EFFECT OF COLLISIONS ON THE NATURE OF SATURATION OF THE VIBRATIONAL ROTATIONAL TRANSITIONS OF THE CO_2 $00^0 1-10^0 0$ *BAND*

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The effect of collisions on the nature of saturation of the $00^0 1-10^0 0$ transition in CO_2 is studied experimentally in a passive cell in a resonator. It is shown that under continuous operation conditions the saturation is uniform despite the fact that $\Delta\nu_L < \Delta\nu_D$. The uniform nature of the saturation is due to diffusion in velocity space. The cross section for the decay of the CO_2 $00^0 1-10^0 0$ levels by collisions with He, Ne, N_2, or CO_2 were measured at 800°K, and the obtained values were respectively 6×10^{-19}, 2.8×10^{-18}, 1.2×10^{-17}, and 6.6×10^{-18} cm^2. Introduction of sufficiently large absorption leads to the appearance of spontaneous pulsations of the output power. Application of a field for a short time leaves the saturation uniform, since the number of collisions experienced by the excited molecules during a short time is not sufficient to smooth out the dip in the velocity distribution.

1. INTRODUCTION

THIS paper is devoted to an investigation of effects of saturation upon collisions on vibrational–rotational transitions of the $00^0 1-10^0 0$ band. When a strong monochromatic field interacts with a Doppler-broadened line, the "hole burning" effect in the amplification line contour takes place.[1] The band hole in the amplification line reflects the increase or a decrease of the density of the excited items in the velocity distribution of the atoms. In a standing-wave field, the formation of dips that are symmetrically disposed relative to the center of the line, leads to the well known dip in the power at the center of the line when scanning the frequency of the resonator. When the field is not very strong and the saturation parameter $\beta = G^2/\Gamma_1\Gamma_2$ is small (Γ_1 is the reciprocal of the sum of the lifetimes on the upper and lower levels and Γ_2 is the Lorentz-half width, $G = d_{12}D/\hbar$), the width of the dip corresponds to the Lorentz width due to the spontaneous saturation and the collisions. In the presence of collisions, the shape of the dip changes.[2]

Observation of the dependence of the generation power on the frequency yields valuable information both concerning the character of the interaction of the field with the atoms upon collision, and on the parameters characterizing the processes of collisions under the conditions when the model of the binary collisions is certainly satisfied, and the broadening and the line shift turn out to be much smaller than the Doppler width. We note that in the optical region of the spectrum the collisions primarily increase the width of the dip and decrease the saturation parameter $\beta = G^2/\Gamma_1\Gamma_2$. The situation may be entirely different upon consideration of the interaction of a strong field with excited molecules in the presence of collisions, for example in such a widely used system as the CO_2 laser. The large lifetimes of the vibrational levels increases to a considerable degree the influence of the collisions, which lead to a change in the velocity of the molecules. These collisions tend to bring the system into equilibrium, i.e., to produce a Maxwellian velocity distribution of the molecules and smooth out the dip in the velocity distribution of the molecules. If the time between the collisions is much smaller than the lifetime of the molecules, then a smoothing of the dip takes place in the distribution of the molecules by velocity, and as a consequence there is no dip in the generation power at the center of the line when the frequency is scanned, even if the Lorentz width $\Delta\nu_L$ is much smaller than the Doppler width $\Delta\nu_D$. In a CO_2 laser, the total pressure is sufficiently large, and the Lorentz width becomes comparable with the Doppler line width, and therefore the absence of a dip is not surprising. As will be shown below, there is no dip also under conditions when $\Delta\nu_L < \Delta\nu_D$.

This does not limit the specific influence of the collisions on the character of the saturation of the vibrational–rotational transitions of the $00^0 1-10^0 0$ band in CO_2. One can expect an anomalous dependence of the saturation parameter on the pressure in the region where the collisions increase the time of interaction of the molecules with the field, owing to the decrease of the diffusion velocity of the molecules towards the walls. There is no doubt that the role of the collisions on the molecule systems is much more varied than the effects of saturation observed heretofore in collisions in an He–Ne laser.

We note that an investigation of the character of the saturation in molecular systems is quite interesting also in connection with the latest independent proposals, made by Lisitsyn and Chebotaev[3] and by Letekhov,[4] of obtaining exceedingly high frequency stabilization in lasers by using the vibrational absorption spectra. In spite of the fact that the concrete methods proposed in [3] and [4] to realize high stability are different, both methods are based on the formation of dips in the absorption line in a standing-wave field.

2. EXPERIMENTAL PROCEDURE AND SETUP

The simplest method of investigating the influence of collisions on the interaction of a field with matter is to scan the generation frequency over the width of the line

in the single-mode regime. This method has very limited possibilities, since a change of the conditions in the active medium can be realized only within those limits in which generation exists. The use of a passive cell greatly broadens the possibilities of the investigations, and the cell can be both in the resonator and outside it. We used saturation of absorption in a cell placed inside a resonator. The relatively large gain margin has made it possible to vary greatly the magnitude of the absorption and the saturation parameter in the cell.

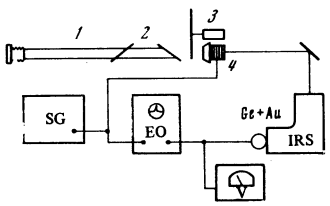

FIG. 1. Diagram of experimental setup: 1 – laser tube, 2 – cell, 3 – mechanical modulator, 4 – piezoceramic, SG – sawtooth voltage generator, V – selective voltmeter, EO – cathode ray oscilloscope, IRS – infrared spectrometer, Ge + Au – photoresistor.

The experimental setup (see Fig. 1) consisted of a laser with sufficiently rigid armature of ~120 cm length. The resonator mirrors and the diameter of the discharge tube (~12 mm) were chosen such that only axial modes were generated. The resonator could be tuned within a range $\lambda/2$ with the aid of a piezoceramic. The active discharge length was 60 cm, and the length of the absorbing cell was approximately 30 cm. The active part was filled with a CO_2–He–N_2 mixture at a total pressure ~10 mm Hg. The presence of strong competition between the rotational sublevels has led to the fact that at each instant of time the emission spectrum of our laser consisted of one mode at one vibrational–rotational transition. When the resonator wavelength was varied within a range $\lambda/2$ it was possible to observe successfully generation at 5–6 vibrational–rotational transitions belonging to the P or R branches of the $00^0 1$–$10^0 0$ band. All the measurements were made on the P(20) line.

The change of the absorption in the cell was effected by changing the temperature of the walls in a range 300–800°K and by varying the pressure of the CO_2.

3. SATURATION IN THE CONTINUOUS REGIME

In molecular systems, the lifetimes of the molecules in vibrational states are usually large compared with the time between the collisions. This can cause the character of the saturation of the transition to be determined by the diffusion in velocity space. In this case the spectrum of each individual molecule represents a sequence of wave trains having different frequencies within a range $k\bar{v}$, i.e., the spectrum of each individual molecule constitutes essentially the spectrum of the entire ensemble. Each molecule is then capable of interacting with the field within the lifetime in the excited state. In spite of the fact that at each instant of time the only molecules interacting effectively with the field are those whose frequency differs from the field frequency by not more than $\Delta\nu_L$, homogeneous saturation takes place.

In our experiments the Lorentz width of the transition in the cell was much smaller than the Doppler width. The CO_2 pressure in the cell ranged from 0.4 to 2 mm Hg at 800°K. According to the data of Gerry and Leonard,[5] the ratio of the Lorentz width to the Doppler width was 0.05–0.2. Estimates based on the data of Lisitsyn and Chebotaev[3] gave grounds for hoping to obtain a power peak at the center of the absorption line in the course of frequency scanning. Nonetheless, no power peak was observed in the continuous regime.

In [6,7] they considered saturation with allowance for diffusion in velocity space in the strong-collision model. The extension of the conclusions of Kol'chenko and Rautian[6] to the case when strong exchange in the rotational sublevel takes place leads to the following expression for the gain or absorption coefficient:

$$\alpha = \alpha_0 \left\{ \left(1 + \frac{G^2\tau_{rot}}{\Gamma + \nu}\right)^{1/2} + \left(\frac{\pi}{\ln 2}\right)^{1/2} + \frac{G^2}{k\bar{v}} [\tau_{2m} W(J) + \tau_{2n} W(J+1)] \right\}^{-1}, \quad (1)$$

where α_0 is the unsaturated gain at the center of the line. τ_{rot} is the lifetime at the rotational sublevel, and W(J) is the Boltzmann distribution of the excited molecules over the rotational sublevels.

The quantity τ_{2m} has the meaning of the lifetime after the first collisions and is determined by the expression

$$\tau_{2m} = \tilde{\nu}_m / (\Gamma_m + \nu_m)(\Gamma_m + \nu_m - \tilde{\nu}_m), \quad (2)$$

where Γ_m is the probability of decay of the vibrational state as a result of spontaneous transitions and diffusion to the walls, ν_m is the probability of collision with change of velocity and rotational quantum number J, and $\tilde{\nu}_m$ is the probability of the inverse collisions. If ν_m and $\tilde{\nu}_m$ differ insignificantly (the difference between these quantities yields the decay probability in the volume) and if each of them is much larger than Γ_m, it is possible to represent (2) in the form

$$\tau_{2m} = 1 / (\Gamma_m + \nu_m - \tilde{\nu}_m). \quad (3)$$

In this case the quantities τ_{2m} and τ_{2n} have the meaning of the lifetime at the corresponding vibrational levels.

Expression (1) is valid in the region of pressures where the broadening due to the collisions is smaller than the doublet broadening. In the case when the broadening due to the collision is comparable with or larger than the Doppler broadening, the saturation parameter should depend quadratically on the pressure of the proper gas itself.[8] A simple estimate shows that in an absorbing cell (at pressures exceeding 0.5 mm Hg) the saturation of the gain and of the absorption is described by the second term of the denominator.

Since the relative numbers of the particles at the upper and lower rotational sublevels differ insignificantly, expression (1) can be represented in the form

$$\alpha = \alpha_0 \left\{ 1 + \left(\frac{\pi}{\ln 2}\right)^{1/2} W(J) G^2 \frac{\tau_{2m} + \tau 2_n}{k\bar{v}} \right\}^{-1}. \quad (4)$$

For CN_2 at 300°K, the lifetime of the $00^0 1$ level is much larger than the lifetime of the $10^0 0$ level. The behavior of the saturation parameter will be determined in this case by the lifetime of the upper level. In our case, the concrete values of these times are unknown, and we shall therefore operate with their sum.

Figure 2 shows the dependence of the absolute power of the laser on the pressure of the admixture gas (He,

Ne, N_2) at CO_2 pressure in the cell 0.5 mm Hg. It is seen from Fig. 2 that the absorption changes more strongly when N_2 is added. It is clear that when the admixture pressure is very high, the magnitude of the absorption in the cell will tend to its unsaturated value. This level is shown by a horizontal line in Fig. 2.

If we now assume that the laser power depends linearly on the losses then, knowing the output power and the transmission of the mirror, it is easy to plot the

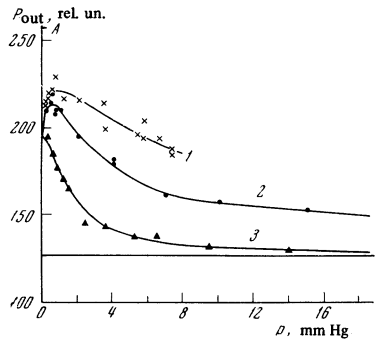

FIG. 2. Dependence of the absolute power on the pressure of the admixture gas in the cell: curve 1 – He, 2 – Ne, 3 – N_2; CO_2 pressure in cell ≈ 0.5 mm Hg; A – power in the absence of absorption.

dependence of the saturation parameter on the pressure of the admixture gas in the cell. The deviation of the dependence of the power and the losses from linearity is insignificant and lies within the limits of the measurement error. An estimate of the deviation from linearity was made both from the approximately known values of the gain and losses in the resonator, and directly from the change of the output power following a change of the CO_2 pressure in the cell in the presence of an appreciable amount of admixture gas. Figure 3 shows the plots of the saturation parameter of the $00^0 1 - 10^0 1$ transition of CO_2 in the cell on the pressure of He, Ne, and N_2. It is seen from the curve that the saturation parameter decreases greatly when 1 mm Hg of He or Ne is added We relate this phenomenon with the decreased rate of diffusion within the limits of the region occupied by the field in the cell.

It can be shown that the time variation of the particle concentration due to diffusion to the walls, in the

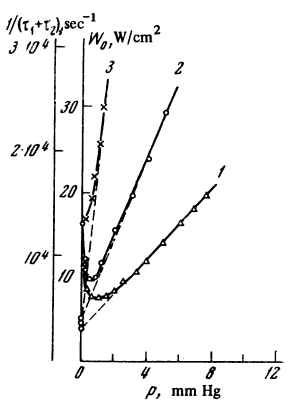

FIG. 3. Dependence of the saturation parameter W_0 and of the quantity $1/(\tau_1 + \tau_2)$ of the P(20) transition of the $00^0 1 - 10^0 0$ band of CO_2 on the pressure of the admixture gas: 1 – He, 2 – Ne, 3 – N_2.

region occupied by the field in the cell, is approximately given by

$$N = \sum_j J_1^2\left(\mu_j \frac{a_1}{a}\right)\exp(-D\lambda_j^2 t), \qquad (5)$$

Here J_1 is a Bessel function of first order, μ_j are the zeroes of the function J_0, a_1 is the radius of the region occupied by the field, a is the cell radius, D is the diffusion coefficient, and $\lambda_j = \mu_j/a$. A calculation based on (5) for the conditions of our experiment (a = 0.9 cm, a_1 = 0.3 cm, pressure $P(CO_2)$ = 0.5 mm Hg at T = 800°K, and D ≈ 200) yields a value ≈ 10^{-4} sec for the time that the molecules are located in the region occupied by the field. This value agrees well with the experimentally obtained one (see Fig. 3).

With further increase of pressure, the diffusion ceases to play any role whatever, and the overwhelming part of the damage occurs in the volume as a result of collisions. From the slope of the curves on the linear sections it is easy to obtain the values of the cross sections for the decay of the $00^0 1 - 10^0 0$ levels. The point of intersection of the lines with the ordinate axis indicates the frequency of the destructive collisions in pure CO_2.

In calculations of the total lifetimes and the cross sections for the decay of the $00^0 1$ and $10^0 0$ levels, the matrix element of the dipole moment was determined by us from the data of Gerry and Leonard.[5] We obtained the following values for the cross sections: $\sigma(CO_2\text{–He}) = 6\times10^{-19}$, $\sigma(CO_2\text{–Ne}) = 2.8\times10^{-18}$, $\sigma(CO_2\text{–}CO_2) = 6.6\times10^{-28}$, and $\sigma(CO_2\text{–}N_2) = 1.2\times10^{-17}$ cm^2. It should be noted that these values are much larger than those measured at 300°K.[9] The fact that the saturation parameter depends linearly on the pressure in an appreciable range of pressures confirms that in our case the homogeneous saturation is due to diffusion in velocity space.

4. SATURATION IN SHORT-DURATION INTERACTION WITH THE FIELD

It is clear that the smoothing of the dip in the amplification or absorption line contour occurred within a time that is larger than or at least equal to the time between collisions. In the case of vibrational–rotational transitions, the collisions lead not only to diffusion in velocity space, but also to an intense exchange between the rotational sublevels. This process will increase the time during which the dip in the center of the absorption or amplification line becomes smoothed out.

An extension of the conclusions of Kol'chenko and Rautian[6] to the case when rotational relaxation is present leads to the following expression for the condition for the smoothing of the dip in the absorption line:

$$W(J)\frac{\nu_m}{\Gamma + \nu_m - \tilde{\nu}_m}\Big/\frac{\Gamma+\nu}{k\bar{v}} > 1, \qquad (6)$$

where $\Gamma + \nu$ and $k\bar{v}$ are the Lorentz and Doppler half widths of the transition. The term $1/(\Gamma_m + \nu_m - \tilde{\nu}_m)$ represents the lifetime at the vibrational level. In the case of a pulsed field, the expression $1/(\Gamma_m + \nu_m - \tilde{\nu}_m)$ in (6) should be taken to mean the time of action of the field τ_f. Then (6) can be represented in the form

$$\tau_f > \frac{k\bar{v}}{W(J)\tilde{\nu}_m(\Gamma+\nu)}. \quad (7)$$

If we now assume that $\tilde{\nu}_m$ is determined mainly by the gas-kinetic collisions, we find that condition (7) is satisfied at $\tau \approx 20-30$ μsec at J = 20, T = 800°K, a pressure 1 mm Hg in the absorbing cell, and a cross section $\sigma = 5.7 \times 10^{-15}$ cm^2 [5] for the broadening of the vibrational–rotational transition.

The obtained time of smoothing of the dip in the absorption line is approximate, since an appreciable contribution can be made to $\tilde{\nu}_m$ by collisions with a change of J and by processes connected with the resonant exchange of excitation.

The character of the saturation in the pulsed regime was studied with the aid of the mechanical modulator, which was introduced into the resonator (modulation frequency ~2 kHz, time during which the generation was turned on ~ 700 μsec). The generation power pulse had a complicated form and consisted of two spikes, following which the generation power reached smoothly a value corresponding to the continuous regime. The first spike, of duration ~10 μsec, is connected with the resonator Q-switching time. We attribute the second maximum to the relaxation time of the lower working level, since the time of its appearance (20–30 μsec after the resonator Q-switching) coincides with the lifetime of the 10^00 level. However, a complete interpretation of such a complex behavior of the output power in the case of resonator Q-switching calls for additional investigations. Nonetheless, when the resonator was scanned in frequency, the envelope of the amplitudes of both the first and second maxima described a power peak at the center of the absorption line.

Figure 4 shows the envelope of the amplitudes of the second generation pulses obtained by frequency scanning in the absence of absorption (curve 1) and in the presence of absorption (curve 2). The level corresponding to the continuous regime is shown by curve 3. The power peak was observed 20–30 μsec after the generation was turned on. This value is in good agreement with that calculated above.

Introduction of sufficient absorption has led to the transition of generation into the continuous regime. In this case the laser generated individual pulses of duration ~10 μsec, the amplitude of which was approximately one order of magnitude higher than the level of the continued generation. In the present investigation we call attention to the fact that in frequency scanning the pulse amplitude described a power peak at the center of the absorption line. Details of the spontaneous-pulsation regime will be reported separately.

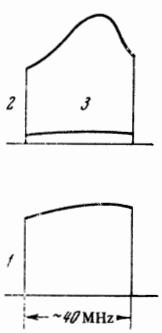

FIG. 4. Power peak at the center of the absorption line upon modulation of the field in the resonator.

The width of the envelope at half the height depends on on the pressure in accordance with the expression

$$\Delta\nu_f = \Delta\nu_L\sqrt{1+G^2/\Gamma_1\Gamma_2}, \quad (8)$$

where Γ_1 is the reciprocal of the time of interaction with the field. Since the duration of the generation pulses is practically independent of the pressure in the range where the power peak was observed in the center of the absorption line, expression (8) can be represented in the form

$$\Delta\nu_f = Ap\sqrt{1+B|E|^2/p}, \quad (9)$$

where A and B are constant coefficients and p is the pressure. The coefficient A can be readily obtained from the data of [5]. In our case it amounts to ~7 MHz/mm. Knowing $\Delta\nu_f$ for any pressure, we obtain the coefficient B.

Figure 5 shows the dependence of the Lorentz width on the pressure and the dependence of the width of the dip $\Delta\nu_f$, constructed in accordance with (9) and normalized to the point C (curve 2). The experimental values of the power-peak width are designated by the point.

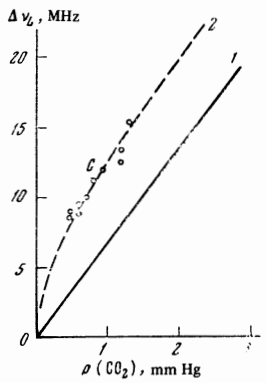

FIG. 5. Dependence of the width of the generation power peak in the center of the absorption line on the pressure: 1 – calculated Lorentz width, 2 – width of generation power peak at the center of the absorption line.

It is seen from Fig. 5 that a power peak is observed at the center of the absorption line in the spontaneous-pulsation regime, and the behavior of its width as a function of the field has the same character as in the case of ordinary inhomogeneous saturation.

5. CONCLUSION

The investigated saturation mechanism is apparently characteristic of molecular lasers operating on vibrational–rotational transitions. The fact that the lifetime in the vibrational state greatly exceeds the time between collisions leads to a homogeneous character of the saturation, in spite of the fact that the Doppler width of the transition greatly exceeds the broadening due to the collisions. A purely inhomogeneous saturation, which makes it possible to realize stabilization with a high degree of accuracy, can be obtained only at very low pressures of the working medium.

Simultaneously with reducing the pressure, it is pos-

sible to reduce the diameter of the laser beam in the resonator. Then the time of interaction of the molecules with the field will be determined by the time that the molecule spins inside the beam. Estimates show that this time can be reduced to a value on the order of 10^{-5}–10^{-6} at pressures 10^{-1}–10^{-2} mm Hg, making it possible to hope to obtain inhomogeneous saturation in the continuous regime.

In conclusion, the authors consider it their pleasant duty to thank S. G. Rautian and G. E. Notkin for valuable discussions. G. I. Shershneva took part in the work.

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