

SATURATION OF ABSORPTION OF THE POWERFUL
RADIATION OF A SYSTEM OF ANHARMONIC OSCILLATORS

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Recently, the theory of nonequilibrium systems simulated by a set of anharmonic oscillators has received significant development. The investigation of such kinds of systems is especially important in the study of problems associated with the stimulation of chemical reactions and the development of effective molecular lasers. The systematic analysis of the kinetics of anharmonic oscillators assumes the simultaneous solution of a large number of nonlinear equations describing the population balance of the vibrational levels. Realization of this approach is associated with cumbersome numerical calculations and does not permit obtaining a qualitative picture of the behavior of the system as a function of the different parameters (pressure, temperature, etc.). An approximate analytical theory has been formulated in [1, 2] which permits finding the distribution function over the vibrational states with the effects of anharmonicity taken into account. We will employ the approach developed in these papers to describe a system of anharmonic oscillators under conditions of powerful optical pumping. This problem was discussed in [3], where it was found that such a system changes into a saturation mode in the case of high pumping levels. The existence of this mode is explained by the fact that the maximum rate of energy input into a vibrational degree of freedom is determined by the rate of distribution of this energy over all the vibrational levels, i.e., by the constant of V-V-exchange. For sufficiently large pumpings the approximation of the Boltzmann distribution function adopted in [3] in connection with the calculation of the saturation parameters is too crude. The goal of this paper is to derive in explicit form expressions for the vibrational energy supply, the absorbed power, and so on, under saturation conditions without the use of the approximation indicated above [3].

Let us write the system of equations for the distribution function over the vibrational states in the approximation of single-quantum exchange in the following form:

$$\sum_{m=0}^{\infty} (Q_{i,i+1}^{m,m+1} f_m f_{i+1} - Q_{i+1,i}^{m+1,m} f_{m+1} f_i) + (A_{i+1} + P_{i+1}) f_{i+1} = \frac{\sigma I}{h\nu} (\delta_0 f_0 - f_i) \delta_{i0}, \quad i = 0, 1, 2, \dots, \quad (1)$$

where f_i is the fraction of the particles on the i -th level, $Q_{i+1,i}^{m,m+1}$ is the frequency of V-V-exchange, A_i is the frequency of spontaneous decay of the i -th state, P_i is the frequency of relaxation from the i -th level, $\delta_0 = \exp\left[\frac{B\Delta j}{T}(2j + \Delta j + 1)\right]$, B is the rotational constant, j is the rotational quantum number, σ is the effective absorption cross section of radiation at the transition $v=0, j \rightarrow v=1, j+\Delta j$ with frequency ω , I is the intensity of the pumping radiation resonant to the transition, T is the temperature of the medium, and δ_{ik} is the Kronecker symbol.

The system (1) is appreciably simplified upon the replacement of the discrete subscript i with a continuous variable. Such an approximation is valid if f_i varies little upon a change of the subscript by unity, i.e., if the condition $|f_i - f_{i+1}| \ll f_i$ is satisfied. As estimates show, this condition is satisfied for sufficiently large values of I .

The approximation [2]

$$P_{i+1} = P_{i0}(i+1) \exp(\delta_{VT} i),$$

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where δ_{VT} is a number independent of the gas composition and its temperature, is usually employed for the quantity P_{i+1} . It is possible to represent the frequency of V-V-exchange in accordance with [4] in the form

$$Q_{i+1, i}^{m, m+1} = Q_{10} (m+1) (i+1) \exp \left\{ -\Delta_{VV} (m-i)^2 + \theta (m-i) \frac{2\Delta E}{T} (m-i) \right\}, \quad (2)$$

where Δ_{VV} is a number independent of the temperature and the kind of molecules; for CO, for example, $\Delta_{VV} = 24/T$; ΔE is the anharmonicity energy, and

$$\theta(x) = \begin{cases} 0, & x < 0, \\ 1, & x > 0. \end{cases}$$

The principle of detailed balancing is taken into account in Eq. (2), so that

$$Q_{i+1, i}^{m, m+1} = Q_{i, i+1}^{m+1, m} \exp \left\{ \frac{2\Delta E}{T} (m-i) \right\}.$$

Switching in (1) to a continuous variable v and using the approximation of resonant V-V-exchange, we obtain the following equation for the distribution function $f(v)$ [2]:

$$v \frac{d}{dv} \left[f^2 (v+1)^2 \left(\frac{\kappa^2}{2} - \frac{d^2 \ln f}{dv^2} \right) \right] = \frac{\sigma I}{\hbar \omega} (f' + \gamma f) \delta(v), \quad (3)$$

where $v = Q_{10} \Delta_{VV}^{-3/2} \frac{\sqrt{\pi}}{4} \left[\frac{1}{2} + \frac{4}{\sqrt{2\pi}} D_{-3} \left(-\frac{2\Delta E}{T\sqrt{2\Delta_{VV}}} \right) \exp \left(\frac{\Delta E^2}{2T^2 \Delta_{VV}} \right) \right]$; $\frac{\kappa^2}{2} = \frac{2\Delta E}{T}$; $D_p(x)$ is the parabolic cylinder function, $\gamma = 1 - \delta_0$; $\delta(v)$ is the Dirac delta function. The V-T-losses are not taken into account in (3), as is valid at very large v . Integrating (3) over v , one can obtain

$$(f^2)'' - \kappa^2 f^2 = \frac{K^2 (f'(0) + \gamma f(0))}{(v+1)^2} + 4(f')^2, \quad (4)$$

where $K^2 = \frac{\sigma I}{\hbar \omega} \cdot \frac{2}{v}$ is the dimensionless magnitude of the intensity of the incident radiation.

Let the distribution function satisfy the condition

$$\left| \frac{K^2 (f'(0) + \gamma f(0))}{(v+1)^2} \right| \gg 4(f')^2. \quad (5)$$

The restrictions which (5) imposes on j and T will be discussed below. Then one can write the solution of Eq. (4) approximately in the following form:

$$f^2(v) = f^2(0) e^{-\kappa v} + \alpha \kappa [e^{-\kappa(v+1)} Ei(\kappa v + \kappa) + e^{\kappa(v+1)} Ei(-\kappa v - \kappa) - e^{-\kappa v} \alpha \kappa [e^{-\kappa} Ei(\kappa) + e^{\kappa} Ei(-\kappa)]], \quad (6)$$

where $\alpha = -\frac{K^2}{2\kappa} (f'(0) + \gamma f(0))$ is the dimensionless magnitude of the power absorbed by the system and $Ei(x)$ is the exponential integral function.

Using (6), it is possible to derive a relation between the absorbed power, the value of the distribution function at the origin, and the radiation intensity

$$\alpha = \frac{K^2 f(0) (1 - 2\gamma/\kappa)}{4 \left[1 + \frac{K^2}{2f(0)} (1 + \kappa e^{\kappa} Ei(-\kappa)) \right]}. \quad (7)$$

As is evident from (6) and (7), saturation of the absorbed energy occurs for large intensities of the pumping radiation

$$\alpha = \frac{\alpha_0}{1 + \frac{K_0^2}{K^2}}, \quad (8)$$

where α_0 is the maximum absorbed energy and K_0^2 is the saturation intensity

$$K_0^2 = \frac{2^{3/2} \alpha_0^{1/2}}{(1 + \kappa e^{\kappa} Ei(-\kappa))^{1/2} (1 - 2\gamma/\kappa)^{1/2}}. \quad (9)$$

It is possible to find the quantity $\alpha_0^{1/2}$ from the normalization condition of the limiting distribution function $f_0(v)$ corresponding to complete saturation. Since it is possible at low temperature to assume $\kappa^2(v+1)^2 \gg 2$ with good accuracy already at $v \geq 2$, it is possible to obtain

$$f(v) |_{v \gg 2} \simeq \left(\frac{2\alpha}{\kappa}\right)^{1/2} \frac{1}{v+1}. \quad (10)$$

from (3) by an expansion in the parameter $2/\kappa^2(v+1)^2$. It is possible at large v to take approximate account of V-T-processes by following [2]. Then, using the normalization conditions, we obtain

$$\alpha_0^{1/2} = 2^{-1/2} \left[\frac{(1 + \kappa e^{\kappa E_i(-\kappa)})^{1/2} (2-\gamma)}{(1-2\gamma/\kappa)^{1/2}} + \frac{1}{\kappa^{1/2}} \ln \left(\frac{1}{3\delta_{VT}} \ln \frac{2^{1/2}}{\kappa^{1/2}} \frac{1}{\mu} \right) \right]^{-1}, \quad (11)$$

where $\mu = P_{10}/v\kappa^2\delta_{VT}$ [2].

The supply of vibrational quanta corresponding to the complete saturation mode is given by the expression

$$\epsilon_0 = f_0(0)(3-2\gamma) - 1 + \left(\frac{2\alpha_0}{\kappa}\right)^{1/2} \frac{\ln \left(\frac{2\alpha_0}{\kappa}\right)^{1/2} \frac{1}{\mu}}{\delta_{VT}}, \quad (12)$$

where $f_0(0)$ is the value of the distribution function at the origin

$$f_0^2(0) = 2\alpha_0(1-2\gamma/\kappa)^{-1}(1 + \kappa e^{\kappa E_i(-\kappa)}). \quad (13)$$

Condition (5) imposes restrictions on the region of applicability of Eqs. (6)-(12). Taking (10) into account, it is sufficient to require continuity of the distribution function at the origin, i.e.,

$$2\gamma^2 < \frac{\kappa - 2\gamma}{1 + \kappa e^{\kappa E_i(-\kappa)}}. \quad (14)$$

In the case of the CO molecule the condition (14) is satisfied when $j < (T/2B)(\Delta j + \sqrt{1+\kappa})$.

Now let us find the relation of the distribution function at the origin to the absorbed power as $I \rightarrow \infty$ on the interval $0 \leq v < 2$ without using the continuous approximation. With (10) taken into account, the system (1) is resolved for $i=0, 1$

$$f_0^2(0) = \alpha_0 D^2, \quad (15)$$

where $D = \frac{1}{2A}(B + \sqrt{B^2 + 4AC})$; $A = Q_{10}^{12}\delta_0^2$; $B = \sqrt{\frac{2}{\kappa}} \left(\frac{Q_{21}^{01}}{3} - \delta_0 \sum_{m=2}^{\infty} Q_{12}^{m+1,m} \frac{1}{m+2} \right)$; $C = \frac{2}{3} \frac{1}{\kappa} \sum_{m=2}^{\infty} \frac{Q_{21}^{m,m+1}}{m+1}$.

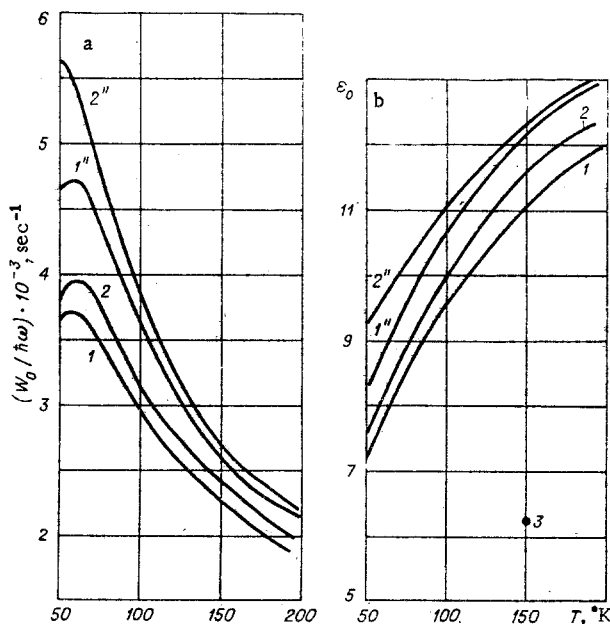


Fig. 1

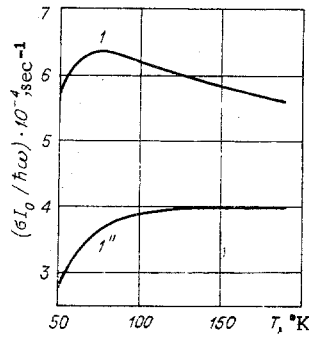


Fig. 2

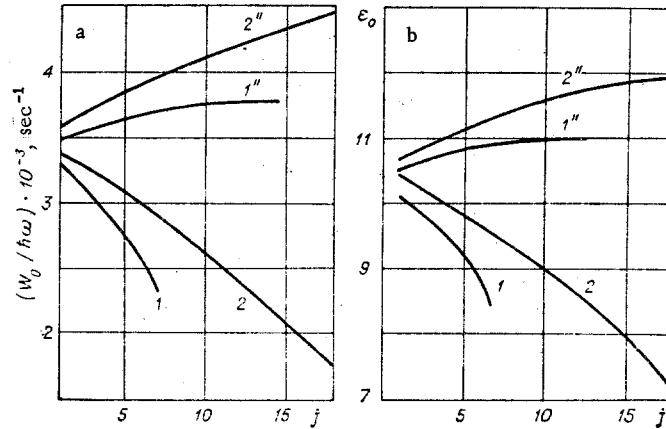


Fig. 3

Now it is possible to obtain from the normalization condition and from (12) the absorbed power and the supply of quanta corresponding to the relation (15).

The expressions for the dimensional values of the power per molecule and the intensity corresponding to complete saturation are of the form

$$W_0 = \hbar\omega\alpha_0\nu\kappa, \quad I_0 = \frac{\hbar\omega}{2\sigma} K_0^2\nu.$$

The dependence of the limiting absorption rate of radiation quanta and the limiting supply of vibrational quanta per CO molecule on the temperature of the medium is presented in Fig. 1 for the cases of absorption with $\Delta j = \pm 1$ (mixture of CO (0.2 mm Hg) + He (6 mm Hg) for $j=4$). Curves 1 ($\Delta j = -1$) and 1'' ($\Delta j = 1$) correspond to Eq. (13); curves 2 ($\Delta j = -1$) and 2'' ($\Delta j = 1$) correspond to Eq. (15); the point 3 corresponds to the result of [3]. The good agreement of the results obtained with the help of (13) and (15) permits using with good accuracy the analytic equations (8)–(13) in connection with the description of the saturation mode. The disagreement with the result of [3] is evidently explained by the crudeness of the approximation adopted in [3] for the distribution function and by the inaccuracy of the expression for the coefficient ν in [3] (see (5)). The dependence of the saturation intensity on temperature is presented in Fig. 2; the conditions and notation are the same as in Fig. 1.

The dependences of the limiting supply of vibrational quanta and the absorbed power per molecule on the rotational number at which pumping occurs are presented in Fig. 3 for a constant temperature ($T=100^\circ\text{K}$; the composition of the mixture and the notations are the same as in Fig. 1).

One should note that in the case of large intensities of the pumping radiation saturation of the absorption can occur in the system under discussion due to the finiteness of the rotational relaxation time. The intensity I necessary for this to occur is given by the estimate

$$\sigma_r I / \hbar\omega \simeq 1/\tau_r,$$

where τ_r is the rotational relaxation time. In order that the indicated effect not dominate the saturation effect produced by the finiteness of the V–V-exchange time, it is necessary that the condition

$$\frac{K_0^2}{2} \nu \tau_r < Z_r (2j + 2\Delta j + 1) e^{-\frac{B}{T} (j + \Delta j)(j + \Delta j + 1)}, \quad (16)$$

be satisfied, where Z_r is the rotational partition function. Condition (16) is realized for a large fraction of absorption bands, with the exception of the far wings corresponding to large j .

The quantity W_0 characterizes the maximum rate of energy input into the system. Since all the energy introduced changes into translational degrees of freedom, it is possible to consider W_0 as the limiting rate of vibrational relaxation.

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ENERGY CHARACTERISTICS OF A CARBON MONOXIDE GASDYNAMIC LASER

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The characteristics of gasdynamic lasers based on mixtures of carbon monoxide with nitrogen and inert gases were investigated and the populations of vibrational levels of CO molecules, the gain of the mixture, and the generation power were determined in [1-8]. But the parameters of a gasdynamic laser (GDL) in the optimum emission mode have not been determined up to now. The difficulties in calculating the optimum energy characteristics are connected with the complexity of the calculating model and the large number of parameters of the system. The energy characteristics of a CO gasdynamic laser are calculated and optimized in the present report on the basis of a simple model.

1. Calculating Model. Let us consider the escape of a binary gas mixture $\psi_{CO} + \psi_{N_2}$ (ψ_{CO} and ψ_{N_2} are the molar fractions of CO and N_2 , respectively) from the flat supersonic nozzle of a gasdynamic laser having a critical cross section with a height h_* and an initial aperture half-angle φ . At a degree of expansion S_0/S_* the expanding part of the nozzle changes into a plane-parallel section where the optical resonator is mounted.

We make the following assumptions, permitting a simplified calculation of the energy characteristics of a CO gasdynamic laser:

1. Losses of vibrational energy as a result of V-T processes occur mainly in the initial section of supersonic escape near the critical cross section of the nozzle.
2. The time the gas spends in the resonator exceeds the characteristic time of establishment of a quasi-steady distribution of molecules over the vibrational energy levels.
3. We consider a plane-parallel Fabry-Perot resonator which forms a nondiverging light flux in a geometrical treatment.

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