Self-oscillation and multistability due to the nonresonant dynamic Stark effect in a resonator with a medium consisting of two-level particles

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It is known that upon intraresonator optical excitation of a quantum-particle medium the dynamic Stark shifts of the resonance levels lead to novel effects under conditions of a relatively short single-mode resonator, viz., to optical multistability and self-oscillations in the "particle + field" system. The effects are due to the nonmonotonic dependence of the real and imaginary parts of the resonant polarizability of the medium on the intensity amplitude of the intraresonator field. They arise in single-resonance excitation of the two-level particles, in contrast to the cases considered by the author with V. A. Kovarskiĭ and I. Sh. Averbukh [JETP Lett. 32, 255 (1980); Sov. Phys. JETP 53, 39 (1981)], in which similar effects appeared in double optical resonance excitation of three-level quantum systems.

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The study of cooperative processes in optical resonators filled with a medium that is nonlinearly polarized, the investigation of the ensuing optical bistabilities, hysteresis, and other phenomena, as well as various aspects of practical applications of the latter, have been the subject of a considerable number of studies during the last decade (see, e.g., the reviews 1 and 2). The most investigated, both theoretically and experimentally, is the situation when there is placed in the resonator a system of quantum particles (a gas of atoms of molecules, a system of impurity atoms or of other elementary excitations in solids), one of the excitation frequencies of which is close to the frequency of one of the longitudinal modes of the resonator, as well as to the frequency of the exciting laser radiation that enters the resonator through a slightly transparent mirror. Such resonance conditions made it possible, when account was taken of the nonlinear polarizability of the medium, to confine oneself to a two-level approximation (model of two-level absorbers[3]). The real and imaginary parts of the polarizability of the medium, calculated in the two-level approximation, are nonlinear monotonic functions of the intensity of the laser radiation, and it is this which ensured the appearance of bistability and hysteresis when such media were placed in not too long single-mode resonators excited by external laser radiation.[1-4]

We have shown earlier[5,6] that if the real and imaginary parts of the polarizability of the medium are nonmonotonic functions of the laser intensity, then threshold phenomena appear that are new for the described conditions (short single-mode resonators). These phenomena can be both stationary (multistability) or nonstationary (self-oscillations in the "particles in resonator + field" system, self-modulation of the light emerging from the resonator). In Refs. 5 and 6 we considered a more complicated system, in which the medium, consisting of three-level quantum particles, was excited by two laser fields whose frequencies were close to the frequencies of adjacent quantum transitions. The already mentioned nonmonotonic dependence of the polarizability of the medium was due in this case to the Autler-Townes optical effect[7] (linear high-frequency Stark effect in double optical resonance). In the present communication we call attention to the fact that the self-oscillations and multistability can be attained also in the simpler case of excitation of two-level particles by one laser beam (at a single resonance). For this purpose it is necessary only to take into account the Stark shift of the resonance levels, which are quadratic in the amplitude of the field intensity of the exciting laser and are connected with the dynamic polarizability. If the calculation of the polarizability of the medium is carried out in a model of only two dipole-coupled levels, such a shift (called in this case the Bloch-Siegert shift[7]) is due to the nonresonant part of the field mixing of the levels. It is small in terms of the parameter $|d_{12}F/(\varepsilon_2-\varepsilon_1)|\ll 1$ (ε_i is the energy of the i-th level, F is the intensity of the laser field, d_{12} is the matrix element of the dipole mixing of levels 1 and 2), and can be disregarded in practice. However, in the case of real (multilevel) quantum systems, nonresonant shifts of two levels singled out by the single-resonance condition can become appreciable (compared with the perturbation due to resonant mixing of these levels). Contributing to this can be the smallness of $|d_{12}|$ (weakly allowed resonant transition). Indeed, the nonresonant shift

$$\delta\varepsilon_i=\sum_{k\neq 1,2}|d_{ik}F|^2\left(\frac{1}{\varepsilon_i-\varepsilon_k-\hbar\omega}+\frac{1}{\varepsilon_i-\varepsilon_k+\hbar\omega}\right)\quad(i=1,2)\tag{1}$$

can be comparable with the value of the resonant perturbation $|d_{12}F|$, inasmuch as the sum over k always contains allowed transitions (with large $|d_{ik}|$). In addition, there can take place the additional quasiresonant conditions $|\varepsilon_k-\varepsilon_i|\sim\hbar\omega$ [but $|\varepsilon_k-\varepsilon_i-\hbar\omega|>\Gamma_i,\ \Gamma_k$, where $\Gamma_{i(k)}$ is the width of the i-th (k-th) level, so that it is possible to retain the quadratic approximation (1) and disregard the population of the k-th level by the laser radiation].

Taking into account the shifts $\delta\varepsilon_i$, the polarizability of the particles $\chi=\chi'+i\chi''$ in a single resonance is of the form[8]

$$\chi(\mathcal{E})=\frac{|d_{12}|^2}{\hbar}\frac{\omega_0-\omega-\alpha\mathcal{E}^2+i\gamma}{(\omega_0-\omega-\alpha\mathcal{E}^2)^2+\gamma^2+4|d_{12}|^2\mathcal{E}^2\gamma\hbar^{-2}\Gamma^{-1}}. \tag{2}$$

Here $\omega_0=(\varepsilon_2-\varepsilon_1)/\hbar$ is the particle excitation frequency, ω is the laser frequency, $\mathcal{E}=|F|$; γ^{-1} and Γ^{-1} are the transverse and longitudinal relaxation times (for simplicity we confine ourselves to the case of homogeneous broadening), and α is the difference between the nonresonant dynamic polarizabilities of the particle in the states 1 and 2. We place the system of particles in a ring resonator of length $\mathcal{L}$ with a mirror transmission coefficient T. We assume that the lifetime of the photon in the resonator $\mathcal{L}/cT$ (c is the speed of light) exceeds considerably the characteristic times of the longitudinal processes ($\mathcal{L}/cT\gg\gamma^{-1},\Gamma^{-1}$), and that the change of the internal field F as a result of a single pass through the resonator is small, so that the mean-field approximation can be used.[3,4,9] Under these assumptions, by adiabatically eliminating the variables that describe the medium from the system of material equations and Maxwell's equations, we can obtain (for details see Ref. 6)

$$\frac{d\mathcal{E}}{d\tau}=-\mathcal{E}\left[1+\frac{2\pi NKL}{T}\chi''(\mathcal{E})\right]+\frac{F_i}{T^{1/2}}\cos\psi, \tag{3}$$

$$\mathcal{E}\frac{d\psi}{d\tau}=\mathcal{E}\left[\Phi+\frac{2\pi NKL}{T}\chi'(\mathcal{E})\right]-\frac{F_i}{T^{1/2}}\sin\psi,\quad \tau=t\left(\frac{cT}{\mathcal{L}}\right). \tag{4}$$

Here $F=\mathcal{E}e^{i\psi}$, $K-\omega/c$, N is the particle density, F_i is the amplitude of the field intensity of the external laser radiation, L is the length of the working arm of the resonator filled with the nonlinearly polarized medium, $\Phi=\Theta/T$, Θ is the measure of the detuning of the frequency ω relative to the natural frequency of the resonator mode ($K\mathcal{L}=\Theta+2\pi m$, where m is an integer and $\Theta\ll 1$). With the aid of (2) and (3), (4) we obtain an equation for the stationary values of the field in the resonator:

$$y_i^2=x^2\left\{\left[1+\frac{C}{(\Delta-\beta x^2)^2+1+x^2}\right]^2+\left[\Phi+\frac{C(\Delta-\beta x^2)}{(\Delta-\beta x^2)^2+1+x^2}\right]^2\right\}, \tag{5}$$

$$x=\frac{\mathcal{E}}{\mathcal{E}_s},\quad \mathcal{E}_s=\frac{\hbar(\Gamma\gamma)^{1/2}}{2|d_{12}|},\quad C=\frac{2\pi NKL|d_{12}|^2}{\hbar\gamma T},\quad \beta=\frac{\alpha\hbar^2\Gamma}{4|d_{12}|^2}$$

$$y_i=\frac{F_i}{\mathcal{E}_s T^{1/2}},\quad \Delta=\frac{\omega_0-\omega}{\gamma}.$$

Obviously, Eq. (5) can have more than one solution. The stability of the stationary solutions of Eqs. (3) and (4), obtained from (5), is determined by the standard linear analysis using the Routh-Hurwitz criterion.[10] This analysis yields the following stability conditions:

$$\partial Q(x)/\partial x>0,\quad \partial P(x)/\partial x>0. \tag{6}$$

Here $Q(x)$ is the right-hand side of Eq. (5), and

$$P(x)=x^2\left[1+\frac{C}{(\Delta-\beta x^2)^2+1+x^2}\right]. \tag{7}$$

In its physical meaning, $P(x)$ is the power dissipated by the internal field on account of the penetrability of the resonator mirrors and the absorption in the medium. Owing to the presence of the dynamic Stark shift ($\sim\beta x^2$), the quantity $P(x)$ is generally speaking a nonmonotonic function of the intensity ($\sim x^2$) of the external field (it contains decreasing sections). On these decreasing sections, the second condition of (6) is violated, and this can lead to instability of the solutions of Eq. (5), including those solutions which correspond to sections of increasing function $Q(x)$. We note that $\beta=0$ and $P(x)$ is a monotonic function of x for the ordinary two-level absorbers previously investigated in the literature in connection with the problem of optical bistability.[1-4] This in turn leads to stability of the solutions (5) corresponding to growing sections of the function $Q(x)$. An example of the situation described above is shown in Fig. 1. As follows from this figure, an entire band (shown shaded in Fig. 1) of values of the amplitude of the external field y_i is produced in which the solutions of (5) [in this example $Q(x)$ is a monotonic function and the solution is unique] are unstable. The presence of a unique and unstable solution of the system of nonlinear equations (3) and (4) leads us to expect the appearance of a limit cycle on the phase plane of the system. Such a limit cycle is indeed observed (Fig. 2, thick line, with the thin line showing an example of a phase trajectory that reaches the limit cycle in the course of time). The presence of the limit cycle means the onset of self-oscillations in the "particles in resonator + field" system, and accordingly amplitude-phase self-modulation of the field emerging from the resonator F_t ($F_t=T^{1/2}F$). As follows from Fig. 2, the modulation depth in the chosen numerical example is appreciable.

Let us dwell in greater detail on the analysis of the stable stationary solutions of system of equations (3) and (4). In the usual dispersion bistability based on resonant polarization of two-level particles,[4] $\beta=0$ and $\chi'(\mathcal{E})$ is a monotonic function of $\mathcal{E}$. Therefore at $\Phi\neq 0$ and if $\mathcal{E}$ varies continuously the natural frequency of the single-mode resonator filled with the nonlinear medium can be tuned only once to resonance with the frequency of the external field. This can lead to the onset of one decreasing section (inflection) of the plot of $Q(x)$ against x, which in turn yields three solutions of Eq. (5) of which two [corresponding to the sections with positive slope of $Q(x)$] are stable (bistability). If account is taken of the dynamic Stark shift of the levels ($\beta\neq 0$), then $\chi'(\mathcal{E})$ is a nonmonotonic function of $\mathcal{E}$. Therefore when $\mathcal{E}$ varies continuously the frequency of

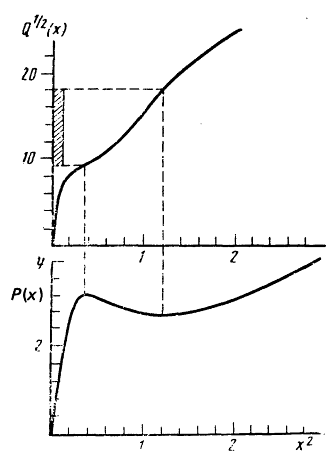

FIG. 1. Onset of the instability zone of stationary states of the system ($C=20$, $\beta^2=10$, $\Phi=20$, $\Delta=0$).

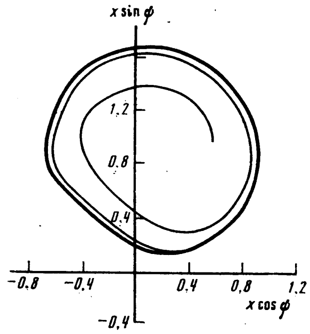

FIG. 2. Limit cycle on the phase plane of the system (C = 20, β^2 = 10, Φ = 20, Δ = 0, y_i = 15).

the resonator becomes tuned more than once to resonance with the frequency of the external field, as a result of which $Q(x)$ acquires additional inflection sections. Equation (5) then has more than three solutions, of which more than two are stable (multistability!). It is easy to see that at $\Delta/\beta \gg 1$ the inequality $|\chi(\mathscr{E})| \ll \chi'(\mathscr{E})$ is satisfied and the multistability has a pure dispersion character. The outward physical manifestations of this multistability are similar to those investigated by us earlier in Refs. 5 and 6.

In conclusion, we make a few estimates. Let there be satisfied a quasi-resonance condition between level 2 and some quantum-system level k connected with level 2 by a dipole-allowed transition ($\hbar\Gamma \ll |\varepsilon_k - \varepsilon_2 - \hbar\omega| \ll \hbar\omega$). Then the polarizability can be approximated by the expression $\alpha \sim |d_{2k}|^2/\hbar|\varepsilon_k - \varepsilon_2 - \hbar\omega$. In this case

$$\beta \sim \frac{\hbar\Gamma}{|\varepsilon_k-\varepsilon_2-\hbar\omega|}\left|\frac{d_{2k}}{d_{12}}\right|^2.$$

The quantity β reaches the values $\beta \gtrsim 1$ needed for the effects described above to manifest themselves, for example, when

$$|d_{2k}/d_{12}| \sim 10, \quad \hbar\Gamma/|\varepsilon_k-\varepsilon_2-\hbar\omega| \gtrsim 10^{-2}.$$

If we chose $\gamma \sim 10^8-10^9$ sec^{-1}, $|d_{12}| \sim 10^{-1}D$, $T \sim 10^{-2}$, and $KL \sim 10^3$, values $C \gtrsim 1$ are reached at particle densities $N \gtrsim 10^{13}-10^{14}$ cm^{-3}. Choosing $\Gamma \sim \gamma$ we find that $y_i \gtrsim 1$ are reached at $F_i \gtrsim 10-10^2$ V/cm. Inasmuch as in the foregoing estimates we chose typical values of the parameters, the effects described above are expected to be observable under nonextremal conditions.

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