
Advanced Laser

SEMI-CLASSICAL MODEL

4.1 Introduction

The rate equations, as they were firstly introduced by Tang, Statz and deMars [1] give access to many valuable information such as the threshold of the laser or the efficiency slope of the laser. However it is a very simple model, which has its limitations.

1. This model cannot describe the operating frequency of the laser. We saw that a Fabry-Perot cavity would allow a comb of discrete frequency only. What if the frequency ν determined by the energy levels through $\Delta E = h\nu$ does not coincide with one of these allowed frequencies? Can the laser work?
2. We also that mode-locked pulses result from a coherent superposition of longitudinal modes. Once again the model of the rate equation cannot describe the spectral bandwidth of the emitted light, which could not yield to pulses!
3. The description of the light-matter interaction in the TSdM model is rather unsatisfactory. In the Tang, Statz and deMars model, light interacts perfectly with the active medium but the reality is unfortunately not as ideal.

Although the field of quantum mechanics is very interesting, even a short introduction should require many hours of lecture. In the present case we only want to introduce a few of the tools that will be necessary to better describe the light-matter interaction and establish a better model than the one introduced by Tang, Statz and deMars.

Newton (1672) considered that light consists of small particles called *corpuscles*. These corpuscles can bound on the surface of the mirror in the case of reflection. Such approach is totally inadequate for the description of interference and diffraction phenomena. The alternative - rival- description of light as a wave given by Christiaan Huygens in 1690 [2] was not at all considered prior the works from Thomas Young and later by Augustin Fresnel, who used this approach to explain the phenomenon of diffraction in 1818. In the frame of electromagnetism, unified by Maxwell's equations, the speed of light is directly linked to electric and magnetic quantities and polarisation phenomena can be interpreted as a consequence of the vectorial properties of the electric field. Electromagnetic theory however could not be used to explain the spectrum of black-body radiation yield Max Planck's hypothesis that *energy was quantified*: For an given electromagnetic wave with a frequency ν the only possible energies are integral number of $h\nu$ where h is a newly introduced fundamental constant. Albert Einstein pushed this hypothesis further, returning toward the corpuscles' theory: light consists of a stream of particles the photons, each of them has an energy $h\nu$. Using this approach Einstein could explain the photoelectric effect, for which he got the Nobel prize in 1921. Corpuscles properties (energy E and momentum \mathbf{p}) and wave parameters of light (angular frequency ω and wavevector \mathbf{k}) are linked by the *Planck-Einstein relations*:

$$E = h\nu = \hbar\omega \tag{4.1a}$$

$$\mathbf{p} = \hbar\mathbf{k} \tag{4.1b}$$

where the $\hbar = (h/2\pi)$ is the reduced Planck constant and $h \simeq 6.62 \times 10^{-34}$ J.s.

In parallel to the discovery of the photons the studies of emission and absorption spectrum of atoms exhibit another peculiar behaviour that cannot be explained by the classical physics: the spectra are discrete. In other words, an atom can only absorb or emit at very well determined frequency. Such a phenomena is more easily understandable if we consider that the energy levels of the atoms are quantified and can only take discrete values $E_1, E_2 \dots E_n$. The conservation of energy implied that

$$h\nu_{ij} = |E_i - E_j| \quad (4.2)$$

In 1923 Louis de Broglie proposes that not only photons but also *material corpuscles* can have an *undulatory behaviour*. This theory was experimentally verified by Davisson and Gremer in 1927 when they reported on the observation of interference pattern in a Young's double-slit experiment using electrons. Considering the Planck-Einstein relations applied to material corpuscles we can associate the de Broglie wavelength¹:

$$\lambda = \frac{2\pi}{\mathbf{k}} = \frac{h}{\mathbf{p}} \quad (4.3)$$

4.1.1 The wave-function

As a consequence of the Young's double-slit experiment the classical notion of trajectory is not appropriate for quantum system and we need to replace this by the notion of *time-dependent state*.

1. The quantum state of a corpuscle² is then characterised by a *wave-function* $\psi(\mathbf{r}, t)$ which contains all the information that we can measure from the corpuscle.
2. The wave-function $\psi(\mathbf{r}, t)$ can be interpreted as an amplitude probability to locate the particle as a given location.
3. The probability to locate a single particle in a volume $d^3r = dx dy dz$ is therefore

$$d\mathcal{P}(\mathbf{r}, t) = C |\psi(\mathbf{r}, t)|^2 d^3r \quad (4.4)$$

where C is normalisation's constant and $|\psi(\mathbf{r}, t)|^2$ is a probability density.

4. Since the probability to find the particle anywhere in the space is unity then

$$\int |\psi(\mathbf{r}, t)|^2 d^3r \quad \text{is finite} \quad (4.5)$$

5. A physical measurable quantity of the quantum system is called an *observable* and is represented by an operator \hat{O} . For every measurement of the observable associated to the operator \hat{O} , the only possible value are the eigenvalues a of the operator. And for each eigenvalue a is associated an eigenfunction $\psi_a(\mathbf{r}, t)$. Of course any arbitrary state described by a given wave-function $\psi(\mathbf{r}, t)$ can be expressed by using the set of eigenvectors $\psi_a(\mathbf{r}, t)$ of the operator \hat{O} such that:

$$\psi(\mathbf{r}, t) = \sum_a c_a \psi_a(\mathbf{r}, t) \quad (4.6)$$

¹Obviously the very short value of the Planck constant prevent the observation of the undulatory behaviour for macroscopic objects.

²In the case of an electron, this does not take into account the existence of the spin.

Note that the coefficient c_a are in general complex numbers and $|c_a|^2$ is the probability to find that the measurement of the wave-function $\psi(\mathbf{r}, t)$ yields the value a . Of course if the state is described by $\psi(\mathbf{r}, t) = \psi_a(\mathbf{r}, t)$, then any measurement of the state yield the value a . On the other hand, if an experiment is repeated many times on an ensemble of particles prepared in the state ψ , the average of the results will be equal to

$$\langle \hat{O} \rangle = \int \psi^* \hat{O} \psi d^3\mathbf{r} \quad (4.7)$$

This average value is called the *expectation value*.

6. The ensemble of eigenfunction $\psi_a(\mathbf{r}, t)$ forms a complete basis of the so-called *Hilbert space* and two eigenfunctions are orthogonal to each other:

$$\int \psi_i^*(\mathbf{r}, t) \psi_j(\mathbf{r}, t) d^3\mathbf{r} = \delta_{ij} \quad (4.8)$$

where δ_{ij} is the Kronecker delta function defined by

$$\begin{cases} \delta_{ij} = 1 & \text{if } i = j \\ \delta_{ij} = 0 & \text{if } i \neq j \end{cases} \quad (4.9)$$

7. The wave-function of particle in a potential $V(\mathbf{r}, t)$ evolves according to the Schrodinger equation

$$i\hbar \frac{\partial}{\partial t} \psi(\mathbf{r}, t) = \hat{H} \psi(\mathbf{r}, t) \quad (4.10)$$

where \hat{H} is the *Hamiltonian* operator representing the total energy of the system:

$$\hat{H} = \hat{T} + \hat{V} \quad (4.11)$$

with \hat{T} and \hat{V} are respectively the kinetic and the potential energy.

4.1.2 The Dirac notation

Introduction and link with the wave-function description

As we just saw the particle can be described by its wave-function. Another way to describe the quantum state is to use the eigenstates, also called the *vector states* in Hilbert space. Dirac proposed a convenient compact notation to represent the state vector of Hilbert space and their properties. In this notation the wave-function is represented by the **ket** vector

$$\psi \equiv |\psi\rangle \quad (4.12)$$

and the corresponding complex conjugate is represented by the **bra** vector

$$\psi^* \equiv \langle \psi| \quad (4.13)$$

There is obviously a strong link between both notations and the expectation value expressed with the Dirac notation is

$$\langle \psi| \hat{O} |\psi\rangle = \int \psi^* \hat{O} \psi d^3\mathbf{r} \quad (4.14)$$

We can also represent the *overlap integral*³ by

$$\langle \psi | \varphi \rangle \equiv \int \psi^* \varphi d^3 \mathbf{r} \quad (4.15)$$

Finally the *matrix element* of the operator \hat{O} is given by

$$\langle n | \hat{O} | m \rangle \equiv \int \psi_n^* \hat{O} \psi_m d^3 \mathbf{r} \quad (4.16)$$

Note that the Dirac notation does not make the calculations simpler, but the appearance of the equations more compact.

Example of the isolated 2-level system

Let consider a gas of N_0 identical particles that can only have two distinct energy level. The states is described as

$$|\psi\rangle = c_1 |1\rangle + c_2 |2\rangle \rightarrow \begin{pmatrix} c_1 \\ c_2 \end{pmatrix} \quad (4.17)$$

c_1 and c_2 are arbitrary complex numbers. From eq. (4.17) we can write the equivalent *bra*-vector

$$\langle \psi | \rightarrow \begin{pmatrix} c_1^* & c_2^* \end{pmatrix} \quad (4.18)$$

and the normalisation condition follows as

$$\langle \psi | \psi \rangle = \begin{pmatrix} c_1^* & c_2^* \end{pmatrix} \begin{pmatrix} c_1 \\ c_2 \end{pmatrix} = |c_1|^2 + |c_2|^2 = 1 \quad (4.19)$$

As we see, this approach can be seen as of scalar product of two vectors analogous to matrix algebra. We see clearly that $|c_i|^2$ is the probability⁴ that the system is in the state $|i\rangle$ and the state can be decomposed by using two orthogonal eigenvectors. Using the matrix analogy we can rewrite the state (eq. (4.17) as

$$\langle \psi | = c_1 \begin{pmatrix} 1 \\ 0 \end{pmatrix} + c_2 \begin{pmatrix} 0 \\ 1 \end{pmatrix} \quad (4.20)$$

Imagine how this matrix form could become for more complicated state... We can clearly see that the Dirac form is much more elegant and compact! Of course we can readily check that the basis is *orthonormal*:

$$\langle 1 | 2 \rangle = \begin{pmatrix} 1 & 0 \end{pmatrix} \begin{pmatrix} 0 \\ 1 \end{pmatrix} = 0 \quad (4.21a)$$

$$\langle 1 | 1 \rangle = \begin{pmatrix} 1 & 0 \end{pmatrix} \begin{pmatrix} 1 \\ 0 \end{pmatrix} = 1 \quad (4.21b)$$

The first equation shows that the two eigenvector are othorgonal and the second one is the normalisation condition, *i.e.* that the quantum states have a unit form.

The Hamiltonian operator \hat{H} of the system represents the possible energy levels of the quantum system. Let E_1 be the energy of the state with the lowest energy and E_2 the

³... and if the states are orthogonal, this integral is null.

⁴Considering the total number of particle, we have $|c_i|^2 = (N_i/N_0)$.

one with the largest energy. In the case of a totally isolated particle the matrix associated with the Hamiltonian of the system must only contain these quantity since these are the only possible physical observable of the system. Moreover, as we saw $|1\rangle$ and $|2\rangle$ are the eigenvectors of the system (of \hat{H}) which is therefore written⁵

$$\hat{H} = \begin{bmatrix} E_1 & 0 \\ 0 & E_2 \end{bmatrix} \quad (4.22)$$

We can interpret the form of the Hamiltonian as follow:

1. the diagonal elements are the values of the possible energy of each state.
2. the non-diagonal elements take into account the possible interactions between the various levels. In the present case the two level do not interact, and therefore the off-diagonal elements are null.

Using the eq. (4.14) to obtain the expectation value of any operator we need to calculate the product

$$\langle \psi | \hat{O} | \psi \rangle = \begin{pmatrix} \psi_1^* & \psi_2^* \end{pmatrix} \begin{bmatrix} a_{11} & a_{12} \\ a_{21} & A_{22} \end{bmatrix} \begin{pmatrix} \psi_1 \\ \psi_2 \end{pmatrix} \quad (4.23)$$

In the current case, we can calculate the expected value if we have the system on the ground state $|1\rangle$:

$$\langle 1 | \hat{H} | 1 \rangle = \begin{pmatrix} 1 & 0 \end{pmatrix} \begin{bmatrix} E_1 & 0 \\ 0 & E_2 \end{bmatrix} \begin{pmatrix} 1 \\ 0 \end{pmatrix} = E_1 \quad (4.24)$$

and the expected value for an arbitrary state $|\psi\rangle = c_1 |1\rangle + c_2 |2\rangle$ is given by

$$\begin{aligned} \langle \psi | \hat{H} | \psi \rangle &= |c_1|^2 \begin{pmatrix} 1 & 0 \end{pmatrix} \begin{bmatrix} E_1 & 0 \\ 0 & E_2 \end{bmatrix} \begin{pmatrix} 1 \\ 0 \end{pmatrix} + |c_2|^2 \begin{pmatrix} 0 & 1 \end{pmatrix} \begin{bmatrix} E_1 & 0 \\ 0 & E_2 \end{bmatrix} \begin{pmatrix} 0 \\ 1 \end{pmatrix} \\ &= |c_1|^2 E_1 + |c_2|^2 E_2 \end{aligned} \quad (4.25)$$

The projector operator \hat{P}

In order to retrieve the information about the physical quantities of the quantum state (position, momentum, angular momentum... etc) we can introduce a specific operator called the projector operator. It is defined as

$$\hat{P} = |k\rangle \langle k| \quad (4.26)$$

Assuming that we have a state defined by ψ , the projector operator *projects* this state one a particular state $|k\rangle$ as follows

$$\hat{P} |\psi\rangle = |k\rangle \langle k | \psi \rangle = c_k |k\rangle \quad (4.27)$$

As we see, the quantity $\langle k | \psi \rangle \equiv c_k$ represents the projection of the state vector ψ onto the vector $|k\rangle$. As previously, this concept is fully analogous to the concept of projection of a vector onto another one since the quantity c_k represents the scalar product between the two vectors.

⁵At thermal equilibrium the distribution of particle on the different levels of energy follows the Gibbs-Boltzmann distribution.

In the case of the isolated 2-level system we can express the projector operator:

$$\hat{P}_1 = |1\rangle \langle 1| = \begin{bmatrix} 1 & 0 \\ 0 & 0 \end{bmatrix} \quad (4.28a)$$

$$\hat{P}_2 = |2\rangle \langle 2| = \begin{bmatrix} 0 & 0 \\ 0 & 1 \end{bmatrix} \quad (4.28b)$$

$$(4.28c)$$

Note that we can readily see that $\hat{P}_1 + \hat{P}_2 = \mathbb{1}$. More generally for a system with n-dimension we have the *closure relation*

$$\sum_k \hat{P}_k = \sum_k |k\rangle \langle k| \equiv \mathbb{1} \quad (4.29)$$

Using a complete set of projector operators we can therefore express any quantum state as

$$|\psi\rangle = \mathbb{1} |\psi\rangle = \sum_k |k\rangle \langle k|\psi\rangle = \sum_k c_k |k\rangle \quad (4.30)$$

Once again the analogy with linear algebra is quite obvious. This simply means that we can change from one system of coordinates (say Cartesian) into another one (say polar).

4.1.3 The density matrix

A special case of projector operator is the *density matrix*. This somehow corresponds to the concept of quantum state but instead of describing with the help of the state vectors, it describes the system as an operator, *i.e.* as a matrix. Knowing the state vector ψ the density matrix is define as

$$\hat{\rho} = |\psi\rangle \langle \psi| \quad (4.31)$$

From the definition of the state $|\psi\rangle = c_1 |1\rangle + c_2 |2\rangle$ we get the density matrix as

$$\hat{\rho} = \begin{bmatrix} |c_1|^2 & c_1^* c_2 \\ c_1 c_2^* & |c_2|^2 \end{bmatrix} = \begin{bmatrix} \rho_{11} & \rho_{12} \\ \rho_{21} & \rho_{22} \end{bmatrix} \quad (4.32)$$

We can notice that in this matrix ($\rho_{12} = \rho_{21}^*$). The diagonal elements of the density matrix are the probability to find the quantum system in the state $|i\rangle$. The off-diagonal elements represent the *coherence terms*. These refer somehow to the coupling between the various states of the system.

Using the density matrix the evolution of the quantum state follows the Heisenberg equation

$$i\hbar \frac{d\hat{\rho}}{dt} = [\hat{H}, \hat{\rho}] \quad (4.33)$$

where

$$[\hat{A}, \hat{B}] = \hat{A}\hat{B} - \hat{B}\hat{A} \quad (4.34)$$

is the commutator between the operators \hat{A} and \hat{B} .

4.1.4 Light-matter interaction for an isolated single particle

So far, the Hamiltonian (eq. (4.22)) describes the situation where the atom has two possible energy level and is totally isolated. To allow the coupling between the light and the atom, we need to modify the Hamiltonian. The standard approach is to use the so-called *dipole approximation*. This consists in describing the atom itself as an electric dipole (i.e., a coupled system formed by a positive charge, the nucleus, and a negative charge, the electron, binded together by a force resulting from the Coulomb potential. The energy for a dipole in an electromagnetic field is

$$V = -\boldsymbol{\mu} \cdot \mathbf{E} \quad (4.35)$$

where $\boldsymbol{\mu} = q\hat{\mathbf{e}}_x$ is the dipole moment of the system, q is the charge and \mathbf{E} the electric field. To study the evolution of the single atom in an electric field we need to use the modified Hamiltonian, which takes into account both its proper energy and the interaction energy E_I :

$$\hat{H} = \hat{H}_0 + \hat{H}_I \quad (4.36)$$

where \hat{H}_I is the Hamiltonian resulting from the interaction⁶

$$\hat{H}_I = \begin{bmatrix} 0 & V_{12} \\ V_{21} & 0 \end{bmatrix} \quad (4.37)$$

Here $V_{ij} = -\mu_{ij}E$ represents the interaction of the electromagnetic field with the quantum system⁷. The idea now is to use this Hamiltonian to describe the situation of atoms interacting with an external electric field. This ultimately yields the so-called *Maxwell-Bloch equations*, always known as the *semi-classical model* since the matter is described quantum mechanically - to obtain the microscopic polarisation, whilst the electric field is classically described by Maxwell's equations.

For N particles we can calculate the macroscopic polarisation $P(z, t)$ from

$$P(z, t) = N [\mu_{12}\rho_{12}(z, t) + \mu_{21}^*\rho_{21}(z, t)] = N\mu\rho_{12} + c.c. \quad (4.38)$$

where $\mu_{12} = \mu_{21}^* = \mu$ is the dipole moment induced by the electric field and ρ_{ij} the off-diagonal elements of the density matrix. Since these terms depend on the relative phases of each state $|i\rangle$ they are called *coherence*. Using the Heisenberg representation we can study the evolution of the density matrix according to

$$i\hbar \frac{d}{dt} \hat{\rho} = [\hat{H}, \hat{\rho}] \quad (4.39)$$

Remembering that the commutator operator is $[\hat{A}, \hat{B}] = \hat{A}\hat{B} - \hat{B}\hat{A}$ and using the full Hamiltonian $\hat{H} = \hat{H}_0 + \hat{H}_I$ we have:

$$\begin{aligned} i\hbar \frac{d}{dt} \begin{bmatrix} \rho_{11} & \rho_{12} \\ \rho_{21} & \rho_{22} \end{bmatrix} &= \begin{bmatrix} E_1 & 0 \\ 0 & E_2 \end{bmatrix} \begin{bmatrix} \rho_{11} & \rho_{12} \\ \rho_{12}^* & \rho_{22} \end{bmatrix} - \begin{bmatrix} \rho_{11} & \rho_{12} \\ \rho_{12}^* & \rho_{22} \end{bmatrix} \begin{bmatrix} E_1 & 0 \\ 0 & E_2 \end{bmatrix} \\ &+ \begin{bmatrix} 0 & V \\ V & 0 \end{bmatrix} \begin{bmatrix} \rho_{11} & \rho_{12} \\ \rho_{12}^* & \rho_{22} \end{bmatrix} - \begin{bmatrix} \rho_{11} & \rho_{12} \\ \rho_{12}^* & \rho_{22} \end{bmatrix} \begin{bmatrix} 0 & V \\ V & 0 \end{bmatrix} \\ &= \begin{bmatrix} V(\rho_{12}^* - \rho_{12}) & V(\rho_{22} - \rho_{11}) - (E_2 - E_1)\rho_{12} \\ V(\rho_{11} - \rho_{22}) + (E_2 - E_1)\rho_{12}^* & V(\rho_{12} - V\rho_{12}^*) \end{bmatrix} \end{aligned} \quad (4.40)$$

⁶In this approach we consider that there is no permanent dipole moment. Therefore the Hamiltonian only exhibits off-diagonal non-zero elements.

⁷Rigorously the value μ_{ij} is the expectation value of the dipole moment operator $\hat{\mu}$ that links the level $|i\rangle$ and $|j\rangle$. According to eq. (4.16) this can be written as $\mu_{ij} = \langle i | \hat{\mu} | j \rangle$. Moreover since we are only considering a 2-level system then $\mu_{21} = \mu_{12}^*$.

Introducing the resonant frequency $\omega_a = (E_2 - E_1) / \hbar$ we have the evolution of the density matrix

$$\begin{bmatrix} \dot{\rho}_{11} & \dot{\rho}_{12} \\ \dot{\rho}_{21} & \dot{\rho}_{22} \end{bmatrix} = \begin{bmatrix} \frac{-i}{\hbar} V (\rho_{12}^* - \rho_{12}) & i\omega_a \rho_{12} - \frac{i}{\hbar} V (\rho_{22} - \rho_{11}) \\ -\frac{i}{\hbar} V (\rho_{11} - \rho_{22}) - i\omega_a \rho_{12}^* & \frac{-i}{\hbar} V (\rho_{12} - \rho_{12}^*) \end{bmatrix} \quad (4.41)$$

Since $\rho_{ii} \in \mathbb{R}$, we can notice that $\dot{\rho}_{21} = \dot{\rho}_{12}^*$. Note that the evolution of the density matrix only

4.1.5 Maxwell-Bloch equations

The eq. (4.41) gives the evolution of each element of the density matrix. However it is important to take into account the lifetime of the level and the possible decoherence processes resulting for instance from collision between atoms in a gas, the generation of phonon in a crystal or simply the spontaneous emission. These can be introduced phenomenologically by

$$\begin{aligned} \dot{\rho}_{11} &= \frac{i}{\hbar} V \rho_{12} + c.c. - \gamma_1 \rho_{11} \\ \dot{\rho}_{22} &= -\frac{i}{\hbar} V \rho_{12} + c.c. - \gamma_1 \rho_{22} \\ \dot{\rho}_{21} &= -i\omega_a \rho_{21} + \frac{i}{\hbar} V (\rho_{22} - \rho_{11}) - \gamma_{\perp} \rho_{21} \end{aligned}$$

where γ_j is the decay rate of the level j and γ_{\perp} is the decay rate of the coherence term. This means that the induced polarisation relaxes over time. Traditionally this lifetime is called T_2 and is equal to γ_{\perp}^{-1} . In gas laser this time would correspond to the mean value between collisions between two atoms.

Additionally we need to take into account that each level j can be pumped and therefore we have an extra term λ_j so that finally we have

$$\dot{\rho}_{11} = \frac{i}{\hbar} V \rho_{12} + c.c. - \gamma_1 \rho_{11} + \lambda_1 \quad (4.42a)$$

$$\dot{\rho}_{22} = -\frac{i}{\hbar} V \rho_{12} + c.c. - \gamma_1 \rho_{22} + \lambda_2 \quad (4.42b)$$

$$\dot{\rho}_{21} = -i\omega_a \rho_{21} + \frac{i}{\hbar} V (\rho_{22} - \rho_{11}) - \gamma_{\perp} \rho_{21} \quad (4.42c)$$

The pumping process does not affect the coherence, and therefore the third equation is not modified.

Interaction with an external field

Suppose now that the system described by eq. (4.42) interacts with an electromagnetic field propagating along z :

$$E(z, t) = F(t) \cos(\omega t - kz) = \frac{1}{2} F(t) e^{i(\omega t - kz)} + c.c. \quad (4.43)$$

This field induces a dipole moment, which allows us to evaluate the density matrix and its evolution ($V = -\mu E$). The influence of this external field appears in the off-diagonal terms of the density matrix. Due to the oscillating behaviour of the external field we can then reformulate these term a

$$\rho_{21} = \tilde{\rho}_{21} e^{i(\omega t - kz)} \quad (4.44)$$

where $\tilde{\rho}_{21}$ is slowly varying in time and its dependence along the propagation is negligible. The derivative of this coherence term is

$$\dot{\rho}_{21} = \frac{d}{dt} [\tilde{\rho}_{21} e^{i(\omega t - kz)}] = i\omega \tilde{\rho}_{21} e^{i(\omega t - kz)} + \dot{\tilde{\rho}}_{21} e^{i(\omega t - kz)} \quad (4.45)$$

but we can also use the eq. (4.42c) to write

$$\dot{\rho}_{21} = -(i\omega_a + \gamma_{\perp})\rho_{21} e^{i(\omega t - kz)} - i \frac{\mu F(t)}{2\hbar} [e^{i(\omega t - kz)} + c.c.] (\rho_{22} - \rho_{11}) \quad (4.46)$$

Using both eq. (4.45) and (4.46) yields

$$\dot{\tilde{\rho}}_{21} = [-\gamma_{\perp} + i(\omega - \omega_0)] \tilde{\rho}_{21} - i \frac{\mu F(t)}{2\hbar} [1 + e^{-2i(\omega t - kz)}] (\rho_{22} - \rho_{11}) \quad (4.47)$$

And the term oscillating at 2ω is dropped since it oscillated too fast in comparison with the other terms. Of course we can also introduce the influence of E in eq. (4.42a) and (4.42b) to finally obtain

$$\dot{\rho}_{11} = \lambda_1 - \gamma_1 \rho_{11} + i \frac{\mu F}{2\hbar} (\tilde{\rho}_{21} - \tilde{\rho}_{12}) \quad (4.48a)$$

$$\dot{\rho}_{22} = \lambda_2 - \gamma_2 \rho_{22} - i \frac{\mu F}{2\hbar} (\tilde{\rho}_{21} - \tilde{\rho}_{12}) \quad (4.48b)$$

$$\dot{\tilde{\rho}}_{21} = [-\gamma_{\perp} + i(\omega - \omega_0)] \tilde{\rho}_{21} - i \frac{\mu F}{2\hbar} (\tilde{\rho}_{22} - \tilde{\rho}_{11}) \quad (4.48c)$$

These are called the Bloch equations.

Stationary state

Inversion of population: As we proceeded many times the station-nary states is obtained when the temporal derivatives are taken equal to zero. Let first analyse the case of the inversion of population. For a density N of atoms the difference of population between the levels is

$$\Delta N = N(\rho_{22} - \rho_{11}) \quad (4.49)$$

And without the perturbation from the external field the system evolves towards equilibrium so that

$$\lambda_i - \hat{\rho}_{ii} = 0 \Rightarrow \hat{\rho}_{ii} = \frac{\lambda_i}{\gamma_i} \quad (4.50)$$

$$\Delta \hat{N} = N(\hat{\rho}_{22} - \hat{\rho}_{11}) = N \left(\frac{\lambda_2}{\gamma_2} - \frac{\lambda_1}{\gamma_1} \right) \quad (4.51)$$

Dividing eq. (4.48a) by γ_1 and eq. (4.48b) by γ_2 and subtracting them yields

$$\left(\frac{\lambda_2}{\gamma_2} - \frac{\lambda_1}{\gamma_1} \right) - (\rho_{22} - \rho_{11}) - i \frac{\mu F}{2\hbar} (\tilde{\rho}_{21} - \tilde{\rho}_{12}) \left(\frac{1}{\gamma_1} + \frac{1}{\gamma_2} \right) \quad (4.52)$$

which can be simplified by introducing the mean relaxation time

$$T_1 = \frac{1}{2} \left(\frac{1}{\gamma_1} + \frac{1}{\gamma_2} \right) = \frac{1}{\gamma_{\parallel}} \quad (4.53)$$

This then leads to

$$\frac{\Delta \widehat{N}}{N} - \frac{\Delta N}{N} - i \frac{\mu F T_1}{2\hbar} (\tilde{\rho}_{21} - \tilde{\rho}_{12}) \quad (4.54)$$

We can now use the eq. (4.48c), which in the stationary state should be null yielding

$$\tilde{\rho}_{21} = \frac{i\mu F}{2\hbar [-\gamma_{\perp} + i(\omega - \omega_0)]} (\rho_{22} - \rho_{11}) = \frac{i\mu F}{2\hbar [-\gamma_{\perp} + i(\omega - \omega_0)]} \frac{\Delta N}{N} \quad (4.55)$$

Finally we should remind that $\rho_{21} = \rho_{12}^*$ and insert it into eq. (4.54) to obtain

$$\frac{\Delta \widehat{N}}{N} - \frac{\Delta N}{N} - i \frac{\mu F T_1}{\hbar} \left\{ \frac{i\mu F}{2\hbar [-\gamma_{\perp} + i(\omega - \omega_0)]} + \frac{i\mu F}{2\hbar [-\gamma_{\perp} - i(\omega - \omega_0)]} \right\} \quad (4.56)$$

$$\Rightarrow \boxed{\Delta N = \Delta \widehat{N} \frac{1 + \left(\frac{\omega - \omega_0}{\gamma_{\perp}} \right)^2}{1 + \left(\frac{\omega - \omega_0}{\gamma_{\perp}} \right)^2 + \frac{\mu^2 F^2}{\hbar^2} T_1 T_2}} \quad (4.57)$$

This reads rather complicated but if we look at it more carefully we see that at perfect resonance the term in (...) vanishes and the remaining term simply reads as

$$\Delta N = \Delta \widehat{N} \frac{1}{1 + S} \quad \text{with} \quad S = \frac{\mu^2 F^2}{\hbar^2} T_1 T_2 \quad (4.58)$$

S is the saturation parameter and it has the form of (I/I_s) where the intensity I is linked to the square of the amplitude F^2 and I_s is the intensity of saturation.

Macroscopic polarisation: The macroscopic polarisation for a density N of atoms is given by eq. (4.55)

$$P(z, t) = N\mu \left[\tilde{\rho}_{12} e^{i(\omega t - kz)} + \tilde{\rho}_{21} e^{-i(\omega t - kz)} \right]$$

and we can now substitute ρ_{21} by its expression (eq. (4.55)) in order to obtain

$$P(z, t) = \frac{\Delta \widehat{N} \mu^2 F}{\gamma_{\perp} \hbar} \frac{\left(\frac{\omega - \omega_0}{\gamma_{\perp}} \right) \cos(\omega t - kz) - \sin(\omega t - kz)}{1 + \left(\frac{\omega - \omega_0}{\gamma_{\perp}} \right)^2 + S} \quad (4.59)$$

As we see, the polarisation has two sinusoidal dependence: one that is in phase with the incident field and one quadrature. To take into account both dependencies we can express the polarisation as

$$P(z, t) = \frac{1}{2} p e^{i(\omega t - kz)} + c.c \quad (4.60)$$

where $p = \epsilon_0 \chi$ where the dielectric susceptibility of the material $\chi(\omega) = \chi_{\text{Re}} + i\chi_{\text{Im}}$ is a complex quantity. We can then identify

$$\chi_{\text{Re}}(\omega) = \frac{\mu^2 \Delta \widehat{N}}{\gamma_{\perp} \epsilon_0 \hbar} \frac{\left(\frac{\omega - \omega_0}{\gamma_{\perp}} \right)}{1 + \left(\frac{\omega - \omega_0}{\gamma_{\perp}} \right)^2 + S} \quad (4.61a)$$

$$\chi_{\text{Im}}(\omega) = \frac{\mu^2 \Delta \widehat{N}}{\gamma_{\perp} \epsilon_0 \hbar} \frac{1}{1 + \left(\frac{\omega - \omega_0}{\gamma_{\perp}} \right)^2 + S} \quad (4.61b)$$

We remind that ω is the frequency of the external electric field and ω_a the central frequency of the transition. The real part of the susceptibility (χ_{Re}), which is in phase with the external field is linked with the dispersion of the material. The imaginary part (χ_{Im}), in quadrature with the external field is linked either with the absorption (for $\Delta \widehat{N} < 0$) or with a gain (for $\Delta \widehat{N} > 0$).

Shape of the transition: In the case of a weak external field ($S \simeq 0$) the expression for the susceptibility can be simplified:

$$\chi_{\text{Re}}(\omega) = \frac{\mu^2 \Delta \widehat{N}}{\gamma_{\perp} \epsilon_0 \hbar} \pi \gamma_{\perp} \left(\frac{\omega - \omega_0}{\gamma_{\perp}} \right) \mathcal{L}(\omega) \quad (4.62a)$$

$$\chi_{\text{Im}}(\omega) = \frac{\mu^2 \Delta \widehat{N}}{\gamma_{\perp} \epsilon_0 \hbar} \pi \gamma_{\perp} \mathcal{L}(\omega) \quad (4.62b)$$

where $\mathcal{L}(\omega)$ is the Lorentzian function⁸

$$\mathcal{L}(\omega) = \frac{1}{\pi \gamma_{\perp}} \frac{1}{1 + \left(\frac{\omega - \omega_0}{\gamma_{\perp}} \right)^2} \quad (4.63)$$

It is remarkable that a classical approach - the so-called *bound electron model* yields the same shape of the resonance (see .1). When the external field however the term due to the saturation S becomes more and more important. In this case the shape of the transition is still a Lorentzian but the normalisation factor differs slightly so that it becomes:

$$\mathcal{L}(\omega) = \frac{1}{\pi \gamma_{\perp} \sqrt{1 + S}} \frac{1}{1 + \left(\frac{\omega - \omega_0}{\gamma_{\perp} \sqrt{1 + S}} \right)^2} \quad (4.64)$$

for which the FWHM becomes $\delta\omega_{\text{sat.}} = 2\gamma_{\perp} \sqrt{1 + S}$. To remain as general as possible it is better to describe the shape of the transition with the non-normalised Lorentzian function

$$\mathcal{S}(\omega) = \frac{1}{1 + \left(\frac{\omega - \omega_0}{\gamma_{\perp}} \right)^2} \quad (4.65)$$

⁸Expressed in this form, the shape of the transition is normalised

$$\int_{-\infty}^{\infty} \mathcal{L}(\omega) = 1$$

and the full width at half maximum $\delta\omega = 2\gamma_{\perp}$.

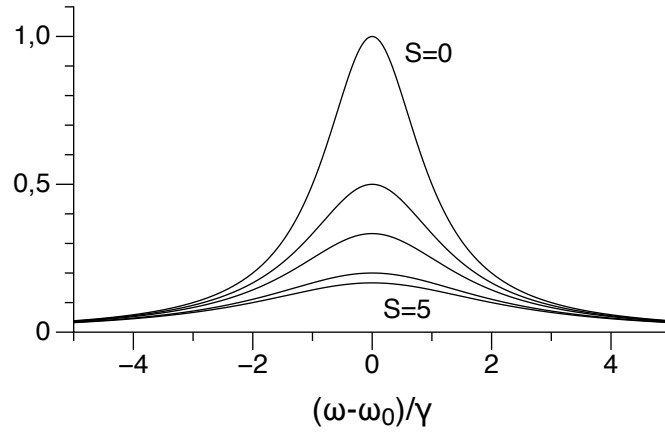


Figure 4.1: Influence of the saturation on the Lorentzian function (eq.)

so that the influence of the saturation is simply introduced by

$$\frac{1}{1 + \left(\frac{\omega - \omega_0}{\gamma_{\perp}}\right)^2 + S} = \frac{\mathcal{S}(\omega)}{1 + S \mathcal{S}(\omega)} \quad (4.66)$$

the influence of the saturation of the line-width of the transition is clearly presented on fig. 4.1

Using the non-normalised Lorentzian yields the real (resp. imaginary) part of the susceptibility:

$$\chi_{\text{Re}} = \frac{\mu^2 \Delta \widehat{N}}{\gamma_{\perp} \epsilon_0 \hbar} \frac{\mathcal{S}(\omega)}{1 + S \mathcal{S}(\omega)} \left(\frac{\omega - \omega_0}{\gamma_{\perp}}\right) \quad (4.67a)$$

$$\chi_{\text{Im}} = \frac{\mu^2 \Delta \widehat{N}}{\gamma_{\perp} \epsilon_0 \hbar} \frac{\mathcal{S}(\omega)}{1 + S \mathcal{S}(\omega)} \quad (4.67b)$$

$$(4.67c)$$

The real part of the susceptibility is an odd-function whilst the imaginary is even. These functions are plotted for the case $S = 0$ and $S = 2.5$ on fig. 4.2. It is clear from this picture that the gain medium will induce dispersion, and the magnitude of this dispersion depends on the external field!

4.1.6 Equation for the field

From Maxwell equations...

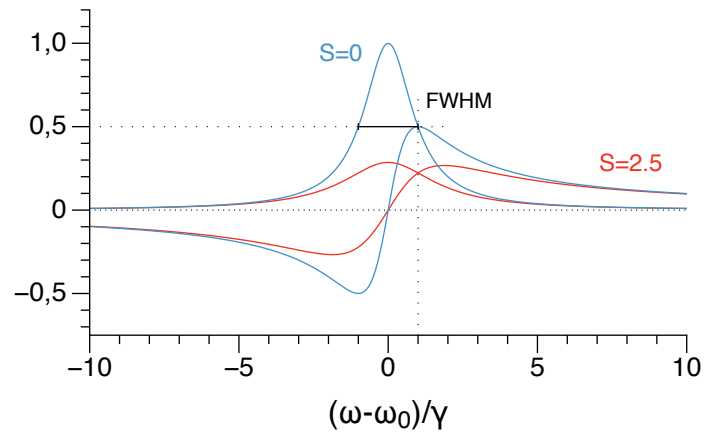


Figure 4.2: Real and imaginary parts of the susceptibility in the case of a gain medium ($\Delta\widehat{N} > 0$).

Appendix A

The bound electron model - the classical picture

The bound electron model: It is common to introduce the origin of the dielectric susceptibility with the help of the bound electron model. In this model the

In this picture, the electron is bound to the nucleus and the motion of that electron¹ can simply be modelled by an harmonic oscillator, which is forced by the incident electric field $E(t)$. Note that in this model the force on the electron is simply given by $\mathbf{F} = -e\mathbf{E}$ where only \mathbf{E} is the electric field and $(-e)$ the elementary charge for the electron. The magnetic field would need to be included if the electrons were relativistic, which is not the case here.

$$\frac{\partial^2 r}{\partial t^2} + \gamma \frac{\partial r}{\partial t} + \omega_0^2 r = -\frac{\bar{e}}{m_e} E(t) \quad (\text{A.1})$$

where r represents the distance between the nucleus and the electron, $-\bar{e}$ is the elementary charge of the electron, γ a damping coefficient, and ω_0 is the resonance frequency of the system. Assuming a monochromatic wave, we can use the complex notation for the electric field

$$E(t) = \frac{1}{2} \mathcal{E}_0 e^{-i\omega t} + c.c \quad (\text{A.2})$$

and insert it in in eq. (68). We should now seek an oscillating solution for r . We readily obtain

$$r(t) = -\frac{\bar{e}}{m_e} \frac{\mathcal{E}_0}{2} \frac{e^{-i\omega t}}{(\omega_0^2 - \omega^2) - i\gamma\omega} + c.c. = \frac{-\bar{e}E(t)}{m_e D(\omega)} \quad (\text{A.3})$$

where we introduce $D(\omega) = \omega_0^2 - \omega^2 - i\gamma\omega$. In the vicinity of a resonance ($\omega \simeq \omega_0$) $\Rightarrow |\omega - \omega_0| \ll \omega_0$ and therefore

$$\begin{aligned} D(\omega) &= (\omega_0^2 - \omega^2) - i\gamma\omega = (\omega_0 + \omega)(\omega_0 - \omega) - i\gamma\omega \\ &\simeq 2\omega_0(\omega_0 - \omega) - i\gamma\omega_0 \\ D(\omega) &\simeq -2\omega_0 \left[\Delta + i\frac{\gamma}{2} \right] \end{aligned}$$

where $\Delta = \omega - \omega_0$ leading to

$$r(t) \simeq \frac{\bar{e} E(t)}{2m_e\omega_0 \left(\Delta + i\frac{\gamma}{2} \right)} \quad (\text{A.5})$$

¹In this picture, the mass of the nucleus $m_n \gg m_e$, the mass of the electron. Therefore only the electronic displacement is considered for the calculation of the dipole.

We can then deduce the macroscopic polarisation $P(t) = N(-\bar{e})r(t) = \epsilon_0\chi^{(1)}E(t)$, where ϵ_0 is the vacuum permittivity, and $\chi^{(1)}$ the *linear susceptibility* is given by:

$$\chi^{(1)} = \frac{-N\bar{e}^2}{2m_e\omega_0\epsilon_0} \left[\frac{\Delta - i\gamma/2}{\Delta^2 + \gamma^2/4} \right]$$

from which we can extract the real and imaginary parts:

$$\text{Re} [\chi^{(1)}] = \frac{-N\bar{e}^2}{2m_e\omega_0\epsilon_0} \frac{\Delta}{\Delta^2 + \gamma^2/4} \quad (\text{A.6a})$$

$$\text{Im} [\chi^{(1)}] = \frac{+N\bar{e}^2}{2m_e\omega_0\epsilon_0} \frac{\gamma/2}{\Delta^2 + \gamma^2/4} \quad (\text{A.6b})$$

Obviously, there exist several electronic resonances across the electromagnetic spectrum, and they will all contribute to the final susceptibility. Each resonance ω_k has not only its own absorption bandwidth γ_k but also contributes differently to the overall susceptibility, with a weight f_k . This results in

$$\chi(\omega) = \frac{N\bar{e}^2}{m_e\epsilon_0} \sum_k \frac{f_k}{(\omega_k^2 - \omega^2) - i\gamma_k\omega} \quad (\text{A.7})$$

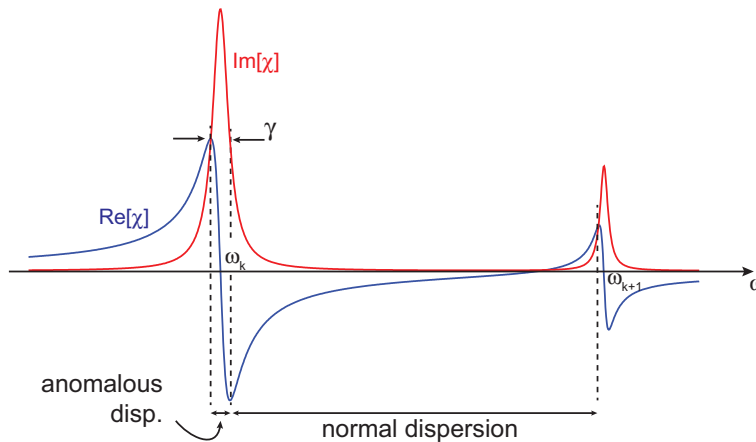


Figure A.1: Overall susceptibility considering multiple resonances ω_k

What is interesting here is that this simple picture of a bound electron model gives the very same results that the quantum mechanics approach.

Bibliography

- [1] C. Tang, H. Statz, and G. deMars, “Spectral output and spiking behavior of solid-state lasers,” *Journal of Applied Physics*, vol. 34, no. 8, p. 2289, 1963.
- [2] C. Huygens, *Traite de la lumiere*. Pierre Vander Aa marchand libraire, 1690.
- [3] G. Stegeman and R. Stegeman, *Nonlinear Optics*, U. o. C. F. Glenn Boreman, Ed. John Wiley & Sons, Inc., 2012.
- [4] P. Mandel, *Nonlinear Optics*. Wiley-VCH Verlag GmbH, 2010.
- [5] R. W. Boyd, *Nonlinear Optics, Third Edition*, 3rd. Orlando, FL, USA: Academic Press, Inc., 2008.
- [6] G. New, *Introduction to Nonlinear Optics*. Cambridge University Press, 2011.
- [7] C. Cohen-Tannoudji, B. Diu, and F. Laloë, *Mécanique quantique*, Hermann, Ed. Paris: 1973, vol. 1.
- [8] *Course of Lecture on Natural Philosophy and the mechanical arts*. Joseph Johnson, 1807.
- [9] G. Grynberg, A. Aspect, and C. Fabre, *Introduction to quantum optics: from the semi-classical approach to quantized light*. New York, NY: Cambridge Univ. Press, 2010.