# Calculation of carbon dioxide absorption spectra in wide spectral regions

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A somewhat unexpected hypothesis of the Fermi distribution of photon fluctuations connected with a two-level system is chosen to explain the exponential shape of the absorption line profile in its far wing. A preference is given to studies of the properties of direct interaction between radiation and a matter being in chaotic thermal motion, rather than to the influence of intermolecular interaction on the line shape. A simple exponential form for the far wings of absorption lines is obtained. Examples of modeling absorption in wide spectral regions from 442 to 9648 cm $^{-1}$  are given for both pure  $CO_2$  and  $CO_2$ –Ymixtures, where Y is one of the buffer gases: N2, H2, He, Ar, or Xe.

# 1. Introduction to the problem and a brief review

The sub-Lorentz character of absorption in the far wings was discovered for many molecular species; for example, it was studied for CO2 by Winters, Silverman, and Benedict (see Ref. 1 and references therein). Experimentally, this problem was thoroughly studied by Burch et al.,2 for the CO2 molecule in a wide spectral region of 2400-7110 cm<sup>-1</sup>. In Ref. 1, the far wing profile is described in the exponential form, and at that time there were no one theoretical explanation to this behavior.

Many investigators (see, for example, Refs. 3-7) used the exponential form in various applications and semi-empirical processing in calculating absorption within atmospheric transmission windows or in the far wings of molecular spectral lines with sub- and super-Lorentz behavior. There exist numerous early (Refs. 8-17) and recent (Refs. 18-21) theoretical explanations of the exponential shape or the fast drop of absorption in the far wings. The theoretical papers mentioned above employed various approaches and ideas, including the hypotheses on random collisions and frequency fluctuations, which led to Gaussian profile<sup>9</sup>; perturbation theory, that yielded the so-called generalized line profile  $^{12-14}$  with the exponential behavior in the wings; kinetic equations and the method of relaxation Fano matrices, which allowed for the line interference 10,11,15,16 or the effect of line mixing; Fano method<sup>22</sup> and the Rosenkranz quasistatistical theory, <sup>23</sup> which also led to interference 19-21 in the far wings; the idea of the finite duration of collisions, 24 which resulted in the exponential spectral dependence as well. 15,16 All these approaches can be divided into two groups, one of which adjusts the profile of an individual line, for example, CKD-function<sup>7</sup> or the generalized line

profile, 12-14 whereas the other lead to mixing or interference of lines in the far wings.

Besides Refs. 7 and 12-14, the first group includes papers by Tvorogov, Nesmelova, Rodimova, and Fomin, 25-29 which, in fact, justify the modified (generalized) profile of an individual line and consider numerous examples of its application. Certainly, the references mentioned above do not cover the entire list of papers, which are explicitly or implicitly connected with this problem. Nesmelova, Rodimova, and Tvorogov in Ref. 28 (see also Refs. 25 and 29) used the method of spatial dispersion in the correlation function that forms the line profile. To describe complex spectra of various molecules in a uniform way, Fomin<sup>27</sup> and Nesmelova, Rodimova, and Tvorogov<sup>28</sup> had to determine the temperature and frequency dependence of the effective intermolecular potential, which enters the exponent. Clear physical meaning of this exponential form as a statistical Gibbs (Boltzmann) exponent can be found in Ref. 29.

It is undoubtedly that the allowance for the statistical intermolecular interaction in a random medium is an important element in the theory of line profile. However, in my opinion, the formation of a line shape is largely caused by local nonequilibrium processes; and the nonequilibrium statistical theory is still far from its completion (see, for example, Ref. 30). It is quite likely that it will give a simple result as applied to elementary systems at the quantum level.

The electromagnetic field should likely be included in the consideration, and even nonlinear processes occurring at its propagation may occur in atmospheric windows at low intensities.  $^{31,32}$  In  $_{2}$ O molecule, high nonlinear absorption may lead to a huge super-Lorentz effect<sup>31</sup> in the IR spectra. One can assume that in CO2 it is much lower, as this molecule demonstrates the properties of the sub-Lorentz profile and the well known problems of the far wings.

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In this paper, we contest against the generally accepted concept that the dispersion (Lorentz) profile does not suit the description of molecular absorption because of the super- and sub-Lorentz behavior. The Lorentzian plays an important role in the theory of the line profile. For example, the profile from Ref. 1 is a product of the exponential and Lorentzian profiles. The super-Lorentz absorption by water vapor was obtained with the use of a nonlinear technique, which deals with a product of the Lorentzian factors. <sup>31,32</sup> Moreover, the many attempts to calculate the fast sub-Lorentz drop based on the Rosenkranz method <sup>22</sup> are in fact the method of obtaining the modified Lorentz profile.

For more than four decades, various papers (some of them are mentioned here) put forward numerous contradictory ideas, but the problem of exponential wings formulated in Ref. 1 is still open in our opinion. In this paper, a different approach to this problem is proposed. We introduce the hypothesis of background fluctuations connected with an elementary two-level system and use nonlinear absorption, but unlike Refs. 31 and 32, we take into account the effect of far wings. Simulation of the absorption coefficient is based on the results of experimental researches 1,2,33 of the absorption by CO2 in mixtures with various buffer gases<sup>1,2,33</sup> in different spectral regions.<sup>2</sup> The attempts to solve the problem of global description of CO<sub>2</sub> absorption in 4.3, 2.7, and 1.4-µm transmission windows have earlier been undertaken in Refs. 12-14, 27, and 28. The explanation of narrowing of the line profile at high pressure is given based on these ideas in Ref. 34.

### 2. Exponential shape of line wings

It is believed that the system of the electromagnetic field propagating through the matter and the matter itself undergo fluctuations and that makes up the subject of the study in statistical mechanics and thermodynamics of the equilibrium and nonequilibrium systems. Further, we are interested in possible photon fluctuations connected with the elementary two-level molecular system. These fluctuations are thought to be characterized by saturation and it is assumed that no more than N photons can be related to a given two-level system. Assume also that the number of photons less than N cannot create fluctuations with the properties needed for interaction with the two-level system. Thus, possible fluctuations stimulating transitions in a two-level system obey the Fermi statistics in the case of thermal equilibrium with thermostat.

A set of particles with the Fermi statistics is characterized by the Fermi level, i.e., the state, in which no thermal perturbations occur when the absolute temperature approaches zero. Suppose that thermal perturbations are not observed under these conditions for a monochromatic radiation with the frequency  $\omega$  equal to the resonance frequency  $\omega_i$  of the two-level system i. If the radiation has the frequency  $\delta \omega_i = \omega - \omega_i = \left| \delta \omega_i \right|$ , then for its interaction with the

two-level system the effect of thermal perturbation with the distribution

$$\overline{n_i} = 1/[\exp(Nhc|\delta\omega_i|/kT) + 1)], \tag{1}$$

is needed, if  $\omega > \omega_i$ . For the case  $\omega = \omega' < \omega_i$ , thermal perturbations are distributed by the law

$$\vec{n_i} = 1 - \vec{n_i}. \tag{2}$$

We use standard designations in Eqs. (1) and (2): h is the Plank's constant, c is the speed of light in vacuum, k is the Boltzmann constant, T is the absolute temperature.

Trapping of photons by the two-level system (detailed discussion can be found in Ref. 34) occurs at absorption with the probability proportional to distributions (1) and (2). For the transition to normalized distribution function in Eqs. (1) and (2), we should introduce the coefficient 2, and the factor in absorption for a line should be represented as  $2\bar{n}_i$  and  $2(1-\bar{n}_i)$ , respectively, for the right-  $\omega > \omega_i$  and left-hand side  $\omega < \omega_i$  wing of the line i.

According to the conclusion  $^{35}$  by Landau and Lifshitz, the partition function  $\Omega_i$  with the variable number of particles  $n_i$  in the state i is determined as

$$\Omega_i = -kT \ln \sum_{n_i} \left\{ \exp \left[ (\mu_i - \varepsilon_i) / kT \right] \right\}^{n_i}.$$
 (3)

The energy of the system  $\varepsilon_i$  in the state i is expressed through the number of the fluctuating photons, i.e.,  $\varepsilon_i = Nhc\omega$  and, as the chemical potential of creation of a fluctuation of N photons calculated per one fluctuation,  $\mu_i = Nhc\omega_i$ . Equation (1) can also be obtained from Eq. (3).

For illustration, the known graphical representation of the Fermi and anti-Fermi factors  $(\overline{n_i})$  and  $(\overline{n_i})$  is shown in Fig. 1a. The solid curve in Fig. 1b shows the distribution of thermal perturbations for various frequencies of the detuning. The right-hand side wing of the solid curve can be interpreted as the presence of "partial" thermal perturbations in the system, and the left wing can be interpreted as the presence of "hole" perturbations. The dashed portion of the curve in Fig. 1b is to be interpreted. For the resonance frequency  $\omega = \omega_i$ , the segments of the solid and dashed curves in Fig. 1b are equal. Here, we can assume, as usually, that induced absorption and emission of photons in a twolevel system, which is connected with the N-photon fluctuation, occur simultaneously. It is reasonable to assume, using the terminology of the ray optics, as one of the possible versions, that the dashed portion of the curve describes deflection (refraction) of the electromagnetic waves. It is known that molecular refraction in the matter is caused by the polarization of the matter, and, according to Ref. 32, the so-called refraction function can deal with the variation of photon density in cavities of the matter.

Thus, if the fluctuation of N photons is connected with the ith two-level system, then  $n_i$  in Eq. (3) equals unity and the solid and dashed curves in Fig. 1b are

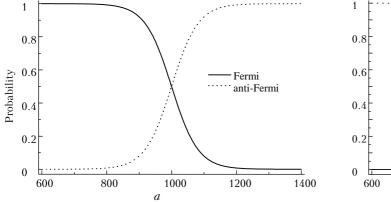
connected, respectively, with the absorption and refraction functions so that for any frequencies these two functions add up to unity. It is clear that normalization of these functions (see Fig. 1b) is not connected with the total number of photons in the selected volume, because our discussion deals only with averaging over a certain type of photon fluctuations connected with the two-level system i. The Fermi level as an upper populated energy level also makes no sense here.

The distribution (1) could be interpreted using the statistical theory of line profile, if the parameter N in Eq. (1) is equal to unity, what is not always the case. For far wings, Eqs. (1) and (2) degenerate into an exponential factor. In this form, in contrast to Refs. 28 and 29 mentioned above, it was applied in Ref. 33, and in Ref. 36 it was used as a factor in the dimer theory of continuum absorption by water vapor. All the results obtained using this exponential factor demonstrate good reconstruction of the temperature dependence of radiation absorption by, for example, dioxide. 19,21,28,29

In contrast to Refs. 25–29, we can avoid using the temperature-dependent effective intermolecular potential to describe all versions of the absorption by

various gases in the presence of buffer gases, since in this approach the term with nonlinear absorption allows for this circumstance. The numerator in the argument of the exponent (1) is independent of temperature and the type of molecules. The experimental data by Dokuchaev et al.<sup>33</sup> partially confirm the latter conclusion, because the logarithm of absorption by carbon dioxide in mixtures with various inert buffer gases in the atmospheric transmission windows is presented by parallel curves.

This hypothesis can be checked most easily in studying the temperature dependence of the absorption by carbon dioxide in a mixture with helium beyond the bands  $v_3$  and  $3v_3$  (Fig. 2b). In contrast to pure carbon dioxide (Fig. 2a), its mixture with helium does not exhibit complex negative or positive temperature dependence, as is seen at different spectral segments of the curves (see Fig. 2a). The negative temperature dependence is observed in the near wings, where the contribution of nonlinear absorption is significant. In the far wings, the temperature dependence is determined by the exponential factor in Eqs. (1) and (2) and it is positive. Quite complicated temperature dependence for pure  $CO_2$  was observed in Ref. 37.



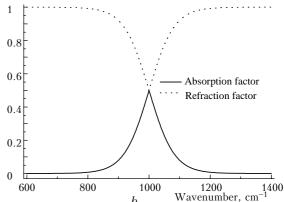
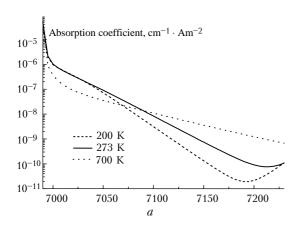


Fig. 1. Fermi (solid curve) and anti-Fermi (dashed curve) distributions (Eqs. (1) and (2), respectively) (a); absorption and refraction factor functions (b). Isolated line centered at  $\omega_i = 1000 \text{ cm}^{-1}$ , parameter N = 5, T = 294 K.



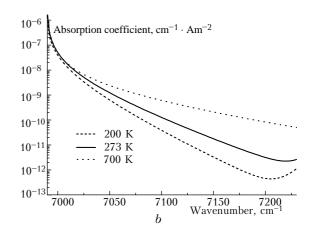
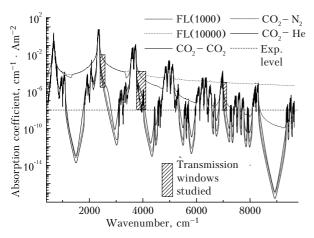


Fig 2. Calculated  $CO_2$  absorption spectra for three values of temperature beyond  $3v_3$  band at  $7000 \text{ cm}^{-1}$  (1.4  $\mu$ m): self-broadening (a) and broadening by helium pressure (b).

# 3. Calculations in different spectral regions

The absorption by carbon dioxide calculated for its mixtures with different buffer gases, He,  $N_2$ , and  $CO_2$  is given in Fig. 3 in comparison with the data calculated by Lorentz profile. The spectral region is determined only by the data available from GEISA<sup>38</sup> or HITRAN<sup>39</sup> databanks used. This is the first attempt to calculate absorption in that wide spectral region. Only relatively narrow spectral regions labeled in Fig. 3 were studied intensely. Many of these regions are inaccessible for observation, because measurements have to be conducted at high pressure due to weak signal, and the pressure-induced absorption largely overlaps other regions. Consequently, extrapolation of the spectrum to these regions (see Fig. 3) is of particular interest.



**Fig. 3.** Spectra of  $CO_2$  in the region of  $442-9648 \, \mathrm{cm}^{-1}$  ( $T=296 \, \mathrm{K}$ ). Two spectra with the Lorentz profile are calculated with line wings of 10000 and 1000  $\, \mathrm{cm}^{-1}$ 

The agreement between the calculated measured data confirms that the shape of the line profile proposed is quite reasonable. It is especially interesting that the absorption by carbon dioxide in the mixture with helium is well described in the mentioned transmission windows (Figs. 4 and 5), if the value of Nas a semi-empirical parameter in Eqs. (1) and (2) is equal to five. Besides this parameter, according to the discussion in Ref. 34, we also introduced the factor 1/20 in calculating the far wings of lines with the Lorentz profile and ordinary collisional halfwidth. The halfwidth for the mixture of  $CO_2$  with helium was chosen as  $\gamma_{\rm CO_2-He}$  = 0.52  $\gamma_{\rm CO_2-N_2}$ ; this value almost coincides with the measured value  $\gamma_{He} = 0.59 \; \gamma_{N_2}$ (Ref. 2). The halfwidths  $\gamma_{N_{2}}$  were taken from the GEISA-97 database. <sup>38</sup>

The exponential shape of the line profile likely follows from the properties of interaction of radiation with the matter being in thermal motion and, to a lower degree, it depends on the intermolecular interaction. Therefore, the line profile considered here

keeps its main properties in the ideal atomic or molecular gases as well. This conclusion is confirmed by analysis of experimental data reported by Dokuchaev et al.,  $^{33}$  because the exponential wings of absorption lines are represented by parallel curves for the mixtures of  $CO_2$  with different inert buffer gases (see Fig. 4). Deviations were noticed in the mixtures with diatomic molecules of nitrogen  $N_2$  and hydrogen  $H_2$  (see Fig. 4), which can be explained by additional hypothetical nonlinear absorption almost absent in mixtures with the atomic inert gases.

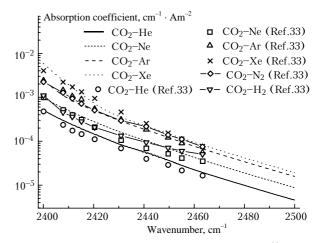
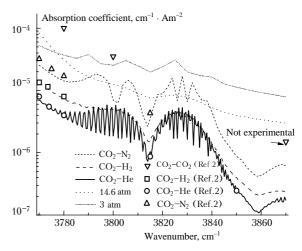


Fig. 4. Comparison of calculated and measured<sup>33</sup> data. Absorption of  $CO_2$  in mixtures with inert gases was calculated with the exponential shape of the curve with the exponent  $5Nhc\delta\omega_i/kT$ .



**Fig. 5.** Reconstruction of the absorption in mixtures  $CO_2$ –Y, where Y is a buffer gas (He, H<sub>2</sub>, N<sub>2</sub>, or  $CO_2$ ) in the region of 3780–3870 cm<sup>-1</sup> near the 2.7- $\mu$ m band.

In spite of that simple shape of the far wings, absorption in  $\mathrm{CO}_2$ —He mixture is well reconstructed in all windows (see Fig. 3). This, in general, confirms the approximation of an isolated line to be quite reasonable. For other inert buffer gases, only the collisional halfwidth in the Lorentz profile is to be changed, and we can state that absorption in the far

wings is described with the use of one additional parameter  $\zeta$  in the equation  $\gamma_{\text{CO}_2-\text{Y}} = \zeta_{\text{Y}}\gamma_{\text{CO}_2-\text{N}_2}$ . The values of this parameter are tabulated below along with the experimental data obtained in Ref. 2 and determining the Lorentz halfwidth in the central part of the profile.

Coefficients  $\zeta_{\rm Y}$  entering the equation  $\gamma_{\rm CO_2-Y}$  =  $\zeta_{\rm Y}$   $\gamma_{\rm CO_2-N_2}$ for calculation of absorption by the Lorentz profile in the central part and far wings. Experimental values<sup>2</sup> correspond to the resonance region. The Lorentz halfwidth  $\gamma_{\mathrm{CO}_2-\mathrm{N}_2}$  has the generally accepted nonlinear dependence on pressure

Y	Far wings	Ref. 2
Xe	2.2	_
$CO_2$	2.7	1.2
$ \begin{array}{c} \operatorname{CO}_2\\ \operatorname{N}_2 \end{array} $	1.7	1.0
Ar	1.8	0.78
$O_2$	_	0.82
$H_2$	1.0	1.41
Ne	1.0	_
Не	0.52	0.59

It is interesting to note that the parameters  $\zeta_Y$  for Y = He,  $N_2$ ,  $CO_2$  in the far wings relate to each other as square experimental values determined in the central part of the profile. These relations for the mixture CO2-CO2 were studied in other transmission windows (Figs. 5 and 6), where the calculated and measured absorption coefficients are in a reasonable agreement as well. Such a dependence is, in our opinion, indicative of significantly nonlinear processes, that could occur in forming the profile caused by the far wings,<sup>34</sup> because this regularity can hardly be a result of triple or multiple-particle collisions at a relatively low pressure. Similar regularity was found by Burch et al., 40-42 as a relation between the values of continuum absorption in pure water vapor and its mixture with nitrogen. The self-broadening halfwidth for water vapor is 3 to 5 times larger than the halfwidth for broadening by the nitrogen pressure, whereas for the continuum absorption this ratio is 13 to 25, i.e., it increases roughly as the square of the halfwidth.

Absorption by CO<sub>2</sub> in mixtures with buffer diatomic molecules CO<sub>2</sub>, N<sub>2</sub>, H<sub>2</sub> includes possible additional nonlinear absorption, which, on the one hand, is small as compared with that in water molecule and allows the far wings to manifest themselves. On the other hand, it explains the excess absorption as compared with the parallel behavior of the curves (see Fig. 4). At the same time, its parameterization is insufficiently unambiguous, and, for example, Ref. 43 considers possible dependence of this absorption on the active gas pressure in a more detail, what changes the parameterization. Unlike the calculations for water molecule,<sup>31</sup> good reconstruction of the absorption is observed for nonlinear interaction with lower resolution achieving tens reciprocal centimeters. In Ref. 31 it was several reciprocal centimeters for water molecule.

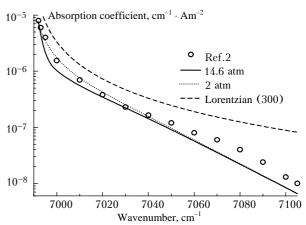


Fig. 6. Reconstruction of absorption by CO2-CO2 in the region of 6995–7110  $cm^{-1}$  beyond the 1.4  $\mu m$  band for two values of pressure: 2 and 14.6 atm.

## Conclusion

The exponential drop of the absorption in the far wings is explained to be the result of thermal equilibrium of possible photon fluctuations connected with a two-level system. As a result, these ideas on the thermal effect are close to those in Refs. 19, 21, and 25-29, where the temperature dependence of absorption in the line wings was reconstructed well when considering thermal factors in the theory. Simplicity is achieved in description of spectra of inert gases with the parameter N equal to five; this value is an empirical fact. We did not need to introduce the effective temperature-dependent intermolecular potential, because we used the idea of existence of nonlinear absorption,<sup>31</sup> responsible for the corrections in the absorption by  $CO_2$  in mixtures with such complex molecules as buffer gases. At the same time, now there is no unambiguous representation of the parameters of such an interaction, and further investigations are desirable, for example, into the pressure dependence of its parameters. Although the thermal (random) motion is essential in establishing the exponential behavior of the far wing profile and the elementary quantum representation of the two-level system is used, we did invoke no ideas of the statistical theory and, as in our earlier paper, 34 the priority was given to the Lorentz profile reflecting the wave character of the interaction between electromagnetic field and the matter. This means that the intermolecular interaction is not considered as a universal means when solving problems of the far-wing line profile, and, the preference is given, in essence, to the collisional phase-break Lorentz-Lenz-Weisskopf theory in the entire region of the frequency shifts. At the same time, the intermolecular interaction must play the main part in the pressure-induced absorption.

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