Processes which are second order in the weak radiation field at a "dressed" atom

G. Yu. Kryuchkov, V. E. Mkrtchyan, M. L. Ter-Mikaelyan, and V. O. Chaltykyan

Institute of Physics Research, Academy of Sciences of the Armenian SSR, Erevan

(Submitted 4 May 1984)
Zh. Eksp. Teor. Fiz. **88**, 30–39 (January 1985)

The simultaneous emission of two photons by an atom in a resonant radiation field (a "dressed" atom) is analyzed. Calculations are carried out for a two-level system in the resonant approximation incorporating intensity effects. The time-varying regime (in which the observation time t is shorter than the lifetime of the excited level, γ^{-1}) and the steady-state regime ($t \gg \gamma^{-1}$) are analyzed. Calculations are carried out to find the probabilities (under the condition $t \gg \gamma^{-1}$) and the spectral correlation functions of the fields and the numbers of photons (in the steady state) for the coherent simultaneous emission of two photons by a dressed atom. The frequencies of the emitted photons, $\omega_{1,2}$, are shown to be symmetric with respect to the frequency of the pump field, ω. The probabilities and correlation functions have peaks as functions of $\omega_{1,2}$ near the frequencies ω and $\omega \pm \Omega$, where Ω is the frequency at which the atom oscillates in the field. The peak widths are calculated for the steady state and found to be equal to the widths for resonant fluorescence. The heights of the peaks are expressed in terms of their widths and in terms of the steady-state populations of the levels of the dressed atom. The relationship between the coherent two-photon emission at a dressed atom and parametric effects in the medium is determined. Probabilities for the incoherent emission of two photons are also derived. These probabilities are shown to have a two-peak structure. The frequencies of the emitted photons are related by $\omega_1 + \omega_2 = 2\omega \mp \Omega$ for transitions involving the excitation and the decay, respectively, of the atom. The scattering of photons by a dressed atom is also studied.

1. INTRODUCTION

The parametric effects which occur as intense optical radiation (the pump field) propagates through a medium have been the object of extensive research for the past 20 yr. The results of this research have been summarized in some well-known monographs.[1,3] The elementary events underlying these parametric effects-events in which the pump field interacts with the individual atoms of the medium-have not been studied adequately (more on this below).

In this paper we consider all possible processes in which one, two, or three pump photons are converted into two other photons in an interaction with a two-level atom. The pump field is assumed to be intense and is taken in the resonant approximation. The field of the other photons is weak.

In Sections 2 and 3 we examine these processes at times $t \gg \gamma^{-1}$, where γ^{-1} is the time for the spontaneous decay of the excited state of the atom. This time interval is of much practical interest because of the development of experimental techniques with picosecond and subpicosecond pump pulses. The problem is solved in the representation of stationary states of the composite system consisting of the atom and the photons of the pump field (the "dressed" atom). The interaction of a dressed atom with the weak radiation field causes transitions between the states of the dressed atom. These transitions result in a conversion of the photons of the pump field into photons at other frequencies. Over short time intervals these transitions can be treated by perturbation theory. Processes which are of first order in the weak radiation field have been studied quite thoroughly (see Refs. 4 and 17, for example). In the present paper we are dealing instead with second-order processes, which determine the parametric effects in a medium. These are processes involving the simultaneous emission (or absorption) of two photons by a dressed atom and processes involving the scattering of the weak radiation field by a dressed atom. In Section 2 we examine the coherent emission and scattering, while in Section 3 we consider the incoherent processes. In Section 4 we derive the relationship between these elementary processes and parametric effects.

In Sections 5–7 the problem is solved for times $t \gg \gamma^{-1}$ (the steady state) by a density-matrix method in the representation in terms of quasienergy states of the composite system consisting of the atom and the classical pump field. In the steady state, it is not sufficient to analyze the processes which are second order in the weak field by perturbation theory; it becomes necessary to consider the many-photon spontaneous emission and absorption in transitions between quasienergy states in all orders of a perturbation theory in the weak radiation field. These processes, like the spontaneous widths of the atomic levels, can be taken into account most comprehensively for a strong field by calculating the correlation functions[5] of the radiation field scattered by the atom.[1)] In Section 5 we derive general expressions for the spectral correlation functions of the field amplitudes and the numbers of photons. In Section 6 we derive a correlation function for the radiation fields for a two-level system for the case in which the frequency of the oscillations of an atomic electron in the strong field, Ω, is considerably higher than the width of the atomic transition ($\Omega \ll \gamma$). In Section 7 we calculate the correlation function for the photon numbers in the same case.

 0038-5646/85/010017-06$04.00

2. COHERENT PROCESSES

We denote by $u_{1,2}$ the coordinate part of the wave functions of a two-level atom in its ground state (with energy E_1) and in its excited state (E_2), respectively (the "bare" atom). In a quantized pump field (in photons of frequency ω; $n \gg 1$) which is nearly at resonance with the frequency of the atomic transition, $\omega_0 = (E_2 - E_1)/\hbar$ (i.e., under the condition that the frequency deviation from the resonance, $\Delta = \omega_0 - \omega$, is small: $|\Delta| \ll \omega$), dressed-atom states form. These dressed-atom states have been used by many investigators (see, for example, Refs. 10, 4, 11, and 17). We denote these states by $|\Phi_{1,2}(n)\rangle$. The corresponding energies are

$$E_{1,2}(n) = E_{1,2} + n\hbar\omega \pm \frac{\hbar}{2}(\Delta - \Omega), \tag{2.1}$$

where $\Omega = (\Delta^2 + 4|V|^2)^{1/2}$, while

$$V = -i(\mathbf{ed})(2\pi\omega n/\hbar v)^{1/2} = -i(\mathbf{ed}/\hbar)(2\pi I/c)^{1/2}$$

in the frequency of the interaction of the atom with the pump field ($\mathbf{e}$ is the polarization vector of the pump photon, $\mathbf{d} = \langle u_2|\hat{\mathbf{d}}|u_1\rangle$ is the matrix element of the dipole moment of the atomic transition, v is the normalization volume, and $I = cn\hbar\omega/v$ is the intensity of the pump field). If the pump field is turned off adiabatically ($V \to 0$), states $|\Phi_{1,2}(n)\rangle$ convert (if $\Delta > 0$) into the states $u_1|n\rangle$ and $u_2|n\rangle$, respectively. The term $\hbar(\Delta - \Omega)/2$ in expressions (2.1), which determines the "binding energy" between the atom and the pump field, also vanishes.

The interaction of the dressed atom with the radiation field causes transitions between the states of the composite system consisting of the dressed atom and the radiation field.[2] We denote these states by $|\Phi_{1,2}(n)\rangle|\{n_i\}\rangle \equiv |\Phi_{1,2}(n)\rangle|\{n_i\}\rangle$, where $|\{n_i\}\rangle \equiv |n_1\rangle|n_2\rangle\ldots$, and n_i is the number of photons of frequency ω_i in the radiation field ($n_i \equiv n_{\mathbf{k}_i\lambda_i}$). The energies of the states $|\Phi_{1,2}(n,\{n_i\})\rangle$ are obviously equal to $E_{1,2}(n) + \Sigma\hbar\omega_i n_i$.

The coherent emission of two photons of frequencies ω_1 and ω_2 occurs when a transition from a state $|\Phi_i(n,n_1,n_2,\ldots)\rangle$ to state $|\Phi_i(n-2, n_1+1, n_2+1,\ldots)\rangle$ ($i = 1,2$) takes place as a result of an interaction with weak radiation field. A calculation in second-order perturbation theory leads to the following expression for the probability per unit time of the spontaneous ($n_1 = n_2 = 0$) emission of two photons in this transition:

$$dW_{11\ \mathrm{em}}^{(2)} = \frac{\omega_1^3\omega_2^3}{8\pi^3\hbar^2c^6}\,d\omega_1\,d\omega_2\,d\omega_1\,|e_{1p}^{*}e_{2q}^{*}\alpha_{pq}(\omega_1)|^2, \tag{2.2}$$

$$\alpha_{pq}(\omega_1) = d_p^{*}d_q^{*}\left(\frac{V}{\Omega}\right)^2\left[2\pi i\delta(\omega-\omega_1) + \frac{1}{\omega_1-\omega_P+i\varepsilon} - \frac{1}{\omega_1-\omega_T-i\varepsilon}\right],$$

where $\mathbf{e}_{1,2}$ are the polarization vectors of the emitted photons, $\omega_{P,T} = \omega \pm \Omega$, p and q are vector indices, and the frequency of the second emitted photon is determined by energy conservation: $\omega_2 = 2\omega - \omega_1$. The infinitesimal imaginary increments in the denominators in (2.2) result from the adiabatic switching on of the interaction with the weak radiation field at $t \to -\infty$. The contribution of the poles in expression (2.2) for the tensor $\alpha_{pq}(\omega_1)$ corresponds to stepped two-photon processes, i.e., processes such that energy conservation also holds in intermediate states. In this case the probability for the process can be broken up into the product of probabilities for two first-order processes (with a factor t). This contribution, however, is considerably smaller than the probability for the first-order processes at the same frequencies, ω, $\omega \pm \Omega$, because of the condition $tW^{(1)} \ll 1$.

Expression (2.2) obviously depends nonlinearly on the pump field intensity. If $|V| \ll |\Delta|$, an expansion of these expressions in the parameter $|V|/|\Delta|$ is equivalent to a perturbation theory in the pump field.[3] The opposite limit, $|V| \gg |\Delta|$, corresponds to a saturation effect.

Working from (2.2) in the usual way, we can go over to equations for stimulated-stimulated ($n_1 \neq 0$, $n_2 \neq 0$), or spontaneous-stimulated ($n_1 \neq 0$, $n_2 \neq 0$) emission of two photons. The stimulated-stimulated emission is accompanied by an absorption process with the same probability. The emission of two photons is thus primarily a spontaneous-stimulated emission, for which the probability is

$$W_{11}(\omega_1) = \frac{8\pi(2\omega-\omega_1)^3}{3\hbar^3c^4}|d|^2 I_{\omega_1}|\mathbf{e}_1^{*}\mathbf{d}^{*}|^2\left(\frac{|V|}{\Omega}\right)^4 \times\left[\frac{1}{\omega_1-\omega_P} - \frac{1}{\omega_1-\omega_T}\right]^2 \tag{2.3}$$

where $I_{\omega_1} = c\hbar\omega_1 n_1/v$ is the intensity of the weak stimulating field at the frequency ω_1. Expression (2.3) is written for regions far from the poles; i.e., it ignores stepwise processes.

Exactly the same result can be derived by using a semiclassical method to calculate the emission of a photon ω_2 caused by two fields, $\mathbf{E}_0$ and $\mathbf{E}_1$ (Section 4).

Coherent (unshifted) scattering of photons occurs in the transitions $|\Phi_i(n,n_1,0,\ldots)\rangle \to |\Phi_i(n,n_1-1,1_{\omega_2},\ldots)\rangle$ ($i = 1,2$); a photon of frequency ω_1 is absorbed, a photon of frequency $\omega_2 = \omega_1$ is emitted, and the number of photons of the pump field remains unchanged. The probability for this process is

$$dW_{11\ \mathrm{sc}}(\omega_1) = \frac{\omega_1^3}{\hbar^3c^4}\,d\omega_2 I_{\omega_1}|e_{1p}e_{2q}^{*}\beta_{pq}(\omega_1)|^2,$$

$$\beta_{pq}(\omega_1) = d_p\,d_q^{*}\left[\left(\frac{|V|}{\Omega}\right)^2 2\pi i\delta(\omega-\omega_1) - \frac{(1+\Delta/\Omega)^2}{4(\omega_1-\omega_p+i\varepsilon)} + \frac{(1-\Delta/\Omega)^2}{4(\omega_1-\omega_T-i\varepsilon)}\right], \tag{2.4}$$

where $\beta_{pq}(\omega_1)$ is the scattering tensor in the presence of the pump field. In the limit of a perturbation theory in the pump field, expression (2.4) corresponds to the Kramers-Heisenberg formula for the case of unshifted scattering by a two-level atom[12] near resonance.

3. INCOHERENT PROCESSES

Incoherent processes—emission, absorption, and scattering—occur in transitions $|\Phi_{1,2}(\ldots)\rangle \to |\Phi_{2,1}(\ldots)\rangle$, i.e., transitions involving a change in atomic state. In this case the probability amplitude for the process depends on the arbitrary phases of the wave functions $u_{1,2}$. As a result, the probabilities for processes in the set of the various atoms will not

contain interference terms from the different atoms after an average is taken over the arbitrary phases. Such processes are sometimes called "combinational processes," in contrast with the parametric processes discussed in Section 2, for which the phase relations are important.

The transition $|\Phi_1(n,n_1,n_2,...)\rangle \to |\Phi_2(n-3,n_1+1,n_2+1)\rangle$ leads to the emission of two photons, ω_1 and ω_2, related by $\omega_1+\omega_2=2\omega-\Omega$. The probability for an emission which is spontaneous in both photons ($n_1=n_2=0$) in this case is

$$dW_{21\ \mathrm{em}}^{(2)}=\frac{\omega_1^3\omega_2^3}{8\pi^3\hbar^2c^6}d\mathbf{o}_1\,d\mathbf{o}_2\,d\omega_1|\,(\mathbf{e}_1^*\mathbf{d}^*)\,(\mathbf{e}_2^*\mathbf{d}^*)\,|^2 \times\frac{|V|^2(1-\Delta/\Omega)^2}{(\omega_1-\omega)^2(\omega_1-\omega_T)^2},\qquad \omega_2=2\omega-\Omega-\omega_1. \tag{3.1}$$

An expression for the probability for emission which is stimulated with respect to both photons can be found from (3.1) by the standard procedure. At the same transition the system will absorb two photons ω_1 and ω_2, related by the conservation law $\omega_1+\omega_2=2\omega+\Omega$. In this case (in contrast with coherent emission processes) the emission which is stimulated with respect to both photons will not compensate for stimulated absorption processes, since they occur at different frequencies.

In the transition $|\Phi_2(n,n_1,n_2,...)\rangle \to |\Phi_1(n-1,n_1+1,n_2+1,...)\rangle$, photons are emitted with frequencies related by $\omega_1+\omega_2=2\omega+\Omega$. In this case the spontaneous emission is described by expression (3.1) with $\Omega\to-\Omega$. In the same transition, photons related by $\omega_1+\omega_2=2\omega-\Omega$ are absorbed. The probability for the absorption of two photons in the transition

$$|\Phi_1(n,n_1,n_2,\dots)\rangle\to|\Phi_2(n+1,n_1-1,n_2-1)\rangle$$

is obviously equal to the probability for the stimulated emission of two photons in the transition

$$|\Phi_2(n,n_1,n_2,\dots)\rangle\to|\Phi_1(n-1,n_1+1,n_2+1,\dots)\rangle,$$

and the probability for the two-photon absorption $|\Phi_2(n,n_1,n_2,...)\rangle\to|\Phi_1(n+3,n_1-1,n_2-1)\rangle$ is accordingly equal to the probability for the stimulated emission $|\Phi_1(n,n_1,n_2,...)\rangle\to|\Phi_2(n-3,n_1+1,n_2+1,...)\rangle$; i.e., it is described by expression (3.1) if we switch to stimulated emission in this expression in the usual fashion. In the weak pump field limit ($|V|\ll|\Delta|$), expression (3.1) is a small quantity of order $|V/\Delta|^6$. The probability for the emission $|\Phi_2(...)\rangle\to|\Phi_1(...)\rangle$ in this case is obviously of order $|V/\Delta|^2$.

An incoherent (combinational) scattering of a weak field occurs in the transitions $|\Phi_1(n,n_1,0,...)\rangle\to|\Phi_2(n-1,n_1-1,1\omega_2,...)\rangle$ and $|\Phi_2(n,n_1,0,...)\rangle\to|\Phi_1(n+1,n_1-1,1\omega_2,...)\rangle$. Calculations lead to the following expression for the scattering probability in the transition $|\Phi_1(...)\rangle\to|\Phi_2(...)\rangle$:

$$dW_{21\ \mathrm{sc}}(\omega_1)=\frac{\omega_2^3}{\hbar^3c^4}I_{\omega_1}|\,(\mathbf{e}_1\mathbf{d})\,(\mathbf{e}_2^*\mathbf{d}^*)\,|^2d\mathbf{o}_2 \times\left(\frac{|V|}{\Omega}\right)^2\left[\frac{1+\Delta/\Omega}{\omega_1-\omega_P}+\frac{1-\Delta/\Omega}{\omega_1-\omega}\right]^2,\qquad \omega_2=\omega_1-\Omega. \tag{3.2}$$

In the transition $|\Phi_2(...)\rangle\to|\Phi_1(...)\rangle$, the emission frequency is $\omega_+ +\Omega$, and the scattering probability is found from (3.2) by changing the sign of Ω.

Incoherent processes do not contain a contribution from stepwise processes, since stepwise processes going through different intermediate states cancel each other out.

4. RELATIONSHIP WITH PARAMETRIC EFFECTS

In this section of the paper we find the relationship between the probabilities of the elementary processes calculated above and the nonlinear susceptibilities. For this purpose we will derive the average dipole moment of an atom in a strong classical pump field of amplitude $\mathbf{E}_0$ and in a weak probe field of amplitude $\mathbf{E}_1$ and frequency ω_1 in the linear approximation in the latter field. In this case, it is convenient to use the well-known quasienergy states[15-17] $|\Phi_{1,2}(t)\rangle$ instead of the states of the dressed atom.

Under the assumption that the system is in the state $|\Phi_1(t)\rangle$ when the interaction with the weak field is turned on at $t\to-\infty$, we find the following expression for the average dipole moment:

$$\langle d_q(t)\rangle=-\frac{V}{\Omega}d_q\,e^{-i\omega t}+\chi_{qp}(\omega_1)E_{1p}(\omega_1)e^{-i\omega_1 t} +\chi_{qp}^{(\mathrm{pr})}(2\omega-\omega_1)\dot{E}_{1p}^*(\omega_1)e^{-i(2\omega-\omega_1)t}+\text{c.c.}, \tag{4.1}$$

where

$$\chi_{qp}(\omega_1)=\frac{1}{4\hbar}d_q^*d_p\left[\frac{(1-\Delta/\Omega)^2}{\omega_1-\omega_T+i\varepsilon}-\frac{(1+\Delta/\Omega)^2}{\omega_1-\omega_P+i\varepsilon}\right], \tag{4.2}$$

$$\chi_{qp}^{(\mathrm{pr})}(2\omega-\omega_1)=d_q^*d_p^*\frac{V^2}{\hbar\Omega^2}\left(\frac{1}{\omega_1-\omega_P-i\varepsilon}-\frac{1}{\omega_1-\omega_T-i\varepsilon}\right). \tag{4.3}$$

The first term in expression (4.1) does not contain the weak field; it leads to the ordinary Rayleigh scattering of the pump field with saturation effects.[17] As can be seen from a comparison of (4.2) and (2.4), the polarizability tensor $\chi_{qp}(\omega_1)$ is the same as the scattering tensor $\beta_{pq}(\omega_1)$ far from resonances.[4)] The obvious explanation for the disagreement of these expressions near resonances (the signs of the imaginary increments are different) is that the width of the atomic-transition line has been ignored in this analysis. The same situation arises in the absence of a pump field.[12] When the width of the excited level is taken into account by adding a finite imaginary part to its enegy, the polarizability becomes equal to the scattering tensor over the entire frequency range. In the presence of a pump field, however, expressions (2.4) and (4.2) may not be the same over the entire frequency range, even if the width of the transition line taken into account, because expression (4.2) has no contribution from coherent Rayleigh scattering.

The third term in (4.1), which describes oscillations of the dipole moment at the frequency $2\omega_1-\omega_1$, results from the coherent emission of two photons, since the polarizability (4.3) is the same as the tensor $\alpha_{pq}(\omega 1)$ in expression (2.2), with the same reservations as in the case of expression (4.2).

Let us examine the propagation of a probe field at the frequency ω_1 through a resonant medium in a pump field. Using dipole moment (4.1) in the truncated Maxwell's equations for the fields at the frequencies ω_1 and $2\omega-\omega_1$, adopt-

ing the one-dimensional approximation along the propagation direction of the waves in the medium (the x axis), and expanding the amplitudes of the weak fields in powers of the distance x in the medium in these equations, we find the following results, which hold to within terms proportional to x^2 (we are assuming that only the amplitude of the weak field at the frequency ω_1 is nonzero at $x=0$):

$$I(2\omega-\omega_1, x)\approx\frac{3\pi}{2}\frac{\hbar c^2}{2\omega-\omega_1}W_{11}(\omega_1)N^2x^2,$$
$$I(2\omega-\omega_1, x)\equiv\frac{c}{2\pi}|\mathbf{E}_1(2\omega-\omega_1, x)|^2. \tag{4.4}$$

We wish to call attention to the fact that the parametric coherent process is proportional to the square of the atomic number density N. It can be seen from (4.4) that the appearance of a field at the "mirror-image" frequency is determined by a two-photon emission which is stimulated at the frequency ω_1 and spontaneous with respect to $2\omega-\omega_1$; the probability for this emission is given by (2.3). Experiments on transmission over short distances can therefore yield information on the elementary processes. First-order incoherent processes determine (4.2).

Second-order incoherent processes do not contribute to propagation effects in an analysis of the propagation of radiation in the problem as formulated above.

5. GENERAL EXPRESSIONS FOR THE SPECTRAL CORRELATION FUNCTIONS

We turn now to two-photon processes for times exceeding the lifetime of the excited level ($t\gg\gamma^{-1}$). To calculate the correlation functions we use the standard apparatus of quantum electrodynamics with a classical pump field (the Furry representation[12]).

A state of the composite system consisting of the atom, the pump field, and the radiation field, $|\psi_i(t)\rangle$, is expressed in terms of a state of the system which does not interact with the radiation field in the limit $T\to-\infty$ by means of a time evolution matrix $S(t)\equiv S(t,-\infty)$ in the representation of quasienergy states (see Ref. 25, for example):

$$|\psi_i(t)\rangle=S(t)|\Phi_i, 0\rangle. \tag{5.1}$$

Here $|\Phi_i,0\rangle\equiv|\Phi_i\rangle|0\rangle$ ($i=1,2$), where $|\Phi_i\rangle$ are the quasienergy states, and $|0\rangle$ is the vacuum state of the radiation field.

The spectral correlation functions of the field amplitudes and of the numbers of photons in which we are interested can be written in terms of this S matrix as follows:

$$g^{(1)}(1,2)=\langle c_1(t)c_2(t)\rangle=\langle\Phi_1, 0|S^+(t)c_1c_2S(t)|\Phi_1, 0\rangle,$$
$$g^{(2)}(1,2)=\langle c_2^+(t)c_1^+(t)c_1(t)c_2(t)\rangle$$
$$=\langle\Phi_1, 0|S^+(t)c_2^+c_1^+c_1c_2S(t)|\Phi_1, 0\rangle,$$

where $c_{1,2}$ and $c_{1,2}^+$ are the annihilation and creation operators for photons with momenta $\mathbf{k}_{1,2}$ and polarizations $\lambda_{1,2}$.

The correlation functions are conveniently written in terms of the density matrix of the quasienergy states,[22-24] which define by[25]

$$\rho_{ij}(t)=S^+(t)|\Phi_i\rangle\langle\Phi_j|S(t)\hat{1}, \tag{5.2}$$

where $\hat{1}$ is the unit operator in the space of photon states.

Evaluating the commutators $[S(t), c_{1,2}]$, we find the following expressions for the correlation functions in the resonant approximation in the frequencies $\omega_{1,2}$:

$$g^{(1)}(1,2)$$
$$=\frac{2\pi}{\hbar v}\omega_1^{1/2}\omega_2^{1/2}e_{1p}^*e_{2q}^*\int_{-\infty}^{t}dt_1\,dt_2\,\theta(t_1-t_2)\exp[i(\omega_1t_1+\omega_2t_2)]$$
$$\times\langle\Phi_1, 0|D_p^{(-)}(t_1)D_q^{(-)}(t_2)|\Phi_1, 0\rangle+(\omega_1, \mathbf{e}_1)\rightleftarrows(\omega_2, \mathbf{e}_2), \tag{5.3}$$

$$g^{(2)}(1,2)$$
$$=\left(\frac{2\pi}{\hbar v}\right)^2\omega_1\omega_2e_{1p}e_{1q}^*e_{2r}e_{2s}^*\int_{-\infty}^{t}dt_1\,dt_2\,dt_3\,dt_4\exp[i\omega_1(t_1-t_3)]$$
$$\times\exp[i\omega_2(t_2-t_4)]\langle\Phi_1, 0|\hat{T}^+[D_p^{(+)}(t_3)D_r^{(+)}(t_4)]$$
$$\times\hat{T}[D_q^{(-)}(t_1)D_s^{(-)}(t_2)]|\Phi_1, 0\rangle, \tag{5.4}$$

where the operators $\hat{T}$ and $\hat{T}^+$ perform a chronological ordering and a chronological antiordering, respectively.

The operators $\mathbf{D}^{(\pm)}(t)$ in (5.3) and (5.4) are given by

$$\mathbf{D}^{(\pm)}(t)=\sum_{i,j}\rho_{ij}(t)\mathbf{d}_{ij}^{(\pm)}(t), \tag{5.5}$$

where $\mathbf{d}_{ij}^{(\pm)}(t)$ are the positive- and negative-frequency parts of the matrix element of the dipole moment between quasienergy wave functions.

The case $t\ll\gamma^{-1}$, discussed in the preceding sections, corresponds in this approach to the approximation $S(t)\approx1$ in expressions (5.2)–(5.5). We can therefore ignore those transitions between quasienergy states which are of higher order in the radiation field. In this case, the quantity $|g^{(1)}|^2$ given by (5.3) describes the coherent emission of two photons in second-order perturbation theory. In the same approximation, the quantity $g^{(2)}$ reduces to the sum of probabilities for the spontaneous coherent and incoherent two-photon emission from the initial state $|\Phi_1\rangle$ over the time t.

We turn now to the limiting case $t\gg\gamma^{-1}$ (the steady-state regime) under the condition $\Omega\gg\gamma$.

6. THE CORRELATION FUNCTION $g^{(1)}$ IN THE STEADY-STATE REGIME

The calculation of the function (5.3) reduces to the calculation of expectation values of the type

$$\langle\Phi_1, 0|\rho_{ij}(t_1)\rho_{kl}(t_2)|\Phi_1, 0\rangle, \tag{6.1}$$

which is carried out by a method analogous to that used in Ref. 23. This method is based on the assertion that in the resonant approximation the quantities in (6.1) satisfy at $t_1>t_2$ the same equations as are satisfied by the expectation values $\langle\Phi_1,0|\rho_{ij}(t_1)|\Phi_1,0\rangle=\langle\rho_{ij}(t_1)\rangle$ (the regression theorem). These equations can be written as follows to within terms of order γ/Ω:

$$\frac{d}{dt}\langle\rho_{11}(t)\rangle=-\Gamma\langle\rho_{11}(t)\rangle+\frac{\gamma}{4}\left(1+\frac{\Delta}{\Omega}\right)^2,$$
$$\frac{d}{dt}\langle\rho_{12}(t)\rangle=-\Gamma_{12}\langle\rho_{12}(t)\rangle, \tag{6.2}$$
$$\langle\rho_{11}(t)\rangle+\langle\rho_{22}(t)\rangle=1,\quad\langle\rho_{21}(t)\rangle=\langle\rho_{12}(t)\rangle^*,$$

where

$$\Gamma_{12}=(\gamma/2)\,(1+2|V|^2/\Omega^2),\quad \Gamma=(\gamma/2)\,(1+\Delta^2/\Omega^2), \tag{6.3}$$

and the width (γ) of the atomic transition is $\gamma = 4\omega_0{}^3|d|^2/3\hbar c^3$.

Using (6.2), we find the following equations for the quantities in (6.1) which we are seeking:

$$\langle\rho_{11}(t_1)\rho_{kl}(t_2)\rangle=\delta_{1k}\langle\rho_{1l}(t_2)\rangle e^{\Gamma(t_2-t_1)}+\rho_{11}\langle\rho_{kl}(t_2)\rangle[1-e^{\Gamma(t_2-t_1)}],$$
$$\langle\rho_{12}(t_1)\rho_{kl}(t_2)\rangle=\delta_{2k}\langle\rho_{1l}(t_2)\rangle e^{\Gamma_{12}(t_2-t_1)}\quad (k,\,l=1,\,2). \tag{6.4}$$

From (5.3), using expressions (6.4) in the steady-state limit, we find[5)]

$$g^{(1)}(1,2)$$
$$=C_1{}^{st}C_2{}^{st}-i\frac{4\pi^2}{(\hbar v)^{1/2}}(\omega_1\omega_2)^{1/2}e_{1p}{}^*e_{2q}{}^*\alpha_{pq}{}^{st}(\omega_1)\,\delta(\omega_1+\omega_2-2\omega), \tag{6.5}$$

$$\alpha_{pq}{}^{st}(\omega_1)=d_p{}^*d_q{}^*\left(\frac{V}{\Omega}\right)^2\left[\frac{(\omega_1-\omega_P)\,(\rho_{22}-\rho_{11})+i\Gamma_{12}}{(\omega_1-\omega_P)^2+\Gamma_{12}{}^2}\right.$$
$$\left.+\frac{(\omega_1-\omega_T)\,(\rho_{11}-\rho_{22})+i\Gamma_{12}}{(\omega_1-\omega_T)^2+\Gamma_{12}{}^2}-i\frac{8\Gamma\rho_{11}\rho_{22}}{(\omega_1-\omega)^2+\Gamma^2}\right],$$

where quantities

$$\rho_{11}\equiv\langle\rho_{11}(t)\rangle_{\gamma t\gg 1}=(1+\Delta/\Omega)^2/2(1+\Delta^2/\Omega^2),$$
$$\rho_{22}\equiv\langle\rho_{22}(t)\rangle_{\gamma t\gg 1}=(1-\Delta/\Omega)^2/2(1+\Delta^2/\Omega^2)$$

are the steady-state value of the populations of the quasienergy states $|\Phi_1\rangle$ and $|\Phi_2\rangle$ at $\Omega\gg\gamma$.

The first term in this expression is the product of the amplitudes for unshifted Rayleigh scattering in the steady-state regime,

$$C_{1,2}^{st}\equiv\langle c_{1,2}(t)\rangle_{\gamma t\gg 1}$$
$$=(2\pi)^{1/2}\left(\frac{\omega_{1,2}}{\hbar v}\right)^{1/2}(\mathbf{e}_{1,2}\mathbf{d}^*)\frac{V}{\Omega}(\rho_{11}-\rho_{22})\,\delta(\omega_{1,2}-\omega), \tag{6.6}$$

and describes a stepwise process. The contribution of this process in the case $|V|\gg|\Delta|$ disappears in this approximation, i.e., with an accuracy $\sim\gamma/|V|$. The resulting correlation function has a three-peak structure (ω, ω_T, ω_P); the peak widths (6.3) agree with the known widths of the peaks in the spectral intensity of resonant fluorescence.

7. THE CORRELATION FUNCTION $g^{(2)}$

We now consider the spectral correlation function of the numbers of photons, which is related in an obvious way to the intensity correlation function.

In calculating this quantity it is convenient to first single out the contribution resulting from stepwise processes. This can be done by writing the chronological products in (5.4) in the form

$$\hat{T}[D(t_1)D(t_2)]=D(t_1)D(t_2)+\theta(t_2-t_1)\,[D(t_2),\,D(t_1)],$$
$$\hat{T}^+[D(t_3)D(t_4)]=D(t_4)D(t_3)+\theta(t_4-t_3)\,[D(t_3),\,D(t_4)].$$

In the steady-state regime, the product of the first terms of these expressions leads to the product $n_1(t)n_2(t)$ of the average numbers of photons of frequencies ω_1 and ω_2, each of which is related to the spectral intensity of resonant fluorescence at $\gamma t\gg 1$ by

$$n_{1,2}(t)=\langle c_{1,2}^+(t)\,c_{1,2}(t)\rangle_{\gamma t\gg 1}=\frac{tI_{\omega_{1,2}}}{\hbar\omega_{1,2}}. \tag{7.1}$$

We can calculate the remaining part of expression (5.4) by replacing the complete set of photon states, $\Sigma|\{n_k\}\rangle\langle\{n_n\}|$ by $|0\rangle\langle 0|$; this approach corresponds to considering only two photons in the scattering field. In this approximation and under the condition $\Omega\gg\gamma$, expression (5.4) contains only terms which grow in time and can be written[6)]

$$g^{(2)}(1,\,2)=n_1(t)\,n_2(t)+|g^{(1)}-C_1{}^{st}C_2{}^{st}|^2, \tag{7.2}$$

where $g^{(1)}$ is given by (6.5). Substituting in this latter expression, we find

$$g^{(2)}(1,2)=n_1(t)\,n_2(t)$$
$$+\frac{8\pi^3\omega_1\omega_2}{\hbar^2v^2}|e_{1p}{}^*e_{2q}{}^*\alpha_{pq}{}^{st}(\omega_1)|^2t\delta(\omega_1+\omega_2-2\omega). \tag{7.3}$$

The first term in this expression corresponds to uncorrelated frequencies ω_1 and ω_2. The second term vanishes outside the region of frequencies $\omega_1+\omega_2=2\omega$; i.e., it describes an effect of the correlation of these frequencies. This correlation is taken into account to within terms of the type $[(\omega_1+\omega_2-2\omega)^2+\gamma^2]^{-1}$, which do not contain the time and which were omitted in the derivation of expression (7.3), as we mentioned above.

The heights of the peaks of the function $g^{(2)}(1,2)$ at the resonant and three-photon frequencies are identical and proportional to $|V|^4/\Omega^4\Gamma_{12}{}^2$, as can be seen from expression (6.5) for α_{pq}^{st}. The height of the peak at the Rayleigh frequency is proportional to $(8|V|^2\rho_{11}\rho_{22}/\Omega^2\Gamma)^2$. In the case $|V|\ll|\Delta|$ (which, because of the approximation $|V|\gg\gamma$ being used here, is meaningful only if $|\Delta|\gg\gamma$), the Rayleigh peak is shorter than the two other peaks by a factor of $16(|V|/\Omega)^8$. In the opposite case, $|V|\gg|\Delta|$, the Rayleigh peak is nine times as high as the three-photon and resonant peaks.

We note in conclusion that in the integral of expression (7.3) over the final states of the photons of frequency ω_2 the basic contribution at $\gamma t\gg 1$ comes from the first term of this expression, which increases in proportion to t^2:

$$\int g^{(2)}(1,2)\frac{vd^3k_2}{(2\pi)^3}=t\gamma n_1(t)\frac{|V|^2}{\Delta^2+2|V|^2}. \tag{7.4}$$

The physical meaning of Eq. (7.4) is obvious, since the quantity $n_1(t)$ represents the probability for resonant fluorescence, the factor $|V|^2/(\Delta^2+2|V|^2)$ is the steady-state population of the excited atomic state, and γt is the probability for decay of the atom.

[1)] A considerable effort has recently been made to calculate and measure the various correlation functions pertinent to resonant fluorescence.[6–9]

[2)] During transitions $|\Phi_i(\cdots)\rangle\to|\Phi_i(\cdots)\rangle$ ($i=1,\,2$), the phases of the wave functions ($u_{1,2}$) of the bare amplitude do not appear in the transition probability amplitude; i.e., such transitions are coherent.

[3)] The problem of two-photon emission was treated in this limiting case in Refs. 13 and 14.

[4)] The effects resulting from the nonlinear polarizability $\chi_{qP}(\omega_1)$ during the scattering of a trial field and during its propagation through a medium have been studied in several places (see Refs. 18–21, for example).

[5)]A general expression for $g^{(1)}(1,2)$ in a slightly different formulation of the problem was derived by a different method in Ref. 26. That expression disagrees with (6.5) in the region of parameters in which the different expressions can be compared.

[6)]A similar relation was derived in Ref. 27 for space-time correlation functions.

[1]S. A. Akhmanov and R. V. Khokhlov, Problemy nelineĭnoĭ optiki (Problems of Nonlinear Optics, Gordon, London 1972) VINITI, Moscow, 1964.

[2]N. Bloembergen, Nonlinear Optics, Benjamin, New York (1965) (Russ. transl. Mir, Moscow, 1966).

[3]D. N. Klyshko, Fotony i nelineĭnaya optika (Photons and Nonlinear Optics, Nauka, Moscow, 1980.

[4]M. L. Ter-Mikaelyan, Preprint IFI 74-11, Nonlinear Resonant Optics, Erevan, 1974.

[5]R. Glauber, Quantum Optics and Quantum Radiophysics (Russ. transl. Mir, Moscow, 1966).

[6]U. J. Carmickael and D. F. Walls, J. Phys. B **9**, 1199 (1976).

[7]H. J. Kimble, M. Dagenais, and L. Mandel, Phys. Rev. Lett. **39**, 691 (1977).

[8]P. A. Apanasevich and Ja. S. Kilin, J. Phys. B **12**, L83 (1979).

[9]A. Aspect, G. Roger, S. Reynaud, *et al.*, Phys. Rev. Lett. **45**, 617 (1980).

[10]C. Cohen-Tannoudji, Optical Pumping and Interaction of Atoms with the Electromagnetic Field. Ecole d'ete de Physique Theorique de Gargence, 1967.

[11]P. L. Knight and P. W. Milonni, Phys. Rep. **66**, 2 (1980).

[12]V. B. Berestetskiĭ, E. M. Lifshitz, and L. P. Pitaevskiĭ, Kvantovaya élektrodinamika (Quantum Electrodynamics), Nauka, Moscow, 1980.

[13]R. I. Sokolovskiĭ, Zh. Eksp. Teor. Fiz. **59**, 799 (1970) [Sov. Phys. JETP **32**, 438 (1971)].

[14]B. R. Mollow, Phys. Rev. **A12**, 1919 (1975).

[15]V. I. Ritus, Zh. Eksp. Teor. Fiz. **51**, 1544 (1966) [Sov. Phys. JETP **24**, 1041 (1967)].

[16]Ya. B. Zel'dovich, Zh. Eksp. Teor. Fiz. **51**, 1492 (1966) [Sov. Phys. JETP **24**, 1006 (1967)].

[17]M. L. Ter-Mikaelyan and A. O. Melikyan, Zh. Eksp. Teor. Fiz. **58**, 285 (1970) [Sov. Phys. JETP **31**, 153 (1970)].

[18]A. M. Bonch-Bruevich, S. G. Przhibel'skiĭ, V. A. Khodovoĭ, and N. A. Chigir', Zh. Eksp. Teor. Fiz. **70**, 445 (1976) [Sov. Phys. JETP **43**, 230 (1976)].

[19]F. Y. Wu, S. Ezekiel, M. Ducloy, and B. R. Mollow, Phys. Rev. Lett. **38**, 1077 (1977)].

[20]V. M. Arutyunyan, E. G. Kanetsyan, and V. O. Chaltykyan, Zh. Eksp. Teor. Fiz. **59**, 195 (1970) [Sov. Phys. JETP **32**, 108 (1971)].

[21]R. W. Boyd, M. G. Raymer, and P. Narum, Phys. Rev. **A24**, 411 (1981).

[22]V. L. Derbov, M. A. Kovner, and S. K. Potapov, Kvantovaya Elektron. (Moscow) **2**, 684 (1975) [Sov. J. Quantum Electron. **5**, 379 (1975)].

[23]C. Cohen-Tannoudji and S. Reynaud, J. Phys. B **10**, 354 (1977).

[24]B. V. Kryzhanovsky and A. O. Melykian, Opt. Commun. **29**, 164 (1979).

[25]G. Yu. Kryuchkov, Zh. Eksp. Teor. Fiz. **83**, 1992 (1982) [Sov. Phys. JETP **56**, 1153 (1982)].

[26]P. A. Apanasevich and S. Ja Kilin, Phys. Lett. **62A**, 83 (1977).

[27]B. R. Mollow, Phys. Rev. **A8**, 2684 (1973).

Translated by Dave Parsons