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Not peer-reviewed version

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Posted Date: 23 July 2024

doi: 10.20944/preprints202407.1771.v1

Keywords: nonlinear optics
absorption of electromagnetic energy
fluorescence
laser emission



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Article

The Dipole-Radiation Interaction Revisited

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Abstract: The phenomena of light absorption, fluorescence and laser emission are accounted for within a *unique* picture. The atomic dipole is dealt with in a *quantum* framework, whereas the electromagnetic field is assumed to be a *classical* sine-wave. The on and off resonance cases are investigated. This is achieved owing to a novel application of Poynting's theorem. Observable predictions are made, regarding absorption of the electromagnetic energy and the frequency bandwidth of light emitted either by fluorescence or laser effect.

Keywords: nonlinear optics; absorption of electromagnetic energy; fluorescence; laser emission

PACS: 42.50.Md, 42.50.Gy

1. Introduction

Though the absorption of light and its emission either by fluorescence or laser effect, all result merely from the resonant interaction of atomic or molecular dipoles with the electromagnetic field [1–3], their respective theoretical accounts are yet quite different. Absorption is analysed with help of the Bloch equations [4], while fluorescence and laser effect are believed to stem respectively from spontaneous and stimulated emission [2] of *incoherent* photons. However such an empirical approach is not even self-consistent, because there is no one to one correspondence between the amplitude of the *coherent* field and the photon number. Therefore this work is aimed at devising a single framework, accounting consistently for light absorption, fluorescence and laser emission, as well. The traditional treatment of absorption will be taken advantage of, as a starting point. It consists [4] of solving the Schrödinger equation, governing the motion of the dipole coupled to the field, and then using the Bloch equations to take into account the irreversible effects, stemming from dipole-lattice relaxation and pumping. Since the field amplitude is given in an absorption experiment, this scheme cannot cope with the light amplification issue, for which the time-dependence of the field amplitude is conversely *unknown*. Nevertheless, by capitalising on a previous work [5], Poynting's theorem [3] will be used to derive the additional equation, needed to assess the unknown time behaviour of the electromagnetic field.

The outline is as follows : the Hamiltonian is presented in section II, while a relationship of prominent significance between the field amplitude and dipolar coordinates is worked out in section III; absorption is addressed in section IV, whereas the master equation, accounting for field amplification, is derived in section V and further applied to the study of fluorescence and laser emission in sections VI,VII; the main results are summarised in the conclusion.

2. The Hamiltonian

Let us consider a set of identical dipoles in concentration ρ , embedded in a transparent medium of refractive index n . Each of them comprises a ground state $|g\rangle$ and an excited one $|e\rangle$, separated by the energy $\hbar(\omega + \delta\omega)$ with $\left|\frac{\delta\omega}{\omega}\right| \ll 1$. They interact with an incoming electromagnetic wave of frequency ω , propagating along the z axis with phase velocity $\frac{c}{n}$ (c stands for the light velocity in vacuum). Its electrical component $E(t, z)$, parallel to the x direction, reads

$$E(t, z) = E_x(t) \cos(\omega t - kz) \quad ,$$

wherein there is $\omega = \frac{c}{n}k$ and $\left|\frac{dE_x}{E_x dt}\right| \ll \omega$ has been assumed.

The time behaviour of a dipole, located at $z = 0$, is expressed by the wave-function $\psi(t)$ ($\langle\psi|\psi\rangle = 1$), solution of the Schrödinger equation

$$\begin{aligned} i\frac{d\psi}{dt} &= H\psi \\ H &= -(\omega + \delta\omega)\sigma_z + \frac{\mu E_x(t)}{\hbar} \cos(\omega t)\sigma_x \end{aligned} \quad (1)$$

The Hamiltonian H has been written in frequency unit and $\mu, \sigma_{i=x,y,z}$ refer respectively to the off-diagonal dipolar matrix element and Pauli's matrices [4], subspanned by the basis $\{|g\rangle, |e\rangle\}$.

It is convenient to apply Rabi's unitary transformation [4,5]

$$R = e^{\frac{i\omega t}{2}}|e\rangle\langle e| + e^{-\frac{i\omega t}{2}}|g\rangle\langle g| \quad ,$$

to $\psi(t)$, which yields the following Schrödinger equation for $\psi_R = R^{-1}\psi$

$$\begin{aligned} i\frac{d\psi_R}{dt} &= H_R\psi_R, \quad H_R = R^{-1}\left(HR - i\frac{dR}{dt}\right) \Rightarrow \\ H_R &= -\delta\omega\sigma_z + \omega_X\sigma_X \end{aligned} \quad (2)$$

with $\omega_X(t) = \frac{\mu E_x(t)}{2\hbar}$ and $\langle\psi_R|\psi_R\rangle = 1$. Terms, showing up in the expression of H_R and oscillating like $\cos(2\omega t)$, are realized to be negligible [4] due to $\left|\frac{\delta\omega}{\omega}\right| \ll 1$ and have thence been discarded. Since the same expression of H_R can be obtained by applying to H a classical rotation around the z axis with angular velocity ω , H_R can be viewed [4,5] as the expression of H in the rotating frame X, Y, z . As a prominent advantage, H_R , unlike H , is seen to include no explicit time dependence, embodied by $\cos(\omega t)$ in Eq.(1), so that solving Eq.(2) for the unknown ψ_R is easy.

To that end, let us introduce [5] the vector $S(t)$, having three dimensionless components

$$S_{j=X,Y,z} = 2\langle\psi_R|\sigma_j|\psi_R\rangle \Rightarrow -1 \leq S_j \leq 1 \quad .$$

In this representation the components of the electric polarization density along the X and Y directions may be expressed as $\frac{\rho\mu}{2}S_{j=X,Y}$, whereas S_z represents the population difference between $|g\rangle$ and $|e\rangle$. Accordingly, $S_z = 1$ ($= -1$) corresponds to $|g\rangle$ ($|e\rangle$) occupied and $|e\rangle$ ($|g\rangle$) empty. A one component vector S can also be defined in the laboratory frame as

$$\begin{aligned} S_x &= 2\langle\psi|\sigma_x|\psi\rangle \Rightarrow \\ S_x &= \cos(\omega t)S_X - \sin(\omega t)S_Y \end{aligned} \quad (3)$$

At last, Eq.(2) can be recast as

$$\begin{aligned} i\frac{d\psi_R}{dt} &= H_R\psi_R \Leftrightarrow \\ \frac{dS_j}{dt} &= 2i\langle\psi_R|[H_R, \sigma_{j=X,Y,z}]\psi_R\rangle \Leftrightarrow \\ \left\{ \begin{aligned} \frac{dS_X}{dt} &= \delta\omega S_Y \\ \frac{dS_Y}{dt} &= -\delta\omega S_X - \omega_X(t)S_z \\ \frac{dS_z}{dt} &= \omega_X(t)S_Y \end{aligned} \right. \end{aligned} \quad (4)$$

Eq.(4) turns out to be identical to the classical equation of motion for S in the X, Y, z frame [4]

$$\frac{dS}{dt} = \Omega \times S$$

with $\Omega_X = \omega_X, \Omega_Y = 0, \Omega_z = -\delta\omega$ being the components of the vector Ω . Though Eqs.(2,4) are fully equivalent, Eq.(4), unlike Eq.(2) as seen elsewhere [6], enables one to cope with irreversible effects, such as dipole-lattice relaxation or pumping. However solving Eq.(4) requires the knowledge of $\omega_X(t)$, which will be achieved below by turning Poynting's theorem into an energy conservation law.

3. Poynting's Theorem

We begin with recalling the expression of Poynting's theorem [3] in a non-magnetic insulator

$$\frac{\partial}{\partial t} \left(\frac{\epsilon_0 n^2 E^2 + \mu_0 H^2}{2} \right) = - \left(E \frac{\partial P_x}{\partial t} + \nabla \cdot F \right) ,$$

wherein ϵ_0, μ_0 designate the electrical permittivity and magnetic permeability of vacuum ($\Rightarrow \epsilon_0 \mu_0 c^2 = 1$) and H refers to the magnetic component of the electromagnetic field, parallel to the y axis, whereas $F = E \times H$ and P_x stand respectively for Poynting's vector and the electric polarization density along the x direction. Likewise we have used the relationship $D = \epsilon_0 n^2 E + P_x$ with D being the electric displacement along the x direction. Besides, E, H are related together through Faraday's law, which implies $H(t, z = 0) = \frac{n}{\mu_0 c} E(t, z = 0)$, so that Poynting's theorem is recast as

$$\epsilon_0 n^2 \frac{\partial E^2}{\partial t} = - \left(E \frac{\partial P_x}{\partial t} + \nabla \cdot F \right) . \quad (5)$$

To get rid of terms oscillating with frequency 2ω , let us average Eq.(5) over one period $\frac{2\pi}{\omega}$, which yields

$$\begin{aligned} A(t) &= \frac{\epsilon_0 n^2 \omega}{2\pi} \int_{t-\frac{\pi}{\omega}}^{t+\frac{\pi}{\omega}} E_x^2(u) \cos^2(\omega u) du \\ &= \frac{\epsilon_0 n^2}{2} E_x^2(t) \Rightarrow \\ \frac{dA}{dt} &= \epsilon_0 n^2 \frac{dE_x}{dt} E_x = \\ &= -\frac{\omega}{2\pi} \int_{t-\frac{\pi}{\omega}}^{t+\frac{\pi}{\omega}} \left(E(u) \frac{\partial P_x}{\partial u} + \nabla \cdot F(u) \right) du \end{aligned} \quad (6)$$

Let us now reckon the average value of $\nabla \cdot F$

$$\begin{aligned} F &= \frac{n}{\mu_0 c} (E_x(t) \cos(\omega t))^2 \Rightarrow \\ \nabla \cdot F &= \frac{nk}{\mu_0 c} E_x^2(t) \sin(2\omega t) \Rightarrow \\ \int_{t-\frac{\pi}{\omega}}^{t+\frac{\pi}{\omega}} \nabla \cdot F(u) du &= 0 \end{aligned} \quad (7)$$

It ensues finally from Eqs.(6,7)

$$\frac{dA}{dt}(t) = -\frac{\omega}{2\pi} \int_{t-\frac{\pi}{\omega}}^{t+\frac{\pi}{\omega}} E(u) \frac{\partial P_x}{\partial u} du . \quad (8)$$

Since A is recognised to represent an energy density, Eq.(8) means that the variation of electromagnetic energy dA over a time-duration $dt \gg \frac{2\pi}{\omega}$ inside a unit volume is equal to the work $dW = -EdP_x$, exchanged between the dipoles and the electromagnetic wave during dt . Thus $dA > 0, dA < 0$ are seen to correspond respectively to amplification or absorption of the electromagnetic field, whence it is concluded that amplification and absorption cannot take place *simultaneously*. Hence Eq.(8) illustrates indeed the significance of Poynting's theorem as an energy conservation law.

Proceeding further by calculating dW/dt with help of Eq.(3) leads to

$$\begin{aligned} P_x &= \frac{\rho \mu}{2} S_x = \frac{\rho \mu}{2} (\cos(\omega t) S_X - \sin(\omega t) S_Y) \Rightarrow \\ \frac{\omega}{2\pi} \int_{t-\frac{\pi}{\omega}}^{t+\frac{\pi}{\omega}} E(u) \frac{\partial P_x}{\partial u} du &= \frac{\rho \mu}{4} E_x \left(\frac{dS_X}{dt} - \omega S_Y \right) \end{aligned} \quad (9)$$

It is inferred finally from Eqs.(8,9)

$$\begin{aligned} \frac{dE_x}{dt} &= \frac{\rho \mu \omega}{4\epsilon_0 n^2} \left(S_Y - \frac{dS_X}{\omega dt} \right) \Rightarrow \\ \frac{d\omega_X}{dt} &= \omega_R^2 \left(S_Y - \frac{dS_X}{\omega dt} \right) , \end{aligned} \quad (10)$$

with $\omega_R = \frac{\mu}{n} \sqrt{\frac{\rho\omega}{8\hbar\epsilon_0}}$. Taking the assignments $n = 1.5, \rho = 10^{26}/m^3, \mu = 1D, \omega = 10^{15}\text{Hz}$ yields $\omega_R \approx 10^{13}\text{Hz}$.

Eq.(10) is the main achievement of this work, insomuch as it provides the first accurate and consistent description of energy exchange between an atomic or molecular dipole and an electromagnetic wave. However it is not appropriate in case of a *single* dipole coupled to a *weak* electromagnetic field, which requires [7,8] conversely to deal with the electromagnetic field as a set of *coherent* photons and thence to substitute the Jaynes-Cummings Hamiltonian to H_R . At last, Eq.(10) enables us to solve Eq.(4) in case of time-dependent $\omega_X(t)$

$$\begin{aligned} \frac{dS_X}{dt} &= \delta\omega S_Y \\ \frac{dS_Y}{dt} &= -\delta\omega S_X - \omega_X S_Z \\ \frac{dS_Z}{dt} &= \omega_X S_Y \\ \frac{d\omega_X}{dt} &= \omega_R^2 \left(S_Y - \frac{dS_X}{\omega dt} \right) = \omega_R^2 S_Y \left(1 - \frac{\delta\omega}{\omega} \right) \end{aligned} \quad (11)$$

Eq.(11) is seen to consist in a system of differential equations, describing the time behaviour of a dipole coupled to an electromagnetic field in case of negligible dipole-lattice relaxation and pumping. Its solution $S_{j=X,Y,Z}(t), \omega_X(t)$ is found [5] to be time-periodic and boils down to a pendulum equation for $\delta\omega = 0$. This is in contrast with a *single* atomic dipole coupled to a *weak* field [7,8], for which $S_{j=X,Y,Z}(t), \omega_X(t)$ are conversely aperiodic. In addition, due to $\left| \frac{\delta\omega}{\omega} \right| \ll 1$, it is inferred from the expression of $\frac{d\omega_X}{dt}$ in Eq.(11) that field absorption $\left(\frac{dA}{dt} < 0 \right)$ and amplification $\left(\frac{dA}{dt} > 0 \right)$ correspond respectively to $S_Y > 0$ and $S_Y < 0$. Moreover the master equations, accounting for absorption, fluorescence and laser emission, to be discussed below, will be derived, merely by complementing Eq.(11) with additional terms, responsible for irreversible effects, stemming from dipole-lattice relaxation and pumping.

4. Absorption

Absorption of the electromagnetic energy by the lattice is a two-stage process [3]: first the dipole takes $\hbar\omega$ from the incoming wave to go from $|g\rangle$ up to $|e\rangle$, which is a reversible mechanism, described by Eq.(11), and then releases $\hbar\omega$ to the lattice in an irreversible manner. Since H_R exhibits no explicit time dependence in the X, Y, Z frame, each dipole is assumed to reach thermal equilibrium for $t \rightarrow \infty$ in a static field Ω with $\Omega_X = \omega_X, \Omega_Y = 0, \Omega_Z = \omega$. Accordingly, the dipole relaxation toward equilibrium will be accounted [4] for, by adding

$$\frac{S(\omega) - S_Z(t)}{T_1},$$

to the right-hand side of $\frac{dS_Z}{dt}$ in Eq.(4). $S(\omega) = \tanh\left(\frac{\hbar\omega}{2k_B T}\right)$ is the thermal equilibrium value of S_Z and k_B, T stand for Boltzmann's constant and temperature, respectively, whereas the relaxation time T_1 is all the shorter, since the dipole-lattice coupling is stronger. At room temperature, there is $S(\omega) \approx 1$ for optical frequencies $\omega \approx 10^{15}\text{Hz}$.

Likewise, S_X, S_Y relaxation is accounted for, by inserting respectively

$$\frac{S(\omega_X) - S_X(t)}{T_2}, \quad -\frac{S_Y(t)}{T_2},$$

into the right-hand side of $\frac{dS_X}{dt}, \frac{dS_Y}{dt}$ in Eq.(4), with $S(\omega_X) = \tanh\left(\frac{\hbar\omega_X}{2k_B T}\right) (\approx \frac{\hbar\omega_X}{2k_B T}$ at room temperature). The relaxation time T_2 is of relevance [4], only if the inter-dipole coupling is stronger than the dipole-lattice one ($\Rightarrow T_2 \ll T_1$), which is unlikely in this context of dilute dipoles. Otherwise there is $T_2 = T_1$.

Since E_x , and thence ω_X too, remain constant for any absorption measurement ($\Rightarrow \frac{d\omega_X}{dt} = 0$), Eq.(4) turns into

$$\begin{aligned}\frac{dS_X}{dt} &= \delta\omega S_Y + \frac{S(\omega_X) - S_X}{T_2} \\ \frac{dS_Y}{dt} &= -\delta\omega S_X - \omega_X S_Z - \frac{S_Y}{T_2} \\ \frac{dS_Z}{dt} &= \omega_X S_Y + \frac{S(\omega) - S_Z}{T_1}\end{aligned} \quad (12)$$

As we are interested in the permanent regime, characterised by $dS_{j=X,Y,Z}/dt = 0$, Eq.(12) yields finally for $S_{j=Y,X,Z}^\infty = S_j(t \rightarrow \infty)$

$$\begin{aligned}S_Z^\infty &= S(\omega) + \omega_X T_1 S_Y^\infty \\ S_X^\infty &= S(\omega_X) + \delta\omega T_2 S_Y^\infty \\ S_Y^\infty &= -T_2 \frac{\delta\omega S(\omega_X) + \omega_X S(\omega)}{1 + (\delta\omega T_2)^2 + \omega_X^2 T_1 T_2}\end{aligned} \quad (13)$$

Note that there is $S_Y^\infty < 0$, as expected for absorption ($\Rightarrow \frac{dA}{dt} < 0$). Accordingly Eq.(13) entails indeed

$$\frac{dA}{dt} = \frac{\hbar\rho\omega}{2} \omega_X S_Y^\infty < 0 \quad (14)$$

Eqs.(12,13) are seen to differ from the Bloch equations, as given in textbooks [4], by $S(\omega_X) \neq 0$, which implies that the absorption curve, associated with the Bloch equations, is an even function of $\delta\omega$, unlike that ensuing from Eq.(12). Consequently $\delta A = \frac{dA}{dt}(\delta\omega) - \frac{dA}{dt}(-\delta\omega)$ should vanish for the Bloch equations, but not for Eq.(13). However, there is

$$\delta\omega S(\omega_X) + \omega_X S(\omega) = \omega_X \left(1 + \frac{\hbar\delta\omega}{2k_B T} \right)$$

with $\left| \frac{\hbar\delta\omega}{2k_B T} \right| \ll 1$ and $\omega_X T_1 \ll 1 \Rightarrow \omega_X T_2 \ll 1$ to avoid saturation effects [4], so that $|\delta A| \ll 1$ is likely to be under the detection threshold. Hence in order to overcome this hurdle and to establish whether experiment validates eventually $\delta A = 0$ or $\delta A \neq 0$, let us propose a new procedure, based on differential lock-in detection. Thus let us apply a quasi static electric field, aimed at making $\delta\omega \propto f(t)$ a periodic function of time, owing to the Stark effect, such that $f(t + t_p) = f(t)$, $f(-t) = -f(t)$, $f\left(t \in \left[0, \frac{t_p}{2}\right]\right) = 1$ and $t_p \approx 10^{-4}s \Rightarrow t_p \gg T_1$. At last, the searched $\delta A \propto S_Y^\infty(\delta\omega) - S_Y^\infty(-\delta\omega)$ is obtained as

$$\delta A \propto \int_{t-\frac{t_p}{2}}^{t+\frac{t_p}{2}} S_Y(u) \sin\left(\frac{2\pi u}{t_p}\right) du \quad ,$$

with $S_Y(t)$ being the measured signal.

5. Master Equation for Amplification

Amplification of the electromagnetic energy is achieved by exciting incoherently the electrons from $|g\rangle$ up to $|e\rangle$ thanks to pumping, which is mimicked by adding $-w_p(S_Z + 1)$ to the right-hand side of $\frac{dS_Z}{dt}$ in Eq.(12) (w_p characterises the pumping rate). Hence $\frac{dS_Z}{dt}$ reads now

$$\frac{dS_Z}{dt} = \omega_X S_Y + \frac{S(\omega) - S_Z}{T_1} - w_p(S_Z + 1) \quad .$$

It is furthermore convenient to substitute effective relaxation time and population difference T_1^*, S_p to $T_1, S(\omega)$

$$\frac{1}{T_1^*} = \frac{1}{T_1} + w_p \quad , \quad S_p(w_p) = \frac{S(\omega) - T_1 w_p}{1 + T_1 w_p} \quad ,$$

whence $S_p(w_p = 0) = S(\omega)$ and $S_p(w_p \rightarrow \infty) = -1$ are inferred to correspond to the cases of vanishing pumping, characterised by $|g\rangle$ occupied and $|e\rangle$ empty, and conversely, maximum pumping,

corresponding to $|e\rangle$ occupied and $|g\rangle$ empty, respectively. Likewise, due to $S(\omega) \approx 1$ at room temperature for optical frequencies, the signature of population inversion is $T_1 w_p > 1 \Rightarrow S_p < 0$. Thus $\frac{dS_z}{dt}$ reads finally

$$\frac{dS_z}{dt} = \omega_X S_Y + \frac{S_p - S_z}{T_1^*} \quad (15)$$

Besides, absorption of the electromagnetic energy by all non-resonant mechanisms, i.e. other than the radiation-dipole coupling at frequency ω discussed here, is accounted for by inserting $-\frac{\omega_X}{T_\phi}$ into the right-hand side of $\frac{d\omega_X}{dt}$ in Eq.(11)

$$\frac{d\omega_X}{dt} = \omega_R^2 \left(S_Y - \frac{dS_X}{\omega dt} \right) - \frac{\omega_X}{T_\phi} \quad ,$$

where $T_\phi \frac{c}{n}$ refers to the absorption length of the electromagnetic energy in the medium, containing the dipoles. This equation is finally recast into

$$\frac{d\omega_X}{dt} = \omega_R^2 \left(S_Y - \frac{dS_X}{\omega dt} - \chi \omega_X \right) \quad , \quad (16)$$

with $\chi = \frac{1}{\omega_R^2 T_\phi}$. Thus the master equation for amplification reads finally, by substituting Eq.(15) to the expression of $\frac{dS_z}{dt}$ in Eq.(12) and adding Eq.(16) to Eq.(12)

$$\begin{aligned} \frac{dS_X}{dt} &= \delta\omega S_Y + \frac{S(\omega_X) - S_X}{T_2} \\ \frac{dS_Y}{dt} &= -\delta\omega S_X - \omega_X S_z - \frac{S_Y}{T_2} \\ \frac{dS_z}{dt} &= \omega_X S_Y + \frac{S_p(w_p) - S_z}{T_1^*} \\ \frac{d\omega_X}{dt} &= \omega_R^2 \left(S_Y - \frac{dS_X}{\omega dt} - \chi \omega_X \right) \end{aligned} \quad (17)$$

The system of nonlinear differential equations in Eq.(17) will be shown below to provide the solutions $S_{j=X,Y,z}(t), \omega_X(t)$ for the fluorescence and laser regimes, each of them being associated with a particular fixed point of Eq.(17), to be studied now.

The fixed points are obtained by solving Eq.(17) in the permanent regime, i.e. in the limiting case $t \rightarrow \infty$, characterised by $dS_{j=X,Y,z}/dt = d\omega_X/dt = 0$, which yields for $S_{j=Y,X,z}^\infty = S_j(t \rightarrow \infty)$ and $\omega_X^\infty = \omega_X(t \rightarrow \infty)$

$$\begin{aligned} \left. \begin{aligned} S_X^\infty &= S(\omega_X) + \delta\omega T_2 S_Y^\infty \\ S_z^\infty &= S_p + \omega_X T_1^* S_Y^\infty \\ S_Y^\infty &= -T_2(\delta\omega S_X^\infty + \omega_X S_z^\infty) \\ S_Y^\infty &= \chi \omega_X^\infty \end{aligned} \right\} \Rightarrow \\ \frac{T_2 \left(\frac{\hbar\delta\omega}{2k_B T} + S_p \right)}{\chi \left(1 + (\delta\omega T_2)^2 + (\omega_X^\infty)^2 T_1^* T_2 \right)} &= -1 \Rightarrow \\ (\omega_X^\infty)^2 &= -\frac{S_p + \frac{\hbar\delta\omega}{2k_B T}}{\chi T_1^*} - \frac{(\delta\omega T_2)^2 + 1}{T_1^* T_2} \end{aligned} \quad (18)$$

It is inferred from the last equation (18) that the condition $(\omega_X^\infty)^2 > 0$ cannot be fulfilled, unless there is

$$S_p + \frac{\hbar\delta\omega}{2k_B T} < 0 \Rightarrow T_1^* w_p > 1 \quad ,$$

which requires finite population inversion and corresponds thence to laser emission. The inequalities $S_Y^\infty > 0$ (consistent with amplification) $\Rightarrow S_z^\infty > S_p$ are further deduced from Eq.(18) in this case.

Conversely, if there is $S_p + \frac{\hbar\delta\omega}{2k_B T} > 0 \Rightarrow T_1^* w_p < 1$, the only solution of Eq.(18) is

$$S_X^\infty = S_Y^\infty = \omega_X^\infty = 0 \quad , \quad S_z^\infty = S_p \quad ,$$

which characterises fluorescence. At last, Eq.(17) will be integrated in the following sections with the assignments $\omega = 10^{15}\text{Hz}$, $\omega_R = 10^{13}\text{Hz}$, $T_1^* = T_2 = 10^{-12}\text{s}$, $\chi = 10^{-15}\text{s}$, $T = 300\text{K}$ and by using the following initial conditions

$$S_X(t=0) = \omega_X(0) = 0, \quad S_Z(0) = S(\omega), \quad S_Y(0) \neq 0, \quad ,$$

with $S_Y(0) = 10^{-4}$ originating from thermal fluctuations.

6. Fluorescence

Eq.(17) has been solved for $S_{j=X,Y,Z}(t)$, $\omega_X(t)$ and $S_p > 0$, corresponding to fluorescence. The results, plotted in Figure 1, illustrate the transient nature of fluorescence, as they display a time behaviour like $e^{-\frac{t}{T_1}} \cos \omega_f t$ with ω_f decreasing with increasing pumping rate $S_p \rightarrow 0$ from $\omega_f \approx \omega_R$ at vanishing pumping $S_p \rightarrow 1$.

The electromagnetic energy $\varepsilon(\delta\omega)$, conveyed by the fluorescence wave-packet, reading

$$\varepsilon(\delta\omega) \propto \int_0^\infty E_X^2(\delta\omega, t) dt, \quad (19)$$

has been plotted in Figure 2 for S_p values, spanning the whole range $1 > S_p > 0$, characterising fluorescence. Noteworthy are the rather weak dependence of $\varepsilon(\delta\omega = 0)$ on S_p and the maximum value of ε showing up at $\frac{\delta\omega}{\omega} \approx 10^{-3}$ rather than $\delta\omega = 0$.

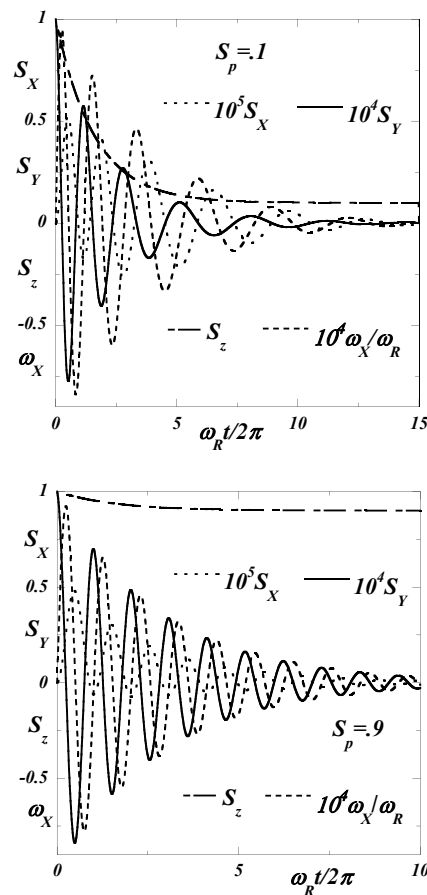


Figure 1. Plot of $S_{j=X,Y,Z}(t)$, $\omega_X(t)$, solutions of Eq.(17) for fluorescence, calculated with $\delta\omega = 0$ and two pumping rates expressed by $S_p > 0$.

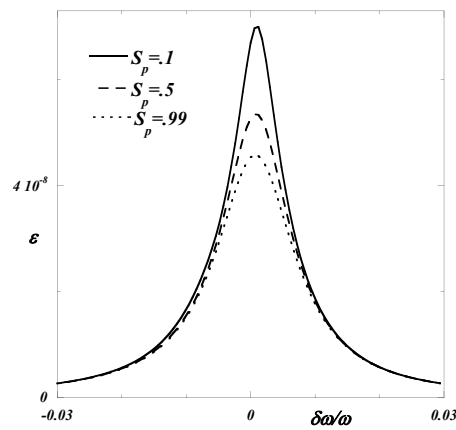


Figure 2. Plot of the electromagnetic energy $\varepsilon(\delta\omega)$, conveyed by the fluorescence wave-packet, in arbitrary units for several pumping rates expressed by $S_p > 0$.

7. Laser Effect

Eq.(17) has been solved for $S_{j=X,Y,z}(t), \omega_X(t)$ with $S_p < 0$, which is typical of laser emission. The results, given in Figure 3, exhibit $S_{j=X,Y,z}(t), \omega_X(t)$ reaching their respective asymptotic values $S_{j=X,Y,z}^\infty, \omega_X^\infty$ within a time-duration $\approx T_1^*$.

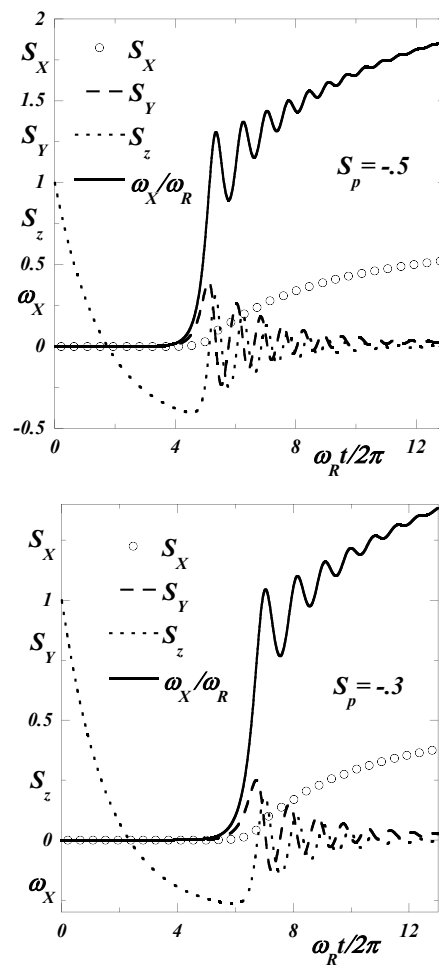


Figure 3. Plot of $S_{j=X,Y,z}(t), \omega_X(t)$, solutions of Eq.(17) for laser emission, calculated with $\delta\omega = 0$ and two pumping rates expressed by $S_p < 0$.

It is inferred from Eq.(18) for ω_X

$$\omega_X(\delta\omega) = \sqrt{\frac{1}{T_1^* T_2} \left(\frac{T_2}{\chi} \left| S_p + \frac{\hbar \delta\omega}{2k_B T} \right| - (\delta\omega T_2)^2 - 1 \right)} \quad (20)$$

The electromagnetic power $P(\delta\omega) \propto (\omega_X^\infty)^2$ has been plotted in Figure 4. P and the *finite* bandwidth of laser emission are seen to grow with the pumping rate, that is S_p decreasing from 0 toward -1 . Besides, P rises to a maximum located at $\frac{\delta\omega}{\omega} \approx -10^{-2}$. Such features lend themselves to a comparison with experimental results.

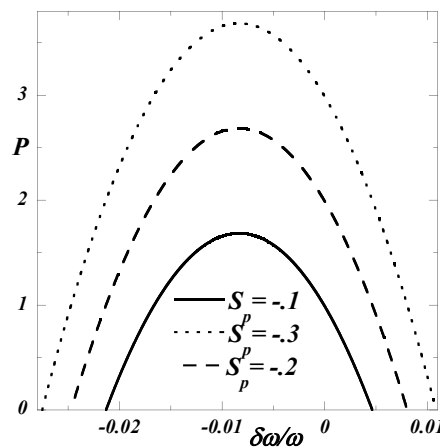


Figure 4. Plot of the electromagnetic power $P(\delta\omega)$, delivered by a laser, in arbitrary units for several pumping rates expressed by $S_p < 0$.

8. Conclusion

The Bloch equations have been slightly altered to allow for thermal relaxation of $S_X(t)$ toward $S(\omega_X) \neq 0$ and applied to the study of the absorption of electromagnetic energy by a set of interacting dipoles. Furthermore Poynting's theorem has been taken advantage of to work out an additional differential equation relating the electromagnetic field to the dipolar coordinates. This has enabled us eventually to account for fluorescence and laser effect, as solutions of a *single* system of non-linear differential equations, each of them being associated with a particular fixed point, characterised by whether there is population inversion between $|g\rangle$ and $|e\rangle$ or not. Besides, the emission bandwidth has been calculated for fluorescence and laser effect. Noteworthy is that the maximum of fluorescence and laser emission is seen to occur at $\delta\omega \neq 0$, which is likely to ensue from $S(\omega_X) \neq 0$. The validity of this analysis could be checked by comparing the results obtained here with experimental data.

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