Optically induced drift of SF_6 molecules

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An experimental search for optically induced drift of $^{32}SF_6$ molecules mixed with He, CH_3F, and $^{34}SF_6$ molecules is described. The effect was not observed in a wide range of experimental conditions. The absence of the effect is attributed to the complicated structure of the SF_6 absorption spectrum, to field saturation of the transition, and to the small difference between the frequencies of the Maxwellizing collisions in the vibrational SF_6 states that interact with the radiation. An estimate $-2.4\times 10^{-4} < \Delta\nu/\nu < 4.2\times 10^{-4}$ is given for the factor $\Delta\nu/\nu$.

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In 1979 Gel'mukhanov and Shalagin theoretically predicted an interesting phenomenon, namely an optically induced drift (OID) of particles in a resonant field of a traveling electromagnetic wave.[1] Its physical gist is the following. Narrow-band radiation interacting with a Doppler-broadened transition of an atomic system produces a dip in the particle velocity distribution on the lower level, and a peak on the upper level. The appearance of the peak and of the dip means the appearance of fluxes of excited and unexcited particles, $\mathbf{J}_m$ and $\mathbf{J}_n$, collinear with the wave vector $\mathbf{k}$. In the absence of a buffer gas we have $\mathbf{J}_m + \mathbf{J}_n = 0$ and the absorbing particles are on the whole at rest. In the presence of a buffer gas the fluxes $\mathbf{J}_m$ and $\mathbf{J}_n$ begin to be subject to pressure, which in the general case is different for the states m and n. Now already $\mathbf{J}_m + \mathbf{J}_n \neq 0$, and the absorbing particles are set in motion.

The flux is determined by the factor $\Delta\nu/\nu$, the relative change of the frequency of the Maxwellizing collisions in the states m and n. The sign of the effect (the direction of the resultant flux $\mathbf{J}_m + \mathbf{J}_n$ of the absorbing particles) is determined by the sign of $\Delta\nu/\nu$ and by the sign of the detuning of the frequency of the electromagnetic field (ω) relative to the frequency of the center of the absorption line ($\omega 0$), $\Omega = \omega - \omega_0$. The antisymmetric dependence, which is peculiar to the OID, of the effect on the frequency (Ω) makes it easy to distinguish it from other strong effect (e.g., laser thermal diffusion[2]), which have a symmetric dependence on Ω.

The first experimental confirmation of the theory of the OID phenomenon was obtained in Ref. 3, where the drift of sodium atoms in a buffer gas (helium, neon) was investigated upon optical excitation of the $^2S_{1/2} - {}^2P_{1/2,3/2}$ transitions (the sodium lines D_1 and D_2).

A number of recent papers are devoted to the OID of molecules under the influence of IR radiation that is at resonance with a vibrational-rotational transitions. The theoretical questions were investigated in detail.[4–6] An experimental study of the effect was initiated. By now, results of investigations of the OID of two molecules, SF_6 and CH_3F, have been published. In the CH_3F molecule the effect was reliably observed.[7,8] Under optimal conditions the OID increases the density of the radiation-absorbing gas component by 70%. A dependence of the effect on Ω, peculiar to the OID, was also observed.

The situation with the observation of the effect in the SF_6 molecule turned out to be much more complicated. In the first paper[9] was reported observation of a very large OID effect following excitation by pulsed radiation. The phenomenon, however, was not observed under cw irradiation under the same conditions and at the same average power density. The measurements in Ref. 10 were made with excitation in a continuous regime. The authors of Ref. 10 observed an increase of the density of the SF_6 molecules following excitation of a CO_2 laser on the lines $P(28)$ and $P(32)$ and a decrease on the lines $P(20)$ and $P(22)$. This phenomenon was attributed to OID. Finally, absence of the effect was reported in Ref. 11. The SF_6 molecules were excited in that case by a CO_2 laser on the $P(18)$ line.

Under these circumstances it seemed of interest to investigate the effect in the SF_6 molecule under conditions that made it possible previously to observe the effect in the CH_3F molecule. The results of these measurements are reported in the present paper. We investigated the drift of $^{32}SF_6$ molecules in a medium of buffer particles CH_3F, He, and $^{34}SF_6$. Principal attention we paid to the investigation of the $SF_6 + \mathrm{He}$ gas mixture, inasmuch as sufficiently detailed data are available on the collisions of these particles. References 9–11 are also devoted to investigations of OID in $SF_6 + \mathrm{He}$.

1. TECHNIQUE AND EXPERIMENTAL RESULTS

The experimental setup is shown in Fig. 1. In this arrangement an optically induced particle flux is produced only during the transient at the start of the excitation. The drift stops after the stationary state is reached and a drop of the density of the absorbing gas component is produced along the beam. The inevitable heating of the gas by the intense radiation makes it inconvenient to determine the magnitude of the effect by directly measuring the change in the density of the absorbing particles. Less subject to the thermal action is the determination of the effect from the optically induced change of the composition of the gas mixture. This change was analyzed in our experiment by mass spectroscopy.

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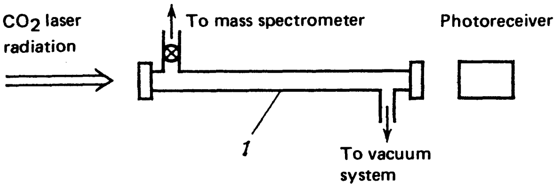

FIG. 1. Experimental setup. 1—Cell (copper tube, inside diameter 4 mm, length 1 m).

The gas was excited in our experiment by a CO_2 laser, using a diffraction grating as the selector of the rotational lines, and with the frequency stabilized to the line center proper. The laser operated in a single-mode regime on lines belonging to the 10.6 μm vibrational band (the 00°1–10°0 transition of CO_2). The radiation (up to 10 W) was focused by a lens ($f = 50$ cm) into a copper cell with inside diameter 4 mm and length 1 m. In some experiments the radiation was not focused. The cell windows were made of NaCl. The radiation loss in the evacuated cell was less than 20%. The gas composition were analyzed at the entrance end (relative to the beam) of the cell. To this end, a gas sample (volume $0.2 - 1$ cm^3) was directed (after the stationary state was reached in the system) to the mass spectrometer. The exit end of the cell was connected to a ballast volume. This cell construction increases the optically induced change of the gas composition at the entrance end of the cell by a factor of two.

The measurement procedure consists of repeated comparisons of the mass spectra of the gas with and without irradiation. To increase the measurement accuracy, as small amount of a reference gas having lines favorably located on the mass spectra was placed in the cell in addition to the absorbing and buffer particles.

The measurement results are summarized in the table, where the following notation is used: S is the incident power density, ΔS the power density absorbed by the gas over the cell length, β the enrichment, equal to $(K_{\rm ill}/K) - 1$, where $K_{\rm ill}$ and K are the ratios of the densities of the absorbing particles to the buffer (or reference) particles with and without illumination. The values of β in each line of the table are the results of averaging over approximately 20 mass-spectrometer measurements of $K_{\rm ill}$ and K.

It can be seen from the experimental results that irradiation of the gas does not change its composition, accurate to 0.2–1%. What conclusions can be drawn from these data on the relative change $\Delta\nu/\nu$ of the frequency of the Maxwellizing collisions of the SF_6 molecule when it is vibrationally excited? To answer this question we consider briefly the the-

TABLE I. Results of measurement of the OID effect of the SF_6 molecules. The measurement errors are taken to be the standard deviations from the mean values.

Experiment No.	S, W/cm^2	ΔS, W/cm^2	CO_2 laser line	β, %	$\frac{\Delta\nu}{\nu}\cdot 10^4$
Partial pressures of components, Torr					
$^{32}SF_6+Ar+^{12}CH_3F=0.055+0.016+0.36$					
1	90	27	$P(18)$	-0.8 ± 1.4	
2	96	28	$P(18)$	0 ± 1.2	
3	103	34	$P(20)$	0 ± 1	
4	95	34	$P(22)$	-2.2 ± 1	
5	90	20	$P(16)$	-0.2 ± 0.6	
6	90	13	$P(14)$	0.4 ± 1	
$^{32}SF_6+Ar+^{12}CH_3F=0.037+0.01+0.9$					
7	99	21	$P(18)$	-0.4 ± 0.5	
$^{32}SF_6+Ar+He=0.082+0.025+2.2$					
8	87	59	$P(22)$	-0.9 ± 0.6	
9	85	58	$P(20)$	-0.9 ± 0.9	
10	92	63	$P(18)$	1.5 ± 1.2	
$^{32}SF_6+^{34}SF_6+He=0.013+0.01+9.6$					
11*	15	6.9	$P(14)$	-0.4 ± 0.5	1 ± 1.2
12*	14	5.5	$P(18)$	-0.08 ± 0.4	0.5 ± 2.5
$^{32}SF_6+^{34}SF_6+He=0.012+0.009+9.5$					
13	81	23	$P(14)$	-0.2 ± 0.5	
14	85	26	$P(18)$	-1.1 ± 0.5	
15	82	17	$P(20)$	0.1 ± 0.3	
$^{32}SF_6+Ar+He=0.01+0.033+20$					
16	82	37	$P(18)$	1.2 ± 0.5	
17	82	20	$P(14)$	-0.4 ± 0.5	
$^{32}SF_6+^{34}SF_6=0.086+0.92$					
18	81	33	$P(14)$	0.7 ± 0.2	
19	41	20	$P(14)$	-0.2 ± 0.2	
20*	27	17	$P(14)$	-0.2 ± 0.2	

*The experiments were performed without focusing the radiation into the cell.

ory of the OID phenomenon for vibrational-rotational molecule transitions.

2. THEORY OF OPTICALLY INDUCED DRIFT OF MOLECULES

We shall solve the problem of the interaction of a strong electromagnetic wave with a gas of moving molecules in the strong-collision model.[12] We take into account in this case collisions of three types: collisions that lead to establishment of the equilibrium distribution over the rotational levels, but do not change the molecule velocity and its vibrational state; collisions that establish an equilibrium distribution in velocity and in the vibrational level (without change of the vibrational state), and collisions that lead to quenching of the vibrational excitation and to establishment of equilibrium distributions in the velocities and in the rotational levels.

In the model assumed, under stationary and homogeneous conditions, we have the following equation for the velocity distributions of the molecule densities in the upper $[\rho_{mJ}(\mathbf{v})]$ and lower $[\rho_{nJ}(\mathbf{v})]$ vibrational-rotational states:

$$\nu_m\rho_{mJ}(\mathbf{v})=Np(\mathbf{v})\delta_{JJ_0}+\nu_{mR}\rho_m(\mathbf{v})W_B(J)+\nu_{mT}\rho_mW_B(J)W(\mathbf{v}),$$

$$\begin{gathered}\nu_n\rho_{nJ}(\mathbf{v})=-Np(\mathbf{v})\delta_{JJ_0'}+\nu_{nR}\rho_n(\mathbf{v})W_B(J)\\+\nu_{nT}\rho_nW_B(J)W(\mathbf{v})+\nu_{mv}\rho_mW_B(J)W(\mathbf{v});\\ \nu_m=\nu_{mR}+\nu_{mT}+\nu_{mv},\qquad \nu_n=\nu_{nR}+\nu_{nT}.\end{gathered}\tag{1}$$

Here N is the density of the absorbing molecules; J_0 and J_0' are the rotational numbers of the levels that interact with the field; ν_{mR} and ν_{nR} are the frequencies of the collisions accompanied by a strong change of the angular momentum of the molecules in the states m and n; ν_{mT} and ν_{nT} are the frequencies of collisions with strong change of the molecule momentum in the states m and n; ν_{mv} is the frequency of the vibrational relaxation, $W_B(J)$ is the Boltzmann distribution over the rotational levels; $W(\mathbf{v})$ is the Maxwell distribution; $Np(\mathbf{v})$ is the velocity distribution of the density of the molecules excited by the radiation per unit time. In (1) we have introduced the notation

$$\rho_i(\mathbf{v})=\sum_J\rho_{iJ}(\mathbf{v}),\quad \rho_i=\int\rho_i(\mathbf{v})\,d\mathbf{v}\quad (i=m,n).\tag{2}$$

It is assumed in Eqs. (1) that the rotational relaxation is faster than the translational. In this respect our analysis differs from that in Ref. 6, where the calculations were made assuming a very fast translational relaxation. A fast rotational relaxation was assumed also in Ref. 5. There, however, in contrast to our model, it was assumed that the collisions that lead to the momentum relaxation are not accompanied by relaxation in J. The vibrational relaxation in the model of Ref. 5, in contrast to our model, is not accompanied by momentum relaxation.

For $p(\mathbf{v})$ we have the equation

$$\begin{gathered}Np(\mathbf{v})=\frac{2|G|^2\Gamma}{\Gamma^2+(\Omega-\mathbf{kv})^2}[\rho_{nJ_0'}(\mathbf{v})-\rho_{mJ_0}(\mathbf{v})],\\ G=Ed_0/2\hbar,\qquad \Omega=\omega-\omega_0,\end{gathered}\tag{3}$$

where ω, $\mathbf{k}$, and E are the frequency, wave vector, and amplitude of the electromagnetic wave; d_0, ω_0, and Γ are the dipole moment, frequency, and impact half-width of the vibrational-rotational $mJ_0\rightarrow nJ_0'$ transition that interacts with the field.

From (1)–(3) we get

$$p(\mathbf{v})=\frac{\Gamma_B{}^2\varkappa W(\mathbf{v})}{\tau_1(1+\varkappa)[\Gamma_B{}^2+(\Omega-\mathbf{kv})^2]}[W_B(J_0')-p\tau_2],\tag{4}$$

where the absorption probability p is of the form

$$p=\int p(\mathbf{v})\,d\mathbf{v}=\frac{W_B(J_0')}{\tau_1}\frac{Y(\Omega)}{1+1/\varkappa+Y(\Omega)\tau_2/\tau_1};\tag{5}$$

here

$$\begin{gathered}\tau_1=\frac{1}{\nu_n}+\frac{1}{\nu_m}+W_B(J_0')\left[\frac{1}{\nu_{nT}}-\frac{1}{\nu_n}\right]+W_B(J_0)\left[\frac{1}{\nu_{mv}+\nu_{mT}}-\frac{1}{\nu_m}\right],\\ \tau_2=W_B(J_0')\left[\frac{1}{\nu_{mv}}-\frac{1}{\nu_{nT}}\right]+W_B(J_0)\left[\frac{1}{\nu_{mv}}-\frac{1}{\nu_{mv}+\nu_{mT}}\right].\end{gathered}\tag{6}$$

The function $Y(\Omega)$ is the Voigt contour of the absorption line

$$\begin{gathered}Y(\Omega)=\int\frac{\Gamma_B{}^2W(\mathbf{v})\,d\mathbf{v}}{\Gamma_B{}^2+(\Omega-\mathbf{kv})^2},\\ \Gamma_B=\Gamma(1+\varkappa)^{1/2},\qquad \varkappa=2|G|^2\tau_1/\Gamma.\end{gathered}$$

We shall find it convenient to express $Y(\Omega)$ in terms of the probability integral

$$Y(\Omega)=\pi^{1/2}y\,\mathrm{Re}[w(z)],\qquad z=x+iy,\qquad x=\Omega/kv_0,\tag{7}$$

$$y=\frac{\Gamma_B}{kv_0},\quad w(z)=e^{-z^2}\left[1+\frac{2i}{\pi^{1/2}}\int_0 e^{t^2}dt\right]$$

[kv_0 is the Doppler parameter, $v_0=(2k_BT/M)^{1/2}$]. Accurate to the specific meaning of the quantities τ_1 and τ_2, the solution (5) agrees with the expression for p in Ref. 6.

The flux of the absorbing particles is easily obtained directly from (1):

$$\mathbf{J}=\int\mathbf{v}[\rho_m(\mathbf{v})+\rho_n(\mathbf{v})]\,d\mathbf{v}=N\frac{\nu_{nT}-\nu_{mv}-\nu_{mT}}{\nu_{nT}(\nu_{mv}+\nu_{mT})}\int p(\mathbf{v})\mathbf{v}\,d\mathbf{v}.\tag{8}$$

The flux $\mathbf{J}$ is determined by the difference between the frequencies of the Maxwellizing collisions in two states: $\Delta\nu=\nu_{mv}+\nu_{mT}-\nu_{nT}$. If this difference is small, we have approximately $\nu_{nT}(\nu_{mv}+\nu_{mt})\approx\nu^2$. It is convenient to represent the integral $\int p(\mathbf{v})\mathbf{v}d\mathbf{v}$ in the form[6]

$$\int p(\mathbf{v})\mathbf{v}\,d\mathbf{v}=\mathbf{k}pv_0\varphi(\Omega)/k,\quad \varphi(\Omega)=\int p(\mathbf{v})(\mathbf{kv})\,d\mathbf{v}/pkv_0.$$

Using (4), we express the function $\varphi(\Omega)$ in terms of the probability integral:

$$\varphi(\Omega)=\mathrm{Re}[zw(z)]/\mathrm{Re}[w(z)].\tag{9}$$

If the experimental conditions are such that there is no gas flow in the stationary state, then the produced drop ΔN of the absorbing gas component along the beam is determined from the condition $\mathbf{J}+\mathbf{J}_{\text{diff}}=0$ ($\mathbf{J}_{\text{diff}}$ is the diffusion flux). From this condition we obtain, assuming the radiation wave vector to be directed along the x axis,

$$-\frac{N\Delta\nu}{\nu^2}pv_0\varphi(\Omega)=\frac{v_0^2}{2\nu}\frac{dN}{dx}. \tag{10}$$

The change of the power density (S) of the radiation along the beam is connected with p:

$$dS/dx=-N\hbar\omega p. \tag{11}$$

From (10) and (11) we obtain the drop ΔN of the concentration of the absorbing gas component:

$$\Delta N=-\frac{\Delta\nu}{\nu}\frac{2\Delta S}{\hbar\omega v_0}\varphi(\Omega); \tag{12}$$

here ΔS is the power density absorbed by the gas. Expression (12) was first derived in Ref. 6. This expression is most convenient for the determination of the parameter $\Delta\nu/\nu$, since it contains easy-to-measure quantities. The value of $\varphi(\Omega)$ can be calculated if Ω and Γ_B are known.

3. OPTIMAL CONDITIONS FOR OBSERVING THE EFFECT IN THE SF$_6$ MOLECULE

We determine now the optimal conditions for observing the effect, with an aim at investigating it in the SF$_6$ molecule. This molecule has a complicated level and transition structure. Thus for example, the $P(18)$ emission line of a CO$_2$ laser is absorbed by a strong SF$_6$ line [the transition $P(33)A\frac{1}{2}$ of the $0\rightarrow\nu_3$ band, $\Omega = 7.18$ MHz, Ref. 13] and by a large number of neighboring weak lines.[13] The weak lines in this section of the SF$_6$ absorption spectrum are arranged with an approximate spacing of 2 MHz between lines. Since the Doppler parameter for these transitions of SF$_6$ is 18 MHz at $T = 300$ K, the absorption-line contours overlap.

We turn now to expression (5) for the absorption probability. We see from it that for the parameters $\varkappa$ such that $\varkappa \gg [1 + Y(\Omega)\tau_2/\tau_1]^{-1}$, the absorption probability no longer depends on $\varkappa$, meaning also on the dipole moment of the transition. In this limit the absorption by the weak and strong lines is of the same order, meaning that the velocity selectivity of the excitation is lost. It is important that this circumstance imposes a stronger restriction on the value of $\varkappa$ than the condition $\Gamma_B \lesssim kv_0$ that holds for isolated absorption lines. The velocity selectivity of the excitation vanishes at large $\varkappa$ also on account of absorption by transitions of hot bands.

The velocity selectivity of the excitation is preserved under the condition

$$S/S_s\equiv\varkappa[1+Y(\Omega)\tau_2/\tau_1]\ll 1. \tag{13}$$

We have introduced here a parameter S_s with dimension of power density. This parameter describes the radiation-power density that saturates the transition. At low radiation intensities, defined by (13), the expression for p is simpler. From (5) we obtain

$$p=\pi^{1/2}W_B(J_0')\varkappa\Gamma\,\mathrm{Re}[w(z)]/\tau_1 kv_0. \tag{14}$$

For the concentration drop ΔN we obtain from (11), (12), and (14)

$$\Delta N=-\frac{\Delta\nu}{\nu}NlW_B(J_0')\pi^{1/2}\varkappa ky^2\,\mathrm{Re}[zw(z)]; \tag{15}$$

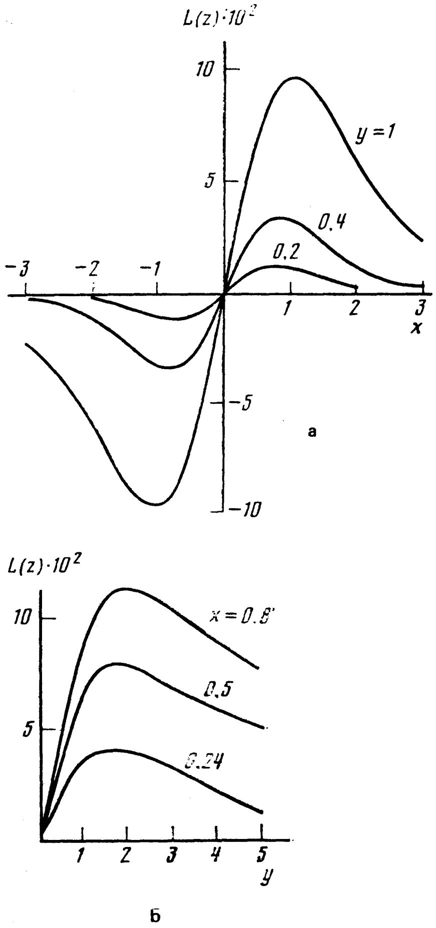

FIG. 2. Examples of plots of the function $L(z) = y^2\,\mathrm{Re}[w(z)]$: a) $y = 0.2$; 0.4; 1, b) $x = 0.24$; 0.5; 0.8.

where we assume $\Gamma\sim 2/\tau_1$ and l is the cell length. Since $\varkappa$ is fixed by the relation (13), variation of the conditions reduces to finding the extrema of the function $L(z) = y^2\,\mathrm{Re}[zw(z)]$. For the case when the SF$_6$ molecules are excited by CO$_2$ laser radiation on the $P(18)$ line we have $x = 0.4$ and $L(z)$ reaches a maximum at $\Gamma \approx 1.5kv_0$. Examples of $L(z)$ plots are shown in Fig. 2. The numerical values of the function $w(z)$ were obtained with the aid of the tables of Ref. 14.

It is important in what follows to determine the permissible radiation-power densities. We estimate the value of S_n for the SF$_6$ absorption line $P(33)A\frac{1}{2}$ of the $0\rightarrow\nu_3$ vibration following excitation of the line by CO$_2$ laser radiation on the $P(18)$ line at a helium pressure 1 Torr. According to the data of Ref. 15 on the vibrational relaxation of SF$_6$ in helium, $\tau_2 \approx 1.2\times 10^{-7}$. The dipole moment of the transition is ~ 0.3 D. The broadening of the SF$_6$ transitions in helium is $\sim 10^{+7}$ sec^{-1}. These values yield $S_n \approx 0.15$ W/cm^2. Thus, at a helium pressure 1 Torr it is necessary to use radiation with a power density $\lesssim 0.15$ W/cm^2. With increasing pressure, the permissible power density increases rapidly ($\sim P^2$). At 10 Torr of helium the radiation power density can be increased to ~ 15 W/cm^2. These estimates agree with the data of Refs. 16–18, devoted to the saturation of the absorption of cw emission of a CO$_2$ laser. We have also measured directly the absorption coefficient at several power-density levels of the emission of a CO_2 laser operating on the $P(18)$ line of the 10.6 μm band. The gas composition was $^{32}SF_6$: $^{34}SF_6$: He = 1:0.8:570. The pressure was 9.7 Torr. The results were

S, W/cm^2:	20	52	71
α, cm^{-1}/Torr:	0.38	0,3	0,23

The linear-absorption coefficient for the $P(18)$ line of the CO_2 laser is ~0.4 cm^{-1}/Torr.[18] It can be seen that the calculated value of S_n agrees qualitatively with the measurement results.

4. DETERMINATION OF THE PARAMETER $\Delta\nu/\nu$. DISCUSSION

To determine the parameter $\Delta\nu/\nu$ we need information on Ω. These data were determined in Refs. 18–20 for a number of CO_2 laser emission lines and strong SF_6 absorption lines. The SF_6 absorption-spectrum sections of interest to us are shown in Fig. 3.

According to the arguments above, the experiments that are sufficiently reliable for the determination of the factor $\Delta\nu/\nu$ are Nos. 11 and 12 (see the table). The ensuing values of $\Delta\nu/\nu$ are given in the same table. Their weighted mean is $\Delta\nu/\nu = (0.91 \pm 1.1)\cdot 10^{-4}$.

Experiments Nos. 16 and 17 were performed at a helium pressure 20 Torr and the field saturation in them is also small. It is difficult, however, to use these results for the analysis of the factor $\Delta\nu/\nu$, for at this pressure it is already necessary to take into account the contribution made to the effect by the neighboring strong absorption lines (see Fig. 3).

Our results allow us therefore to establish the possible range of the factor $\Delta\nu/\nu$. For a significance level 0.003 we obtain

$$-2.4\cdot10^{-4}<\Delta\nu/\nu<4.2\cdot10^{-4}. \qquad \text{(b)}$$

So small a value of the factor $\Delta\nu/\nu$ for the SF_6 molecule (more accurately, for the pair of particles SF_6 and He) do not allow us to interpret the results of the experiments of Refs. 9 and 10 as a manifestation of the OID effect. Indeed analysis of the data of Refs. 9 and 10, similar to that carried out above, yields a value of $\Delta\nu/\nu$ that is 10–100 times larger than the limit established by us.

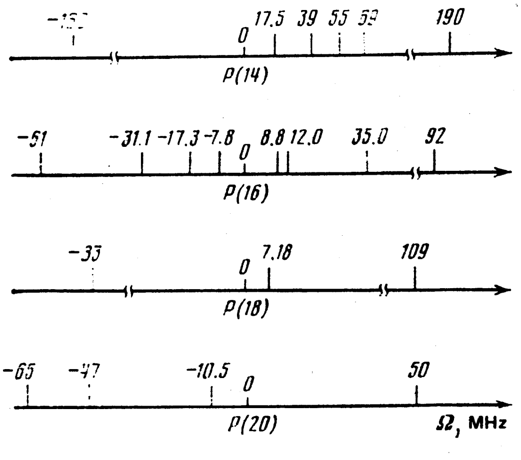

FIG. 3. Sections of SF_6 absorption spectra near the lines $P(14)$ to $P(20)$ of CO_2 laser emission in the 10.6 μm band.

The authors of Ref. 11, who did not observe, as we did, the OID effect in the gas mixture SF_6 + He, cite an estimate $\Delta\nu/\nu = 3\cdot10^{-4}$–$6\cdot10^{-4}$. They conclude that this value does not conform to the value $\Delta\nu/\nu = 5\times10^{-2}$ determined from spectroscopic measurements of the broadening constants of the SF_6 lines in waves that move in the same direction and in opposite directions, namely 4 ± 0.2 and 3.8 ± 0.3 MHz/Torr.[21] In our opinion this conclusion is in error for a number of reasons. The first two of them are more readily of methodological character.

When the factor $\Delta\nu/\nu$ was determined in Ref. 11 from the spectroscopic data,[21] the errors of the latter were not taken into account. When the measurement errors of Ref. 21 are taken into account we obtain $\Delta\nu/\nu = 5\cdot10^{-2} \pm 9\cdot10^{-2}$, which no longer contradicts the value $\Delta\nu/\nu = 3\cdot10^{-4}$–$6\cdot10^{-4}$ of Ref. 11.

The spectroscopic data of Ref. 21 cannot be used directly to determine the factor $\Delta\nu/\nu$, since they pertain to broadening of the SF_6 lines in SF_6 itself, but only collisions of SF_6 with He are of importance for the OID effect in Ref. 11.

We note further that the line broadening constants have no direct bearing on the frequencies of the Maxwellizing collisions. This can be easily verified by calculating the self-diffusion coefficient of SF_6 for the collision frequency corresponding to a broadening 4 MHz/Torr.[21] At a pressure 1 Torr we obtain $D\approx 6.5$ cm^2/sec. Actually, however, the self-diffusion coefficient is $D\approx 44$ cm^2/sec.[22]

The experimental conditions under which the estimate $\Delta\nu/\nu = 3\cdot10^{-4}$–$6\cdot10^{-4}$ was obtained in Ref. 11 were chosen such that strong field saturation took place. This leads, as shown in the present paper, to a lower excitation selectivity in velocity and makes the interpretation of the measurement results much more complicated.

Thus, the small value of OID effect of the SF_6 molecules is due apparently to two factors: the small difference between the frequencies of the Maxwellizing collisions in two vibrational states, and the complicated absorption-spectrum structure, which leads even at relatively low radiation-power densities to a decrease of the excitation selectivity in velocity.

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