\omega_{\ell,1}t + \varphi_1(t)] \\ &+ \mathcal{E}_2(t)\cos[\omega_{\ell,2}t + \varphi_2(t)] + \ldots . \end{aligned} \tag{46}$$

The relevant three level system is sketched in Fig. 3. The detunings are defined as:

$$\begin{aligned} \Delta_1 &= \omega_{01} - \omega_{\ell,1} \\ \Delta_2 &= \omega_{02} - (\omega_{\ell,1} + \omega_{\ell,2}) \end{aligned} \tag{47}$$

The coupling with the multilevel (three) system is through the dipole interaction term in the time dependent Schrödinger equation:

$$H\psi = i\hbar\frac{\partial\psi}{\partial t}, \tag{48}$$

with:

$$H = H_0 + H' = H_0 - p \cdot E(t) \tag{49}$$

where p is the dipole moment. The wave function ψ is written as a linear combination of the wave function of the unperturbed atomic system ψ_k:

$$\psi(t) = \sum_k a_k(t)\psi_k \tag{50}$$

which leads to a system of differential equations for the coefficients $a_k(t)$:

$$\frac{da_k}{dt} = -i\omega_k a_k + \sum_j \frac{i}{2\hbar} p_{k,j}[\tilde{\mathcal{E}}_1 e^{i\omega_{\ell,1}t} + \tilde{\mathcal{E}}_2 e^{i\omega_{\ell,2}t} + c.c.]a_j \tag{51}$$

The "rotating frame" approximation for this particular situation is:

$$\begin{aligned} a_0 &= c_0 \\ a_1 &= e^{-i\omega_{\ell,1}t}\, c_1 \\ a_2 &= e^{-i(\omega_{\ell,1}+\omega_{\ell,2})t}\, c_2 \end{aligned} \tag{52}$$

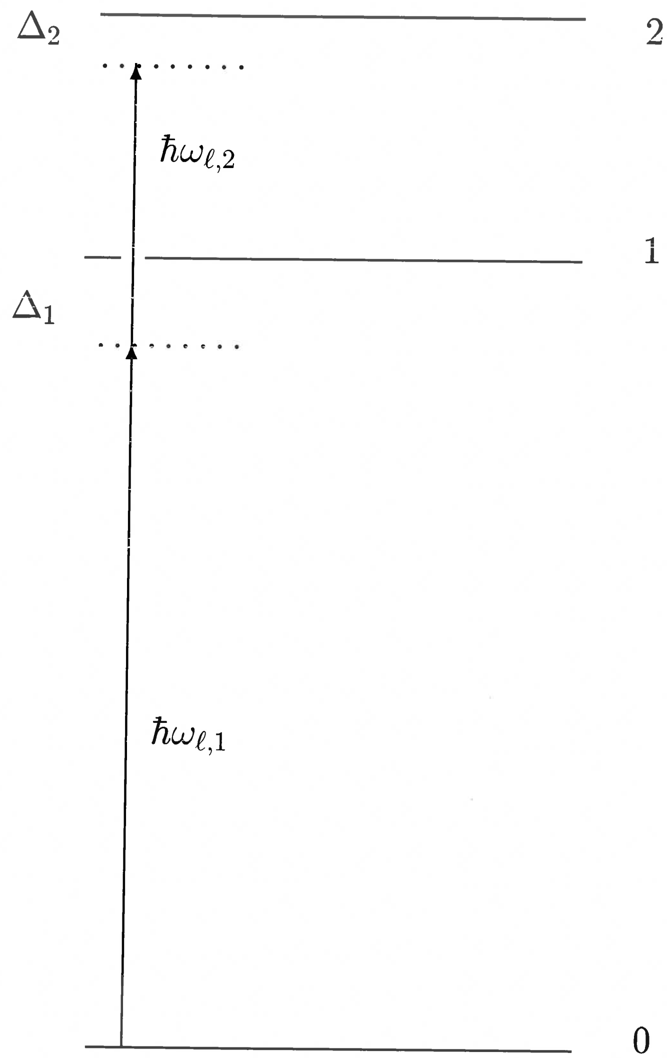

Figure 3: Energy levels and detunings

Substituting in Eqs. (51) leads to:

$$\begin{aligned}\frac{dc_0}{dt} &= \frac{i}{2\hbar}p_{1,0}\tilde{\mathcal{E}_1}(t)\ c_1 \\ \frac{dc_1}{dt} &= -i\Delta_1 c_1 + \frac{i}{2\hbar}p_{0,1}\tilde{\mathcal{E}_1^*}(t)c_0 + \frac{i}{2\hbar}p_{2,1}\tilde{\mathcal{E}_2}(t)c_2 \\ \frac{dc_2}{dt} &= -i\Delta_2 c_2 + \frac{i}{2\hbar}p_{1,2}\tilde{\mathcal{E}_2^*}(t)c_1 \end{aligned} \tag{53}$$

This systems takes a simpler form is we define the Rabi frequencies as:

$$\begin{aligned}\tilde{E_1} &= \frac{i}{\hbar}p_{1,0}\tilde{\mathcal{E}_1} \\ \tilde{E_2} &= \frac{i}{\hbar}p_{2,1}\tilde{\mathcal{E}_2}. \end{aligned} \tag{54}$$

Substituting:

$$\frac{d}{dt}\begin{pmatrix} c_0 \\ c_1 \\ c_2 \end{pmatrix} = \begin{pmatrix} 0 & \frac{1}{2}\tilde{E_1} & 0 \\ -\frac{1}{2}\tilde{E_1^*} & -i\Delta_1 & \frac{1}{2}\tilde{E_2} \\ 0 & -\frac{1}{2}\tilde{E_2^*} & -i\Delta_2 \end{pmatrix} \cdot \begin{pmatrix} c_0 \\ c_1 \\ c_2 \end{pmatrix} \tag{55}$$

8.1.2 Adiabatic approximation

We next consider a three level system where the detuning of the intermediate level 1 is much larger than the transition rates. We can then use a stationary phase approximation to solve the second Eq. (55):

$$c_1 = \frac{i}{2\Delta_1}\left(E_1^* c_0 - E_2 c_2\right). \tag{56}$$

Substituting in the other two equations:

$$\begin{aligned}\dot{c}_0 &= \frac{i}{4\Delta_1}\left(E_1^* c_0 - E_2 c_2\right) \\ \dot{c}_2 &= -\frac{i}{4\Delta_1}E_1^* E_2^* c_0 + \frac{i}{4\Delta_1}E_2 E_2^* c_2 - i\Delta_2 c_2 \end{aligned} \tag{57}$$

Defining:

$$\begin{aligned}\tilde{Q}_2 &= -ic_0c_2^* \\ W_2 &= c_2c_2^* - c_0c_0^* \end{aligned} \tag{58}$$

leads to the following set of equations:

$$\begin{aligned}\dot{\tilde{Q}}_2 &= i\left\{\Delta_2 + \frac{1}{4\Delta_1}\left[|E_1|^2 - |E_2|^2]\right]\right\}\tilde{Q}_2 - \frac{E_1E_2}{2\Delta_1 W_2} \\ \dot{W}_2 &= \frac{1}{2\Delta_1}Re\left[E_1E_2\tilde{Q}^*\right]. \end{aligned} \tag{59}$$

This set of equations is reminiscent of Bloch's equation for a single photon two-level system. Let us assume that there is only one optical frequency ($\omega_1 = \omega_2 = \omega$). The two photon analogue of the Rabi frequency $\kappa\mathcal{E}$ is a two photon Rabi frequency $\kappa_2\mathcal{E}^2$, where:

$$\kappa_2 = \frac{\kappa_1\kappa_2}{2\Delta_1} = \frac{p_{01}p_{12}}{\hbar^2\Delta_1}. \tag{60}$$

Note a small complexity appearing in the detuning: a time dependent detuning $\Delta\omega_2(t)$ has to be substituted to the constant detuning Δ_2:

$$\Delta\omega_2(t) = \Delta_2 + \frac{1}{4\Delta_1}\left[|E_1|^2 - |E_2|^2\right]. \tag{61}$$

This is the dynamic Stark shift associated with a two-photon resonance. It leads to interesting phenomena such as "self-induced adiabatic rapic passage", or a convenient way to completely invert a two-photon resonance [17].

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