away from resonance, under the influence of an optical oscillating field at ω, the motion of the electron follows the frequency of the applied field, in phase, and is thus $d = d_0 \cos \omega t$. At a point of observation at a distance R from the dipole, the field due to the dipole is:

$$\Delta E = \frac{q^2}{4\pi R^2}\left[1 - \frac{R^2}{(R+d)^2}\right] \approx \frac{2q^2 d}{4\pi R^3} \tag{3.27}$$

Putting that in Maxwell's propagation equation:

$$\frac{\partial^2 E}{\partial z^2} - \frac{1}{c^2}\frac{\partial^2 (E + \Delta E)}{\partial t^2} = 0 \tag{3.28}$$

or

$$\frac{\partial^2 E}{\partial z^2} - \frac{1}{c^2}\frac{\partial^2 E}{\partial t^2} = \frac{1}{c^2}\frac{\partial^2 \Delta E}{\partial t^2} = \frac{\omega^2}{c^2}\Delta E. \tag{3.29}$$

Using:

$$\begin{aligned} E &= \frac{1}{2}\mathcal{E} e^{i(\omega t - kz)} \\ \Delta E &= \frac{1}{2}\Delta\mathcal{E} e^{i(\omega t - kz)} \end{aligned}$$

we find:

$$-2ik\frac{\partial \mathcal{E}}{\partial z} - 2i\frac{\omega}{c^2}\frac{\partial \mathcal{E}}{\partial t} = \frac{\omega^2}{c^2}\Delta\mathcal{E}, \tag{3.30}$$

and

$$\frac{\partial \mathcal{E}}{\partial z} + \frac{1}{c}\frac{\partial \mathcal{E}}{\partial t} = -i\frac{\omega}{2c}\Delta\mathcal{E}, \tag{3.31}$$

Even though we started from a $\Delta\mathcal{E}$ in phase with the applied field, after insertion in the propagation equation it appears that its envelope is adding 90 degrees out of phase with the applied field, as is the case of an index of refraction.

It appears as if, by the time the electron re-radiates, the wave has already moved by a distance of $\lambda/4$.

3.3 Semi-classical approach to light matter interaction

In a semi-classical approach, the field is treated classically, and the atom quantum mechanically. The basic physics is essentially the same as discussed in the previous classical section: the electromagnetic field of light excites the motion of electrons bound to the atom. The electrons being bound to the atom/molecule, the

re-radiation into the field is that of the dipole term in Eq. (3.10). In that dipole radiation term, the only varying parameter is the distance d between charges, which is found by solving the time dependent Schrödinger equation for the atomic system,

$$H\psi = i\hbar\frac{\partial\psi}{\partial t}, \tag{3.32}$$

where the Hamiltonian H is the atomic system Hamiltonian H_0 perturbed by the dipolar term:

$$H = H_0 + (qd)E. \tag{3.33}$$

The atomic system is characterized by a set of energy levels $\hbar\omega_k$, eigenvalues of the equation:

$$H_0\psi_k = \hbar\omega_k\psi_k. \tag{3.34}$$

The wave function solution ψ is found by inserting in Eq. (3.32) the expansion:

$$\psi(t) = \sum a_k(t)\psi_k, \tag{3.35}$$

and solving for the coefficients $a_k(t)$. The reaction field per electron $(qd(t)/\epsilon_0)$ that radiates back into the applied field is calculated by taking the expectation value of the position r:

$$\Delta E = \langle\psi|qr|\psi\rangle/\epsilon_0. \tag{3.36}$$

The polarization $P = \epsilon_0\Delta E$ is in general defined by a differential equation, where the driving term is the total electric field applied to the atomic system, which can have components at different frequencies. The initial conditions are given by the state of the system *prior* to the application of the field. In practical situation, the total field may be given by a combination of m pulses a various frequencies:

$$E(t) = \frac{1}{2}\sum_{j=1}^{j=m}\left\{\tilde{\mathcal{E}}_j(t)e^{i\omega_{\ell,j}-\vec{k}_j\cdot\vec{r}}\right\} \tag{3.37}$$

where some of the fields $\tilde{\mathcal{E}}_j$ may be generated from the time dependent polarization. One should not forget that the interaction will always have a particular aim, which is either to create a particular state of matter characterized by the wave function ψ [or equivalently the set of time dependent coefficients $a_k(t)$], or create a particular reaction field $\Delta E(t)$ or polarization $P(t)$. In the latter case, one will want to compress or modulate the applied field, or create new frequency components. One will generally seek a particular combination of atomic system (levels) and fields depending on the goal that one seeks to achieve, most often seeking near resonance or proximity of certain transition frequencies between levels and light frequencies.

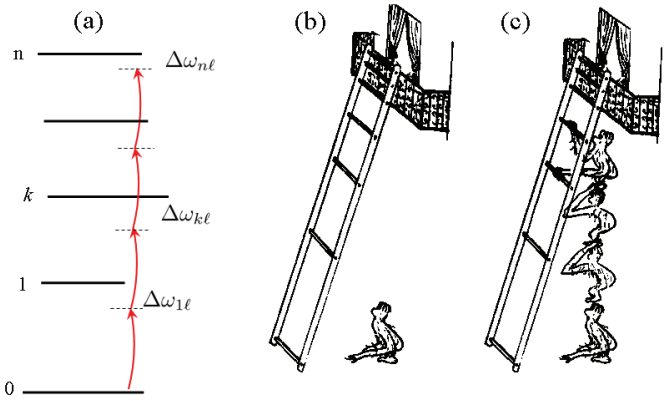

Figure 3.3: (a) The main approximation in this approach is to consider the interaction of the light only with n levels connected by a combination of photons of frequency $\omega_{\ell,j}$. The important parameters are the dipole moments of the transitions, and the detunings $\Delta\omega_{k,\ell} = \omega_{k,\ell} - \sum_{j=1}^{j=k} \omega_{\ell,j}$. (b) A typical problem will be to create an inversion with a stack of unequally spaced levels, or for a "Romeo" to reach the balcony of his "Juliet" with a lousy ladder. The solution (c) is the multiphoton approach.

We will first investigate the situation of *cascade transitions*, applicable when each photon of frequency $\omega_{\ell,k}$ finds a near resonance with a pair of levels, as sketched in Fig. 3.3(a). An example of related physical situation is to excite a stack of rotational levels. These levels are in general an anharmonic ladder. A single pulse excitation may only reach to the first step, as illustrated by the "Romeo" of Fig. 3.3(b) trying to reach his "Juliet" with an anharmonic ladder. The smart approach that can be taken is to create a properly timed and phased sequence of pulses or "Romeos" to reach the top of the ladder, as in Fig. 3.3(c). Rotational level inversion can be engineered with ultrashort IR pulses [12]. They are also taking place in the propagation of ultrashort intense pulses in air [13]. Cascade excitation can be exploited to create a complete population inversion in atomic vapors [14].

Situations can be created where most detuning — except one — can be neglected. The interaction with the off-resonant levels can be considered nearly instantaneous: the response time is of the order of the inverse of the detuning. One can find a stationary ("adiabatic") solution for the coefficients a_k associated with the off-resonant levels. The interaction reduces to a set of differential equations involving the near resonant levels, which, for times sufficiently short that relaxation effects (radiative and non-radiative decays, collisional relaxation) are negligible, can often be represented by a "Bloch vector" model. We will see under which condition these equations reduce first to rate equations, next to the classical non-

resonant instantaneous linear and nonlinear polarization.

Semi-classical approach applied to cascade excitation of multilevel systems 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.P For simplicity, we will limit ourselves here to a three-level system. The procedure followed here is easily generalized to n-levels.

We consider a bichromatic laser pulse described by:

$$\begin{aligned} E(t) &= \mathcal{E}_1(t)\cos[\omega_{\ell,1}t+\varphi_1(t)] \\ &+ \mathcal{E}_2(t)\cos[\omega_{\ell,2}t+\varphi_2(t)]+\ldots. \end{aligned} \tag{3.38}$$

Note that we are not using at this point the complex notations. As it is often the case in nonlinear optics, one has to be careful to include both the positive and negative frequencies at the onset.

The relevant three level system is sketched in Fig. 3.4. The detunings are defined as:

$$\begin{aligned} \Delta_1 &= \omega_{01}-\omega_{\ell,1} \\ \Delta_2 &= \omega_{02}-(\omega_{\ell,1}+\omega_{\ell,2}) \end{aligned} \tag{3.39}$$

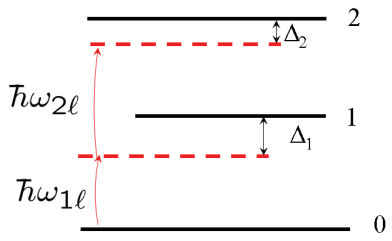

Figure 3.4: Sketch showing the three levels 0, 1 and 2, the light frequencies $\hbar\omega_{1\ell}$ and $\hbar\omega_{2\ell}$ and the detunings.

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

$$H\psi = i\hbar\frac{\partial\psi}{\partial t}, \tag{3.40}$$

with:

$$H = H_0 + H' = H_0 - p \cdot E(t) \tag{3.41}$$

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:

$$\psi(t) = \sum_k a_k(t)\psi_k \tag{3.42}$$

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

$$\frac{da_k}{dt} = -i\omega_k a_k + \sum_j \frac{i}{2\hbar} p_{k,j}[\tilde{\mathcal{E}}_1 e^{i\omega_{\ell,1}t} + \tilde{\mathcal{E}}_2 e^{i\omega_{\ell,2}t} + c.c.]a_j \tag{3.43}$$

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

$$\begin{aligned} a_0 &= c_0 \\ a_1 &= e^{-i\omega_{\ell,1}t}\, c_1 \\ a_2 &= e^{-i(\omega_{\ell,1}+\omega_{\ell,2})t}\, c_2 \end{aligned} \tag{3.44}$$

In substituting in Eqs. (3.43), it is important to keep only the slowly varying terms (as compared to the light frequency or transition frequency). This is the step where we see the importance of having defined the field as a real quantity, i.e. with both positive and negative frequencies. The positive field frequencies combine with negative going frequencies, and vice versa, to give:

$$\begin{aligned} \frac{dc_0}{dt} &= \frac{i}{2\hbar} p_{1,0}\tilde{\mathcal{E}}_1(t)\, c_1 \\ \frac{dc_1}{dt} &= -i\Delta_1 c_1 + \frac{i}{2\hbar} p_{0,1}\tilde{\mathcal{E}}_1^*(t)c_0 + \frac{i}{2\hbar} p_{2,1}\tilde{\mathcal{E}}_2(t)c_2 \\ \frac{dc_2}{dt} &= -i\Delta_2 c_2 + \frac{i}{2\hbar} p_{1,2}\tilde{\mathcal{E}}_2^*(t)c_1 \end{aligned} \tag{3.45}$$

or in general, applicable to a n-level system:

$$\frac{dc_k}{dt} = -i\Delta_k c_k + \frac{i}{2\hbar} p_{k-1,k}\tilde{\mathcal{E}}^*(t)c_{k-1} + \frac{i}{2\hbar} p_{k,k+1}\tilde{\mathcal{E}}(t)c_{k+1}. \tag{3.46}$$

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

$$\begin{aligned} \tilde{E}_1 &= \frac{i}{\hbar} p_{1,0}\tilde{\mathcal{E}}_1 \\ \tilde{E}_2 &= \frac{i}{\hbar} p_{2,1}\tilde{\mathcal{E}}_2. \end{aligned} \tag{3.47}$$

Substituting:

$$\boxed{\begin{array}{rcl}\dfrac{dc_0}{dt} &=& 0+\dfrac{1}{2}\tilde{E}_1 c_1+0\\ \dfrac{dc_1}{dt} &=& -\dfrac{1}{2}\tilde{E}_1^* c_0 - i\Delta_1 c_1+\dfrac{1}{2}\tilde{E}_2 c_2\\ \dfrac{dc_2}{dt} &=& 0-\dfrac{1}{2}\tilde{E}_2^* c_1 - i\Delta_2 c_2\end{array}}$$

or in matrix form:

$$\boxed{\frac{d}{dt}\begin{pmatrix} c_0\\ c_1\\ c_2\end{pmatrix}=\begin{pmatrix} 0 & \frac{1}{2}\tilde{E}_1 & 0\\ -\frac{1}{2}\tilde{E}_1^* & i\Delta_1 & \frac{1}{2}\tilde{E}_2\\ 0 & \frac{1}{2}\tilde{E}_2^* & -i\Delta_2\end{pmatrix}\begin{pmatrix} c_0\\ c_1\\ c_2\end{pmatrix}} \tag{3.48}$$

This is the basics of the treatment of a cascade of multilevel systems. The extension to a larger number of transitions is straightforward. A basic approximation is that any level k is connected by a dipole transition to a level $k+1$ and $k-1$.

The system of equations (3.48) is easy to solve numerically. One is generally not interested in expressing the results as a matrix of c coefficients, but instead the 3×3 matrix of the density matrix elements $\rho_{ij} = c_i c_j^*$. The diagonal elements $c_i c_i^*$ represent the populations of the level i. The off-diagonal elements $c_i c_j^*$ are a measure of the amplitude excitation at the frequency $\omega_j - \omega_i$, and will be directly connected to the polarization, as we have already seen in the case of the two level system.

This matrix formalism is most useful in reaching a desired population distribution. This approach can be used in systems where the density of levels is such that one can generally find a "ladder" of levels to climb. An example of application is given in reference [12]. It is shown how a properly phased sequence of pulses can create a complete population inversion in the vibro-rotational level structure of CH_3F. It can also be applied to the less crowded level structure of atomic transitions. The example of inverting a two-photon transition in sodium vapor, with the purpose of creating a bichromatic artificial guide-star, is treated in reference [14,15]. In the case of atomic transitions, the more often considered case is to have the intermediate level – or intermediates levels – far off resonance. In that case, the equation for that particular (or these particular) intermediate level(s) can be considered to be steady state, and the system of equation is reduced. This is the "adiabatic approximation", which will be solved in Section 3.3.1.

3.3.1 Adiabatic approximation; multiphoton Bloch model

If the detuning of the intermediate level 1 is larger than the transition rates: the second Eq. (3.48) can be considered to be in steady state, and one can solve for the

coefficient c_1:

$$c_1 = \frac{i}{2\Delta_1}\left(E_1^* c_0 - E_2 c_2\right). \tag{3.49}$$

By substituting this solution in the other two equations, the three level system has been reduced to a two-level system, where the ground and upper state are not connected by a dipole transition. These equation can be represented by a "Bloch vector" model, in which a "pseudo-polarization" vector rotates around a "pseudo-electric-field" vector with an angular velocity given by a Rabi frequency that is now proportional to the square of the electric field amplitude. Substituting the solution (3.49) into the other two equations:

$$\begin{aligned} \dot{c}_0 &= \frac{i}{4\Delta_1}\left(E_1^* c_0 - E_2 c_2\right) \\ \dot{c}_2 &= -\frac{i}{4\Delta_1} E_1^* E_2^* c_0 + \frac{i}{4\Delta_1} E_2 E_2^* c_2 - i\Delta_2 c_2 \end{aligned} \tag{3.50}$$

Defining:

$$\begin{aligned} \tilde{Q}_2 &= -i c_0 c_2^* \\ W_2 &= c_2 c_2^* - c_0 c_0^* \end{aligned} \tag{3.51}$$

leads to the following set of equations:

$$\boxed{\begin{aligned} \dot{\tilde{Q}}_2 &= i\left\{\Delta_2 + \frac{1}{4\Delta_1}\left[|E_1|^2 - |E_2|^2\right]\right\}\tilde{Q}_2 - \frac{E_1 E_2}{2\Delta_1} W_2 \\ \dot{W}_2 &= \frac{1}{2\Delta_1} Re\left[E_1 E_2 \tilde{Q}^*\right]. \end{aligned}} \tag{3.52}$$

We recognize here Bloch's equations for a two-level system [16], if we define a two photon Rabi frequency $\kappa_2 \mathcal{E}^2$, where:

$$\boxed{\kappa_2 = \frac{\kappa_1 \kappa_2}{2\Delta_1} = \frac{p_{01} p_{12}}{\hbar^2 \Delta_1}.} \tag{3.53}$$

In general, more than one intermediate level may be involved in the calculation of the two-photon Rabi frequency. This simply means than instead of the single term in the right hand side of Eq. (3.53), there will be a sum over i, the latter designing the index of an intermediate level with detuning Δ_i [essentially replacing all indices "1" by "i" in Eq. (3.53)].

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:

$$\Delta\omega_2(t) = \Delta_2 + \frac{1}{4\Delta_1}\left[|E_1|^2 - |E_2|^2\right]. \tag{3.54}$$

The substitution leads to the Maxwell'Bloch multiphoton system of equations:

$$\dot{\tilde{Q}}_2 = i\Delta\omega_2(t)\tilde{Q}_2 - \kappa_2\tilde{\mathcal{E}}^2 W - \frac{\tilde{Q}_2}{T_2} \quad (3.55)$$

$$\dot{W} = Re\left[\kappa_2\tilde{\mathcal{E}}^2\tilde{Q}^*\right] - \frac{W - W_0}{T_1} \quad (3.56)$$

$$\frac{\partial\tilde{\mathcal{E}}}{\partial z} = -\alpha_2\tilde{Q}_2\tilde{\mathcal{E}}^* - \frac{\alpha}{2}\tilde{\mathcal{E}} + \eta\alpha_2\tilde{\mathcal{E}}_3\tilde{Q}^* \quad (3.57)$$

$$\frac{\partial\tilde{\mathcal{E}}_3}{\partial z} = -\eta\alpha_2\tilde{Q}_2\tilde{\mathcal{E}} - \frac{\alpha_3}{2}\tilde{\mathcal{E}}_3 \quad (3.58)$$

Q_2 is the amplitude of some atomic "excitation" oscillating at the frequency of the two-photon transition, or 2 ω. It is thus natural to expect that the field opposing the driving field (hence responsible for two-photon transition) is given by the combination $Q_2\tilde{\mathcal{E}}^*$ which would be the amplitude of an oscillation at the frequency $2\omega - \omega = \omega$. A rigorous derivation of the polarization shows that this is indeed the case [17]. An additional contribution to a time varying polarization comes from the fact that the atom has a different polarizability in the ground state versus the upper state. Therefore, as the populations swing up and down under the influence of the field, there will be a modulation of the polarization proportional to the population difference. This has a negative impact on phase matching in third harmonic generation. A fourth equation has been added to the set (3.52), with the combination of $Q_2\tilde{\mathcal{E}}$, expected to oscillate at the frequency $3\omega_\ell$. This is the term responsible for two-photon resonant third harmonic generation discussed in the next section.

The system of Eqs. (3.55) through (3.57) can easily be generalized to multi-photon resonant interaction, where n- rather than 2- photons are near resonance with two atomic levels [18, 19]. In most of the cases, a geometric representation applies, as sketched in Fig. 3.5(a). The n-photon have created a matter excitation at frequency $n\omega$, which is represented by a a three dimensional "pseudo-vector" $\vec{Q}(Q_r, Q_i, W)$, where W is proportional to the population difference between the resonant levels. The geometric interpretation of the Eqs. (3.55,3.56) is that the time evolution of the vector $\vec{Q}$ results from a rotation of the $\vec{Q}$ around a pseudo-vector $\vec{\mathcal{E}}(\mathcal{E}_r.\mathcal{E}_r, \Delta\omega)$ with an angular velocity proportional to $|\vec{\mathcal{E}}|$. The third component of the pseudo-vector $\vec{\mathcal{E}}$ is the detuning between $n\omega_\ell$ and the near resonant level, as modified by an eventual Stark shift.

In the set of Eqs. (3.55) through (3.57), the phase $\varphi(t)$ of the field does not appear explicitly. An equivalent form of equations that is preferred for analytical treatments is obtained by the substitution $Q = (iu + v)\exp(i\varphi)$. The geometrical representation of the interaction is that given in Fig. 3.5(b), where the medium resonance is described by the vector $\vec{\mathcal{P}}(u, v, W)$. Here also the motion of the vector $\vec{\mathcal{P}}$ is a rotation about the pseudo-electric field vector $\vec{\mathcal{E}}(\mathcal{E}^n, 0, \Delta\omega - dot\varphi)$. This

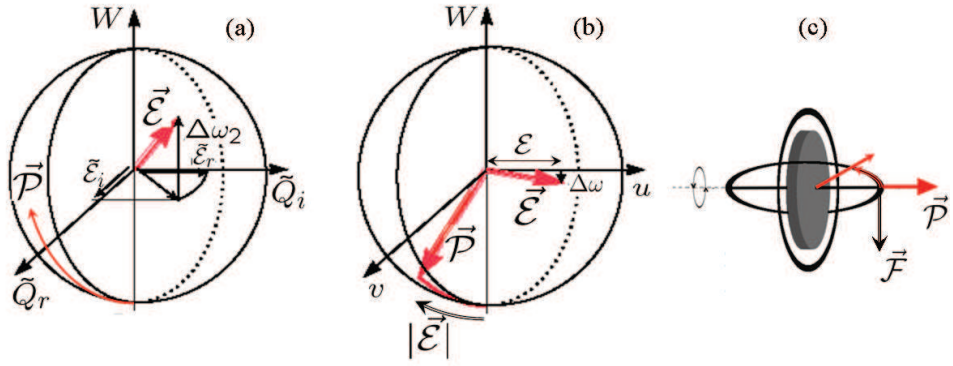

Figure 3.5: Bloch vector model for the near resonant interaction between light and an atomic system. The first two components of $\vec{Q}$ are $-iQ_r + Q_i = \rho_{1n}$ where ρ_{1n} is the single- or multi-photon resonant matrix element between the ground state and the resonant level. In (a), the field is represented by its real and imaginary parts. In (b), the field is represented by its amplitude $\mathcal{E}$ and phase φ, and $Q = (iu + v)\exp(i\varphi)$. The closest analogy to the generalized polarization is not an oscillating spring, but a gyroscope as represented in (c).

particular model will be dealt with in more details when discussing linear optics as a limiting case of coherent interaction (Section 3.3.4). It is seen here that the fundamental mechanical analogy of resonant excitation of matter by a light pulse is not an harmonic force driving a spring near its resonance, but a gyroscope. Indeed, as shown in Fig. 3.5(c), the vector $\vec{\mathcal{P}}$ is represented by the shaft of a gyroscope of angular momentum $I\omega$, If a force $\vec{\mathcal{F}}$ is applied to the cage of the gyroscope, the axis $\vec{\mathcal{P}}$ will precess following an equation of motion $\partial\vec{\mathcal{P}}/\partial t \propto \vec{\mathcal{P}} \times \vec{\mathcal{F}}$.

3.3.2 Optimizing harmonic conversion

Long wavelength lasers being generally more efficient, frequency conversion by harmonic generation is often used to generate shorter wavelength. Gases and atomic vapors have often been used as nonlinear media because of their higher damage threshold as compared to nonlinear crystals. The set of equations presented in the previous section provides a guide to the choice of nonlinear material. If the medium has a two photon resonance, the generation length for a maximum third harmonic generation will be reduced.In the set of Eqs. the function Q has a maximum value, limited by the radius of the Bloch sphere. At the maximum value of Q, the propagation equations appear to be describing just linear gain or linear absorption.

3.3.3 Coherent Raman scattering

It is essentially the same model/equations as in the previous section, with the level 2 "folded down". The notations are $\tilde{\mathcal{E}}_1$ at $\omega_{\ell,1}$ for the Raman pump, $\tilde{\mathcal{E}}_2$ at $\omega_{\ell,2}$ for the Stokes signal; the ground level is "0", the upper level (connected by dipole transition) "1", and the Raman transition is $0 \to 2$.

Stimulated Stokes Backward Raman Scattering

The system of Maxwell-Bloch equations reduces now to:

$$\begin{aligned}
\dot{\tilde{Q}}_r &= i\Delta\omega_2(t)\tilde{Q}_r - \kappa_2\tilde{\mathcal{E}}_1\tilde{\mathcal{E}}_2^* W - \frac{\tilde{Q}_r}{T_2} \\
\dot{W} &= Re\left[\kappa_2\tilde{\mathcal{E}}_1\tilde{\mathcal{E}}_2^*\tilde{Q}_r^*\right] - \frac{W - W_0}{T_1} \\
\frac{\partial\tilde{\mathcal{E}}_2}{\partial z} - \frac{1}{c}\frac{\partial\tilde{\mathcal{E}}_2}{\partial t} &= -\alpha_s\tilde{Q}_r^*\tilde{\mathcal{E}}_1 \\
\frac{\partial\tilde{\mathcal{E}}_1}{\partial z} + \frac{1}{c}\frac{\partial\tilde{\mathcal{E}}_1}{\partial t} &= -\alpha_p\tilde{Q}_r\tilde{\mathcal{E}}_2
\end{aligned} \tag{3.59}$$

The fields, with their complete exponential dependence, are

$$\begin{aligned}
E_1 &= \tilde{\mathcal{E}}_1 e^{i(\omega_{\ell,1}t - k_1 z)} \\
E_2 &= \tilde{\mathcal{E}}_1 e^{i(\omega_{\ell,2}t + k_2 z)}
\end{aligned} \tag{3.60}$$

The Raman excitation has the harmonic dependence:

$$\tilde{Q}e^{i[(\omega_{\ell,1} - \omega_{\ell,2})t - (k_1 + k_2)z]} \tag{3.61}$$

All these exponential dependence are consistent with the above equations.

The coefficients α_s and α_p should contain the respective optical frequencies, in order to satisfy energy conservation.

3.3.4 Single photon coherent propagation

Whether we are dealing with molecular or atomic transitions, the situation can arise where the ultrashort duration of the optical pulse becomes comparable with – or even less than – the phase relaxation time of the excitation. In the frequency domain, the pulse spectrum is broader than the homogeneous linewidth defined in the first section of Chapter 3. If the pulse is so short that its spectrum becomes much larger than the inhomogeneous linewidth, the medium response becomes similar to that of a single atom. It may seem like a simplified situation when the excitation occurs in a time shorter than all inter-atomic interaction. It is in fact quite to the contrary: in dealing with longer pulses, the faster phase relaxation time of the induced excitation simplifies the light matter response. One is used to dealing with a steady state rather than the "transient" response of light-matter interaction.

We will start from the semi-classical equations for the interaction of near resonant radiation with an ensemble of two-level systems inhomogeneously broadened around a frequency ω_{ih}. The extension to multilevel systems will be discussed in the next section. We refer to the book by Allen and Eberly for more detailed developments [20].

In this section we chose a density matrix approach to derive the interaction equations for a near resonant two-level system, of ground state $|0\rangle$ and upper state $|1\rangle$, excited by the field $E(t)$. The density matrix equation for this two-level system is:

$$\dot{\rho} = \frac{1}{i\hbar}\left[H_0 - pE,\, \rho\right] \tag{3.62}$$

where H_0 is the unperturbed Hamiltonian, and p the dipole moment which is parallel to the polarization direction of the field. Introducing the complex field through $E = \tilde{E}^+ + \tilde{E}^-$ in Eq. (3.62) leads to the following differential equations for the diagonal and off-diagonal matrix elements:

$$\dot{\rho}_{11} - \dot{\rho}_{00} = \frac{2p}{\hbar}\left[i\rho_{01}\tilde{E}^- - i\rho_{10}\tilde{E}^+\right] \tag{3.63}$$

$$\dot{\rho}_{01} = i\omega_0\rho_{01} + \frac{ip\tilde{E}^+}{\hbar}\left[\rho_{11} - \rho_{00}\right] \tag{3.64}$$

where ω_0 is the resonance frequency of the two-level system. It is generally convenient to define a complex "pseudo polarization" amplitude $\tilde{Q}$ by

$$i\rho_{01}p\bar{N} = \frac{1}{2}\tilde{Q}\exp(i\omega_\ell t) \tag{3.65}$$

where $\bar{N} = \bar{N}_0 g_{inh}(\omega_0 - \omega_{ih})$ and $\bar{N}_0$ is the total number density of the two-level systems. The real part of $\tilde{Q}$ will describe the attenuation (or amplification for an

initially inverted system) of the electric field. Note that $\tilde{Q} = i\tilde{\mathcal{P}}$ where $\tilde{\mathcal{P}}$ is the slowly varying polarization envelope defined in Fig. 3.5 [1]. Further we introduce a normalized population inversion:

$$w = p\bar{N}(\rho_{11} - \rho_{00}). \tag{3.66}$$

The complete system of interaction and propagation equations can now be written as:

$$\dot{\tilde{Q}} = i(\omega_0 - \omega_\ell)\tilde{Q} - \kappa\tilde{\mathcal{E}}w - \frac{\tilde{Q}}{T_2} \tag{3.67}$$

$$\dot{w} = \frac{\kappa}{2}[\tilde{Q}^*\tilde{\mathcal{E}} + \tilde{Q}\tilde{\mathcal{E}}^*] - \frac{w - w_0}{T_1} \tag{3.68}$$

$$\frac{\partial\tilde{\mathcal{E}}}{\partial z} = -\frac{\mu_0\omega_\ell c}{2n}\int_0^\infty \tilde{Q}(\omega_0')g_{inh}(\omega_0' - \omega_{ih})d\omega_0'. \tag{3.69}$$

The quantity $\kappa\mathcal{E}$ with $\kappa = p/\hbar$ is the Rabi frequency. T_1 and T_2 are respectively the energy and phase relaxation times. Most of the energy conserving relaxations are generally lumped in the phase relaxation time T_2. Equation (3.69) has been obtained by integrating over the polarization of subensembles with resonance frequency ω_0'. The set of Eqs. (3.67)–(3.69) is generally designated as Maxwell–Bloch equations.

Another common set of notations to describe the light-matter interaction uses only real quantities, such as the in-phase (v) and out-of phase (u) components of the pseudo-polarization $\tilde{Q}$, and, for the electric field $\tilde{\mathcal{E}}$, its (real) amplitude $\mathcal{E}$ and its phase φ. Defining

$$\tilde{Q} = (iu + v)e^{i\varphi} \tag{3.70}$$

and substituting in the above system of equations leads to the usual form of Bloch equations[2] for the subensemble of two-level systems having a resonance frequency ω_0.

$$\dot{u} = (\omega_0 - \omega_\ell - \dot{\varphi})v - \frac{u}{T_2} \tag{3.71}$$

$$\dot{v} = -(\omega_0 - \omega_\ell - \dot{\varphi})u - \kappa\mathcal{E}w - \frac{v}{T_2} \tag{3.72}$$

$$\dot{w} = \kappa\mathcal{E}v - \frac{w - w_0}{T_1} \tag{3.73}$$

[1] More details about the polarisation $\tilde{\mathcal{P}}$ in the remainder of this section.

[2] These equations are the electric-dipole analogues of equations derived by F. Bloch [21] to describe spin precession in magnetic resonance.

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

$$w_0 = p\bar{N}(\rho_{11}^{(e)} - \rho_{00}^{(e)}). \tag{3.74}$$

The propagation equation Eq. (3.69), in terms of $\tilde{\mathcal{E}}$ and φ, becomes

$$\boxed{\begin{aligned} \frac{\partial \mathcal{E}}{\partial z} &= -\frac{\mu_0 \omega_\ell c}{2n} \int_0^\infty v(\omega_0') g_{inh}(\omega_0' - \omega_{ih}) d\omega_0' \qquad (3.75) \\ \frac{\partial \varphi}{\partial z} &= -\frac{\mu_0 \omega_\ell c}{2n} \int_0^\infty \frac{u(\omega_0')}{\mathcal{E}} g_{inh}(\omega_0' - \omega_{ih}) d\omega_0'. \qquad (3.76) \end{aligned}}$$

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 $\vec{\mathcal{E}}$.

The vector representation of Feynman *et al.* [16], for the interaction equations is particularly useful in the description of coherent phenomena. The representation is a cinematic representation of the set of equations (3.71), (3.72), and (3.73). For simplicity, we consider first an undamped isolated two-level system ($T_1 = T_2 = T_3 = \infty$), and construct a fictitious vector $\vec{\mathcal{P}}$ of components (u, v, w), and a pseudo-electric field vector $\vec{\mathcal{E}}$ of components $(\kappa\mathcal{E}, 0, -\Delta\omega)$. The detuning is defined as $\Delta\omega = \omega_0 - \omega_\ell - \dot{\varphi}$. The system of Eqs. (3.71)–(3.73) are then the cinematic equations describing 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. 3.5(a)]. The vectorial form of Eqs. (3.71)- (3.73) is thus:

$$\boxed{\partial\vec{\mathcal{P}}/\partial t = \vec{\mathcal{E}} \times \vec{\mathcal{P}}} \tag{3.77}$$

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. Since we have assumed 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 (3.71), (3.72) and (3.73) multiplied by u, v, and w, respectively, yields after integration:

$$u^2 + v^2 + w^2 = w_0^2 \tag{3.78}$$

which is satisfied for each subensemble of two-level systems. As shown in Fig. 3.5(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 phase 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 $\Delta\omega$.

A similar representation can be made for the system of Eqs. (3.67)–(3.68). 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. 3.5(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).

3.4 From transient to stationary interaction

Most classical linear and nonlinear optics, which is treated in the next chapter, treats the linear and nonlinear polarizations as being instantaneous. Therefore, it be understood as a steady-state approximation of the equations covered in the preceding sections.

3.4.1 Rate equations

We have seen how the semiclassical interaction in multilevel systems can be reduced to a two-level system, described by Bloch's equations, if the near resonance of a pair of levels dominates. The next most common situation is when dealing with pulses long compared with the phase relaxation time. If the light field envelope is slowly varying with respect to T_2, Bloch's equations reduce to the standard rate equations. For pulses longer than the dephasing time T_2, the two first Bloch equations (3.71), (3.72) are stationary on the time scale of the pulse. Solving these equations for u, v, and substituting v into the third equation (3.73) for the population difference, leads to the rate equation:

$$\dot{w} = -\frac{\mathcal{E}^2(\kappa^2 T_1 T_2)}{1 + \Delta\omega^2 T_2^2} \frac{w}{T_1} - \frac{w - w_0}{T_1} = -\frac{I}{I_{soff} T_+ 1} w - \frac{w - w_0}{T_1} \tag{3.79}$$

Equation (3.79) defines a saturation field at resonance $\tilde{\mathcal{E}}_{s0} = 1/(\kappa\sqrt{T_1 T_2})$. Off resonance, a larger field $\tilde{\mathcal{E}}_{soff} = \tilde{\mathcal{E}}_{s0}\sqrt{1 + \Delta\omega^2 T_2^2}$ is required to saturate the same transition. To that off-resonance saturation field corresponds a saturation intensity I_{soff}.

For pulses much shorter than the energy relaxation time $\tau_p \ll T_1$ and purely homogeneoulsy broadened media the rate equation (3.79) can be integrated together

with the propagation equation (3.69) which yields for the transmitted intensity

$$I(z,t) = I_0(t)\frac{e^{W(t)/W_s}}{e^{-a} - 1 + e^{W(t)/W_s}} \tag{3.80}$$

In this last equation $W(t) = \int_{-\infty}^{t} I_0(t)dt$, and $a = \sigma_{01}^{(0)} w_0 z/p$ is the linear gain/absorption coefficient. Equation (3.80) corresponds can be written in terms of photon flux F:

$$\boxed{F(z,t) = F_0(t)\frac{e^{2\sigma_{01}\bar{W}_0(t)}}{e^{-a} - 1 + e^{2\sigma_{01}\bar{W}_0(t)}}} \tag{3.81}$$

where $\bar{W}_0(t) = \int_{-\infty}^{t} F_0(t')dt' = 1/(\hbar\omega_\ell)\int_{-\infty}^{t} I_0(t')dt'$ (I_0 intensity of the incident pulse), cf. Eqs. (1.29), (1.30), and

$$a = \sigma_{01}\Delta N^{(e)} z \tag{3.82}$$

is the absorption ($\Delta N^{(e)} < 0$) or amplification ($\Delta N^{(e)} > 0$) coefficient corresponding to a sample of length z. $\bar{W}_0(t)$ is a measure of the incident pulse energy (area) density until time t in units of (photons)/cm^2. The total incident energy density is $\hbar\omega_\ell \bar{W}_0(t = \infty) = \hbar\omega_\ell \bar{W}_{0,\infty} = W_0$. The transmitted energy density $W(z,t) = \hbar\omega_\ell \bar{W}(z,t)$ is obtained by integrating Eq. (3.81) with respect to time and can be written as

$$\boxed{W(z,t) = \hbar\omega_\ell \int_{-\infty}^{t} F(z,t')dt' = W_s \ln\left[1 - e^{a}\left(1 - e^{W_0(t)/W_s}\right)\right],} \tag{3.83}$$

where $W_s = \hbar\omega_\ell/(2\sigma_{01})$ is the saturation energy density of the medium. With Eq. (3.79), in the limit $\tau_p \ll T_1$, we can express the population inversion as

$$\boxed{\Delta N(z,t) = \Delta N^{(e)} e^{-2\sigma_{01}\bar{W}(z,t)} = \frac{\Delta N^{(e)}}{1 - e^{a}[1 - e^{W_0(t)/W_s}]}.} \tag{3.84}$$

Femtosecond pulse propagation through a homogeneously broadened saturable medium in the limit of $T_2 \ll \tau_p \ll T_1$ is completely determined by two parameters: the saturation energy density W_s and the linear absorption (gain) coefficient a. Equation (3.80) is particularly useful in calculating pulse propagation in amplifiers, as further detailed in Chapter 8.1.

3.4.2 Steady-state approximation: linear and nonlinear optics

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).

$$\tilde{Q} = \frac{\kappa\tilde{\mathcal{E}}T_2 w}{1 - i\Delta\omega T_2}. \tag{3.85}$$

or, in terms of u and v:

$$u = -\frac{\Delta\omega T_2 \kappa \mathcal{E} T_2 w}{1+\Delta\omega^2 T_2^2} \tag{3.86}$$

$$v = -\frac{\kappa \mathcal{E} T_2 w}{1+\Delta\omega^2 T_2^2}. \tag{3.87}$$

Substituting v in the third Bloch equation (3.73) leads to the rate equation discussed in the previous Section [Eq. (3.79)]

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

$$u = -\frac{\Delta\omega T_2 \kappa \mathcal{E} T_2 w}{1+\Delta\omega^2 T_2^2+\kappa^2\mathcal{E}^2 T_1 T_2} \tag{3.88}$$

$$v = -\frac{\kappa \mathcal{E} T_2 w}{1+\Delta\omega^2 T_2^2+\kappa^2\mathcal{E}^2 T_1 T_2} \tag{3.89}$$

$$w = \frac{w_0(1+\Delta\omega^2 T_2^2)}{1+\Delta\omega^2 T_2^2+\kappa^2\mathcal{E}^2 T_1 T_2} = \frac{w_0}{1+\frac{I}{I_{soff}}} \tag{3.90}$$

where we used the off-resonance saturation intensity defined in Eq. (3.79).

3.5 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 (3.67) can be written in the integral form:

$$\tilde{Q}(t) = \int_{-\infty}^{t} \kappa \mathcal{E} w e^{-i[(\omega_0-\omega_\ell)t'-\varphi(t')]} dt' \tag{3.91}$$

For weak pulses $w \approx w_0$) and the right hand side of Eq. (3.91) at $t = \infty$ is proportional to the Fourier transform of $\kappa\tilde{\mathcal{E}}w$. Thus we have:

$$|\tilde{Q}|^2 = u^2+v^2 = \kappa^2 w_0^2 |\tilde{\mathcal{E}}(\omega_0-\omega_\ell)|^2 \tag{3.92}$$

$$\approx -2w_0(w_\infty - w_0) \tag{3.93}$$

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:

$$(w_\infty - w_0) = -\frac{\kappa^2 w_0}{2}|\tilde{\mathcal{E}}(\omega_0 - \omega_\ell)|^2. \tag{3.94}$$

This is a close connection to linear optics. Equation (3.92) 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. (3.94) 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.