INTERACTION OF COHERENT RADIATION WITH ATOMS

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The interaction of coherent radiation with a system of two-level atoms is considered. A general expression for the time dependence of the average number of photons is derived. The coefficients in this expression [formula (12)] are determined by a system of differential equations [formula (10)]. The cases of one and two atoms are considered in detail and exact solutions of the equations are obtained. Approximate solutions of the equations for a system with an arbitrary number of atoms are also considered.

THE appearance of such sources of electromagnetic radiation as lasers has opened up new possibilities for experiments aiming at the observation of the finer effects of optical coherence. Recently, a number of theoretical papers (cf., for example, [1]) on the problem of the interaction of radiation with a resonant medium have appeared. In these papers, the emission and absorption of a definite fixed number of photons were considered. No type of phase relations and hence, no coherence effects of the photon current can be considered in such an approach. These problems have been discussed in [2] on the basis of a quasiclassical theory, and the results agree in some limiting cases with the results obtained from quantum-electrodynamical considerations.

The mathematical apparatus of quantum electrodynamics which is built up in the occupation number representation, i.e., in a representation where the number of photons is diagonal, does not permit one to obtain information on the phase of the photons. The statistical properties of the photon current have been discussed in recent papers of Glauber, Sudarshan, and several other authors.

In the present paper we consider the interaction of completely coherent radiation with two-level atoms in the single-mode approximation (the relaxation of the atoms is neglected). The method of calculation used here is analogous to the one applied by Arutyunyan.[3] The complete Hamiltonian for the system of atoms and the photon field in the single-mode approximation has the form

$$H = \frac{\hbar\omega_0}{2}J_z + \hbar\omega c^+c + \hbar(c\beta^*J_+ + c^+\beta J_-) \qquad (1)$$

here we have assumed that the transition matrix elements are the same for all atoms. The quantity ω_0 is the transition frequency in the atom;

$$J_z = \sum_j \sigma_z{}^j, \quad J_+ = \sum_j \sigma_+{}^j, \quad J_- = \sum_j \sigma_-{}^j,$$

where the summation goes over the number of atoms:

$$\beta^* = \left(\frac{2\pi}{\hbar\omega V}\right)^{1/2}(\mathbf{Me}), \qquad \beta = \left(\frac{2\pi}{\hbar\omega V}\right)^{1/2}(\mathbf{M}^*\mathbf{e}),$$

ω is the frequency, **e** the polarization vector, c and c^+ are the photon annihilation and creation operators, respectively, **M** is the matrix element for the transition of an isolated atom from a lower level to a higher level with simultaneous absorption of a photon, **M*** is the matrix element for the inverse transition with emission of a photon, and V is the volume within which the processes take place.

The initial state of the photon current will be described in a representation where the annihilation operator c is diagonal:

$$c|z\rangle = z|z\rangle, \qquad (2)$$

hence

$$\langle z|c^+ = z^*\langle z|. \qquad (2')$$

Expanding the function $|z\rangle$ in the number of photons,

$$|z\rangle = \sum_{n=0}^{\infty} A_n|n\rangle, \qquad (3)$$

we find for the coefficients A_n:[4]

$$A_n = \exp(-\tfrac{1}{2}|z|^2)z^n / \sqrt{n!} \qquad (4)$$

Let us now assume that at the initial moment (t = 0) the first N_2 atoms are excited while $N_1 = N - N_2$ atoms are in the ground state. Then the wave function of the N atoms at the time t = 0 can be written in the form

$$\Phi(0) = \Phi_1(j_1, -j_1)\Phi_2(j_2, j_2), \quad (5)$$

where $j_2 = N_2/2$, $j_1 = N_1/2$; or, expanding in a Clebsch-Gordan series in analogy to the theorem for the addition of angular momenta, we obtain

$$\Phi(0) = \sum_{j=|j_1-j_2|}^{j_1+j_2} C_j \Phi(j, j_2 - j_1),$$

$$C_j = \left[\frac{(2j+1)(2j_1)!(2j_2)!}{(j_1+j_2+j+1)!(j_1+j_2-j)!}\right]^{1/2}. \quad (6)$$

The complete initial wave function for the system of N atoms and the coherent photon field therefore has the form

$$\Psi(0) = \Phi(0)|z\rangle = \sum_{j=|j_1-j_2|}^{j_1+j_2} C_j \Phi(j, j_2 - j_1) \sum_{n=0}^{\infty} A_n |n\rangle. \quad (7)$$

Let us write the wave function at time t in the form

$$\Psi(t) = \sum_{j=|j_1-j_2|}^{j_1+j_2} C_j \Big\{ \sum_{n=0}^{j-(j_2-j_1)} A_n \sum_{m=0}^{n+j+j_2-j_1} B_m^{n;j}(t)|m\rangle \Phi(j, n - m + j_2 - j_1) + \sum_{n=j-(j_2-j_1-1)}^{\infty} A_n \sum_{m=n-[j-(j_2-j_1)]}^{n+j+j_2-j_1} B_m^{n;j} \times (t)|m\rangle \Phi(j, n-m+j_2-j_1)\Big\}, \quad (8)$$

where the limits of the summation over m are determined by

$$-j \leqslant n - m + j_2 - j_1 \leqslant j. \quad (9)$$

Substituting the function (8) in the Schrödinger equation with the full Hamiltonian (1), we obtain the following system of differential equations for the coefficients $B_m^{n;j}(t)$ [with an accuracy up to an inessential phase factor

$$\exp[-i(n + j_2 - j_1)\omega_0 t](\beta/\beta^*)^{m/2}]:$$

$$i\frac{dB_m^{n;j}(x)}{dx} = -m\varepsilon B_m^{n;j}(x) + [(m+1)(j+n-m+j_2-j_1)\cdot(j-n+m-j_2+j_1+1)]^{1/2} B_{m+1}^{n;j}(x) + [m(j-n+m-j_2+j_1)(j+n-m+j_2-j_1+1)]^{1/2} B_{m-1}^{n;j}(x) \quad (10)$$

with the initial conditions

$$B_m^{n;j}(0) = (\beta^*/\beta)^{m/2}\delta_{m,n}, \quad (11)$$

where we have introduced the notation

$$x = t|\beta|, \quad \varepsilon = (\omega_0 - \omega)/|\beta|.$$

The average number of photons at time t, defined by

$$\bar{n}(t) = \langle\Psi(t)|c^+c|\Psi(t)\rangle,$$

has the form

$$\bar{n}(t) = \sum_{n=0}^{\infty} |A_n|^2 \rho_n(t), \quad (12)$$

where

$$\rho_n(t) = \sum_{j=|j_1-j_2|}^{j_1+j_2} C_j^2 \begin{cases} \sum_{m=0}^{n+j+j_2-j_1} m|B_m^{n,j}(t)|^2 & \text{for } n \leqslant j-(j_2-j_1) \\ \sum_{m=n-[j-(j_2-j_1)]}^{n+j+j_2-j_1} m|B_m^{n;j}(t)|^2 & \text{for } n > j-(j_2-j_1) \end{cases} \quad (12')$$

is the average number of photons at time t if there were n photons at the initial time; $|A_n|^2$ is the distribution of the number of photons at the initial time, which for complete coherence, starting from formula (4), has the form of a Poisson distribution:

$$|A_n|^2 = e^{-\bar{n}}(\bar{n})^n/n!, \quad (13)$$

where $\bar{n} = |z|^2$ is the average number of photons in the incident beam.

Let us now consider a few of the simplest cases, in which the system of equations (10) with the initial conditions (11) permits an exact solution. For the case of a single atom which is in an excited state at the initial moment, the formula for the average number of photons at time t has the following form if the incident photon beam is completely coherent:

$$\bar{n}(t) = \bar{n} + \sum_{n=0}^{\infty} \frac{e^{-n}(\bar{n})^n}{n!}(n+1)\frac{1}{\Omega_{n+1}^2\tau^2}\sin^2\frac{\Omega_{n+1}}{2}t, \quad (14)$$

An analogous formula is obtained if the atom is initially in the ground state:

$$\bar{n}(t) = \bar{n} - \sum_{n=0}^{\infty} \frac{e^{-n}(\bar{n})^n}{n!} n \frac{1}{\Omega_n^2\tau^2}\sin^2\frac{\Omega_n}{2}t, \quad (15)$$

where

$$\Omega_n = [(\omega_0 - \omega)^2 + n/\tau^2]^{1/2}, \quad (16)$$

and $\tau = 1/2|\beta|$ is the time for a spontaneous transition. At resonance, where $\omega_0 = \omega$ and hence $\Omega_n = \sqrt{n}/\tau$, formulas (14) and (15) take the form

$$\bar{n}(t) = \bar{n} + \sum_{n=0}^{\infty} \frac{e^{-n}(\bar{n})^n}{n!}\sin^2\sqrt{n+1}\frac{t}{2\tau},$$

$$\bar{n}(t) = \bar{n} - \sum_{n=0}^{\infty} \frac{e^{-\bar{n}}(\bar{n})^n}{n!} \sin^2 \sqrt{n} \frac{t}{2\tau}. \qquad (17)$$

The first formula has been obtained by a different method in a recent paper of Cummings.[5]

For the case of two atoms, one of which is in the ground state while the other is an excited state, we obtain the following expression for the average number of photons at resonance, where $\omega_0 = \omega$:

$$\bar{n}(t) = \bar{n} + \sum_{n=0}^{\infty} \frac{e^{-n}(\bar{n})^n}{2(2n+1)n!} \sin^2 \sqrt{2(2n+1)} \frac{t}{2\tau}. \qquad (18)$$

We shall not quote the general formula outside the resonance because of its complexity. In going over to perturbation theory it is easy to see the connection between (17) and (16).

Let us now consider the approximate solution of the system of equations (10) with the initial conditions (11) for a system with an arbitrary number of atoms. Formulas (12) and (12′) for the average number of photons can be rewritten in the form

$$\bar{n}(t) = \sum_{n=0}^{\infty} |A_n|^2 \sum_{j=|j_1-j_2|}^{j_1+j_2} C_j^2 \rho_n{}^j(t), \qquad (19)$$

where

$$\rho_n{}^j(t) = \sum_m m |B_m^{n,j}(t)|^2, \qquad (20)$$

where the limits of the summation over m are determined by (9).

Let us rewrite (10) in matrix form:

$$i\frac{dB^{n,j}(x)}{dx} = H^{n,j} B^{n,j}(x), \qquad (21)$$

where

$$H^{n,j} = -\varepsilon l + g^{n,j} + f^{n,j},$$
$$l_{m,k} = m\delta_{m,k}, \quad f^{n,j} = (g^{n,j})^+,$$
$$(g^{n,j})_{m,k} = [m(j-n+m-j_2+j_1)(j+n-m + j_2 - j_1 + 1)]^{1/2}\delta_{m-1,k}.$$

It is easy to see that the formal solution of (21) with the initial conditions (11) has the form

$$B_m^{n,j}(x) = \left(\frac{\beta^*}{\beta}\right)^{n/2} (e^{-ixH^{n,j}})_{mn}. \qquad (22)$$

Substituting this solution in (20), we obtain the function

$$\rho_n{}^j(x) = \sum_m m (e^{ixH^{n,j}})_{nm} (e^{-ixH^{n,j}})_{mn}, \qquad (23)$$

which is the diagonal term $(L^{n,j})_{nn}$ of the Heisenberg matrix

$$L^{n,j}(x) = e^{ixH^{n,j}} l e^{-ixH^{n,j}}. \qquad (24)$$

Hence

$$\rho_n{}^j(x) = (L^{n,j}(x))_{nn}. \qquad (25)$$

Direct differentiation of the operator $L^{n,j}(x)$ with respect to x and use of the commutation relations

$$[l, g^{n,j}] = g^{n,j}, \quad [l, f^{n,j}] = -f^{n,j},$$
$$[g^{n,j}, f^{n,j}] = 3l^2 - [4(n+j_2-j_1) - 1] \cdot l - (j+n+j_2-j_1)(j-n-j_2+j_1+1)$$

lead to a differential equation for the operator $L^{n,j}(x)$:

$$\frac{d^2L^{n,j}}{dx^2} = -6(L^{n,j})^2 + \{-\varepsilon^2 + 2[4(n+j_2-j_1)-1]\} L^{n,j} + 2(j+n+j_2-j_1)(j-n-j_2+j_1+1) - \varepsilon H^{n,j} \qquad (26)$$

In order to obtain a closed equation for the function $\rho_n^j(x)$ from the formula (25), we rewrite (26) for the diagonal matrix element $(L^{n,j}(x))_{nn}$, neglecting the nondiagonal elements of the matrix $L^{n,j}$:

$$[(L^{n,j})^2]_{nn} = [(L^{n,j})_{nn}]^2.$$

Finally we obtain the following equation:

$$\rho_n{}^{j\prime\prime}(t) = -\frac{3}{2|\Delta|\tau^2} \rho_n{}^j(t)^2 + \left\{-(\omega_0-\omega)^2 + \frac{2n}{|\Delta|\tau^2} + \frac{\Delta}{|\Delta|\tau^2} - \frac{1}{2|\Delta|\tau^2}\right\} \cdot \rho_n{}^j(t) + \frac{1}{2|\Delta|\tau^2}\left(j+n+\frac{\Delta}{2}\right) \times \left(j-n-\frac{\Delta}{2}+1\right) + n(\omega_0-\omega)^2 \qquad (27)$$

with the initial conditions

$$\rho_n{}^j(0) = n, \quad \rho_n{}^{j\prime}(0) = 0 \qquad (28)$$

[the initial conditions are obtained from (11) and (20), using (10)].

The quantity τ in (27) is the characteristic time

$$1/\tau = 2|\beta|\sqrt{|\Delta|},$$

and Δ is the initial excess population:

$$\Delta = 2(j_2 - j_1) = N_2 - N_1.$$

It is convenient to introduce instead of $\rho_n^j(t)$ the new function

$$f_n{}^j(t) = \rho_n{}^j(t) - n, \qquad (29)$$

Then formula (19) for the average number of photons at time t has the form

$$\bar{n}(t) = \bar{n} + \sum_{n=0}^{\infty} |A_n|^2 \sum_{j=|j_1-j_2|}^{j_1+j_2} C_j^2 f_n{}^j(t), \qquad (30)$$

where the function $f_n^j(t)$ satisfies the differential equation

$$f_n^{j\prime\prime}(t) = -\frac{3}{2|\Delta|\tau^2} f_n^j(t)^2 + \left\{-(\omega_0-\omega)^2 + \frac{\Delta}{|\Delta|\tau^2} - \frac{1}{|\Delta|\tau^2}\left(n+\frac{1}{2}\right)\right\} \cdot f_n^j(t) + \frac{\Delta}{2|\Delta|\tau^2}\left(n+\frac{1}{2}-\frac{\Delta}{4}\right) + \frac{\langle j^2\rangle}{2|\Delta|\tau^2} \quad (31)$$

with the initial conditions

$$f_n^j(0) = 0, \quad f_n^{j\prime}(0) = 0, \quad (32)$$

where

$$\langle j^2 \rangle = j(j+1).$$

Equation (16) coincides exactly with the equation for the two cases $j_2 = 0$, $j = j_1$ and $j_1 = 0$, $j = j_2$ obtained in [1]. It also agrees completely with the equation obtained by Arutyunyan for $n = 0$.

Equation (16) permits the following solution:

$$t = \tau\sqrt{|\Delta|} \int_0^{f_n^j(t)} \frac{dx}{[x(x-x_1)(x_2-x)]^{1/2}}, \quad (33)$$

where x_1 and x_2 are the roots of the quadratic equation

$$x^2 - |\Delta|\left(\frac{\Delta}{|\Delta|} - (\omega_0-\omega)^2\tau^2 - \frac{1}{|\Delta|}\left(n+\frac{1}{2}\right)\right) \cdot x - \Delta\left(n+\frac{1}{2}-\frac{\Delta}{4}\right) - \langle j^2\rangle = 0. \quad (34)$$

Let us consider two cases in detail.

1. $\Delta > 0$ and hence $\Delta(n + 1/2 - \Delta/4) + \langle j^2 \rangle > 0$. In this case Eq. (34) has roots of different signs and the solution (33) can therefore be written in the form

$$2\tau\sqrt{\frac{\Delta}{x_2-x_1}} F\left(\arcsin\left[\frac{(x_2-x_1)f_n^j(t)}{x_2(f_n^j(t)-x_1)}\right]^{1/2}, \left[\frac{x_2}{x_2-x_1}\right]^{1/2}\right) = \begin{cases} t & \text{for } 0 \leqslant t \leqslant T/2 \\ T-t & \text{for } T/2 \leqslant t \leqslant T, \end{cases} \quad (35)$$

where the function F is the incomplete elliptic integral of the first kind, x_2 and x_1 are the positive and negative roots of the quadratic equation (34), respectively, and T is the period of the function $f_n^j(t)$:

$$T = 4\tau\sqrt{\frac{\Delta}{x_2-x_1}} F\left(\frac{\pi}{2}, \sqrt{\frac{x_2}{x_2-x_1}}\right). \quad (36)$$

The function $F(\pi/2, x)$ is the complete elliptic integral of the first kind. Since $x_2 \geq f_n^j(t) > 0 > x_1$, it follows that $\max f_n^j(t) = x_2$.

2. $\Delta < 0$.

a) If $\Delta(n + 1/2 + \Delta/4) - \langle j^2 \rangle < 0$ ($\Delta \equiv |\Delta|$), then (34) has again roots with different signs, and hence this case is similar to the first case;

b) if $\Delta(n + 1/2 + \Delta/4) - \langle j^2 \rangle > 0$ ($\Delta \equiv |\Delta|$), then (34) has two negative roots $x_1 < x_2 < 0$, and therefore the solution (33) has the form

$$2\tau\sqrt{\frac{\Delta}{-x_1}} F\left(\arcsin\sqrt{\frac{f_n^j(t)}{x_2}}, \sqrt{\frac{x_2}{x_1}}\right) = \begin{cases} -t & \text{for } 0 \leqslant t \leqslant T/2 \\ -T+ & \text{for } T/2 \leqslant t \leqslant T; \end{cases} \quad (37)$$

where the period of the function $f_n^j(t)$ is equal to

$$T = 4\tau\sqrt{\frac{\Delta}{-x_1}} F\left(\frac{\pi}{2}, \sqrt{\frac{x_2}{x_1}}\right). \quad (38)$$

Since $0 > f_n^j(t) \geq x_2 > x_1$, we have $\min f_n^j(t) = x_2$.

It is easy to see that formulas (35) and (37) for a single atom agree with formulas (14) and (15) for large intensities ($\bar{n} \gg 1$).

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