Stark instability and cooperative threshold phenomena in double optical resonance

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(Submitted 29 May 1980)
Zh. Eksp. Teor. Fiz. **80**, 80–95 (January 1981)

It is shown that an instability resulting in a number of cooperative threshold optical phenomena appears in a system of atoms or molecules in a resonator under double optical resonance conditions. This instability is due to the high-frequency Stark effect caused by the collective field. When the parameters of the exciting resonance fields exceed a certain critical value the stationary state of the system becomes multivalued. Transitions between the various stable states occur by jumps representing a first-order kinetic phase transition. The dependences of the optical characteristics of the system on the intensities of the exciting field are hysteretic, indicating that the system exhibits an optical memory. Under certain conditions the "atoms + field" system exhibits spontaneous oscillations resulting in self-modulation of the radiation emerging from the resonator when the intensities of the exciting fields are fixed. An analysis is also made of the case when a combination process results in amplification of one of the resonance fields by the medium. It is found that there can be a hard oscillation regime in which a field of finite amplitude appears abruptly in a resonator.

PACS numbers: 32.60. + i, 33.55. + c, 33.40.Hp

Recent years have seen an upsurge of interest in cooperative optical phenomena in light-excited quantum systems which have a distinguishing characteristic that multiple stable states appear when the parameters of a given system exceed certain critical values. These phenomena include optical bistability in resonators with saturable absorbers,[1-4] dispersive optical bistability,[5,6] effects occurring in lasers with a nonlinear absorber inside the resonator,[7-10] vibrational bistability in optically excited molecular gases,[11-13] carrier-density bistability in optically excited semiconductors,[14] etc. This range of phenomena is very interesting from the practical point of view—because they can be used in optical memory cells, optical amplifiers, limiters, etc.[15-19]—and also from the theoretical point of view—because all these phenomena are examples of "dissipative structures" that appear in open systems far from a state of thermodynamic equilibrium.[20-24] It is easy to show that the common cause of all these effects is the existence of feedback in a system resulting in various instabilities. In each of the examples given above[1-14] the ability of a system to absorb the energy of an external agency depends on the degree of its nonequilibrium which itself is governed by the absorbed power. For example, in the absorptive optical bistability[1-4] this feedback is due to a saturation effect which reduces the absorption in a resonator as the field increases.

We shall draw attention to the fact that under double optical resonance conditions the high-frequency Stark effect gives rise to a specific feedback mechanism that produces several new cooperative threshold optical phenomena.

FORMULATION OF THE PROBLEM AND QUALITATIVE ANALYSIS

We shall consider a system of three-level atoms or molecules (to be specific, we shall speak of atoms) placed in a resonator tuned to one of the atomic transitions subjected to optical pumping E_p corresponding to another transition. An additional electromagnetic field E_i of frequency Ω close to the resonator mode is injected into the resonator. We shall consider the case of such pumping intensities which do not yet saturate the transition in question. The internal field E of frequency Ω can then be considerably stronger than E_i when the Q factor of the resonator is high. The resonator field is governed also by the induced polarization of the medium. The internal field shifts the atomic levels as a result of the high-frequency Stark effect. This shift then alters the degree of absorption of the pump field E_p which governs the induced polarization. This provides feedback which can result in an instability of the system. A specific manifestation of this feedback depends strongly on the relationship between the widths of the pump wave and of the atomic levels, and on the detuning of the central pump frequency from the relevant transition under double optical resonance conditions. Two typical situations are shown in Fig. 1.

Figure 1a corresponds to the case of a narrow-band quasimonochromatic pump wave whose central frequency is $\Omega_p = \omega_{21} + \Delta$ (ω_{ij} are the frequencies of the atomic transitions) and whose spectral width is much less than the widths of the two excited atomic levels 2 and 3. It is well known that in the presence of strong

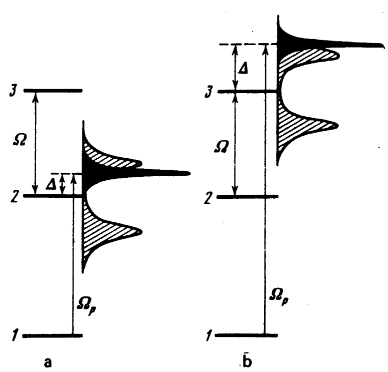

FIG. 1. Double optical resonance in a three-level quantum system. The intermediate level (a) or the upper level (b) is pumped. The components of the Stark absorption band doublet are shaded. The spectral profile of the pump is shown black.

resonance radiation corresponding to the 2–3 transition the light-absorption band Ω_p splits into two components whose widths are of the order of the widths of the levels 2 and 3 and which are separated by an interval that increases on increase in the field E. Since the levels 2 and 3 are not populated in the absence of pumping, the polarization of the medium at the frequency Ω is not possible only as a result of a combination process involving absorption of the pump field. We can thus see that the refractive index of the medium and of the absorption coefficient at this frequency are governed by the pump absorption. For simplicity, we shall assume that the empty resonator and the external field E_i are tuned exactly to the atomic transition frequency ($\Omega = \omega_{32}$). Then, the pump-induced change in the refractive of the medium alters the optical length of the resonator and detunes the resonator from the frequency ω_{32}. This effect and the appearance of the absorption at the frequency Ω reduce the internal field E in the resonator for a constant value of E_i. We shall show how an instability appears in this situation. Let the field E be such that $\Omega_R > \Delta$ ($\Omega_R = |d_{23}E|\hbar$, where d_{ij} is the dipole matrix element). Then, a slight increase in E increases even more the separation between the components of the pump absorption band, reduces this absorption, and consequently causes a further rise of the field E. An instability appears and it then becomes stabilized because of the finite value of the transmission coefficient of the mirrors. The instability appears when the field E_i exceeds a critical value corresponding to the condition $\Omega_R \approx \Delta$. In weaker fields E_i such that $\Omega_R < \Delta$ the state of the system is stable. An allowance for the field losses in the resonator mirrors gives rise to a threshold of the appearance of the instability not only in respect of the value of the field E_i but also in respect of the intensity of the pump field. The appearance of the instability when the external agencies exceed the threshold values is typical of "trigger" systems far from equilibrium undergoing a first-order kinetic phase transition.[20,21] The resultant set of stable states of the system will be called, stressing its origin, the Stark multistability.

A qualitatively different situation (Fig. 1b) can appear as a result of pumping via the 1–3 transition (the resonator is still tuned to the frequency ω_{32}). In this case we can expect amplification of the field E as a result of absorption of the pump wave if certain relationships are satisfied between the relaxation times of the levels 2 and 3. This amplification increases on increase of the pump absorption. We can easily see that in this case again an instability appears and it is associated with a characteristic self-tuning of the system to a resonance with the pump field. Let us assume that a narrow-band pump wave is detuned somewhat from the 1–3 transition, as shown in Fig. 1b. The process of pump absorption increases the internal field and this results in an increase in the interval of the Stark doublet components. Then, the detuning between the pump frequency Ω_p and, for example, the upper Stark component (Fig. 1b) decreases and this results in a resonant increase of the pump absorption and in a further rise of the internal field. Such an instability becomes stabilized when the system is completely resonance-tuned ($\Omega_R \approx \Delta$). It is interesting to note that a similar effect can occur even in the absence of external radiation of frequency $\sim\omega_{32}$ when the internal field appears only as a result of absorption of the pump wave. In contrast to the usual lasing, representing a second-order kinetic phase transition,[22-24] the effect in question can be regarded—as shown below—as a new example of a first-order kinetic phase transition in an optical amplifying medium.

PRINCIPAL DYNAMICS EQUATIONS OF A SYSTEM AND CONDITIONS OF STABILITY OF STATIONARY STATES

Complications associated with the appearance of standing waves in a resonator with parallel mirrors will be avoided by considering a ring resonator, as shown in Fig. 2. Mirrors 3 and 4 will be regarded as perfectly reflecting, whereas mirrors 1 and 2 will be considered to have finite transmission T and reflection R coefficients ($R+T=1$). For simplicity, the atomic levels will be regarded as homogeneously broadened and the thermal motion of atoms will be ignored. The equations for the density matrix of the medium σ_{ij} then have the following form in the resonance approximation ($\Omega_p \sim \omega_{21}$, $\Omega \sim \omega_{32}$)

$$\dot\sigma_{11}=w_{21}\sigma_{22}+w_{31}\sigma_{33}+i(A_{12}\lambda_{21}-\lambda_{12}A_{21}), \tag{1}$$

$$\dot\sigma_{22}=-w_{21}\sigma_{22}+w_{32}\sigma_{33}+i(A_{21}\lambda_{12}-\lambda_{21}A_{12})+i(A_{23}\lambda_{32}-\lambda_{23}A_{32}), \tag{2}$$

$$\dot\sigma_{33}=-(w_{31}+w_{32})\sigma_{33}+i(A_{32}\lambda_{23}-\lambda_{32}A_{23}), \tag{3}$$

$$\dot\lambda_{31}=(i\Delta_{31}-\gamma_{31})\lambda_{31}+i(A_{32}\lambda_{21}-\lambda_{32}A_{21}), \tag{4}$$

$$\dot\lambda_{21}=(i\Delta_{21}-\gamma_{21})\lambda_{21}+iA_{21}(\sigma_{11}-\sigma_{22})+iA_{23}\lambda_{31}, \tag{5}$$

$$\dot\lambda_{32}=(i\Delta_{32}-\gamma_{32})\lambda_{32}+iA_{32}(\sigma_{22}-\sigma_{33})-iA_{12}\lambda_{31}, \tag{6}$$

where

$$\sigma_{11}+\sigma_{22}+\sigma_{33}=1,$$

$$\lambda_{ij}=\lambda_{ji}^* \quad (i\neq j), \quad A_{ij}=A_{ji}^*,$$

$$\lambda_{21}=\sigma_{21}\exp(i\Omega_p t), \quad \lambda_{32}=\sigma_{32}\exp(i\Omega t),$$

$$\lambda_{31}=\sigma_{31}\exp[i(\Omega_p+\Omega)t].$$

Here, $\Delta_{21}=\Omega_p-\omega_{21}$, $\Delta_{32}=\Omega-\omega_{32}$, $\Delta_{31}=\Delta_{21}+\Delta_{32}=\Omega_p+\Omega-\omega_{31}$, $A_{12}=d_{12}E_p/\hbar$, and $A_{23}=d_{23}E/\hbar$. The quantities w_{21}, w_{31}, w_{32} and γ_{21}, γ_{31}, γ_{32} are the reciprocal longitudinal and transverse relaxation times of the relevant transitions. The system of equations (1)–(6) is written down on the assumption that the temperature of the system under investigation is insufficient for the thermal excitation of the atomic levels.

The amplitude of the internal field $E(z,t)$ satisfies the reduced Maxwell equation

$$\frac{\partial E}{\partial z}+\frac{1}{c}\frac{\partial E}{\partial t}=2\pi i k N \mathscr{P}(z,t). \tag{7}$$

Here, $k=\Omega/c$, N is the density of atoms, and $\mathscr{P}(z,t)$ is

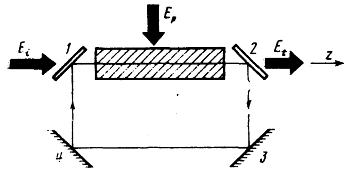

FIG. 2. Sample (shown shaded) placed in a ring resonator.

the amplitude of a wave of the polarization of the medium of frequency Ω:

$$\mathscr{P}(z,t)=\lambda_{32}(z,t)d_{23}. \tag{8}$$

We shall ignore attenuation of the pump field in the medium. Equations (1)–(8) form a closed system which can be solved allowing for the boundary condition on the mirror 1:

$$E(0,t)=T^{1/2}E_i+RE\left(L,t-\frac{\mathscr{L}-L}{c}\right)\exp(ik\mathscr{L}), \tag{9}$$

where $\mathscr{L}$ is the total optical path in the empty resonator and L is the length of the sample containing the investigated substance (Fig. 2). In the stationary case, we have

$$\mathscr{P}(z,t)=[\chi'(E)+i\chi''(E)]E,\ E=E(z), \tag{10}$$

where $\chi=\chi'+i\chi''$ is the polarizability of the atoms at the frequency Ω.

We shall assume that the medium is not too dense and the resonator not too long so that the change in the amplitude of the light wave E as a result of a single trip through the resonator is small. Therefore, the medium has a significant influence on the field only if $T\ll 1$ (i.e., when the Q factor of the resonator is high) and this will be assumed in our treatment. We shall integrate Eq. (7) with respect to the coordinate z from $z=0$ to $z=L$ in the stationary case and we shall use the smallness of the change in $E(z)$ in taking the polarization $\mathscr{P}$ outside the integral for $E=E(0)$. We shall use the boundary condition (9) to obtain

$$E=\frac{T^{1/2}E_i}{1-R\exp(ik\mathscr{L})[1-2\pi kNL(\chi''-i\chi')]}\ . \tag{11}$$

We shall assume that the resonator is tuned almost exactly to the frequency Ω, i.e., $k\mathfrak{L}=2\pi m+\theta$ (m is an integer and $\theta\ll 1$). It is convenient to go over to equations for the modulus $\mathscr{E}$ and the argument ψ of the field E ($E=\mathscr{E}e^{i\psi}$). It readily follows from Eq. (11) that

$$Q(\mathscr{E})-E_i^2/T=0, \tag{12}$$

$$Q(\mathscr{E})=\mathscr{E}^2\left\{\left[1+\frac{2\pi NkL}{T}\chi''(\mathscr{E})\right]^2+\left[\frac{\theta}{T}+\frac{2\pi NkL}{T}\chi'(\mathscr{E})\right]^2\right\},$$
$$\operatorname{tg}\psi=\left[\frac{\theta}{T}+\frac{2\pi NkL}{T}\chi'(\mathscr{E})\right]\left[1+\frac{2\pi NkL}{T}\chi''(\mathscr{E})\right]^{-1}. \tag{13}$$

Since the quantities $\chi'(\mathscr{E})$ and $\chi''(\mathscr{E})$ are complex nonlinear functions of $\mathscr{E}$, Eq. (12) can have more than one solution and this means that there can be many stationary states. However, not every solution of Eq. (12) corresponds to a stable state of the system. In investigating the stability of stationary states it is necessary to return to the secular equations (1)–(8), linearize them near each of the solutions, and apply the standard Hurwitz–Routh criterion.[25] However, in order to obtain clear results, we shall investigate only the case of a high-Q resonator when the atomic relaxation time is much shorter than the characteristic time $\sim\mathscr{L}/cT$ of a change in the field in the resonator. Then, the polarization of the medium follows adiabatically the field E in accordance with Eq. (10), where however E is time-dependent. Integrating again the secular equation (7) for z from $z=0$ to $z=\mathscr{L}$ and applying the condition (9), we obtain—subject to the same assumptions as in the stationary case—the equations

$$\frac{d\mathscr{E}}{d\tau}=-\mathscr{E}\left[1+\frac{2\pi NkL}{T}\chi''(\mathscr{E})\right]+\frac{E_i}{T^{1/2}}\cos\psi,$$
$$\mathscr{E}\frac{d\psi}{d\tau}=\mathscr{E}\left[\frac{\theta}{T}+\frac{2\pi NkL}{T}\chi'(\mathscr{E})\right]-\frac{E_i}{T^{1/2}}\sin\psi, \tag{14}$$
$$\tau=t(\mathscr{L}/cT)^{-1}.$$

The stationary solutions of the system (14) are naturally identical with Eqs. (12) and (13). The stability conditions of these solutions are

$$dP(\mathscr{E})/d\mathscr{E}>0,\ P(\mathscr{E})=\mathscr{E}^2[1+2\pi NkLT^{-1}\chi''(\mathscr{E})], \tag{15}$$
$$dQ(\mathscr{E})/d\mathscr{E}>0. \tag{16}$$

It should be noted that the condition (15) has a simple physical meaning: a state is stable if near this state the power dissipated by the internal field increases on increase in the field. The condition (16) identifies regions of a positive slope of the function $Q(\mathscr{E})$ which occurs in the "equation of state" (12). A further analysis of Eqs. (12) and (13) cannot be made without the knowledge of the explicit form of the polarizability $\chi(\mathscr{E})$.

CRITICAL EFFECTS AND STARK MULTISTABILITY IN THE CASE OF PUMPING TO AN INTERMEDIATE LEVEL

For simplicity, we shall assume that $\gamma_{21}=\gamma_{31}=\gamma$ and $\Omega=\omega_{32}$ ($\Delta_{21}=\Delta_{31}=\Delta$). Employing the assumption that the pump is weak, we shall find steady-state solutions of Eqs. (1)–(6) in the lowest order with respect to E_p, with exact allowance for the field E. We can easily demonstrate that $\sigma_{11}\sim 1$; on the other hand, $\sigma_{22}\propto\sigma_{33}\propto\lambda_{32}\propto E_p^2$ and $\lambda_{31}\propto\lambda_{21}\propto E_p$. Dropping in Eq. (4) the term with λ_{32}, we shall solve simultaneously Eqs. (4) and (5). Substituting the result in Eq. (6) and employing Eqs. (1)–(3), we then find that

$$\chi(\mathscr{E})=\frac{|d_{32}|^2}{\hbar\gamma_{32}}\frac{|d_{12}|^2E_p^2}{\hbar^2}[(\Delta^2-\Omega_R^2)^2+2\gamma^2(\Delta^2+\Omega_R^2)+\gamma^4]^{-1}$$
$$\times\{-2\gamma\Delta+i(w_{31}+w_{32})[w_{21}(w_{31}+w_{32})+2\Omega_R^2(w_{31}+w_{21})/\gamma_{32}]^{-1}$$
$$\times[2\gamma(\Delta^2+\Omega_R^2+\gamma^2)+w_{21}(\Omega_R^2-\Delta^2+\gamma^2)]\},\quad \Omega_R=|d_{32}E|/\hbar. \tag{17}$$

The real and imaginary parts of Eq. (17) are generally nonmonotonic functions of $\mathscr{E}$, a consequence of the high-frequency Stark effect. This nonmonotonic behavior is manifested clearly if $\Delta\gtrsim\gamma$ and then both $\chi'(\mathscr{E})$ and $\chi''(\mathscr{E})$ have extrema at $\Omega_R\sim|\Delta|$. If $|\Delta|\ll\gamma$, we have $\chi'\approx 0$ and χ'' decreases monotonically on increase in $\mathscr{E}$. Consequently, we shall consider the cases $|\Delta|\ll\gamma$ and $|\Delta|\gg\gamma$ separately

Case $|\Delta|\ll\gamma$. We shall introduce dimensionless variables

$$x=\frac{1}{T^{1/2}}\frac{E_t}{\mathscr{E}_s}\ (E_t=T^{1/2}\mathscr{E}),\quad y_i=\frac{1}{T^{1/2}}\frac{E_i}{\mathscr{E}_s},$$
$$\mathscr{E}_s=\frac{\hbar}{|d_{32}|}\left[\frac{\gamma_{32}w_{21}}{2}\frac{w_{31}+w_{32}}{w_{31}+w_{21}}\right]^{1/2},\quad \beta=\frac{w_{21}\gamma_{32}}{2\gamma^2}\frac{w_{31}+w_{32}}{w_{31}+w_{21}}, \tag{18}$$
$$C_1=\frac{\pi NkL}{T}\left(\frac{2\gamma}{w_{21}}+1\right)\frac{|d_{32}|^2}{\hbar\gamma_{32}}\frac{|d_{12}|^2E_p^2}{\hbar^2\gamma^2},\quad \Phi=\frac{\theta}{T},$$

where $\mathscr{E}_s$ is the saturation field of the 2–3 transition and E_t is the amplitude of the field of frequency $\Omega=\omega_{32}$ emerging from the resonator via the mirror 2. It follows from Eq. (12) that

$$y_i^2=x^2\left\{\left[1+\frac{2C_1}{(1+x^2)(1+\beta x^2)}\right]^2+\Phi^2\right\}. \tag{19}$$

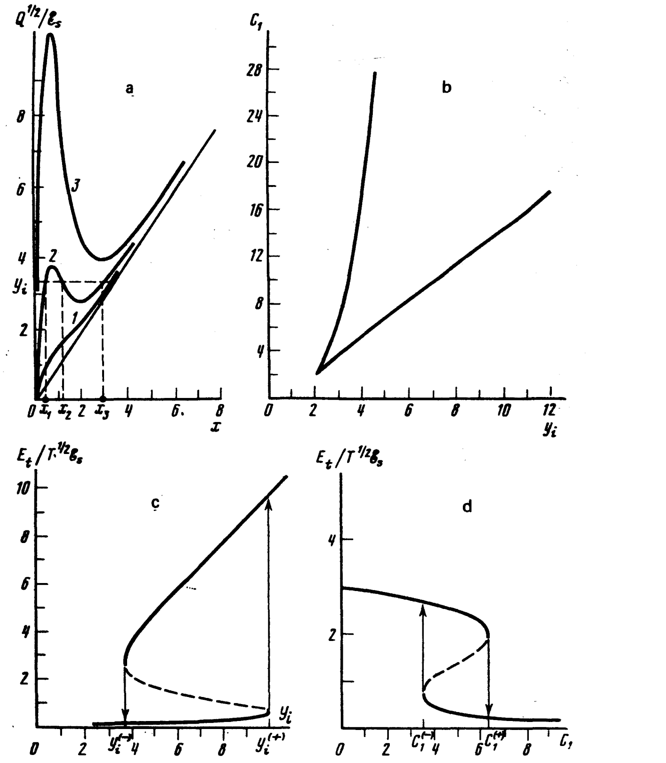

FIG. 3. Threshold phenomena in the $|\Delta| \ll \gamma$, $\theta = 0$, $\beta = 1$ case: a) graphical solution of the "equation of state" (19) for three values of the parameter $C_1 = 1, 5, 15$ (curves 1, 2, and 3, respectively); b) bifurcation diagram; c) hysteretic dependence of E_t on y_i for $C_1 = 15$; d) hysteretic dependence of E_t on C_1 for $y_i = 3$. The arrows identify kinetic phase transitions.

We shall determine first the stationary states of the system corresponding to $\theta = 0$. A graphical solution of Eq. (19) obtained for this case is shown in Fig. 3a for three values of the parameter $C_1(E_p)$. At pump intensities lower than the critical value (curve 1) this equation has one root. When the quantity E_p (parameter C_1) exceeds its critical value, this equation can have three solutions (curve 2). When E_p is sufficiently high (but E_i is fixed), Eq. (19) again has just one root (curve 3). In accordance with the stability criterion (16), the root x_2 (Fig. 3a) on the part of curve 2 with a negative slope is unstable. Since for $\chi' = 0$ and $\theta = 0$ the inequality (16) results automatically in satisfaction of the condition (15), the roots x_1 and x_3 on parts with a positive slope are stable. The bifurcation diagram of the system is shown in Fig. 3b. In the case of the parameters lying outside the wedge bounded by thick lines (bifurcation lines) Eq. (19) has one solution, whereas outside it it has three solutions, one of which is unstable. The occurrence of two stable states of the system is manifested by the double-valued hysteresis of its optical characteristics. Figures 3c and 3d show the dependences of the amplitude $E_t = T^{1/2}\mathscr{E}$ of the field transmitted by the resonator on y_i in the case when $C_1 = \text{const}$ (Fig. 3c) and when $C_1(E_p)$ for $y_i = \text{const}$ (Fig. 3d). When the values of the parameters are $y_i^{\pm}$ (Fig. 3c) or $C_1^{\pm}$ (Fig. 3d) the system undergoes an abrupt transition from one stable state to another. Near these values the optical properties of the system are nonanalytic functions of the external field intensities, i.e., these properties cannot be represented by expansions as power series of deviations from such values. Indeed, it is easy to show that, for example, close to E_i^{+} (Fig. 3c) we have

$$E_t - E_t(E_i^{(+)}-0) \sim -(E_i^{(+)}-E_i)^{1/2} \quad \text{for} \quad E_i \leq E_i^{(+)},$$

$$E_t - E_t(E_i^{(+)}+0) \sim (E_i - E_i^{+}) \quad \text{for} \quad E_i \geq E_i^{(+)},$$

where $E_t(E_i^{(+)}+0) - E_t(E_i^{(+)}-0)$ is the magnitude of the jump (discontinuity). Thus, near these values of the parameters the observed properties of the system cannot be deduced by perturbation theory from the magnitude of the interaction with external fields.

In the specific case when $\beta \ll 1$, Eq. (19) is formally identical with the equation of state describing optical bistability in a resonator with a saturable absorber.[1-4] However, the parameter C_1 describing the behavior of the system is then proportional to the pump intensity and is controlled by this intensity, whereas in the case described in Refs. 1–4 it is governed solely by the density of atoms. If $\beta \ll 1$, the threshold of the disappearance of such a bistability is naturally the same as in Refs. 1–4, i.e., $C_1^{\text{cr}} = 4$. If $\beta \gtrsim 1$ and $\theta = 0$ the behavior is basically still the same but the magnitude of the threshold decreases. For example, if $\beta = 1$, we find that $C_1^{\text{cr}} = 2$.

A basically new situation may arise when $\theta \neq 0$. An increase in $|\theta|$ at a fixed value of C_1 results in a gradual disappearance of the characteristic kink exhibited by the $Q(\mathscr{E})$ curve and in narrowing of the region where the inequality (16) is disobeyed. Since the inequality (15) is independent of θ, it follows that when θ exceeds a certain value the regions with a positive slope of the curve $Q(\mathscr{E})$ may become unstable since they lie in the region where the criterion (15) is violated (Figs. 4a and 4b). A bifurcation diagram corresponding to this situation is shown in Fig. 4c. As before, inside the wedge-shaped regions there are three stationary solutions and outside it there is only one solution. The thick line is also a boundary of the appearance of saddle-type stationary states. Within the region bounded by the thin line the system has unstable nodes or foci. Thus, for example, in the shaded region of the parameters there is a single unstable stationary state. All the phase trajectories (paths) of the system of nonlinear equations (14) beginning from the unstable equilibrium state move away from this state with time. On the other hand, it follows from Eq. (14) that in the case of sufficiently high values of $\mathscr{E}$ we have $\dot{\mathscr{E}} < 0$, i.e., the phase trajectories cannot escape to infinity either (this is physically evident from the dissipative nature of the system). As is known,[26] in a situation of this kind we can expect limit cycles in the phase plane. Figure 4d shows the results of a numerical integration of the system (14) for parameters lying within the shaded region of the bifurcation diagram in Fig. 4c. A limit cycle (represented by a closed thick line in Fig. 4d) can be observed and it surrounds a state of unstable equilibrium identified by a point. All the phase trajectories beginning inside and outside the cycle approach it asymptotically with time. The existence of a limit cycle results in amplitude-phase self-modulation of light emerging from the reso-

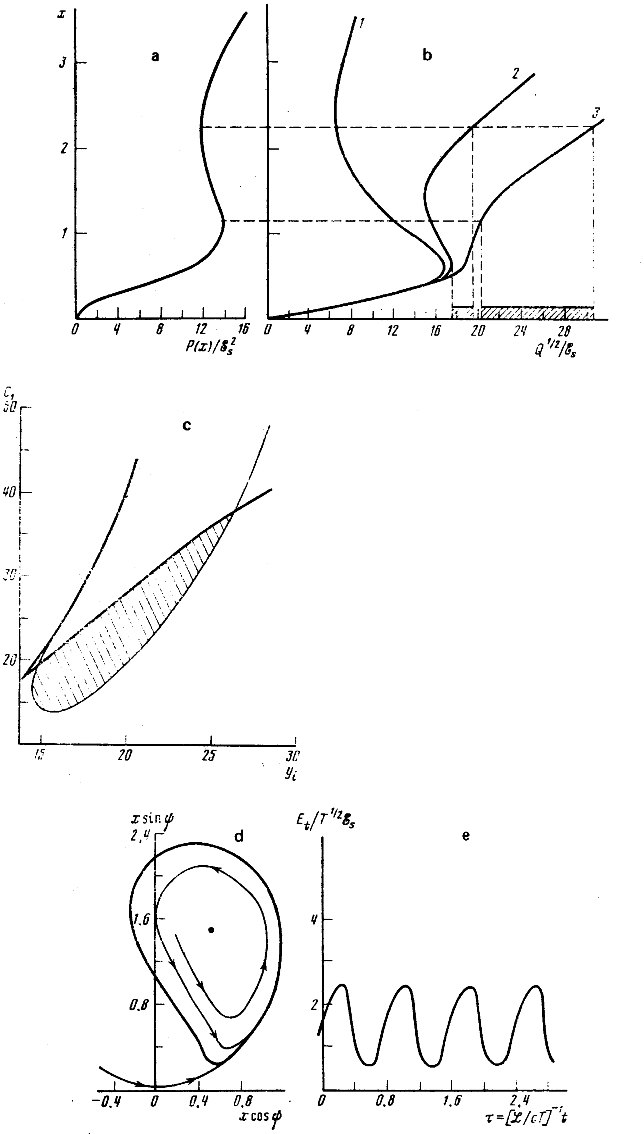

FIG. 4. Self-modulation of the field in a resonator ($|\Delta| \ll \gamma$, $\theta \neq 0$, $\beta = 1$). a), b) Appearance of an instability zone of stationary states of the system ($C_1 = 25$). The curves in Fig. 4b correspond to different values of Φ: 1) 2; 2) 9; 3) 14. The zones of instability with respect to y_i are shown shaded. c) Bifurcation diagram ($\Phi = 10$). The shaded region shows the range of existence of the only stable state. d) Limit cycle (closed thick curve) in the phase plane of the system ($C_1 = 20$, $y_i = 17$, $\Phi = 10$). The point represents an unstable equilibrium state. The thin curves are examples of phase trajectories. e) Time dependence of the amplitude of light emerging from the resonator and corresponding to the limit cycle in Fig. 4d.

nator when the intensities of the external fields E_i and E_p are constant. Figure 4e shows the time dependence of the amplitude of the field emerging from the resonator and this dependence corresponds to the limit cycle in Fig. 4d. The time ordering which appears in the system when the external agencies exceed critical values represents a time-dependent dissipative structure.[20,21]

It should be noted that the instability zones associated with the presence of a falling region in the dependence of the absorbed power on the field $\mathscr{E}$ cannot, for fundamental reasons, appear in the models of optical bistability with saturable absorbers,[1-4] because the power absorbed by such saturable substances varies monotonically.

Case $|\Delta| \gg \gamma$. To be specific, we shall assume that $\Delta > 0$. It follows from Eq. (17) that the medium becomes strongly polarized in the $\Omega_R \sim \Delta$ case. We shall also bear in mind that if $\Omega_R \sim \Delta$, then the 2–3 transition is saturated. Separating the principal terms of the polarizabilities $\chi'(\mathscr{E})$ and $\chi''(\mathscr{E})$ near their extrema, we shall write down the equation of state (12) in the form

$$y_i^2 = x^2 \left\{ \left[1 + \frac{2\tilde{C}_2}{(x-1)^2+\delta^2} \right]^2 + \left[\Phi - \frac{2C_2}{(x-1)^2+\delta^2} \right]^2 \right\}. \qquad (20)$$

Here,

$$x = \frac{1}{T^{1/2}} \frac{|d_{32}|E_t}{\hbar\Delta}, \quad y_i = \frac{1}{T^{1/2}} \frac{|d_{32}|E_i}{\hbar\Delta},$$

$$C_2 = \frac{1}{2} \frac{\pi NkL}{T} \frac{|d_{32}|^2}{\hbar\gamma_{32}} \frac{|d_{12}|^2 E_p^2}{\hbar^2} \frac{\gamma}{\Delta^3},$$

$$\tilde{C}_2 = \frac{w_{31}+w_{32}}{w_{31}+w_{21}} \frac{\gamma_{32}}{\gamma} \delta C_2, \quad \delta = \frac{\gamma}{\Delta}.$$

In the case under consideration when $\delta \ll 1$, we find that $|\chi''/\chi'| \ll 1$ ($\tilde{C}_2/C_1 \sim \delta$). Therefore, we can have a situation when $\tilde{C}_2/\delta^2 \ll 1$, but $C_2/\delta^2 \gtrsim 1$ ($\delta \gg C_2 \gtrsim \delta^2$). Then,

$$y_i^2 = x^2 \left\{ 1 + \left[\Phi - \frac{2C_2}{(x-1)^2+\delta^2} \right]^2 \right\}. \qquad (21)$$

This equation can have more than one solution because of the nonmonotonic nature of the expression in the brackets of Eq. (21) in a region of width $\sim\delta$ near $x = 1$. Interesting singularities appear for $0 < \Phi < 2C_2/\delta^2$. Then, the expression in Eq. (21) vanishes twice as a function of x so that Eq. (21) has not only three but five stationary solutions depending on E_i (Fig. 5a). However, the only stable roots are those lying on regions with a positive slope of the curves in Fig. 5a. Physically, the occurrence of three stable states is associated with the fact that on increase of the internal field because of the nonmonotonic dependence $\chi'(\mathscr{E})$ for the specified values of Φ and C_2 the condition of resonance transmission of light through the resonator (equality of the optical path to an integral number of wavelengths) is obeyed twice. In the usual dispersive bistability associated with the saturation effect[4,5] the value of χ' depends on $\mathscr{E}$ monotonically and this can result in just one tuning to a resonance.

The hysteresis of the amplitude of the light emerging from the resonator as a function of the fields E_i and E_p, which appears in this case, are illustrated in Figs. 5b and 5c. It should be noted that a similar hysteresis is observed also when the pump frequency (i.e., Δ) is varied and the external field intensities are fixed.

If $\Phi < 0$ and $\Delta > 0$, the polarization of the medium simply results in an additional detuning of the system from resonance and in this case the number of stable states does not exceed two (Fig. 5a). The threshold of appearance of the Stark multistability cannot be found in an analytic form in the case of an arbitrary relationship between the quantities Φ and δ. Using the smallness of the parameter δ, we can obtain explicit expressions for the threshold in the following two limiting

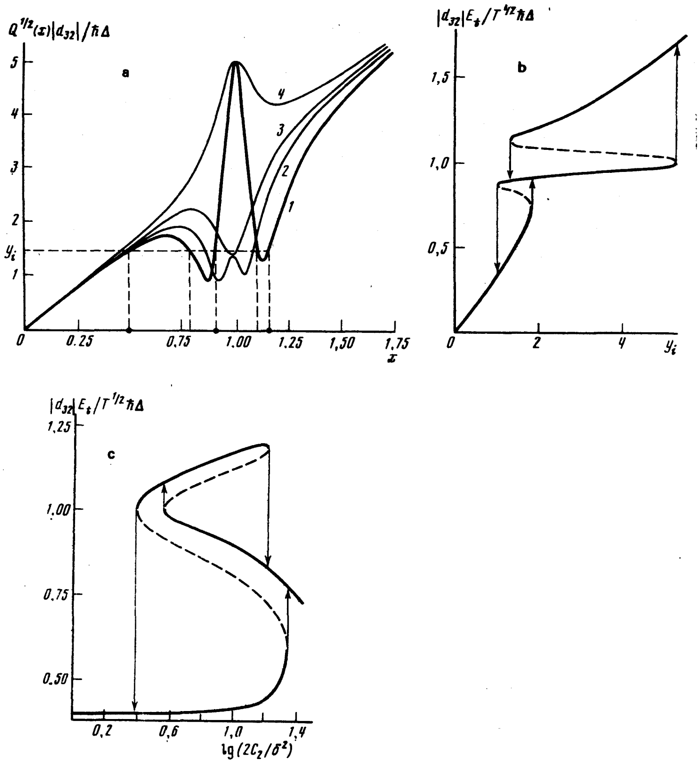

FIG. 5. Threshold phenomena in the $|\Delta| \gg \gamma$ ($\delta = 0.1$) case. a) Graphical solution of the "equation of state" (21) obtained for different values of the parameters: 1) $2C_2/\delta^2 = 8$, $\Phi = 3$; 2) $2C_2/\delta^2 = 4$, $\Phi = 3$; 3) $2C_2/\delta^2 = 2$, $\Phi = 3$; 4) $2C_2/\delta^2 = 2$, $\Phi = -3$. The points on the x axis identify the stable solutions. b) Hysteretic dependence of E_t on y_t for $2C_2/\delta^2 = 8$, $\Phi = 3$. c) Hysteretic dependence of E_t on C_2 for $y_t = 1.2$, $\Phi = 3$.

cases:

$$C_2^{cr} = \frac{3}{5}\left(\frac{6}{5}\sqrt{5}\right)^{1/2}\delta^{5/2}, \quad |\Phi| \ll \delta^{1/2},$$
$$C_2^{cr} = \frac{4\sqrt{3}}{9}\frac{1+\Phi^2}{|\Phi|}\delta^3, \quad |\Phi| \gg \delta^{1/2}. \qquad (22)$$

If $C_2 \gtrsim \delta$, we have to allow for the contribution of $\chi''(\mathscr{E})$ to Eq. (20) but this does not result in any qualitative changes.

CRITICAL PHENOMENA UNDER CONDITIONS OF FIELD GENERATION IN A RESONATOR

We shall consider the case shown in Fig. 1b when the upper level 3 is pumped. The polarizability of atoms at the frequency $\Omega = \omega_{32}$ then has the form

$$\chi(\mathscr{E}) = \frac{|d_{32}|^2}{\hbar\gamma_{32}}\frac{|d_{13}|^2 E_p^2}{\hbar^2}[(\Delta^2-\Omega_R^2)^2+2\gamma^2(\Delta^2+\Omega_R^2)+\gamma^4]^{-1}$$
$$\times\Big\{-2\gamma\Delta - i[2(w_{21}-w_{32})\gamma(\Delta^2+\Omega_R^2+\gamma^2)+w_{21}(w_{31}+w_{32})(\Omega_R^2-\Delta^2+\gamma^2)]$$
$$\times\Big[w_{21}(w_{31}+w_{32})+2(w_{31}+w_{21})\frac{\Omega_R^2}{\gamma_{32}}\Big]^{-1}\Big\}. \qquad (23)$$

It is clear from Eq. (23) that in this case the quantity $\chi''(\mathscr{E})$ can become negative and this amplifies the electromagnetic field of frequency Ω. As before, we shall consider separately the cases $|\Delta| \ll \gamma$ and $|\Delta| \gg \gamma$.

Case $|\Delta| \gg \gamma$ ($\Delta > 0$), $E_t = 0$. We shall assume, for simplicity, that a resonator is tuned to the frequency ω_{32}. We shall introduce dimensionless variables

$$x = \frac{1}{T^{1/2}}\frac{|d_{32}|E_t}{\hbar\Delta}, \quad \tilde{C}_3 = \frac{w_{21}-w_{32}}{w_{31}+w_{21}}\frac{\gamma_{32}}{\gamma}\delta C_3,$$
$$C_3 = \frac{1}{2}\frac{\pi NkL}{T}\frac{|d_{32}|^2}{\hbar\gamma_{32}}\frac{|d_{13}|^2 E_p^2}{\hbar^2}\frac{\gamma}{\Delta^3}. \qquad (24)$$

It then follows from Eq. (12) that if $E_t = 0$, we find that

$$xy(x) = 0, \quad y(x) = 1 - \frac{2\tilde{C}_3}{(x-1)^2+\delta^2} - \frac{2C_3}{(x+1)^2+\delta^2}, \qquad (25)$$

$$\left[\Phi - \frac{2C_3}{(x-1)^2+\delta^2} - \frac{2C_3}{(x+1)^2+\delta^2}\right]x = 0. \qquad (26)$$

Nontrivial solutions of Eq. (25) are possible only if $\tilde{C}_3 > 0$ ($w_{21} > w_{32}$), when the medium is active [$\chi''(\mathscr{E}) < 0$]. Figure 6a shows the dependence $y(x)$ obtained for various values of the parameter $\tilde{C}_3$. If $\tilde{C}_3 < \tilde{C}_3^{cr}$ ($2\tilde{C}_3^{cr}/\delta^2 \approx 1$) (curve 1), Eq. (25) has only the trivial solution $x = 0$. If $\tilde{C}_3 = \tilde{C}_3^{cr}$, an additional solution is obtained. For $\tilde{C}_3^{cr} < \tilde{C}_3 < C_*$, Eq. (25) has two nontrivial solutions (curve 2) and one solution for $\tilde{C}_3 > C_*$ ($C_* \approx 0.25$) (curve 3). The appearance of these additional roots reflects the possibliity of generation by this system of a field of frequency $\sim\omega_{32}$ governed by Eq. (26). We can easily show that only the larger of the roots of $y(x) = 0$ corresponds to stable oscillation. The trivial solution $x = 0$ is stable for $\tilde{C}_3 < C_*$ and becomes unstable for $\tilde{C}_3 \geq C_*$.

In the derivation of Eqs. (25) and (26) we have used, for the sake of simplicity, an expression for the polarizabilities $\chi'(\mathscr{E})$, $\chi''(\mathscr{E})$ at a frequency $\Omega = \omega_{32}$. This approximation is valid if the shift of the oscillation frequency is $|\omega - \omega_{32}| \ll \Omega_R \sim \Delta$. It follows from Eq. (26) that

$$\Phi \sim |\omega - \omega_{32}|\mathscr{L}/cT \sim 2C_3/\delta^2,$$

i.e., it is essential that

$$C_3 \ll \delta^2\mathscr{L}\Delta/cT. \qquad (27)$$

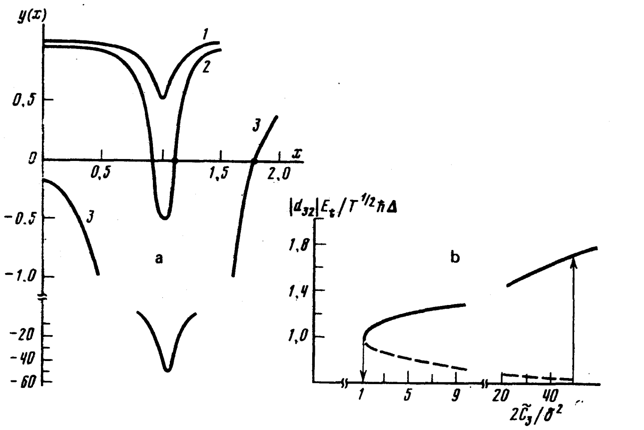

FIG. 6. Threshold phenomena in the course of pumping of the upper level in the $|\Delta| \gg \gamma$ ($\delta = 0.1$) case. a) graphical solution of the equation $y(x)=0$ [see Eq. (25)]: 1) $2\tilde{C}_3/\delta^2 = 0.5$; 2) $2C_3/\delta^2 = 1.5$; 3) $\tilde{C}_3 = 0.3$. b) Hysteretic dependence of E_t on $\tilde{C}_3$.

On the other hand, the condition $\theta = \Phi T \ll 1$ used above leads to the inequality

$$C_3 \ll \delta^2/T. \tag{28}$$

Oscillation appears if $C_3 \sim \delta$. Therefore, as long as

$$T \ll \delta \ll 1 \text{ and } T \ll \gamma\mathscr{L}/c, \tag{29}$$

we can ignore the difference of the oscillation frequency from ω_{32}.

Figure 6b shows the dependence of the field $T^{1/2}\mathscr{E}$ of frequency $\sim\omega_{32}$ emerging from the resonator on the parameter $\tilde{C}_3$ (pump intensity). It is clear from Fig. 6b that if $\tilde{C}_3 > \tilde{C}_3^{cr}$, the emitted field may appear abruptly beginning from a finite value, in contrast to the usual lasing when the field rises continuously from zero when a certain threshold is exceeded. Moreover, the absorption of the pump field changes abruptly at the moment of appearance of oscillations and the dependence of the absorbed power on the intensity of pumping also shows a hysteresis.

The conventional lasing can be interpreted as a second-order kinetic phase transition.[22-24] The hard excitation of oscillations corresponding to Fig. 6 represents a first-order kinetic phase transition. Only a few examples of such transitions in the generation of light in active media are known at present.[7-10,27]

Case $|\Delta| \ll \gamma$. We can easily show that if $E_i = 0$, only the soft oscillation regime, which does not differ qualitatively from the conventional lasing, can be observed. However, an interesting critical effect appears if $E_i \neq 0$. We shall consider only the most interesting situation when the pump intensity ensures the possibility of generating an internal field at a frequency $\sim\omega_{32}$. We shall also assume that the frequency of the external field differs little from the oscillation (lasing) frequency. If E_i is sufficiently small compared with the intensity of the generated field $\mathscr{E}_g$, the dynamics of changes in the field E will naturally represent a superposition of oscillations of two frequencies: the internal oscillation frequency and the frequency of the external field, which results in beats of the intensity of light transmitted by the resonator.

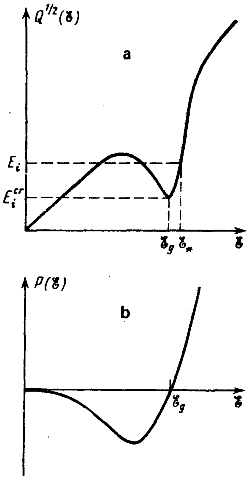

FIG. 7.

However, as is well known from the theory of nonlinear oscillations[26] when a self-oscillatory system is subjected to an external harmonic force, frequency locking occurs when the force exceeds a certain threshold value: the oscillations of the system are then synchronized with the oscillations of the external force. Let us assume that E_i exceeds this locking threshold. We can then seek a solution of an internal field of frequency Ω. The equation for the amplitude of the internal field has the form of Eq. (12) where $\chi' < 0$ and $\chi' = 0$. If $E_i > E_i^{cr}$ (Figs. 7a and 7b), Eq. (12) has one stable stationary solution $\mathscr{E}_*$ lying near $\mathscr{E}_g$ and corresponding to oscillations of the system locked (synchronized to the external field E_i. When E_i is reduced right down to E_i^{cr}, the stable solution merges with the unstable one and it disappears. If $E_i < E_i^{cr}$, the locked solution of amplitude $\sim\mathscr{E}_g$ is impossible and then oscillations with two periods appear in the system. The locking threshold E_i^{cr} corresponding to small values of Φ can be found by substituting $\mathscr{E} = \mathscr{E}_g$ in Eq. (12):

$$E_i^{cr} = \mathscr{E}_g|\theta|T^{-1/2}.$$

We have considered critical phenomena in double optical resonance from the dynamic point of view. In the presence of several stable states a system exhibiting such a resonance assumes in a determined way one of these states in accordance with the previous history of changes in the parameters of the external agency. As is well known, fluctuations causing transitions from one state to another play an important role in systems with more than one equilibrium state. Therefore, after a sufficiently long time, we can only speak of the probability of finding a system in one or another equilibrium state and the statistical approach is needed. In the case of optical bistability this problem has been considered in the case of quantum[28,29] and technical[30,31] noise. This program may be realized also for a range of phenomena described in the present paper: for example, one can employ the method of the Langevin equations.[32] However, since in the majority of the cases considered here the dispersive effects are important, the condition of detailed equilibrium is not obeyed in the relevant Fokker–Planck equation[32] and this makes it difficult to solve it even in the steady-state case. Then, the probability of fluctuation-induced transitions can be found only by asymptotic methods[33,34] that require extensive numerical calculations.

CONCLUSIONS

We have considered a number of new cooperative threshold optical phenomena which appear in a system of three-level atoms or molecules because of the high-frequency Stark effect caused by the collective field. The thresholds of these critical phenomena depend on the intensities of both exciting fields and, in contrast to Refs. 1-4, these thresholds are controllable. This may be of interest in the construction of multifunction optical devices with tunable parameters. In contrast to the saturation effects which underlie the phenomenon of optical bistability in a system of two-level absorbers,[1-4] the high-frequency Stark effect results, under double optical resonance conditions, in a nonmonotonic dependence of the refractive index of the medium of the power absorbed by the medium on the field intensity. Then, the number of stable states of the system may exceed two even when the induced change in the optical length of the resonator is much less than the wavelength of light. The monotonic behavior of the absorbed power results in spontaneous oscillations of the "atom + field" system.

The effects discussed above should be detectable experimentally. The requirements to be satisfied in such detection are in practice no more stringent than those in the case of observation of bistability in resonators with saturable absorbers. The latter effect has already been observed experimentally under nonextremal conditions (see, for example, Refs. 5 and 35).

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Translated by A. Tybulewicz