



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

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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.

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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 λ , and give explicit results for the polarization field, occupancy numbers and energy in the lowest, most relevant orders of λ , in the presence of an external electromagnetic 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 current 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

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

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, $\Delta \chi(\mathbf{r}) = -\kappa^2 \chi(\mathbf{r})$, κ being a constant. With the notation $\omega_0^2 = c^2 \kappa^2$, the second equation (1) becomes

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

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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$\langle \partial^2 p / \partial t^2 \rangle = -\omega_1^2 \langle p \rangle$, 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 generally 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

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

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

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

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

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

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

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

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

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

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

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

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

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

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 ω_0 subjected to an external force of frequency ω_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 ω_0 and the characteristic frequency ω_1 of the ensemble of particles. The term x_1 represents the oscillations of the ensemble

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