DECAY INSTABILITY OF VIBRATIONAL RELAXATION IN MOLECULAR GASES

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It is shown that with constant pumping in molecules of the CO_2 type the oscillatory transfer of energy between different modes of vibrations is possible. This effect can lead to a peaked mode of generation in lasers based on CO_2.

In lasers based on molecular gases such as CO_2, N_2O, H_2O, etc., the size of the inversion is determined by the kinetics of the energy exchange between different modes of vibrations and translational degrees of freedom. Under certain conditions the process of energy exchange can have a self-oscillatory nature. The development of self-oscillations in the case under consideration is analogous to the self-excitation of oscillations in nonlinear mechanical, biological, and chemical systems [1-3].

A system developed in [4] is usually used in the analysis of vibrational relaxation in mixtures of molecular gases. A virtue of this system is the reduction of the kinetic problem to an examination of a system of equations of energy balance for individual modes of vibrations, which is justified by the smallness of the times of energy exchange between the state of an individual mode compared with the times of intermode interaction.

Let us write the simplified equations describing the kinetics in a CO_2 laser in the form

$$dn_3/dt = q - W\,[n_3\,(n_2+1)^3 - \exp(-\delta E/T)\,n_2^3\,(n_3+1)] \tag{1}$$

$$dn_2/dt = {}^3/_2\,W\,[n_3\,(n_2+1)^3 - \exp(-\delta E/T)\,n_2^3\,(n_3+1)] - n_2/\tau_r \tag{2}$$

Here n_3 and n_2 are the average numbers of vibrational quanta in the asymmetrical and the doubly degenerate deformational modes of the CO_2 molecule, respectively. These values are related to the notation adopted in [4] in the following way: $2n_2 = \varepsilon_2$, $n_3 = \varepsilon_3$. The rate of formation of vibrational quanta, designated as q, effectively accounts for the excitation of the asymmetrical mode by electron collision or resonant energy transfer from N_2 or CO molecules. The source is omitted in (2) since with the appropriate choice of conditions, such as the composition of the mixture, its influence on the phenomenon under consideration can be neglected; W is the "effective" probability of decay of a quantum of the asymmetrical mode into three quanta of the deformational mode. As in [4], we assume the equality of the temperatures of the deformational and asymmetrical modes. Of the two channels of decay of an asymmetrical quantum ($n_3 \to 3n_2$, $n_3 \to n_1 + n_2$) the first is left. For small occupation numbers, taking account of both channels does not alter the form of the equations, but as W one must understand the total probability of decay by both channels. For large occupation numbers the contribution of the second channel is unimportant if the probabilities of decay in the two reactions are comparable. The energy defect in the reaction $n_3 \to 3n_2$ is designated as δE, and τ_r^{-1} is the probability of vibrational-translational relaxation of the deformational mode.

The stationary state of system (1), (2) is determined by the equations

$$\begin{cases} \varepsilon_2^\circ = 3q\tau_r \\ \varepsilon_3^\circ = [8q/W + \exp(-\delta E/T)(\varepsilon_2^\circ)^3]/[(\varepsilon_2^\circ+2)^3 - \exp(-\delta E/T)(\varepsilon_2^\circ)^3] \end{cases} \tag{3}$$

Let us examine this stationary state for stability. As a function of the parameters

$$\alpha \equiv q\tau_r,\quad \beta \equiv 8/W\tau_r,\quad \gamma \equiv \exp(-\delta E/T)$$

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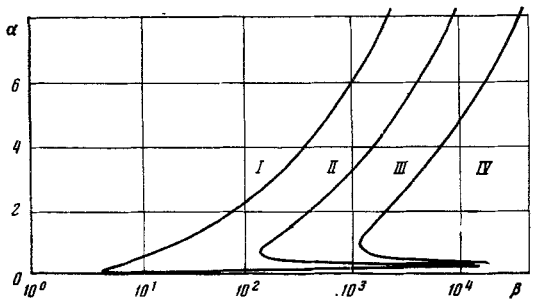

Fig. 1

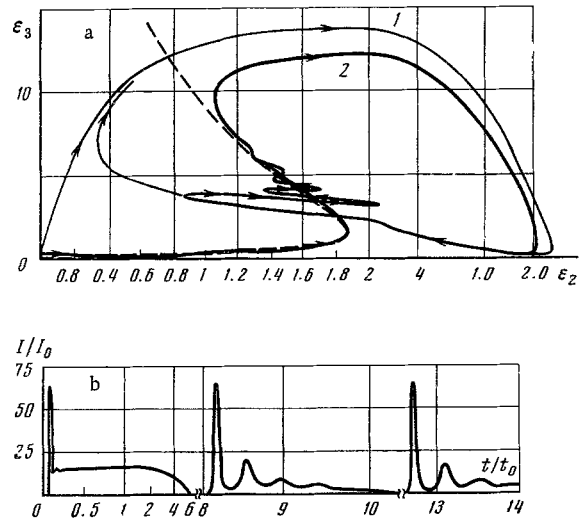

Fig. 2

the roots of the characteristic equation correspond to different types of stationary points. The nature of the stationary point changes upon crossing through the curves in the α, β plane shown in Fig. 1 (for the case of $\gamma = 0.16$): in region I there is a stable node, in II a stable focus, in III an unstable focus, and in IV an unstable node. Actually the parameters α and β vary as a function of the composition of the gas mixture and the degree of ionization in the electrical discharge ($25 \leq \beta \leq 3 \cdot 10^3$, $0 < \alpha \leq 5$), and one can select conditions in which systems I, II, and III are realized. In cases III and IV self-oscillations with a limiting cycle surrounding the unstable stationary point develop in the system. Physically the instability has a decay nature and has the increment

$$\mathrm{Im}\,\nu = -\tfrac{1}{16} W [(2 + \varepsilon_2^\circ)^3 - \gamma (\varepsilon^\circ_2)^3 - 9\,\varepsilon_3^\circ (\varepsilon_2^\circ + 2)^2 + 9\,\gamma\,(\varepsilon_2^\circ)^2 \times (\varepsilon_3^\circ + 1) + \beta]$$

This effect can exert an important influence on the nature of light generation in molecular lasers. The system of equations for the reduced energies ε of the vibrational modes and the light intensity J averaged over the length of the resonator [5] has the form

$$d\varepsilon_3/dt = q - \tfrac{1}{8} W [\varepsilon_3 (\varepsilon_2 + 2)^3 - \gamma\,\varepsilon_2^3 (\varepsilon_3 + 1)] - J\,\sigma\Delta / \hbar\omega \quad (4)$$

$$d/dt\,(\varepsilon_2 + 2\varepsilon_1) = \tfrac{1}{8} 3W [\varepsilon_3 (\varepsilon_2 + 2)^3 - \gamma\,\varepsilon_2^3 (\varepsilon_3 + 1)] + 2J\sigma\Delta / \hbar\omega - \varepsilon_2 / \tau_r \quad (5)$$

$$dJ/dt = cJ\,\sigma\,N\,(\Delta - \Delta_0) \quad (6)$$

Considering the generation of light at the wavelength of 10.6 μ to be specific, for the value of Δ we have

$$\Delta = 16\,(\varepsilon_2 + 1)\,[\varepsilon_3 / (\varepsilon_3 + 1) - \varepsilon_2^2 / (\varepsilon_2 + 2)^2] / [(\varepsilon_2 + 2)^4 (\varepsilon_3 + 1)]$$

In (4)-(6) $\varepsilon_1 = \varepsilon_2^2/4\,(\varepsilon_2 + 1)$, σ is the cross section of resonant light absorption, ω is the frequency, N is the concentration of active molecules, and $N\Delta_0$ is the threshold inversion. As an analysis of system (4)-(6) shows, for a given value of Δ_0 as a function of the parameters α and β either a stationary state with $J \neq 0$ is realized (in which case self-oscillations do not develop) or a stationary state with $J \neq 0$ is absent ($\alpha > \alpha_*$). For $\Delta_0 \ll 1$ the boundary of the region separating these two cases is determined by the condition $\alpha_* \sim \Delta_0^{-1/5}$. If in this case α and β lie in region III or IV (Fig. 1) then system (4)-(6) has a limiting cycle in which the peaked generation of light is possible against the background of self-oscillations in ε_2 and ε_3. The projection of the phase trajectory of system (4)-(6) on the ε_2, ε_3 plane calculated for the case $\gamma = 0.16$, $\alpha = 1$, $\beta = 8 \cdot 10^2$, and $\Delta_0 = 0.03$ is shown in Fig. 2a. Curve 1 is the phase trajectory of system (4)-(5) when $J = 0$ and curve 2 pertains to the full system (4)-(6). The dashed curve is the boundary of the region in which light generation is possible. The scale along the ε_2 axis from 0 to 2 is linear and for $\varepsilon_2 > 2$ it is logarithmic. The time dependence of the intensity of the light signal is presented in Fig. 2b in relative units, $J_0 = 0.1 N c h \omega \Delta_3$. The time scale is $t_0 = 100/Nc\sigma\Delta_0 \approx 10^{-6}$ sec; in the section $1 < t < 6$ the scale is logarithmic and for $t < 1$ and $t > 6$ it is linear. Such a mode is realized if the limiting phase trajectory of system (4)-(5) when $J = 0$ crosses the boundary of the region $\Delta = \Delta_0$; otherwise the generation of a single light pulse occurs.

The phenomenon of self-oscillations can be detected experimentally either in the form of oscillations in the coefficient of amplification of a weak signal or in the properties of the generation modes of the system in the resonator (the peaked mode).

Under real conditions the nature of the phenomenon discussed is influenced by various effects not taken into account in the model adopted: variation in the gas temperature, on which τ_r, W, and γ depend; the degree of ionization and collision of the second kind, which affects q. Chemical reactions proceeding in the system can alter the composition of the mixture and with it the parameters α and β. Trial calcula-

tions taking into account temperature variations and collisions of the second kind were carried out in which new effects appeared. In particular, because of collisions of the second kind with $\alpha > \alpha_c$ ($\alpha_c \approx 2.4$) three stationary points develop for fixed α and β. The maxima of α and β are strongly decreased but the characteristic properties of intermodal transfer are retained qualitatively.

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