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**APPROXIMATE EMISSIVITY CALCULATIONS
FOR POLYATOMIC MOLECULES. I. CO₂**

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**APPROXIMATE EMISSIVITY CALCULATIONS FOR
POLYATOMIC MOLECULES. I. CO₂.***

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Approximate emissivity calculations for CO₂ have been carried out, as a function of optical density, at 300 and at 600°K. The calculations involve the assumption that the rotational lines overlap extensively. This condition appears to be satisfied at total pressures above about 1 atmos. Comparison of the values calculated from spectroscopic data with the emissivities tabulated by Höttel and his collaborators shows satisfactory agreement. The analysis presented in this manuscript emphasizes the fact that it is possible to obtain reasonable estimates for the engineering emissivity without performing extensive analytical work, provided the physical principles are understood and the needed spectroscopic data are available.

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I. INTRODUCTION

In a series of recently published papers we have attempted to estimate the engineering emissivities of diatomic molecules by utilizing basic spectroscopic constants. Tractable methods of calculation have been developed for (a) complete overlapping between rotational lines¹ and for (b) separated rotational lines.² Although accurate theoretical calculations of gas emissivities from spectroscopic data for polyatomic molecules involve formidable computational difficulties, it is to be expected that approximate calculations can be carried out with relatively little effort by utilizing approximations similar to those introduced for the study of diatomic molecules.^{1, 2} For CO₂ it has been found that the total absorptivity at room temperature is substantially independent of pressure at pressures exceeding about 1 atmos.,³ thus indicating extensive overlapping between rotational lines. Accordingly, it is not unreasonable to base the present preliminary calculations on the assumption that extensive overlapping between rotational lines does occur. The results would be expected to apply, for example, at room temperature for total pressures in excess of 1 atmos. and at 3000°K at pressures in excess of 11 atmos.³ Since it appears quite likely that the

¹ (a) S. S. Penner, J. Appl. Phys. 21, 685 (1950);
(b) J. Appl. Mech. 18, 53 (1951); (c) S. S. Penner and D. Weber, J. Appl. Phys. 22, 1164 (1951).

² (a) S. S. Penner, M. H. Ostrander, and H. S. Tsien, J. Appl. Phys. 23, 256 (1952); (b) S. S. Penner, J. Appl. Phys. 23, 825 (1952).

³ R. J. Holm, D. Weber, and S. S. Penner, J. Appl. Phys. 23, 1283 (1952).

actual range of validity of the results covers a larger range of total pressures, we shall not hesitate to show that calculated emissivities agree reasonably well with empirical data obtained⁴ at 1 atmos. at a temperature of 600°K as well as at 300°K. Perhaps the most important conclusion which can be derived from the present analysis is the statement that the analytical labor involved in making approximate emissivity calculations for a polyatomic molecule with overlapping rotational lines is trivial provided the important physical principles are understood and the needed spectroscopic data are available. It is only fair to note, however, that all of the needed integrated intensity measurements have not yet been performed for CO₂ in spite of the fact that several papers dealing with intensity measurements have been published recently.^{5,6,7}

II. BASIC RELATIONS

Theoretical considerations of intensities for various transitions of the CO₂ molecule were summarised by Dennison a number of years ago.⁸ We shall reproduce here the parts of the analysis which are useful for making approximate emissivity estimates on CO₂.

⁴ W. H. McAdams, Heat Transmission, McGraw-Hill Book Co., New York 1942, Chapter III by H. C. Hottel.

⁵ A. M. Thorndike, J. Chem. Phys. 15, 868 (1947).

⁶ D. F. Eggers, Jr., and B. L. Crawford, Jr., J. Chem. Phys. 19, 1554 (1951).

⁷ D. Weber, R. J. Holm, and S. S. Penner, J. Chem. Phys. 20, 1820 (1952).

⁸ D. M. Dennison, Rev. Mod. Phys. 3, 280 (1931).

Frequencies (ν) and wave numbers (ω) corresponding to transitions between fixed energy levels are given by the Bohr frequency condition

$$\nu = c \omega = (\Delta W'_V + \Delta W'_R)/h \quad (1)$$

where $\Delta W'_V$ and $\Delta W'_R$ represent, respectively, the changes in vibrational and rotational energy corresponding to the frequency ν , h is Planck's constant, and c represents the velocity of light. According to the results of Dennison and Adel,^{9, 10} the rotational (W'_R) and vibrational (W'_V) energies for CO_2 are given, respectively, by the expressions

$$W'_R = hc(j^2 + j - \ell^2) \left[0.3925 - 0.00058 (n_1 + \frac{1}{2}) + 0.00045 (n_2 + 1) - 0.00307 (n_3 + \frac{1}{2}) \right] + 1.7 hc(\ell^2 - 1) \quad (2)$$

and

$$W'_V = hc \left[1351.2 (n_1 + \frac{1}{2}) + 672.2 (n_2 + 1) + 2396.4 (n_3 + \frac{1}{2}) - 0.3 (n_1 + \frac{1}{2})^2 - 1.3 (n_2 + 1)^2 - 12.5 (n_3 + \frac{1}{2})^2 + 5.7 (n_1 + \frac{1}{2}) (n_2 + 1) - 21.9 (n_1 + \frac{1}{2}) (n_3 + \frac{1}{2}) - 11.0 (n_2 + 1) (n_3 + \frac{1}{2}) \right] \quad (3)$$

⁹ D. M. Dennison, Rev. Mod. Phys. 12, 175 (1940).

¹⁰ Slight corrections to the listed numerical values have been noted by W. S. Benedict and his collaborators. For the present purposes these corrections are negligibly small. Recent work on frequencies of lines belonging to CO_2 is described in the following papers: W. S. Benedict, R. C. Herman, and S. Silverman, J. Chem. Phys. 19, 1325 (1951); J. H. Taylor, W. S. Benedict, and J. Strong, Progress Report on "Infrared Spectra of H_2O and CO_2 at 500°C ", Contract Nonr. 248-01, The Johns Hopkins University, March 1952.

Here n_1 , n_2 , and n_3 are the vibrational quantum numbers associated with the fundamental vibration frequencies ν_1 , ν_2 , and ν_3 , respectively. The quantum number ℓ measures the angular rotation, in units of $h/2\pi$, which is associated with the degenerate (bending) ν_2 -vibration. The rotational energy levels are identified by the rotational quantum number j .

The selection rules⁹ for the transitions $n_1 n_2 \ell n_3 \rightarrow n'_1 n'_2 \ell' n'_3$ may be summarized as follows. For the perpendicular bands, Δn_2 odd, Δn_3 even, $\Delta \ell = \pm 1$. For the parallel bands, Δn_2 even, Δn_3 odd, $\Delta \ell = 0$. The rotational selection rules are $\Delta j = \pm 1, 0$.

The amplitude factors⁸ for given rotational transitions are¹¹

$$\left(A_j^j \begin{smallmatrix} \ell \\ \ell \end{smallmatrix} \right)^2 = \frac{\ell^2}{j(j+1)}, \quad (4)$$

$$\left(A_{j-1}^j \begin{smallmatrix} \ell \\ \ell \end{smallmatrix} \right)^2 = \frac{j^2 - \ell^2}{j(2j+1)} = \frac{2j-1}{2j+1} \left(A_j^{j-1} \begin{smallmatrix} \ell \\ \ell \end{smallmatrix} \right)^2 \quad (4a)$$

$$\left(A_j^j \begin{smallmatrix} \ell \\ \ell \pm 1 \end{smallmatrix} \right)^2 = \frac{(j \pm \ell)(j \mp \ell + 1)}{4j(j+1)}, \quad \ell \neq 0, \quad (4b)$$

$$\left(A_j^j \begin{smallmatrix} 0 \\ 1 \end{smallmatrix} \right)^2 = 1/2, \quad (4c)$$

$$\left(A_{j-1}^j \begin{smallmatrix} \ell \\ \ell \pm 1 \end{smallmatrix} \right)^2 = \frac{(j \pm \ell)(j \pm \ell - 1)}{4j(2j+1)}, \quad \ell \neq 0, \quad (4d)$$

$$\left(A_{j-1}^{j-1} \begin{smallmatrix} 0 \\ 1 \end{smallmatrix} \right)^2 = \frac{j-1}{2(2j+1)}, \quad (4e)$$

$$\left(A_j^{j-1} \begin{smallmatrix} \ell \\ \ell \pm 1 \end{smallmatrix} \right)^2 = \frac{(j \pm \ell)(j \pm \ell + 1)}{4j(2j-1)}, \quad \ell \neq 0, \quad (4f)$$

¹¹ The amplitude factors given in Eqs. (4) to (4g) are 4 times as large as those listed by Dennison⁸ because we are using a Fourier series in time of the form $\cos(2\pi\nu t)$ rather than $\exp(2\pi i \nu t)$.

$$\left(A_j^{j-1} \begin{smallmatrix} 0 \\ 1 \end{smallmatrix} \right)^2 = \frac{j+1}{2(2j-1)} \quad (4g)$$

In Eqs. (4) to (4g) the convention has been adopted that the amplitude factor corresponding to the transition $j, \ell \rightarrow j', \ell'$ has been written as $\left(A_j^{j'} \begin{smallmatrix} \ell' \\ \ell \end{smallmatrix} \right)$.

The integrated absorption for a given transition from the lower energy level $n_1 n_2 \ell n_3; j$ to the upper energy level $n'_1 n'_2 \ell' n'_3; j'$ is designated as $S \equiv S(n_1 n_2 \ell n_3; j \rightarrow n'_1 n'_2 \ell' n'_3; j')$ and is given⁸ by the approximate relation

$$S = (8\pi^3 \nu N_T / 3hc Q'_V Q'_R) \left\{ \exp \left[- \left[W'_V(n_1, n_2, n_3) + W'_R(j, \ell) \right] / kT \right] \right. \\ \left. \times g_{j', \ell'} \left(A_j^{j'} \begin{smallmatrix} \ell' \\ \ell \end{smallmatrix} \right)^2 \beta^2 \left[1 - \exp(-h\nu/kT) \right] \right\} \quad (5)$$

In Eq. (5) $\nu \equiv \nu(n_1 n_2 \ell n_3; j \rightarrow n'_1 n'_2 \ell' n'_3; j')$ is the Bohr frequency corresponding to the indicated change in the quantum numbers;* N_T = total number of molecules per unit volume per unit pressure; $g_{j', \ell'}$ = statistical weight of the upper state with $g_{j', \ell'} = 2j' + 1$ for $\ell' = 0$ and $g_{j', \ell'} = 2(2j' + 1)$ for $\ell' \neq 0$; $\beta \equiv \beta(n_1 n_2 \ell n_3 \rightarrow n'_1 n'_2 \ell' n'_3)$ is a factor which must be determined empirically and corresponds to the matrix element of the electric moment in the molecule associated with the indicated change in (vibrational) quantum numbers;* $Q'_R = \sum_j \sum_{\ell} g_{j, \ell} \exp \left[- W'_R(j, \ell) / kT \right] =$ complete rotational partition function; $Q'_V = \sum_{n_1} \sum_{n_2} \sum_{n_3} \exp \left[- W'_V(n_1, n_2, n_3) / kT \right] =$ complete vibrational partition function.

* Although we shall not indicate explicitly the changes in the (vibrational) quantum numbers involved, both ν and β do, of course, vary from one vibration-rotation band to another.

The amplitude factors given in Eqs. (4) to (4g) are based on the assumption that vibration-rotation interactions have a negligibly small influence on amplitude factors. For this reason, the use of Eq. (5) involves somewhat cruder calculations than were employed in the calculations on diatomic molecules with non-overlapping rotational lines.²

For the purposes of approximate radiant heat transfer calculations it is convenient to use the integrated absorption coefficient for a given band. Let

$$\alpha(n_1 n_2 \ell n_3 \rightarrow n'_1 n'_2 \ell' n'_3) = \sum_j \sum_{j'} S(n_1 n_2 \ell n_3; j \rightarrow n'_1 n'_2 \ell' n'_3; j'). \quad (6)$$

For the parallel bands $\Delta \ell = 0$ and $\sum_{j'} g_{j' \ell'} (A_j^{j'} \ell')^2 = g_{j \ell}$ according to the Burger and Dorgelo summation rules, which may be verified by use of Eqs. (4) to (4g) by utilizing the selection rules. Hence we obtain from Eqs. (5) and (6) the useful result

$$\begin{aligned} \alpha(n_1 n_2 \ell n_3 \rightarrow n'_1 n'_2 \ell' n'_3) &= (8\pi^3 \rho^2 \nu^4 N_T / 3hc Q_V) g_\ell \left\{ \exp \left[-W_V(n_1, n_2, n_3, \ell) / kT \right] \right. \\ &\quad \times \left[1 - \exp(-h\nu/kT) \right] \text{ with } g_\ell = 1 \text{ for } \ell = 0 \text{ and } g_\ell = 2 \\ &\quad \text{for } \ell \neq 0. \end{aligned} \quad (7)$$

where we have set $W_V'(n_1 n_2, n_3) + W_R'(j, \ell) = W_V(n_1, n_2, n_3, \ell) + W_R(j)$,

$$Q_V = \sum_{n_1, n_2, n_3, \ell} \exp(-W_V/kT), \quad Q_V' Q_R' = Q_V Q_R \text{ with } Q_R = \sum_j g_j \exp[-W(j)/kT],$$

and $g_{j \ell} = g_j \times g_\ell$. If the assumption is not made that

$\nu \approx \nu' \approx \nu(n_1 n_2^{\ell} n_3; 0 \rightarrow n_1' n_2'^{\ell} n_3'; 0)$ a slight correction is obtained to Eq. (7).^{12, 13}

For the perpendicular bands $\Delta \ell \neq 0$ and $\sum_{j', \ell'} S_{j', \ell'} (\Lambda_j^{j'} \ell')^2 = g_{j \ell}$.

$$\sum_{j'} S_{j', \ell'} (\Lambda_j^{j'} \ell' \pm 1)^2 = \frac{1}{2} g_{j \ell} \cdot \sum_{j', \ell'} S_{j', \ell'} (\Lambda_j^{j'} \ell'_0)^2 = g_{j \ell}. \quad \text{Hence}$$

$$\alpha(n_1 n_2^{\ell} n_3 \rightarrow n_1' n_2'^{\ell+1} n_3') = (4\pi^3 \rho^2 \nu' N_T / 3hcQ_V) g_{j \ell}$$

$$\times \left\{ \exp - [W_V(n_1, n_2, n_3, \ell) / kT] \right\} [1 - \exp(-h\nu/kT)]$$

where $g_{j \ell} = 1$ for $\ell = 0$ and $g_{j \ell} = 2$ for $\ell \neq 0$, (8)

and

$$\alpha(n_1 n_2^{\ell} n_3 \rightarrow n_1' n_2'^{\ell-1} n_3') = (4\pi^3 \rho^2 \nu' N_T / 3hcQ_V) g_{j \ell}$$

$$\times \left\{ \exp - [W_V(n_1, n_2, n_3, \ell) / kT] \right\} [1 - \exp(-h\nu/kT)]$$

where $g_{j \ell} = 1$ for $\ell = 0$ and $g_{j \ell} = 2$ for $\ell \neq 0$. (9)

In using Eqs. (8) and (9) it should be noted particularly that the quantities ρ^2 are generally different for the transitions involving $\ell \rightarrow \ell + 1$ and

¹² B. L. Crawford, Jr., and H. L. Dinmore, J. Chem. Phys. 18, 983, 1682 (1950).

¹³ Eggers and Crawford⁶ have shown how to relate for CO₂ the matrix coefficients to the coefficients of Taylor series expansions of the potential energy and dipole moment about their respective equilibrium values.

$l \rightarrow l-1$. The ratios of the β^2 can be calculated theoretically to the harmonic oscillator approximation for harmonic bands (see Appendix I for details).

In the same manner as for emissivity calculations on diatomic molecules with overlapping rotational lines,¹ we shall find Eq. (5) useful for the determination of "effective band-widths" whereas Eqs. (7) to (9) lead directly to "average absorption coefficients" for vibration-rotation bands. For the calculation of S it may be convenient to combine Eqs. (4) to (4g), (5), and (7) to (9). For example, for the positive branch of the ν_2 - fundamental of CO_2 , we obtain

$$S \approx \left[\omega(01^1 0; j \rightarrow 00^0 0; j-1) / \omega(01^1 0; 0 \rightarrow 00^0 0; 0) \right] \times (\Omega_R)^{-1} \\ \times \alpha(01^1 0 \rightarrow 00^0 0) \times (j+1) \left[\exp - (W_R/kT) \right] \quad (5a)$$

where ω denotes a wave number.

III. APPROXIMATE EMISSIVITY CALCULATIONS AT 300°K

The positions and approximate intensities of the stronger vibration-rotation bands of CO_2 at 300°K are well-known.^{5-10, 14} A first approximation to the effective band-width is obtained by utilizing relations such as Eq. (5a) and defining the effective band-width as the wave number range for which S exceeds 10^{-3} of its maximum value.¹ Utilizing this definition of band-width, which we shall refine presently, a summary of vibration-rotation bands for CO_2 at 300°K has been drawn in Figs. 1 to 3.

¹⁴ G. Herzberg, Infrared and Raman Spectra of Polyatomic Molecules, D. Van Nostrand Co., New York 1951, Table 56 on p. 274. Herzberg's designations of band intensities as vs (very strong), s (strong), m (medium), w (weak), and vw (very weak) has been used in Figs. 1 to 3 to identify the approximate strengths of bands at room temperature.

In these diagrams the abscissa is the wave number whereas the ordinate represents, in arbitrary units which vary from one band to another, the intensity. Integrated intensities at 300°K are given in Table I in so far

Table I. Integrated intensities for vibration - rotation bands of CO₂.⁷

Band Center (cm ⁻¹)	Transition	$\alpha(\text{cm}^{-2}\text{-atmos}^{-1})$
5109	00 ⁰ 0 → 20 ⁰ 1	0.426
4984	00 ⁰ 0 → 12 ⁰ 1	1.01
4861	00 ⁰ 0 → 04 ⁰ 1	0.272
3716	00 ⁰ 0 → 10 ⁰ 1	42.3
3609	00 ⁰ 0 → 02 ⁰ 1	28.5
2349	00 ⁰ 0 → 00 ⁰ 1	2706
2137 } 2094 } 2077 }	01 ¹ 0 → 20 ⁰ 0 } 01 ¹ 0 → 12 ² 0 } 00 ⁰ 0 → 11 ¹ 0 }	.147
1933 } 1886 }	00 ⁰ 0 → 03 ¹ 0 } 01 ¹ 0 → 04 ⁰ 0 }	.083
721 } 668 } 667 } 648 } 618 }	01 ¹ 0 → 10 ⁰ 0 } 01 ¹ 0 → 02 ² 0 } 00 ⁰ 0 → 01 ¹ 0 } 02 ⁰ 0 → 03 ¹ 0 } 01 ¹ 0 → 02 ⁰ 0 }	171.5
961	10 ⁰ 0 → 00 ⁰ 1	0.0219*
1064	02 ⁰ 0 → 00 ⁰ 1	0.0532*

* Unpublished data obtained by D. Weber.

as they are available.⁷ For the strongest bands (ν_2 - and ν_3 - fundamentals) the contributions from the isotopic molecule $C^{13}O_2^{16}$ are also shown. This chemical species constitutes about 1.1% of all of the CO_2 present and makes contributions to the total emissivity which are not negligibly small for moderate and large optical densities of emitter. Also shown in Figs. 1 to 3 are the spectral intensities of radiation emitted by a blackbody at 300, 600 and 1500°K, again utilizing arbitrary scales along the ordinates, which are, however, consistent for any one of the given temperatures.

Reference to Fig. 1 shows that the principal contribution to the total radiant heat transfer at room temperature must occur from the spectral region around the ν_2 - fundamental. Thus it is readily shown¹ that the "limiting emissivity" for CO_2 at 300°K is in the neighborhood of 0.4 with more than 90% of the total contributions made by the wave number region extending from 551 to 830 cm^{-1} .

It is physically obvious that the engineering emissivity E at 300°K is determined by the $01^10 \rightarrow 00^00$ transition in the limit of zero optical density of the emitter. As was pointed out in an earlier publication,¹ the emissivity as a function of optical density (partial pressure p of emitter $\times$ optical pathlength L) for very small values of pL can be computed quite accurately either by utilizing a treatment applicable for non-overlapping lines² or by using average absorption coefficients. The emissivity E_{667} of the band with center at 667 cm^{-1} , as calculated from Eq. (30) of Ref. 2a, is plotted as a function of pL in Fig. 4 and labelled "approximation for non-overlapping lines". The corresponding

quantity obtained by using the average absorption coefficient⁷
 $PX = (171 \text{ cm}^{-2} \text{ atmos}^{-1}) / (106 \text{ cm}^{-1})$ over the effective band-width
 extending from 600 to 706 cm^{-1} gives results which are practically
 identical with those obtained for non-overlapping rotational lines as pL
 is reduced below about 0.02 ft-atmos. As pL is increased Eq. (30) of
 Ref. (2a) rapidly fails to apply and yields excessively large values not
 only for E_{667} but also for the total engineering emissivity E . Need-
 less to say, for $pL \leq 0.02$ ft-atmos, the numerical value of E_{667} is
 to be identified with the value of E .

As the optical density is increased to moderate values of pL ,
 it becomes necessary first to include the $01^1_0 \rightarrow 00^0_0$ transition for
 $\text{C}^{13}\text{O}_2^{16}$, which is assumed to represent 1.1 % of the total CO_2 ,
 and for which the integrated intensity is roughly the same as for the
 $\text{C}^{12}\text{O}_2^{16}$ species. In treatments using average absorption coefficients
 it is, of course, necessary to add absorption coefficients rather than
 emissivities in regions in which partial overlapping between effective
 band-widths occurs. Finally, the contributions made by the $10^0_0 \rightarrow 01^1_0$
 and $02^0_0 \rightarrow 01^1_0$ bands must be included; both of these are designated
 by Hersberg¹⁴ as being of "medium" intensity whereas the $03^1_0 \rightarrow 02^0_0$
 and $11^1_0 \rightarrow 02^2_0$ bands are labelled "weak" and the $11^1_0 \rightarrow 02^0_0$
 band is said to be "very weak".

Strictly speaking, room temperature emissivity calculations for
 CO_2 cannot be carried further because the integrated intensities for the
 weaker bands are not available. However, it is easily shown that
 results in fairly good agreement with Hottel's data⁴ are obtained if reason-

able estimates are made for the strongest of the bands which has not yet been included in determining E .

It is known from rough unpublished measurements⁷ that the integrated intensity α_{721} for the $10^0 0 \rightarrow 01^1 0$ band is of the order of $1 \text{ cm}^{-2} \text{ atmos}^{-1}$. Emissivity calculations treating α_{721} as a variable parameter between 0 and $10 \text{ cm}^{-2} \text{ atmos}^{-1}$ are shown in Fig. 4. The curves labelled $\alpha_{721} = 0$, $\alpha_{721} = 1$, and $\alpha_{721} = 10$ with $\alpha_{667} = 171$ correspond to the calculated values of E for the various assumed values of pL . In making these calculations the concept of the effective band-width was refined in so far as the effective band-width for the ν_2 -fundamental was set equal to the wave number range for which $SpL/0.7 \geq 0.1$. This definition of band-width for strong bands has the important advantage of making the band-width a weak function of pL , in agreement with empirical measurements.^{1b} The contributions to E made by the $00^0 1 \rightarrow 10^0 0$ and $00^0 1 \rightarrow 02^0 0$ bands are very small but have also been included. To these bands Weber assigns the following values at 300°K : $\alpha_{961} = 0.0219 \text{ cm}^{-2} \text{ atmos}^{-1}$, $\alpha_{1064} = 0.0532 \text{ cm}^{-2} \text{ atmos}^{-1}$.

In addition to the calculated emissivities, the results of empirical measurements⁴ are also shown in Fig. 4. For $pL \leq 0.06 \text{ ft-atmos}$. the "empirical" data are really extrapolated results. Reference to Fig. 4 shows fair agreement between calculated and observed values of E for the reasonable values $\alpha_{667} = 171$ and $\alpha_{721} = 1 \text{ cm}^{-2} \text{ atmos}^{-1}$. At very small values of pL the calculated emissivities* are larger than

* The dotted curve in Fig. 2 corresponds to $\alpha_{667} = 29$ and $\alpha_{721} = 1 \text{ cm}^{-2} \text{ atmos}^{-1}$. Although this curve represents a good fit of the empirical data, the result cannot be considered to be significant since α_{667} is known to be much larger than $29 \text{ cm}^{-2} \text{ atmos}^{-1}$.

the extrapolated empirical data but must be considered to be more reliable since they depend only on the numerical value of α_{667} , which is known with fair accuracy.⁵⁻⁷ Our ability to calculate E at very large values of pL is limited by the lack of adequate intensity data for the transitions $02^00 \longrightarrow 01^10$, $11^10 \longrightarrow 02^20$, and $11^10 \longrightarrow 02^00$. However, it is clear that as pL is increased sufficiently above 3 ft-atmos, E must approach about 0.4 and will then increase only very slowly as exceedingly large values of pL are obtained.

The discussion of emissivity calculations on CO_2 at $300^\circ K$ as a function of pL is mostly of academic interest. However, it is instructive in so far as it exemplifies the inherent simplicity of the analysis if adequate spectroscopic data are available.

IV. APPROXIMATE EMISSIVITY CALCULATIONS AT $600^\circ K$

It is evident from Fig. 2 that the major contributions to radiant heat transfer at $600^\circ K$ are made by vibration-rotation bands in the spectral region between 890 and 2400 cm^{-1} . As pL goes to zero it is expected that the total emissivity E will become substantially equal to the emissivity of the intense ν_3 - fundamental of $C^{12}O_2^{16}$ with appropriate corrections for contributions from the isotopic species $C^{13}O_2^{16}$. Although we are unable to complete the emissivity calculations at $600^\circ K$ because the needed spectroscopic data are not available, the engineering emissivity is estimated correctly for small values of pL . This result emphasizes the fact that relatively accurate emissivity calculations for polyatomic molecules can be performed without difficulty as soon as the necessary infrared intensity measurements are available.

For the weaker bands, i.e.; for all bands but the ν_3 - fundamental, we shall define the effective band-width as the wave number region for which S exceeds about 10^{-3} of its maximum value. For the ν_3 - fundamental we set the effective band-width equal to the wave number range for which $(SpL/0.7) \geq 0.1$.* The long wave-number limit of the ν_3 - fundamental is set equal to 2410 cm^{-1} , i.e., 11 cm^{-1} beyond the band head. This choice allows for the many tails of rotational lines which "spill" across the band-head limit. We proceed by calculating separately the partial emissivities for selected wave number regions.

(A). Contributions of the $00^{\circ}1 \rightarrow 10^{\circ}0$ and $00^{\circ}1 \rightarrow 02^{\circ}0$ Bands

The criterion $(S/S_{\text{max}}) \geq 10^{-3}$ for the weaker bands leads to band-widths of the order of 140 cm^{-1} at 600°K . The integrated intensity for a given vibration-rotation band at the arbitrary temperature T , divided by the measured value at 300°K , is obtained by the use of Eqs. (7), (8) or (9). From these relations we find for the integrated intensity α of the band whose ground state is identified by the set of quantum numbers $1n_2^2n_3$ the result

* Although the definitions of the effective band-widths are somewhat arbitrary, it is easily shown that calculated values of E are quite insensitive to the chosen band-widths. This result is caused by automatic choice of too small absorption coefficients for band-widths which are too large, and conversely (Compare Ref. 1a). Use of the quantity $SpL/0.7 \geq 0.1$ assures inclusion within the band-width of the ν_3 - fundamental of all of the rotational lines with average emissivities in excess of 0.1. Substantially the same numerical values for the effective band-widths of the ν_3 - fundamental are obtained if the lines with $SpL/0.7 \geq 10^{-1}n$ are included, where n is a number which does not differ from unity by more than a factor of three or four.

$$\begin{aligned} \alpha(T^{\circ}\text{K})/\alpha(300^{\circ}\text{K}) &= (300/T) \left\{ Q_V(300^{\circ}\text{K}) \exp \left[W_V(00^{\circ}0)/300k \right] \right\} \\ &\times \left\{ Q_V(T^{\circ}\text{K}) \times \exp \left[W_V(00^{\circ}0)/kT \right] \right\}^{-1} \\ &\times \left\{ \exp - \left[W_V(n_1 n_2 n_3) - W_V(00^{\circ}0) \right] \left[(kT)^{-1} - (300k)^{-1} \right] \right\} \\ &\times \left[1 - \exp(-h\nu/kT) \right] \left[1 - \exp(-h\nu/300k) \right] \quad (10) \end{aligned}$$

where

$$\begin{aligned} Q_V(T) \exp \left[W_V(00^{\circ}0)/kT \right] &\simeq \left[1 - \exp(-1388 hc/kT) \right]^{-1} \\ &\times \left[1 - \exp(-667 hc/kT) \right]^{-2} \\ &\times \left[1 - \exp(-2349 hc/kT) \right]^{-1} \quad (11) \end{aligned}$$

The use of Eqs. (10) and (11) leads to the following results for the $00^{\circ}1 \rightarrow 10^{\circ}0$ and $00^{\circ}1 \rightarrow 02^{\circ}0$ bands, respectively: $\alpha_{961}(600^{\circ}\text{K}) \simeq 9.29$ $\alpha_{961}(300^{\circ}\text{K}) = 0.204 \text{ cm}^{-2} \text{ atmos}^{-1}$; $\alpha_{1064}(600^{\circ}\text{K}) \simeq 7.22$ $\alpha_{1064}(300^{\circ}\text{K}) = 0.385 \text{ cm}^{-2} \text{ atmos}^{-1}$. Hence the total contribution to the emissivity obtained from the two bands under discussion is

$$\begin{aligned} E_{961} + E_{1064} &= 0.052 \left[1 - \exp(1.40 \times 10^{-3} \text{ pL}) \right] \\ &+ 0.017 \left[1 - \exp(4.15 \times 10^{-3} \text{ pL}) \right] \\ &+ 0.054 \left[1 - \exp(2.75 \times 10^{-3} \text{ pL}) \right] \quad (12) \end{aligned}$$

where pL is expressed in cm-atmos.

(B). Contributions Made by the Bands Between 1800 and 2210 cm^{-1}

It is known from the recent work of Taylor, Benedict, and Strong¹⁵ that an appreciable number of vibration-rotation bands, in addition to the bands shown in Fig. 1 in this region, are observable at 500°C.* Since all of these bands involve transitions to excited energy levels, their integrated intensity¹⁶ would be expected to increase as the temperature is raised. Rough absolute intensity estimates for several bands in the 5 micron region are given in Table II at 300 and 600°K. The relative intensity estimates were obtained by using the greatly oversimplified harmonic oscillator approximation described in Appendix I. Thus the contributions of both mechanical and of electrical anharmonicities to the intensity have been neglected.

As is evident from the experimental data shown in Reference 15, a large number of vibration-rotation bands remain for which even rough intensity data are not available. For this reason no result can be given for the contributions made to the engineering emissivity by the vibration-rotation bands between 1800 and 2100 cm^{-1} .

¹⁵ J. H. Taylor, W. S. Benedict, and J. Strong, Progress Report on "Infrared Spectra of H₂O and CO₂ at 500°C", Contract Nonr 248-01, The Johns Hopkins University, Baltimore, Md., March 1952.

* Data of the type presented in Ref. 15 are valuable aide in performing emissivity calculations. For example, the spectroscopic transmission records indicate, at least qualitatively, the vibration-rotation bands which must be included, at temperatures up to 773°K.

Table II. Absolute intensities at 300 and at 600°K for several vibration-rotation bands in the 5 micron region, based on intensity calculations using harmonic oscillator approximations (Cf. Appendix I.). The value $0.147 \text{ cm}^{-2}\text{-atmos}^{-1}$ for the sum of the bands centered at 2137, 2094, and 2077 cm^{-1} served as standard to fix the absolute intensity scale.

Transition	α at 300°K ($\text{cm}^{-2}\text{-atmos}^{-1}$)	α at 600°K ($\text{cm}^{-2}\text{-atmos}^{-1}$)
$00^0_0 \rightarrow 11^1_0$	0.12	0.040
$01^1_0 \rightarrow 12^2_0$	0.020	0.032
$01^1_0 \rightarrow 20^0_0$	0.0045	0.0075
$01^1_0 \rightarrow 12^0_0$	0.00048	0.00079
$02^2_0 \rightarrow 13^3_0$	0.0012	0.0099
$03^3_0 \rightarrow 14^4_0$	0.000038	0.0020
$02^2_0 \rightarrow 21^1_0$	0.00019	0.0015
$10^0_0 \rightarrow 21^1_0$	0.00029	0.0026
$11^1_0 \rightarrow 30^0_0$	0.0000053	0.00026

(C). Contributions from the ν_3 - Fundamental

From the measured integrated intensity⁷ for the ν_3 -fundamental at 300°K and by use of Eqs. (10) and (11) we find $\alpha_{2349} = 1060 \text{ cm}^{-2}\text{atmos}^{-1}$ at 600°K. Of the total integrated intensity per $(\text{cm}^{-1}) \times (\text{cm}^{-1}\text{-atmos}^{-1} \text{ of } \text{CO}_2)$ we associate 98.9 % with $\text{C}^{12}\text{O}_2^{16}$ and 1.1 % with $\text{C}^{13}\text{O}_2^{16}$. For the intense ν_3 -fundamental the effective band-width corresponds to the wave number range $\Delta\omega_{2349}$ for which $(\text{SpL}/0.7) \geq 0.1$ with the band-width of the isotopic species determined similarly. In general, the

$C^{12}O_2^{16}$ - band is black before the isotopic band makes appreciable contributions. The emissivities at 600°K, E_{2349} , calculated by the use of average absorption coefficients $\bar{P} = \alpha/\Delta\omega$, are summarized in Table III as a function of pL. Comparison of E_{2349} (total) with the

Table III. Emissivities E_{2349} for $C^{12}O_2^{16}$ and $C^{13}O_2^{16}$ at 600°K as a function of optical density. *

pL, cm-atmos.	pL, ft-atmos.	$E_{2349}(C^{12}O_2^{16})$	$E_{2349}(C^{13}O_2^{16})$	$E_{2349}(\text{total})$
0.1	0.0033	0.018	0.0005	0.019
0.5	0.0164	0.033	0.0016	0.035
1.0	0.033	0.037	0.0025	0.040
5.0	0.164	0.050	0.0070	0.057
15	0.492	0.051	0.011	0.062
50	1.64	0.052	0.011	0.063
100	3.28	0.053	0.012	0.065
200	6.56	0.055	0.012	0.067

* The tabulated values of $E_{2349}(C^{13}O_2^{16})$ correspond to the values which must be added to $E_{2349}(C^{12}O_2^{16})$ in order to obtain $E_{2349}(\text{total})$. They do not include emissivities in the (black) region of the ν_3 -fundamental of $C^{12}O_2^{16}$ which is overlapped by the ν_3 -fundamental of $C^{13}O_2^{16}$.

empirical data⁴ plotted in Fig. 5 shows that for pL ≤ 0.033 ft.-atmos, $E_{2349}(\text{total})$ is practically identical with the engineering emissivity E.

The result is in accord with expectations and shows clearly that useful emissivity data can be obtained with practically no analytical work once the physical principles are understood clearly.

(D). Contribution of the $02^{\circ}1 \rightarrow 00^{\circ}0$ and $10^{\circ}1 \rightarrow 00^{\circ}0$ Bands

From the measured integrated intensities⁷ we find $\alpha_{3609} = 9.70 \text{ cm}^{-2} \text{atmos}^{-1}$ for the $02^{\circ}1 \rightarrow 00^{\circ}0$ - band and $\alpha_{3716} = 14.4 \text{ cm}^{-2} \text{atmos}^{-1}$ for the $10^{\circ}1 \rightarrow 00^{\circ}0$ - band at 600°K . The effective band-widths extend from 3539 cm^{-1} to 3679 cm^{-1} and from 3646 cm^{-1} to 3716 cm^{-1} , respectively. Using average absorption coefficients the following results is obtained:

$$\begin{aligned} E_{3609} + E_{3716} \approx & 0.0047 \left[1 - \exp(-6.92 \times 10^{-2} \text{ pL}) \right] \\ & + 0.0017 \left[1 - \exp(-1.72 \times 10^{-1} \text{ pL}) \right] \\ & + 0.0017 \left[1 - \exp(-1.03 \times 10^{-1} \text{ pL}) \right]. \end{aligned} \quad (13)$$

(E). The Total (Engineering) Emissivity

The total (engineering) emissivity is obtained by adding the partial emissivities from selected spectral regions. Since estimates of partial emissivities were not included for all of the important vibration-rotation bands, no general expression for the engineering emissivity can be given although useful results have been obtained for small values of the optical density (compare $\int \epsilon_{\text{EC}}$ and Fig. 5).

APPENDIX I. RELATIVE INTENSITY CALCULATIONS FOR CO₂ USING HARMONIC OSCILLATOR APPROXIMATIONS*

It is well known that harmonic bands with non-zero matrix components are predicted even to the harmonic oscillator approximation. Calculations of this sort are useful for rough relative intensity estimates although mechanical as well as electrical anharmonicity corrections are neglected. Representative relative intensity estimates have been given, for example, by Benedict¹⁵ and by Kaplan.¹⁶ For the present applications it will be convenient to present a general equation for relative intensities of harmonic bands involving changes in the vibrational quantum number ν_2 .

The desired result is obtained readily by using the normalised wave functions for the isotropic plane oscillator and an integral involving associated Laguerre polynomials first obtained by Schrödinger.¹⁷ In this manner it is easily shown that

$$\left[\beta(n_1 n_2^{\ell} n_3 \rightarrow n_1' n_2'^{\ell'} n_3) \right]^2 / \left[\beta(n_1 0^0 n_3 \rightarrow n_1 1^1 n_3) \right]^2 = R(n_2^{\ell} \rightarrow n_2'^{\ell'}) =$$

$$\left[k! k'! / (k + \ell)! \right] (p!)^2 \left\{ \sum_{\tau=0}^{\min(k, k')} \binom{p - \ell}{k - \tau} \binom{p - \ell'}{k' - \tau} \binom{-p - 1}{\tau} \right\}^2 \quad (A-1)$$

* The author is indebted to Drs. W. S. Benedict and L. D. Kaplan for helpful correspondence concerning intensity estimates for harmonic bands of CO₂.

¹⁶ L. D. Kaplan, J. Chem. Phys. 18, 186 (1950).

¹⁷ E. Schrödinger, Ann. d. Physik. 80, 483 (1926).

where $k = (1/2)(n_2 - l)$, $k' = (1/2)(n_2' - l')$, $p = (1/2)(l + l' + 1)$, and $\binom{n}{k}$ denotes a binomial coefficient.

By combining Eqs. (7) to (9) with Eq. (A-1) the following useful result is obtained:

$$\begin{aligned} \alpha(n_1 n_2^l n_3 \rightarrow n_1 n_2'^{l'} n_3) / \alpha(n_1 0^0 n_3 \rightarrow n_1 1^1 n_3) &= \xi(n_2^l \rightarrow n_2'^{l'}) \\ &= g_2 R(n_2^l \rightarrow n_2'^{l'}) \left[\gamma(n_1 n_2^l n_3 \rightarrow n_1 n_2'^{l'} n_3) / \gamma(n_1 0^0 n_3 \rightarrow n_1 1^1 n_3) \right] \\ &\quad \times \left\{ \exp \left[\left(W_V(n_1, n_2, n_3, l) - W_V(n_1, 0, n_3, 0) \right) / kT \right] \right\} \\ &\quad \times \left\{ 1 - \exp \left[-h \nu(n_2^l \rightarrow n_2'^{l'}) / kT \right] \right\} \times \left\{ 1 - \exp \left[-h \nu(0^0 \rightarrow 1^1) / kT \right] \right\}^{-1}. \quad (A-2) \end{aligned}$$

Numerical values of $R(n_2^l \rightarrow n_2'^{l'})$ calculated from Eq. (A-1) are identical with Benedict's estimates,¹⁵ which utilized data given by Shaffer.¹⁸

Following Benedict we shall assume that resonance between $2\nu_2$ and ν_1 is exact, an approximation which will not change the order of magnitude of the calculated results although it is not in accord with observed infrared and Raman intensities. By utilizing Eqs. (A-1) and (A-2) relative intensities have been computed for a number of bands in the 5μ region at 300 and at 600°K. The results are summarized in Table (A-1).

¹⁸ W. H. Shaffer, Rev. Mod. Phys. 16, 245 (1944).

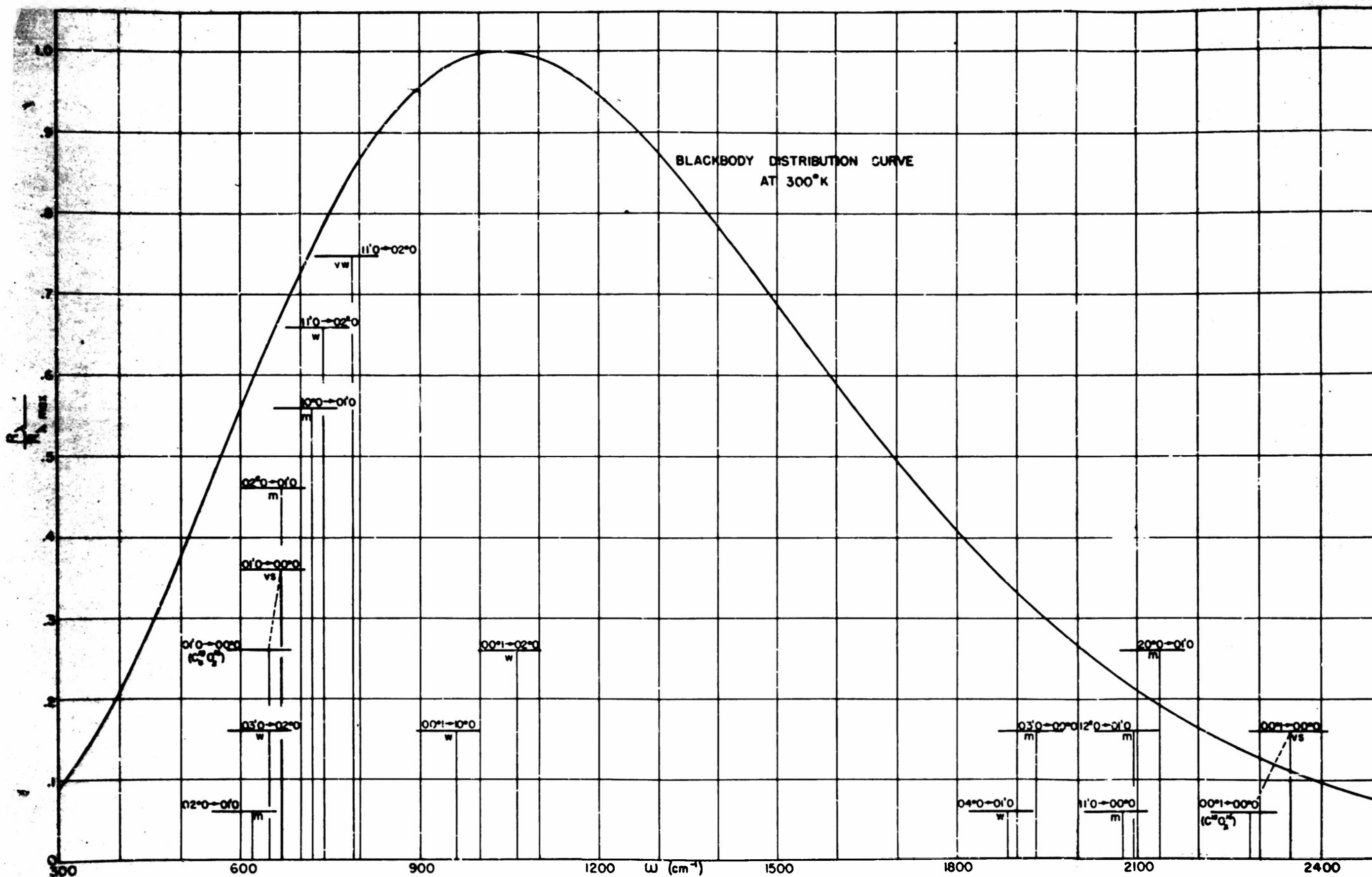
Table (A-1). Relative intensities for harmonic bands in the 5μ region based on harmonic oscillator wave functions.

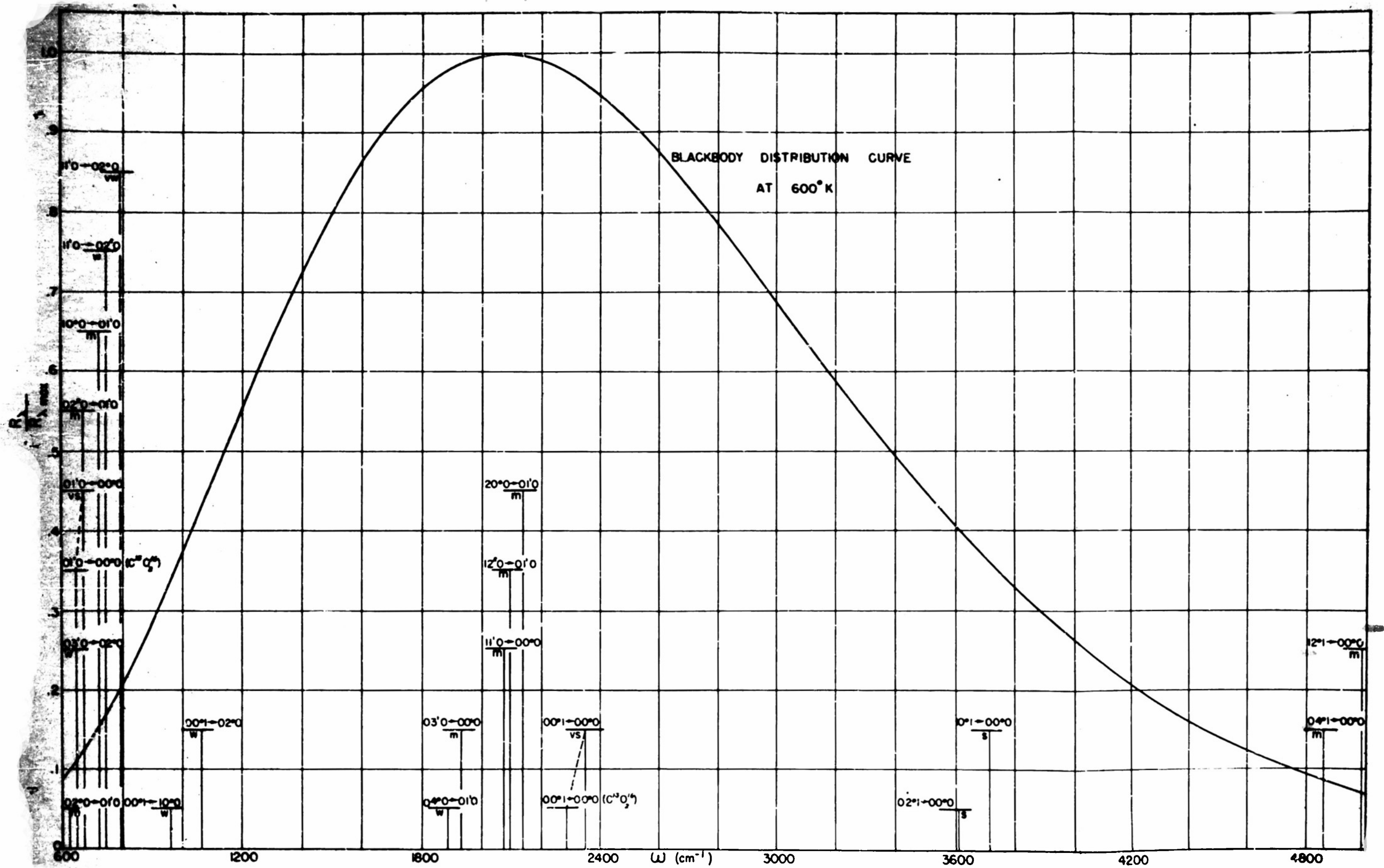
Transition	$S_L R(n_1 n_2 \ell_0 \rightarrow n_1' n_2' \ell_0')$	$S_L R(n_1 n_2 \ell_0 \rightarrow n_1' n_2' \ell_0')$ at 300°K	$S_L R(n_1 n_2 \ell_0 \rightarrow n_1' n_2' \ell_0')$ at 600°K
$00^0 0 \rightarrow 11^1 0$	1	1	1
$01^1 0 \rightarrow 12^2 0$	4	0.164	0.810
$01^1 0 \rightarrow 20^0 0$	0.9*	0.0376	0.187
$01^1 0 \rightarrow 12^0 0$	0.1*	0.00395	0.0196
$02^2 0 \rightarrow 13^3 0$	6	0.0101	0.243
$03^3 0 \rightarrow 14^4 0$	8	0.000315	0.0507
$02^2 0 \rightarrow 21^1 0$	0.9*	0.00155	0.0382
$10^0 0 \rightarrow 21^1 0$	1.8*	0.00238	0.0656
$11^1 0 \rightarrow 30^0 0$	0.9*	0.0000438	0.00638

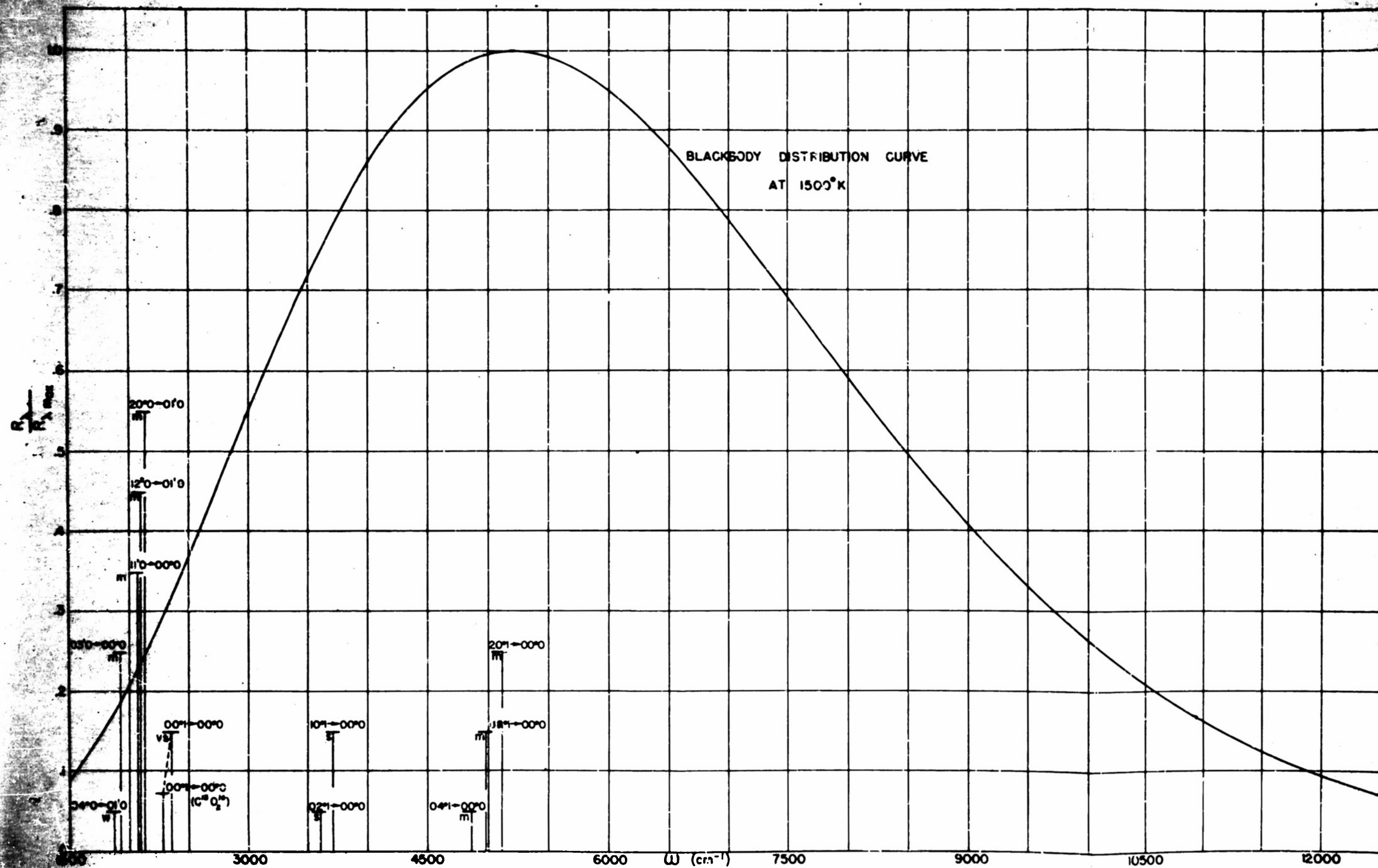
Adjusted according to Benedict to concentrate most of the intensity of resonating bands in the highest frequency member. 15

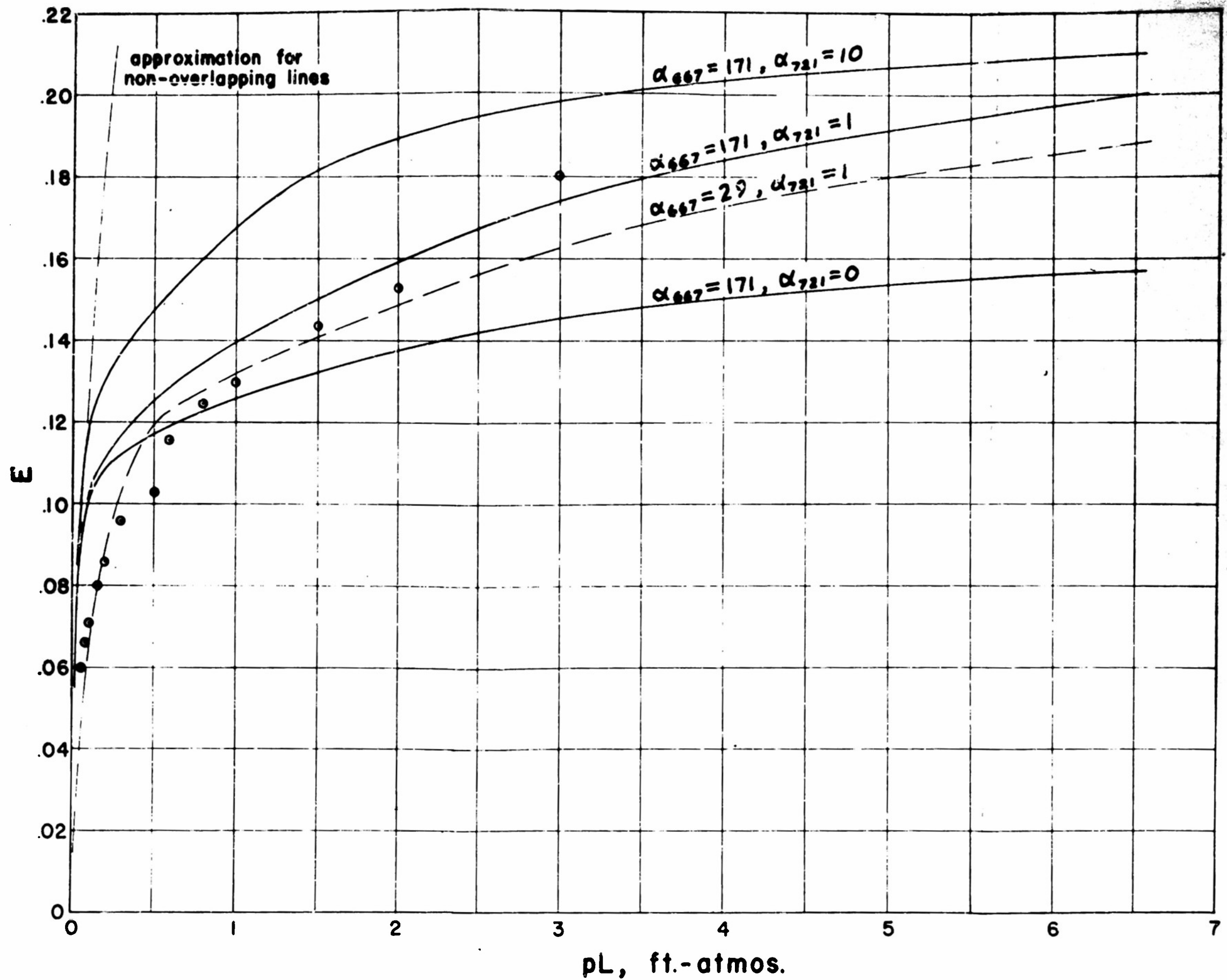
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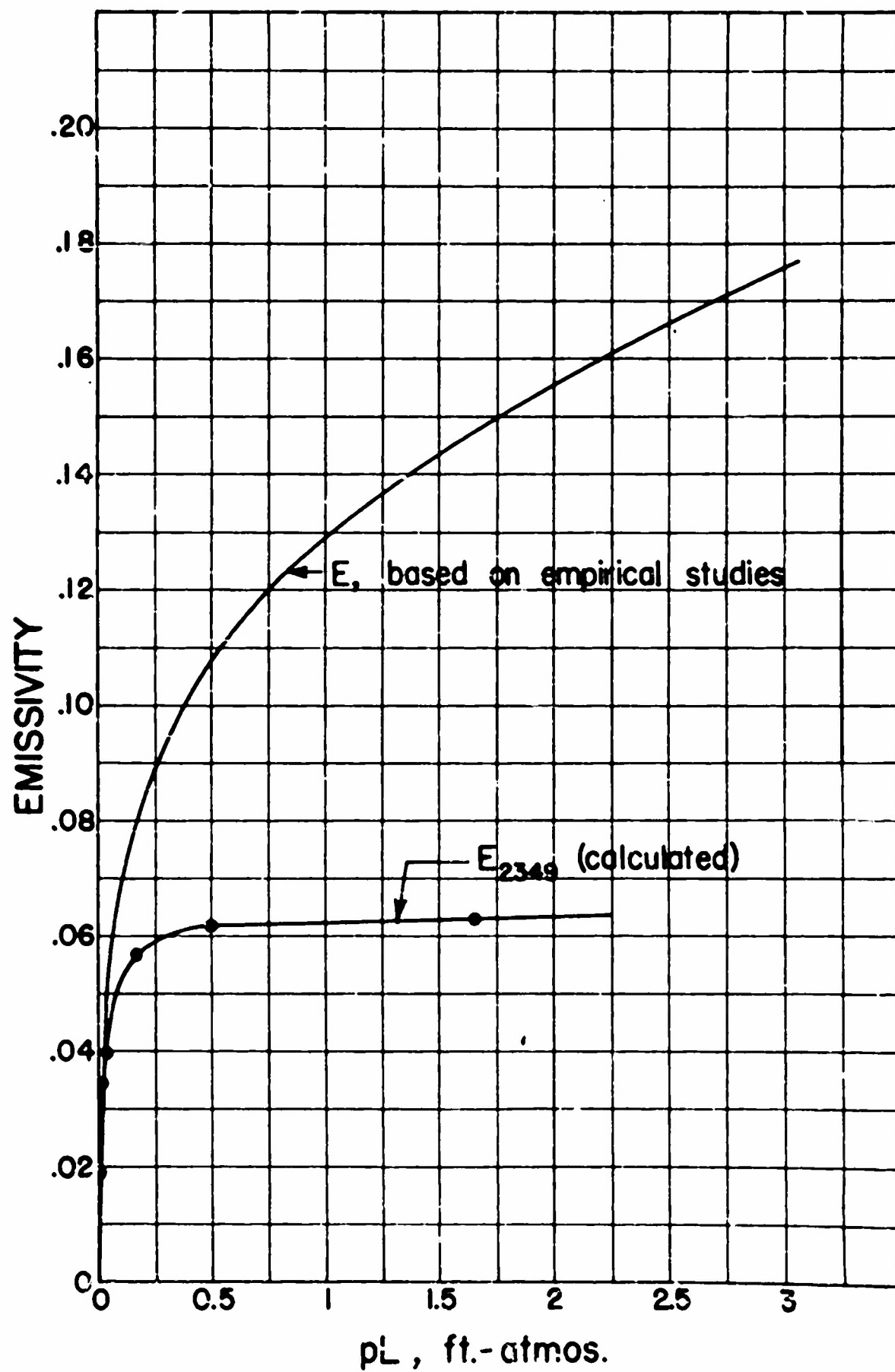
- Fig. 1.** Positions of centers of vibration-rotation bands observed at room temperature between 300 and 2400 cm^{-1} . The designations vs (very strong), s (strong), m (medium), w (weak), and vw (very weak) are those of Herzberg.¹⁴ Also shown is the intensity ratio $R_{\lambda}/R_{\lambda \text{ max}}$ for a blackbody at 300°K.
- Fig. 2.** Positions of centers of vibration-rotation bands observed at room temperature between 600 and 4800 cm^{-1} . Also shown is the intensity ratio $R_{\lambda}/R_{\lambda \text{ max}}$ for a blackbody at 600°K.
- Fig. 3.** Positions of centers of vibration-rotation bands observed at room temperature between 1500 and 12,000 cm^{-1} . Also shown is the intensity ratio $R_{\lambda}/R_{\lambda \text{ max}}$ for a blackbody at 1500°K.
- Fig. 4.** Engineering emissivity E as a function of pL for CO_2 at 300°K. Integrated intensities α are expressed in $\text{cm}^{-2}\text{-atmos}^{-1}$. The results based on empirical measurements (Cf. reference 4) are shown as circles.
- Fig. 5.** Engineering emissivity E as a function of pL for CO_2 at 600°K, based on the empirical correlations of H. C. Hottel and R. B. Egbert. Also shown are the calculated contributions to the total emissivity made by the ν_3 - fundamental.











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