

# Resonant interaction of two-level atoms in an electromagnetic field

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The radiative mechanism for the relaxation of two-level atoms is discussed, using the Keldysh technique. It is shown that an equilibrium state is established in strong fields as a result of radiative transitions between quasi-energy states and that complete saturation is not reached.

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To describe two-level atoms in an external field one usually uses the density-matrix formalism proposed by Lamb<sup>1</sup> together with a specified relaxation mechanism. Radiation damping is one of the possible relaxation mechanisms, and, although collisions provide the principal relaxation mechanism for real gases, it is also important to have a consistent treatment of radiative processes. However, as will be shown below, the standard density-matrix formalism, in which radiative processes are taken into account by introducing the width  $\gamma_0$  of the upper level, is valid only for rather weak fields:

$$\mathcal{E} \ll \frac{\hbar}{d} [\omega_0(\gamma_0 + |\delta|)]^{1/2}, \quad \gamma_0 = \frac{2}{3} \frac{\omega_0^3 d^2}{\hbar c^3},$$

where  $\omega_0$  is the transition frequency,  $d$  is the dipole-moment matrix element between the upper and lower levels,  $\hbar$  is Planck's constant,  $c$  is the velocity of light, and  $\delta$  is the frequency mismatch. For stronger fields one must take account of the splitting of the atomic levels in the external alternating field, for the transition probability between the levels depends on that splitting.

In this paper we give a systematic treatment of the interaction of two-level atoms with a quantized electromagnetic field within the framework of the Keldysh diagram technique<sup>2,3</sup> for the cases of weak and strong fields. In the linear approximation in the gas density, the lower atomic level can be described by a nonrelativistic field  $\hat{\varphi}$ , and the upper level, by a nonrelativistic vector field  $\hat{\chi}_j$ . In the dipole approximation, the interaction with the field is described by the Hamiltonian

$$\hat{H}_{int} = -d\mathcal{E}_j(\hat{\varphi}\hat{\chi}_j^+ + \hat{\varphi}^+\hat{\chi}_j).$$

As was shown in Ref. 4, the diagram expressions in the Keldysh technique for the equations for the Green's functions in the presence of the average field or of a Bose condensate are the same as those in the Belyaev technique<sup>5,6</sup> for a Bose liquid. Neglecting the renormalization of the vertices and limiting ourselves to the resonance approximation, we obtain the set of diagram equations shown in Fig. 1 for the Green's functions in the Keldysh technique. In the figure all the heavy lines correspond to exact Green's functions, and the thin lines to free Green's functions. The Green's function for the lower atomic level is represented graphically by an ordinary arrow, that for the upper level, by a double arrow, that for the electromagnetic field in the  $A_0 = 0$  gauge, by a dashed line, and that for the average field, by a wavy line with an asterisk at the end. The Green's function for the polarization

$$P_j = -i \langle T_K \hat{\varphi} \hat{\chi}_j^+ \rangle, \quad P_j^* = -i \langle T_K \hat{\varphi}^+ \hat{\chi}_j \rangle,$$

where the  $T_K$  are time ordered along the double contour  $K$  (Ref. 2) are represented by "forked" arrows. We shall limit ourselves to finding only stationary and spatially uniform solutions, for which it is convenient to work in the Fourier representation, since then all the Green's functions will depend only on the differences between the arguments. Such solutions exist in the resonance approximation when the external field is a traveling wave, or when the atomic system is considered in a region that is smaller than the wavelength of the field. In what follows, therefore, we shall consider only plane monochromatic waves of the form

$$\mathcal{E}_{ij} = 2\mathcal{E}_{ij}(\omega, \mathbf{k}) \cos(\omega t - \mathbf{k}\mathbf{r}), \quad \mathcal{E}_{ij} = 2\mathcal{E}_{ij}(\omega) \cos(\omega t).$$

In the case of a plane wave, only one component of the vector field of the upper level takes part in the interaction with the external electromagnetic field, so that in the case of a plane wave we can replace the vector Green's function for the upper level by the Green's function for a scalar field. After all these simplifications we can obtain an explicit solution of the diagram equations without considering the kinetic equation, which would have to be taken into account in the case of fields of a more general type. It is known that the renormalization of the retarded electromagnetic Green's function  $D_0^R$  in the medium reduces to taking the dielectric constant  $\kappa(\omega)$  into account,<sup>7</sup> and the dielectric constant can be found from the equation for the average field. The complete electromagnetic Green's function therefore has the following form:

$$D_{ij}^R(\omega, \mathbf{k}) = 4\pi \left( \delta_{ij} - \frac{k_i k_j}{\omega^2 \kappa(\omega)} \right) (\omega^2 \kappa(\omega) - \mathbf{k}^2 + i0 \text{ sign } \omega)^{-1},$$

$$D_{ij}^F(\omega, \mathbf{k}) = D_{ij}^+ + D_{ij}^- = (D_{ij}^R - D_{ij}^A) (1 + 2\tilde{d}(\omega)) \text{ sign } \omega,$$

where  $\tilde{d}(\omega)$  is the photon distribution function in the medium (here and below we use for all the intermediate calculations units in which  $\hbar = c = 1$ ).

Generally speaking, it is necessary to solve the problem of finding the Green's functions for atoms with arbitrary values of  $\kappa(\omega)$  and  $\tilde{d}(\omega)$  and then to find  $\kappa(\omega)$  and  $\tilde{d}(\omega)$  from the last two diagram equations. In this manner we can obtain self-consistent equations for  $\kappa(\omega)$  and  $\tilde{d}(\omega)$ , which can be easily solved for the region of transparency, where  $|\kappa - 1| \ll 1$ . We can then neglect the deviation of  $\kappa(\omega)$  from unity in calculating the Green's functions for the atoms. In that case, as will be shown below, the calculation yields

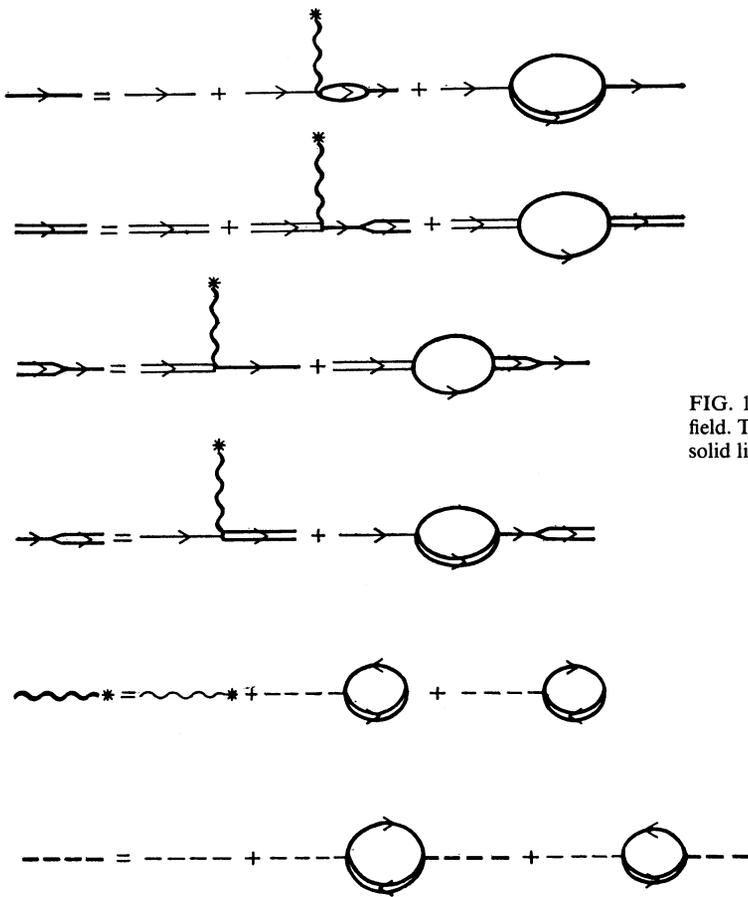


FIG. 1. A set of diagram equations for the interaction of atoms with a field. The photon Green's functions in the mass operators are denoted by solid lines.

$$\kappa - 1 = 4\pi n d^2 \left( \frac{\delta - i\gamma_0}{\delta^2 + \gamma_0^2 + 2\alpha} + \frac{3}{\omega} \right), \quad \alpha = d^2 |\mathcal{E}(\omega)|^2,$$

where  $n$  is the gas density and  $\mathcal{E}(\omega)$  is the average field. The condition  $|\kappa - 1| \ll 1$  is therefore satisfied for rarefied gases for which  $nd^2 \ll \gamma_0$ , or far from the resonance where  $nd^2 \ll |\delta|$ , or for sufficiently strong fields

$$d\mathcal{E} \gg [\omega_0(\gamma_0 + |\delta|)]^{1/2}, \quad |\kappa - 1| \sim nd^2/\omega_0 \sim na_0^3 \ll 1,$$

where  $a_0$  is the characteristic size of an atom. The last case is the most interesting, since in weak fields the approach under consideration is equivalent to the standard density-matrix formalism and yields no new results. In calculating the Green's functions for the atoms in the region of transparency, therefore, we may use a free Green's function for the electromagnetic field with a Planck distribution for the photons that arise from interactions with the thermostat. Taking the motion of the atoms into account reduces, in the density matrix formalism, to correcting  $\delta$  for the Doppler effect (if recoil is neglected), so it is sufficient to obtain a solution for the case in which the atoms are stationary in the reference system of the traveling wave and then to take the nonuniform broadening into account.

In the Keldysh technique, the Green's function for the lower level  $G$  and that for the upper level  $F$  have a matrix representation, for example

$$G = \begin{pmatrix} 0 & G^A \\ G^R & G^F \end{pmatrix},$$

where  $G^A$  and  $G^R$  are the advanced and retarded Green's functions and  $G^F = G^+ + G^-$ .<sup>2,3</sup> The mass operators  $M_1$  and  $M_2$  corresponding to the diagrams of Fig. 2 can also be represented by matrices. In the first approximation one can neglect the Lamb shift and analyze the imaginary mass operators in the form

$$M_1(E) = -i\gamma_1(E) \begin{pmatrix} 2(2g(E)+1) & 1 \\ -1 & 0 \end{pmatrix},$$

$$M_2(E) = -i\gamma_2(E) \begin{pmatrix} 2(2f(E)+1) & 1 \\ -1 & 0 \end{pmatrix}.$$

Now we have to find a solution of the algebraic set of diagram equations for arbitrary values of  $\gamma_1$ ,  $\gamma_2$ ,  $f$ , and  $g$  and calculate the mass operators from it; this enables us to obtain self-consistent equations for  $\gamma_1$ ,  $\gamma_2$ ,  $f$ , and  $g$ , whose solution enables us to find the nonlinear response of the medium to the average field. In the presence of radiation damping, the stationary solution of the algebraic set of diagram equations

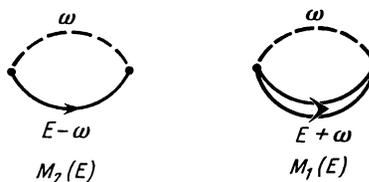


Fig. 2. The mass operators.

does not depend on the free distribution functions that occur in  $G_0^F$  and  $F_0^F$ . The solution of the diagram equations for  $F^R$  and  $G^R$  are as follows:

$$F^R(E) = \frac{E' + \delta + i\gamma_1(E' + \delta)}{[E' + \delta + i\gamma_1(E' + \delta)][E' + i\gamma_2(E')] - \alpha},$$

$$E' = E - \omega_0, \quad \delta = \omega_0 - \omega,$$

$$G^R(E) = \frac{E - \delta + i\gamma_2(E - \delta)}{[E - \delta + i\gamma_2(E - \delta)][E + i\gamma_1(E)] - \alpha},$$

$$\alpha = d^2 |\varepsilon(\omega)|^2.$$

With accuracy up to terms of the order of  $\gamma_0/\omega_0$  and  $|\delta|/\omega_0$ , these equations reduce to the form

$$F^R(E) = \frac{1}{2} \left(1 - \frac{\Delta}{s}\right) \left[E' + \frac{\delta}{2} + s + i\gamma_+\right]^{-1} + \frac{1}{2} \left(1 + \frac{\Delta}{s}\right) \times \left[E' + \frac{\delta}{2} - s + i\gamma_+\right]^{-1}, \quad \gamma_+ = \frac{\gamma_0}{2}, \quad \Delta = \frac{\delta}{2} - i\gamma_+,$$

$$G^R(E) = \frac{1}{2} \left(1 + \frac{\Delta}{s}\right) \left[E - \frac{\delta}{2} + s + i\gamma_+\right]^{-1} + \frac{1}{2} \left(1 - \frac{\Delta}{s}\right) \left[E - \frac{\delta}{2} + i\gamma_+ - s\right]^{-1}, \quad s = (\Delta^2 + \alpha)^{1/2}.$$

The solutions for  $F^F$  and  $G^F$  are as follows:

$$F^F(E) = [F^R(E) - F^A(E)] [1 + 2\tilde{f}(E)],$$

$\tilde{f}(E)$

$$= \frac{\gamma_2(E') [(E' + \delta)^2 + (\gamma_1(E' + \delta))^2] f(E) + \alpha \gamma_1(E' + \delta) g(E - \omega)}{\gamma_2(E') [(E' + \delta)^2 + (\gamma_1(E' + \delta))^2] + \alpha \gamma_1(E' + \delta)} \quad (1)$$

$$G^F(E) = [G^R(E) - G^A(E)] [1 + 2\tilde{g}(E)],$$

$\tilde{g}(E)$

$$= \frac{\gamma_1(E) [(E - \delta)^2 + (\gamma_2(E - \delta))^2] g(E) + \alpha \gamma_2(E - \delta) f(E + \omega)}{\gamma_1(E) [(E - \delta)^2 + (\gamma_2(E - \delta))^2] + \alpha \gamma_2(E - \delta)}.$$

In calculating the mass operators with these Green's functions we have to consider the problem of cutting off the integrals that arise in the calculation, since the interaction is treated in terms of a derivative and the integrals are strongly divergent. The dipole approximation is valid only when  $\omega \lesssim c/a_0$ , where  $a_0$  is the characteristic dimension of the atom, so all the integrals must be cut off at the frequency  $\omega_{\max} \sim c/a_0$ . As an example, let us show the calculation of the width of the upper level in the absence of an external field, in which, under the condition  $d \sim ea_0$ , where  $e$  is the electron charge, the integral is independent of  $\omega_{\max}$  up to terms of the order of  $x \ll 1$ :

$$\omega_{\max} \sim \frac{c}{a_0} \sim \omega_0 \left( \frac{\omega_0 e^2}{\gamma_0 \hbar c} \right)^{1/2}, \quad \gamma_0 \sim \frac{\omega_0^3 e^2 a_0^2}{\hbar c^3},$$

$$x = \left( \frac{\gamma_0 e^2}{\omega_0 \hbar c} \right)^{1/2} \ll 1,$$

$$\gamma = \frac{\gamma_0}{\omega_0^3} \int_{-\infty}^{\infty} \frac{\gamma}{\pi} \frac{\omega^3 d\omega}{(\omega - \omega_0)^2 + \gamma^2} = \frac{\gamma_0}{\omega_0^3} \int_{-\omega_{\max}}^{\omega_{\max}} d\omega \frac{\gamma}{\pi} \frac{\omega_0^3 + 3\omega_0 \omega^2}{\omega^2 + \gamma^2}.$$

The ratio of the second term to the first in the expression for  $\gamma$  is of the order  $\gamma \omega_{\max} / \omega_0^2 \sim x \ll 1$ , so  $\gamma = \gamma_0(1 + O(x))$  and with good accuracy is independent of  $\omega_{\max}$ . Since the two-level atoms are treated only in fields  $d\varepsilon \ll \omega_0$ , the integrals are calculated in the external field with the same accuracy.

Let us consider the explicit form of the self-consistency conditions, omitting the arguments for brevity:

$$-\gamma_1 \begin{pmatrix} 2(2g+1) & 1 \\ -1 & 0 \end{pmatrix} = \frac{d^2}{2} \int \frac{d^3 q}{(2\pi)^4} (F^R - F^A) (D^R - D^A) \begin{pmatrix} 2(\tilde{d} + \tilde{f} + 2\tilde{d}\tilde{f}) & \tilde{d} - \tilde{f} \\ \tilde{f} - \tilde{d} & 0 \end{pmatrix}, \quad (2)$$

$$-\gamma_2 \begin{pmatrix} 2(2f+1) & 1 \\ -1 & 0 \end{pmatrix} = \frac{d^2}{2} \int \frac{d^3 q}{(2\pi)^4} (G^R - G^A) (D^R - D^A) \times \begin{pmatrix} 2(1 + \tilde{d} + \tilde{g} + 2\tilde{d}\tilde{g}) & 1 + \tilde{d} + \tilde{g} \\ -(1 + \tilde{d} + \tilde{g}) & 0 \end{pmatrix}.$$

From the solution of the diagram equations (1) we can express  $f$  and  $g$  in terms of  $\tilde{f}$  and  $\tilde{g}$  and obtain a set of integral equations for  $\tilde{f}$  and  $\tilde{g}$ . Having chosen the left-hand sides of the self-consistency conditions (2) at the poles of the Green's functions and having calculated the integrals, which are expressed in terms of the quantities  $\tilde{f}$  and  $\tilde{g}$  at the poles of the Green's functions, we obtain a set of eight algebraic equations for the occupation numbers and widths. In the presence of the average field of frequency  $\omega$ , however, the part played by the thermal photons with the distribution function  $\tilde{d}(\omega) \ll 1$  is insignificant, so we shall first examine the solution of the self-consistency equations (2) for  $\tilde{d} = 0$  and then find the correction due to the thermal photons.

It is easy to see that when  $\tilde{d} = 0$  the solution of the self-consistency conditions yields  $f = 0$  and  $g = -1$ . There remain two equations for  $\gamma_1$  and  $\gamma_2$ , from which it is evident that  $\gamma_2 \approx \gamma_0$  and  $\gamma_1 \approx -\gamma_0 \tilde{f}$ . In the linear approximation in the occupation numbers, therefore, Eqs. (1) for the occupation numbers  $\tilde{f}$  and  $\tilde{g}$  simplify as follows:

$$\tilde{f}(E) = -\frac{\alpha \gamma_1(E' + \delta)}{\gamma_2(E') (E' + \delta)^2},$$

$$\tilde{g}(E) = -\frac{\gamma_1(E) [(E - \delta)^2 + (\gamma_2(E - \delta))^2]}{\alpha \gamma_2(E - \delta)}. \quad (3)$$

We can set  $\gamma_1 = 0$  and  $\gamma_2 = \gamma_0$  in the expressions for the retarded Green's functions for the integration. The mass operators at energies corresponding to the poles of the retarded Green's functions are calculated using the residues of the poles of the retarded Green's functions  $G^R$  and  $F^R$  with accuracy to terms of the order of  $x \ll 1$ . After the integration we obtain the self-consistency conditions in the following form:

$$\gamma_1 \left( E + \frac{\delta}{2} \right) = -\gamma_2 \left( E - \frac{\delta}{2} \right) \tilde{f} \left( E + \omega + \frac{\delta}{2} \right) \frac{(E + \delta/2)^2}{\alpha}$$

$$= -\frac{\gamma_0}{2\omega_0^3} \left[ (\omega - E + s - i\gamma_+)^3 \left( 1 + \frac{\Delta}{s} \right) \tilde{f}_+ + (\omega - E - s - i\gamma_+)^3 \right. \\ \left. \times \left( 1 - \frac{\Delta}{s} \right) \tilde{f}_- \right], \quad \tilde{f}_{\pm} = \tilde{f} \left( \omega_0 - \frac{\delta}{2} - i\gamma_{\pm} s \right),$$

$$\gamma_2 \left( E - \frac{\delta}{2} \right) = \frac{\gamma_0}{2\omega_0^3} \left[ (\omega + E - s + i\gamma_+)^3 \left( 1 - \frac{\Delta}{s} \right) + (\omega + E + s + i\gamma_+)^3 \left( 1 + \frac{\Delta}{s} \right) \right]$$

(in the resonance approximation we neglect transitions with widths

$$\gamma_0 (s^3/\omega^3) \ll \gamma_0.$$

On substituting the expression for  $\gamma_2$  into the first equation we obtain an equation for  $\tilde{f}(E)$ . Taking the values of the function  $\tilde{f}(E)$  at the poles of the retarded Green's function, we obtain the following relation between  $\tilde{f}_+$  and  $\tilde{f}_-$ :

$$\tilde{f}_+ (s + \Delta)^2 (\omega + 2s)^3 = \tilde{f}_- (s - \Delta)^2 (\omega - 2s)^3. \quad (4)$$

It is also easy to find from (3) that

$$\tilde{g}_\pm = \tilde{g} \left( \frac{\delta}{2} - i\gamma_+ \pm s \right) = \tilde{f}_\pm \left( 1 \mp \frac{4i\gamma_+}{s \mp \Delta} \right). \quad (5)$$

Calculating the diagrams in the equation for the average field yields the following values for the polarization:

$$P(\omega) = \frac{d^2}{2s} (\tilde{f}_- - \tilde{f}_+) \mathcal{E}(\omega), \quad P^*(\omega) = \frac{d^2}{2s} (\tilde{g}_- - \tilde{g}_+) \mathcal{E}^*(\omega). \quad (6)$$

After finding the occupation numbers we can neglect the difference between  $\gamma_2$  and  $\gamma_0$ , as well as that between  $\gamma_1$  and zero. The occupation numbers are obtained from the following equation for the total number of particles:

$$N = \frac{1}{2} \left( 1 + \frac{\Delta}{s} \right) (\tilde{f}_+ + \tilde{g}_-) + \frac{1}{2} \left( 1 - \frac{\Delta}{s} \right) (\tilde{f}_- + \tilde{g}_+) = \tilde{f}_+ \left( 1 - \frac{i\gamma_0}{s} \right) + \tilde{f}_- \left( 1 + \frac{i\gamma_0}{s} \right).$$

Having found the expressions for  $\tilde{f}_+$ ,  $\tilde{f}_-$ ,  $\tilde{g}_+$ , and  $\tilde{g}_-$  in the linear approximation in  $s/\omega$  from the above equations and Eqs. (4) and (5), we obtain the following expression for the polarization from (6):

$$P(\omega) = \frac{nd^2}{\hbar} \left( \frac{\delta - i\gamma_0}{\delta^2 + \gamma_0^2 + 2\alpha} + \frac{3}{\omega} \right) \mathcal{E}(\omega), \quad \alpha = \frac{d^2 |\mathcal{E}(\omega)|^2}{\hbar^2}, \quad (7)$$

$$P^*(\omega) = \frac{nd^2}{\hbar} \left( \frac{\delta + i\gamma_0}{\delta^2 + \gamma_0^2 + 2\alpha} + \frac{3}{\omega} \right) \mathcal{E}^*(\omega).$$

To simplify the notation we introduce the quantities  $\mathcal{E}_1$  and  $\mathcal{E}_2$  defined as follows:

$$d\mathcal{E}_1 = \hbar [\gamma_0 (\gamma_0 + |\delta|)]^{1/2}, \quad d\mathcal{E}_2 = \hbar [\omega_0 (\gamma_0 + |\delta|)]^{1/2}.$$

In fields  $\mathcal{E} \ll \mathcal{E}_2$ , Eq. (7) for the polarization reduces to a formula that can easily be obtained in the density-matrix formalism. However, the expression for the polarization is substantially different in fields such that  $d\mathcal{E}_2 \ll d\mathcal{E} \ll \hbar\omega_0$ . Let us consider in more detail why Eq. (7) differs from the standard density-matrix formula. For this purpose we depict the structure of the levels of a two-level atom in an external field in Fig. 3, using the solution (1) found above. As should be expected on the basis of general considerations, the spectrum of the atom in an external field has a quasi-energy form and consists of two harmonics corresponding to the quasi-

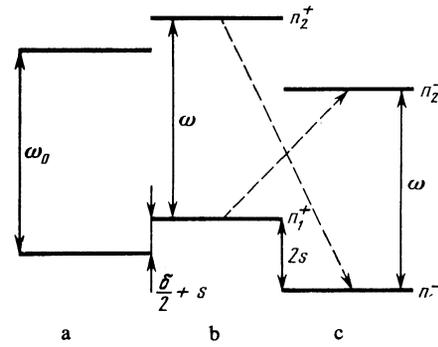


FIG. 3. Level structure of a two-level atom: a—levels of the atom in the absence of an external field; b, c— quasi-energy levels corresponding to the quasi-energies  $\delta/2 \pm s$ . A dashed arrow corresponds to a transition from the state of quasi-energy  $\delta/2 + s$  to a state of quasi-energy  $\delta/2 - s$ ; the arrow directed downward (upward) corresponds to the factor  $\tilde{d} + 1$  ( $\tilde{d}$ ), where  $\tilde{d}$  is the number of thermal photons.

energies  $\delta/2 \pm s$ . However, the numbers of particles in different harmonics corresponding to the same quasi-energy are different. The number  $n_{1,2}^\pm$  of particles on each quasi-energy harmonic is shown at the right of that harmonic in Fig. 3. These numbers may be expressed as follows in terms of the  $\tilde{f}_\pm$  and  $\tilde{g}_\pm$ :

$$n_2^\pm = \text{Re} \left[ \frac{1}{2} (1 \pm \Delta/s) \tilde{f}_\pm \right], \quad n_1^\pm = \text{Re} \left[ \frac{1}{2} (1 \mp \Delta/s) \tilde{g}_\pm \right].$$

As is evident from Eq. (5), the expressions for the occupation numbers simplify only in sufficiently strong fields  $\mathcal{E} \gg \mathcal{E}_1$ , since  $\tilde{f}_\pm \approx \tilde{g}_\pm$ , and one can even speak of a pair of quasi-energy levels, each of which is characterized by its own occupation number  $\tilde{f}_\pm$ . As before, however, different harmonics of the same quasi-energy level have different occupation numbers. The self-consistency condition is a balance condition by which transitions from the states  $\tilde{f}_+$  and  $\tilde{f}_-$  depicted in Fig. 3 by arrows are compensated by transitions from  $\tilde{f}_-$  to  $\tilde{f}_+$ . Only in the limit  $\mathcal{E} \gg \mathcal{E}_2$  do both harmonics of the quasi-energy level have the same occupation number  $\tilde{f}_\pm/2 = n_2^\pm = n_1^\pm$ , while the self-consistency condition reduces to the form

$$\tilde{f}_+ (\omega + 2s)^3 = \tilde{f}_- (\omega - 2s)^3.$$

Because the transition energies between the split levels are unequal, the transition probabilities between them are also unequal; as a result, the occupation numbers  $\tilde{f}_+$  and  $\tilde{f}_-$  of these levels are unequal and this results in the appearance of the polarization

$$P(\omega) = (3nd^2/\hbar\omega) \mathcal{E}(\omega). \quad (8)$$

We note that for any relaxation mechanism of width  $\Gamma$  in sufficiently strong fields,

$$\hbar [\omega (\Gamma + |\delta|)]^{1/2} \ll d\mathcal{E} \ll \hbar\omega_0.$$

The main contribution to the polarization will come from radiative processes, and the simple expression (8) for the polarization will be valid. The nonuniform broadening can be treated in the same way as in the density-matrix formalism, but in the case of a strong field it can be simply neglected.

Now let us consider the contribution to the polarization from thermal photons with the arbitrary distribution function  $\tilde{d}(\omega) \ll 1$ . For simplicity we shall consider only the case in which  $|\delta| \gg \gamma_0$  and  $\mathcal{E} \gg \mathcal{E}_1$ , for in that case we can simplify the Green's functions and obtain the following self-consistency condition in the linear approximation in the gas density in place of Eq. (4):

$$\begin{aligned} & \tilde{f}_+ [(\delta+2s)^2(\omega+2s)^3 \tilde{d}(\omega+2s) + 1] \\ & + (\delta-2s)^2(\omega-2s)^3 \tilde{d}(\omega-2s)] \\ = & \tilde{f}_- [(\delta-2s)^2(\omega-2s)^3 \tilde{d}(\omega-2s) + 1] \\ & + (\delta+2s)^2(\omega+2s)^3 \tilde{d}(\omega+2s)]. \end{aligned}$$

This self-consistency condition is also the balance condition for transitions between quasi-energy levels, the factors  $\tilde{d} + 1$  and  $\tilde{d}$  corresponding, respectively, to the transitions indicated in Fig. 3 by the downward- and upward-directed dashed arrows. The factors  $(\delta \pm 2s)^2$  are, respectively, proportional to the probability that the corresponding quasi-energy level is the upper or lower harmonic of the level involved in the transition.

Let us consider the contribution of thermal photons to the polarization for the case of a strong field with  $\mathcal{E} \gg \mathcal{E}_2$ . It is easy to find from Eq. (6) that in this case

$$P(\omega) = \frac{3nd^2}{\hbar\omega} [1 + \tilde{d}(\omega+2s) + \tilde{d}(\omega-2s)]^{-1} \mathcal{E}(\omega), \quad s = \frac{d\mathcal{E}}{\hbar},$$

$$\frac{\tilde{f}_+}{\tilde{f}_-} = 1 - \frac{12s}{\omega} [1 + \tilde{d}(\omega+2s) + \tilde{d}(\omega-2s)]^{-1} \neq \exp\left(-2\frac{\hbar s}{kT}\right).$$

For the case  $\mathcal{E}_1 \ll \mathcal{E} \ll \mathcal{E}_2$  we similarly find that

$$P(\omega) = \frac{nd^2}{\hbar} \delta\mathcal{E}(\omega) [\delta^2 + 2\alpha + (\delta+2s)^2 \tilde{d}(\omega+2s) + (\delta-2s)^2 \tilde{d}(\omega-2s)]^{-1},$$

$$S = \left(\frac{\delta^2}{4} + \alpha\right)^{1/2}.$$

It is evident from these formulas that if the photon distribution function  $\tilde{d}(\omega)$  is small ( $\tilde{d}(\omega) \ll 1$ ) and has the form of Planck's distribution for the temperature  $kT \ll \hbar\omega_0$ , where  $k$  is Boltzmann's constant, it will not contribute significantly to the polarization and will not lead to a Gibbs distribution for the quasi-energies:

$$\tilde{f}_+ \neq \tilde{f}_- \exp\left(-\frac{2\hbar s}{kT}\right), \quad \tilde{d}(\omega) = \exp\left(-\frac{\hbar\omega}{kT}\right).$$

In concluding, let us consider a simple derivation of the polarization equation (8) for the case of resonance in a fairly strong field  $\mathcal{E} \gg \mathcal{E}_2$ . The complete wave function of the two-level atom breaks up into eigenfunctions  $|1\rangle$  and  $|2\rangle$  of the internal Hamiltonian. The Schrödinger equation for the stationary atom in the external field has the following form in the resonance approximation:

$$i\hbar \frac{\partial}{\partial t} \begin{pmatrix} c_2 \\ c_1 \end{pmatrix} = - \begin{pmatrix} 0 & d\mathcal{E} \\ d\mathcal{E} & 0 \end{pmatrix} \begin{pmatrix} c_2 \\ c_1 \end{pmatrix}, \quad |\psi\rangle = c_1|1\rangle + c_2 e^{-i\omega_0 t}|2\rangle,$$

$$\mathcal{E}(t) = 2\mathcal{E} \cos(\omega_0 t).$$

On transforming to the functions  $2^{1/2}c_{\pm} = c_1 \mp c_2$ , this

equation breaks up into two independent equations with the energies  $\pm d\mathcal{E}$ , and the average value of the dipole moment in the states  $\psi_{\pm}$ , for which  $c_1 = \mp c_2$ , is

$$\langle \psi_{\pm} | \hat{d} | \psi_{\pm} \rangle = \mp d \cos(\omega_0 t).$$

To describe the stationary state of a gas of two-level atoms one must know the number  $\tilde{f}_+$  of atoms in the state  $\psi_+$  and the number  $\tilde{f}_-$  of atoms in the state  $\psi_-$ ; then the stationary polarization of the gas is

$$P(t) = (\tilde{f}_- - \tilde{f}_+) d \cos(\omega_0 t).$$

This equation is equivalent to Eq. (6), which was obtained above for the polarization. The relation between  $\tilde{f}_+$  and  $\tilde{f}_-$  in the stationary case can be obtained from the balance condition  $\tilde{f}_+ \Gamma_{+-} = \tilde{f}_- \Gamma_{-+}$ , where  $\Gamma_{+-}$  and  $\Gamma_{-+}$  are the probabilities for the transitions  $\psi_+ \rightarrow \psi_-$  and  $\psi_- \rightarrow \psi_+$ , respectively. Let us consider the radiative damping mechanism in which the widths of the levels are proportional to the third power of their separation. As is evident from Fig. 3, the quasi-energy states  $\psi_{\pm}$  correspond to two levels that are shifted from the corresponding levels of the unperturbed atom by the respective amounts  $\pm d\mathcal{E}$ . A transition between two quasi-stationary states corresponds to the transition between the upper harmonic of the first quasi-energy level and the lower harmonic of the second quasi-energy level, since for radiative transitions the probability for such a process is proportional to

$$(\hbar\omega_0 \pm 2d\mathcal{E})^3 \gg (2d\mathcal{E})^3, \quad \frac{\Gamma_{+-}}{\Gamma_{-+}} = \frac{(\hbar\omega_0 + 2d\mathcal{E})^3}{(\hbar\omega_0 - 2d\mathcal{E})^3}.$$

Knowing the total number  $\tilde{f}_+ + \tilde{f}_-$  of atoms in the gas and the ratio  $\tilde{f}_-/\tilde{f}_+ = \Gamma_{+-}/\Gamma_{-+}$ , it is easy to find the following expression for the polarization of the medium:

$$P(t) = nd \cos(\omega_0 t) \frac{(\hbar\omega_0 + 2d\mathcal{E})^3 - (\hbar\omega_0 - 2d\mathcal{E})^3}{(\hbar\omega_0 + 2d\mathcal{E})^3 + (\hbar\omega_0 - 2d\mathcal{E})^3} \approx \frac{3d^2 n}{\hbar\omega_0} \mathcal{E}(t). \quad (9)$$

Equation (9) agrees with Eq. (8), which was obtained earlier for the polarization in the case of a strong field. However, Eq. (9) cannot be obtained using the density-matrix formalism, since in that formalism one considers transitions between levels that are not split and have a constant width  $\gamma_0$ , while the correction to the polarization obtained here is based on the behavior of transitions between split levels.

Thus, proper allowance for radiative processes leads to the simple expression (9) for the polarization in strong fields and to the conclusion that complete saturation is not reached in strong fields.

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