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BAND MODEL PARAMETERS FOR THE 4.3-MICRONS FUNDAMENTAL  
BAND OF CO<sub>2</sub> IN THE 100-3000 K TEMPERATURE RANGE

Stephen J. Young

Aerospace Corporation

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# Band Model Parameters for the 4.3- $\mu$ m Fundamental Band of CO<sub>2</sub> in the 100-3000°K Temperature Range

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19 February 1976

## Interim Report

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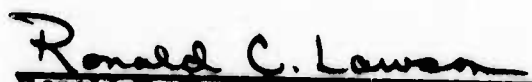


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parameters). The former set adequately describes the low-temperature variations of the parameters, but is inadequate for high-temperature applications. The latter set is suitable for high-temperature applications, but fails for low-temperature cases. Examples of the deficiencies of these two sets are presented by comparison of predicted spectra with experimental absorption and emission spectra for low- and high-temperature gas samples. The adequacy of the combined band model parameter set (COMBCO2 parameters) is demonstrated by comparison with the same experimental data. Examples of the construction of the combined set are given, and a tabulation of the parameter set is included as an Appendix.

## CONTENTS

I.	INTRODUCTION . . . . .	3
II.	COMPONENT BAND MODEL PARAMETER SETS . . . . .	5
	A. Band Model Parameters from Line Data (LINAVECO2 Parameters) . . . . .	5
	B. NASACO2 Parameters . . . . .	5
	C. Evaluation of Parameter Sets . . . . .	6
III.	COMBINED PARAMETER SET (COMBCO2) . . . . .	13
	APPENDIX. COMBCO2 PARAMETER SET LISTING . . . . .	17

## FIGURES

1.	Low-Temperature Transmission Spectra for Moderate CO <sub>2</sub> Absorption . . . . .	8
2.	Low-Temperature Transmission Spectra for Strong CO <sub>2</sub> Absorption . . . . .	9
3.	High-Temperature Emission Spectra for CO <sub>2</sub> . . . . .	11
4.	Variation of $\bar{k}$ with Temperature for Selected Spectral Intervals . . . . .	14
5.	Variations of $1/\delta_e$ with Temperature for Selected Spectral Intervals . . . . .	15

## I. INTRODUCTION

In a previous report,<sup>1</sup> the need for sets of band model parameters for infrared active species that are internally consistent for the entire temperature range from near atmospheric ( $\sim 250^{\circ}\text{K}$ ) to gas combustion values ( $\sim 2500^{\circ}\text{K}$ ) was discussed. In that report, parameter sets for the  $2.7\text{-}\mu\text{m}$  bands of  $\text{H}_2\text{O}$  and  $\text{CO}_2$  were constructed by combining the low temperature variations of parameters derived from the Air Force Cambridge Research Laboratories (AFCRL) atmospheric absorption line data compilation<sup>2</sup> with the high-temperature variations published by General Dynamics in the NASA Handbook of Infrared Radiation from Combustion Gases.<sup>3</sup> The former parameters are valid near atmospheric temperatures and the latter for high temperatures. Conversely, outside the temperature region for which they are valid, both sets are decidedly inadequate. The combined sets were constructed to provide a consistent set that could be applied for all temperatures in the  $100$  to  $3000^{\circ}\text{K}$  region. The present report extends this work to the  $4.3\text{-}\mu\text{m}$  band of  $\text{CO}_2$ .

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<sup>1</sup>S. J. Young, Band Model Parameters for the  $2.7\text{-}\mu\text{m}$  Bands of  $\text{H}_2\text{O}$  and  $\text{CO}_2$  in the  $100$  to  $3000^{\circ}\text{K}$  Temperature Range, TR-0076(6970)-4, The Aerospace Corp., El Segundo, Calif. (31 July 1975).

<sup>2</sup>R. A. McClatchey, W. S. Benedict, S. A. Clough, D. E. Burch, R. F. Calfe, K. Fox, L. S. Rothman, and J. S. Garing, AFCRL Atmospheric Absorption Line Parameters Compilation, AFCRL-TR-73-006, Air Force Cambridge Research Laboratories, Bedford, Mass. (25 January 1973).

<sup>3</sup>C. B. Ludwig, W. Malkmus, J. E. Reardon, and J. A. L. Thompson, Handbook of Infrared Radiation from Combustion Gases, eds. R. Goulard and J. A. L. Thompson, NASA SP-3080, Marshall Space Flight Center, Huntsville, Ala. (1973).

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## II. COMPONENT BAND MODEL PARAMETER SETS

### A. Band Model Parameters From Line Data (LINA VE Parameters)

The procedure for deriving band model parameters appropriate to a Lorentz line statistical band model from line data is discussed in detail in Ref. 1. This procedure was applied to the AFCRL line data\* in the 4.3- $\mu\text{m}$  region for  $\text{CO}_2$  to obtain the band model parameters  $\bar{k}$  (mean absorption coefficient) and  $1/\delta_e$  (mean effective line density) for the 97 spectral intervals from  $\nu = 2010$  to  $2490 \text{ cm}^{-1}$  by steps of  $5 \text{ cm}^{-1}$  with a spectral resolution  $\Delta\nu = 5 \text{ cm}^{-1}$  and for the 14 temperature values  $T = 100, 150, 200, 250, 300, 350, 400, 500, 750, 1000, 1500, 2000, 2500,$  and  $3000^\circ\text{K}$ . The line broadening parameter  $\bar{\gamma}_0$  for nonresonant self-broadening was derived for the same spectral intervals. The third band model parameter  $\bar{\gamma}$  (mean line half width at half height) is given in terms of  $\bar{\gamma}_0$  by Eq. (30) or (32) of Ref. 1 and the  $\text{CO}_2$  data of Table 1 of Ref. 1. The parameter set is designated for identification in this report as LINA VE $\text{CO}_2$ .

### B. NASA Parameters

The band model parameters for  $\text{CO}_2$  from the NASA Handbook<sup>3</sup> are based on quantum mechanical calculations by Malkmus,<sup>4</sup> and are intended primarily for high-temperature application. The data for  $\bar{k}$  are complete for the spectral region from 1900 to  $2395 \text{ cm}^{-1}$ . For  $1/\delta_e$ ,\*\* the data are given for the spectral region from 2000 to  $2390 \text{ cm}^{-1}$ . For both parameters, the data are given at the seven temperatures

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\*The line data compilation version dated 4 Feb 1975 by AFCRL was used in this work.

\*\*This parameter is from the NASA Handbook tabulation for the single line grouping (SLG) model.

<sup>4</sup>W. Malkmus, J. Opt. Soc. Am. 53, 951 (1963).

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$T = 300, 600, 1200, 1500, 1800, 2400, \text{ and } 3000^{\circ}\text{K}$  and reflect a spectral resolution of  $\sim 5 \text{ cm}^{-1}$ . From this data compilation, a band model parameter set (designated NASACO2 in this report) was constructed\* for  $\nu = 1900 \text{ to } 2400 \text{ cm}^{-1}$  by  $5 \text{ cm}^{-1}$  steps and for the same seven temperatures as the NASA Handbook tabulation. The NASA unit for  $\bar{k}$  is  $\text{cm}^{-1}$  at STP and was converted to the unit  $\text{cm}^{-1}/\text{atm}$  by multiplication by  $273/T$ . The  $\bar{\gamma}_0$  coefficient is taken as a constant for all  $\Delta\nu$  intervals with the value (Table 1, Ref. 1)  $\bar{\gamma}_0 = 0.09 \text{ cm}^{-1}/\text{atm}$ .

### C. Evaluation of Parameter Set

An evaluation of the LINAVERCO2 and NASACO2 parameter sets was made by comparing absorption and emission spectra predicted by the respective sets with experimental absorption and emission measurements made on homogeneous, isothermal gas samples. The evaluation was made for both room-temperature and high-temperature gas samples.

#### 1. Low-Temperature Evaluation ( $296^{\circ}\text{K}$ )

Gryvnak et al.<sup>5</sup> have made high-resolution measurements of the absorption spectra of the  $4.3\text{-}\mu\text{m}$  fundamental band of  $\text{CO}_2$  at  $296^{\circ}\text{K}$  for a wide variety of optical thickness,  $\text{CO}_2$  partial pressure, and total pressure (with  $\text{N}_2$  as the foreign gas). For most of the sample experimental case, extensive tables are given from which the integrated absorptance between any two wavenumbers can be calculated.

\*When required, linear interpolations with respect to  $\nu$  were used to obtain the parameters for spectral positions not listed in the Handbook tabulation. The  $\delta_e$  data at  $\nu = 2000 \text{ cm}^{-1}$  was assumed to prevail between  $\nu = 1900$  and  $2000 \text{ cm}^{-1}$ . The  $\delta_e$  values at  $2395 \text{ cm}^{-1}$  were assumed to be  $1/2$  the values at  $2390 \text{ cm}^{-1}$ . All  $\bar{k}$  and  $\delta_e$  values were set to zero at  $\nu = 2400 \text{ cm}^{-1}$ . The spurious temperature variation of  $\bar{k}$  between  $1500$  and  $2400^{\circ}\text{K}$  in the  $1990$  to  $2090 \text{ cm}^{-1}$  spectral region was modified by defining the parameter values at  $1800^{\circ}\text{K}$  to be the semilogarithmically interpolated value between the data at  $1500$  and  $2400^{\circ}\text{K}$ .

<sup>5</sup>D. A. Gryvnak, R. A. Patty, D. E. Burch, and E. E. Miller, Absorption by  $\text{CO}_2$  between  $1800$  and  $2850 \text{ cm}^{-1}$ , U-3857, Aeronutronic Div., Philco-Ford Corp., Newport Beach, Calif. (15 Dec. 1966).

The solid curve of Figure 1 is the result obtained for their sample No. 100. The relevant data for this sample are:  $p = 0.132$  atm,  $c = 2.5 \times 10^{-4}$ , and  $L = 12110$  cm. The optical depth is  $u = 0.399$  atm cm and represents a case of moderate absorption. The curve was constructed to reflect a spectral resolution  $\Delta\nu = 5$   $\text{cm}^{-1}$ . Figures 1a and 1b show the comparison of the experimental spectrum with the spectra computed with the LINAVERCO2 and NASACO2 parameters, respectively. Both of the parameter sets give results that underestimate the experimental absorption near the band center. This underestimation may be due to the use of a statistical rather than a regular line spacing band model. Over the whole of the band, the LINAVERCO2 parameters give an excellent fit to the experimental data, whereas the NASACO2 parameters give a poor fit.

A similar comparison was made for a more strongly absorbing sample (sample No. 40) in order to make a more sensitive comparison in the band wing region below  $2300$   $\text{cm}^{-1}$ . For this sample,  $p = 0.132$  atm,  $c = 4.0 \times 10^{-3}$ ,  $L = 46,900$  cm,  $u = 24.8$  atm cm, and, again,  $\Delta\nu = 5$   $\text{cm}^{-1}$ . The results are presented in Figure 2 in the same format as Figure 1. Again, the excellency of the LINAVERCO2 and the poorness of the NASACO2 parameter sets are demonstrated.

## 2. High-Temperature Evaluation ( $1500^{\circ}\text{K}$ )

A high-temperature emission comparison was made between calculated emissivity spectra and an experimental spectrum of Burch and Gryvnak.<sup>6</sup> The experimental conditions were:  $p = 0.249$  atm,  $c = 1.00$ ,  $L = 7.75$  cm, and  $T = 1500^{\circ}\text{K}$ .

<sup>6</sup> D. E. Burch and D. A. Gryvnak, Infrared Radiation Emitted by Hot Gases and its Transmission Through Synthetic Atmospheres, U-1929, Aeronutronic Div., Philco-Ford Corp. Newport Beach, Calif. (31 October 1962).

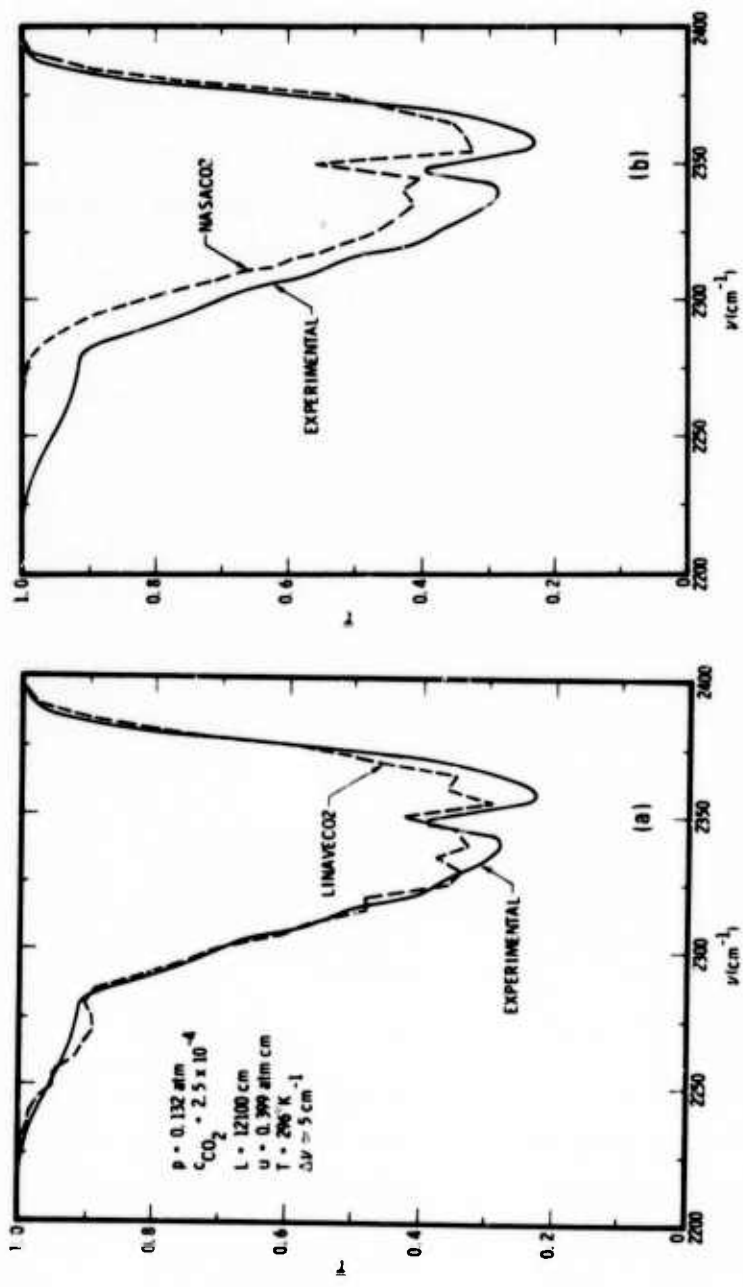


Figure 1. Low-Temperature Transmission Spectra for Moderate CO<sub>2</sub> Absorption. The LINAVECO<sub>2</sub> (a) and NASACO<sub>2</sub> (b) curves show spectra computed with the indicated band model parameter set. The EXPERIMENTAL curve is derived from the tables of Ref. 5 for sample No. 100 and for Δν = 5 cm<sup>-1</sup>.

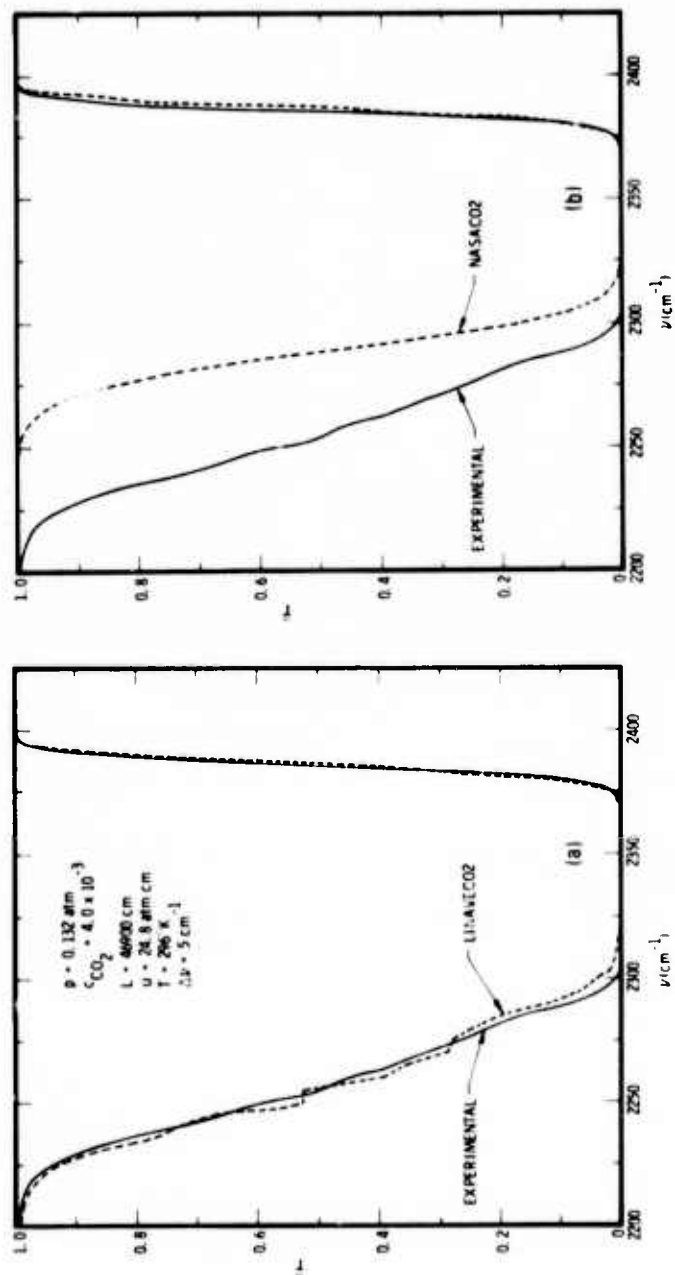


Figure 2. Low-Temperature Transmission Spectra for Strong CO<sub>2</sub> Absorption. The LINAVCO<sub>2</sub> (a) and NASACO<sub>2</sub> (b) curves show spectra computed with the indicated band model parameter set. The EXPERIMENTAL curve is derived from the tables of Ref. 5 for sample No. 40 and for  $\Delta\nu = 5 \text{ cm}^{-1}$ .

The optical depth is 1.93 atm cm and  $\Delta\nu \sim 8\text{cm}^{-1}$ . The comparison is shown in Figure 3. Here, we see the serious failing of the LINAVERCO2 parameters when applied to high-temperature gases. Not only is the band-center emission seriously underestimated, but the entire band wing emission below  $\sim 2200\text{cm}^{-1}$  is missing. The NASACO2 parameters, on the other hand, provide excellent agreement over the whole emission band.

These comparisons give the same qualitative result that was obtained for the  $2.7\text{-}\mu\text{m}$  band of  $\text{CO}_2$  in Ref. 1; namely, that the LINAVERCO2 parameters are valid for low-temperature applications but not for high-temperature applications, whereas the validity of the NASA parameters is reversed.

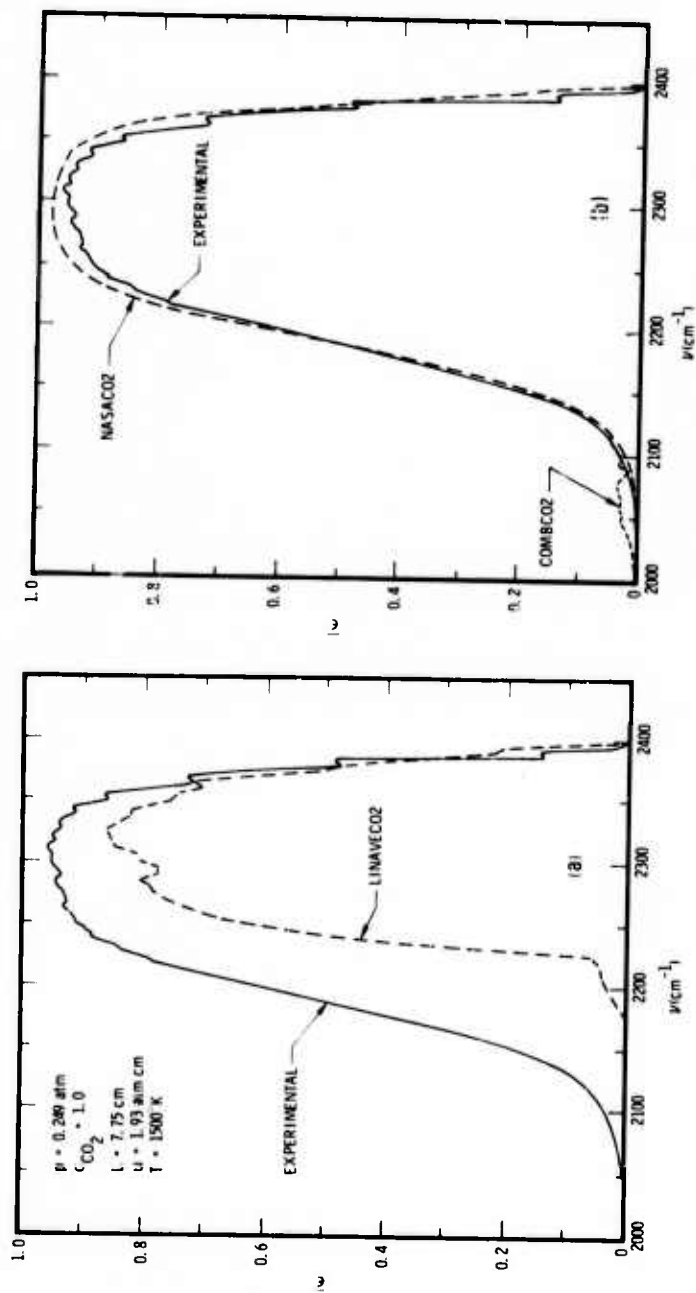


Figure 3. High-Temperature Emission Spectra for CO<sub>2</sub>. The LINAVECO2 (a), NASACO2 (b) and COMBICO2 (b) curves show spectra computed with the indicated band model parameter set. The EXPERIMENTAL curve is taken from Ref. 6.

### III. COMBINED PARAMETER SET (COMBCO2)

The synthesis of a consistent band model parameter set for the 4.3- $\mu\text{m}$  band of  $\text{CO}_2$  followed the general procedure given in Ref. 1. For each spectral interval, the LINAVERCO2 and NASACO2 parameters were plotted as a function of temperature, and an interpolation was made between some low-temperature point on the LINAVERCO2 curve to some high-temperature point on the NASACO2 curve. Some representative constructions are shown in Figures 4 and 5. The spectral positions  $\nu = 2100, 2225, \text{ and } 2350 \text{ cm}^{-1}$  represent positions in the far wing, near wing, and band center, respectively. As a general rule, the choice of interpolation line (dashed curves in Figures 4 and 5) was relatively self-evident, and little iteration was required to get a best fit between spectra computed with the synthesized parameter set and the experimental spectra of Section IIC.

The final version of the synthesized set is designated COMBCO2 in this report. The band model parameters  $\bar{k}$  ( $\text{cm}^{-1}/\text{atm}$ ) and  $1/\xi_e$  ( $1/\text{cm}^{-1}$ ) are given for the 81.5  $\text{cm}^{-1}$  spectral intervals from 2000 to 2400  $\text{cm}^{-1}$  and the 10 temperatures  $T = 100, 200, 300, 500, 750, 1000, 1500, 2000, 2500, \text{ and } 3000^\circ\text{K}$ . The broadening parameter  $\bar{\gamma}_0$  from the LINAVERCO2 set was used to represent this parameter in the combined set. A listing of the final parameter set is given in the Appendix.

Spectra for the homogeneous path conditions of Section IIC were generated with the final set to verify its validity. For both of the low-temperature cases, the spectra generated with the COMBCO2 parameters were indistinguishable (in plots) from those generated with the LINAVERCO2 set. For the high-temperature case, the only difference between the spectra

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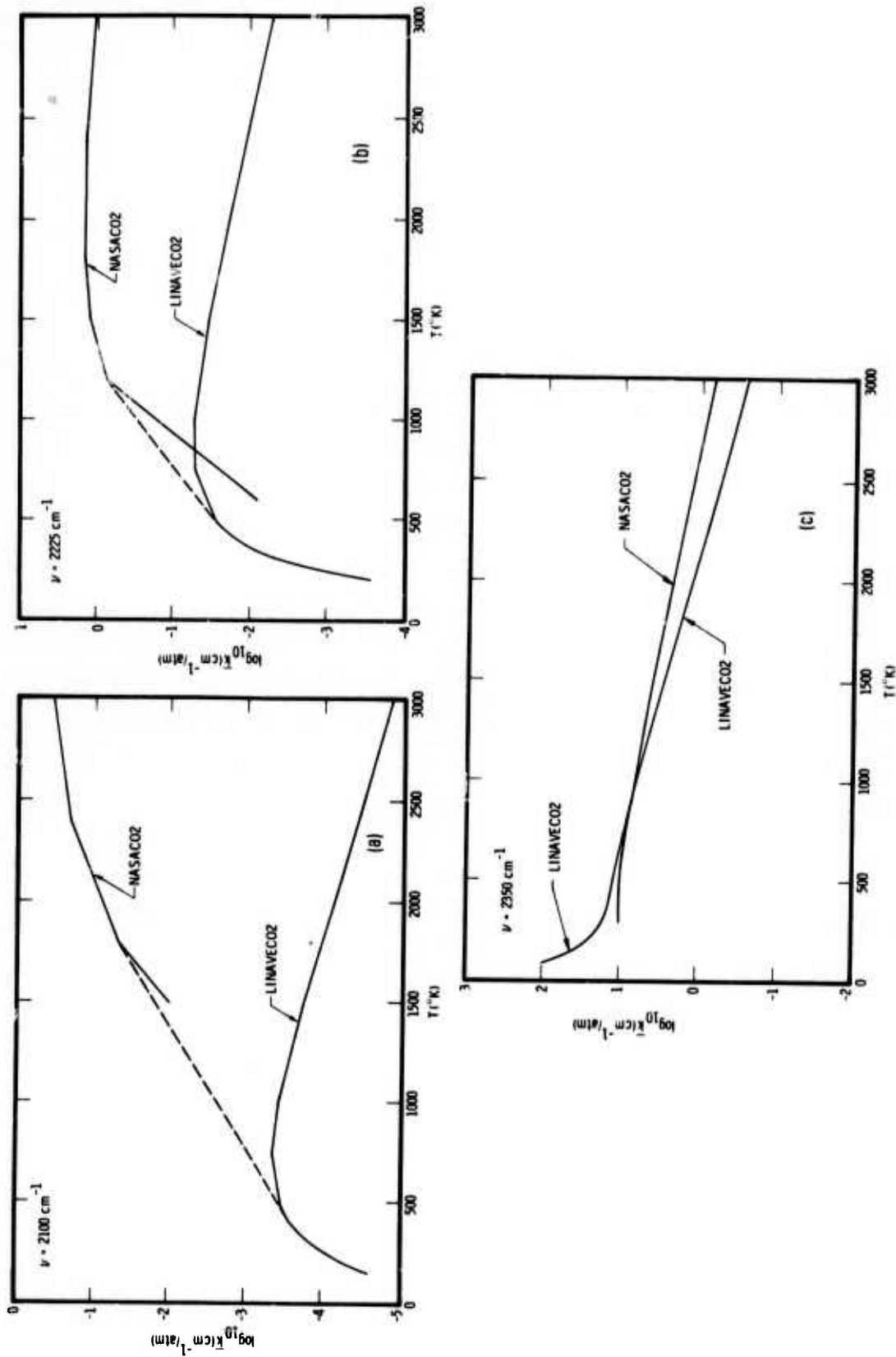


Figure 4. Variation of  $\bar{k}$  with Temperature for Selected Spectral Intervals. The LINA/VECO2 and NASAC02 curves are the variations for the indicated band model parameter set; dashed curves show assumed transitions from one curve to the other.

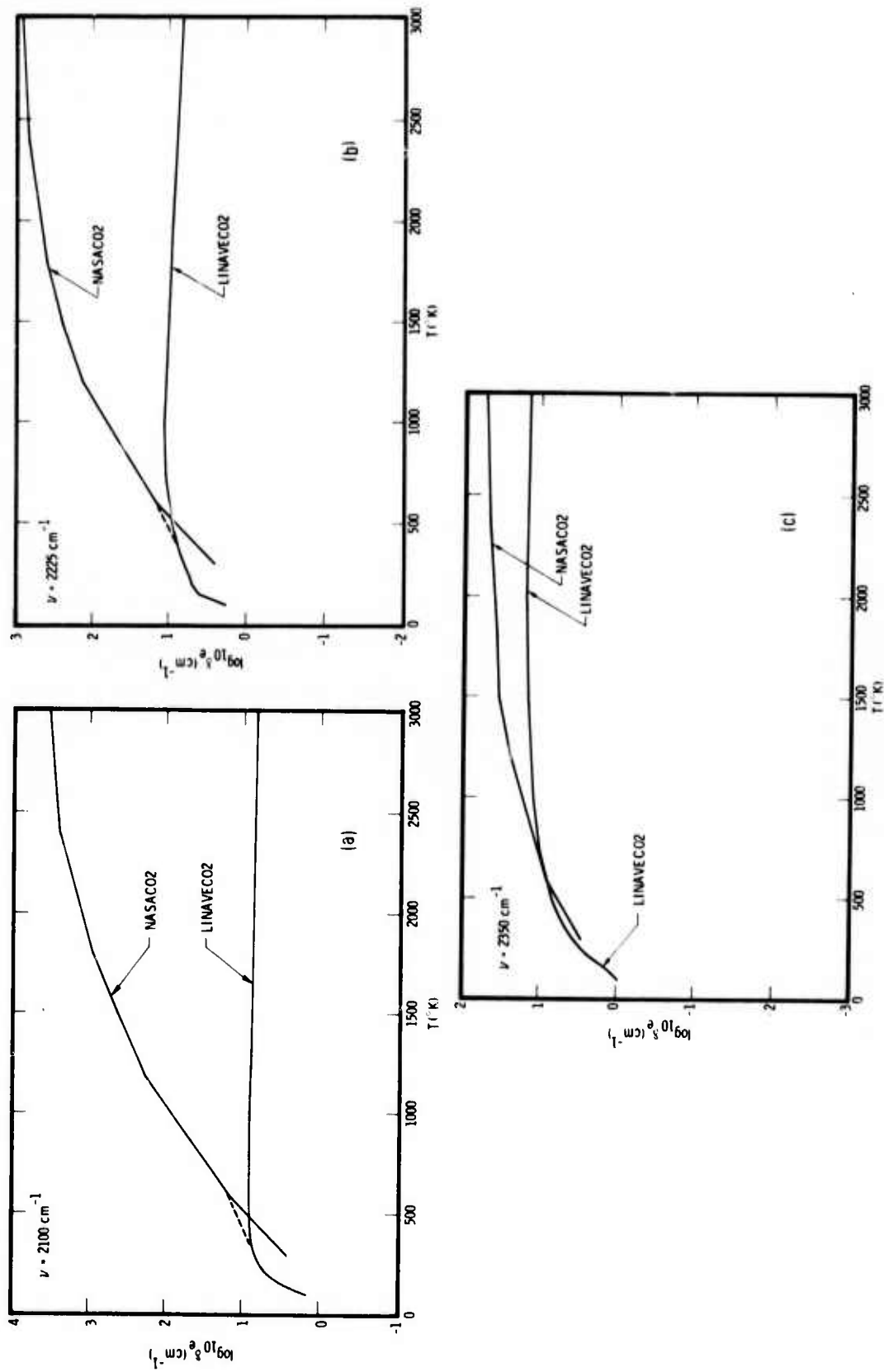


Figure 5. Variations of  $1/\delta_e$  with Temperature for Selected Spectral Intervals. The LINAVECO2 and NASACO2 curves are the variations for the indicated band model parameter set; dashed curves show assumed transitions from one curve to the other.

computed with the COMBCO2 and NASACO2 sets is an increase in emission for the former set in the far band wing below  $\sim 2100 \text{ cm}^{-1}$ . This difference is indicated in Figure 3b by the short-dashed curve. Thus, the COMBCO2 parameter sets successfully reproduce spectra for both high- and low-temperature applications and can be expected to give adequate results for intermediate temperatures.

## APPENDIX. COMBCO2 PARAMETER SET LISTING

A listing of the COMBCO2 band model parameters is given in the following table. IDNAME is simply an identification name for the parameter set. RESOLUTION is the value of  $\Delta\nu$  appropriate for the set. ALPHA (1) through ALPHA (5) are the ratios of the efficiency of pressure broadening by the indicated mechanisms to that of nonresonant self-broadening. ALPHA (6) is the atomic mass of the absorbing species. the WAVENUMBER array lists the center values of all the  $\Delta\nu$  intervals included in the set. NW is the number of such intervals. The TEMPERATURE array similarly lists the temperatures for which the data are tabulated. NT is the number of such temperatures. The ABSORPTION COEFFICIENT array is  $\bar{k}$ . The first column of this array is the interval center wavenumber, and the rows are the values at the temperatures of the TEMPERATURE array. The EFFECTIVE LINE DENSITY array is the parameter  $1/\delta_e$  and is presented in the same format as  $\bar{k}$ . The MEAN LINE WIDTH array is the parameter  $\bar{\gamma}_0$  for nonresonant self-broadening at STP. The values correspond to the wavenumbers listed in the WAVENUMBER array.









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