## Squeezed-vacuum effects in the quasienergy structure of atoms in a laser field

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A theory is developed of nonlinear optical processes involving isolated atomic systems in a strong external field and in a squeezed vacuum. The theory is based on transitions between quasienergy states of the system considered. For three systems—a two-level atom in a monochromatic field and in a bichromatic field, and a three-level atom under conditions of two-photon excitation—quantum phase-dependent effects are found in the transition probabilities between quasienergy states under the influence of the squeezed-vacuum field. It is shown that for certain parameters of the fields, these probabilities can be strongly suppressed. This circumstance is also manifested in the stationary interaction regime in our calculations of the resonance fluorescence spectra and in the populations of the atomic levels. It is shown that squeezed-vacuum effects lead to anomalous behavior of the atomic populations and to the possibility of their complete inversion for a three-level atom. © 1996 American Institute of Physics. [S1063-7761(96)00601-3]

## **1. INTRODUCTION**

The interaction of squeezed light with atomic systems has a number of unusual properties. It is well known that the spontaneous emission spectrum is changed significantly when an atom is placed in a squeezed vacuum, i.e., a wideband electromagnetic field in a vacuum squeezed state. For such a field, the quantum-mechanical expectation value of the amplitude is zero, the quantum noise of one of the quadrature components of the amplitude is less than the vacuum level, and the width of the spectrum greatly exceeds the natural width of the atomic transition considered. As was first shown in Ref. 1, for a two-level atom interacting with a squeezed vacuum, the decay of the atomic polarizability is described by two transverse damping constants, and these may be much greater and much less, respectively, than the transverse damping constant of the atomic polarizability in the ordinary vacuum. This effect has the consequence, in particular, that the width of the central peak of the resonance fluorescence spectrum becomes less than the natural width of the atomic transition.<sup>2</sup> At the present time, there are a fair number of results relating to the changes in the properties of radiative decays of atomic systems due to their interaction with a squeezed-vacuum field, and also for nonlinear optical processes in a squeezed vacuum. We merely mention some of these studies for isolated two-level<sup>1-7</sup> and three-level<sup>8-13</sup> atomic systems. We also emphasize that the interest in this problem has certainly grown in connection with recent experiments on atomic spectroscopy with squeezed light.<sup>14</sup>

The present paper is devoted to an investigation of some novel aspects of the interaction of isolated atomic systems with a strong laser field and with a squeezed vacuum. To describe such problems, we propose to use an approach that was developed earlier to describe nonlinear processes in an ordinary vacuum. We are referring here to the density-matrix formalism in the representation of quasienergy states of the compound system consisting of an atom and the laser field.<sup>15</sup> In such an approach, we assume that quasienergy states of the composite system are formed in a strong laser field,  $^{16-18}$  and the radiation processes are described by transitions between them due to the interaction of the composite system with the squeezed vacuum.

We recall that the dynamics of a quantum system in a squeezed vacuum is usually described as the interaction of a system with a reservoir of  $\delta$ -correlated squeezed white noise.<sup>1,2,4</sup> Elimination of such a reservoir leads to an equation for the reduced density matrix of the system. Proceeding similarly in this paper, we arrive at an equation for the reduced density matrix of the system consisting of an atom and the laser field. The advantage of such an approach compared with the density-matrix method in the space of atomic states is manifested in the region of strong laser fields with energy of interaction with the atom greatly exceeding the energy of the interaction of the atom with the squeezed vacuum and the characteristic relaxation widths. In this case, the spectral emission lines of the atom are separated in frequency, it becomes possible to identify the parts rapidly oscillating in time, and the equations simplify considerably (secular approximation). Such an approach permits a comparatively simple investigation of complicated nonlinear systems that are difficult to study by means of the density-matrix formalism in the space of atomic states.

In the framework of such an approach, we investigate in this paper squeezed-vacuum effects for a two-level atom excited by a resonant monochromatic field, and separately by a bichromatic field. As another application, we also consider a three-level atom in a monochromatic field under conditions of two-photon excitation when it is coupled to a squeezed vacuum.

For all these systems and for the case of small time intervals of excitation of the atoms, we find quantum phasedependent effects in the radiative transition probabilities between quasienergy states. For strong squeezing, these effects significantly change the probabilities. As follows from the results given below, the squeezed-vacuum effects in the quasienergy structure of the atoms are also manifested in the stationary regime of interaction in the fluorescence spectra. They also lead to anomalous behavior of the atomic level populations and to their complete inversion for a three-level atom. We note that these effects are absent both for the ordinary vacuum and for the interaction of atoms with a thermal reservoir.

## 2. EQUATIONS FOR THE DENSITY MATRIX

This section is devoted to the derivation of equations for the density matrix of an isolated atom interacting with a laser field that is described classically, and with the field of a squeezed vacuum. These equations are represented in a basis of quasienergy states and describe transitions between these states due to the interaction of the system consisting of the atom and the laser field with the squeezed vacuum.

We use an expression for the density matrix operator

$$\rho(t) = S(t) |\psi_0\rangle \langle\psi_0|S^+(t)$$
(2.1)

expressed in terms of the S matrix  $S(t) = S(t, -\infty)$  in the representation of quasienergy states<sup>15</sup> with interaction operator in the dipole approximation:

$$\mathbf{d}(t)\mathbf{E}(t) = \mathbf{d}(t)[\mathbf{E}^{(-)}(t) + \mathbf{E}^{(+)}(t)],$$
$$\mathbf{E}^{(+)}(t) = \int d\omega g(\omega)\mathbf{e}(\omega)a(\omega)e^{-i\omega t}.$$
(2.2)

Here  $|\psi_0\rangle = |\phi_0\rangle |0\rangle_S$ , where  $|\phi_0\rangle$  is the initial quasienergy state and  $|0\rangle_S$  is the state of the squeezed vacuum;  $\mathbf{d}(t)$  is the dipole moment operator in the representation of quasienergy states;  $g(\omega)$  is the spectral function;  $\mathbf{e}(\omega)$  is the polarization vector.

The correlation functions of the creation and annihilation operators with respect to the squeezed-vacuum state have the form<sup>1,4</sup>

$$\langle a(\omega_1)a^+(\omega_2)\rangle = [N(\omega_1)+1]\delta(\omega_1-\omega_2), \langle a(\omega_1)a(\omega_2)\rangle = M(\omega_1)\delta(2\omega_S-\omega_1-\omega_2),$$
(2.3)

where  $N(\omega)$  is the number of photons with frequency  $\omega$  in the mode in the noise reservoir, and  $M(\omega)$ , which satisfies the symmetry relation  $M(\omega_1) = M(2\omega_s - \omega_1)$ , characterizes the correlation between the amplitudes of the two modes of the radiation field with frequencies  $\omega_1$  and  $2\omega_s - \omega_1$ , where  $\omega_s$  is the characteristic center frequency of the spectrum. The following inequality holds between these parameters:

$$|M(\omega_1)|^2 \leq N(\omega_1)N(2\omega_S - \omega_1) + \min[N(\omega_1), N(2\omega_S - \omega_1)].$$
(2.4)

The equation for the density matrix operator follows from the equation for the S matrix. It can then be transformed into an equation for the reduced density matrix averaged over the squeezed-vacuum state:  $\hat{\sigma}(t) = \text{Tr}[\hat{\rho}(t)]$ . Writing the coefficients of this equation in the lowest  $e^2$ approximation in the radiation field and in the Markov approximation, which does not take into account memory effects, for the elements  $\sigma_{\alpha\beta} = \langle \alpha | \hat{\sigma}(t) | \beta \rangle$  of the density matrix between the quasienergy states we obtain the system of equations

$$i \frac{\partial}{\partial t} \sigma_{\alpha\beta}(t) = \sum_{k} \left[ m_{\alpha k}(t) \sigma_{k\beta}(t) - m^{*}_{\beta k}(t) \sigma_{\alpha k}(t) \right] \\ + \sum_{k,p} \sigma_{kp}(t) \left[ m_{\alpha k,p\beta}(t) - m^{*}_{\beta p,k\alpha}(t) \right].$$
(2.5)

Note that in these equations we have, for simplicity, omitted the vector polarization structure, which will be restored in the final results. The coefficients of the equations have the form

$$m_{\alpha\beta}(t) = -\sum_{k} m_{k\beta,\alpha k}(t), \qquad (2.6)$$

$$m_{\alpha k, p\beta}(t) = i \int_{-\infty}^{t} dt_1 d_{\alpha k}(t_1) d_{p\beta}(t) \langle E(t)E(t_1) \rangle.$$
 (2.7)

At the same time, they can be expressed in terms of the matrix elements of dipole transitions between quasienergy wave functions:

$$d_{\alpha\beta}(t) = \exp[i(E_{\alpha} - E_{\beta})t] \int d^3x \phi_{\alpha}^*(x,t) \phi_{\beta}(x,t). \quad (2.8)$$

In the expression (2.8),  $E_{\alpha,\beta}$  are the quasienergies, and  $\phi_{\alpha,\beta}(x,t) = \phi_{\alpha,\beta}(x,t+T)$  are functions that are periodic with respect to the time.<sup>16</sup> We recall that the quasienergies contain field shifts and splitting of the atomic levels, and the wave functions contain the effects of mixing of the atomic wave functions.

We obtain Eqs. (2.5) for the important case of an arbitrary atomic system perturbed by a monochromatic field with frequency  $\omega$ . For this, we write the coefficients (2.6) and (2.7) in terms of the positive-frequency  $[d_{\alpha\beta}^{(+)}(t)]$  and negative-frequency  $[d_{\alpha\beta}^{(-)}(t)]$  parts of the matrix elements (2.8). We also use the expansion

$$d_{\alpha\beta}^{(-)}(t) = \exp(iE_{\alpha\beta}t)\sum_{q} d_{\alpha\beta}^{(q)} \exp(-iq\,\omega t)$$
(2.9)

with respect to the harmonics for values  $E_{\alpha\beta} - q\omega < 0$ . This is a consequence of the periodicity of the wave functions  $\phi\alpha(x,t)$  with period  $T=2\pi/\omega$ . In the resonance approximation with respect to the radiation field, we obtain

$$m_{\alpha k, n \beta}(t) = \sum_{q} \exp[i(E_{\alpha k} + E_{n\beta} - q\omega)t] [m_{\alpha k, n\beta}^{(q)} + \exp(-2i\omega_{s}t)Q_{\alpha k, n\beta}^{(q)} + \exp(2i\omega_{s}t)P_{\alpha k, n\beta}^{(q)}],$$
(2.10)

$$m_{\alpha k,n\beta}^{(q)} = \int d\omega_1 |g(\omega_1)|^2 \sum_{q_1} \left[ \frac{d_{\alpha k}^{(q_1)} d_{\beta n}^{(q_1-q)*}[N(\omega_1)+1]}{E_{\alpha k} - q_1 \omega + \omega_1 - i\varepsilon} + \frac{d_{k\alpha}^{(-q_1)*} d_{n\beta}^{(q-q_1)}N(\omega_1)}{E_{\alpha k} - q_1 \omega - \omega_1 - i\varepsilon} \right], \qquad (2.11)$$

 $Q^{(q)}_{\alpha k,n\beta}$ 

$$=\int d\omega_{1}\sum_{q_{1}}\frac{d_{k\alpha}^{(-q_{1})^{*}}d_{\beta n}^{(q_{1}-q)^{*}}g(\omega_{1})g(2\omega_{S}-\omega_{1})M(\omega_{1})}{E_{\alpha k}-q_{1}\omega-\omega_{1}-i\varepsilon},$$
(2.12)

 $P^{(q)}_{\alpha k,n\beta}$ 

$$=\int d\omega_1 \sum_{q_1} \frac{d_{\alpha k}^{(q_1)} d_{\alpha \beta}^{(q-q_1)} g^*(\omega_1) g^*(2\omega_5 - \omega_1) M^*(\omega_1)}{E_{\alpha k} - q_1 \omega + \omega_1 - i\varepsilon}$$
(2.13)

An expansion analogous to the expression (10) with respect to harmonics holds for the coefficients  $m_{\alpha\beta}$ 

$$m_{\alpha\beta} = \sum_{q} \exp[i(E_{\alpha\beta} - q\omega)t][m_{\alpha\beta}^{(q)} + \exp(-2i\omega_{S}t)Q_{\alpha\beta}^{(q)} + \exp(2i\omega_{S}t)P_{\alpha\beta}^{(q)}].$$
(2.14)

where the expansion coefficients are readily obtained from (2.11)-(2.13) by means of the relations (2.6).

The coefficients of Eqs. (2.6) and (2.7) describe the radiative processes of lowest order in the squeezed vacuum (see Sec. 3) with allowance for the effects of the laser field. It is readily seen that these effects can be of two types. The quantities in (2.6) and (2.7) contain the effects of the mixing of the atomic states through the matrix elements  $d_{\alpha\beta}^{(g)}$ , and also the effects of the field displacement of the atomic levels through the spectrum of quasienergies. In the general case, the dependence of the coefficients (2.6) and (2.7) on the time is determined by a sum of exponentials in which, in actual calculations, we shall omit the contributions of rapid oscillations in the region of large time intervals. The expressions (2.10)-(2.14) also describe special cases: the ordinary vacuum for N = |M| = 0, a thermal field for  $N \neq 0$ , M = 0, and the squeezed-vacuum state for which the minimum of the uncertainty relation and, therefore, Eq. (2.4) holds.

# 3. TRANSITIONS BETWEEN QUASIENERGY STATES IN THE SQUEEZED VACUUM. BALANCE EQUATIONS

For the case of strong laser fields, it is possible to ignore in Eqs. (2.5) the coupling between the diagonal and offdiagonal elements of the density matrix. In order of magnitude, this coupling is equal to the ratio of the matrix elements of the atomic transitions in the squeezed vacuum to the matrix elements of the transitions in the laser field. If at the same time it is assumed that the time dependence in the coefficients can be omitted, the equations take the form

$$\frac{\partial}{\partial t} \sigma_{\alpha\alpha}(t) = -\sigma_{\alpha\alpha}(t) \sum_{k \neq \alpha} W_{\alpha k} + \sum_{k \neq \alpha} W_{k\alpha} \sigma_{kk}(t), \quad (3.1)$$
$$\frac{\partial}{\partial t} \sigma_{\alpha\beta}(t) = [i(\delta E_{\alpha} - \delta E_{\beta}) - \Gamma_{\alpha\beta}] \sigma_{\alpha\beta}(t), \quad \alpha \neq \beta.$$
$$(3.2)$$

In (3.1) and (3.2), we have adopted the notation

$$W_{\alpha k} = -i(\bar{m}_{k\alpha,\alpha k} - \bar{m}^*_{k\alpha,\alpha k}), \qquad (3.3)$$

where the bar over the coefficients denotes averaging over the fast time oscillations, and we have used the relation (2.6).

We give some explanations regarding the derivation of these equations for the case of a monochromatic excitation field with frequency  $\omega$ . Ignoring the fast time oscillations in (3.3) and (3.4), as follows from the expressions (2.10) and (2.14), for  $\pm q\omega = 2\omega_s$  it is necessary to retain the terms with the exponential dependence  $\exp[i(q\omega \mp 2\omega_s)t]$ . If the frequency of the excitation field is close to the frequency of the squeezed field ( $\omega \approx \omega_s$ ), then the harmonics with  $q = \pm 2$ make a nonvanishing contribution to (3.3) and (3.4) that contains slowly varying (in time) exponentials of the form  $\exp[\pm 2i(\omega - \omega_s)t]$ . Therefore, Eqs. (3.1) and (3.2) for this case are valid in the case of exact resonance,  $\omega = \omega_s$ , when their coefficients are constant in time after averaging over the fast oscillations.

## 3.1. Transition probabilities in the squeezed vacuum

Equations (3.1) describe the population kinetics of the quasienergy states. It is readily surmised that the coefficients  $W_{\alpha\beta}$  of the equations are the probabilities for transitions  $|\alpha\rangle \rightarrow |\beta\rangle$  in unit time between the quasienergy states in the squeezed vacuum, summed over all final states  $|R\rangle$  of the radiation field. Indeed, the transition amplitude from the initial state  $|\alpha\rangle|0\rangle_S$  of the system to the final state  $|\beta\rangle|R\rangle$  is equal in lowest-order perturbation theory to

$$A_{\alpha\beta}(R) = -i \int_{-\infty}^{t} dt_1 \langle R | \langle \beta | E(t_1) d(t_1) | \alpha \rangle | 0 \rangle_S, \quad (3.5)$$

whence, bearing in mind the definition (2.7), we obtain for the corresponding probability per unit time

$$W_{\alpha\beta} = \frac{d}{dt} \sum_{R} |A_{\alpha\beta}(R)|^2 = -i(\bar{m}_{\beta\alpha,\alpha\beta} - \bar{m}^*_{\beta\alpha,\alpha\beta}). \quad (3.6)$$

We give this expression for the case of a multilevel atom in a monochromatic excitation field for  $\omega = \omega_S$ . Calculations in accordance with (2.10)–(2.13) give

$$W_{\alpha\beta} = 2\pi \sum_{q} \{ |d_{\beta\alpha}^{(q)}|^{2} |g(\omega_{1})|^{2} [N(\omega_{1}) + 1] + |d_{\alpha\beta}^{(q)}|^{2} |g(\omega_{2})|^{2} N(\omega_{2}) + 2 \operatorname{Re}[d_{\alpha\beta}^{(2-q)} d_{\beta\alpha}^{(q)} g^{*}(\omega_{1}) g^{*} \times (2\omega_{S} - \omega_{1}) M^{*}(\omega_{1})] \}, \qquad (3.7)$$

where  $\omega_1 = E_{\alpha\beta} + q\omega$ ,  $\omega_2 = E_{\beta\alpha} + q\omega$ , and the summation contains the harmonics for which  $\omega_1 > 0$ ,  $\omega_2 > 0$ . Note that in writing this expression in compact form, we have used the relation  $M(2\omega_S - \omega_1) = M(\omega_1)$ .

It is readily seen that the transition probabilities contain, in addition to the vacuum and stimulated contributions proportional to N, phase-dependent contributions with parameter  $M = |M| \exp(i\psi_S)$ , where  $\psi_S$  is the phase of the anomalous correlation function (2.3) of the squeezed field. It is readily seen that the phase-dependent contribution to the transition probability is due to the interference between the amplitude for the two-photon transition  $|\alpha\rangle \rightarrow |\beta\rangle \rightarrow |\alpha\rangle$  between the quasienergy states and the amplitude M, which characterizes two correlated photons of the squeezed field.

# 3.2. Quasienergy widths and shifts. Spectral widths of the transitions

Equations (3.2) describe the transition currents between the quasienergy states and contain radiative shifts of the quasilevels and the widths of the spectral lines in the transition  $|\alpha\rangle \rightarrow |\beta\rangle$ . It is convenient to represent the spectral widths in the form

$$\Gamma_{\alpha\beta} = \Gamma_{\alpha} + \Gamma_{\beta} + \gamma_{\alpha\beta}^{(1)} + \gamma_{\alpha\beta}^{(2)}.$$
(3.8)

Here  $\Gamma_{\alpha}$  is determined together with the quasienergy shift  $\delta E_{\alpha}$  in terms of  $\bar{m}_{\alpha\alpha}$ , which is the diagonal element of the mass operator of an electron bound in the atom in the presence of the excitation field and the squeezed-vacuum field as follows:<sup>1)</sup>  $\bar{m}_{\alpha\alpha} = \delta E_{\alpha} - i\Gamma_{\alpha}$ . As calculations show, and as is also clear from general considerations, the quasilevel width  $\Gamma_{\alpha}$  is equal to the sum of the probabilities (3.7) of transitions from the quasienergy states:

$$2\Gamma_{\alpha} = \sum_{k \neq \alpha} W_{\alpha k} \,. \tag{3.9}$$

The other contributions to the spectral width (3.8) are obtained from the coefficients (2.7) of crossed type, which, in accordance with (2.10), are equal to

$$\bar{m}_{\alpha\alpha,\beta\beta} = m^{(0)}_{\alpha\alpha,\beta\beta} + Q^{(-2)}_{\alpha\alpha,\beta\beta} + P^{(2)}_{\alpha\alpha,\beta\beta}.$$
(3.10)

At the end of the calculations, we obtain

$$\gamma_{\alpha\beta}^{(1)} = -2\pi \sum_{q>0} |g(q\omega)|^2 \{ d_{\alpha\alpha}^{(q)} d_{\beta\beta}^{(q)*} [N(q\omega) + 1] + d_{\alpha\alpha}^{(q)*} d_{\beta\beta}^{(q)} N(q\omega) \}, \qquad (3.11)$$

$$\gamma_{\alpha\beta}^{(2)} = -4\pi \sum_{q>0} \operatorname{Re}[d_{\alpha\alpha}^{(q)}d_{\beta\beta}^{(2-q)}g^{*}(q\omega)g^{*} \times (2\omega_{s}-q\omega)M^{*}(q\omega)].$$
(3.12)

Thus, we can obtain for the quasilevel shifts an expression in the form

$$\delta E_{\alpha} = \int d\omega_{1} |g(\omega_{1})|^{2} \sum_{k,q} \left[ \frac{|d_{k\alpha}^{(q)}|^{2} [N(\omega_{1}) + 1]}{E_{\alpha k} + q \omega - \omega_{1}} + \frac{|d_{\alpha k}^{(q)}|^{2} N(\omega_{1})}{E_{\alpha k} - q \omega + \omega_{1}} \right] + \int d\omega_{1} \sum_{k,q} \left[ \frac{R_{\alpha k}^{(q)}}{E_{\alpha k} + q \omega - (2 \omega_{S} - \omega_{1})} + \frac{R_{\alpha k}^{(q)}}{E_{\alpha k} + q \omega - \omega_{1}} \right].$$
(3.13)

where

$$R_{\alpha k}^{(q)} = d_{\alpha k}^{(2-q)} d_{k\alpha}^{(q)} g^{*}(\omega_{1}) g^{*}(2\omega_{5} - \omega_{1}) M^{*}(\omega_{1}).$$

The quasienergy shifts are due to the interaction of the atom in the monochromatic excitation field with the squeezed vacuum. For N = |M| = 0, they reduce to the Lamb

shift of the quasienergy in the nonrelativistic approximation.<sup>15</sup> In the general case they contain not only Lamb shifts but also Stark level shifts proportional to the numbers of photons in the mode and phase-dependent parts due to the correlation of the squeezed-vacuum modes.

# 4. INTERFERENCE EFFECTS OF THE SQUEEZED VACUUM FOR A TWO-LEVEL ATOM

Squeezed-vacuum effects in resonance fluorescence for a two-level atom in the stationary regime are well known (see Refs. 1-7 and the references there to other studies). Therefore, in this section we mainly give results for time intervals of the perturbation that are short from the point of view of transitions between the quasienergy states. We note that the concept of a "dressed atom" for resonance fluorescence in a squeezed vacuum has been constructed in a number of studies (see, for example, Refs. 7 and 13) on the basis of stationary states consisting of an atom and a given number of photons.<sup>19</sup> In its physical meaning, such a description differs fundamentally from the one used by us based on nonstationary quasienergy states, and is more appropriate in problems with cavities. In addition, a description by means of nonstationary quasienergy states can also be used for the case of pulsed excitation fields with allowance for the time dependence of the field amplitude.

We consider a two-level atom in an electromagnetic excitation field with carrier frequency  $\omega$  close to the atomic transition frequency  $\omega_0$  and with amplitude E(t). For the case of adiabatic excitation under the condition that the offset from resonance  $\varepsilon = \omega_0 - \omega$  is much greater than the reciprocal  $\tau^{-1}$  of the characteristic time of growth and decay of its intensity, the quasienergy states  $\Phi_1$  and  $\Phi_2$  of the system are well known, and the negative-frequency parts of the dipole moments of the transitions between them are

$$d_{12}^{(-)}(t) = a^{2}d^{*} \exp\left\{-i\int^{t} dt' [\omega + \Omega(t')]\right\},$$
  

$$d_{21}^{(-)}(t) = -\frac{V}{|V|} b^{2}d^{*} \exp\left\{-i\int^{t} dt' [\omega + \Omega(t')]\right\},$$
  

$$d_{11}^{(-)} = -d_{22}^{(-)}(t) = \frac{V}{|V|} abd^{*} \exp(-i\omega t).$$
(4.1)

Here

$$a = \frac{1}{\sqrt{2}} \sqrt{1 + \frac{\varepsilon}{\Omega}}, \quad b = \frac{1}{\sqrt{2}} \sqrt{1 - \frac{\varepsilon}{\Omega}},$$

 $\Omega = (\varepsilon^2 + 4|V|^2)^{1/2}$  is the Rabi frequency, V = E(t)d, d is the dipole moment of the transition between the atomic states, and  $\varepsilon > 0$ .

In the nonstationary case, the treatment is difficult on account of the calculation of the coefficients of Eqs. (2.5) with the dipole moments (4.1) with complicated time dependence. Nevertheless, results can be obtained if the method of stationary phase is used in the integration over time. How-

ever, these questions go beyond the scope of the present paper, and here we restrict ourselves to presenting the results for the limiting case of short pulses  $\tau\gamma \ll 1$ , when the time dependence of the amplitudes *a* and *b* and of the Rabi frequency can be omitted.

#### 4.1. Interference effects in transition probabilities

We assume that the excitation field is turned on adiabatically, as a result of which there is formed a definite quasienergy state:  $\Phi_1$  or  $\Phi_2$ . As is well known,<sup>18</sup> the transition  $\Phi_1 \rightarrow \Phi_2$  in the ordinary vacuum is accompanied by spontaneous emission at the frequency  $\omega - \Omega$ ; the transition  $\Phi_2 \rightarrow \Phi_1$ , by emission at the frequency  $\omega + \Omega$ ; and diagonal transitions, by emission at the frequency  $\omega$ . The probabilities of these transitions per unit time are, respectively,

$$W_{12}^{(0)} = \gamma b^4, \quad W_{21}^{(0)} = \gamma a^4, \quad W_{11}^{(0)} = W_{22}^{(0)} = \gamma a^2 b^2 \quad (4.2)$$

and they determine the intensities of the corresponding emission lines in the case of short time intervals of the excitation  $(\tau\gamma \ll 1)$ .

In the squeezed vacuum, the transition probabilities between the quasienergy states can be calculated in accordance with (3.7), in which we ignore the field shifts of the atomic levels. This means that when we take into account the effect of the laser field on the transition probabilities we take into account the effects of the mixing of the atomic states as a function of the parameter  $E\omega_0/E_{at} \varepsilon \gtrsim 1$  ( $E_{at}$  is the characteristic atomic field strength), while the nonresonant corrections  $\simeq E/E_{at}$  are ignored. The calculations lead to the results

$$W_{12} = \gamma [b^4 (N+1) + a^4 N - 2a^2 b^2 |M| \cos \psi], \qquad (4.3)$$

$$W_{21} = \gamma [b^4 N + a^4 (N+1) - 2a^2 b^2 |M| \cos \psi], \qquad (4.4)$$

$$W_{11} = W_{22} = 2 \gamma a^2 b^2 (N + \frac{1}{2} + |M| \cos \psi). \tag{4.5}$$

where  $N = N(\omega_0)$ ,  $M(\omega_0) = |M| \exp(i\psi_S)$ , and  $\psi = \psi_S - 2\psi_L$ is the phase difference of the squeezed-vacuum field and the excitation field  $\psi_L$ . These expressions are written in a form convenient for analyzing the contributions of the various elementary processes. Thus, in (4.3) the first term describes the probability of the spontaneous-stimulated transition  $\Phi_1 \rightarrow \Phi_2$ and the second the probability of the stimulated transition  $\Phi_2 \rightarrow \Phi_1$  in the ordinary vacuum. The third term contains the interference between the amplitude of the two-photon transition  $\Phi_1 \rightarrow \Phi_2 \rightarrow \Phi_1$  and the correlation function of the amplitude of the squeezed-vacuum field. We note that the above two-photon transition amplitudes are given in Ref. 20.

The interference is greatest for squeezing with minimum uncertainty relation: for  $|M|^2 = N(N+1)$ . It is readily seen that in this case even when  $\psi = 0$  the  $|1\rangle \rightarrow |2\rangle$  transition probability vanishes,  $W_{12}=0$ , if

$$N = (\sqrt{1 + \eta^2} - 1)^2 / 4\sqrt{1 + \eta^2}, \qquad (4.6)$$

where  $\eta = 2|V|/\varepsilon$ . For large offsets from resonance, this relation is realized for small photon numbers, and in the region of resonance (but not for  $\varepsilon = 0$ ) for  $N \ge 1$ . For the reverse transition  $|2\rangle \rightarrow |1\rangle$ , this effect is absent.

Analysis of the expressions (4.3)–(4.5) shows that for  $|M|^2 = N(N+1)$ ,  $\psi = 0$ , and  $N \ge 1$  there is suppression of the



FIG. 1. Dependence of normalized probabilities of the transitions  $|2\rangle \rightleftharpoons |1\rangle$  between the quasienergy states as a function of the parameter  $\eta$  for the ordinary vacuum (continuous curves) and the squeezed vacuum for  $\psi = 0$ , N=0.5 (dashed curves).

probabilities  $W_{12}$ ,  $W_{21}$  of the off-diagonal transitions compared with the probability of spontaneous decay in the ordinary vacuum when  $\varepsilon^2/\Omega^2 \ll 1/N \ll 1$  and a growth of the probabilities of the diagonal transitions with increasing N. In this case even when  $\psi = \pi$  there is suppression of the probabilities of the diagonal transitions and an increase of the probabilities of the off-diagonal transitions. A plot of the probabilities (4.3), (4.4) as a function of the parameter  $\eta$  is shown in Fig. 1.

## 4.2. Populations and radiative widths

We give some results from which it is possible to see the role of the probabilities obtained here in the stationary regime, i.e., for  $t \gg \gamma^{-1}$ . To describe this regime, we must use Eqs. (3.1) and (3.2) for the populations  $\sigma_{11}$  and  $\sigma_{22}$  and the current of the transition  $\sigma_{12}$ ; in this case, these relations are valid when  $\Omega \gg (N+1)\gamma$ . For population inversion of the quasienergy states, we obtain

$$\Delta(t) = \sigma_{11}(t) - \sigma_{22}(t) = \Delta_S + [\Delta(0) - \Delta_S]$$
  
 
$$\times \exp[-\Gamma_{11}(t)], \qquad (4.7)$$

where  $\Delta_S = (W_{21} - W_{12})/(W_{21} + W_{12})$  is the steady-state value of the inversion,  $\Delta(0)$  is its initial value, and  $\Gamma_{11} = W_{12} + W_{21}$  is the damping constant. The steady-state populations of the atomic levels turn out to be equal,

$$\delta_{11,(22)} = \frac{1}{2} \left( 1 \pm \frac{\epsilon}{\Omega} \Delta_S \right) \tag{4.8}$$

both when (4.6) holds and when  $\psi = 0$ , and they are identical to the corresponding populations of the atomic states in the quasienergy state  $\Phi_1$ , i.e.,  $\delta_{11,(22)} = (1 \pm \varepsilon/\Omega)/2$ . (Here and in what follows, we write two expressions simultaneously, separated by a comma.)

The radiative widths  $\Gamma_{\alpha} = -\text{Im } \bar{m}_{\alpha\alpha}$  of the quasienergies are calculated in accordance with (2.14). It should be noted that the result obtained,

$$\Gamma_1 = \frac{\gamma}{2} (N + b^2), \quad \Gamma_2 = \frac{\gamma}{2} (N + a^2)$$
 (4.9)

does not contain phase-dependent terms, which cancel each other when the contributions of the two transition channels are added. It is readily seen that the spectral width of the central fluorescence line is equal to the damping constant of the population inversion (see also Sec. 5). As is well known,<sup>2</sup> this width for fluorescence in a squeezed vacuum can be less than the spontaneous width of the atomic transition. The interpretation of this effect in the language of quasienergy states is very transparent and, as follows from the results given above and the relation  $\Gamma_{11} = (W_{12} + W_{21})$ , is due to phase-dependent interference in the transition probabilities (4.3) and (4.4). The spectral widths of the two side peaks of the fluorescence spectrum are equal to the width  $\Gamma_{12}$ , which can be calculated in accordance with the expressions (3.8), (3.11), (3.12), and (4.9) and by means of the matrix elements (4.1). The result has the form

$$\Gamma_{12} = \gamma [N + \frac{1}{2} + 2a^2b^2(N + \frac{1}{2} + |M|\cos\psi)].$$
(4.10)

## 5. SQUEEZED-VACUUM EFFECTS FOR A TWO-LEVEL ATOM IN A BICHROMATIC FIELD

In this section, we consider a two-level atom in a bichromatic field

$$\mathbf{E} = \mathbf{E}_0 \operatorname{Re}[\exp\{i(\omega_0 + \delta)t - 2i\Phi\} + \exp\{i(\omega_0 - \delta)t\}],$$
(5.1)

which possesses components with equal amplitudes and with frequencies  $\omega_0 + \delta$  and  $\omega_0 - \delta$  symmetrically disposed about the frequency  $\omega_0$  of the atomic transition, which also interacts with the squeezed vacuum.

As has been shown experimentally,<sup>21</sup> in a bichromatic excitation field the fluorescence spectrum contains new resonances that are absent in the case of monochromatic excitation. The spontaneous emission of a two-level atom in a field of the form (5.1) was investigated in Refs. 22-24, and the fluorescence spectrum was calculated in Ref. 25 with allowance for the effect of the excitation field on the spectral widths. The spectrum has a multipeak structure with maxima at the frequencies  $\omega_q = \omega_0 + q \delta$   $(q = 0, \pm 1, \pm 2, ...)$  that are separated from each other by the offset from resonance. The fluorescence spectrum for the more general case of asymmetric offsets from resonance is given in Ref. 26, and it was measured in Ref. 27, the results of which agree well with those of Refs. 25, 26, and 28. In Ref. 29, the effects of the cavity on the fluorescence spectrum were investigated, and it was shown that this process is very effective for the generation of single-mode squeezed light. New effects in the dynamics of the transitions of two-level Rydberg atoms in a microwave cavity under the influence of a bichromatic field were found in Ref. 30. The present section is devoted to an investigation of the radiative transitions, fluorescence spectra, and absorption of a two-level atom in a field of the form (5.1) in the presence of a squeezed vacuum.

## 5.1. Transition probabilities and spectral widths

In the special case of the excitation field (5.1), the quasienergies are equal to zero, and the quasienergy wave

functions are given in Refs. 22–25. The negative-frequency parts of the dipole transitions between the quasienergy states  $|1\rangle$  and  $|2\rangle$  have the form

$$d_{12}^{(-)}(t) = A^{2}(t)de^{-i\omega_{0}t},$$
  

$$d_{21}^{(-)}(t) = B^{2}(t)de^{-i\omega_{0}t},$$
  

$$d_{11}^{(-)}(t) = d_{22}^{(-)}(t) = -iA(t)B(t)de^{-i\omega_{0}t}.$$
(5.2)

Here

$$A(t) = \cos\left[\frac{\xi}{2}\sin(\delta t + \Phi)\right],$$
  
$$B(t) = \sin\left[\frac{\xi}{2}\sin(\delta t + \Phi)\right]e^{-i\Phi}$$
(5.3)

and we have adopted the following notation:  $\xi = 2V/\delta$ ,  $V = \mathbf{d} \cdot \mathbf{E}_0$  is the matrix element of the interaction of the atom with the field (5.1), and  $d = \langle \varphi_1 | \hat{d} | \varphi_2 \rangle$ , where  $|\varphi_1\rangle$  and  $|\varphi_2\rangle$  are atomic states. [These expressions generalize the results of Refs. 22–25 to the case of an arbitrary phase  $\Phi$  between the constituent amplitudes of the fields in (5.1).] In this case, the expansions with respect to the harmonics (2.9) have the form

$$d_{ij}^{(-)}(t) = \sum_{q} \exp[-i(\omega_0 + q\,\delta)t] d_{ij}^{(q)}$$
(5.4)

and, in particular, determine the spectral emission lines at the frequencies  $\omega_a = \omega_0 + q \delta$ .

We now obtain equations for the density matrix (2.5) that apply to the case considered. The coefficients of the equations are calculated in accordance with the expressions (2.6) and (2.7) by means of the matrix elements (5.2), with only the effects of the mixing of the atomic states by the bichromatic field being taken into account (the frequency shifts  $q\delta$  relative the frequency of the atomic transition are ignored). We illustrate the calculations with the example of the spontaneous contribution to  $m_{12,21}(t)$ , which in the resonance approximation is

$$m_{12,21}(t) = i \int_{-\infty}^{t} dt_1 \int d\omega_1 |g(\omega_1)|^2 d_{12}^{(-)}(t) d_{21}^{(+)}(t)$$
  
  $\times \exp[i\omega_1(t_1-t)].$  (5.5)

Using the expressions (5.2) and the harmonic expansion for the coefficient A(t) (5.3),

$$A^2(t) = \sum_q e^{-iq\,\delta t} A_q^2,$$

we obtain for the principal part of the integral over  $\omega_1$ 

$$m_{12,21}(t) = i \pi |\mathbf{d}|^2 \int d\omega_1 \sum_{q_q, q_q} A_{q_1}^2, A_{q_2}^2$$
  
 
$$\times \exp[-i(q_1 + q_2) \, \delta t] |g(\omega_1)|^2 \, \delta(\omega_1 - \omega_0 - q_1 \, \delta).$$
(5.6)

In what follows, we ignore the frequency shifts  $q \delta$  compared with the transition frequency  $\omega_0$  in the spectral density  $|g(\omega_1)|^2$  and the fast time oscillations. We then obtain

$$\overline{m_{12,21}(t)} \simeq \frac{i}{2} \gamma \overline{A^4(t)},$$
 (5.7)

where the bar denotes averaging over time.

As such an analysis shows, the equations for the density matrix in this case in the approximation of nonoverlapping spectral lines when  $\delta \gg (N+1)\gamma$  can be reduced to the standard form

$$\frac{\partial}{\partial t} \sigma_{11} = -W_{12} \sigma_{11} + W_{21} \sigma_{22}, \qquad (5.8)$$

$$\frac{\partial}{\partial t}\sigma_{12} = [i(\delta E_1 - \delta E_2) - \Gamma_{12}]\sigma_{12}$$
(5.9)

with coefficients having the same form as the expressions (4.3)-(4.5) and (4.10) but with different amplitudes of the mixing of the atomic states. Namely, the results for  $W_{12}$ ,  $W_{21}$ ,  $\Gamma_{12}$  can be obtained from (4.3), (4.4), (4.10) by the substitution

$$a^{4} \rightarrow \overline{A^{4}(t)} = \frac{1}{8} [3 + 4J_{0}(\xi) + J_{0}(2\xi)],$$
  

$$b^{4} \rightarrow \overline{|B(t)|^{4}} = \frac{1}{8} [3 - 4J_{0}(\xi) + J_{0}(2\xi)],$$
  

$$a^{2}b^{2} \rightarrow \overline{A^{2}(t)|B(t)|^{2}} = \frac{1}{8} (1 - J_{0}(2\xi)),$$
(5.10)

where  $J_0(\xi)$  is a Bessel function, and they have the form

$$W_{12,(21)} = \frac{\gamma}{8} \left[ 3 \mp 4J_0(\xi) + J_0(2\xi) \right] + \frac{\gamma N}{4} \left[ 3 + 4J_0(2\xi) \right]$$
$$- \frac{\gamma}{4} \left| M \right| \left[ 1 - J_0(2\xi) \right] \cos \psi,$$
$$W_{11} = W_{22} = \frac{\gamma}{4} \left[ 1 - J_0(2\xi) \right] (N + \frac{1}{2} + |M| \cos \psi), \quad (5.11)$$
$$\Gamma_{12} = \frac{\gamma}{8} \left( 2N + 1 \right) \left[ 5 - J_0(2\xi) \right]$$
$$+ \frac{\gamma}{4} \left| M \right| \left[ 1 - J_0(2\xi) \right] \cos \psi, \quad (5.12)$$

Here  $N = N(\omega_0)$ ,  $M(\omega_0) = |M| \exp(i\psi_S)$ , and  $\psi = \psi_S - 2\Phi$ .

(5.12)

By means of these results, we also obtain the damping rate  $\Gamma_{11}$  of the populations of the quasienergy states and the inversion of their steady-state values as the solution of Eq. (5.8):

$$\Gamma_{11} = W_{12} + W_{21} = \gamma \Big\{ 2N + 1 - \frac{1}{2} \big[ 1 - J_0(2\xi) \big] \\ \times (N + \frac{1}{2} + |M| \cos \psi) \Big\},$$
(5.13)

$$\Delta_{S} = \sigma_{11}^{S} - \sigma_{22}^{S} = \gamma J_{0}(\xi) / (W_{12} + W_{21}).$$
(5.14)

The transition to the basis of atomic states is made by means of the expressions for the mean populations of the atomic states  $|\varphi_1\rangle$  and  $|\varphi_2\rangle$  in the quasienergy state  $|1\rangle$ , which, as is readily seen, have the form

$$n_{1} = |\langle \varphi_{1} | 1 \rangle|^{2} = \frac{1}{2} [1 + J_{0}(\xi)],$$
  

$$n_{2} = \overline{|\langle \varphi_{2} | 1 \rangle|^{2}} = \frac{1}{2} [1 - J_{0}(\xi)].$$
(5.15)

By means of these expressions, we obtain for the steady-state populations of the atomic levels

$$\delta_{11,(22)} = \frac{1}{2} [1 \pm J_0(\xi) \Delta_S].$$
(5.16)

It is readily verified that for the ordinary vacuum the expressions obtained for the spectral widths and populations go over into the well-known results.<sup>25</sup>

## 5.2. Resonance fluorescence spectrum

The fluorescence spectrum is given by the Fourier transform of the two-time correlation function of the operators of the dipole moment in the Heisenberg picture:

$$S(\omega_k) = \frac{2}{T} \operatorname{Re} \int_{-\infty}^{T} dt_2 \int_{-\infty}^{T} dt_1 \times \exp[i\omega_k(t_1 - t_2)] \langle D^{(+)}(t_2) D^{(-)}(t) \rangle, \quad (5.17)$$

where T is the integration time. In the steady-state, this is proportional to the number of photons with frequency  $\omega_k$ emitted per unit time.

The correlation function of the dipole moments can be expressed in terms of the variables in the representation of the quasienergy states and can be calculated by means of the fluctuation regression theorem and Eqs. (5.8) and (5.9). For the resonance fluorescence in the bichromatic field in the absence of squeezed-vacuum effects, such a method of calculations was used in Ref. 25. For the case considered here, we obtain

$$\langle D^{(+)}(t_2)D^{(-)}(t_1) \rangle = d^{(+)}_{11}(t_2)d^{(-)}_{11}(t_1)[\Delta_S^2 + (1 - \Delta_S^2)\exp[-\Gamma_{11}(t_2 - t_1)]] + \frac{1}{2}\exp[-\Gamma_{12}(t_2 - t_1)][d^{(+)}_{12}(t_2)d^{(-)}_{21}(t_1)(1 + \Delta_S) + d^{(+)}_{21}(t_2)d^{(-)}_{12}(t_1)(1 - \Delta_S)].$$
(5.18)

Using also (5.2) and the corresponding expansions (5.4), we arrive at an expression for the fluorescence spectrum in the form of the sum of the contributions of the elastic and inelastic scattering:

$$S(\omega_{k}) = S_{1}(\omega_{k}) + S_{2}(\omega_{k}),$$

$$S_{1}(\omega_{k}) = \pi^{2} |\mathbf{d} \cdot \mathbf{e}|^{2} \sum_{q} J_{2q+1}^{2}(\xi) \Delta_{S}^{2} \delta(\omega_{k} - \omega_{2q+1}),$$

$$S_{2}(\omega_{k}) = \pi |\mathbf{d} \cdot \mathbf{e}|^{2} \sum_{q} \left\{ \left[ \delta_{q0}(1 - 2J_{0}(\xi)\Delta_{S}) + J_{2q}^{2}(\xi) \right] \frac{\Gamma_{12}}{(\omega_{k} - \omega_{2q})^{2} + \Gamma_{12}^{2}} + J_{2q+1}^{2}(\xi) \frac{(1 - \Delta_{S}^{2})\Gamma_{11}}{(\omega_{k} - \omega_{2q+1})^{2} + \Gamma_{11}^{2}} \right\}.$$
(5.19)

The elastic part  $S_1$  of the spectrum has peaks at the frequencies  $\omega_0 \pm \delta$ ,  $\omega_0 \pm 3\delta$ ,..., i.e., at frequencies of odd order relative to the central fluorescence line at the frequency of the atomic transition. The inelastic part of the spectrum has peaks at all frequencies  $\omega_q = \omega_0 + q \delta$ . The squeezed-vacuum effects are manifested in the spectrum through the difference



FIG. 2. Inelastic parts of the resonance fluorescence spectra in the bichromatic field in relative units for  $\xi=4$ ,  $\delta=10$  for the ordinary vacuum (curve I) and the squeezed vacuum for  $\psi=0$ , N=1 (curve 2). For the given value of the parameter  $\xi$ , peaks at the frequencies  $\omega_0 \pm \delta$  are absent.

of the populations of the quasienergy states and the widths, which are given by the phase-dependent expressions (5.12)– (5.14). It is readily seen that a phase-dependent decrease of the spectral widths occurs for the peaks with odd order. They all have the same spectral width  $\Gamma_{11}$ , which for  $\psi = 0$  and the largest negative value of the Bessel function  $J_0(2\xi)$  can reach the smallest value

$$\Gamma_{11/\gamma} = 0.65(2N+1) - 0.7|M| \tag{5.20}$$

at  $\xi = 1.9$ .

It is readily seen that  $\Gamma_{11}$  cannot be less than  $\gamma/2$ , i.e., in the resonance fluorescence in the bichromatic field a reduction of the widths of the emission lines below the spontaneous transition width does not occur.

The fluorescence spectra for the case of the ordinary vacuum and squeezed vacuum for  $|M|^2 = N(N+1)$  are given in Fig. 2.

We note also that the total integrated intensity is obtained by integrating the spectrum and is

$$S = \int d\omega_k S(\omega_k) = 4\pi^2 |\mathbf{d} \cdot \mathbf{e}|^2 \delta_{22}.$$
 (5.21)

As one would expect, this quantity is equal to the product of the population of the excited atomic level (5.16) and the transition probability of the atom from the excited level to the ground level.

## 5.3. Absorption spectrum of a test field

We consider the squeezed-vacuum effects in the absorption spectrum of a test field in the case of an isolated atom excited by a bichromatic field. For the case of an atom in a monochromatic field, these effects were investigated in Ref. 3.

The time average of the power absorbed by the atom from the test field with amplitude  $\mathbf{e}_0$  and frequency  $\nu$  is equal in the lowest order in the test field to

$$P(\nu) = \frac{2\nu}{\hbar} \operatorname{Re}(\mathbf{e}_{0n} \mathbf{e}_{0m}^* \overline{g_{n,m}(\nu,t)}),$$

$$g_{n,m}(\nu,t) = \int_{-\infty}^{t} dt_1 \exp[i\nu(t_1-t)] \\ \times \langle [D_n^{(+)}(t_1), D_m^{(-)}(t)] \rangle, \qquad (5.22)$$

where n and m are vector indices, and the bar denotes averaging over time. Using the method of calculations given in Sec. 5.2, we obtain for the two-time correlation function of the dipole moments the expression

$$\langle [D_n^{(+)}(t_1), D_m^{(-)}(t)] \rangle = -\Delta_s(\xi) J_0(\xi) d_n d_m \\ \times \exp[(i\omega_0 - \Gamma_{12})(t_1 - t)],$$
(5.23)

which leads to the following result for the absorption spectrum:

$$P(\nu) = -\frac{\sigma}{2} \left(\frac{\nu}{\omega_0}\right) \frac{\gamma J_0(\xi) \Delta_s \Gamma_{12}}{(\nu - \omega_0)^2 + \Gamma_{12}^2}.$$
 (5.24)

In this expression,  $\sigma = 4\pi\omega_0 |\mathbf{d} \cdot \mathbf{e}_0|^2 / \hbar \gamma$  is the coefficient of absorption of the test field at the frequency of the atomic transition in the absence of the excitation field.

As follows from this result, for an atom in a strong bichromatic field there is absorption of the test field in the vicinity of the atomic transition frequency, but there is no amplification. Such a situation is specific to the bichromatic field in the form (5.1) and can be readily interpreted in the language of transitions between quasienergy states (see Ref. 29, where such an interpretation is given for the coefficients of the kinetic equations). We recall that in the case of a monochromatic field there is both absorption of the test field and amplification of it at frequencies symmetric about the frequency of the excitation field.<sup>31</sup>

At the parameter values  $\xi = 2.3$ , 5.5, 8.7,..., equal to the roots of the equation  $J_0(\xi) = 0$ , the population difference of the quasienergy states (5.14) vanishes, and there is no absorption. This also occurs in the ordinary vacuum. In the squeezed vacuum, the values of the parameter  $\xi$  at which absorption is absent are the same as in the ordinary vacuum, but the peak values of the absorption and the spectral width of the absorption region are significantly changed. We note that at the indicated values of  $\xi$  there is also no contribution of the elastic scattering in the fluorescence spectrum (5.19).

## 6. THREE-LEVEL ATOM UNDER CONDITIONS OF TWO-PHOTON RESONANCE AND IN A SQUEEZED VACUUM

In this section, we consider a three-level atom in the configuration of two-photon excitation through a virtual level, as shown in Fig. 3. The matrix element of the twophoton transition is

$$W(t) = We^{2i\omega t} + \text{c.c.} \quad W = V_{23}V_{31}/(\omega_{31} - \omega),$$
 (6.1)

where  $V_{23}$  and  $V_{31}$  are the matrix elements of the singlephoton transitions, and  $\omega_{ij}$  are the frequencies of the atomic transitions. The system decays as a result of interaction with the squeezed vacuum through the level  $|3\rangle$ , which is not perturbed by the excitation field.

The negative-frequency parts of the matrix elements of the dipole transitions are (see, for example, Ref. 18)



FIG. 3. Scheme of atomic states (a) and of the quasienergy states (b) of the atom under conditions of two-photon excitation. The wavy lines in the diagrams correspond to spontaneous transition frequencies.

$$d_{31}^{(-)}(t) = -\frac{W}{|W|} A_2 d_1 \exp(-i\nu_1 t),$$
  

$$d_{13}^{(-)}(t) = A_1 d_2 \exp(-i\nu_4 t),$$
  

$$d_{32}^{(-)}(t) = A_1 d_1 \exp(-i\nu_2 t),$$
  

$$d_{23}^{(-)}(t) = -\frac{W}{|W|} A_2 d_2 \exp(-i\nu_3 t),$$
(6.2)

where  $d_1$  and  $d_2$  are the matrix elements of the atomic dipole transitions  $|\varphi_2\rangle \rightarrow |\varphi_3\rangle$  and  $|\varphi_3\rangle \rightarrow |\varphi_1\rangle$ ,  $A_{1,2} = (1 \pm \delta/\Omega_2)^{1/2}/\sqrt{2}$ ,  $\delta = \omega_{21} - 2\omega > 0$  is the two-photon offset from resonance, and  $\Omega_2 = (\delta^2 + 4|W|^2)^{1/2}$  is the Rabi frequency. The spectral lines of this system are determined by the transitions between the quasienergy states (6.2) and have the frequencies

$$\nu_{1,2} = \omega_{23} - \frac{1}{2}(\delta \pm \Omega_2), \quad \nu_{3,4} = \omega_{31} - \frac{1}{2}(\delta \pm \Omega_2).$$
 (6.3)

Our aim is to investigate effects due to the interaction of the perturbed atomic system with the squeezed vacuum at  $\omega_s = \omega$  in the transition probabilities between quasienergy states of this system, in the resonance fluorescence spectrum, and in the populations of the atomic energy levels. We note that in the ordinary vacuum this system was considered in Ref. 32 and, for a step configuration, in Ref. 33.

## 6.1. Interference effects in the transition probabilities

The calculations of the transition probabilities per unit time between the quasienergy states of the considered system in accordance with (3.7) and by means of the expressions (6.2) lead to the results

$$W_{31,(32)} = A_{1,2}^{2} \gamma_{2} (N_{2} + 1) + A_{2,1}^{2} \gamma_{1} N_{1}$$
  

$$\mp 2 \sqrt{\gamma_{1} \gamma_{2}} A_{1} A_{2} |M| \cos \psi,$$
  

$$W_{13,(23)} = A_{1,2}^{2} \gamma_{2} N_{2} + A_{2,1}^{2} \gamma_{1} (N_{1} + 1)$$
  

$$\mp 2 \sqrt{\gamma_{1} \gamma_{2}} A_{1} A_{2} |M| \cos \psi.$$
(6.4)

Here  $N_1$  and  $N_2$  are the numbers of photons at the frequencies  $\omega_{23}$  and  $\omega_{31}$  of the atomic transitions,  $\gamma_1$  and  $\gamma_2$  are the spontaneous partial widths of the atomic transitions

 $|\varphi_2\rangle \rightarrow |\varphi_3\rangle$  and  $|\varphi_3\rangle \rightarrow |\varphi_1\rangle$ , respectively, and  $\psi = \psi_s - 2\psi_L$ , where  $\psi_L$  is the phase of the excitation field; the probabilities of the dipole transitions  $|2\rangle \rightarrow |1\rangle$  are zero.

In obtaining these results, we have used the symmetry property  $M(\omega_{31}) = M(\omega_{23}) = |M| \exp(i\psi_s)$  of the correlation function, and in the parameters  $N_i$  and M the shifts of the atomic levels have been omitted.

The phase-dependent contribution to (6.4) leads to interference effects that under certain conditions suppress the probabilities of specific transitions. These effects are most clearly manifested for squeezing with the minimum of the uncertainty relation. In this case, it follows from (2.4) that  $|M|^2 = N_2(N_1+1)$  if  $N_1 > N_2$  and that  $|M|^2 = N_1(N_2+1)$  if  $N_2 > N_1$ . In these cases and when  $\psi = 0$  the probabilities of the transitions  $|3\rangle \rightleftharpoons |1\rangle$  can be suppressed, namely:

$$W_{31} = 0$$
 for  $N_2 > N$ , if  $\frac{q}{\eta^2} (\sqrt{1 + \eta^2} + 1)^2 = \frac{N_1}{N_2 + 1}$ ,  
(6.5)

$$W_{13} = 0$$
 for  $N_1 > N_2$ , if  $\frac{q}{\eta^2} (\sqrt{1 + \eta^2} + 1)^2 = \frac{N_1 + 1}{N_2}$ ,  
(6.6)

where  $\eta = 2|W|/\delta$  and  $q = \gamma_2/\gamma_1$ .

For the different relationship  $\psi = \pi$  between the phases, the probabilities of the transitions  $|2\rangle \rightleftharpoons |3\rangle\rangle$  can be suppressed:

$$W_{23} = 0$$
 for  $N_1 > N_2$ , if  $\frac{q}{\eta^2} (\sqrt{1 + \eta^2} - 1)^2 = \frac{N_1 + 1}{N_2}$ ,  
(6.7)

$$W_{32}=0$$
 for  $N_2 > N_1$ , if  $\frac{q}{\eta^2} (\sqrt{1+\eta^2}-1)^2 = \frac{N_1}{N_2+1}$ .  
(6.8)

The transition probabilities determine the radiative widths of the quasienergies in accordance with (3.9):

$$\Gamma_{1} = W_{13}/2, \quad \Gamma_{2} = W_{23}/2,$$
  

$$\Gamma_{3} = \frac{1}{2} (W_{31} + W_{32}) = \frac{\gamma_{2}}{2} (N_{1} + 1) + \frac{\gamma_{1}}{2} N_{1}. \quad (6.9)$$

where the phase-dependent contributions cancel for the width  $\Gamma_3$ .

We note that the resulting phase-dependent effects in the transition probabilities between the quasienergy states can be investigated in the case of excitation of atoms by short pulses  $(\tau \ll \gamma_1^{-1}, \gamma_2^{-1})$  in the regime of adiabatic onset of the interaction. In addition, they can be investigated through the widths  $\Gamma_1$  and  $\Gamma_2$  by a measurement of the spectral distributions of the light intensity in transitions from the quasienergy state  $|1\rangle$  or  $|2\rangle$  to any other states. In the following subsection, we give another consequence of these effects in the steady-state populations of the atomic levels.

## 6.2. Interference mechanism of population inversion of atomic levels

The equations for the density matrix in the secular approximation in the case of nonoverlapping spectral lines with  $\Omega_2 \gg \gamma_1(N_1+1)$ ,  $\gamma_2(N_2+1)$  have the same structure as Eqs. (3.1) and (3.2) and for the populations of the quasienergy states have the form

$$\dot{\sigma}_{11}(t) = -W_{12}\sigma_{11}(t) + W_{31}\sigma_{33}(t),$$
  

$$\dot{\sigma}_{22}(t) = -W_{23}\sigma_{22}(t) + W_{32}\sigma_{33}(t).$$
  

$$\dot{\sigma}_{33}(t) = -(W_{31} + W_{32})\sigma_{33}(t) + W_{13}\sigma_{11}(t) + W_{23}\sigma_{22}(t).$$
  
(6.10)

Solving these equations in the steady-state for  $t \gg \gamma_1^{-1}, \gamma_2^{-1}$  and then going over in the solutions to the basis

of the atomic states, we obtain after some manipulation the following results for the steady-state populations of the atomic levels:

$$\rho_{11,(22)} = \frac{1}{2(1+R_1+R_2)} \left( R_1 + R_2 \pm \frac{R_1 - R_2}{\sqrt{1+\eta^2}} \right),$$
  
$$\rho_{33} = \frac{1}{1+R_1 + R_2}, \qquad (6.11)$$

where

$$R_{1} = \frac{W_{31}}{W_{13}} = 1 + \frac{1 + q + (q - 1)\sqrt{1 + \eta^{2}}}{\sqrt{1 + \eta^{2}(1 + N_{1} + qN_{2}) + qN_{2} - N_{1} - 1 - 2\eta\sqrt{q}|M|\cos\psi}},$$

$$R_{2} = \frac{W_{32}}{W_{23}} = 1 - \frac{1 + q - (q - 1)\sqrt{1 + \eta^{2}}}{\sqrt{1 + \eta^{2}(1 + N_{1} + qN_{2}) - qN_{2} + N_{1} + 1 + 2\eta\sqrt{q}|M|\cos\psi}}.$$
(6.12)

It is readily seen that the quantum effects due to the interaction of the considered composite system with the squeezed vacuum are manifested in the steady-state populations through the transition probabilities between the quasienergy states. These effects lead to anomalous behavior of the populations in the parameter regions in which the probabilities (6.4) vanish.

We consider the case of squeezing with the minimum of the uncertainty relation, when  $N_1 > N_2$  and  $\psi = \pi$ , under the condition (6.7). In this parameter region, we have  $W_{23}=0$ , and from (6.11) and (6.12) we obtain

$$\rho_{22} - \rho_{11} = \frac{1}{\sqrt{1 + \eta^2}}, \quad \rho_{33} = 0.$$
(6.13)

Thus, in this parameter region the population of the first excited level is exactly zero,  $\rho_{11}$  and  $\rho_{22}$  depend only on  $\eta$ , and for  $\eta \leq 1$  we obtain complete inversion of the atomic levels ( $\rho_{22}\approx 1$ ,  $\rho_{11}\approx 0$ ,  $\rho_{33}=0$ ). It is readily verified that the conditions (6.7) are satisfied when  $\gamma_2 > \gamma_1$ . Such a result is explained by the fact that the interference of the phase-dependent effects completely suppresses the transition  $|2\rangle \rightarrow |3\rangle$  between the quasienergy states, while the transitions  $|2\rangle \rightarrow |1\rangle$  are forbidden in the dipole approximation. Therefore, in the steady-state regime  $\sigma_{11}=\sigma_{33}=0$  and  $\sigma_{22}=1$ , and this leads to a large population in the second excited atomic level.

We now consider the parameter region determined by the conditions (6.5) for  $\psi = 0$ . The conditions (6.5) can hold for atomic systems for which  $\gamma_2 < \gamma_1$ . In this parameter region,  $W_{31}=0$ , and, as a consequence, the ground atomic level has a very low population, while the populations of both excited levels are high. In this case, as follows from (6.11) and (6.12),

$$\rho_{22} - \rho_{11} = \frac{1}{(2+S)\sqrt{1+\eta^2}}, \quad \rho_{33} = \frac{1+S}{2+S},$$
(6.14)

where

$$S = \frac{1}{4N_1(1+\eta^2)} \left[ (\sqrt{1+\eta^2}+1)^2 - q \eta^2 \right].$$

Note that S>0 for all values of the parameters by virtue of the condition (6.5), and therefore there is a population inversion between the atomic levels coupled by the twophoton excitation:  $\rho_{22} > \rho_{11}$ . The maximum of the inversion is attained at large offsets from resonance and for  $N_1 \ge 1$ . In this case  $\rho_{11} \simeq 0$ ,  $\rho_{22} \simeq 1/2$ ,  $\rho_{33} \simeq 1/2$ . In the derivation of these results, it was important that interference suppresses the transition  $|3\rangle \rightarrow |1\rangle$  between the quasienergy states, leading, as a consequence, to vanishing of the steady-state population:  $\sigma_{11}=0$ .

The expressions (6.11) and (6.12) also describe the ordinary vacuum and for  $N_1 = N_2 = |M| = 0$  are identical to the results of Ref. 32 in the present approximation  $\Omega_2 \gg \gamma_1$ ,  $\gamma_2$ . In this case, we have for all finite values of the parameters qthe inequalities  $\rho_{11} > \rho_{22} > \rho_{33}$  for  $\gamma_2 > \gamma_1$  and  $\rho_{11} > \rho_{22}$ ,  $\rho_{33} > \rho_{22}$  for  $\gamma_2 < \gamma_1$ . A similar situation also occurs for a three-level atom in the step configuration.<sup>33</sup>

Figure 4 gives the dependence of the atomic populations on the parameter  $\eta$  for two regions of their anomalous behavior and, for comparison, for the ordinary vacuum. Note also that the anomalous behavior of the populations and the inversion of the atomic levels are due to the effects of the squeezed vacuum and occur neither in the ordinary vacuum nor in a thermal reservoir.

#### 6.3. Resonance fluorescence

It is readily seen that for the present system, the contributions of crossed type to the widths are equal to zero:  $\gamma_{\alpha\beta}^{(1)} = \gamma_{\alpha\beta}^{(2)} = 0$  ( $\alpha, \beta = 1, 2, 3$ ). This is due to the fact that these contributions (3.11) and (3.12) are determined by diagonal transitions between the quasienergy states, and the latter van-



FIG. 4. Dependence of populations of the atomic levels (curves 1, 2, 3, respectively, for  $\rho_{11}$ ,  $\rho_{22}$ ,  $\rho_{33}$ ) on the parameter  $\eta$  for the regions of their anomalous behavior for the following parameter values:  $N_1=9$ ,  $N_2=6$ , q=10,  $\psi=\pi$  (a);  $N_1=50$ ,  $N_2=83$ , q=0.1,  $\psi=0$  (b) and for the ordinary vacuum (c).

ish due to the fact that the dipole transition  $|\varphi_2\rangle \rightarrow |\varphi_1\rangle$  is forbidden. Therefore, the equations for the transition currents have the form

$$\dot{\sigma}_{13}(t) = -\Gamma_{13}\sigma_{13}(t),$$
  
$$\dot{\sigma}_{23}(t) = -\Gamma_{23}\sigma_{23}(t),$$
 (6.15)

where

$$\Gamma_{13} = \frac{1}{2}(W_{13} + W_{31} + W_{32}), \quad \Gamma_{23} = \frac{1}{2}(W_{23} + W_{31} + W_{32})$$
(6.16)

and the transition probabilities are given by (6.4).

The above results are sufficient to calculate the resonance fluorescence spectrum in accordance with (5.15) in the approximation of nonoverlapping spectral lines. The scheme of the calculations is described in Sec. 5.2 and leads to the following result for the spectrum:<sup>2</sup>

$$S(\omega_{k}) = \pi |\mathbf{e}^{*}(k)\mathbf{d}_{1}|^{2} \left[ \frac{A_{2}^{2}\sigma_{11}\Gamma_{13}}{(\omega_{k}-\nu_{1})^{2}+\Gamma_{13}^{2}} + \frac{A_{1}^{2}\sigma_{22}\Gamma_{23}}{(\omega_{k}-\nu_{2})^{2}+\Gamma_{23}^{2}} \right] + \pi |\mathbf{e}^{*}(k)\mathbf{d}_{2}|^{2}\sigma_{33} \left[ \frac{A_{1}^{2}\Gamma_{13}}{(\omega_{k}-\nu_{4})^{2}+\Gamma_{13}^{2}} + \frac{A_{2}^{2}\Gamma_{23}}{(\omega_{k}-\nu_{3})^{2}+\Gamma_{23}^{2}} \right],$$
(6.17)

in which the steady-state populations of the quasienergy states are

$$\sigma_{11,(22)} = \frac{R_{1,(2)}}{1 + R_1 + R_2}, \quad \sigma_{33} = \frac{1}{1 + R_1 + R_2}.$$
 (6.18)

The spectrum has four peaks at the frequencies of the transitions between the quasienergy states (6.3), and is very sensitive to the choice of parameters of the squeezed vacuum. In particular, it is a consequence of the interference effects of the squeezed vacuum that the spectrum becomes a single-peak spectrum in the vicinity of the frequency  $\nu_1$  when 6.6 holds, when  $W_{13}=0$ , and, therefore,  $\sigma_{22}=\sigma_{33}=0$ ,  $\sigma_{11}=1$ , and a single-peak spectrum in the vicinity of  $\nu_2$  when (6.7) holds, when  $W_{23}=0$  and, therefore,  $\sigma_{11}=\sigma_{33}=0$ ,  $\sigma_{22}=1$ . In other parameter regions, the spectrum may have three peaks. Thus, if (6.5) holds, yielding  $W_{31}=0$  and therefore  $\sigma_{11}=0$ , the emission line at  $\nu_1$  disappears, and in the range of parameter values determined by (6.8), when  $W_{32}=0$  and therefore  $\sigma_{22}=0$ , the emission line at  $\nu_2$  vanishes. The squeezed-vacuum effects also lead to a change in the spectral

widths of the fluorescence lines compared with the ordinary vacuum, but they do not decrease below the spontaneous widths  $\gamma_1$  and  $\gamma_2$ .

## 7. CONCLUSIONS

The results obtained in this paper show that the transitions between the quasienergy states of an atomic system in a laser field acquire some unusual properties under the influence of the field of a squeezed vacuum. A fundamentally new circumstance is that the transition probabilities per unit time between the quasienergy states contain quantum, phasedependent effects [see the expression (3.7)], by virtue of which they can be strongly suppressed compared with the ordinary vacuum. These effects have been found for all the three nonlinear optical systems considered in the paper [see the expressions (4.4), (4.5), (5.11), and (6.4)] and are reflected, in particular, in Fig. 1. They are also manifested in the steady-state regime of interaction of the atom in the resonance fluorescence spectra [expressions (5.19) and (6.17), Fig. 4]. On the basis of the phase-dependent effects in the transition probabilities between the quasienergy states, it is easy to understand why the width of the central line of the resonance fluorescence spectrum is reduced in a two-level atom to a value below the spontaneous width of the atomic transition.<sup>2</sup> The interference effects in the transition probabilities [the expressions (6.5)-(6.8)] also lead to anomalous behavior of the steady-state populations of the atomic levels (Fig. 4). It may be said that we have found a new mechanism for occurrence of population inversion of atomic levels based on suppression of the probabilities of certain transitions between quasienergy states.

I thank K. V. Kheruntsyan for helpful discussions. This work was in part supported by the International Science Foundation (Grant RY-6000).

<sup>1)</sup>For an atom in a monochromatic field and in an ordinary vacuum, this element was investigated in Ref. 15.

<sup>2)</sup>The fluorescence spectrum for the considered system in the ordinary vacuum is calculated in Ref. 34.

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Translated by Julian B. Barbour