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Research Article

The rotational–translational spectra of N_2 and CO_2 and their mixtures with argon

Abstract

Rotational–translational absorption spectra of carbon dioxide, nitrogen and their mixtures with argon in the wavelength range 20–190 cm^{-1} induced by pair collisions of molecules are obtained and analyzed. It is shown that, in contrast to absorption in noble gas mixtures, terahertz spectra of N_2 a CO_2 are mainly caused by quadrupole interactions, The role of the overlapping mechanism in the formation of these spectra was estimated, and the quadrupole moments of the N_2 and CO_2 molecules, equal to 1.5–1.7·10^{-26} CGSE and 4.25·10^{-26} CGS, respectively, were determined from the integrated intensity of the recorded spectra.

Keywords: terahertz spectra, collision induced absorption, quadrupole interactions, quadrupole moments of the N_2 and CO_2

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Introduction

The inclusion of N_2 and CO_2 molecules in the range of investigated objects makes it possible to approach the study of a wider class of interaction–induced spectra: rotational–translational spectra. In contrast to absorption in mixtures of noble gases,[1] the FIR N_2 and CO_2 spectra are mainly caused by quadrupole interactions, since the dipole moment of overlapping in the collision of two identical molecules, as well as of two identical atoms, is zero.

On the other hand, the difference between these spectra and the spectra of H_2 and its mixtures with Ar, Kr and Xe, also due to quadrupole interactions,[2] lies in the fact that in them the rotational part, due to the much smaller rotational constant of the molecules N_2 a CO_2 (B_{N2}=2cm^{-1}, Bco_2=0.4cm^{-1}, and B_{H2}=40cm^{-1}), will not be basically separated from the translational part of a, overlapping with it, forms one absorption band. The long–wavelength spectra of N_2 a CO_2 have been studied in detail in[3,4] the spectra of mixtures of these gases with Ar have been studied in less detail.[5]

The study of the spectra of their mixtures with argon, carried out in parallel with the study of the spectra of N_2 and CO_2, is interesting in that it allows us to evaluate the role of the overlapping mechanism in the formation of the rotational–translational spectrum.

Materials and methods

The recorded absorption spectra of N_2 a CO_2 and their mixtures with argon are shown in Figure 2–7. Spectra in the 20–180 cm^{-1} region were obtained using a vacuum long–wavelength spectrometer Hitachi FIS–21 with resolution of 1–2 cm^{-1}. The investigated mixture of gases was placed in a multi–way cell, collected according to the White scheme, with a base of 34.2 cm, which made it possible to obtain an optical layer of 6.9 m at 20 intersections of the working volume. The windows of crystalline quartz 6 mm thick were installed on a cuvette withstanding the pressure of 70 atm. Temperature regulation in the interval 135–300°K was carried out by changing the flow rate of cold nitrogen in the heat exchanger assembled on the casing. Thermal insulation was achieved by placing the cooled part of the cuvette in a vacuum.

The temperature was measured with copper–constantan thermocouple; its stability was not lower than + 2°. Gas purification was performed on a high–pressure unit assembled from absorbers with KOH and zeolite and low–temperature traps. The purity of the investigated gases was controlled by the absence of absorption in each component separately. General view of the experimental setup is shown in Figure 1.

The experimental conditions for pressures (p), density (ρ) and temperatures, and also (ν_{max}), (I_{max}) and the integrated intensities (A) of the recorded absorption bands are given in Table 1.

Table 1 The experimental conditions of the recorded absorption bands.

	T (^{0}K)	p (atm)	ρ (amagat)	ν_{max} (cm^{-1})	$I_{max}\cdot10^7$ (cm$^{-1}\cdot$am^{-2})	$A\cdot10^4$ (am$^{-2}\cdot$cm^{-2})
N_2	138	7.2 N_2	17.0 N_2[6]	88±5	75.0	5.65
	298	18.0 N_2	17.0 N_2[6]	110±10	50.0	6.45
N_2+Ar	138	3.9 N_2 12.6 Ar	8.0 N_2[6] 28.5 Ar[7]	82±5	55.0	5.25
	298	14.5 N_2 14.5 Ar	13.8 N_2[6] 13.5 Ar [6,4]	100±10	40.0	5.80
CO_2	215	3.5 CO_2	4.75 CO_2[8]	42.5	110.0	71.5
CO_2+Ar	215	1.75 CO_2 10.25 Ar	1.95 CO_2[9] 13.5 Ar[8]	47±5	37.0	18.5

An analysis of the errors in the measurement and processing of the measurement results showed that the error in determining the intensities of the recorded absorption bands does not exceed 10–15%.

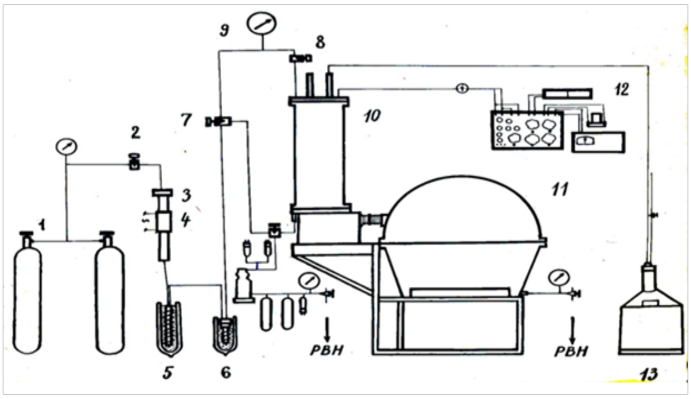

Figure 1 General view of the experimental setup. 1–10: cleaning system, 10: multi–way cuvette, 11: long–wave infrared spectrometer, 12, 13: temperature adjustment and measurement system. PBH: systems of evacuation of a cuvette and a spectrometer.

Results and discussions

The first thing that attracts attention when considering these spectra is that unlike the translational spectra of noble gases and mixtures of H_2 with Ar, Kr, and Xe,[2] practically the entire absorption band with a maximum and wings up to a level of 5–10% with respect to absorption at the maximum. An exception is the spectra of N_2 and N_2 + Ar at room temperature, whose high–frequency wing at the boundary of the investigated range had an intensity only half the intensity at the maximum.

The spectra of CO_2 and CO_2 with argon, because of the large quadrupole moment of the CO_2 molecule, are almost an order of magnitude stronger than the spectra of N_2 and N_2 +Ar. Spectra of pure N_2 and CO_2 more intense than the spectra of the mixtures with argon. While the intensity of the spectrum of the CO_2 + Ar mixture differs several times from the intensity of the CO_2 spectrum, the intensity of the N_2 + Ar spectrum differs insignificantly from the N_2 spectrum.

This fact reflects the difference in the polarizability of CO_2 and argon: $\alpha_{co2}=2.93\cdot10^{-24}cm^3$, $\alpha_{Ar}=1.63\cdot10^{-24}cm^3$, and the proximity of the polarizability of N_2 and argon ($\alpha_{N2}=1.74\cdot10^{-24}cm^3$). In this case, as in the case of mixtures of H_2 with Ar, Kr and Xe, we are dealing here with spectra induced by the electrostatic mechanism–quadrupole induction, but in contrast to the spectra of mixtures of H_2 with Ar, Kr, and Xe in the spectra of N_2 and CO_2 and their mixtures with argon, individual rotational lines will no longer be resolved and not because of the insufficient resolution of the spectrometer $\Delta\upsilon\left(\Delta\upsilon_{spect}=1-2\ cm^{-1}\right)$, but because these lines, for example, in the case of N_2 from each other by 8 cm^{-1},[10] will be significantly broadened by the translational effect. According to,[11] $\Delta\nu_J\cong 1/d\cdot\sqrt{(T/M)}$, where d is the collision diameter, M is the reduced mass of the colliding particles. Assuming the value of d to be proportional to the value of the parameter σ of the Lennard–Jones potential, it is easy to compare it with the width of the rotational lines of H_2: for nitrogen, $\Delta\nu_J=20\ cm^{-1}$, for carbon dioxide $\Delta\nu_J=20\ cm^{-1}$.

The temperature behavior of the spectra is in agreement with their assignment to the rotational–translational class induced by the quadrupole field. With decreasing temperature, the spectrum shifts to the low–frequency region in accordance with the redistribution of molecules along rotational levels. It should, however, be pointed out that the maxima of the recorded absorption bands, due to the asymmetric broadening of the rotational lines, by the translational effect, are shifted to the high–frequency region with respect to those predicted by the theoretical rotational spectrum. (In Figure 2–7, the theoretical rotational spectrum is shown by vertical lines). To learn from the spectra of further information, a computer calculation of the integrated absorption intensities was carried out. From the theory of rotational–translational spectra developed by Kiss & Van Kranendonk[12], it follows that the binary absorption coefficient of a mixture of diatomic molecules with monatomic

$$A_{rot.-tr.}=\beta\cdot\{L(J)\cdot[\lambda^2\cdot(I+4\pi\sigma^2\cdot I')+\mu^2\cdot(F+\frac{6mc\sigma^2}{i}\cdot F')+\lambda\cdot\mu\cdot(K+\frac{7.75m\sigma^2}{i}\cdot K'+D\cdot(\mu')^2\cdot F'\},\qquad(1)$$

where $\beta=\frac{\pi e^2\sigma^3 n_0^2}{3mc^2},\lambda=\frac{\xi}{e\sigma}\cdot\exp\left(-\frac{\sigma}{\rho}\right),\mu=Q\cdot\frac{\alpha}{e\sigma5},\mu'=\sqrt{\frac{88}{255}\cdot\frac{\gamma Q}{R^4}}$
,and $L(J)$ is the result of averaging over the rotational states of the molecule; I, I', F, F', K, K', and D are configuration integrals, similar to the integrals F, F' in expression (7); Q is the quadrupole moment of the molecule; α is the polarizability; γ–anisotropy of polarizability; i is the moment of inertia.

The first term in (1), proportional to the integral I–is the contribution of the overlapping mechanism, the second and third, proportional to the integrals F and F', are respectively translational and rotational contributions to the spectrum due to the quadrupole induction mechanism. The terms proportional to the integrals K and K' are respectively translational and rotational contributions to the interference spectrum between the quadrupole mechanism and the overlapping mechanism. Finally, the last term, in square brackets, is the term proportional to the integral F' is the contribution of the double transitions due to the anisotropy of the polarizability: $\Delta J_1=2$, $\Delta J_2=2$ or $\Delta J_1=2$, $\Delta J_2=0$.

Since the contribution to the band is due to absorption due to quadrupole induction, according to formula (2),

$$A_{rot.}(J)=\frac{48\pi^4\nu}{hc}\cdot Q^2\cdot\alpha^2\left\{\frac{p(J)}{2J+1}-\frac{p(J+2)}{2J+5}\right\}\cdot Z(J,J+2)\cdot\int_0^\infty R^{-6}\cdot exp\left[-V(R)/kT\right]dR,\qquad(2)$$

where p are the Boltzmann factors and Z is the Racah`s coefficients for the quantum numbers J, $V(R)$ is the pair interaction potential of the Lennard–Jones or Kihara type, which is obtained from (1) when only the electrostatic induction mechanism is taken into account, the relative intensities of the rotational lines of the branches S(J), O(J) and Q(J) are determined. (The branch O(J)–(transitions ΔJ=–2) corresponds to stimulated emission, which plays an important role in the formation of the total contour of the band at not too low temperatures, especially near zero frequencies. The branch Q(J)–

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(transitions ΔJ=0) gives a line at one frequency ν=0; however, due to broadening, its high–frequency wing must also be taken into account when calculating the total spectrum. In principle, the branch Q(J) corresponds to purely translational transitions). Each rotational line was then "broadened" on the PC and described purely formally, as in the case of spectra of mixtures of H_2 with Ar, Kr and Xe, a curve of the form:

$$\grave{A}(\nu) = \hat{A} \cdot \nu \cdot th\left(h\nu / 2kT\right) \cdot exp\,[-\sqrt{£^2 + \left(\frac{í}{í_0}\right)^2}\,], \qquad (3)$$

Proposed in Kouzov[13], The parameters of this contour *B*, *£*, and ν_0 were varied so that the experimental contour was combined with the calculated one in the best way. The values of *B*, *£*, and ν_0, together with the values of the integrated intensities found in the calculation, are given in Table 2.

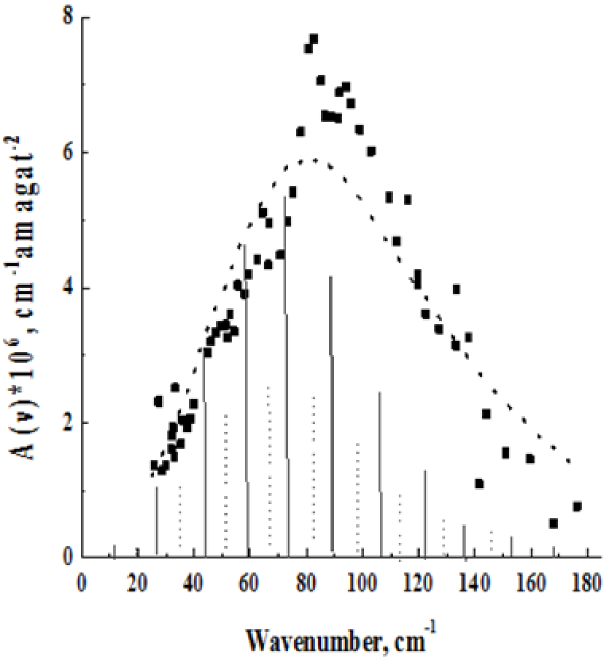

Figure 2 Rotational–translational spectrum N_2 at T=138⁰ K.

Having the values of the integral intensities now it is possible, if the quadrupole moments of the N_2 and CO_2 molecules are known, to assess the role of the induction mechanisms in the formation of the rotational–translational spectrum. For the N_2 molecule, the values of the quadrupole moments[1] obtained by different methods differ somewhat from each other.

So, from the quantum mechanical calculations, we obtain $Q_{N2} = 1.9 - 2.0{\cdot}10^{-26}\,CGSE$ [14] obtained from the second virial coefficient and from measurements of the spin relaxation of $Q_{N2} = 1.9 - 2.0{\cdot}10^{-26}\,CGSE$.[15,16] Intermediate values were found using data on induced spectra: on the vibrational–rotational spectrum of $H_2 + N_2$ at: room temperature $1.64.10^{-26}\,CGSE$;[17] on the rotational–translational spectrum of N_2 at room temperature– $1.47{\cdot}10^{-26}\,CGSE$,[11] $1.47{\cdot}10^{-26}\,CGSE$ [18] and $1.58{\cdot}10^{-26}\,CGSE$.[19]

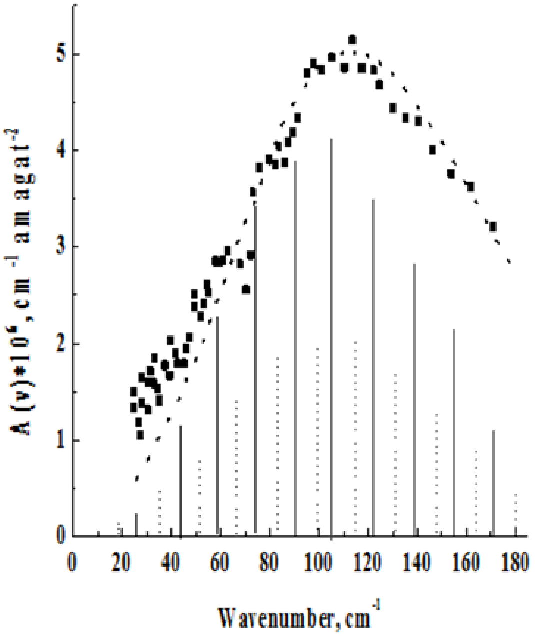

Figure 3 Rotational–translational spectrum of N_2 at T=298⁰ K.

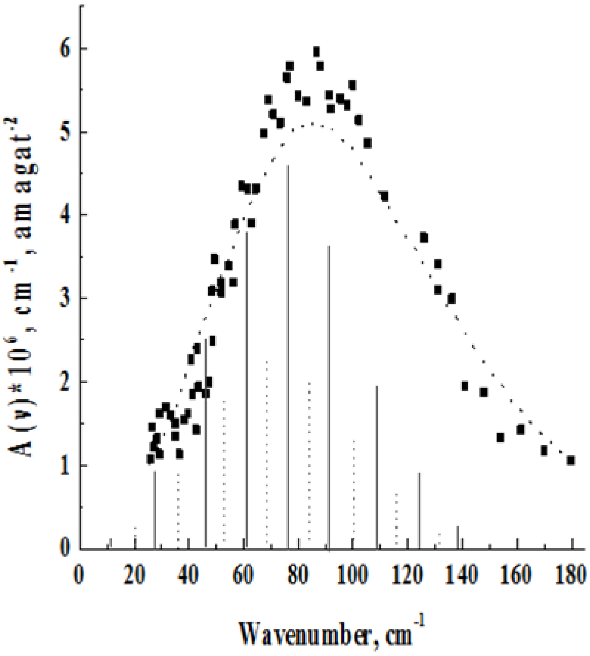

Figure 4 Rotational–translational spectrum N_2+Ar at T=138⁰ K.

We set ourselves the task, having set the induction mechanism, to determine the quadrupole moment of the N_2 molecules from the integrated intensity of the spectra recorded by us. We recall that

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[1] In this paper, the molecular quadrupole moment is determined by the expression:

$$Q = 1\ /\ 2\int r^2 \cdot \rho\ \left(r,\ \theta\right) \cdot \left[3cos^2\theta - 1\right]\ d\tau, \qquad (4)$$

where ρ is the charge density, r and θ are the polar coordinates with respect to the center of mass molecule (the polar axis is directed along the internuclear axis), and $d\tau$ is the volume element.

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according to (1), the following induction mechanisms take part in the formation of the rotational–translational spectrum. The first term in (1), which is proportional to the integrals *I* and *I'*, is the contribution of the overlapping mechanism. The second and third terms proportional to the integrals *F* and *F'* are, respectively, translational and rotational contributions to the spectrum due to the quadrupole induction mechanism. The terms proportional to the integrals *K* and *K'* are translational and rotational contributions of the interference between the quadrupole induction mechanism and the overlapping mechanism. Finally, the last term, in parentheses, is the term proportional to the integral *F'*, which is the contribution of the double transitions due to the anisotropy of the polarizability: $\Delta J_1=2$, $\Delta J_2=2$ or $\Delta J_1=2$, $\Delta J_2=0$.

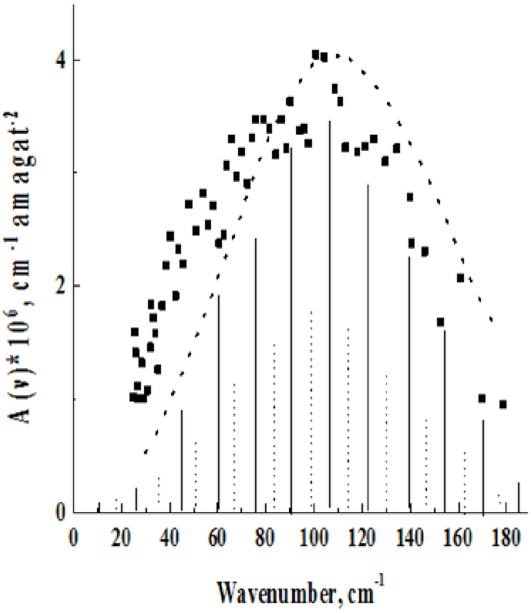

Figure 5 Rotational–translational spectrum of N_2 + Ar at T=298⁰ K.

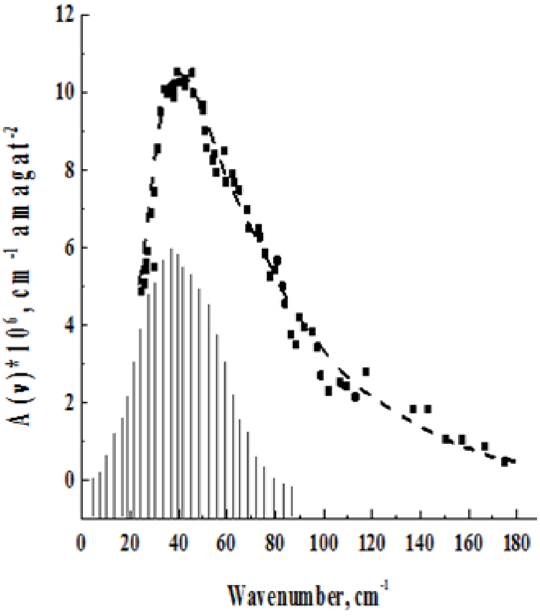

Figure 6 Rotational–translational spectrum CO_2 at T=215⁰ K.

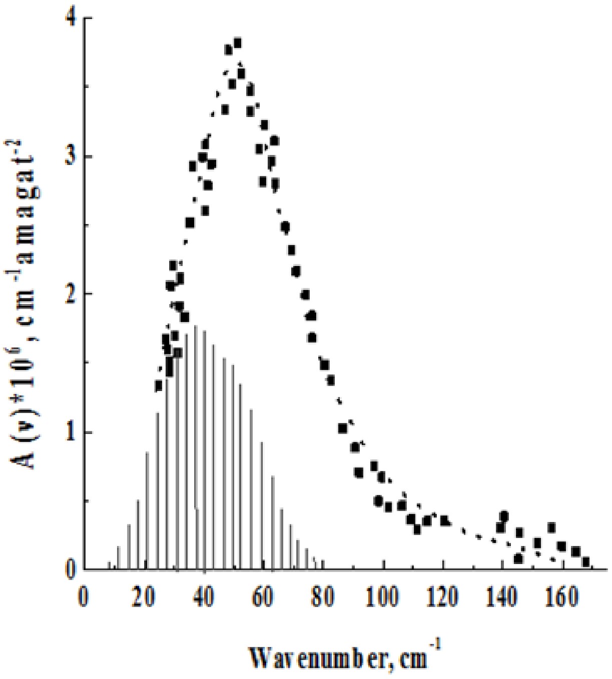

Figure 7 Rotational–translational spectrum of CO_2 + Ar at T = 215⁰ K.

In,[11,18,19] calculating the quadrupole moment of the N_2 molecule, it was assumed that the contribution of the overlapping mechanism to the spectrum of pure nitrogen is negligible. Indeed, in the collision of two identical particles, the coefficient characterizing the main part of the dipole moment of overlap is zero for symmetry reasons.

The contribution of transitions due to polarizability anisotropy to the total absorption intensity does not exceed, apparently, 1–2%. Thus, according to the data of,[3] in the induced rotational spectrum of CO_2, the polarizability anisotropy of which is larger than for N_2, only 5% of the intensity is associated with these transitions.

Neglecting the contribution of the overlap mechanism and the contribution of the transitions due to the anisotropy of the polarizability, from (1) we obtain the following expression for the binary absorption coefficient of the rotational–translational band:

$$\grave{A}_{1,rot.-tr.} = \frac{\pi Q^2 \alpha^2 n_0^2}{c^2} \cdot L \left[\frac{F}{3m\sigma^7} + \frac{2F'}{i\sigma^5} \right] \qquad (5)$$

Hence, knowing the integral intensity of the N_2 spectrum, we can determine Q_{N2}. Using the integrals *F* and *F'* calculated with the Lennard–Jones potential, tabulated,[12,20] we obtain from the intensity of the N_2 spectrum at room temperature: $Q_{N2} = 1.7 \cdot 10^{-26}\,CGSE$, the intensity of the N_2 spectrum at T = 138⁰ K is $Q_{N2} = 1.5 \cdot 10^{-26}\,CGSE$.

In determining Q_{N2} from the spectra of the N_2 + Ar mixture, the assumption of a negligible contribution of the overlapping mechanism should apparently remain in force: in the case of collision of N_2 with Ar, practically isoelectronic particles interact. If we use the correlation we found between the difference between the polarizabilities of the colliding atoms and the dipole moment parameter λ, from the graph of Figure 6 of the article[21] with $\Delta\alpha = 1.74(\mathring{A})^3 - 1.63(\mathring{A})^3 = 0.11(\mathring{A})^3$

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, we can obtain $\lambda = 0.1 \cdot 10^{-4}$. The intensity calculated for this λ is no more than 1% of the recorded absorption[2]. Thus, if the overlapping mechanism is neglected, using equation (4) for the intensity of the N_2 + Ar spectrum at room temperature, we obtain $Q_{N2} = 1.6 \cdot 10^{-26}\,CGSE$, and from the intensity of the N_2 + Ar spectrum at $T = 138^0 K - Q_{N2} = 1.4 \cdot 10^{-26}\,CGSE$. These values are close to those obtained by other methods.

Moreover, the values determined from the low–temperature spectra of N_2 and N_2 + Ar agree better. A few overestimated values, found from spectra recorded at room temperature, seem to reflect the effect of absorbing impurities, which could not be eliminated during nitrogen purification. If we assume that the assumption of the predominant role of the quadrupole induction mechanism in the formation of the rotational–translational spectrum is also valid in the case of CO_2, calculation by formula (4), analogous to the calculation of Q_{N2}, leads to the following Q_{CO2} values: from the induced spectrum of pure $CO_2 - QCO_2 = 5.9 \cdot 10^{-26}\,CGSE$, from the spectrum of a mixture of CO_2 with argon $-Q_{CO_2} = 5.6 \cdot 10^{-26}\,CGSE$.

These quantities, generally speaking, are close to those obtained by other authors from the rotational–translational spectrum of CO_2: $6.6 - 8.2 \cdot 10^{-26}\,CGSE$;[22] $6.6 - 5.2 \cdot 10^{-26}\,CGSE$,[3] but slightly differ from the most accurate of the currently known values of $Q_{CO2} = 4.5 \cdot 10^{-26}\,CGSE$.[23]

To clarify our calculation of Q_{CO2} in terms of the intensity of the spectrum of the CO_2 + Ar mixture, we took into account the fact that this spectrum is due to its origin in addition to quadrupole induction, also the induction of overlap. Indeed, in contrast to the interaction of two isoelectronic particles of type N_2 and Ar in the collision of CO_2 and Ar, the isotropic part of the dipole moment of overlap is obviously not equal to zero. Evidence for this can be found in Bar-Ziv[24], in which a high–frequency wing from the center of a mixture of CO_2 + He was investigated at room temperature. It shows that in the region of 250 cm^{-1}, the binary absorption coefficient of the CO_2 + He mixture is only 3 times lower than in the pure CO_2 spectrum, whereas in the case of the small contribution of the overlapping mechanism it must differ in $\left(\alpha_{Ñî\,2} \middle/ \alpha_{Íå}\right) = (12.5)^2$ times. The intensity of the rotational–translational spectrum with allowance for the contribution of the induction of overlap is determined by the expression (1). From it, roughly estimating the graph of Figure 6 of the previous work, the parameter $\lambda = 2.10^{-4}$ and using the integrals *I*, *I′*, *F*, *F′*, *K* and *K′* tabulated in Poll et al.[21] and,[12] we have: $Q_{Ñî\,2} = 4.8 \cdot 10^{-26}\,CGSE$.

Table 2 The integrated intensities of spectra.

Spectra (Parameters of the contourof the line in equation (3))	**N_2 (T=138⁰K)**	**N_2 (T=298⁰K)**	**N_2+Ar (T=138⁰K)**	**N_2+Ar (T=298⁰K)**	**CO_2 (T=215⁰K)**	**CO_2+Ar (T=215⁰K)**
$Â \cdot 10^5$	3,3	4,2	2.9	4,0	400,0	103,4
£	1,0	0,7	1,0	0,3	1,0	1,0
ν_0	21,6	25,7	23,5	25,3	13,2	25,0
$A \cdot 10^4\ (am^{-2} \cdot cm^{-2})$	5,65	6,45	5,25	5,80	71,0	18,6

To correct the calculation of the Q_{CO2} value from the intensity of the rotational–translational spectrum of pure CO_2 as the interaction potential, instead of Lennard–Jones, the potential of Kihara was used:

$$V(R) = 4\varepsilon\{[(\frac{\sigma - 2a}{R - 2a})^{12} - (\frac{\sigma - 2a}{R - 2a})^{6}]\} \tag{6}$$

with the following parameters: $\varepsilon = 441.7^0\,Ǻ;\ \sigma a = 3.72\ Å;2\ = 1.46$,[26] which, as was shown in Kihara[27] & Datta et al.[28] more precisely than the Lennard–Jones potential, conveys the features of the interaction of molecules of similar CO_2.

When the Lennard–Jones potential G(R)=exp [–V(R)/ kT] is replaced by the Kihara potential, the integrals *F* and F^1 in expression (4) become:

$$F = 336\pi \int_0^\infty x^{-8} \cdot exp\left[\frac{\left(\frac{1-b}{x-b}\right)^6 - \left(\frac{1-b}{x-b}\right)^{12}}{0.25\,T^*}\right]dx,$$
$$F^1 = 12\pi \int_0^\infty x^{-6} \cdot exp\left[\frac{\left(\frac{1-b}{x-b}\right)^6 - \left(\frac{1-b}{x-b}\right)^{12}}{0.25\,T^*}\right]dx, \tag{7}$$

where $x = R / \sigma$; $b = 2a / \sigma$.

Calculating them on a PC and substituting in (4), we have obtained $Q_{CO2} = 4.25 \cdot 10^{-26}\,CGSE$.

Conclusion

Such results indicate the prospects of studying the rotational–translational spectra of simple molecules for determining their

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[2] Recall that the correlation between λ and *Δα*, shown in work[21] was established from the translational spectra of mixtures of He and Ar, Kr and Xe, and we use it here only for estimating calculations. In the calculations, $\lambda_{isotropic}$. and $\lambda_{anisotropic}$. relied on the basis of the data of ,[25] are the same.

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molecular constants. The rotational–translational spectra also undoubtedly contain information about the dynamics of molecular interactions and the intermolecular potential.

The laboratory study of such spectra is also important because induced infrared absorption plays an important role in controlling the energy balance of the terrestrial atmosphere.[29,30]

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None.

Conflict of interest

Author declares there is no conflict of interest.

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