Investigation of Narrow Resonances in Absorption Saturation of SF_6 Molecules by CO_2 Laser Emission

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Narrow resonances within the Doppler line of ν_3 band vibrational-rotational transitions of the SF_6 molecule are investigated for absorption saturation by CO_2 laser emission. The characteristics of a nonlinear laser spectrometer based on the absorption cross modulation method are investigated and the structure of narrow resonances at low SF_6 pressure is resolved. An experiment is performed to observe narrow molecular resonances in saturation of absorption of spatially separated opposed light waves.

1. INTRODUCTION

AN earlier paper[1] (henceforth cited as I) reported on a nonlinear spectroscopy method based on absorption saturation within a Doppler line by a quasi-traveling light wave (see also[2]).

The SF_6 molecule was selected as the object of investigation, while a CO_2 laser emitting several lines of the P branch in the 10.6 μ region served as the source of monochromatic coherent radiation with a frequency tunable within a small region. The quasi-traveling wave method proposed in[3] has an advantage over the usual standing-wave method[4-6] in that it furnishes large-amplitude resonances especially when optically dense absorption cells are used. The method is sensitive enough for work at pressures of 10^{-2} Torr with a resolution of 0.6 MHz (see I).

The present paper reports on the results of further investigation of the narrow resonances discovered earlier in the Doppler line of rotational-vibrational transitions of the ν_3 band of the SF_6 molecule by the method of absorption saturation by CO_2 laser emission. In contrast to I, we now use an improved method of absorption saturation, which allows us to observe narrow nonlinear resonances within the Doppler line with a much higher sensitivity. Our method can be called the method of nonlinear cross modulation since it is based on traveling wave transmission modulation achieved in interaction with molecules whose absorption is periodically varied by an opposed wave with modulated intensity. This method was previously used with success to record narrow molecular resonances reported in[7,8].

Section 2 of this paper deals with the characteristics of a nonlinear laser spectrometer using the absorption cross modulation method. Section 3 reports on the use of this method to investigate the structure of narrow resonances at low pressures of SF_6. Finally Sec. 4 describes an experiment to observe narrow molecular resonances in absorption saturation by spatially separated opposed light waves, made possible for the first time by the high sensitivity of the new method.

2. NONLINEAR SPECTROSCOPY BY THE ABSORPTION CROSS MODULATION METHOD

Figure 1 shows the experimental setup. The tunable CO_2 laser with a water-cooled sealed tube 60 m long and 10 mm in diameter operated in a single-mode regime at the wavelength of 10.6 μ. The laser was pumped by a dc discharge. The resonator (100 cm long) was formed by two plane mirrors with reflection coefficients of 99 and 78%. The frequency of the CO_2 laser was tunable within the limits of a Doppler line by scanning the output mirror mounted on a piezoceramic tube. The emission output power reached 1 W. Special measures were taken to eliminate mechanical vibration and to provide acoustic and thermal decoupling of the resonator from the ambient medium. The relatively high time stability of laser frequency allowed us to use long frequency scan times (tens of seconds) using synchronous detection at the intensity modulation frequency of one of the beams.

Beam splitter R_1 and a mirror system transformed the CO_2 laser emission into two opposed beams incident on the external SF_6 cell. The length of the cell was L_{abs} = 120 cm and its diameter was 36 mm. The intensity ($I_1(\Omega)$) of one of the two opposed beams saturating the absorption was controlled with attenuator A_1 representing a short cell filled with gaseous sulfur hexafluoride at high pressure (~1 Torr); it was also modulated at a frequency Ω = 900 Hz by mechanical shutter S. The necessary attenuation of the second beam intensity (I_2) was accomplished by attenuator A_2 also filled with SF_6 at high pressure. To eliminate back scattering, beam I_2 (test beam) was allowed to enter the SF_6 cell at a small angle ($\sim 3 \times 10^{-3}$) to the opposed (forward) beam $I_1(\Omega)$. After reflection from semi-transparent plate R_4, the I_2 beam was intercepted by a Ge : Au detector. The detector signal was amplified by a narrow-band amplifier at the frequency Ω and after synchronous detection was relayed to an automatic recorder.

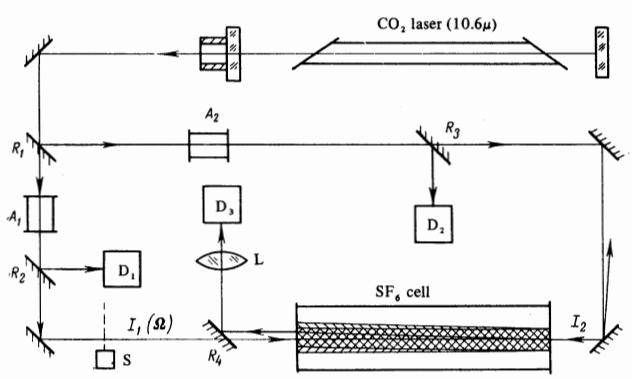

FIG. 1. Experimental setup for the investigation of narrow molecular resonances. R_1, R_2, R_3, R_4—semitransparent mirrors; A_1, A_2—attenuators; S—mechanical shutter; L = lens; D_1, D_2, D_3—Ge: Au photocells.

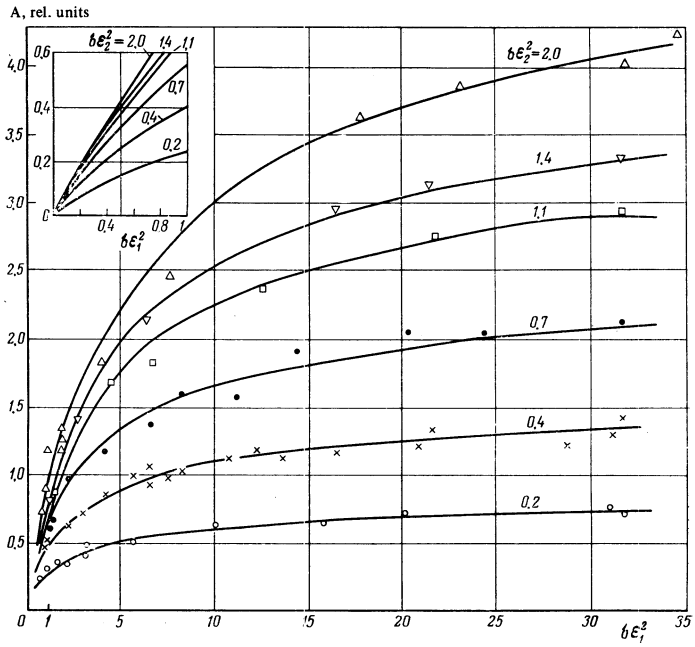

FIG. 2. Transmission peak amplitude as a function of degree of absorption saturation by forward beam at cell input for various values of test beam intensity. Points denote experimental data, lines are theoretical plots. Insert shows theoretical dependence of resonance amplitude in weak field of forward beam.

The P(18) line of the CO_2 laser emission was selected for the study of narrow resonance parameters in a low pressure SF_6 cell. The SF_6 pressure in the cell was 5 mTorr. At this pressure the cell can be considered optically thin ($d = \kappa_0 L_{absp} = 0.3$) enough to neglect the change in the dip shape due to absorption in the cell.

Figure 2 shows transmission peak amplitude A as a function of degree of absorption saturation by the forward beam with amplitude ϵ_1 at the input for various values of intensity ϵ_2^2 of the test beam.

The results of this experiment may be compared with theory within the framework of a model that neglects polarization effects; this is possible in principle when absorption saturation is strong (see[9] for example). In this case the transmission peak amplitude ϵ_2^2 of the test beam is determined by the difference between absorption in the center of the line $|\nu - \omega_0| \ll \Delta\nu$ ($\Delta\nu$ is the width of the resonance and ω_0 is the center of the Doppler line) and outside the center $\Delta\nu_{Dop} \gg |\nu - \omega_0| \gg \Delta\nu$ ($\Delta\nu_{Dop}$ is the Doppler width). The coefficient of absorption of a wave with amplitude ϵ_2 outside the center of the Doppler line, in the approximation considered here, equals[10,11]

$$\varkappa_{\text{off resonance}} = \varkappa_0 / (1 + b\varepsilon_2^2)^{1/2}, \tag{1}$$

where κ_0 is the initial absorption in the line center and b is the absorption saturation parameter. We assume that absorption saturation at resonance is determined simply by the total intensity of the two beams:

$$\varkappa_{\text{in resonance}} = \varkappa_0 / [1 + b(\varepsilon_1^2 + \varepsilon_2^2)]^{1/2}. \tag{2}$$

The transmission peak amplitude of the test beam equal to the modulated portion of intensity then is

$$A = \varkappa_0[(1 + b\varepsilon_2^2)^{-1/2} - (1 + b(\varepsilon_1^2 + \varepsilon_2^2))^{-1/2}]\varepsilon_2^2. \tag{3}$$

If the absorption saturation by the test beam is small ($b\epsilon_2^2 \ll 1$), i.e., in the case of a quasi-traveling wave[3]

$$A = \varkappa_0[1 - (1 + b\varepsilon_1^2)^{-1/2}]\varepsilon_2^2. \tag{4}$$

Thus the magnitude of the dip is proportional to the intensity of the test beam and tends towards the limiting value of $A_{max} = \kappa_0 \epsilon_2^2$ as the absorption saturation by the strong forward beam is increased.

To compare with experiment it is convenient to transform (3) into

$$A = \frac{\varkappa_0 x_2}{b(1+x_2)^{1/2}}\left(1 - \frac{1}{[1 + x_1/(1+x_2)]^{1/2}}\right), \tag{5}$$

where $x_i = b\epsilon_i^2$ (i = 1, 2). In the approximation of a weak field of the forward beam ($x_1 \ll 1$), or if it is small compared to the test beam field ($x_1 \ll x_2$), this expression reduces to the following:

$$A = \frac{\varkappa_0}{2b}\frac{x_1 x_2}{(1+x_1)^{3/2}}. \tag{6}$$

Testing this relationship for an extremum with respect to x_2 we can readily find that it has a maximum for $x_2 = 2$. This means that in the indicated range of values of the forward beam field the transmission peak amplitude of the opposed beam has a maximum value for a saturation parameter of $b\epsilon_2^2 = 2$.

Figure 2 shows theoretical values of the resonance amplitude as a function of absorption saturation by the forward beam (solid lines) computed for the same values of opposed beam intensity as those of the experimental plot. The insert in Fig. 2 shows the theoretical dependence of the transmission peak amplitude for the same values of the test beam intensity in the region of small parameters of saturation by the forward beam. For all six values of $b\epsilon_2^2 = 0.2$, 0.4, 0.7, 1.1, 1.4, and 2, the theoretical curves coincide with the experimental data within the experimental error determined by system noise. The minimum value of the relative contrast of the transmission peak recorded in the experiment was 0.25%.

We also measured the width $2\Delta\nu$ of the transmission peak at half-altitude as a function of the absorption saturation $b\epsilon_1^2$ by the forward beam at the cell input, for various values of opposing beam intensity ϵ_2^2. The curves are represented within experimental error (10%) by the expression

$$\Delta\nu = {}^1/_2\Delta\nu_0[(1 + b\varepsilon_1^2)^{1/2} + (1 + b\varepsilon_2^2)^{1/2}], \quad (7)$$

which follows from the simple model under consideration ($\Delta\nu_0$ is the homogeneous half-width of the line at half-altitude). In fact, the width of the resonance at the center is determined in our approximation by the convolution of a Doppler contour saturated by a wave ϵ_1, in which a "hole" $2\Delta\nu_0(1 + b\epsilon_1^2)^{1/2}$ wide is burned at the frequency ν, with a Lorentz contour at the mirror-symmetry frequency $2\omega_0 - \nu$, defining a frequency region $2\Delta\nu_0(1 + b\epsilon_2^2)^{1/2}$ wide and interacting with the opposing beam ϵ_2. The exponent $1/2$ is due to the fact that the distance between the frequencies $2\omega_0 - \nu$ and ν is twice the deviation of the beam frequency from the center of the Doppler contour. The results are in good agreement with experimental data given in I. The width of the narrowest transmission resonance observed in the experiment amounted to $2\Delta\nu = 0.45 \pm 0.05$ MHz at half-altitude at an SF_6 pressure 5 mTorr.

The results of the above experiment are in good agreement with a simple approximation in which polarization effects are neglected, although according to theory developed in[9] their contribution under our conditions should have been significant. What is the cause of this discrepancy? We think that a description of an interaction between a field and rotational-vibrational transitions of the molecule must take into account the degeneracy of the transition. For example, in the case of the SF_6 molecule the employed CO_2 laser lines coincide with transitions that have high values of the angular momentum J (according to[12] J = 50—80, although the type of transition is not identified). Because of level degeneracy, the field interacts with a large number of ensembles of two-level particles with different transition dipole moments. In such a case the approximation of a two-level nondegenerate system is a priori invalid. For example, for a strong degeneracy ($J \gg 1$) the change of the population difference for levels ρ_1 and ρ_2 is described by[13]

$$\frac{\rho_1 - \rho_2}{\rho_1^0 - \rho_2^0} = \frac{\pi}{2} E_1\left(\frac{\mu\varepsilon}{\hbar} t\right),$$

$$E_1(x) = \frac{1}{\pi}\int_0^{\pi} \sin(\theta - x\sin\theta)\, d\theta, \quad (8)$$

where μ is some average dipole transition momentum and $E_1(x)$ is a Weber function. This means that the dependence is an oscillating, nonperiodic, and above all, damped function. It differs significantly from the periodic undamped function $\cos(\mu\epsilon\tau/\hbar)$ for a nondegenerate two-level system. Strong degeneracy seems to be able to smooth the population and polarization oscillations in the molecular transition, and to validate a velocity approximation that takes only noncoherent saturation of the level-population difference into account.

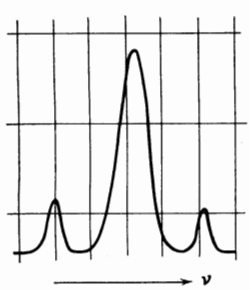

FIG. 3. Structure of triple resonance observed in the contour of Doppler line P(16) when absorption in SF_6 is saturated by CO_2 laser emission. SF_6 pressure is 5 mTorr. One division corresponds to 0.5 MHz.

3. STRUCTURE OF TRIPLE RESONANCE OBSERVED IN THE P(16) LINE

The resolution achieved in the experiment permitted us to return to the structure of the anomalously broad peak in the contour of the Doppler P(16) line observed when absorption in SF_6 gas is saturated with CO_2 laser emission. Figure 3 shows the peak structure obtained in the quasi-traveling wave regime at a scanning time of 20 sec. We observe that the resonances are not equidistant and have different intensities. The side components are removed from the central peak by 1.0 and 1.25 MHz respectively (the experimental error was 10%), while the intensity and half-altitude width of the central component are approximately 4—5 times larger than those of the side satellites.

The unequal spacing of the resonances cannot be attributed to the effect of "pulling" resonance frequencies toward the center of the P(16) emission line of the CO_2 laser. In fact, if ν_i^0 and $\Delta\nu_r$ are the actual center frequency and resonance width, and ν_l and $\Delta\nu_l$ are the center frequency and amplification linewidth of the laser P(16) line, the observed frequency ν_i^{obs} of the narrow resonance is shifted relative to the actual frequency ν_i^0 by the magnitude

$$\frac{\nu_i^{abs} - \nu_i^0}{\Delta\nu_r} \simeq \frac{\nu_l - \nu_i^0}{\Delta\nu_l}\frac{\Delta\nu_r}{\Delta\nu_l}. \quad (9)$$

In this case $\nu_l - \nu_i^0 = 9$ MHz, $\Delta\nu_r = 0.5$ MHz, $\Delta\nu_l = 60$ MHz, and the amount of "pulling" is 10^{-3} MHz, i.e., much less than the experimentally observed deviation from equal spacing. It was proposed in[5] that three equidistant resonances can be produced by the interaction of a standing wave with two close transitions having a common level[15]. In this case the two outside resonances occur when the wave frequency coincides with the center of each transition, while the center resonance is due to the coincidence of two holes belonging to different transitions. Experiments with SF_6 performed so far showed that three components in SF_6 observed with the aid of the P(16) line of the CO_2 laser were equidistant[1,6,14]. The present experiment showed that they are not equidistant. This permits us to reject the above mechanism and to postulate that the resonances occur in three independent close transitions. An analogous conclusion was based in[14] on experiments with absorption saturation in a two-frequency light field.

4. NARROW RESONANCES IN SPATIALLY SEPARATED BEAMS

Of particular interest is the observation of narrow molecular resonances within the Doppler line when absorption saturation occurs in the field of two parallel spatially separated light beams. The narrow resonance is due to nonlinear interaction of the fields with molecules intersecting both light beams without changing their velocity direction.

Absorption saturation by a coherent traveling wave results in a Doppler-broadened transition in a "hole" burned in the line contour. The hole is due to absorption saturation of molecules in resonance with the field,

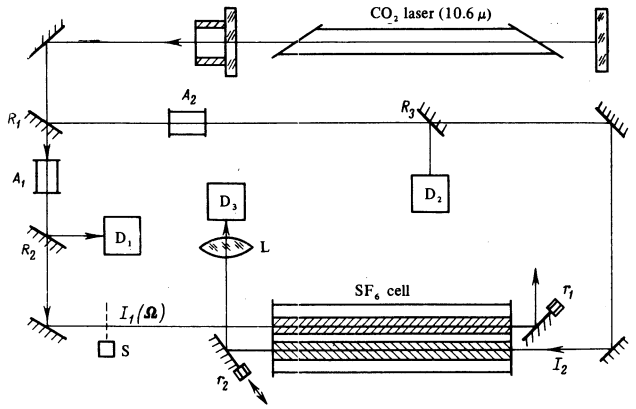

FIG. 4. Setup for observing molecular resonances when absorption is saturated in separated light beams. R_1, R_2, R_3–semitransparent mirrors; A_1, A_2–attenuators; S–mechanical shutter; L–lens; r_1, r_2–totally reflecting mirrors; D_1, D_2, D_3–Ge: Au detectors.

i.e., molecules characterized by a definite projection of velocity **v** on the direction of wave propagation **k**:

$$|\omega_0 - \nu - \mathbf{kv}| \lesssim \Gamma \ll \Delta\omega_{Dop} \qquad (10)$$

where ω_0, Γ and $\Delta\omega_{Dop}$ are center frequency, homogeneous width, and total width of the Doppler line, respectively. Given a long lifetime, the excited molecules can travel in space around the beam. Collisions between the molecules change their velocity direction and the "hole" in their velocity distribution is very quickly dissipated. Even weak collisions scattering the molecules at small angles on the order of $\Gamma/\Delta\omega_{Dop}$ play a significant role in the dissipation of the hole. However, if the gas pressure is low enough and the mean free path of the molecules is comparable with the beam diameter, the "hole" moves in space over a perceptible distance. Of course, to observe this effect the pressure must be quite low, since the small-angle scattering cross section of the molecules is approximately two orders higher than the gaskinetic cross section.

We can observe the shift of the "hole" in space with the aid of a coherent test beam of the same frequency but propagating in the opposite direction. Such a beam interacts with the molecules whose velocity projection on the direction of a strong beam satisfies another condition:

$$|\omega_0 - \nu + \mathbf{kv}| \lesssim \Gamma. \qquad (11)$$

If $|\omega_0 - \nu| \gtrsim \Gamma$, then there are no molecules that satisfy resonance conditions (10) and (11) simultaneously, except for the frequency region $|\omega_0 - \nu| \lesssim \Gamma$. Therefore only when passing through the Doppler line center can the test beam interact with molecules previously saturated by a strong opposing beam and crossing from one beam region to the other without collisions. A resonance drop in test beam absorption occurs at that time, i.e., a narrow dip is formed in its absorption.

The "hole" shift effect was predicted in the early papers suggesting the presence of narrow resonances in the absorption saturation of long-lived molecular transitions[16]. A detailed theory of absorption saturation in spatially separated fields is presented in[17].

The experimental setup for the observation of this effect is shown in Fig. 4. It is similar in principle to that shown in Fig. 1. A CO_2 laser (10.6 μ) operated at the P(16) line frequency where sulfur hexafluoride has maximum absorption (κ_0 = 1.3 cm^{-1}-Torr). This allowed us to use low SF_6 pressures at which the mean free path of the molecules is maximal.

A system of beam splitters and mirrors directed the laser radiation, in the form of two opposing traveling waves, to the external SF_6 cell. The length of the absorption cell was L_{abs} = 120 cm, and its diameter was 36 mm. One of the two opposing beams, $I_1(\Omega)$, saturated the absorption; its intensity was controlled by attenuator A_1 and modulated by mechanical shutter S at a frequency Ω = 900 Hz. The intensity of the second beam I_2 was reduced by attenuator A_2. Both attenuators were short cells filled with SF_6 under high pressure. A weak test beam passed through the cell and was reflected by rectangular plate r_2 onto a Ge : Au photocell. The detector signal was amplified by a narrow-band amplifier operating at the frequency Ω and after synchronous detection was fed to the automatic recorder. As in the case of the intersecting beams, the recording of the portion of test beam intensity that was modulated at the frequency Ω permitted us to isolate only that portion of the absorption coefficient function which was due to the saturating beam modulated at the frequency Ω.

The illuminated spot diameter at cell input was 7 mm for the saturating beam and 9 mm for the test beam. The rectangular plates r_1 and r_2 shaped the fields so as to have the cross sections of both beams in the form of two segments (Fig. 5a) limited on one side at the same power level ($I \approx 0.8 I_{max}$). Such a field configuration greatly enhanced the effectiveness of field interaction since it increased the number of excited molecules capable of crossing the field of the test beam. The distance separating the beams was selected taking the directivity pattern ($\theta_{0.5} \approx 1.5 \times 10^{-3}$) and wave diffraction into account. Initially it was 4 mm in the center section of the cell. The distance between the separated light beams was varied by parallel displacement of plate r_2 across the strong beam.

The main results of the experiment are given in Fig. 5b, which shows the amplitude of molecular resonances obtained in the separated light fields as a function of the parameter d determining the effective distance between the beams (Fig. 5a). In the range of the above pressures (35–4 mTorr) the peaks were due to the unavoidable weak diffractional overlapping of the fields. The magnitude of the saturation parameter of the SF_6 molecules was rigidly maintained against varying pressure in the course of the experiment; for this purpose the dependence of absorption saturation on the SF_6 pressure was plotted in advance. Since the signal at a fixed saturation parameter is proportional to the initial absorption coefficient or to the cell pressure, the resonance amplitudes at the maximum were normalized to unity.

The figure shows that the dependence of the resonance amplitude on the beam separation parameter d shifts towards the larger values of d as the pressure decreases. This shift amounts approximately to 0.8 mm when the SF_6 pressure in the absorption cell changes from 35 to 4.5 mTorr; it is inversely proportional to the pressure, a fact that can be attributed only to the shift of the "dip" from one light wave to the other by a distance equal to the mean free path of the molecules.

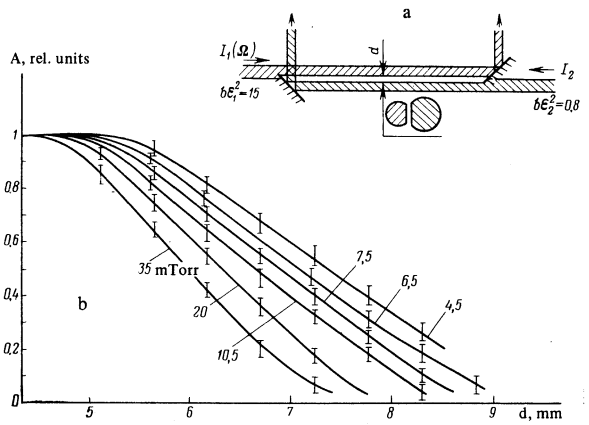

FIG. 5. Normalized resonance amplitude as a function of beam separation parameter for various pressures of SF_6.

Variation of the SF_6 saturation parameter in the cell with separated light fields could not have an effect, because the curves were not affected when this parameter was decreased with calibrated attenuators at the same pressure. We also performed a control experiment to eliminate the effect of variation in optical density $D = \kappa_0 L_{abs} p_{SF_6}$ due to pressure changes in the SF_6 cell. For this purpose the absorption was saturated at a constant value of the saturation parameter by the CO_2 laser emission at the P(18) line for which $\kappa_0 = 0.5\ cm^{-1} \cdot Torr^{-1}$. It is thus 2.6 times smaller for the same pressure. At the same time, the position and shape of the curves did not change when the beams were separated.

The magnitude of the "dip" shift was determined from the shift of the curves as a result of a reduction of pressure in the cell. The "dip" shift turned out to be $p\Lambda_{test} = (4.5 \pm 1.0) \times 10^{-4}$ cm · Torr, i.e., it coincided with the mean free path obtained from the data of[1,6].

5. CONCLUSION

Additional improvement in the resolving power of nonlinear spectroscopy methods is possible if the molecular gas pressure is significantly reduced (to 10^{-4} Torr). In this case the mean free path of the molecules increases to several centimeters, which in turn permits a more detailed investigation of transit effects in separated (opposed and having the same direction) traveling waves. Since the saturation power decreases in proportion to the pressure, the emission intensity in such an experiment is low enough to render the internal noise of 10 μ receivers the principal contribution. In nonlinear spectrometers based on CO_2 lasers this difficulty can be avoided by using quantum amplifiers at the outputs of low-pressure gas cells. In principle a gain of the order of 10^3—10^2 can be obtained in several passes in the CO_2 amplifier at low temperature[18]. This approach promises further extension of the possibilities of nonlinear spectroscopy to investigate the structure within the Doppler lines of molecules. For example, at low gas pressures the resolving power of nonlinear laser spectroscopy of molecules can be increased up to the natural width[19], thus making spectroscopy within the homogeneous width possible.

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