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Power Saturation of the $J=1\rightarrow 2$ Rotational Transition of OCS

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The power saturation of seven lines arising from the $J=1\rightarrow 2$ rotational transition in OCS has been investigated with a Stark-cavity spectrometer. The experimental results on all investigated lines confirm the theory of Karplus and Schwinger adapted in this paper to the case of gas enclosed in a cylindrical cavity absorption cell oscillating in a TE_{0mn} mode with an arbitrary n-value, and m-value from 1 to 10.

INTRODUCTION

A QUANTUM-STATISTICAL treatment of power saturation of molecular transitions in a uniform radiation field has been given by Snyder and Richards,[1] and by Karplus and Schwinger.[2] The experimental results[3,4] on the saturation of the 3,3 line of ammonia ($N^{14}H_3$), thus far the only line investigated extensively, substantiate the theory of Karplus and Schwinger. This theory treats the saturation of each Zeeman component of the line independently and predicts an additional broadening of the line and a corresponding decrease of the peak intensity. The saturated peak intensity is obtained simply by adding the saturated intensities of individual Zeeman components.

In this paper the uniform-field theory of Karplus and Schwinger is applied to the case of a gas enclosed in a cavity absorption cell oscillating in a cylindrical TE_{0mn} mode with m ranging from 1 to 10, and n arbitrary.[5] The present theory is an extension and modification of the semiclassical treatment of power saturation in a cylindrical TE_{011} cavity given by Bleaney and Penrose.[4] Predictions of the theory are compared with results of measurements on the saturation of a series of lines arising from the $J=1\rightarrow 2$ rotational transition in carbonyl sulphide (OCS).

THEORETICAL NOTES

The theory of power saturation as developed by Karplus and Schwinger is applicable both to magnetic and electric dipole moment transitions. For microwave spectroscopy the latter transitions are by far the most important since only a few molecules have strong magnetic dipole moments. According to Karplus and Schwinger[2] the absorption intensity $\alpha^{(s)}(\nu)$ of an isolated line, resulting from electric dipole moment transition between molecular states a and b in the presence of a strong monochromatic radiation, can be written

$$\alpha^{(s)}(\nu) = C_\alpha \sum_\kappa \frac{\Delta\nu |p_{ab}{}^\kappa|^2}{(\nu-\nu_0)^2+(\Delta\nu)^2+(|p_{ab}{}^\kappa|\,|\mathbf{E}|/h)^2}\ \mathrm{cm}^{-1}, \quad (1)$$

where ν, $\mathbf{E}$ = frequency and electric field vector of the exciting field, respectively; ν_0 = resonance frequency of the line; κ = degeneracy index of the transition; $\Delta\nu$ = half-width of the line at half of its maximum intensity ("half-width" in the following) due to molecular collisions only; $p_{ab}{}^\kappa$ = nonvanishing dipole moment matrix element for transition between the degenerate states a and b; h = Planck's constant; and C_α = a constant independent of $\mathbf{E}$ and κ. The expression (1) is derived assuming that the line is broadened by molecular collisions only,[6] that each collision restores thermal equilibrium distribution, that the period of the exciting radiation is large compared to the duration of a molecular collision but small compared to the mean time between molecular collisions, and that the exciting external field is very weak compared to the internal molecular field.

The Lorentzian shape of a collision-broadened line is not affected by the strength of the exciting field. All desired information about the power saturation of a transition is thus obtained by considering only the power dependence of the peak intensity $\alpha_{\max}{}^{(s)}$. For $|\mathbf{E}|\rightarrow 0$, and $\nu=\nu_0$, $\alpha_{\max}{}^{(s)}$ becomes the unsaturated peak intensity $\alpha_{\max}$. Since, $\alpha_{\max}=\sum_\kappa(\alpha_\kappa)_{\max}$, it follows from Eq. (1) that

$$R(\alpha)=\frac{\alpha_{\max}{}^{(s)}}{\alpha_{\max}}=\frac{\sum_\kappa |p_{ab}{}^\kappa|^2 R_\kappa(\alpha)}{|p_{ab}|^2}, \quad (2)$$

where

$$|p_{ab}|^2=\sum_\kappa |p_{ab}{}^\kappa|^2,$$

$$R_\kappa(\alpha)=\frac{(\alpha_\kappa{}^{(s)})_{\max}}{(\alpha_\kappa)_{\max}}=\left[1+\left(\frac{|p_{ab}{}^\kappa|\,|\mathbf{E}|}{h\Delta\nu}\right)^2\right]^{-1}, \quad (3)$$

and $(\alpha_\kappa{}^{(s)})_{\max}$, $(\alpha_\kappa)_{\max}$ are the saturated and unsaturated

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[1] H. S. Snyder and P. I. Richards, Phys. Rev. **73**, 1178 (1948).

[2] R. Karplus and J. Schwinger, Phys. Rev. **73**, 1020 (1948).

[3] W. V. Smith and R. L. Carter, Phys. Rev. **72**, 638 (1947); R. L. Carter and W. V. Smith, Phys. Rev. **73**, 1053 (1948).

[4] B. Bleaney and R. P. Penrose, Proc. Phys. Soc. (London) **60**, 83 (1948).

[5] A. Dymanus, Rev. Sci. Instr. **30**, 191 (1959).

[6] This assumption implies that the pressure of the gas must be sufficiently high to make the broadening due to Doppler effect and to collisions with the walls of the absorption cell negligibly small compared to the broadening due to molecular collisions. The natural line width is of the order of 10^{-4} cps at microwave frequencies and can thus be disregarded, except for transitions between excited electronic states where it may become much larger than the collision width.

peak intensity of the κth component of the line, respectively. In a cavity the exciting field, and hence the amount of saturation, depends on the position of the absorbing molecule in the cell. To obtain the observable value of $\alpha_{\max}{}^{(s)}$ an average value of this quantity has to be computed over the volume of the cavity. This average value $\langle\alpha_{\max}{}^{(s)}\rangle_{\mathrm{Av}}$ is given by

$$\langle\alpha_{\max}{}^{(s)}\rangle_{\mathrm{Av}}=\int_V \alpha_{\max}{}^{(s)}(r,\theta,z)E_s(r,\theta,z)dV \Big/ \int_V E_s(r,\theta,z)dV. \quad (4)$$

Herein, $\alpha_{\max}{}^{(s)}(r,\theta,z)$ and $E_s(r,\theta,z)$ are the peak intensity of the line and the energy density, respectively, in an infinitesimal volume dV at a point described by cylindrical coordinates r, θ, z.

In the Appendix to this paper $\langle R_\kappa(\alpha)\rangle_{\mathrm{Av}}$ is calculated for the case of a gas enclosed in a cylindrical cavity oscillating in a TE_{0mn} mode with $m=1-10$ and an arbitrary n. This type of cavity absorption cell has been used in the present investigation. As shown in the Appendix $\langle R(\alpha)\rangle_{\mathrm{Av}}$ at the power P_c dissipated in the cavity is obtained from an appropriate curve in Fig. 2 and from Eq. (2) if the saturation constant $\Omega_{m\kappa}$ with

$$\Omega_{m\kappa}=\frac{4P_cQ_L|p_{ab}{}^\kappa|^2C_{0m}}{\nu_0Vh^2C_\nu{}^2P^2},$$

is known for all κ components of the line. In the expression for $\Omega_{m\kappa}$, Q_L and V are the loaded Q-factor and volume of the cavity, respectively, C_{0m} the mode constant listed in the Appendix Table I, C_ν the halfwidth constant, and P the pressure in the absorption cell.

The results given in the Appendix hold for all transitions which comply with the assumptions mentioned at the beginning of this section. For rotational transitions between molecular states (J, K, M) and (J', K', M') of molecules with a permanent dipole moment p, the dipole moment matrix elements $|p_{ab}{}^\kappa|$ are

$$|p_{ab}{}^\kappa|=p|D_{J,K,M}{}^{J',K',M'}|. \quad (5)$$

Here $|D_{J,K,M}{}^{J',K',M'}|$ is the direction cosine matrix element connecting the two states; J is the rotational quantum number, K the quantum number of component of **J** along molecular axis, and M the magnetic quantum number. The unprimed quantities J, K, M, and the primed ones J', K', M', correspond to the initial and the final state of the transition, respectively. The degeneracy index κ is now M, and the sum in Eq. (5) is taken over all Zeeman components of the line.

For p in Debye units, ν_0 in Mc/sec, C_ν in (Mc/sec)/mm Hg, P in mm Hg, and P_c in watts, the expression for Ω_{mM} ($=\Omega_{m\kappa}$ with $\kappa=M$, see above) can be written

$$\Omega_{mM}=9.1\times10^5\frac{p^2P_cQ_LC_{0m}}{V\nu_0C_\nu{}^2P^2}|D_{J,K,M}{}^{J',K',M'}|^2. \quad (6)$$

EXPERIMENTAL PROCEDURE

A superheterodyne spectrometer, with Stark modulation at a frequency of 400 cps, and a pill-box shaped cylindrical Stark cavity absorption cell[5] were used throughout this investigation. The cavity was designed for use in any cylindrical TE_{0m1} mode with m from about 5 to about 12, but all present measurements were performed in the TE_{061} mode. The modulating Stark voltage (zero-biased square wave) was applied in this cavity between plunger and bottom. Owing to the high sensitivity of the spectrometer (minimum detectable absorption intensity $\alpha_{\min}\simeq2\times10^{-10}\mathrm{cm}^{-1}$) saturation measurements could also be performed on weak lines ($\alpha_{\max}\simeq3\times10^{-7}\ \mathrm{cm}^{-1}$) and at low pressures.

The unsaturated peak intensities $\alpha_{\max}$, and the ratios $\langle R(\alpha)\rangle_{\mathrm{Av}}$ at a given power and pressure of the gas were determined by the antimodulation method[7] with an error of about 2.5 and 1%, respectively. The largest contribution to the latter error came from incomplete splitting[8] of the lines.

The OCS lines possessing an essentially first order Stark effect (l-doublet lines) can easily be split completely and offer no difficulties. However, the lines possessing a second order Stark effect are hard to split. In the present investigation the maximum available Stark field was about 1650 v/cm. In the absence of power saturation the second order lines could be split completely by this field only at pressures lower than ~0.03 mm Hg, the error in $\alpha_{\max}$ increasing to several percent at higher pressures. At strong saturation the error in $\langle R(\alpha)\rangle_{\mathrm{Av}}$ due to incomplete splitting can thus be considerable since it is roughly equal to $\alpha_{\max}/\alpha_{\max}{}^{(s)}$ times the error in $\alpha_{\max}$ due to the same phenomenon. By using special extrapolation methods[7,9] this error could be reduced to about one percent.

The pressure used in the present measurements varied from 0.02 to about 0.06 mm Hg. It was measured with a precision micromanometer with an accuracy of about one percent. Errors in $\langle R(\alpha)\rangle_{\mathrm{Av}}$ due to pressure drift have been eliminated by the procedure described elsewhere.[9] The loaded Q-factor of the cavity was determined by the modified dynamic method[9] and was accurate to about one percent.

The determination of the power P_c was by far the the most difficult problem. Absolute power measurements were performed with a calibrated crystal detector and could be in error up to 25%. Relative power measurements were performed with a calibrated attenuator. The error in the latter measurements did not exceed 5%.

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[7] A. Dymanus, Physica (to be published).

[8] The ratio, $A(F)=[(\alpha_{\max})_{F\to\infty}-(\alpha_{\max})_{F=F}]/(\alpha_{\max})_{F\to\infty}$, where $(\alpha_{\max})_{F=F}$ and $(\alpha_{\max})_{F\to\infty}$ are the peak intensities measured at a Stark field F and at an infinitely strong field, respectively, is frequently used as a measure of the line splitting. In this paper a complete splitting corresponds to $A(F)<0.01$.

[9] A. Dymanus, thesis, Utrecht, 1958.

RESULTS AND DISCUSSION

Measurements on power saturation of OCS were performed on seven lines arising from the rotational transition $J=1\rightarrow 2$. Five of these lines originate from the most abundant species $O^{16}C^{12}S^{32}$ in five vibrational states: $(v_1v_2^{|l|}v_3)=(000)$, (100), $(01_1{}^10)$, $(01_2{}^10)$, (02^00) (lines Nos. 1, 3, 4, 5, and 6, respectively, in the following). The remaining two lines come from the isotopic species $O^{16}C^{13}S^{32}$ and $O^{16}C^{12}S^{34}$, both in the ground vibrational state (lines Nos. 2 and 7, respectively, in the following). The lines $(v_1v_2^{|l|}v_3)=(01_1{}^10)$, $(01_2{}^10)$ form the well-known l doublet.

The measured and the calculated ratio $\langle R(\alpha)\rangle_{\text{Av}}$ of the most intense line (line No. 1, the main line in the following) and of the $1_1{}^1$ component of the l doublet is shown in Fig. 1 as a function of the power P_c for a pressure of 0.025 mm Hg. The unit power in this figure corresponds to $P_c \simeq 1.4\times 10^{-4}$ w. All experimental points in Fig. 1 have been shifted to the right by about 0.1 P_c so as to make the measured and the calculated values of $\langle R(\alpha)\rangle_{\text{Av}}$ equal at $P_c=1$. The theoretical curves have been calculated with the aid of Eqs. (2) and (6) and Fig. 2, assuming the following values for the various constants in the expression (6) for Ω_{mM}: $C_{06}=61.6$ (Table I, TE_{061} mode of the cavity), $V=137$ cm^3, $Q_L=5300$, $C_\nu=6.30$ (Mc/sec)/mm Hg,[10,11] $p=0.700$ Debye unit for the l-doublet lines, and $p=0.709$ Debye unit for all other lines[12]; the matrix elements involved[13] are given by

$$|D_{J,K,M}{}^{J',K',M'}|^2=|D_{J,l,M}{}^{J+1,l,M\pm 1}|^2 =\frac{[(J+1)^2-l^2](J\pm M+1)(J\pm M+2)}{(J+1)^2(2J+1)(2J+3)},$$

with $K=l=1$, 0 for the l-doublet lines and for all other lines, respectively.[14]

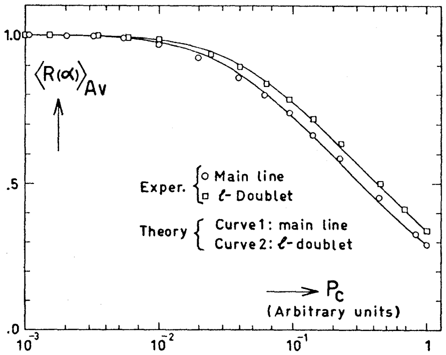

FIG. 1. Power saturation of the $J=1\rightarrow 2$ rotational transition of OCS; the measured and calculated peak intensity ratio $\langle R(\alpha)\rangle_{\text{Av}}$ of the main line and of the $1_1{}^1$ component of the l-doublet as a function of the power P_c dissipated in the cavity at a pressure of 0.025 mm Hg. The experimental points are shifted to the right by about $0.1P_c$ so as to make the measured and the calculated values of $\langle R(\alpha)\rangle_{\text{Av}}$ equal at $P_c=1$.

In the expression for $|D_{J,K,M}{}^{J',K',M'}|^2$, l is the quantum number of angular momentum due to excited degenerate bending vibration; the upper (+) sign corresponds with $M\rightarrow M+1$ transitions, and the lower (−) sign with $M\rightarrow M-1$ transitions.

The assumption of $C_\nu=6.3$ (Mc/sec)/mm Hg for all lines of OCS seems well justified. Recent measurements on the half-widths of the main line and of the l doublet performed with a new method[11] yield for C_ν the values of 6.27 ± 0.18 and 6.17 ± 0.23 (Mc/sec)/mm Hg, respectively. The former value agrees well with the mean value of 6.2 (Mc/sec)/mm Hg of the measurements of Feeny *et al.* (6.4 ± 0.15) and of Johnson and Slager (6.1 ± 0.35). Within our knowledge l-doublet C_ν has not been measured previously. As for the other lines, good agreement between the measured and the calculated peak intensities makes C_ν for these lines outside the limits of 6.3 ± 0.2 rather improbable.[7]

Within the total experimental error (~7%) of pressure, $\langle R(\alpha)\rangle_{\text{Av}}$, and relative power measurements, there is a good agreement between the experimental results and the predictions of the theory of Karplus and Schwinger if the shift of experimental points in Fig. 1 is disregarded. It has been found that the saturation (at a given P_c and P) of the lines Nos. 2, 3, 6, and 7, was the same as of the main line, and of the $1_2{}^1$ component of the l doublet the same as of the $1_1{}^1$ component. These results confirm that saturation of a rotational transition is independent of isotopic substitution or of vibrational state, provided the dipole moment matrix elements are the same. The power P_c required for a given saturation was found to be proportional to P^{-2} [Eq. (6)] for all investigated lines.

The theory of Karplus and Schwinger is based on the assumption that each collision restores thermal equilibrium distribution. In this investigation a systematic deviation of about 10% was found for all lines and at all pressures and powers. The power P_c needed for a given value of $\langle R(\alpha)\rangle_{\text{Av}}$ was about 10% lower than the power calculated with the theory of Karplus and Schwinger. These results would suggest that not all collisions are effective in restoring thermal equilibrium distribution thus making the effective value of Ω_{mM} larger than the theoretical value. In terms of relaxation times this would mean that the thermal relaxation time[4] (the mean time between collisions which are effective in restoring thermal equilibrium) is longer than the mean time between collisions which are effective in broadening the line. However, no definite conclusion can be drawn on this point in view of the large uncertainty of the absolute power level in this investigation.

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[10] Feeny, Lackner, Moser, and Smith, J. Chem. Phys. **22**, 79 (1954); C. M. Johnson and D. M. Slager, Phys. Rev. **87**, 677 (1952).

[11] Dymanus, Dÿkerman, and Zÿderveld, J. Chem. Phys. (to be published).

[12] R. G. Shulman and C. H. Townes, Phys. Rev. **77**, 500 (1950).

[13] In a TE_{0m1} cavity only the $\Delta M=\pm 1$ transitions are detected.

[14] G. H. Townes and A. L. Schawlow, *Microwave Spectroscopy* (McGraw-Hill Book Company, Inc., New York, 1955), Chap. 2.

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APPENDIX. CALCULATION OF $\langle R_\kappa(\alpha)\rangle_{\mathrm{Av}}$

In a cylindrical cavity oscillating in a TE_{0mn} mode the radial, azimuthal, and axial components of $\mathbf{E}$ (E_r, E_θ, and E_z, respectively) are[15]:

$$E_r = E_z = 0,$$

$$E_\theta = E_0 J_1(x_{0m} r/R)\sin(n\pi z/L), \qquad \text{(A,1)}$$

where R and L = radius and length of the cavity, respectively, $J_1(x)$ = Bessel function of the first kind and first order, and x_{0m} = mth zero of $J_1(x)$. Since $\alpha_{\max}$ and $(\alpha_\kappa)_{\max}$ are independent of r, θ, z, and E_s is proportional to $|\mathbf{E}|^2$, it follows from Eqs. (3), (4), and (A,1) that

$$\langle R_\kappa(\alpha)\rangle_{\mathrm{Av}} = I_{m\kappa}/I_{0m}, \qquad \text{(A,2)}$$

with

$$I_{0m} = 2\pi\int_0^L dz\int_0^R E_\theta^2 r dr,$$

and

$$I_{m\kappa} = 2\pi\int_0^L dz\int_0^R \frac{E_\theta^2 r dr}{1+[E_\theta|p_{ab}^\kappa|/(h\Delta\nu)]^2}.$$

The subscripts 0 and m of the integrals I_{0m} and $I_{m\kappa}$ are the same as used in the designation of the oscillation mode of the cavity.

By a straightforward calculation one obtains for the integral I_{0m}:

$$I_{0m} = VE_0^2 I_{0m}^*/x_{0m}^2,$$

where

$$I_{0m}^* = \int_0^{x_{0m}} sJ_1^2(s)ds, \qquad \text{(A,3)}$$

and

$$s = x_{0m} r/R.$$

The integral I_{0m}^* can be solved with the aid of the relation:

$$\int_{\lambda_1}^{\lambda_2} sJ_k^2(s)ds = \tfrac{1}{2}[s^2 J_k'^2(s) + (s^2-k^2)J_k^2(s)]_{\lambda_1}^{\lambda_2},$$

which is readily derived from the differential equation for the Bessel function of the kth order. With $k=1$, $\lambda_1=0$, $\lambda_2=x_{0m}$, and the recurrence relations

$$2J_k'(s) = J_{k-1}(s) - J_{k+1}(s),$$

$$(2k/s)J_k(s) = J_{k-1}(s) + J_{k+1}(s),$$

the integrals I_{0m}^* and I_{0m} become

$$= \tfrac{1}{2}x_{0m}^2 J_0^2(x_{0m}), \qquad \text{(A,4)}$$

$$I_{0m} = \tfrac{1}{2}VE_0^2 J_0^2(x_{0m}). \qquad \text{(A,5)}$$

TABLE I. Values of the constant C_{0m} and of the integral I_{0m}^* for $m=1-10$.

m	1	2	3	4	5	6	7	8	9	10
I_{0m}^*	1.19	2.22	3.22	4.23	5.24	6.24	7.24	8.24	9.24	10.2
C_{0m}	12.3	22.2	32.1	41.5	51.8	61.6	71.5	81.5	91.3	101

Numerical calculations of the values of I_{0m}^* for $m=1-10$ lead to the results given in Table I.

By introducing a new variable: $v=\cot(n\pi z/L)$, integration of the axial part of the integral $I_{m\kappa}$ can be performed in the ordinary way. The resulting expression for $I_{m\kappa}$ is then

$$I_{m\kappa} = \frac{VE_0^2}{x_{0m}^2\Omega_{m\kappa}}\left[x_{0m}^2 - 2\int_0^{x_{0m}} \frac{sds}{[1+\Omega_{m\kappa}J_1^2(s)]}\right], \qquad \text{(A,6)}$$

where

$$\Omega_{m\kappa} = \left(E_0\frac{|p_{ab}^\kappa|}{h\Delta\nu}\right)^2, \qquad \text{(A,7)}$$

is a constant, the saturation constant of the κth component of the line in the mth TE_{0mn} mode of the cavity.

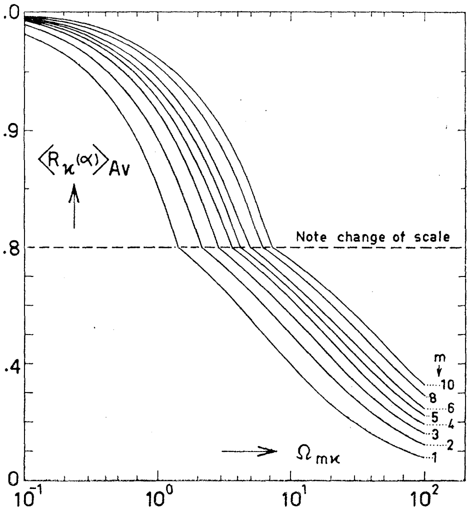

FIG. 2. Averaged peak-intensity ratio $\langle R_\kappa(\alpha)\rangle_{\mathrm{Av}}$ of the κth component of the line as a function of the saturation constant $\Omega_{m\kappa}$ for a cylindrical cavity oscillating in a TE_{0mn} mode with an arbitrary n-value and m-value from 1 to 10.

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[15] C. G. Montgomery, *Technique of Microwave Measurements*, Massachusetts Institute of Technology Radiation Laboratory Series (McGraw-Hill Book Company, Inc., New York, 1947), Vol. 11, Chap. 5.

From Eqs. (A,2) and (A,4–6), it follows that

$$\langle R_\kappa(\alpha)\rangle_{\mathrm{Av}}=\frac{1}{\Omega_{m\kappa}I_{0m}{}^*}\left[x_{0m}{}^2-2\int_0^{x_{0m}}\frac{sds}{[1+\Omega_{m\kappa}J_1{}^2(s)]^{\frac{1}{2}}}\right].\tag{A,8}$$

Apparently there is no known method to solve the integral in Eq. (A.8). This integral has been computed numerically for the values of $\Omega_{m\kappa}$ from 0.1 to 100 and for the m-values from 1 to 10. The resulting curves of $\langle R_\kappa(\alpha)\rangle_{\mathrm{Av}}$ *versus* $\Omega_{m\kappa}$ are given in Fig. 2. With the aid of the appropriate curve in this figure and Eq. (2) the saturated intensity of the line is readily obtained if $\Omega_{m\kappa}$ is known for all components of the line.

It is convenient to relate the saturation constant $\Omega_{m\kappa}$ to the power P_c, dissipated in the cavity as a result of conduction and dielectric losses. The total energy E_s stored in the cavity is connected with P_c by the well-known relation[15]:

$$E_s=P_cQ_L/(2\pi\nu_a),\tag{A,9}$$

where ν_a and Q_L are resonance frequency and loaded Q-factor of the cavity, respectively. On the other hand, from the definition of E_s and I_{0m} and from Eqs. (A,4) and (A,5) it follows that

$$E_s=(1/8\pi)I_{0m}=(1/8\pi)VE_0{}^2C_{0m}{}^{-1},\tag{A,10}$$

with

$$C_{0m}=x_{0m}{}^2/I_{0m}{}^*.\tag{A,11}$$

The constant C_{0m} in Eq. (A,10), in the following called "mode constant," depends only on the oscillation mode of the cavity. The value of C_{0m} for m from 1 to 10 is listed in Table I.

As the line is broadened only by molecular collisions, the half-width $\Delta\nu$ can be written

$$\Delta\nu=C_\nu P,\tag{A,12}$$

where, C_ν is the half-width constant and P pressure in the absorption cell. From Eqs. (A,7) and (A,9–12) the resulting expression for $\Omega_{m\kappa}$ is

$$\Omega_{m\kappa}=\frac{4P_cQ_L|p_{ab}{}^\kappa|^2C_{0m}}{\nu_0Vh^2C_\nu{}^2P^2},\tag{A,13}$$

in which ν_a is replaced by ν_0, since cavity is always tuned to the resonance frequency of the gas.