Optik 123 (2012) 193–196

Contents lists available at ScienceDirect

Optik

journal homepage: www.elsevier.de/ijleo

# Classical interaction of the electromagnetic radiation with two-level polarizable matter

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ARTICLE INFO

*Article history:*
Received 2 August 2010
Accepted 23 October 2010

*PACS:*
41.20.−q
42.55.Ah
42.50.Ct
42.50.Pq

*Keywords:*
Matter interacting with radiation
Polarization
Lasers
Perturbation theory
Non-linear equations

ABSTRACT

The interaction of the classical electromagnetic field with an ensemble of polarizable, identical, atomic particles with two energy levels is investigated, and the coupled non-linear equations of motion for the polarization field and the amplitudes of the level occupancies are solved by a perturbation-theoretical method. A small coupling constant is identified, and the solution is represented as a power series in this coupling constant. Explicit results are given for the leading contributions to the solution. In particular, it is shown that an external electromagnetic field may induce a lasing effect in such an ensemble of particles, by populating the (initially empty) upper level.



The interaction of the classical electromagnetic radiation with an ensemble of polarizable, identical, atomic particles with two energy levels is the core of the "semi-classical theory" of the laser (see, for instance, Refs. [1–3]). The problem has been extensively investigated, by various approaches and from many angles [4–17]. Usually, the equations of motion for the electromagnetic field and the occupancies of the two levels are solved by means of some approximations which, among other particular assumptions, discard the fast oscillating terms. However, such terms may bring relevant contributions in the stationary regime. It is generally believed that an exact solution of the coupled, non-linear equations of the semi-classical theory of the laser would be impossible (see, for instance, Ref. [2], p. 459, Ref. [3], p. 98). We present here a fully computable solution, represented as a power series in a (small) coupling constant $\lambda$, and give explicit results for the polarization field, occupancy numbers and energy in the lowest, most relevant orders of $\lambda$, in the presence of an external eletromagnetic field. We show that a lasing effect can be induced in the ensemble of particles, driven by the external field which can populate the (initially empty) upper level.

We consider a uniform distribution of polarizable, identical particles, each with two quantum energy levels $\varepsilon_{0,1}$, subjected to an external electromagnetic field and to their own polarization field. The ensemble of particles exhibits a fluctuating curent density $\mathbf{j}(\mathbf{r}, t)$, and a polarization $\mathbf{P}(\mathbf{r}, t)$, related by $\mathbf{j}(\mathbf{r}, t) = \partial\mathbf{P}(\mathbf{r}, t)/\partial t$, which, in turn, give rise to a polarization field, according to the well-known wave equations with sources

$$\frac{1}{c^2}\frac{\partial^2\mathbf{A}}{\partial t^2} - \Delta\mathbf{A} = \frac{4\pi}{c}\mathbf{j}, \quad \frac{1}{c^2}\frac{\partial^2\mathbf{E}}{\partial t^2} - \Delta\mathbf{E} = -\frac{4\pi}{c^2}\frac{\partial^2\mathbf{P}}{\partial t^2}, \tag{1}$$

where $\mathbf{A}$ is the vector potential and $\mathbf{E} = -(1/c)\partial\mathbf{A}/\partial t$ is the polarization electric field (we assume a transverse radiation field). We take only one polarization, oriented along one coordinate axis, and look for a separable solution of the form $E(\mathbf{r}, t) = E(t)\chi(\mathbf{r})$, $P(\mathbf{r}, t) = P(t)\chi(\mathbf{r})$, where $\chi(\mathbf{r})$ is an eigenfunction of the laplacian, $\Delta\chi(\mathbf{r}) = -\kappa^2\chi(\mathbf{r})$, $\kappa$ being a constant. With the notation $\omega_0^2 = c^2\kappa^2$, the second equation (1) becomes

$$\ddot{E}(t) + \omega_0^2 E(t) = -4\pi\frac{\partial^2 P(t)}{\partial t^2}. \tag{2}$$

We envisage a classical polarization field $E$; consequently, the source in the *rhs* of Eq. (2) can be written as $4\pi n\langle\partial^2 p/\partial t^2\rangle$, where $p$ is the dipole momentum of a particle and the brackets denote the quantum average; the spatial average is taken into account by the (uniform) density $n$ of the ensemble of particles. We take

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0030-4026/$ – see front matter 
doi:10.1016/j.ijleo.2010.10.053

$\langle \partial^2 p/\partial t^2 \rangle = -\omega_1^2 \langle p \rangle$, where $\hbar\omega_1 = \varepsilon_1 - \varepsilon_0$. Eq. (2) can then be written as

$$\ddot{E}(t) + \omega_0^2 E(t) = 4\pi n \omega_1^2 \langle p \rangle. \quad (3)$$

In general, $\langle \partial^2 p/\partial t^2 \rangle$ depends on the internal dynamics of the particles, and can be kept as such in Eq. (3), or it may be expressed in terms of other conventional parameters. We note also that the electric field source, given generaly by $\partial j(t)/\partial t$, may not originate only in orbital currents (as we assumed here), but it may have also other origins, like the spin, for instance.

The two quantum states $\varphi_{0,1}$ are defined by the free hamiltonian $H_0$ of the internal degrees of freedom of each individual particle, $H_0\varphi_{0,1} = \varepsilon_{0,1}\varphi_{0,1}$. The interaction hamiltonian for one particle placed at $\mathbf{r}$ is given by

$$H_{int} = -p\chi(\mathbf{r})[E_0(\mathbf{r}, t) + E(\mathbf{r}, t)] = -pE_t(t)\chi^2(\mathbf{r}), \quad (4)$$

where the external field $E_0$ has been introduced, as well as the total field $E_t = E_0 + E$. We assume the fields and the (orthogonalized) eigenfunctions real. The spatial average of Eq. (4) gives an interaction hamiltonian

$$(H_{int})_{av} = -pE_t(t). \quad (5)$$

The interacting state $\varphi = c_0\varphi_0 + c_1\varphi_1$ is a superposition of the two free states $\varphi_{0,1}$, with coefficients $c_{0,1}$ satisfying the Schrodinger equation

$$i\hbar\frac{\partial c_0}{\partial t} = \varepsilon_0 c_0 - p_{01}E_t c_1, \qquad i\hbar\frac{\partial c_1}{\partial t} = \varepsilon_1 c_1 - p_{01}^* E_t c_0. \quad (6)$$

The quantum average of the dipole momentum is given by

$$\langle p \rangle = p_{01}c_0^* c_1 + p_{01}^* c_1^* c_0, \quad (7)$$

where we have assumed $p_{00} = p_{11} = 0$, as for stationary states. Moreover, we assume for simplicity $p_{01} = p_{01}^* = p$. We set $\varepsilon_0 = 0$ and introduce the parameter

$$x(t) = \frac{2p}{\hbar\omega_1}E_t(t), \quad (8)$$

so that Eq. (6) become

$$i\frac{\partial c_0}{\partial t} = -\frac{1}{2}\omega_1 x(t) c_1, \quad i\frac{\partial c_1}{\partial t} = \omega_1 c_1 - \frac{1}{2}\omega_1 x(t) c_1 \quad (9)$$

and Eq. (3) can be written as

$$\ddot{E}(t) + \omega_0^2 E(t) = 4\pi n \omega_1^2 p(c_0^* c_1 + c_1^* c_0). \quad (10)$$

In Eq. (8) we may recognize the well-known Rabi "frequency" $pE_t/\hbar$. Usually, the system of Eq. (9) is transformed into a system of equations for the occupancies $|c_{0,1}|^2$ and the associated matrix density [2,3]. We adopt a different route, and focus on the system of Eq. (9) for the occupancy amplitudes $c_{0,1}$.

The system of Eq. (9) can be solved formally with $c_{0,1} = C_{0,1}e^{i\theta}$; we get immediately $\dot{C}_{0,1} = 0$ and

$$c_0 = C_0 e^{i\theta_0} - fC_1 e^{i\theta_1}, \quad c_1 = fC_0 e^{i\theta_0} + C_1 e^{i\theta_1}, \qquad \dot{\theta}_{0,1} = \frac{1}{2}\omega_1(-1 \pm \sqrt{x^2(t) + 1}), \quad (11)$$

where

$$f(t) = \frac{x(t)}{\sqrt{x^2(t) + 1} + 1}. \quad (12)$$

The coefficients $C_{0,1}$ are determined by requiring the initial values of the occupancy numbers $|c_{0,1}(t = 0)|^2$ be equal with $n_{0,1}$ ($n_0 + n_1 = 1$). We get the amplitudes

$$C_{0,1} = \frac{1}{1 + f^2(t)}[\sqrt{n_{0,1}} \pm f(t)\sqrt{n_{1,0}}] \quad (13)$$

and the occupancy numbers

$$|c_{0,1}|^2 = n_{0,1} \pm \frac{1}{2}\frac{x(t)}{x^2(t) + 1}[2\sqrt{n_0 n_1} - x(t)(n_0 - n_1)] \times [1 - \cos(\theta_0 - \theta_1)], \quad (14)$$

where the phase difference $\theta_0 - \theta_1$ is given by

$$\Delta\theta = \theta_0 - \theta_1 = \omega_1 \int_0^t dt \sqrt{x^2(t) + 1}. \quad (15)$$

The oscillations of the occupancies given by Eq. (14) are reminiscent of the well-known Rabi oscillations, exhibited, for instance, by the Jaynes–Cummings model (see, for instance, Refs. [4,15]). We take the time averages of all the relevant quantities given above. We can see, by Eq. (11), that the energy levels $\varepsilon_{0,1}$ are changed by interaction into the mean values of $\hbar\dot{\theta}_{0,1}$, and, in addition, the interaction mixes up the two states, as expected. We can see also that the mean values of the coefficients $C_{0,1}$, as well as the mean values of the coefficients $fC_{0,1}$ entering Eq. (11), are constants, as it is required by a stationary solution; it becomes apparent that $n_{0,1}$ are constants of integration.

From Eqs. (11)–(13) we get

$$c_0^* c_1 + c_1^* c_0 = \frac{1}{x^2 + 1}\{x[2x\sqrt{n_0 n_1} + n_0 - n_1] + [2\sqrt{n_0 n_1} - x(n_0 - n_1)]\cos\Delta\theta\}, \quad (16)$$

which can be inserted into Eq. (10); we can add the external field $E_0$, which satisfies the free wave equation $\ddot{E}_0 + \omega_0^2 E_0 = 0$, such that Eq. (10) becomes

$$\ddot{x} + \omega_0^2 x = \lambda^2\omega_1^2 \frac{1}{x^2 + 1}\{x[2x\sqrt{n_0 n_1} + n_0 - n_1] + [2\sqrt{n_0 n_1} - x(n_0 - n_1)]\cos\Delta\theta\}, \quad (17)$$

where $\lambda^2 = 8\pi n p^2/\hbar\omega_1$. We note that $x = \lambda E_t/e$, where $e = \sqrt{2\pi n\hbar\omega_1}$ is a characteristic electric field.

Eq. (17) is a non-linear (integro-differential) equation. We assume $\lambda \ll 1$, and seek the solution as a power series in $\lambda$,

$$x = \lambda x_0 + \lambda^2 x_1 + \lambda^3 x_2 + \ldots, \quad (18)$$

where $x_0 = B\cos\tilde{\omega}_0 t$, $B = E_0/e$ and $\tilde{\omega}_0$ remains to be determined. We get straightforwardly

$$x_1 = 2\sqrt{n_0 n_1}\frac{\omega_1^2}{\omega_0^2 - \omega_1^2}\cos\tilde{\omega}_1 t, \qquad x_2 = \frac{1}{2}(n_0 - n_1)B\left[\frac{\omega_1}{2\omega_0 + \omega_1}\cos(\tilde{\omega}_0 + \tilde{\omega}_1)t - \frac{\omega_1}{2\omega_0 - \omega_1}\cos(\tilde{\omega}_0 - \tilde{\omega}_1)t\right], \quad (19)$$

where

$$\tilde{\omega}_0 = \omega_0 - \lambda^2\frac{\omega_1^2}{2\omega_0}(n_0 - n_1), \quad \tilde{\omega}_1 = \omega_1\left(1 + \frac{1}{4}\lambda^2 B^2\right), \quad (20)$$

for $\omega_1 \neq \omega_0$, $\pm 2\omega_0$. These restrictions can be related to the parametric resonances $2\omega_0 \simeq n\omega_1$ (where $n \neq 0$ is any integer), occurring for an associated Mathieu equation which is a close representation of the linearized form of Eq. (17) for $n_1 = 0$ (though not a fully correct approximation to Eq. (17)) [18]. Leaving aside the (weak) frequency renormalization, the resonances exhibited by Eq. (19) are in fact what we may expect from a non-linear oscillator with the basic frequency $\omega_0$ subjected to an external force of frequency $\omega_1$. As it is well known, such an oscillator exhibits the combined-frequency phenomenon, as reflected in the occurrence of frequencies of the form $\omega_0 \pm \omega_1$ and denominators $2\omega_0 \pm \omega_1$, etc. (arising from terms like $\omega_0^2 - (\omega_0 \pm \omega_1)^2$).

We can see that the interaction renormalizes both the field frequency $\omega_0$ and the characteristic frequency $\omega_1$ of the ensemble of particles. The term $x_1$ represents the oscillations of the ensemble of particles (for $n_{0,1} \neq 0$); the effect of the external field appears only in the next order (the term $x_2$), with combined frequencies $\tilde{\omega}_0 \pm \tilde{\omega}_1$. For $n_{1,0} = 0$ the polarization process is governed entirely by the external field, as expected (and the constraint $\omega_0 \neq \omega_1$ is removed).

Having known the parameter $x(t)$, the mean values (time averages) of all the relevant quantities can be computed, as given by Eqs. (11)–(15). We get, for instance, the frequencies

$$\Omega_0 = \bar{\dot{\theta}}_0 = \frac{1}{8}\lambda^2\omega_1 B^2, \quad \Omega_1 = \bar{\dot{\theta}}_1 = -\omega_1 - \frac{1}{8}\lambda^2\omega_1 B^2 \tag{21}$$

and the mean occupancies

$$\overline{|c_{0,1}|^2} = n_{0,1} \mp \lambda^2 n_0 n_1 \frac{\omega_1^2}{\omega_0^2 - \omega_1^2} \mp \frac{1}{4}\lambda^2(n_0 - n_1)B^2. \tag{22}$$

One can see that the external field can pump, or deplete, the upper level, depending on the parameters $n_{0,1}$ and $\omega_{0,1}$. Particularly interesting is the case $n_1 = 0$ (corresponding to an upper level which is empty at the initial moment $t = 0$). In this case, the occupancy of the upper level is given by

$$\overline{|c_1|^2} = \frac{1}{4}\lambda^2 B^2 = \frac{1}{4e^2}\lambda^2 E_0^2 = \left(\frac{pE_0}{\hbar\omega_1}\right)^2; \tag{23}$$

the external field leads to a macroscopic occupation of this level. The release of the corresponding energy $E_s = \hbar\omega_1\overline{|c_1|^2}$ (per particle) is a lasing effect, driven by the external field. We note in Eq. (23) the occurrence of the Rabi frequency $pE_0/\hbar$.

The polarization can be computed by making use of the solution $x(t)$ given here (Eq. (19)) in Eq. (7) and (16). Within this approximation, the polarization is a complicate function, involving quadratic dependence on the strength of the external field, frequency doubling, combined frequencies, etc., as expected for a non-linear equation. We collect here a few relevant terms:

$$\begin{aligned} &c_0^*c_1 + c_1^*c_0 = 2\sqrt{n_0 n_1}\cos\tilde{\omega}_1 t + \lambda B(n_0 - n_1)(1 - \cos\tilde{\omega}_1 t)\cos\tilde{\omega}_0 t - \\ &+2\lambda^2\sqrt{n_0 n_1}\left[B^2\cos^2\tilde{\omega}_0 t - (n_0 - n_1)\frac{\omega_1^2}{\omega_0^2 - \omega_1^2}\cos^2\tilde{\omega}_1 t\right] - \\ &-\lambda^3\frac{\omega_1 B^2}{16\omega_0}(n_0 - n_1)\sin\tilde{\omega}_0 t\sin\tilde{\omega}_1 t. \end{aligned} \tag{24}$$

The mean value of the polarization is given by

$$\bar{P} = np\overline{(c_0c_1^* + c_1c_0^*)} = \lambda^2 np\sqrt{n_0 n_1}\left[B^2 - (n_0 - n_1)\frac{\omega_1^2}{\omega_0^2 - \omega_1^2}\right], \tag{25}$$

where the quadratic dependence on the external field is to be noted. It is worth noting that $\bar{P} = 0$ for $n_{0,1} = 0$. The $\tilde{\omega}_0$-component of the polarization gives the permittiviy $\kappa = (2np^2/\hbar\omega_1)(n_0 - n_1)$ and the particle polarizability $\alpha = 2p^2/\hbar\omega_1$ (for $n_1 = 0$). We can see that the coupling constant $\lambda = \sqrt{8\pi np^2/\hbar\omega_1}$ can be related to the polarizability $\alpha$ through $\lambda^2 = 4\pi n\alpha$. It follows that we are justified in assuming $\lambda \ll 1$ as far as the polarizability per unit volume is small, which is a usual situation.

By Eq. (18), the total electric field can be represented as a $E_t = e(x_0 + \lambda x_1 + \lambda^2 x_2 + \ldots)$. Hence, we can obtain the vector potential $A_t(t) = -c\int_0^t dtE_t$ and the magnetic field $\mathbf{H} = curl\mathbf{A}_t(\mathbf{r}, t)$ (where we take into account the transversality condition $div\mathbf{A}_t = 0$). It is then easy to compute the mean value of the total field energy density (per particle) $E_f^t$. We get

$$E_f^t = \frac{1}{8\pi n}E_0^2 + \frac{\lambda^2}{4\pi n}\left[n_0 n_1\frac{\omega_1^2(\omega_0^2 + \omega_1^2)}{(\omega_0^2 - \omega_1^2)^2}e^2 + (n_0 - n_1)\frac{\omega_1^2}{4\omega_0^2}E_0^2\right]. \tag{26}$$

From Eqs. (5), (7) and (16) we can compute the interaction energy (per particle)

$$E_{int} = -\langle p\rangle E_t = -\frac{1}{2}\hbar\omega_1 x(c_0^*c_1 + c_1^*c_0) \tag{27}$$

and its mean value

$$\bar{E}_{int} = -\lambda^2\hbar\omega_1\left[n_0 n_1\frac{\omega_1^2}{\omega_0^2 - \omega_1^2} + \frac{1}{4}(n_0 - n_1)B^2\right]. \tag{28}$$

As it was said above, the energy of the ensemble of particles (per particle) is given by

$$E_s = \hbar\omega_1\overline{|c_1|^2} = \hbar\omega_1\left[n_1 + \lambda^2 n_0 n_1\frac{\omega_1^2}{\omega_0^2 - \omega_1^2} + \frac{1}{4}\lambda^2(n_0 - n_1)B^2\right]. \tag{29}$$

Particularly interesting are these equations for $n_1 = 0$:

$$\begin{aligned} E_f^t &= E_f^0\left(1 + \lambda^2\frac{\omega_1^2}{2\omega_0^2}\right), \\ E_s &= -\bar{E}_{int} = \frac{1}{4}\hbar\omega_1\lambda^2 B^2 = \lambda^2 E_f^0, \end{aligned} \tag{30}$$

where $E_f^0 = E_0^2/8\pi n$ is the energy (per particle) of the (bare) external field. One can see that the total energy $E_t = E_f^t + E_s + \bar{E}_{int}$ reduces to the total field energy $E_f^t$, the polarization energy ($E_s$) being entirely compensated by the interaction energy ($\bar{E}_{int}$), as expected. The efficiency quotient of this lasing process is $\simeq\lambda^2$.

In conclusion, after setting up the equations of motion for the polarization field and the amplitudes of the level occupancy for an ensemble of identical, polarizable particles with two energy levels interacting with a classical electromagnetic radiation, we have identified a small coupling constant $\lambda$ which allows the solution of these coupled non-linear equations to be obtained by a theoretical-perturbation method. The solution is represented as a power series in $\lambda$. The coupling constant $\lambda$ is related to the polarizaton $n\alpha$ per unit volume of the ensemble of particles, $\lambda = \sqrt{4\pi n\alpha}$. Explicit results have been given for the leading contributions to the polarization field, occupancy numbers and energy in the presence of an external field. It was shown that an external field can induce a lasing effect, by pumping the upper energy level which was empty at the initial moment of time.

Finally, it is worthwhile commenting upon the absence of the external field in the problem presented in this paper. The equations of motion (3) and (6) have a completely different solution in this case. These equations can be solved exactly, and the solution, which is independent of time and exists only for $\lambda$ exceeding a critical value, is non-analytic in the coupling constant $\lambda$. For a strong enough coupling the ensemble of particles is unstable, and it undergoes a phase transition to a state known as a super-radiance state. Beside such a criticality condition upon the coupling constant, for finite temperatures the super-radiance (second-order) phase transition occurs below a certain critical temperature. The exact solution was obtained in a previous paper [19], where the super-radiance transition was also characterized, under the assumption of a coherent interaction of matter with radiation. It was shown there that the two energy levels and the photon states are macroscopically occupied, leading to a spatially long-range ordered state which involves the quantum phases of the internal motion of the particles. The transition process is initiated, and carried on, for $\omega_0 = \omega_1$, as corresponding to quantum transitions. In the classical case the condition of coherent interaction is implicitly included. The solution of the classical equations of motion (3) and (6) in the absence of the external field is identical with the solution obtained in Ref. [19] within the coherent interaction theory.

## Acknowledgements

The author is indebted to the Romanian-European Governmental Research Project “Extreme Light Infrastructure” (ELI) for giving him the opportunity of studying this problem.

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