INVESTIGATION OF NARROW RESONANCES WITHIN THE DOPPLER LINE OF THE ROTATIONAL-VIBRATIONAL TRANSITIONS OF THE SF_6 MOLECULE DURING ABSORPTION SATURATION

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We consider theoretically the properties of a nonlinear spectroscope with a quasitraveling wave. Results are presented of an experimental investigation of the narrow resonances within the Doppler line of rotational-vibrational transitions of the SF_6 molecule upon saturation of the absorption by the quasitraveling wave of the CO_2 laser. The attained resolution of the spectroscope makes it possible to investigate the hyperfine structure of the molecule. The method is particularly convenient for the stabilization of the frequency of the CO_2 laser.

1. INTRODUCTION

THE widths of the spectral lines of a gas are determined usually by the Doppler broadening, whereas the emission (absorption) line width of an individual atom or molecule, i.e., the homogeneous width, may be appreciably smaller. The development of gas lasers[1] has uncovered the possibility of investigating the structure within the Doppler line, at a resolution determined in practice by the homogeneous width. The gist of the method is as follows. Upon saturation of the gain of a Doppler spectral line by a strong traveling light wave of frequency ν, a "hole" is produced inside the line. The width of this hole is equal to the homogeneous width, apart from a certain broadening by the field[2]. If we now measure the contour of the saturated Doppler line with the aid of a weak trial wave parallel to the strong wave, by scanning its frequency, it is possible to find the width of the hole, its depth, etc.

A more interesting effect arises when the Doppler line is saturated by strong standing light wave of frequency ν. In this case each of the opposing traveling waves burns out its own "hole" in positions that are symmetrical with respect to the center of the line, as shown in Fig. 1a. The depths and widths of the "holes" increase with increasing field amplitude. When the frequency of the standing wave is scanned along the Doppler contour, the saturated gain will have a dip in the center of the Doppler line ($\nu = \omega_0$), for in this case the particles interact with both waves and the degree of saturation increases[3]. The width of the dip, apart from the broadening by the field, is equal to the homogeneous width. The possibilities of using the Lamb dip for spectroscopic purposes were discussed in[4,5].

Particularly interesting possibilities of this method were uncovered after it was proposed to use nonlinearly absorbing cells with low-pressure gas inside the laser[6-8]. In this case, when the generation frequency passes through the center of the absorption line, owing to the saturation effect, a power peak appears, the width of which, especially in the case of absorption by the rotational-vibrational transitions of the molecules[6,7], can be exceedingly small. The possibilities of such a method were demonstrated in[9], where a power peak with a width on the order of 0.3 MHz was obtained upon saturation of the absorption of the rotational-vibrational transition of CH_4 by He-Ne laser radiation at $\lambda = 3.39\ \mu$. In[10] it was proposed to use an external nonlinearly-absorbing cell, thereby illuminating the influence of the nonlinear absorption on the operation of the laser.

A shortcoming of the method of saturation of absorption by a standing wave is the relatively small magnitude of the dip. In the optimal case, the depth of the dip is approximately 20–30% and decreases with the decreasing or increasing field amplitude. When the field power increases, the depth of the dip decreases because the decrease of the saturated absorption is negligibly small in the case of an overlap of very deep "holes" (Fig. 1a). This shortcoming disappears in the case of saturation of the absorption by a quasitraveling wave[11]. In this method, one traveling wave of frequency ν (forward) is strong and saturates the absorption, while the backward wave is weak and interacts linearly with the particles within the limits of the homogeneous width at the mirror-symmetry frequency $2\omega_0 - \nu$ (Fig. 1b). In this case the transmission of the backward weak wave increases strongly on passing through the center of the Doppler line, when it interacts with particles saturated by the forward strong wave. The amplitude of the transition peak can be very

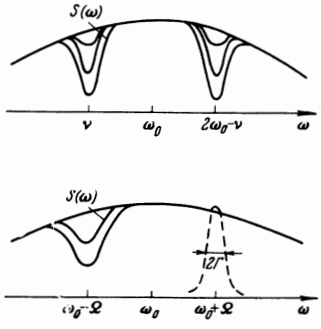

FIG. 1. Shape of absorption line at different degrees of saturation of the absorption: a–saturation by a standing wave (top) and b–saturation by a quasitraveling wave (bottom).

large, especially when optically dense absorbing cells are used. The possibilities of this method were demonstrated in[12], when narrow resonances were observed upon saturation of the absorption of rotational-vibrational transitions of the SF_6 molecule by CO_2-laser radiation.

In this article we report the results of an investigation of the structure of the rotational-vibrational lines of the ν_3 band of the SF_6 molecule by using radiation from a CO_2 laser to saturate the absorption of a quasitraveling wave in an external cell. In Sec. 2 we consider theoretically the properties of a nonlinear spectrometer with a quasitraveling wave, and in Sec. 3 we describe the experimental setup and the results of measuring the amplitudes and the widths of narrow resonances with the aid of the P(18) line of the CO_2 laser emission. The structure of narrow resonances observed with the aid of several P lines of CO_2 laser emission at $\lambda = 10.6\ \mu$ is described.

2. NONLINEAR SPECTROSCOPY WITH THE AID OF A QUASITRAVELING WAVE

A strong traveling wave of frequency ν and amplitude E_+ saturates the Doppler line in the following manner:

$$S(\omega)=\varkappa_0(\omega)\left[1+bE_+^2\frac{\Gamma^2}{(\omega-\nu)^2+\Gamma^2}\right]^{-1}, \tag{1}$$

where $\kappa_0(\omega)$ is the shape of the Doppler absorption line for a weak field, b is the absorption saturation parameter, 2Γ is the homogeneous width of the transition. The quantity bE_+^2 determines the degree of saturation of the absorption. The absorption of the backward traveling wave is determined by the interaction with the molecules within the limits of the homogeneous width at the frequency $\omega_0 + \Omega$, where $\Omega = (\omega_0 - \nu)$ is the frequency deviation of the wave relative to the center ω_0 of the Doppler line. If the saturation of absorption by the backward wave is small.

$$bE_-^2 \ll 1, \tag{2}$$

then the saturation coefficient of the backward wave is determined by the expression

$$\varkappa_-(\Omega)=\frac{1}{\pi}\int_{-\infty}^{\infty}S(\omega)\frac{\Gamma}{\Gamma^2+(\omega-\omega_0-\Omega)^2}d\omega. \tag{3}$$

Assuming smallness of the homogeneous width 2Γ compared with the width $\Delta\omega_{Dop}$ of the Doppler contour $\kappa_0(\omega)$, expression (3) reduces to

$$\varkappa_-(\Omega)=\varkappa_0(\Omega)\frac{1}{\pi}\int_{-\infty}^{\infty}\left[1+bE_+^2\frac{\Gamma^2}{(\omega-\nu)^2+\Gamma^2}\right]^{-1}+\frac{\Gamma}{\Gamma^2+(\omega-\omega_0-\Omega)^2}d\omega. \tag{4}$$

After cumbersome calculations we get

$$\varkappa_-(\Omega)=\varkappa_0(\Omega)\left\{\left(\frac{2\Omega}{\Gamma}\right)^2\left[\left(\frac{2\Omega}{\Gamma}\right)^2+bE_+^2+4\right]+\frac{bE_+^2}{\sqrt{1+bE_+^2}}\left[bE_+^2-\left(\frac{2\Omega}{\Gamma}\right)^2\right]\right\}\left\{\left[bE_+^2-\left(\frac{2\Omega}{\Gamma}\right)^2\right]^2+4\left(\frac{2\Omega}{\Gamma}\right)^2(1+bE_+^2)\right\}^{-1}. \tag{5}$$

In the approximation in which the field of the forward wave is assumed to be weak ($bE_+^2 \ll 1$), this expression reduces to the ordinary expression for the Lamb dip in first approximation in the field:

$$\varkappa_-(\Omega)\simeq\varkappa_0(\Omega)\left(1-\frac{1}{2}E_+^2\frac{\Gamma^2}{\Gamma^2+\Omega^2}\right). \tag{6}$$

The transmission of the backward wave by the absorbing cell $\eta - (\Omega)$ is determined by the general expression

$$\eta_-(\Omega)=\exp\left[-\int_0^L\varkappa_-(\Omega,x)dx\right], \tag{7}$$

where L is the length of the cell; the dependence of $\kappa_-(\Omega)$ on the coordinate is the result of the attenuation of the forward wave after passing through the cell. In the case of an optically thin cell ($\kappa_0 L \ll 1$) we have

$$\eta_-(\Omega)=1-\varkappa_-(\Omega)L, \tag{8}$$

i.e., the shape of the dip is not distorted by the dependence on the absorption in the cell. In the case of an optically dense cell this is not the case, and it is necessary to take into consideration the attenuation of the forward wave.

The attenuation of the forward wave is described by the equation

$$\frac{dE_+^2}{dx}=-\frac{\varkappa_0}{\sqrt{1+bE_+^2}}E_+^2, \tag{9}$$

where we have used the well known expression for the coefficient of nonlinear absorption of a traveling wave by the Doppler-broadened transition[5,13]. An implicit expression for the solution of (9) is

$$2(z-z_0)+\ln\left(\frac{z-1}{z_0-1}\frac{z_0+1}{z+1}\right)=-\varkappa_0 x, \tag{10}$$

where $z = \sqrt{1+bE_+^2(x)}$, $z_0 = \sqrt{1+bE_+^2(0)}$, and $E_+(0)$ and $E_+(x)$ are the wave field intensities at the entrance to the cell and at a distance x from the entrance. Figure 2 shows plots of the transmission of the forward wave

$$\eta_+ = E_+^2(L)/E_+^2(0) \tag{11}$$

against the absorption-saturation level at the entrance $bE_+^2(0)$ for different optical thicknesses of the absorbing cell $d = \kappa_0 L$, obtained by solving Eq. (10).

Substituting the distribution of the field amplitude of the forward wave along the cell, obtained by solving Eq. (10), into the general expression (5) and integrating in accordance with (7), we can determine the dependence of the transmission of the backward wave $\eta_-(\Omega)$ on the frequency, on the field intensity of the entering

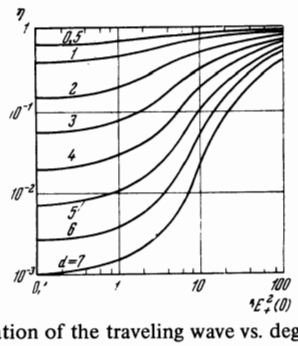

FIG. 2. Attenuation of the traveling wave vs. degree of saturation of the absorption at the entrance to the cell for different initial optical densities of the cell $d = \kappa_0 L$.

wave, on the cell thickness, etc. In the general case, this can be done only by numerical integration with a computer. A typical plot by $\eta_-(\Omega)$ for $\Delta\omega_{Dop} \gg 2\Gamma$ is shown in the insert of Fig. 3. In the center of the Doppler absorption line ($\Omega = 0$) there is a transmission peak, the parameters of which depend on the degree of saturation at the entrance $dE_+^2(0)$ and on the optical density of the cell $d = \kappa_0 L$.

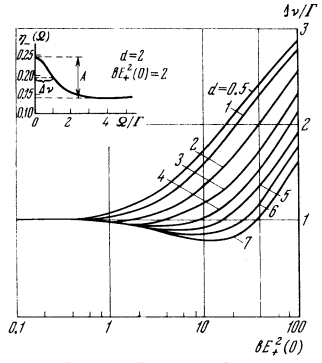

FIG. 3. Dependence of the half-width of the transmission peak on the degree of saturation of the absorption at the entrance for different initial optical densities of the cell. In the upper left corner is shown a typical form of the transmission peak of the backward weak wave: A—absolute contrast of the peak, Ω—field frequency deviation from the center of the Doppler line, Γ—half-width of the homogeneous line.

Figure 3 shows the dependence of the half-width $\Delta\nu$ of the transmission peak at half height on the degree of saturation by the forward absorption wave at the entrance of the cell, $bE_+^2(0)$, for different values of the optical density d of the cell. At small optical density ($d < 1$) the width of the transition resonance increases with increasing saturation, owing to the broadening of the "hole" in $S(\omega)$. The half width of the "hole" in $S(\omega)$ at the field frequency increases in proportion to $\delta\nu = \Gamma\sqrt{1 + bE_+^2}$ [5,13]. The half-width of the transmission resonance $\kappa_-(\Omega)$ increases approximately like

$$\Delta\nu = {}^1/_2(\delta\nu + \Gamma) = {}^1/_2\Gamma(1 + \sqrt{1 + bE_+^2}). \quad (12)$$

We note that for an exact determination of the width of the transmission resonance at the half height it is necessary to measure the value of η_- in the wings. In practice this is impossible, owing to the finite width of the Doppler line. Therefore we considered in the calculation the finite region $|\Omega| \leq 10\Gamma$ and chose in place of $\eta_-(\infty)$ the value of η_- at the point $\Omega = 10\Gamma$. This leads to less broadening by the field than in accordance with (12). With increasing optical density, the broadening of the transmission peak is offset somewhat by the narrowing due to the exponential attenuation of the wings of $\kappa_-(\Omega)$ during the propagation. This effect is perfectly analogous to the narrowing of the band of an optical amplifier with a large gain, inasmuch as for the backward wave a narrow transmission band of the saturated molecules is equivalent to a resonant amplification line.

An important role in the transmission peak is played by such parameters as the absolute contrast of the peak A and the relative contrast α, which are determined by the relations

$$A = \eta_-(0) - \eta_-(\infty), \quad (13)$$

$$\alpha = \eta_-(0)/\eta_-(\infty). \quad (14)$$

The dependence of the absolute contrast A on the degree of saturation at the input for different optical thicknesses of the cell is shown in Fig. 4. We note that at the maximum the transmission of the backward weak wave is equal to the transmission of the strong forward wave:

$$\eta_-(0) = \eta_+. \quad (15)$$

From the theoretical data it follows that the most pronounced peaks are obtained in cells with optical density $d = 1.0-3.0$ with a degree of saturation $bE_+^2(0) \approx 4-10$ at the entrance. In this case the absolute contrast reaches 50% and the relative contrast reaches several units, while the broadening of the peak by the field does not exceed 50%. Consequently, the sensitivity of registration of narrow peaks in the saturation of the absorption by a quasitraveling strong wave in an external cell may be much higher than in a cell located in the field of a standing wave inside a resonator.

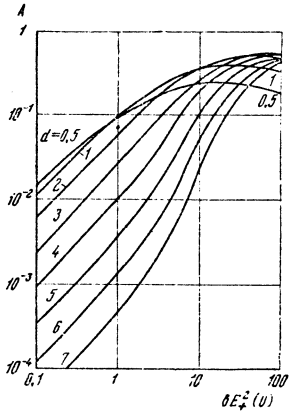

FIG. 4. Absolute contrast of the peak at different degrees of saturation of the absorption at the entrance and at different initial optical densities of the cell.

3. EXPERIMENT

The experimental setup is shown in Fig. 5. We used a flow-through tunable CO_2 laser 195 cm long with a mixture $CO_2:N_2:He = 1:1:3$ at a total pressure of 7 Torr. The amplifying tube was 110 cm long and had an inside diameter of 9 mm; it was cooled with water. The excitation was by means of a dc discharge. One of the mirrors was a diffraction grating with 150 lines/mm, operating in the autocollimation regime, and the output mirror of the laser was a plane-parallel

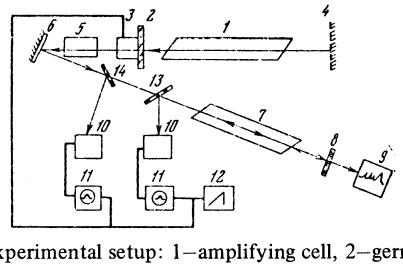

FIG. 5. Experimental setup: 1—amplifying cell, 2—germanium window, 3—piezoceramic tube, 4—diffraction grating, 5—power attenuator, 6—total-reflection mirror, 7—external absorbing cell, 8—weakly reflecting mirror, 9—spectrophotometer, 10—Ge:Au detector, 11—oscilloscope, 12—sawtooth voltage generator, 13, 14—semitransparent mirrors.

germanium plate. The emission frequency of the CO_2 laser was tuned within the limits of the Doppler line by scanning the position of the resonator output mirror, which was mounted on a piezoceramic tube. It was possible to apply to the piezoceramic voltages from a sawtooth generator as well as a sinusoidal voltage. These voltages could produce mirror oscillations with amplitudes larger than $\lambda/2$.

The laser emission output power could reach 1 W. The necessary attenuation of the output power was effected with the aid of a short cell filled with gaseous SF_6 at high pressure.

The radiation of the CO_2-N_2-He laser was passed through an external absorbing cell (length 10—80 cm, diameter 2 cm) with SF_6. A small fraction ($\sim 3\%$) of the radiation passed by the cell was reflected at a small angle in the backward direction, and was registered, after passing again through the cell, by means of a Ge:Au photoreceiver operating at 77°K. After amplification, the output signal was applied to the vertical input of the oscilloscope: the horizontal sweep of the oscilloscope was effected by a generator signal controlling the piezoceramic. For identification of the rotational-vibrational lines of the CO_2 laser we used a spectrophotometer with a resolution of 50 Å. All the measurements were carried out at 25°C.

Spectroscopic data. These data on the absorption of the CO_2 laser radiation at $\lambda = 10.6\ \mu$ by the SF_6 were published in[14,15]. The absorption coefficient at the maximum, measured in[15] for a number of lines, turned out to be

Line:	P (16)	P (18)	P (20)
κ_0, $cm^{-1}\,torr^{-1}$:	1.3	0.46	0.34

The Doppler width of the SF_6 line at 25°C amounts to $\Delta\nu_{Dop} = 30$ MHz. The SF_6 absorption saturation intensity at relatively high pressures (0.28—0.82 Torr), when the line broadening is close to homogeneous, was measured in[14]. For the P(20) line, the saturation intensity is $I = 7 \pm 2\ W/cm^2$ Torr.

The narrow resonances in the lines P(16), P(18), P(20), etc, upon saturation of the SF_6 absorption by the CO_2 laser radiation, were first observed in[12] and recently in[16,17].

For a detailed study of the parameters of the narrow resonances in the low-pressure SF_6 cells, we chose the P(18) line of the CO_2 laser radiation. An oscillogram of the transmission of the backward wave as a function of the frequency within the limits of part of the Doppler line P(18) is shown in Fig. 6.

Nonlinear absorption of traveling wave. To measure the dependence of the parameters of the transmission peak of the weak reflected wave on the degree of saturation of the absorption by the forward wave it is necessary to know the saturation parameter in the region of low pressures of SF_6, when the absorption line is inhomogeneously broadened. To this end we measured the transmission of the forward wave as a function of the level of its intensity at the entrance to the cell for different SF_6 pressures in the absorbing cell (Fig. 7). Extrapolating this dependence to zero intensity, we obtain the optical density of the cell $d = \kappa_0 L$, and from the latter we obtain directly the absorption coefficient of SF_6. The measured value of the absorption coefficient of SF_6 on the P(18) line is 0.5 $cm^{-1}Torr^{-1}$, in agreement with the data of[15]. By aligning the experimental transmission curve of the forward wave with the calculated one (Fig. 2), at equal values of the optical density d, we obtain the corresponding values of the saturation intensity. The procedure for finding the saturation parameter with the aid of the calculated curves is explained by using cells with optical density $d \gtrsim 1$.

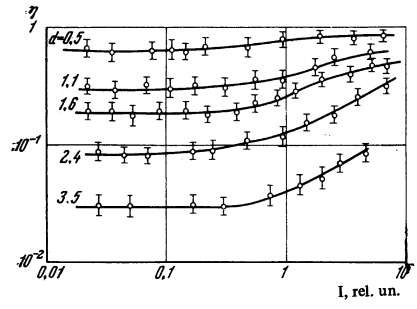

FIG. 7. Attenuation of traveling wave as a function of the power at the entrance to the cell for different values of the initial optical density of the cell.

Within the pressure range 10—70 mtorr, the dependence of the saturation parameter on the SF_6 pressure is linear. Such a dependence is due to the fact that even in the region of the lowest pressures the mean free path of the SF_6 molecule was much lower than the beam diameter, and molecule relaxation could be produced only by their collisions. A deviation from the linear dependence can be expected only at an SF_6 pressure on the order of 1 mtorr, when the mean free path of the molecules exceeds the beam diameter.

Nonlinear absorption of quasitraveling wave [P(18)]. For the transmission peak shown in Fig. 6, it is of interest to investigate the variation of the peak parameters with increasing field power and with increasing optical density of the absorbing cell.

Figure 8 shows the experimental dependence of the width $2\Delta\nu$ of the transmission peak at half-height on the degree of saturation of the absorption by the forward wave at the input of the cell for different optical densities of the cell d. The course of the curves within the limits of the measurement error (20%) is described by expression (12). Since the measurement is carried out by increasing the pressure p, the width of the peak in a weak field depends only on p. Approximating the curves in the region of the weak field, we

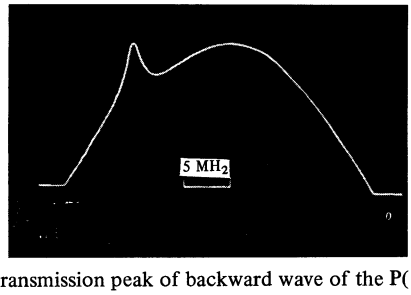

FIG. 6. Transmission peak of backward wave of the P(18) line. SF_6 pressure 40 mtorr.

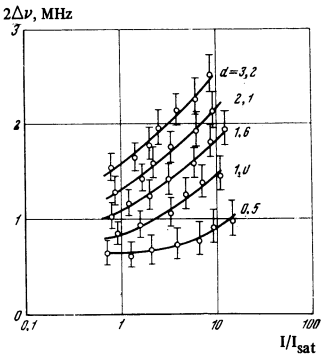

FIG. 8. Width of the transmission peak as a function of the degree of saturation of the absorption by the forward wave at the entrance to the cell for different values of the optical density d.

can find the dependence of the width of the transmission peak on the SF_6 pressure in the cell. A similar dependence can be found also by another method, knowing the broadening of the peak by the strong field, but in this case it is necessary to use the calculated dependence of the peak width on the field amplitude and on the optical density of the cell in accordance with Fig. 3. The results obtained in both cases are the same. For SF_6, the homogeneous width 2Γ of the transition depends on the pressure, with a proportionality coefficient equal to 17 ± 4 MHz/Torr. In the Lorentz-broadening model, the time of collisions between molecules is given by $pT_2 = 1.9 \times 10^{-8}$ sec-Torr, and the mean free path by $pl_{path} \sim 4 \times 10^{-4}$ cm-Torr.

The width of the narrowest transmission peak observed in the experiment was $2\Delta\nu = 0.6 \pm 0.1$ MHz at half height, at an SF_6 pressure 10 mtorr. The experimental resolution was 0.5 ± 0.1 MHz. This value consists of the broadening due to the finite time of flight through the beam (0.1 MHz), the broadening connected with the fact that the forward and backward beams are not parallel, a fact necessary to eliminate self-excitation of the laser with internal absorbing cell (~0.15 MHz), the broadening due to the collisions (0.2 MHz), and the broadening due to saturation (~0.15 MHz).

We measured the dependence of the relative contrast α on the degree of saturation by the forward wave and on the optical thickness of the cell. The results are shown in Fig. 9. They agree qualitatively with the theoretical calculations. The observed relative contrast in cells with large optical density reached 200%, thus confirming the high sensitivity of the method. The lowering of the relative contrast in the region of the strong field, compared with the theoretical value, is due to the fact that it is very difficult to measure in the experiment the wings of the transmission peak over a considerable section of the Doppler absorption line of SF_6, the width of which is only 30 MHz.

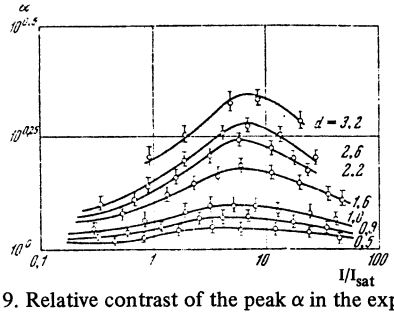

FIG. 9. Relative contrast of the peak α in the experiment.

Narrow resonances on other P lines. Narrow resonances were observed within the limits of the Doppler line for several lines of the P branch of the CO_2 laser emission. On the P(20) line there are observed four weak peaks. Within the limits of the Doppler line P(18), one can see one strong resonance belonging to SF_6. On the contour of the Doppler line P(16), which has the maximum absorption, there are four resonances at saturation of the SF_6 absorption. The width of the most intense dip at the minimal power levels is approximately twice as large as the width of any other dip observed at the same pressure (10—30 mtorr). We succeeded in resolving the structure of the anomalously broad peak. It turned out that it consists of three narrow resonances (Fig. 10). In addition, the high resolution of the spectroscope has made it possible to observe within the limits of the Doppler width of the P(14) line, not one peak, as indicated heretofore[17], but four peaks of transmission upon saturation of the SF_6 absorption.

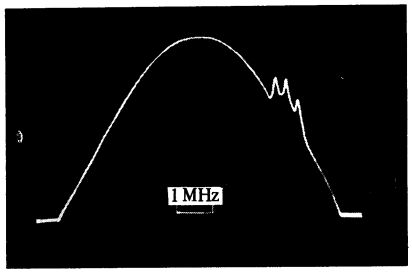

FIG. 10. Structure of anomalously broad resonance belonging to SF_6 on the contour of the P(16) emission line of a CO_2 laser. The SF_6 pressure is 8 mtorr. The scale indicated in the figure corresponds to 1 MHz.

4. CONCLUSION

We have demonstrated the great possibilities of spectroscopy within the Doppler line, by the method of saturating the absorption with a quasitraveling wave. The saturation by a quasitraveling wave in an external cell makes it possible to determine the time of collisions between the SF_6 molecules. We have measured, apparently for the first time, the effects of broadening of molecular resonances by a strong field. The experimentally obtained large amplitude of the peak, together with its narrowness, make this method particularly effective for stabilization of the frequency of a CO_2 laser. The high resolving power of the method is also evidenced by the resolution of the hyperfine structure of SF_6 at the P(16) line and by the observation of weak resonances in the P(14) line of the CO_2 laser emission. The resolving power of the method can be increased by one more order of magnitude. This makes it possible to investigate subtle effects connected with coherent interaction of the field with molecules and with the interaction of the molecules with one another.

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