ANALYTICAL THEORY OF STEADY GENERATION ON AN OVERTONE OF THE CO MOLECULE

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An analytical theory of steady generation of an overtone of the CO molecule is constructed on the basis of a continuous quasiresonance approximation. The analytical theory is compared with the results of a numerical calculation based on the complete set of kinetic equations.

Theoretical [1, 2], and experimental [3, 4] reports have recently appeared devoted to the study of the characteristics of a laser based on the first overtone of the CO molecule, permitting the expansion of the generation spectrum into the little-developed range of ~3 μm. In [1, 2] the amplification factor, power, and generation spectrum are investigated on the basis of a numerical solution of the equations of multilevel vibrational kinetics together with the equations for the emission intensity. The results of these reports do not make it possible to obtain simple parametric functions for the generation characteristics. The analytical theory of a steady CO laser at the fundamental frequency constructed in [5-7] allows one to find the generation spectrum and the efficiency as functions of the pumping power and the properties of the working medium and the resonator. At the basis of this theory lies the quasiresonance continuous approximation [8, 9], which makes it possible to find the distribution function over the vibrational levels of anharmonic molecules under conditions of strong nonequilibrium.

In the present report the approach developed in [5-7] is used for an analytical description of steady generation on the first overtone of the CO molecule.

First let us consider the case when radiation generation at the fundamental frequency is absent. Let W(v) be the number of quanta emitted per second by a CO molecule in the transition $(v + 2, j_v - 1 \to v, j_v)$ with a frequency $\omega(v)$, where v and j_v are the vibrational and rotational quantum numbers, respectively. For the quantity W(v) by definition we have

$$W(v) = \frac{J(v)\,\sigma(v)}{h\omega(v)}\,[f(v+2) - \delta_v f(v)], \tag{1}$$

where J(v) and σ(v) are the intensity and the effective cross section for stimulated emission in the transition under consideration, respectively; f(v) is the fraction of CO molecules which are at the v-th vibrational level; $\delta_v = \exp(-2B_v j_v/T)$; B_v is the rotational constant, which depends weakly on v; T is the temperature of the medium. Using (1), the system of equations for f(v) in the generation mode can be written in the form (cf. [5-7])

$$2W(v) + W(v-1) + W(v+1) = \Phi[v,\ f(v)] - \Phi[v+2,\ f(v+2)], \quad v = 0,\ 1,\ 2,\ \ldots, \tag{2}$$

where Φ[v, f(v)] is the flux of quanta in the cross section v of the space of vibrational numbers,

$$\Phi[v,\ f(v)] = \sum_{m=v}^{\infty}\Big\{\sum_{n=0}^{\infty}[Q_{m,m+1}^{n+1,n} f(m) f(n+1) - Q_{m+1,m}^{n,n+1} f(m+1) f(n)] - \\ -(P_{m+1} + A_{m+1}^{(1)} + A_{m+1}^{(2)}) f(m+1) + A_{m+2}^{(2)} f(m+2) + \sum_{n=0}^{m} q_n\Big\}. \tag{3}$$

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Here $Q^{n,n+1}_{m+1,m}$ is the frequency of V–V exchange; P_m is the frequency of relaxation from the m-th level; $A^{(k)}_m$ is the frequency of spontaneous emission in the transition ($m \to m - k$); q_n is the frequency of excitation of the n-th level by the outside source, Converting to the continuous variable v and using the approximation of resonance V–V exchange, we can reduce (2) to the form [5-9]

$$4W(v) = \widetilde{\Phi}[v, f(v)] - \widetilde{\Phi}[v+2, f(v+2)], \tag{4}$$

where

$$\widetilde{\Phi}[v, f(v)] = \nu(T)\left[(v+1)^2 f^2(v)\left(1 - \frac{T}{2\Delta E}\frac{d^2 \ln f}{dv^2}\right)\right] + \int [P(v) + A(v)] f(v)\, dv, \tag{5}$$

$\nu(T)$ is the effective frequency of V–V exchange [7]; ΔE is the anharmonism energy; $A(v) = A^{(1)}_{v+1} + 2A^{(2)}_{v+2}$; $P(v) = P_v$. In the derivation of (4)-(5) it was assumed that the excitation by the outside source takes place at lower vibrational levels, where generation is absent. Simple estimates show that both inside the generation region and outside it the logarithmic derivative in the expression for the flux $\tilde{\Phi}[v, f(v)]$ in (5) can be neglected. Then (5) is simplified and takes the form

$$\widetilde{\Phi}[v, f(v)] = \nu(T)(v+1)^2 f^2(v) + \int f(v)[P(v) + A(v)]\, dv. \tag{6}$$

Equation (4) is supplemented by the condition of steady generation

$$\sigma(v)[f(v+2) - \sigma_v f(v)] = \Delta / N_{CO}, \tag{7}$$

where Δ is the threshold amplification factor; N_{CO} is the density of CO molecules, Equations (4)-(7) fully solve the problem of determining the generation spectrum if the left edge l of the generation spectrum and the value of the flux $\Phi[l, f(l)]$ of quanta in the cross section l are known. Neglecting V–T losses and spontaneous emission at $v \leqslant l$, the flux of quanta can be equated to the power of the outside source:

$$\widetilde{\Phi}[v, f(v)]|_{v<l} = \eta_V \frac{W_0}{E_1 N_{CO}} \equiv \nu(T) c^2, \tag{8}$$

where W_0 is the pumping power; η_V is the fraction of the power going into the excitation of vibrations; E_1 is the energy of a lower vibrational quantum. If the flux of vibrational quanta in the cross section v is known, then the quantity f(v) can be found from (6);

$$f(v) = -Z + \sqrt{Z^2 + \frac{\Phi[v, f]}{\nu(v+1)^2}} \equiv F[v, \Phi], \tag{9}$$

where

$$Z = \frac{1}{2\nu(v+1)^2 f} \int f(P + A)\, dv.$$

Using (7)-(9), (4) can be reduced to the form

$$4W(v) = \nu c^2 - \sum_{i=l}^{v-1} 4W(i) - \tilde{\Phi}\left[v+2, \frac{\Delta}{\sigma(v) N_{CO}} + \delta_v F\left(v, \nu c^2 - \sum_{i=l}^{v-1} 4W(i)\right)\right], \tag{10}$$

where $\tilde{\Phi}$ and F are defined in (6) and (9), respectively,

The summation in (10) is carried out only over even i if $(v - l)$ is even and over odd i if $(v - l)$ is odd. The recurrent equation (10) is convenient for calculation, but it contains one unknown parameter, the number l. The left edge l of the generation spectrum can be found using the amplication factor $K_0(v)$ for a weak signal, calculated from the distribution function $f_0(v)$ undisturbed by emission:

$$K_0(v) = N_{CO} \frac{\sigma(v) c}{v+1}\left(\frac{v+1}{v+3} - \delta_v\right). \tag{11}$$

Since $\sigma(v)$ grows approximately quadratically with the number v, $K_0(v)$ is a growing function of v, and the number j_v for which the condition $K_0(l, j_l) \geqslant \Delta$ is satisfied should first appear at $v = l$. The rotational quantum number j_v at which generation takes place and on which $\sigma(v)$ and δ_v depend in (10) should correspond to the maximum amplification factor K(v) calculated from the distribution function f(v) disturbed by emission:

$$K(v) = N_{CO}\sigma(v)\left\{F\left[v+2,\ vc^2 - \sum_{i=l}^{v} 4W(i)\right] - \delta_v F\left[v,\ vc^2 - \sum_{i=l}^{v-1} 4W(i)\right]\right\}, \tag{12}$$

where, just as in (10), the summation is carried out over even or odd i, depending on the parity of the number $(v - l)$. As is seen from (12), j_v itself depends on W(v). For the final calculation of W(v) we can use the following iteration process (its convergence is obvious):

$$W_n(v) = W\{v,\ j_v[W_{n-1}(v)]\}, \tag{13}$$

taking $W_0(v) = 0$ as the zeroth approximation. In Fig. 1 the generation spectrum calculated by the above-described method is compared with the results of an exact numerical solution of the system (2) by the method presented in [10]. As seen from Fig. 1, the analytical theory describes the generation spectrum well enough. The calculations were made on a BÉSM-6 computer, with two orders of magnitude more computer time being required for the realization of the method of numerical solution of the system (2) of [10] than for the calculation by Eq. (13). The generation spectrum calculated from Eq. (13) without allowance for V–T processes and losses to spontaneous emission is presented in Fig. 1 for comparison. It is seen from the figure that V–T processes affect the quantitative results. Let r be the right edge of the generation spectrum and $W(r) = 0$; then from (10) we can obtain an expression for the generation efficiency, assuming that V–T processes weakly distort the distribution function:

$$\eta \simeq 1 - \frac{(r+3)^2}{c^2}\left[\frac{\Delta}{\sigma(r)(1-\delta_r)} + Z(r)\right]^2, \tag{14}$$

where Z(r) is defined in (9): $Z(r) \simeq \dfrac{1}{2v(r+1)^2}\int (P+A)\,dv$. In the mode beyond the threshold the number r depends weakly on the pumping power and is determined mainly by the rate of the V–T processes. The efficiencies as functions of the threshold amplification factor at different temperatures, calculated from the analytical and exact theories, are compared in Fig. 2. As seen from Figs. 1 and 2, the agreement of the numerical calculations with the analytical theory permits its use in the construction of parametric functions for steady lasers based on the first overtone of the CO molecule.

In the experiments of [3, 4] generation took place simultaneously at both the fundamental frequency and the first overtone. Let the threshold amplification factor for the fundamental frequency equal Δ_1. Then in the region of values of v covered by generation at the fundamental frequency the function f(v) must satisfy the condition of steady generation

$$\sigma_1(v)[f(v+1) - \delta_1(v)f(v)] = \Delta_1/N_{CO}, \tag{15}$$

where $\sigma_1(v)$ is the cross section for stimulated emission on the transition $(v + 1, j_v - 1 \to v, j_v)$. Since in general one function f(v) cannot satisfy Eqs. (7) and (14) at once, the regions of generation of the fundamental frequency and the second harmonic do not overlap.

Since $\sigma_1(v)$ grows faster than $\sigma(v)$ with an increase in v, the generation of the fundamental frequency obviously takes place at lower vibrational levels. Therefore, the presence of generation at the overtone does not affect the generation of the fundamental frequency, and hence all the results involving generation of the fundamental frequency presented in [5–7] remain in force. Let η_1 be the efficiency of generation at the fundamental frequency; then the flux $\tilde{\Phi}[v, f]$ of quanta beyond the limits of the generation region is

$$\tilde{\Phi}[v,\ f]|_{r_1 \leqslant v \leqslant l} = vc^2(1 - \eta_1), \tag{16}$$

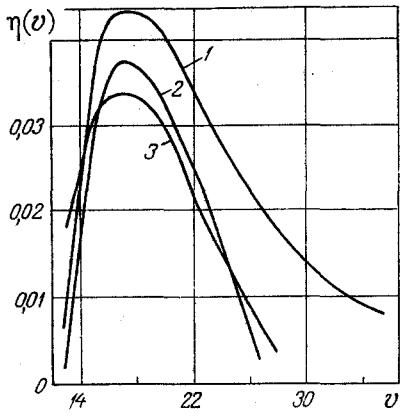

Fig. 1

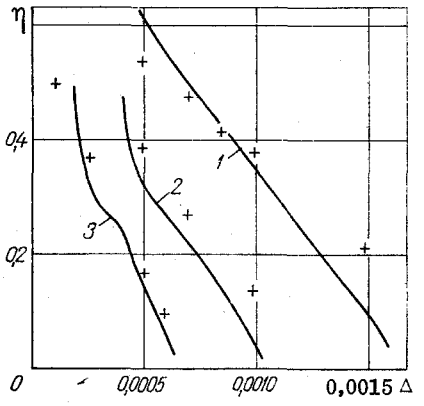

Fig. 2

Fig. 1. Dependence on v of the efficiency η(v) of generation in an individual transition (mixture of CO:He = 8.8:91.2, P = 20 torr, T = 100°K); 1) analytical theory without allowance for V–T processes and spontaneous emission; 2) with allowance for losses; 3) numerical calculation. $W = 1.5$ W/cm^3, $\Delta = 10^{-3}$ cm^{-1}.

Fig. 2. Dependence of generation efficiency on threshold amplification factor (CO:He = 8.8:91.2; P = 20 torr, T = 100°K): 1) T = 100°K; 2) 150; 3) 200; points) results of numerical calculation. $W = 1.5$ W/cm^2, cm^2,* Δ, cm^{-1}.

where r_1 is the right edge of the spectrum of generation of the fundamental frequency. It is seen from a comparison of (16) and (8) that all the equations involving generation at the overtone remain in force; one must only replace c by $\sqrt{1 - \eta_1}c$.

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