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RESONANCE INTERACTION OF RADIATION WITH A MEDIUM

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Resonance interaction of radiation with a medium as determined by collective processes is considered. It is shown that for small radiation energies the interaction results in a modulation of the electromagnetic field. The de-excitation time of the non-equilibrium medium due to collective processes is also determined.

THE resonance interaction of radiation with a medium is usually described with the help of the dielectric permittivity ε. This refers also to the case of a medium with negative absorption coefficient, where ε, averaged over the oscillation frequency of the field, depends on the field energy as on a parameter.[1,2] In calculating the reaction of the medium to the radiation (transition probabilities) it is assumed that the time dependence of the electromagnetic field is given by $e^{i\omega t}$ with a constant field amplitude. This approximation is justified, however, only for a strong electromagnetic field when the energy of emission is large compared to the excitation energy of the medium, i.e., when

$$\gamma = \mathbf{E}^2 / 4\pi N\omega_0 \gg 1. \tag{1}$$

Here ω_0 is the transition frequency, N the density of atoms in the states between which the transition takes place, and $c = \hbar = 1$. In the opposite case, $\gamma \lesssim 1$, the change of the radiation energy during the interaction with the medium must be taken into account. In this case it is no longer possible to obtain a closed expression for ε, and one must consider a self-consistent solution of Maxwell's equations and the equations of motion for the medium. In the present paper such a solution is found (in a number of special cases) for arbitrary γ.

For small radiation energies the interaction with the medium leads to a modulation of the field amplitude with a characteristic frequency Ω [see formula (11) below]. It is clear that consideration of this effect is meaningful only for a medium with sufficiently weak absorption,

$$\Omega\tau \gtrsim 1, \tag{2}$$

where τ is the characteristic relaxation time. The condition $\gamma \lesssim 1$ and (2) define thus the range of parameters within which collective processes in the medium may become of importance.

In general, the proposed problem is rather complicated. To simplify the calculations, we shall therefore assume that (1) the medium is infinite and sufficiently rarefied, so that $\Omega \ll \omega_0$, (2) the medium represents a two-level system, and (3) there is no dissipation in the medium ($\tau = \infty$). The last condition implies that we are interested in the behavior of the medium and of the electromagnetic field during times small compared with τ. Treated in this way, the problem reduces to the consideration of the behavior in time of the electromagnetic field, given its initial value.

In Sec. 2 we consider the case where the electromagnetic field is a plane wave. In Sec. 3 we compute the de-excitation time of the excited medium for $\gamma \ll 1$.

1. As usual, we shall describe the behavior of the medium with the help of the density matrix ρ_{mn}. The basic equation for ρ_{mn} and the Maxwell equations have the form

$$i\partial\rho_{mn} / \partial t = \omega_{mn}\rho_{mn} + \sum_l (\mathbf{E}\mathbf{D}_{ml}\rho_{ln} - \rho_{ml}\mathbf{E}\mathbf{D}_{ln}), \tag{3}$$

$$\Delta\mathbf{E} - \frac{\partial^2\mathbf{E}}{\partial t^2} = \frac{\partial^2}{\partial t^2}\left\langle 4\pi N \sum_{m,n} \mathbf{D}_{mn}\ \rho_{nm}\right\rangle, \tag{4}$$

where $\mathbf{D}_{mn}$ is the dipole moment between the states m and n; $\langle\ldots\rangle$ means averaging over all possible orientations of the atoms.

We shall seek the electromagnetic field in the form of a superposition of plane waves with a slowly time dependent amplitude,

$$\mathbf{E}(t, \mathbf{r}) = \sum_{\mathbf{k}} \mathbf{e}_{\mathbf{k}} E_{\mathbf{k}} e^{i(\omega_{\mathbf{k}}t + \mathbf{kr})} + \text{c.c.}, \tag{5}$$

where $\mathbf{e}_{\mathbf{k}}$ is the unit polarization vector. By a slow time dependence of the amplitude we mean

$$\dot{E}_{\mathbf{k}}(t) \ll \omega_{\mathbf{k}} E_{\mathbf{k}}(t), \quad \omega_{\mathbf{k}} = |\mathbf{k}|.$$

A criterion for slowness will be given further below.

Let us introduce the following notation

$$\rho_{22}-\rho_{11}=\lambda(t),\quad \rho_{12}=\rho_{21}^{*}=\sum_{\mathbf{k}}\rho_{\mathbf{k}}(t)\,\mathbf{e}_{\mathbf{k}}\,\mathbf{d}e^{-i(\omega_{\mathbf{k}}t+\mathbf{kr})}$$
$$\mathbf{D}_{12}=\mathbf{D}_{21}=\mathbf{d}D,\quad |\mathbf{d}|=1. \tag{6}$$

Assuming that $\mathbf{D}_{11}=\mathbf{D}_{22}=0$ and averaging (3) and (4) over the high-frequency oscillations with frequency $\omega_{\mathbf{k}}\cong\omega_0$ and over the space oscillations, we obtain a system of equations for the separate harmonics ($\delta_{\mathbf{k}}=\omega_{\mathbf{k}}-\omega_0$):

$$\dot{\lambda}=\frac{4D}{3}\,\mathrm{Im}\sum_{\mathbf{k}}E_{\mathbf{k}}\rho_{\mathbf{k}}, \tag{7}$$

$$i\dot{\rho}_{\mathbf{k}}+\delta_{\mathbf{k}}\rho_{\mathbf{k}}=\lambda DE_{\mathbf{k}}^{*}, \tag{8}$$

$$i\dot{E}_{\mathbf{k}}=\frac{2\pi}{3}\,\omega_{\mathbf{k}}ND\rho_{\mathbf{k}}^{*}. \tag{9}$$

In averaging over the orientations of the atoms we have made the approximation $\langle\rho_{\mathbf{k}}|\mathbf{e}_{\mathbf{k}}\mathbf{d}|^2\rangle\cong(1/3)\rho_{\mathbf{k}}$. The average value symbol around λ and $\rho_{\mathbf{k}}$ has been omitted in (7) to (9) for brevity, as will also be done in what follows.

Equations (7) to (9) have the following first integral:

$$\lambda(t)=\lambda_0-\sum_{\mathbf{k}}\frac{|E_{\mathbf{k}}(t)|^2-|E_{\mathbf{k}}^0|^2}{\pi N\omega_{\mathbf{k}}}. \tag{10}$$

This is an adiabatic invariant of the problem under consideration. However, in the sum of this last expression only the resonance harmonics are important, for which the frequency difference is small ($\delta_{\mathbf{k}}\ll\omega_0$). Therefore, (10) practically coincides with the energy integral, i.e., the energy stored in the medium and the energy of the field are conserved in the sum:

$$\lambda(t)N\omega_0+E^2(t)/4\pi=\mathrm{const},\quad |\lambda|\leqslant 1. \tag{10$'$}$$

Eliminating $\rho_{\mathbf{k}}(t)$ from (8) and (9), we find

$$\ddot{E}_{\mathbf{k}}+i\delta_{\mathbf{k}}\dot{E}_{\mathbf{k}}-\lambda(t)\,\Omega_{\mathbf{k}}^2E_{\mathbf{k}}=0,\quad \Omega_{\mathbf{k}}^2={}^2/_3\pi ND^2\omega_{\mathbf{k}}. \tag{11}$$

We note that the characteristic value of $\Omega_{\mathbf{k}}$ in the optical region is $\Omega_{\mathbf{k}}\sim\omega_{\mathbf{k}}\sqrt{Na^3}$, where a is the linear dimension of the atom. Thus the slowness condition for the change of the field amplitude, $\Omega_{\mathbf{k}}\ll\omega_{\mathbf{k}}$, becomes $Na^3\ll 1$ ("gas approximation").

2. Let us first consider the case where the field contains only one harmonic. Leaving out the index $\mathbf{k}$ in (10) and (11) and setting $|E^0|^2/2\pi N\omega_0=\gamma_0$, we have the equation

$$\ddot{E}+i\delta\dot{E}-[\lambda_0+2\gamma_0(1-|E/E^0|^2)]\,\Omega^2E=0. \tag{12}$$

As initial conditions for $E(t)$ we take the following $E(0)=E^0$ and $\dot{E}(0)=\rho(0)=0$, i.e., the medium is initially in a mixed state. Noting that (12) is equivalent to the equation of motion for a nonlinear oscillator in a magnetic field, we find in virtue of the conservation laws for the generalized angular momentum and energy

$$d\arg E/dt={}^1/_2\delta(1-I^{-1}), \tag{13}$$

$$(dI/dt)^2+4\Omega^2(I-1)\,[\gamma_0I^2$$
$$-(\gamma_0+\lambda_0-\delta^2/4\Omega^2)\,I-\delta^2/4\Omega^2]=0, \tag{14}$$

where $I(t)=|E(t)/E^0|^2$ is the dimensionless field energy.

The roots of the second term on the left-hand side of (14) are

$$I_1=1,$$

$$I_\pm=\frac{1}{2}\left\{\gamma_0+\lambda_0-\left(\frac{\delta}{2\Omega}\right)^2\pm\left[\left(\gamma_0+\lambda_0-\frac{\delta^2}{4\Omega^2}\right)^2+\frac{\gamma_0\delta^2}{\Omega^2}\right]^{1/2}\right\}. \tag{15}$$

Since $I_\pm\gtrless 0$, the field energy $I(t)$ oscillates periodically between 1 and I_+. As is seen from (15), $I_\pm\gtrless 1$ for $\lambda_0\gtrless 0$. The case $\lambda_0<0$ corresponds to the equilibrium state of the medium, while $\lambda_0>0$ corresponds to a non-equilibrium state (inverted state): at the initial moment there are more particles on the upper level than on the lower one. Thus the electromagnetic energy decreases (increases) as a result of the interaction with a medium in equilibrium (not in equilibrium).

Let us consider some limiting cases. If the radiation energy is small, $\gamma_0\ll 1$, and the medium is in equilibrium, $\lambda_0<0$, then we find approximately from (13) and (14)

$$\lambda(t)=\lambda_0\left\{1+\frac{2\gamma_0}{|\lambda_0|+\delta^2/\Omega^2}\sin^2\alpha t\right\},$$

$$I(t)=\cos^2\alpha t+\varkappa^2\sin^2\alpha t, \tag{16}$$

$$\arg E(t)=\arg E^0+\frac{\delta}{2}\left[t-\frac{1}{\alpha\varkappa}\tan^{-1}(\varkappa\tan\alpha t)\right];$$

$$\alpha=\sqrt{\delta^2/8+|\lambda_0|\,\Omega^2/2},\qquad \varkappa=\delta/\sqrt{\delta^2+4|\lambda_0|\,\Omega^2}. \tag{17}$$

In this case the population of the levels changes slightly under the influence of the radiation. At precise resonance ($\delta=0$) the field energy $I(t)$ oscillates between 0 and 1; far from the resonance ($\delta\gg 2\sqrt{|\lambda_0|}\,\Omega$) the field energy is constant.

If the medium was initially in a non-equilibrium state ($\lambda_0>0$) and the intensity of the radiation is small ($\gamma_0\ll\lambda_0$), then the maximal energy of the field I_+ as a function of the frequency difference is approximately given by

$$I_+=\begin{cases}\lambda_0/\gamma_0-\delta^2/4\Omega^2\gamma_0, & \delta<2\sqrt{\lambda_0}\,\Omega,\\ \sqrt{\lambda_0/\gamma_0}, & \delta=2\sqrt{\lambda_0}\,\Omega,\\ \delta^2/(\delta^2-4\Omega^2\lambda_0) & \delta>2\sqrt{\lambda_0}\,\Omega.\end{cases} \tag{18}$$

As should be expected, the oscillations become essentially nonlinear for sufficiently small frequency differences. The expression for the oscillation period t_0 for $\delta = 0$ and $\gamma_0 \ll \lambda_0$ is

$$t_0 = \frac{1}{\sqrt{\gamma_0}\Omega} \int_1^{I_+} \frac{dI}{\sqrt{I(I-1)(I_+ - I)}} \simeq \frac{\ln(4\lambda_0/\gamma_0)}{\sqrt{\lambda_0}\Omega}. \quad (19)$$

We further note the special case $\delta = 0$ and $\gamma_0 = |\lambda_0|$, $\lambda_0 < 0$. Integrating (14), we find

$$\lambda(t) = |\lambda_0|\,[1 - 2\,\mathrm{ch}^{-2}(\tfrac{1}{2}\Omega\sqrt{|\lambda_0|}\,t)], \quad (20)^*$$

$$I(t) = \mathrm{ch}^{-2}(\tfrac{1}{2}\Omega\sqrt{|\lambda_0|}\,t). \quad (21)$$

This solution can be interpreted as the "spontaneous" radiation from a non-equilibrium medium $[\lambda(-\infty) > 0]$ and absorption by a medium in equilibrium $[\lambda(0) < 0]$ separated (in time) by the electromagnetic pulse. The period of the pulse is of order $(\sqrt{|\lambda_0|}\,\Omega)^{-1}$.

Finally, in the case of a strong electromagnetic field ($\gamma_0 \gg 1$, $\delta \gg \Omega$, $N \to 0$) we obtain from (14) and (10) the known result of the "single particle" approximation [3] averaged over the orientations of the atoms:

$$\lambda(t) = \lambda_0\left[1 - \frac{2|E^0D|^2}{|E^0D|^2 + 3\delta^2/4}\sin^2\left(\frac{t}{2}\sqrt{\delta^2 + \tfrac{4}{3}|E^0D|^2}\right)\right], \quad (22)$$

$$I(t) = 1 + \frac{2\pi\omega_0\lambda_0 N}{|E^0|^2 + 3\delta^2/4D^2}\sin^2\left(\frac{t}{2}\sqrt{\delta^2 + \tfrac{4}{3}|E^0D|^2}\right). \quad (23)$$

3. In this section we discuss the spontaneous radiation from a non-equilibrium medium. In the "single particle" approximation ($\gamma \gg 1$) the solution of this problem is well known. It is of interest to consider the other limiting case, when the de-excitation process is determined by the vibrational properties of the medium, i.e., $\gamma \ll 1$. The characteristic feature of this case is that the radiation which starts out as spontaneous, quickly goes over into stimulated emission owing to the large density of atoms in the medium.

Thus let us assume that at $t = 0$ the medium is in the non-equilibrium state ($\lambda_0 > 0$); the electromagnetic field is a random quantity, where the energy of the resonance harmonics, for which $|\delta_{\mathbf{k}}| < \sqrt{\lambda_0}\,\Omega_{\mathbf{k}}$, is small compared to the excitation energy of the medium. Setting $\lambda = \lambda_0$ in (11), we may obtain the energy of the resonance harmonics at the initial stage of the increase of the radiation:

$$|E_{\mathbf{k}}(t)|^2 \simeq \tfrac{1}{4}|E^0_{\mathbf{k}}|^2 \exp\left[t\Omega_{\mathbf{k}}\sqrt{2\lambda_0 - \tfrac{1}{2}(\delta_{\mathbf{k}}/\Omega_{\mathbf{k}})^2}\right]. \quad (24)$$

Here it is assumed that the exponential is large.

Let us now determine the time for the transition of the medium into the equilibrium state, t_i. It is natural to assume that for $t = t_i$ the radiation energy becomes equal to the excitation energy of the medium at $t = 0$, i.e., $\lambda(t_i) = 0$. Computing the sum in (10) by the method of steepest descent, we obtain with logarithmic accuracy

$$t_i = \ln\Lambda/\sqrt{2\lambda_0}\,\Omega, \quad \Lambda = \sqrt{\lambda_0}N/\omega_0\Omega_0 \sum_{|\mathbf{k}|=\omega_0} |E^0_{\mathbf{k}}|^2. \quad (25)$$

The summation in (25) goes over a sphere of radius ω_0. The last expression is only valid for $\Lambda \gg 1$.

If the initial electromagnetic field is given by the zero order vacuum oscillations ($|E^0_{\mathbf{k}}|^2 \sim |\mathbf{k}|$), t_i is approximately determined as

$$t_i \sim \ln(\lambda_0 N a^3)/\sqrt{\lambda_0 N a^3}\,\omega_0 \quad (26)$$

in the optical range.

In conclusion we note that the condition for the applicability of (25) and (26) in a realistic situation, where the dissipation is finite, is, of course, $t_i \ll \tau$.

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*ch = cosh.