DESCRIPTION OF THE TEMPERATURE DEPENDENCE OF HALF–WIDTHS AND SHIFTS OF SPECTRAL LINES OF LINEAR MOLECULES USING THE RATCF MODEL

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The temperature dependence of half–widths and shifts of the spectral lines of CO_2 *and* CO *molecules is analyzed within the scope of the refined Anderson–Tsao–Curnutte–Frost (RATCF) model. It is shown that the temperature coefficients* n_γ *which enter in* $\gamma(T)$ *are dependent of the type of colliding molecules, quantum transition, and temperature interval.*

In this paper we analyze the temperature of half–widths and shifts of spectral lines as functions of the interacting molecular system, of the quantum transition $i\{V_i, J_i\} \to f\{V_f, J_f\}$, and of the temperature interval within the scope of the RATCF model.[1]

EFFECT OF THE TEMPERATURE ON THE LINE HALF–WIDTHS OF CO_2 AND CO MOLECULES

At present the large quantity of experimental and theoretical data have been accumulated concerning the broadening of linear molecules in a wide temperature interval. In addition, the temperature dependence $\gamma_{if}(T)$ is usually described in the form

$$\gamma_{if}(T) = \gamma_{if}(300)\,(T/300)^{-n_\gamma}\,. \qquad (1)$$

The analytic formulas for n_γ can be obtained only in simple model cases[2-5] of fixed intermolecular interaction types and validity of the law $\bar{v} \sim \sqrt{T}$, which is independent of the line number.

In contrast to the well–known analytic models used for determining n_γ, in the RATCF model it is assumed that the most probable velocity of translational motion of the interacting molecules depends on their states. In this case for most numbers J_i the velocity $v(J_i)$ will not obey the law $\bar{v} \sim \sqrt{T}$.

In our previous papers[1,6,7] with the help of the RATCF approach we have demonstrated a good reproduction of the experimental data (γ and δ) at the temperature $T = 300\ K$ in different frequency ranges for the three absorbing molecules CO_2, CO, and NO. In the present paper the calculated values of γ are given for CO_2 and CO molecules at different temperatures, for which experimental data are available.

TABLE I. $\gamma_{CO_2-N_2}$ *for the lines with large values of* J_i *in the* ν_3 *band at different temperatures* ($10^{-3}\cdot cm^{-1}\cdot atm^{-1}$).

T, K	$R(38)$				$R(42)$				$R(54)$			
	Our results	Ref. 8	Ref. 9	Ref. 10	Our results	Ref. 8	Ref. 9	Ref. 10	Our results	Ref. 8	Ref. 9	Ref. 10
198	96.7	–	–	93.5	94.7	–	–	92.6	88.5	–	–	87.2
295	72.0	73.0	71.9±0.7	–	72.0	73.0	70.9±0.3	–	–	–	–	–
297	–	–	–	–	–	–	–	–	69.0	72.0	68.3±0.5	–
371	61.8	62.0	60.5±0.8	–	–	–	–	–	–	–	–	–
379	–	–	–	–	60.9	62.5	59.2±0.2	–	–	–	–	–
411	–	–	–	–	–	–	–	–	57.1	58.0	53.9±0.8	–
435	–	–	–	–	55.4	55.0	53.2±0.2	–	–	–	–	–
495	50.4	51.0	49.2±0.3	–	–	–	–	–	–	–	–	–
586	–	–	–	–	–	–	–	–	46.1	47.0	41.8±0.6	–
605	–	–	–	–	44.3	43.5	42.0±0.6	–	–	–	–	–
613	43.3	43.0	42.6±0.3	–	–	–	–	–	–	–	–	–
736	–	–	–	–	–	–	–	–	40.3	43.0	35.6±0.5	–
814	–	–	–	–	36.2	34.0	35.0±0.6	–	–	–	–	–
$N_{\gamma>300}$	0.71	–	0.73	–	0.68	–	0.72	–	0.61	–	0.72	–

Table I shows the model and experimental values of $\gamma_{CO_2-N_2}(T)$ for three lines in the ν_3 band. Our data presented in Table I were calculated in the following way. We calculated the parameters $\gamma_{CO_2-N_2}$ based on the RATCF model for the temperature interval 300–500 K. This was followed by determining the temperature coefficients n_γ according to

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formula (1) (for the temperature interval T = 300–500 K the obtained parameters n_γ are denoted by $n_{\gamma>300}$ and for T = 100–300 K, $n_{\gamma<300}$) and the values of $\gamma_{CO_2-N_2}$ for all of the indicated values of T. Table I also shows the values of γ calculated from the Bonami–Robert model.[8] As can be seen, from Table I, for the lines $R(J_i$ = 38, 42, and 54) our calculated values of γ agree rather well with the experimental data of Refs. 9 and 10 and the calculations of Ref. 8. Table I gives the temperature coefficients n_γ, which have been determined based on our data and from experiment. The former differ insignificantly from the latter. However, in contrast to the experimental data, our n_γ are dependent of the rotational quantum number J_i. The lack of the experimental dependence $n_\gamma(J_i)$ can be attributed to a significant experimental error for the lines with $I_i \geq 42$, because with rise of the temperature from 300 to 800 K there occurs narrowing of these lines by a factor of nearly two, and this naturally leads to an increase in the experimental error when the values of γ are determined. Table II, alongside with our data, presents the experimental values of γ from Refs. 11–13. In spite of the fact that our values of $\gamma_{CO_2-CO_2}$ and $\gamma_{CO_2-O_2}$ agree well with experiment, the coefficients n_γ substantially differ in a number of cases which may be attributed to the accuracy of determining γ for each line. For example, according to Refs. 11 and 13 the experimental values of $\gamma_{CO_2-CO_2}$ for P(8) and R(15) lines at normal temperature (see Table II) differ by ~10%.

TABLE II. The values of half–widths (cm^{-1}·atm^{-1}) *and* $n_{\gamma>300}$ *for* CO$_2$ *in the* 001–100 (9.6 μ) *band.*

Mixture	T, K	P(8)	R(15)	P(20)	P(30)	
		0.113	0.107	0.106	0.096	Our results
		n_γ= 0.59	n_γ= 0.65	n_γ= 0.66	n_γ= 0.53	
	300	0.11	0.105	0.099	0.092	Ref. 13
		n_γ= 0.86	n_γ= 0.80	n_γ= 0.83	n_γ= 0.80	
		0.11	0.102	0.095	0.089	Ref. 11
		–	n_γ= 0.77	–	–	
CO$_2$–CO$_2$	296	0.121	0.114	0.109	0.098	Ref. 12
		n_γ= 0.87	n_γ= 0.85	n_γ= 0.74	n_γ= 1.02	
		0.135	0.130	0.129	0.114	Our results
	220	0.149	0.143	0.138	0.124	Ref. 12 with $\overline{n}_\gamma$= 0.86
		0.076	0.066	0.065	0.066	Our results
		n_γ= 0.58	n_γ= 0.54	n_γ= 0.61	n_γ= 0.71	
	300	0.071	0.067	0.063	0.06	Ref. 13
		n_γ= 0.81	n_γ= 0.74	n_γ= 0.76	n_γ= 0.79	
CO$_2$–CO$_2$		0.076	0.074	0.070	0.062	Ref. 12
	296	n_γ= 0.93 (8)	n_γ= 0.94 (11)	n_γ= 0.94 (7)	n_γ= 1.09 (7)	
	220	0.091	0.078	0.0787	0.082	Our results
		0.1047	0.0991	0.0933	0.0826	Ref. 12 with $\overline{n}_\gamma$= 0.94

Moreover, the experiments aimed at obtaining the coefficients n_γ, which have been carried out during recent years, for the most part were carried out at elevated temperatures (329–423 K according to Ref. 11, 200–300 K according to Ref. 12, 300–523 K according to Ref. 13, and 373–620 K according to Refs. 14 and 15). Comparison of our value of $\overline{n}_\gamma = 0.56$ (P(20) and P(30) lines near 9.6 μ at T = 300–500 K in a CO$_2$–CO$_2$ mixture) with other experimental data, $n_\gamma = 0.58$ (P(16) and P(32) lines at T = 373–620 K) from Ref. 14 and $n_\gamma = 0.56$ (P(16) line at T = 373–620 K) from Ref. 15, which have been obtained in nearly the same temperature intervals, also indicates that these parameters are close in values.

In accordance with the RATCF model, the relation for the temperature coefficient

$$n_\gamma = 1 + x_v - 2x_b(p - 2) \tag{2}$$

depends on the powers x_v and x_b which describe the temperature dependence of the velocity $v\left(v \sim T^{x_v}\right)$ and gas–kinetic distance $b_{gk}\left(b_{gk} \sim T^{x_b}\right)$. The parameter p describes the shape of the potential of intermolecular interaction ($V(r) \sim -C_p r^{-p}$). x_b is found from the analytic relation $b_{gk} = a\sqrt{1 + g/T}$, where the values of a and g are different for each type of molecules. The velocity index x_v for each state of molecule absorbing the radiation is determined from our collisional RATCF model.[1] Varanasi[5] has formulated a problem of obtaining the temperature dependence $\gamma(T)$ by integrating over all of the collisional velocities. In our opinion, the relation of n_γ to coefficient x_v may serve as an answer to this problem. For a specific gaseous mixture the parameter x_b is a constant quantity and in our case $x_b = 0.157$. Under the assumption that the quadruple–quadruple interaction is the basic intermolecular interaction for the CO$_2$–CO$_2$ molecular medium in the region of resonant collisions, i.e., $p = 5$, the temperature coefficient becomes dependent solely of the quantity x_v,

$$n_\gamma = 0.06 + x_v\ . \tag{3}$$

Simplified relation (3) shows that the parameter n_γ, for a specific medium, depends on two quantities: the rotational state J_i and the temperature interval ΔT.

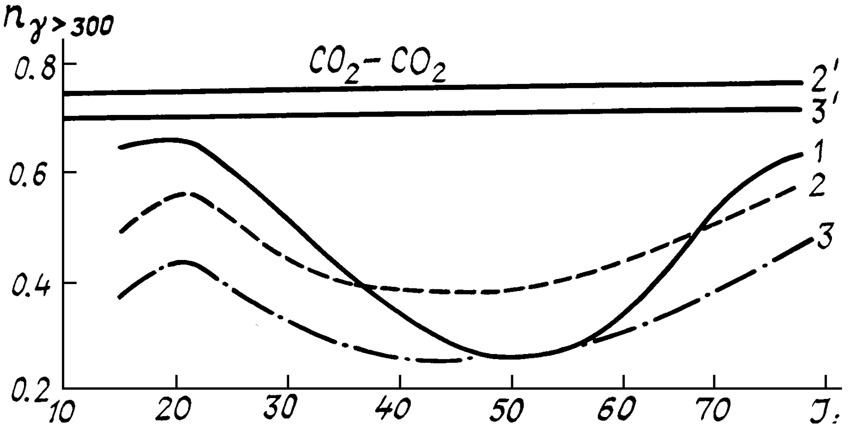

FIG. 1. The parameter $n_{\gamma>300}$ *as a function of the rotational number* J_i *for self–broadened* CO_2 *(R branch near* 9.6 μ*). 1) formula (1) of the RATCF model; 2) formula (2) with* V_{qq}*; 3) formula (2) with* V_{dis}*;* $n_\gamma = 0.5 + 1/(P - 1)$ *for curves 2' with* V_{qq} *and 3' with* V_{dis}*.*

The behavior of $n_\gamma(J_i)$ is shown in Fig. 1. At first glance, it is evident from the behavior of the curves (*1*, *2*, and *3*) that for any region of the quantum numbers J_i the resonant collisions of the molecules contribute mainly to the temperature dependence of the velocity of their motion ($\nu(T)$). When comparing these curves one can see that for $J_i < 20$, the nonresonant collisions with potential V_{qq} are predominant, because for these values of J_i curve *2* lies closer to curve *1* in comparison with curve *3*. This is followed for $J_i > 20$ by a transition to the resonant collisions with the same potential up to $J_i \approx 43$. When J_i further increases it can be seen that the potential V_{qq} is replaced by V_{dis} (curve *1* is superimposed on curve 3 $n_\gamma(V_{dis})$). In spite of the fact that for large $J_i > 65$ the basic intermolecular interaction type is V_{dis}, the values of n_γ given by formula (1) and shown by curve *1* significantly exceed those given by formula (2) and shown by curve *3*. Here most probably the nonresonant collisions start playing an increasing role, whose contribution to the broadening gives rise to such a behavior of curve *1* for large J_i. By comparing the curves, which have been obtained under the same assumptions (based on the formula

$$n_\gamma = 0.5 + 1/(p - 1) \qquad (4)$$

and formula (2)), we can see that formula (4) in some cases results, e.g., for the interval $43 < J_i < 65$, in a considerable overestimation of n_γ (by a factor of ~ 3 as can be seen from curves 2 and 2' in Fig. 1).

Comparison of the values of $\gamma_{CO_2-CO_2}$ for T = 220 K with values of n_γ, which we have obtained for T = 300–500 K, ($n_{\gamma>300}$) indicates that in the case of self–broadening our values of $\gamma(J_i)$ support the experimental data and, as a result, make it possible to extend the interval of the temperatures, for which our coefficients $n_{\gamma>300}$ preserve a predicted behavior. It should be noted that, because other experimental data are not available in Ref. 12, the values of γ (T = 296 K) were taken recalculated at T = 220 K with the help of the mean experimental value $\bar{n}_\gamma$.

For the same lines, for broadening by other molecules (CO_2–O_2) our calculated values of γ for the P(8, 20) and R(15) lines differ from those of Ref. 12 by 10–15%. The lack of experimental data on $\gamma_{CO_2-O_2}$ (J_i, T = 220 K) makes further analysis of $\gamma_{CO_2-O_2}$ (T = 220 K) more difficult.

TABLE III. The line half–width corresponding to the 0 → 1 *rotational transition of* CO *(*10^{-1},$cm^{-1}\cdot atm^{-1}$*).*

<table>
<tr><td colspan="6">CO–CO</td></tr>
<tr><td>T, K</td><td>Our results
$n_{\gamma<300}$= 0.66
$n_{\gamma>300}$= 0.74</td><td>Ref. 16
n_γ= 0.94
–</td><td>Ref. 17
–
–</td><td colspan="2">Ref. 18
–
–</td></tr>
<tr><td>300</td><td>1.04</td><td>–</td><td>0.89</td><td colspan="2">–</td></tr>
<tr><td>293</td><td>–</td><td>0.87</td><td>–</td><td colspan="2">–</td></tr>
<tr><td>195</td><td>1.43</td><td>1.32</td><td>–</td><td colspan="2">1.22</td></tr>
<tr><td>77</td><td>2.845</td><td>3.0</td><td>–</td><td colspan="2">2.4</td></tr>
<tr><td colspan="6">CO–N_2</td></tr>
<tr><td></td><td>Our results
$n_{\gamma<300}$= 0.55
$n_{\gamma>300}$= 0.70</td><td>Ref. 16
n_γ= 0.86</td><td>Ref. 19
–</td><td>Ref. 20
–</td><td>Ref. 21
–</td></tr>
<tr><td>300</td><td>0.83</td><td>–</td><td>0.84</td><td>0.80</td><td>0.821</td></tr>
<tr><td>293</td><td>0.84</td><td>0.83</td><td>–</td><td>–</td><td>–</td></tr>
<tr><td>200</td><td>1.04</td><td>1.16 (9)</td><td>1.21 (6)</td><td>–</td><td>–</td></tr>
<tr><td>190</td><td>1.07</td><td>1.22 (10)</td><td>–</td><td>–</td><td>–</td></tr>
<tr><td colspan="6">CO–O_2</td></tr>
<tr><td></td><td>Our results
$n_{\gamma<300}$= 0.54</td><td colspan="2">Ref. 16</td><td colspan="2">Ref. 20</td></tr>
<tr><td>300</td><td>0.75</td><td colspan="2">–</td><td colspan="2">0.713 (6)</td></tr>
<tr><td>293</td><td>0.76</td><td colspan="2">0.66 (1)</td><td colspan="2"></td></tr>
<tr><td>250</td><td>0.827</td><td colspan="2">–</td><td colspan="2"></td></tr>
<tr><td>200</td><td>0.933</td><td colspan="2">–</td><td colspan="2"></td></tr>
<tr><td colspan="6">CO–air</td></tr>
<tr><td></td><td>Our results
$n_{\gamma<300}$= 0.55</td><td colspan="2">Ref. 16</td><td colspan="2">Ref. 22</td></tr>
<tr><td>300</td><td>0.81</td><td colspan="2">–</td><td colspan="2">0.809</td></tr>
<tr><td>293</td><td>0.821</td><td colspan="2">0.75</td><td colspan="2">–</td></tr>
<tr><td>250</td><td>0.89</td><td colspan="2">0.92</td><td colspan="2">–</td></tr>
<tr><td>200</td><td>1.01</td><td colspan="2">1.08</td><td colspan="2">1.09</td></tr>
</table>

For the CO molecules we will start our analysis of the dependences of the line half–widths on temperature using the experimental data corresponding to the purely rotational transition 0 → 1. In spite of the fact that our temperature coefficients $n_{\gamma<300}$ are substantially different from those of Ref. 16, a good agreement of the parameters γ with experiment is observed at different temperatures (see Table III). The deviation of our values from n_γ published in Ref. 16 is associated first of all with the fact that our $n_{\gamma<300}$ have been obtained in the temperature interval 100–300 K, while in Ref. 16 the temperature changes from 220 to 293 K. Note that we have also considered the interval of the elevated temperatures 300–500 K, for which the values of $n_{\gamma>300}$ somewhat differ from $n_{\gamma<300}$ (see Table III). For the vibrational band 0–1 of CO broadened by N_2 and CO, we have calculated the parameters $\gamma(T)$ for the temperature interval 400–600 K based on formula (1) and our values of $n_{\gamma>300}$ and compared them with the available experimental values.[23,24] Just as previously, our values of $\gamma(T)$ agree well with experiment (see Tables IV and V). In the same band, but for large values of the rotational quantum numbers $J_i \geq 25$, for self–broadening Table V gives the values of $\gamma(T)$ and our coefficients $n_{\gamma>300}$ and $n_{\gamma<300}$. As one can see in Table V, these coefficients differ by 30% for some values of J_i. The values of $\gamma(T)$ by themselves, obtained with the help of formula (1) and $n_{\gamma>300}$ reproduce the experimental values of γ. For the $P(29)$ and $P(31)$ lines, it is pointless to test the dependence of $\gamma(T)$, because in the experiment of Ref. 25 for $J_i > 27$ the values of γ started to increase, even though for these J_i the contribution of the nonresonant collisions to the line half–width, as compared to the resonant interactions, must increase with increase of J_i. This conclusion is supported by the parameters $n_{\gamma>300}$, which, as shown in Fig. 1, started to increase after reaching a minimum (these are the P(18, 22, and 25) lines for CO–CO). Meanwhile, such a behavior of n_γ indicates that the contribution of nonresonant collisions increases.

TABLE IV. The temperature dependence of γ_{CO-N_2} *(*10^{-3}, $cm^{-1} \cdot atm^{-1}$*) in the* 0–1 *band and the parameters* $n_{\gamma>300}$.

T, K	399	501	601	–
$P^*(2)$ $n_\gamma = 0.67$	61.4	52.56	46.57	–
Ref. 23	61.2	53.3	49.5	–
T, K	400	500	601	–
$P^*(3)$ $n_\gamma = 0.66$	57.9	49.97	44.3	–
Ref. 23	56.0	48.7	43.1	–
T, K	360	436	525	600
$P^*(7)$ $n_\gamma = 0.74$	56.34	48.9	42.6	38.62
Ref. 23	52.3	46.7	41.0	37.3
T, K	401	499	600	–
$P^*(12)$ $n_\gamma = 0.67$	50.73	43.82	38.7	–
Ref. 23	46.2	40.2	35.2	–
T, K	406	465	582	762
$P^*(4)$ $n_\gamma = 0.65$	55.07	50.46	43.53	36.63
Ref. 23	56.0±0.5	50.0±0.5	43.2±0.4	35.7±0.9

*Our results.

TABLE V. The temperature dependence of γ_{CO-CO} *(*10^{-1}, $cm^{-1} \cdot atm^{-1}$*) in the* 0–1 *band.*

T, K	300	293	313	333	353	373	393	413
P^* (25)								
$n_{\gamma<300} = 0.43$								
$n_{\gamma>300} = 0.44$	0.45	–	0.43	–	0.42	0.41	0.40	0.39
Ref. 25	–	0.48	0.47	0.45	0.43	0.42	0.40	0.39
P^* (26)								
$n_{\gamma<300} = 0.44$								
$n_{\gamma>300} = 0.48$	0.44	0.45	–	0.42	–	0.40	–	0.38
Ref. 25	–	0.46	–	0.44	–	0.40	–	0.36
P^* (27)								
$n_{\gamma<300} = 0.45$								
$n_{\gamma>300} = 0.52$	0.44	0.44	–	0.41	–	0.39	–	0.37
Ref. 25	–	0.46	–	0.42	–	0.39	–	0.37
P^* (29)								
$n_{\gamma<300} = 0.42$								
$n_{\gamma>300} = 0.59$	0.43	–	–	–	–	–	–	–
Ref. 25	–	0.47	–	–	–	–	–	–
P^* (31)								
$n_{\gamma<300} = 0.49$								
$n_{\gamma>300} = 0.66$	0.41	–	–	–	–	–	–	–
Ref. 25	–	0.53	–	–	–	–	–	–

*Our results.

TEMPERATURE DEPENDENCE OF SPECTRAL LINE FOR CO_2 AND CO

Based on the same temperature law (formula (1)) we have examined the variations of the line centres shifts (δ) of CO_2 with rise or fall of temperature. As shown in Figs. 2 and 3, for the broadening by different molecules in different branches and bands the line shifts values decrease with rise of temperature. In addition, for broadening by an air molecule the shift values are less dependent of temperature as compared to the case of CO_2–CO_2 (compare the dashed and solid curves in Fig. 3). In the latter case, in different spectral intervals near 9.6 μ and in the ν_3 band the values of δ are much smaller (by a factor of ~ 1.5) with rise of temperature from 300 to 500 K. It can therefore be concluded that, for carbon dioxide when $T > 300$ K, one can choose a temperature, at which the line shape undergoes such transformations that the line becomes narrow and, in addition, a frequency shift is virtually no longer observed.

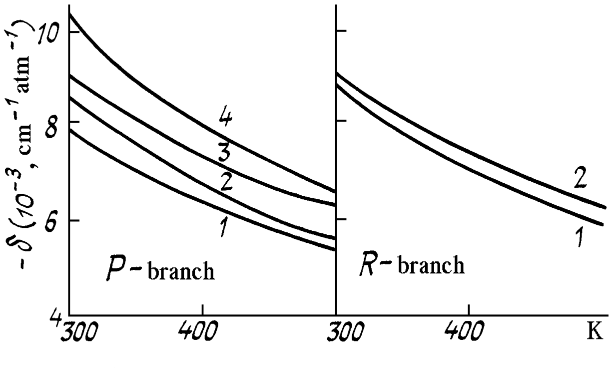

FIG. 2. The temperature dependence of the parameters $\delta_{CO_2-CO_2}$ *near* 9.6 μ. *P–branch:* $J_i = 8$ *(1),* 10 *(2),* 20 *(3), and* 30 *(4); R–branch:* $J_i = 10$ *(1) and* 18 *(2).*

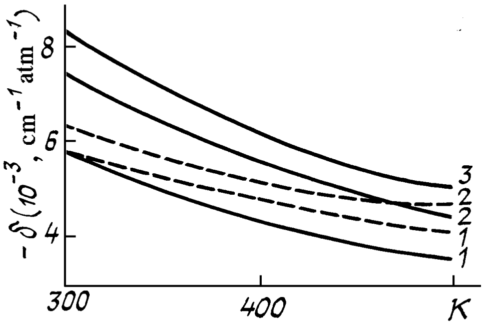

FIG. 3. The values of δ_{CO_2-CO} and air *as function of T in the* ν_3 band *(R–branch).* CO_2–O_2 *(solid) and* CO_2–*air (dashed);* $J_i = 36$ *(1),* 50 *(2), and* 60 *(3).*

For the molecular CO–CO medium the temperature coefficients given in Table VI show that the shift values may both increase and decrease at low temperatures.

TABLE VI. The values of δ_{CO-CO} *and the temperature coefficients in the* 0–1 *band corresponding to the* $0 \to 1$ *rotational transition (*10^{-3}, $cm^{-1} \cdot atm^{-1}$*).*

(J_i)	$R(0)$	$P(3)$	$P(4)$	$P(12)$	$P(18)$	$P(22)$	$P(27)$
δ	0.67	−0.92	−1.5	−1.8	−2.3	−3.0	−3.2
$n_{\delta<300}$	1.0	0.4	0.4	1.0	0.96	0.75	0.7
$n_{\delta>300}$	−0.3	0.58	1.37	1.3	0.90	1.2	0.92

In conclusion, it should be noted that comparison of our data with the experimentally observed half–widths of the spectral lines for the broadening by different molecules shows that the RATCF model reproduces well the temperature dependence $\gamma(T)$. Unfortunately, sufficiency of the experimental data on the dependence of the line center shifts on T for CO_2 and CO made it impossible to compare the values of $\delta(T)$ with our results.

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