DYNAMICS OF THE INTRACAVITY ABSORPTION SPECTRA OF A THREE – LEVEL SYSTEM WITH AN ACCOUNT OF A BLEACHING EFFECT

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The qualitative behavior of the laser whose frequency is continuously tuned within time over which the pulse acts and a medium inside the cavity has bleaching, is studied by numerical simulation. The medium is modeled as a closed three–level system with ground state and intermediate metastable level. It is shown that in comparison with the situation without bleaching of the medium, the distortions of the temporal dependence of lasing intensity in the region of selective absorption are much more pronounced. It is found that the position of the dip in lasing power as well as the frequency and the amplitude of oscillations caused by a nonlinear laser response on nonadiabatic frequency scanning throughout the line of selective absorption nontrivially depend on the scanning speed. The regions of quasicontinuous lasing are found, in which the distortions of the absorption line profile are negligible.

The method of intracavity laser spectroscopy (ICLS) has been successfully used in investigations of molecular and atomic systems (see, e.g., Ref. 1). The broad–band modification of the ICLS is most commonly used in applications because of its relative simplicity, high sensitivity and fast speed response. A narrow–band modification of the technique though being more complicated in experimental performance, provides much better spectral resolution if a single–mode tunable laser[2–4] is used. At the same time, theoretical models for description of experiments are simplified, in the condition under which the medium interacts with a single mode of laser radiation, and the continuous frequency tuning in the process of recording of the spectrum allows one to observe explicitly the origin and the temporal evolution of some nonlinear transient phenomena.[5]

A sufficiently general and frequently used model of absorbing medium is a three–level model which takes into account a great number of processes in actual molecular and atomic quantum systems. Metal vapor, if introduced into the cavity of a broad–band laser,[6] can strongly affects the lasing spectrum due to the so–called effect of spectrum condensing, while the time–dependent absorption of the type considered in Ref. 5 can change the lasing kinetics of a narrow–band tunable laser so that oscillations in lasing power may appear. These oscillations in their turn can be superimposed on the oscillations in lasing intensity caused by Q–switching as a result of frequency scanning throughout the selective absorption line.[2,7] Thus, the lasing of a laser with a three–level medium inside the cavity having the bleaching has a complex character specified by various physical reasons.

In this paper the qualitative behavior of lasing of a CW tunable laser and the medium inside the cavity having the bleaching (the narrow–band ICL spectrometer) are studied by numerical simulation. The laser is considered as a single–mode, the medium consisted of three–level atoms with the ground state and intermediate metastable level. From the practical point of view, this problem allows one to estimate the contribution of nonlinearity of light absorption in the distortion of an individual line profile recorded by the ICL spectrometer and to determine the boundaries of the region of the linear response of the spectrometer, which guarantee the record of the undistorted spectrum.

The derivation of the equations for lasing power of a single–mode sweep–frequency laser with the cavity filled with a three–level medium was based on the dynamic mode approach for the cavity with a uniformly moving mirror.[8] Standard derivation of the curtate equations for the amplitude of electric field has shown that sufficiently slow variation of lasing frequency affects only the frequency and phase characteristics of lasing, whereas the lasing power is well described by the ordinary Statz – de Wars equations.[9] For the description of the dynamics of the closed three–level system in the process of absorption of radiation resonant with an optical transition from the ground state, we used the balance equations for the level populations discussed in Ref. 5. Thus, the equations for lasing of a laser in the proposed model have the form

$$\dot{W} + \gamma_p W = \zeta_a WN - \zeta W(N_0 - N_1) K(t) ;$$

$$\dot{N} + \gamma_a (1 + \xi W)N = \gamma_a N_e ;$$

$$\dot{N}_0 + [\gamma + WK(t)] N_0 - [A - \gamma + WK(t)] N_1 = \gamma;$$

$$\dot{N}_1 + [1 + WK(t)] N_1 = WN_0 K(t) ; \qquad (1)$$

$$W = 4|d_{10}E/\hbar|^2/(\gamma_1\Gamma) ; \quad \zeta = \omega_{10}nl|d_{10}|^2/(L \hbar \gamma_1\Gamma) ;$$

$$\zeta_a = \omega_{10}n_a l_a |d_a|^2/(L\hbar\gamma_1\Gamma_a) ; \quad K(t) = 1/(1 + \Omega_2) ;$$

$$\xi = 2\gamma_1\Gamma \; |d_a/d_{10}|^2/(\gamma_a\Gamma_a) ; \; \Omega = \varepsilon t - \Omega_0; \; \gamma_p = \omega_{10}/2Q .$$

Here W is the dimensionless lasing power, which means simultaneously the parameter of saturation of the absorption on the resonance transition 0–1; N is the difference between the populations of the upper and lower levels, combining with the field, in the active medium of the length l_a; N_0 and N_1 are the populations of the ground– and upper–excited states of the absorbing three–level medium of the

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length l; ζ and ζ_a are the parameters which determine the rate of the absorption saturation and the coefficient of amplification of laser radiation, proportional to the concentrations of the absorbing $\eta(\zeta)$ and active $\eta_a(\zeta_a)$ media, in addition, ζ and ζ_a are inversely proportional to the dimensionless power of the absorption saturation in the passive and active media; E is the electric field intensity of the light wave; d_{10} and d_a are the matrix elements of the dipole moments of transitions resonant with the radiation in the absorbing and active media; ω_{10} is the frequency of the resonant transition in the absorbing medium; γ_1 and Γ are the rates of decay of the population of the upper level and of the ligth–induced polarization on the resonant transition of the three–level system; $\hbar$ is Planck's constant; Ω is the time–dependent frequency detuning of the laser radiation from the center of the absorption line in the passive medium; ε is the speed of frequency tuning; Ω_0 is the initial frequency detuning at time at which laser frequency sweeping is turned on; A is the first Einstein coefficient corresponding to the resonant transition 1–0 in the absorbing medium; γ is the rate of decay of the population of intermediate metastable levels of absorbing atoms in the passive medium; γ_a and Γ_a are the rates of decay of the population of the upper level and the light–induced dipole moment corresponding to the working transition of the active medium; ζ is the ratio of the saturation parameters for the active and passive media; L is the length of the laser cavity; N_e is the equilibrium difference between populations of levels in the active medium; Q is the cavity Q–factor. All the quantities entering into Eq. (1), which have the dimension of time (frequency), here and further are expressed in the units of $\gamma_1^{-1}(\gamma_1)$.

The estimates of the laser parameters were chosen based on the well–known experimental results, published in Ref. 2, and from the notion of the absorbing medium as the metallic vapors in a buffer gas at pressure lower than the atmospheric pressure. But the use of the initially chosen parameters in model (1) resulted in the nonstationary lasing even in the case of adiabatically slow variation of the tuning frequency of the laser cavity. Therefore, the values of the parameters were changed in the model under consideration in such a way that the regime of lasing could be assumed classic and could be described by the Statz – de Mars equations. This regime was realized in Ref. 2, namely, the periodic rapidly decaying oscillations occurred at the leading edge of the pulse, while the quasicontinuous lasing was recorded for the most part of the pulse. The value of correction for the initial parameters was less than an order of their magnitude.

In the numerical simulation the lasing power, the population inversion in the active medium, and the populations of the upper and lower levels of the absorbing medium were calculated as functions of time. With the goal of investigation of the effect of the bleaching of the medium, the populations of the absorbing medium levels were fixed in some cases. The only factor which affected the lasing dynamics was Q – switching, considered in Ref. 7, and the obtained results were similar. Their subsequent analysis has shown that nonlinearity of the absorbing medium leads to much more complex dynamics of lasing.

The steady–state solution of system (1) at the frequency detuning $\Omega = -\Omega_0$ has been chosen as the initial condition for making mass calculations, i.e., it is assumed that tuning of the laser frequency starts after the system reaches its steady state. This assumption allowed us to avoid the study of the strongly unstable regime of switching of the laser which was additionally perturbed by the nonlinearly absorbing medium. As a rule, the initial detuning Ω_0 was set equal to $10\gamma_1$. The special–purpose calculations have shown that all above–discussed phenomena contribute not less than 85 – 90 % of their maxima.

In this paper much attention is devoted to the effect of the speed of laser frequency tuning on the character of these processes. For low speeds of frequency tuning ($\varepsilon < 10^{-3}$), the interaction between radiation and absorbing medium was adiabatic, and the profile of the dip in lasing power (i.e., the signal recorded by the ICL spectrometer) coincided with the corresponding profile of the absorption line in the cases of both regarding and neglecting the effect of the bleaching of the medium. But in the first case the half–width δ of the dip was much greater because of light–induced broadening of the absorption line

$$\delta = [1 + (\gamma_1 - A + 2\gamma)\, W/\gamma_1\gamma]^{1/2} . \qquad (2)$$

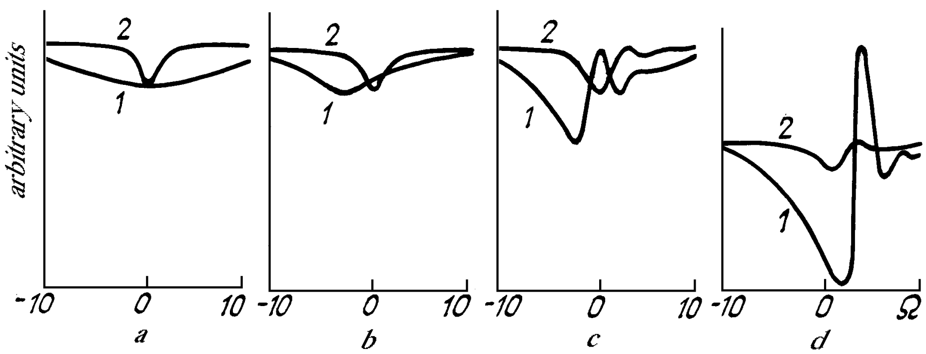

FIG. 1. The temporal dependence of lasing intensity of the laser with the absorbing medium inside the cavity: 1) with an account of the bleaching of the medium, 2) without an account of the bleaching of the medium. a) $\varepsilon \simeq 0$; b) $\varepsilon = 5\cdot10^{-3}$; c) $\varepsilon = 10^{-2}$; d) $\varepsilon = 1.6\cdot10^{-2}$. The values of the rest of the parameters are $\gamma_c = 7\cdot10^{-2}$, $\gamma_a = 10^{-2}$, $\gamma = 10^{-3}$, $A = 10^{-1}$, $\zeta = 7\cdot10^{-1}$, $\zeta_a = 5\cdot10^{2}$, $\zeta = 9$, $N_e = 3\cdot10^{-4}$, and $\Omega_0 = 10$.

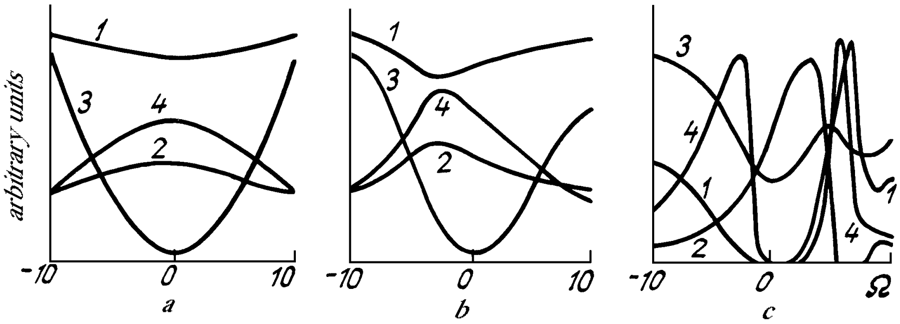

FIG. 2. The time dependences of the lasing power (1), the difference between the populations of the working levels of the active medium (2), and the populations of the lower (3) and upper (4) levels of the absorbing medium: a) $\varepsilon = 0$; *b)* $\varepsilon = 5\cdot10^{-3}$; *c)* $\varepsilon = 2\cdot10^{-2}$. *The rest of the parameters are the same as in Fig. 1.*

The shape of the dip deformed with increase of the tuning speed in such a way that its center was displaced with respect to the center of the absorption line, and the oscillations appeared in its trailing edge (Fig. 1). The distortion of the temporal dependence of lasing intensity in the region of selective absorption was much more pronounced when the nonlinearity of the absorbing medium and the "twisting" of populations were taken into account. The sharp decrease of the power, naturally, leads to strong oscillations in the regime of steady–state lasing established after frequency scanning throughout the absorption line. Build–up of quasistationary lasing in this case differs from that in the beginning of the pulse because the absorption is periodically switched on due to the light–induced line broadening effect at the moment when the radiation intensity is high (Fig. 2). Such an interaction had a character of a negative feedback and resulted in faster damping of oscillations.

The dependence of the dip position with respect to the center of the absorption line on the tuning speed ε turned out to be unexpectedly strong (Fig. 3). It is very important that the displacement is greater at low tuning speeds when the distortions of the dip shape are practically absent. The boundary line depicted in Fig. 3 corresponds to the appearance of the first maximum in lasing intensity after the frequency scanning throughout the dip, and thus it indicates the regions where distortion of the line profile is qualitatively different. Neglect of the bleaching of the medium, as can be seen from Fig. 3, yields an alternative result.

The period of oscillations arising after the frequency scanning throughout the absorption line weakly depends on the tuning speed (and practically remain unchanged for linear absorption), being 30% shorter in the first case and 8% shorter in the second for three–fold increase of the speed ε. Note that the period of oscillations is determined primarily by the parameters of the laser.

The large displacement of the dip center with respect to the center of the absorption line is explained as follows. When the speed of tuning is low, interaction of the tuning frequency with the broadened absorption line takes place during the time being long enough for establishing the bleaching of the medium before the lasing frequency approaches the line center (this is illustrated by Fig. 2*b* which shows that the upper–state population in the absorbing medium is most sensitive to the increase in the tuning speed). As a consequence, the absorption line "vanishes" for the laser, the dip is no longer growing, and the radiation power starts to increase. At hegher speed of tuning, the same bleaching is achieved when the lasing frequency approaches closer to the line center. And, finally, if tuning is very fast, then the medium has no time for establishing the bleaching. This leads to a sharp decrease in lasing power and practically to complete depletion of lasing (Figs. 2*c* and 1*d*). Note that the drop of the power continued even after the frequency scanning throughout the absorption line.

Thus, the dynamics of level populations in the absorbing medium inside the cavity of the single–mode tunable laser has a very strong qualitative effect on the characteristics of the signal recorded by the ICL spectrometer, namely, on the profile and depth of the dip in the temporal dependence of lasing power, and on the position of the dip center with respect to the absorption line center. We found that the displacement of the dip center nontrivially depends on the speed of frequency tuning, and its value may reach several absorption linewidths, under conditions of weak distortion of the dip shape.

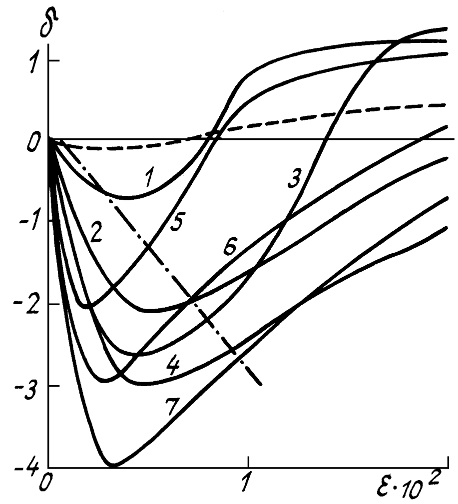

FIG. 3. The dependence of the displacement of the dip center on the speed of frequency tuning ε *without an account of the bleaching of the medium (the dynamics of level populations) (dashed curve) and with an account of it (solid curve). 1)* $\zeta = 0.3$, $A = 0.2$, *and* $\gamma = 4\cdot10^{-3}$; *2)* $\zeta = 0.3$, $A = 0.25$, *and* $\gamma = 1.5\cdot10^{-3}$; *3)* $\zeta = 0.7$, $A = 0.1$, *and* $\gamma = 10^{-3}$; *4)* $\zeta = 0.3$, $A = 0.1$, *and* $\gamma = 10^{-3}$; *5)* $\zeta = 0.3$, $A = 0.75$, *and* $\gamma = 5\cdot10^{-4}$; *6)* $\zeta = 0.3$, $A = 0.55$, *and* $\gamma = 5\cdot10^{-4}$; and, *7)* $\zeta = 0.7$, $A = 0.1$, *and* $\gamma = 5\cdot10^{-4}$. *The boundary of the region of occurrence of the oscillations is denoted by the dot – dash line: to the left of it the dip shapes have no oscillations. The values of the rest of the parameters are the same as in Figs. 1 and 2.*

And, finally, though the calculations were carried out for the parameters which are typical of the atomic systems, the general conclusions are also valid for the molecular systems by virtue of the similarity of the dynamics of bleaching of the three–level medium and rovibrational transitions of molecules.[5] Hence, for the quantitative analysis of measurements obtained with the help of the narrow–band ICL spectrometers, it is necessary to analyze the possible contribution of the above – considered phenomena.

REFERENCES

1. S.F. Luk'yanenko, M.M. Makogon, and L.N. Sinitsa, *Intracavity Laser Spectroscopy* (Nauka, Novosibirsk, 1985), 121 pp.
2. V.P. Kochanov, L.N. Sinitsa, and A.M. Solodov, Zh. Prikl. Spectrosk. **41**, No. 2, 335–338 (1984).
3. S.A. Batishche, V.A. Mostovnikov, and A.N. Rubinov, Kvant. Elektron. **3**, No. 11, 2516–2519 (1976).
4. S.V. Sidorov and A.I. Khizhnyak, in: *Kvantovaya Elektronika* (Naukova Dumka, Kiev, 1978), No. 14, pp. 46–53.
5. M.S. Zubova and V.P. Kochanov, Pis'ma Zh. Eksp. Teor. Fiz. **50**, No. 9, 376–378 (1989).
6. V.V Vasil'ev, V.S. Egorov, and I.A. Chekhonin, in: *Abstracts of Reports at the First Conference on Nonlinear and Coherent Phenomena in the Method of Intracavity Laser Spectroscopy*, A.S. Pushkin State Pedagogical Institute, Kirovograd (1988), pp. 10–11.
7. S.F. Luk'yanenko and A.M. Solodov, Zh. Prikl. Spektrosk. **49**, No. 2, 206–209 (1986).
8. R.I. Baranov and Yu.M. Shirokov, Zh. Eksp. Teor. Fiz. **53**, No. 6 (12), 2123–2130 (1967).
9. Ya.I. Khanin, *Quantum Radiophysics.* V. **2**. *Dynamics of Quantum Generators* (Sov. Radio, Moscow, 1975), 496 pp.