Influence of nonlinear dynamic relaxation on the absorption and emission spectra of a two-level atom in a strong electromagnetic field

A. A. Panteleev and A. N. Starostin

Troitsk Institute of Innovative and Thermonuclear Research, 142092 Troitsk, Moscow Region, Russia
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The standard equations for the density matrix describing a two-level atom in a strong electromagnetic field have been corrected for the variation of the radiative relaxation structure and the low-frequency component of the Lamb shift. It is shown that two-level atoms may exist in an inverted state when there is high-frequency detuning of the strong electromagnetic field from resonance. The influence of nonlinear dynamic relaxation on the emission and absorption (or amplification) spectra of a two-level atom in a strong electromagnetic field has been investigated. © *1994 American Institute of Physics.*

1. INTRODUCTION

The investigation of the properties of the interaction of intense radiation with a two-level system (or a two-level atom) is of fundamental significance for understanding processes involving the interaction of radiation with matter, as well as in applications associated with quantum and nonlinear optics, laser physics, etc. The numerous phenomena appearing as a result of the interaction of an intense electromagnetic field with a two-level system have been the subject of an enormous number of investigations (see, for example, Refs. 1–3). The most distinct manifestations of the interaction of a strong resonant electromagnetic field with a two-level system include bleaching of the medium (saturation) and alteration of the emission and absorption spectra of the two-level system.[3–6]

Two-level atoms in an electromagnetic field are usually described using the density-matrix formalism[3] or the optical Bloch equations,[1] in which the interaction of a two-level atom with an electromagnetic wave is taken into account in the dipole approximation and the relaxation is described by a set of constants that characterize the lifetime of the excited state (T_1) and the polarization relaxation time (T_2).

A strong electromagnetic field can have a significant influence on the relaxation of atoms. The dependence of the relaxation constants, which are determined by the interaction with a thermostat or with impurity particles through collisions, on the intensity of the electromagnetic field was discussed in detail in Pestrov's work[7] (see also the references therein). It was shown in Ref. 7 that the relaxation of the atomic subsystem has a highly nonlinear character in a strong electromagnetic field as a consequence of field narrowing:[8]

$$\Omega\tau_c \gg 1, \tag{1}$$

(where Ω is the Rabi frequency of the two-level system and τ_c is the collision time or the noise correlation relaxation time).

We note that the influence of a strong electromagnetic field on the free induction decay rate has been observed in the optical region in $LaF_3:Pr^{3+}$ (Ref. 9) and ruby (Ref. 10) crystals. It was established experimentally that the observed plots of the phase relaxation contradict the theoretical results following from the standard Bloch equations (or the density matrix) with the constant relaxation parameters T_1 and T_2. These experiments lead to the appearance of several theoretical papers[11–17] based on the dependence of the relaxation constants on the intensity of the electromagnetic field.

In the present work we investigated the influence of the effects of nonlinear dynamic radiative relaxation on the properties of a two-level atom in a strong electromagnetic field. It is usually assumed that an electromagnetic field does not influence the spontaneous relaxation of atoms. However, it was shown in a paper by Fanchenko[18] that the standard description based on the density-matrix formalism in fairly strong fields, i.e., on the relation

$$\Omega \geqslant \sqrt{\omega_0|\Delta|}, \tag{2}$$

has restrictions associated with the dependence of the radiative relaxation constants on the intensity of the pump field. Here we have set $\Delta=\omega_L-\omega_0+i\gamma/2$, where ω_0 and ω_L are the transition frequency and the frequency of the incident radiation, respectively, and γ is the spontaneous relaxation rate: $\gamma=1/T_1=2/T_2$. As was shown in Ref. 18, a dynamic equilibrium state is established under condition (2) due to variation of the values of the frequencies of the radiative transitions between the quasienergy states which appear as a consequence of the high-frequency Stark effect. This keeps the refractive index of the medium from reaching complete saturation and makes it follow a constant asymptote. This result was confirmed by the investigations of Pestov[7] and Vinogradov.[19] The present work is devoted to deriving equations for a two-level atom in a strong electromagnetic field and to analyzing the photon absorption and emission spectra under condition (2).

2. EQUATIONS FOR THE DENSITY MATRIX OF A TWO-LEVEL ATOM IN A STRONG ELECTROMAGNETIC FIELD

Fanchenko[18] used the Keldysh diagram technique for nonequilibrium Green's functions.[20] This description provides a graphic picture of the quasienergy states of an atom. However, calculations of the emission and absorption spectra of an atom in an intense electromagnetic field are fairly com-

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plicated: second-order perturbation theory with respect to the coupling constant (the single-loop approximation) is inadequate, i.e., ladder-type vertex functions must be inserted to solve the problem exactly.[21,22] This remark also applies to the mass operators, which describe the relaxation of the atomic subsystem. Fanchenko[18] used only second-order perturbation theory with respect to the coupling constant, which, as was indicated above, is inadequate for an exact description of the radiative relaxation of an atom in treating the real emission spectrum. Moreover, Fanchenko[18] did not obtain modified equations of the density matrix incorporating the effects of nonlinear dynamic relaxation.

In the present work we used the Scully–Lamb atom–photon density matrix formalism[23] to describe the properties of a two-level atom in an intense electromagnetic field. One advantage of this approach over the Keldysh diagram technique is the relative simplicity of the calculation of the emission and absorption spectra of an atom (see, for example, Refs. 24 and 25) and the description of relaxation processes. In addition, it is convenient for generalizing the theory to the case of the interaction between an electromagnetic wave and a multilevel medium. We note that the atom–photon density matrix formalism underwent considerable development in the work of Sargent *et al.*[25–27]

The equation for an atom-photon density matrix has the form (in rad/s)

$$i\dot{\rho}_{\text{a-ph}}=[\hat{H};\rho_{\text{a-ph}}]+i\Gamma(\rho_{\text{a-ph}}), \tag{3}$$

where the operator $\Gamma(\rho_{\text{a-ph}})$ describes the relaxation processes. We represent the Hamiltonian of the system under consideration in the form

$$\hat{H}=\hat{H}_0+\hat{H}_{\text{r}}+\hat{H}_{\text{i}}+\hat{H}_{\text{q}}, \tag{4}$$

where the first two terms describe the Hamiltonian of the unperturbed atomic subsystem and the free radiation field:

$$\hat{H}_0=-\Delta_{\text{L}}\sigma_z; \quad \hat{H}_{\text{r}}=\sum_k \nu_k a_k^+ a_k. \tag{5}$$

Here a_k^+ and a_k are the creation and annihilation operators of photons with a wave vector $\mathbf{k}$ and a frequency ω_k; σ is an ordinary Pauli matrix. The energy of the atoms and photons is calculated relative to the energy of the quanta of a monochromatic electromagnetic field with a frequency ω_{L}: $\nu_k=\omega_k-\omega_{\text{L}}$; $\Delta_{\text{L}}=\omega_{\text{L}}-\omega_0$. The last two terms in expression (4) describe the interaction of the atomic subsystem with the electromagnetic wave and the quantized radiation field, which we represent using the rotating wave approximation in the form

$$\hat{H}_1=\sigma^- V^*+\sigma^+ V, \tag{6}$$

$$\hat{H}_{\text{q}}=\sum_k (g\sigma^+ a_k+g^*\sigma^- a_k^+), \tag{7}$$

where $V=-\mu_{21}E_{\text{L}}/2\hbar$, E_{L} is the amplitude of the electromagnetic field intensity, μ_{21} is the matrix element describing the dipole moment of the $2\rightarrow1$ transition, $g=-i\mu_{21}\sqrt{2\pi\omega_k/\hbar W}$ is the coupling constant (the vacuum Rabi frequency), and W is the quantization volume.

Performing the convolution in (3) with respect to the photon variables, we obtain an equation for the atomic density matrix ρ, and, conversely, taking the trace with respect to the atomic states, we have the equation for the photon field operator P. In the zeroth approximation, for the atomic density matrix we have the ordinary equation

$$i\dot{\rho}^{(0)}=[H_0+H_{\text{i}};\rho^{(0)}]+i\Gamma(\rho^{(0)}). \tag{8}$$

Here the operator Γ has the standard structure[3] corresponding to ordinary relaxation processes, and the interaction with the quantized radiation field is not taken into account. For electromagnetic fields of low intensity, i.e., when the line profile in the spontaneous emission spectrum of a two-level atom in an electromagnetic field is not distorted significantly, the introduction of radiative relaxation constants is a sound procedure (see, for example, Refs. 3 and 23). To systematically take into account the radiative relaxation processes of a two-level atom in an intense electromagnetic field, in the next step we take into account the interaction of the atomic subsystem with the quantized radiation field. The phenomenologically introduced relaxation operator should be omitted. Following the approach developed in Ref. 28, where the formalism used here was employed to describe radiation transfer processes in a resonant medium in the presence of a laser field, we have the following expressions for the components ρ_{js} of the atomic density matrix:

$$\begin{aligned}
i\dot{\rho}_{22}&=V\rho_{12}-\rho_{21}V^*+\sum_k (g\langle\Phi_{k12}^-\rangle-g^*\langle(\Phi_k^-)_{21}^+\rangle),\\
i\dot{\rho}_{11}&=V\rho_{21}-\rho_{12}V+\sum_k (g^*\langle\Phi_{k21}^+\rangle-g\langle(\Phi_k^+)_{12}^+\rangle),\\
i\dot{\rho}_{12}&=\Delta_{\text{L}}\rho_{12}+V^*(\rho_{22}-\rho_{11})+\sum_k g^*(\langle\Phi_{k22}^+\rangle\\
&\quad-\langle(\Phi_k^-)_{11}^+\rangle),\\
i\dot{\rho}_{21}&=-\Delta_{\text{L}}\rho_{21}+V(\rho_{11}-\rho_{22})+\sum_k g(\langle\Phi_{k11}^-\rangle\\
&\quad-\langle(\Phi_k^+)_{22}^+\rangle),
\end{aligned} \tag{9}$$

where the operators Φ_k are defined in the following manner: $\Phi_K^-=a_k\rho_{\text{a-ph}}$, $\Phi_K^+=a_k^+\rho_{\text{a-ph}}$. When these operators are calculated (see Refs. 25 and 26), the solution of the problem of deriving the equations for ρ and Φ_k must be self-consistent. We solve this problem by iterations. In a first approximation we adopt the usual radiative relaxation structure in the equations for Φ_k, and we use the standard solution of Eq. (8) for the values of the components of the atomic density matrix. Then for Φ_k^- (see Refs. 25 and 26, as well as 28) we have the expressions

$$\begin{aligned}
\Phi_{k22}^-&=g^*a_k a_k^+ P(m_{21}\rho_{21}^{(0)}-m_{31}\rho_{22}^{(0)})\\
&\quad+g^*a_k P a_k^+(m_{11}\rho_{21}^{(0)}+m_{31}\rho_{11}^{(0)}),\\
\Phi_{k11}^-&=-g^*a_k a_k^+ P(m_{22}\rho_{21}^{(0)}-m_{32}\rho_{22}^{(0)})\\
&\quad-g^*a_k P a_k^+(m_{12}\rho_{21}^{(0)}+m_{32}\rho_{11}^{(0)}),
\end{aligned}$$

$$\Phi_{k12}^{-}=g^{*}a_{k}a_{k}^{+}P(m_{23}\rho_{21}^{(0)}-m_{33}\rho_{22}^{(0)})$$
$$+g^{*}a_{k}Pa_{k}^{+}(m_{13}\rho_{21}^{(0)}+m_{33}\rho_{11}^{(0)}),$$
$$\Phi_{k21}^{-}=-g^{*}a_{k}a_{k}^{+}P(m_{24}\rho_{21}^{(0)}-m_{34}\rho_{22}^{(0)})$$
$$-g^{*}a_{k}Pa_{k}^{+}(m_{14}\rho_{21}^{(0)}+m_{34}\rho_{11}^{(0)}), \qquad (10)$$

where $m_{js}=M_{js}/\nu f(\nu)$, and $f(\nu)=(\nu-\Delta)(\nu+\Delta^{*})(\nu-i\gamma)-2|V|^{2}(2\nu-i\gamma)$ is Mollow's well-known cubic polynomial.[5] For M_{js} we have the expressions

$$M_{11}=\nu(\nu-\Delta)(\nu+\Delta^{*})-|V|^{2}(2\nu-i\gamma),$$
$$M_{13}=-V^{*}(\nu-\Delta)(\nu+i\gamma),$$
$$M_{12}=(\nu-\Delta)(\nu+\Delta^{*})i\gamma+|V|^{2}(2\nu-i\gamma),$$
$$M_{14}=-V(\nu+\Delta^{*})(\nu+i\gamma),$$
$$M_{21}=|V|^{2}(2\nu-i\gamma), \quad M_{22}=-|V|^{2}(2\nu-i\gamma)+(\nu-\Delta)\times(\nu+\Delta^{*})(\nu-i\gamma),$$
$$M_{23}=-V^{*}(\nu-\Delta)(\nu-i\gamma), \quad M_{24}=-V(\nu+\Delta^{*})(\nu-i\gamma); \quad M_{31}=-V\nu(\nu-\Delta),$$
$$M_{33}=\nu[(\nu-\Delta)(\nu-i\gamma)-2|V|^{2}],$$
$$M_{34}=2V^{2}\nu; \quad M_{32}=M_{31}. \qquad (11)$$

Here the index k has been omitted in the detuning of ν. The value of γ is determined by the rate of radiative decay of the excited state, and $\Delta=\Delta_{L}+i\gamma/2$. For the components of the atomic density matrix $\rho_{js}^{(0)}$, which comprise the stationary solution of Eq. (8), we have the well-known expressions

$$\rho_{22}^{(0)}=\frac{|V|^{2}}{|\Delta|^{2}+2|V|^{2}}, \quad \rho_{12}^{(0)}=\frac{V^{*}\Delta}{|\Delta|^{2}+2|V|^{2}},$$
$$\rho_{11}^{(0)}=1-\rho_{22}^{(0)}, \quad \rho_{21}^{(0)}=(\rho_{12}^{(0)})^{*}. \qquad (12)$$

In expressions (10) we have omitted the terms corresponding to forward four-wave mixing processes,[25,26,28] which are beyond the scope of our treatment. The quantities $\langle\Phi_{k}\rangle$ contain terms of two types: terms which describe the emission of radiation by atoms and are proportional to $\bar{n}_{k}+1$, and terms which describe the absorption of radiation and are proportional to $\bar{n}_{k}$. Here $\bar{n}=\langle Pa^{+}a\rangle=\Sigma_{n}P_{n}n$ is the photon occupation number of the mode in question; P_{n} is the probability of finding n photons, $\Sigma P_{n}=1$. In expressions (10) Φ_{kjs}^{-} is $\propto a_{k}a_{k}^{+}P$ and $\propto a_{k}Pa_{k}^{+}$.

For moderate values of these quantities we have

$$\langle aa^{+}P\rangle=\sum_{n}P_{n}(n+1)=\bar{n}+1, \qquad (13)$$

$$\langle aPa^{+}\rangle=\sum_{n}P_{n+1}(n+1)=\bar{n}. \qquad (14)$$

Here the index k has been omitted.

To investigate the influence of a strong electromagnetic field on radiative relaxation processes we neglect the driven processes in Eqs. (9), i.e., we set $\bar{n}_{k}=0$. Using expressions (10) and (11) and making the transition in (9) from summation to integration, we obtain the equations

$$i\dot{\rho}_{22}=+V\rho_{12}-\rho_{21}V^{*}+\kappa\int_{-\omega_{L}}^{\tilde{\omega}-\omega_{L}}\frac{d\nu_{k}}{2\pi}\,\omega_{k}^{3}\left\{\frac{\rho_{21}V^{*}(\nu_{k}-i\gamma)(\nu_{k}-\Delta)+\rho_{22}\nu_{k}[(\nu_{k}-i\gamma)(\nu_{k}-\Delta)-2|V|^{2}]}{\nu_{k}f(\nu_{k})}-\text{c.c.}\right\}, \qquad (15)$$

$$i\dot{\rho}_{21}=\Delta_{L}\rho_{21}+V(\rho_{11}-\rho_{22})-\kappa\int_{-\omega_{L}}^{\tilde{\omega}-\omega_{L}}\frac{d\nu_{k}}{2\pi}\,\omega_{k}^{3}\left\{\frac{\rho_{21}(\nu_{k}-i\gamma)(\nu_{k}-\Delta)(\nu_{k}+\Delta^{*})+2\rho_{22}V\nu_{k}(\nu_{k}-\Delta)}{\nu_{k}f(\nu_{k})}\right\}. \qquad (16)$$

Here $\kappa=4|\mu|^{2}/3\hbar c^{3}$, and $\tilde{\omega}$ is the cut-off frequency. The last terms in these equations describe the radiative decay of a two-level atom. In the integrands in Eqs. (15) and (16) the components of the density matrix $\rho_{js}^{(0)}$ have been replaced by the ρ_{js} appearing in these equations. This procedure is substantiated by the second iteration of the equations for the operators Φ_{k}, in which the values of the components of the density matrix ρ_{js} are used. Our analysis shows that the ordinary radiative relaxation structure can be used for this iteration of Φ_{k} as well, since consideration of the terms corresponding to effects of nonlinear dynamic relaxation [see Eqs. (19) and (20) below] produces only minor corrections in Eqs. (15) and (16). In expression (15) the part of the integrand in curly brackets describes the resonance fluorescence spectrum of the two-level atom. We note that the polarization decay spectrum, i.e., the integrand in (16), differs from the fluorescence spectrum.

In weak fields the introduction of phenomenological relaxation constants, i.e., the Wigner–Weisskopf procedure, and consideration of the electrodynamic level shift imply the following replacement

$$I(\Delta_{L},\Gamma)=\kappa\int_{-\omega_{L}}^{\tilde{\omega}-\omega_{L}}\frac{d\nu_{k}}{2\pi}\,\frac{1}{\nu_{k}-\Delta_{L}-i\Gamma}\rightarrow i\gamma/2+\Delta_{LFLS}, \qquad (17)$$

where the resonant approximation can be used to move ω_{k}^{3} outside the integral sign at a pole of the integrand. Here Δ_{LFLS} describes the low-frequency part of the Lamb shift[29] of a two-level atom. We note that the constant Γ, which describes the width of the emission line, appears when relaxation is described with consideration of the real profile of the emission line. The value of Γ depends on the conditions under which the atom is excited. In a purely radiative decay

regime with excitation of the atom by a weak monochromatic field we have $\Gamma \to 0$, while in the case of excitation by broad-band radiation we have $\Gamma = \gamma/2$. If the atoms undergo collisions, Γ is determined by the collisional broadening. However, when the cut-off frequency is properly introduced, the rate of spontaneous emission does not depend on Γ. Following Fanchenko,[18] we determined the cut-off frequency for a two-level atom from the condition for validity of the dipole approximation $\omega_k \leqslant c/a_0$, where a_0 is the characteristic dimension of the atom. In this case we have

$$\tilde{\omega} \sim c/a_0 \sim \omega_0 \sqrt{\alpha \frac{\omega_0}{\gamma}} \sim \frac{\omega_0}{\alpha}, \tag{18}$$

where α is the constant of the fine structure. We note that in the widely adopted description of the Wigner–Weisskopf procedure the integration limits in expressions like (17) are extended to infinity,[3,23,29] and it is assumed that $\Gamma \to 0$ in the denominator in the integrand in (17). The introduction of a cut-off frequency under condition (18) makes it possible to avoid moving ω_k^3 outside the integral sign. Then the usual expression $\gamma = 4|\mu|^2\omega_0^3/3\hbar c^3[1+O(x)]$, and $x = \sqrt{\alpha\gamma/\omega_0}$ hold, to within corrections which may be neglected.

Using these rules for evaluating the integrals, we find corrections to the relaxation processes with an accuracy to $o(V/\omega_L)$:

$$i\dot{\rho}_{22} = -i\gamma\rho_{22} + V\rho_{12} - \rho_{21}V^* + \{\Sigma(\rho_{21}V^* - \rho_{22}\Delta_L) - \text{c.c.}\}, \tag{19}$$

$$i\dot{\rho}_{21} = -\Delta\rho_{21} + V(\rho_{11} - \rho_{22}) - 2V\rho_{22}\Sigma, \tag{20}$$

$$\rho_{11} = 1 - \rho_{22}, \quad \rho_{12} = \rho_{21}^*, \tag{21}$$

where

$$\Sigma = \left[I\left(-2V, \frac{3\gamma}{4}\right) - I\left(2V, \frac{3\gamma}{4}\right)\right] \frac{1}{2V} \approx \frac{3\gamma}{2\omega_L}\left(i + \frac{\Lambda}{\pi}\right),$$

and $\Lambda = \ln(\tilde{\omega}/\omega_L)$ is the cut-off logarithm. It is assumed that the usual parameter Δ_{LFLS}, as well as the high-frequency component of the Lamb shift, contribute to the transition frequency of the two-level atom ω_0. Equations (19) and (20) are similar to the equations obtained in Pestov's work,[7] but along with the corrections due to nonlinear dynamic relaxation, they contain additional corrections due to the nonlinear low-frequency component of the Lamb shift. The latter also did not appear in Ref. 18.

The stationary solution of Eq. (20) has the form

$$\rho_{21} = \frac{V(\Delta^* + 2|V|^2(\Sigma^* - \Sigma)/i\gamma)}{|\Delta|^2 + 2|V|^2}, \tag{22}$$

and for the population difference in this case we have the expression

$$\rho_{11} - \rho_{22} = \frac{|\Delta|^2 - 2|V|^2(\Sigma\Delta^* - \Sigma^*\Delta)/i\gamma}{|\Delta|^2 + 2|V|^2}. \tag{23}$$

In sufficiently strong fields, i.e., under condition (2), expression (22), which specifies the polarizability of the atom, has the form

$$\rho_{21} \approx 3V/\omega_0, \tag{24}$$

and coincides with the result in Ref. 18, where it was shown for a strong electromagnetic field that the refractive index does not achieve complete saturation. It can be seen from expression (22) that this result does not depend on the corrections due to the nonlinear low-frequency component of the Lamb shift; therefore, the result obtained in Ref. 18 remained unchanged.

Under condition (2) it can be seen from (23) that population inversion can occur when there is high-frequency detuning ($\Delta_L > 0$) of the strong electromagnetic field:

$$\rho_{11} - \rho_{22} \approx 3\left(\frac{\Lambda\gamma}{\pi\omega_0} - \frac{\Delta_L}{\omega_0}\right). \tag{25}$$

In this case radiation is absorbed [see (22)]. In quantum optics there are numerous effects (for example, amplification of a test signal in the presence of a strong field in two-level systems,[6] various laser schemes without population inversion,[30,31] etc.) under which radiation is amplified without inversion, and the situation under discussion here has the opposite character, i.e., despite the occurrence of inversion, radiation is absorbed.

3. INFLUENCE OF THE EFFECTS OF NONLINEAR DYNAMIC RELAXATION ON THE ABSORPTION AND EMISSION SPECTRA OF A TWO-LEVEL ATOM IN A STRONG ELECTROMAGNETIC FIELD

A direct experimental investigation of manifestations of the effects of nonlinear dynamic radiative relaxation upon the interaction of a strong electromagnetic field with a two-level atom would be difficult. It follows from expression (22) that the absorption coefficient does not differ from the usual result [compare with (12)]. Measuring the refractive index of a medium in this case is a technically complicated task. Direct measurement of the populations of the levels and, in particular, the observation of population inversion, which is possible according to Eq. (23), are also scarcely feasible. For example, the population of the upper level can be found from the total intensity (integrated over all frequencies) of the resonance fluorescence, which is proportional to $\hbar\omega_0\rho_{22}$; however, the difference between ρ_{22} and $\rho_{22}^{(0)}$ is so small that it is very difficult to detect experimentally. Therefore, experimental investigations of indirect manifestations of these effects would be of interest. They include measurements of the resonance fluorescence spectra and the absorption coefficient of a test signal.

It is well known[5,6] that the absorption and emission spectra of a two-level atom undergo significant changes in an intense monochromatic electromagnetic field ($\Omega > |\Delta|$). The resonance fluorescence spectrum then consists of a triplet of Lorentzian lines separated by the generalized Rabi frequency $\Omega' = \sqrt{\Delta_L^2 + 4|V|^2}$. The absorption spectrum of the test signal also changes considerably, and absorption may be replaced by amplification in some regions of the spectrum. We note that according to the results in Refs. 22 and 25, in a gas of two-level atoms in the presence of an intense monochromatic electromagnetic field the absorption coefficients (or gains) for a classical test signal and for a quantized radiation field coincide.

Let us examine the influence of nonlinear dynamic relaxation on the absorption and emission spectra of a two-level atom in a strong electromagnetic field. The procedure for calculating these quantities with the atom-photon density matrix formalism is described in Refs. 25 and 26. Using this approach we find that the emission spectrum $A(\nu_k)$ (the resonance fluorescence spectrum) of a two-level atom is determined by the portion of the integrand in curly brackets in Eq. (15). The components of the atomic density matrix ρ_{js} then comprise the stationary solution of system of equations (19)–(21) [see (22) and (23)]. The result obtained is similar to Mollow's spectrum,[5] which is obtained by replacing ρ_{js} by $\rho_{js}^{(0)}$. In strong fields the inelastic component of the fluorescence spectrum dominates. The inelastic part of $A(\nu_k)$ is then nearly identical [to $o(V/\omega_L)$] with the inelastic component of Mollow's spectrum. There is a significant difference in the behavior of the elastic or coherent component of these spectra. The expression for the elastic component of $A(\nu_k)$ has the form

$$A^{el} \propto 2\pi|\rho_{21}|^2\delta(\nu_k), \tag{26}$$

where ρ_{21} is specified by expression (22) in the case under consideration here and is replaced by $\rho_{21}^{(0)}$ in the case of Mollow's spectrum [see (12)]. Here $\delta(\nu_k)$ is the Dirac delta function. Since under condition (2) $\rho_{21}^{(0)} \to 0$, and $\rho_{21} \to 3V/\omega_L$, the influence of the effects of nonlinear dynamic relaxation cause the elastic component of the fluorescence spectrum not only not to achieve saturation, in contrast to Mollow's results, but also to begin to increase. However, as before, its magnitude is much smaller than the contribution of the inelastic component. Therefore, it is difficult to observe this effect experimentally.

The expression for the absorption (or amplification) spectrum of a two-level atom in a strong electromagnetic field [the absorption coefficient (or gain) of the test signal] has the form

$$\mathcal{K}(\nu) = \mathcal{K}_0 \operatorname{Re}\left(i\gamma \frac{\rho_{21}V^*(\nu - i\gamma) - (\rho_{11} - \rho_{22})[(\nu - \Delta)(\nu - i\gamma) - 2|V|^2]/2}{f(\nu)} \right), \tag{27}$$

where $\mathcal{K}_0$ is the saturated absorption coefficient at the center of the line. We note that this result can be obtained from Eqs. (19)–(21) by means of perturbation theory by adding the test field to them. Like the expressions for the spectra discussed above, expression (27) is also distinguished from the familiar result[6] by the replacement of ρ_{js} by $\rho_{js}^{(0)}$. The influence of the effects of nonlinear dynamic relaxation on the absorption spectra of a two-level atom in a strong electromagnetic field is demonstrated in Fig. 1, which reveals that consideration of these effects can result not only in changes in the absolute values of the absorption coefficient, but also in variation of the spectral regions for amplification of the radiation. Calculations were performed for typical parameters of the optical range, viz., $\omega_0/\gamma = 5\times10^7$ and $\Lambda/\pi = 2$, when $V/\gamma = 3\times10^3$. Even under these conditions, when we have $\Omega \approx 2V \sim \sqrt{\omega_0|\Delta|}$, the role of these effects is appreciable. Under condition (2) expression (27) has the following asymptotic form:

$$\mathcal{K}(\nu) \approx \frac{\Omega\mathcal{K}_0}{2\omega_0}\left(\frac{9\gamma^2/16}{(\nu+\Omega)^2 + 9\gamma^2/16} - \frac{9\gamma^2/16}{(\nu-\Omega)^2 + 9\gamma^2/16} \right). \tag{28}$$

From this formula it can be seen that, first, the absorption coefficient (or gain) of the test signal increases with increasing intensity. Second, the positions of the amplification and absorption regions do not depend on the sign of the detuning Δ_L of the pump field from resonance: low-frequency components are amplified, and high-frequency components are absorbed. On the other hand, if the effects of nonlinear dynamic relaxation at the spectral maxima are disregarded under condition (2), $\mathcal{K}(\nu) \propto 1/\Omega$, and the positions of the amplification and absorption regions are strongly dependent on the magnitude and sign of Δ_L.

The widely accepted interpretation of the emission and absorption spectra of a two-level atom in a strong electromagnetic field was based on the properties of the high-frequency Stark effect: the atomic levels split into four quasienergy levels.[2] The resonance fluorescence spectrum may be interpreted as spontaneous transitions between these levels, and the absorption (or amplification) of the test signal may be interpreted as Raman scattering on the system of quasienergy levels. The interesting physical interpretation of the absorption coefficient (or gain) of the test signal proposed by Agarwal in Ref. 32 is also noted. The investigation performed in that work showed that in sufficiently strong fields [see (2)] the differences in the rates of the radiative transitions between the quasienergy states, which cause changes in their population dynamics, must be taken into account.

4. CONCLUSIONS

Thus, in the present work the standard equations for the density matrix describing a two-level atom in a strong electromagnetic field have been corrected for the variation of the radiative relaxation structure and the low-frequency component of the Lamb shift. Fanchenko's conclusion[18] that the refractive index cannot be saturated by a strong electromagnetic field in a gas of two-level atoms has been confirmed. It has been shown under these conditions that two-level atoms may exist in an inverted state when there is high-frequency detuning of the strong electromagnetic field from resonance. The pump field is then absorbed, and the absorption coeffi-

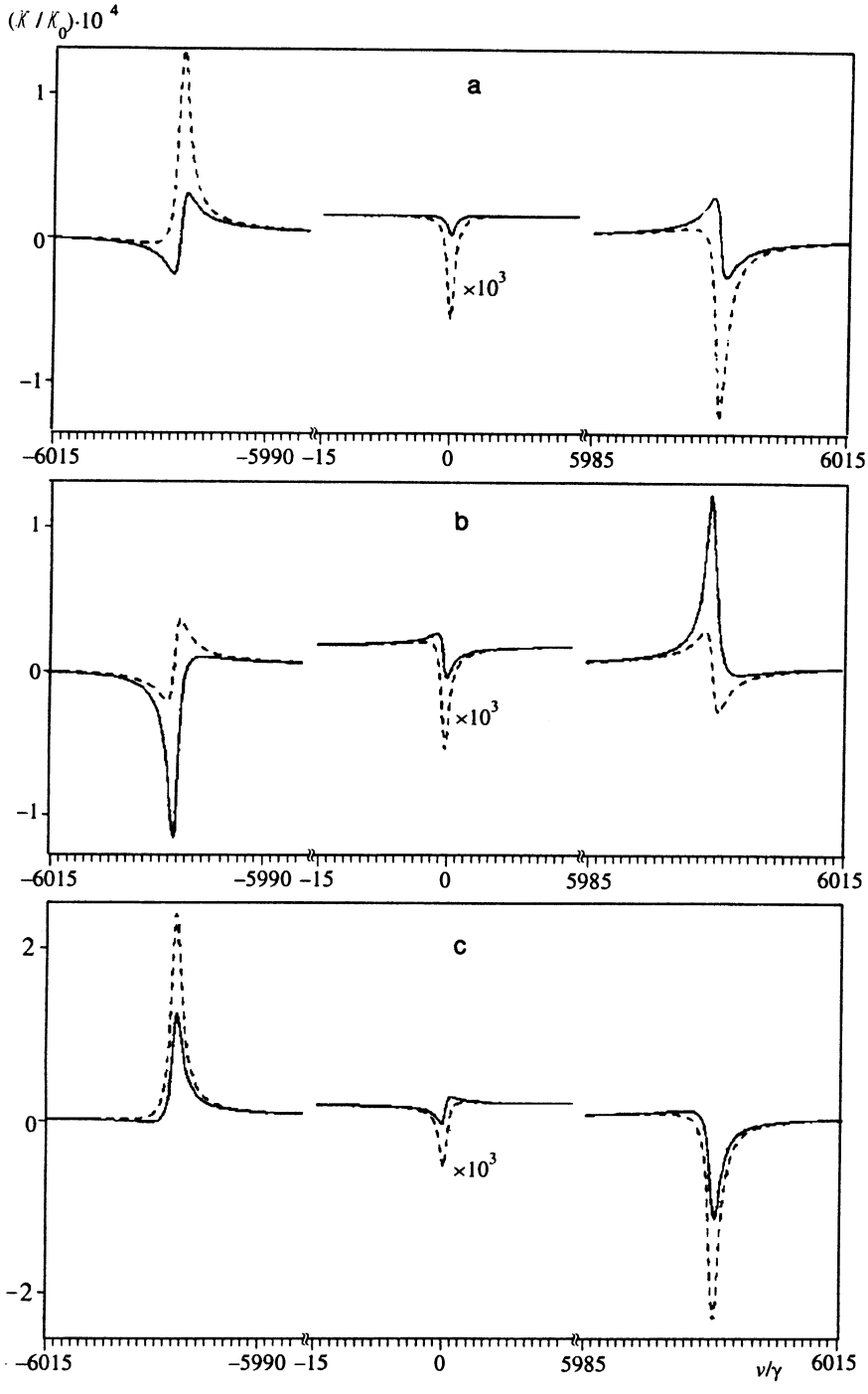

FIG. 1. Spectral dependence of the absorption coefficient (or gain) of the test signal for $\omega_0/\gamma=5\times10^7$, $\Lambda/\pi=2$, $V/\gamma=3\times10^3$: a) $\Delta_L/\gamma=0$; b) $\Delta_L/\gamma=2$; c) $\Delta_L/\gamma=-2$. Solid lines—results of calculations performed according to the standard theory;[6] dashed lines—results of the present work using expressions (27), (22), and (23). The results have been magnified by a factor of 10^3 for $|\nu/\gamma|\leq 15$.

cient achieves saturation and coincides with the result following from the standard equations for the density matrix.

The influence of the effects of nonlinear dynamic relaxation on the emission and absorption (or amplification) spectra of a two-level atom in a strong electromagnetic field has been investigated. A comparison of the emission spectrum of a two-level atom that we obtained with Mollow's resonance fluorescence spectrum[5] has revealed that the expressions for the inelastic components are nearly identical and that there are significant differences in the behavior of the elastic components. The influence of the effects of nonlinear dynamic relaxation is manifested most strongly in the absorption (or amplification) spectra of a test signal. It has been shown that consideration of these effects significantly alters both the absolute values of the absorption coefficient (or gain) of the test signal and its spectral dependence.

The effects of nonlinear dynamic relaxation may be displayed experimentally in the optical region at fairly moderate intensities. If we set $\gamma=6\times10^7$ rad/s and $\omega_0=3\times10^{15}$ rad/s, the condition $\Omega\sim\sqrt{\omega_0\gamma}$ holds for intensities of the order of 0.5–0.6 MW/cm^2. While this value is considerable for continuous lasers, it is easily achieved under pulsed conditions. In that case the pulse length must be greater than the lifetime

of the excited state for reliable detection of the predicted effects.

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