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TEMPERATURE DEPENDENCE OF OPTICALLY PUMPED FAR-INFRARED (FIR) LASER OUTPUT POWER

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ABSTRACT

The temperature dependence of the small signal gain and saturation power are derived using temperature-dependent rates in a four-level model. An expression is developed for the output power of a far-infrared oscillator as a function of temperature for both fixed pressure and fixed density. The results are valid in the regime of homogeneous broadening of the rotational transition and Doppler broadening of the pump transition. It is shown that, for most lasers, both the small signal gain and the saturation power decrease with increasing temperature. These effects have the overall result of increasing output power with decreasing temperatures.

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Introduction

This paper describes the temperature dependence of the power output of an optically pumped far-infrared (FIR) laser. The derived expressions are based on a rate-equation model with temperature-dependent rates.

The model used is a four-level scheme in which one level represents the lumped effects of all the nonlasing rotational levels in the upper vibrational manifold [1, 2, 3]. Figure 1 is a schematic representation of the model.

The rate equations describing the physical processes involved are given by:

$$dN_0/dt = W_v N_3 - W_p N_0 , \tag{1a}$$

$$dN_1/dt = B_{21} n(N_2 - g_2/g_1 N_1) - W_r N_1 , \tag{1b}$$

$$dN_2/dt = W_p N_0 - W_r N_2 - B_{21} n(N_2 - g_2/g_1 N_1) , \tag{1c}$$

$$dn/dt = B_{21} n(N_2 - g_2/g_1 N_1) - 2\pi n\delta , \tag{1d}$$

$$N = N_0 + N_1 + N_2 + N_3 . \tag{1e}$$

where

N = population of the 0-state in the absence of pumping,

N_0, N_1, N_2, N_3 = population densities of states 0, 1, 2, and 3, respectively,

n = number of FIR photons,

W_v, W_r = vibrational and rotational relaxation rates, respectively,

W_p = rate of pumping,

B_{21} = Einstein coefficient for stimulated interaction given by:

$$B_{21} = \frac{c^3 f(\nu)}{4\pi^2 \nu^2 t_{sp} \Delta\nu_N}, \tag{2}$$

where

ν = FIR frequency,

t_{sp} = spontaneous lifetime of the 2-state,

$\Delta\nu_N$ = homogeneous linewidth of the normalized lorentzian $f(\nu)$ for the 1→2 transition,

g_1, g_2 = degeneracy factors for levels 1 and 2,

δ = FIR cavity half-width.

These equations neglect spontaneous emission because the spontaneous lifetime of the FIR states is of the order of seconds. Moreover, it is assumed that W_v is smaller than the rotational relaxation rate, W_r, but is still large enough that the collisional populations of 1 and 2 can be neglected.

The rate equations (1a through 1d) can be solved for the steady-state case by setting all the time derivatives equal to zero. This results in an equilibrium FIR photon density given by:

$$n = \frac{g_1}{g_1 + g_2} \frac{W_r}{B_{21}} \left(\frac{N B_{21} W_p}{2\pi\gamma W_r (1 + W_p/W_\nu)} - 1 \right) \tag{3}$$

Using Rigrod's analysis of a homogeneously broadened oscillator [4], the FIR power emitted may be cast in the form:

$$P_{FIR} = \frac{1}{2} t P_s \left(\frac{2L\alpha}{t + a} - 1 \right), \tag{4}$$

where

t = relative coupling loss per double pass,

a = loss from other mechanisms,

α = small signal gain per unit length,

L = active length of the laser,

P_s = saturation power.

Defining the active cavity volume as V, equation 4 and the steady-state solutions yield:

$$\alpha = \frac{B_{21}}{cW_r} \frac{NW_p}{1 + W_p/W_v} , \tag{5}$$

$$P_s = \frac{h\nu V_c}{L} \frac{g_1}{g_1 + g_2} \frac{W_r}{B_{21}} \tag{6}$$

Typical values for some of the foregoing parameters are $t = 0.2$, $a = 0.5$, and $L = 1.7$ m, $\alpha = 2\ \mathrm{m}^{-1}$.

Temperature Dependences–Fixed Pressure

To obtain the temperature dependence of the power output of an FIR oscillator relative to its room-temperature performance, each component of P_s and α in equation 4 must be considered in detail. The dependences will be expressed in terms of a constant γ, which defines the operating temperature, T, by:

$$T = \gamma T^{\circ} \tag{7}$$

where T° and all quantities with the $^{\circ}$ superscript refer to $T^{\circ} = 300$K values. It is implicit to all results that the pressure and volume remain constant. The temperature dependence of each parameter is given and freely used in this section; all derivations are given in the Appendix.

The rate-equation model described refers to three rates, all of which vary with temperature. Their various dependences are given by:

$$W_r(\gamma) = W_r^{\circ}/\gamma\,, \tag{8}$$

$$W_v(\gamma) = W_v^{\circ}\gamma^{3/2}\,, \tag{9}$$

$$W_p(\gamma) = \frac{W_p^o}{\sqrt{\gamma}} \exp\left[Z\left(1 - \frac{1}{\gamma}\right)\right] , \tag{10}$$

$$Z = \frac{4 \ln(2) (\nu_p - \nu_p^o)^2}{(\Delta\nu^o_D)^2} , \tag{11}$$

where ν_p^o is the line-center frequency of the pump transition ($0 \rightarrow 2$), ν_p is the pump frequency, and $\Delta\nu_D^o$ is the Doppler linewidth of the inhomogeneously broadened transition.

The temperature dependence of the Einstein coefficient, B_{21}, is related to the variation of $W_r(\gamma)$ because of the relationship between the homogeneous linewidth, $\Delta\nu_N$, and the rotational relaxation rate,

$$W_r = \pi \Delta\nu_N . \tag{12}$$

Therefore, equation 12 can be substituted into equation 2 to obtain for $B_{21}(\gamma)$:

$$B_{21}(\gamma) = \frac{\theta(\nu)}{2\pi\left[(\nu - \nu_0)^2 + \left(\frac{\Delta\nu_N^o}{2\gamma}\right)^2\right]} , \tag{13}$$

where $\theta(\nu) = c^3/(4\pi^2\nu^2 t_{sp})$, $\Delta\nu_N^o$ is the room-temperature linewidth, and ν_0 is the line-center frequency of the FIR transition. At line center, equation 13 reduces to:

$$B_{21}(\gamma) = \frac{2\theta(\nu_0)\gamma^2}{\pi(\Delta\nu_N^o)^2} . \tag{14}$$

The dependence of N on temperature is given by:

$$N_{JK}(\gamma) = \frac{\rho^{\circ}}{\gamma} f_{v}^{\circ} f_{JK} \quad , \tag{15}$$

where ρ° is the room-temperature molecular density, f_{v}° is the fraction of molecules in the ground vibrational manifold, and f_{JK} is the fraction of molecules with rotational quantum numbers J and K. The f_{JK} characterize symmetric top molecules, but, in general, if the molecule is not a symmetric top, an appropriate distribution function for rotation can be used.

For a symmetic top with rotational constants B and C,

$$f_{JK}(\gamma) = \frac{T(I,K)\,(2J+1)}{\gamma^{3/2}} \left[\frac{B^{2}Ch^{3}}{\pi(kT^{\circ})^{3}}\right]^{1/2} \exp{}^{-}\left[\frac{hBJ(J+1)+(C-B)\,K^{2}}{kT^{\circ}\gamma}\right] . \tag{16}$$

T(I,K) is a statistical weight factor due to the nuclear spins and the degeneracy with respect to inversion. For molecules with three or more atoms, T(I,K) is bounded by 2 [5].

The thermal equilibrium number density of molecules in the 0-state with rotational quantum numbers J and K is given by:

$$N_{JK}(\gamma) = \frac{\rho^{\circ}}{\gamma}\left(1 - e^{\frac{-E_{02}}{kT^{\circ}\gamma}}\right) f_{JK}(\gamma) \quad , \tag{17}$$

where E_{02} is the energy separation between levels 0 and 2. Expanding the vibrational exponent and noting the dominant $\gamma^{-3/2}$ dependence of f_{JK} for low J and K results in the approximate expression:

$$N_{JK}(\gamma) \approx \frac{N^{\circ}_{JK}}{\gamma^{3/2}} \; , \tag{18}$$

where N°_{JK} is the room-temperature population density.

However, equation 18 is not appropriate for high J and K values because, at some critical point, the exponential portion of f_{JK} begins to dominate its behavior. When the $\gamma - 1$ term due to the ideal gas law is omitted, $N_{JK}(\gamma)$ will begin to grow with $\gamma > 1$ when

$$\left[\frac{d}{d\gamma}\left(N_J(\gamma)\right)\right]_{\gamma=1} > 0 \; . \tag{19}$$

This leads to the condition,

$$J(J+1) > \frac{5}{2}\,\frac{kT^{\circ}}{hB} \; . \tag{20}$$

For a typical molecule that is emitting in the submillimeter wavelength region, $\frac{kT^{\circ}}{hB} \approx 300$, which leads to $J > 20$. When constant pressure is maintained, the criterion becomes:

$$J(J+1) > \frac{7}{2}\,\frac{kT^{\circ}}{hB} \; , \tag{21}$$

which results in $J > 30$.

Figures 2 and 3 illustrate the dependence of $N_J(\gamma)$ for constant pressure and constant density on γ and J. The expression derived thus far can be substituted in equations 5 and 6 to determine $P_s(\gamma)$ and $\alpha(\gamma)$ at line center:

$$\alpha_{JK}(\gamma) = \frac{2\vartheta(\nu_0)\,W_v^o\,\rho^o\,E_{02}}{kT^o c\pi^2(\Delta\nu_N^o)^3}\,\gamma^{5/2}\,f_{JK}(\gamma)\left[\frac{W_p^o - W_v^o\gamma^2\exp\left[Z\left(\frac{1}{\gamma}-1\right)\right.}{W_p^o - W_v^o}\right] \qquad (22)$$

In the limit, $W_p^o \gg W_v^o$, and low J and K, the simple expression,

$$\alpha(\gamma) = \alpha^o\gamma \ , \qquad (23)$$

may be used, where α^o is the room-temperature value.

At constant pressure, the saturation power is given by:

$$P_s(\gamma) = \frac{P_s^o}{\gamma^3} \ . \qquad (24)$$

Equations 22, 23, 24, and 4 can be used to predict the temperature behavior. The result is particularly simple for low J values and is given by:

$$P_F(\gamma) = \frac{P_F^o}{(\phi - 1)}\left(\frac{\phi\gamma - 1}{\gamma^3}\right) . \qquad (25)$$

where $\phi = \dfrac{2L\alpha^o}{t + a}$, and P_F^o is the power emitted at room temperature. Figure 4 shows the power dependence of the constant-pressure system for low and high J values.

Temperature Dependences – Constant-Number Density

For a fixed molecular-number density in the laser tube, the following temperature dependences apply (line center):

$$W_r(\gamma) = W_r^\circ \quad , \tag{26}$$

$$B_{21}(\gamma) = B_{21}^\circ \quad , \tag{27}$$

$$W_v(\gamma) = W_v^\circ \gamma^{1/2} \quad , \tag{28}$$

$$W_p(\gamma) = \frac{W_p^\circ}{\gamma^{1/2}} \exp\left[Z\left(1 - \frac{1}{\gamma}\right)\right] , \tag{29}$$

$$N_{JK}(\gamma) = \rho^\circ \left[1 - e^{\frac{-E_{02}}{kT^\circ \gamma}}\right] f_{JK}(\gamma) \quad . \tag{30}$$

The foregoing equations give:

$$\alpha(\gamma) \approx \frac{\alpha^\circ}{\gamma^2} \quad \text{(low J values only)} \quad , \tag{31}$$

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$$P_s(\gamma) = P_s^\circ \quad . \tag{32}$$

Equations 28 and 29 result in a power-output dependence given by:

$$P_F(\gamma) \approx \frac{P_F^\circ}{\phi - 1}\left(\frac{\phi}{\gamma^2} - 1\right) \quad . \tag{33}$$

Figure 5 illustrates the power output as a function of temperature for a sealed-off laser tube. It is evident that low J values are favored by low temperatures, whereas high J transitions improve their output with increasing temperatures.

Discussion

It has been shown that the small signal gain at the FIR transition is highly dependent on temperature for both constant-pressure and constant-density operations, primarily because of the sensitivity of the ground-state population on temperature. Moreover, it is shown that this population, and therefore α, are dependent on the J values of the ground-state level. Lowering the temperature favors the low J values, whereas raising the temperature favors high J values. However, because most of the transitions observed are restricted to moderate values of J, it is expected that most FIR lasers would have increased small signal gains at lower temperatures. This is especially important for traveling-wave FIR amplifers.

The power coupled out of the FIR oscillator is more temperature-sensitive in constant-pressure systems because the saturation power is proportional to T^{-3}. This makes it harder to saturate the transition at lower temperature. In both constant-pressure and constant-density systems, the net power coupled out as a function of temperature is dependent on J. This effect should be most pronounced for the constant-density system.

The analysis suggests that, for high-gain FIR amplifiers, cooling an FIR gas (operating on a $J < 10$) would result in dramatic increases in single-pass gains. The same suggestions are recommended for a constant-pressure oscillator system.

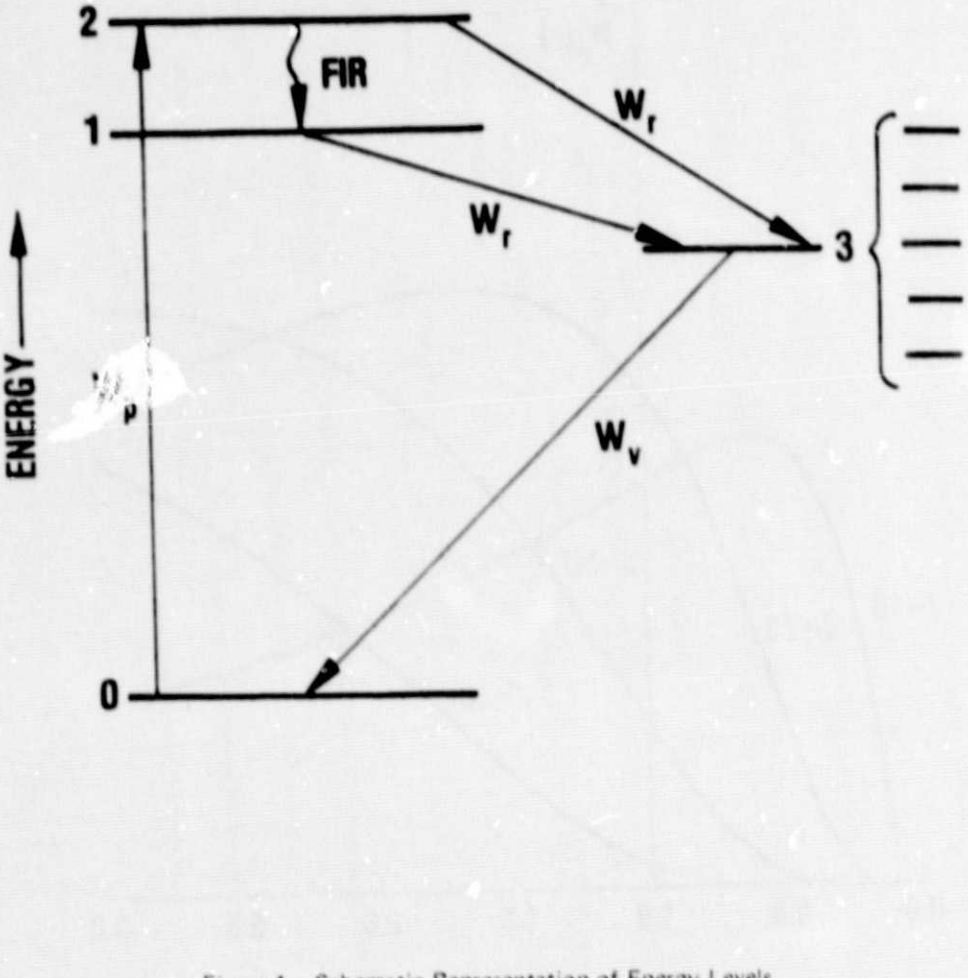

Figure 1. Schematic Representation of Energy Levels Used in the Four-Level Rate Model

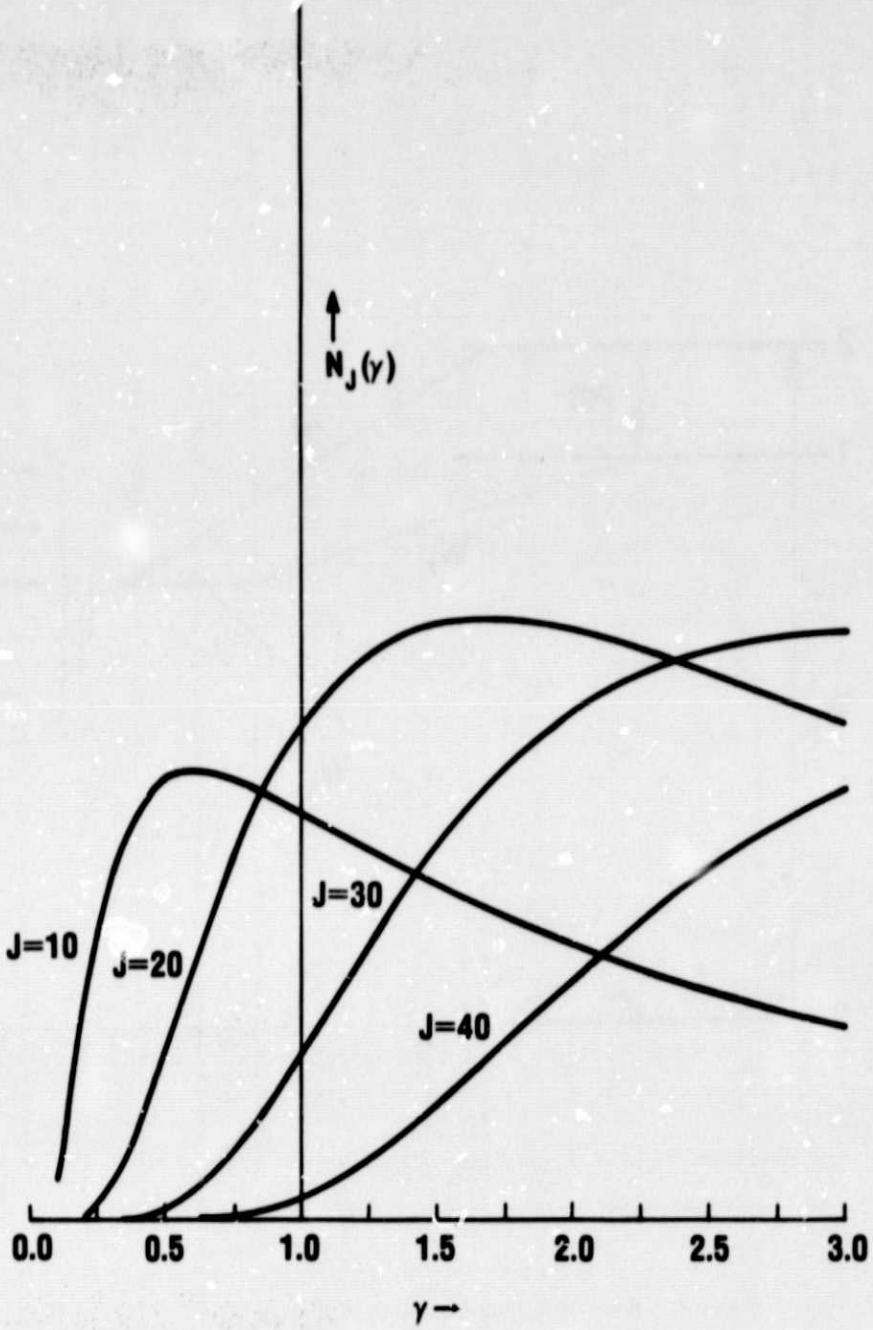

Figure 2. Population Density of 0-State in the Absence of Pumping (constant density)

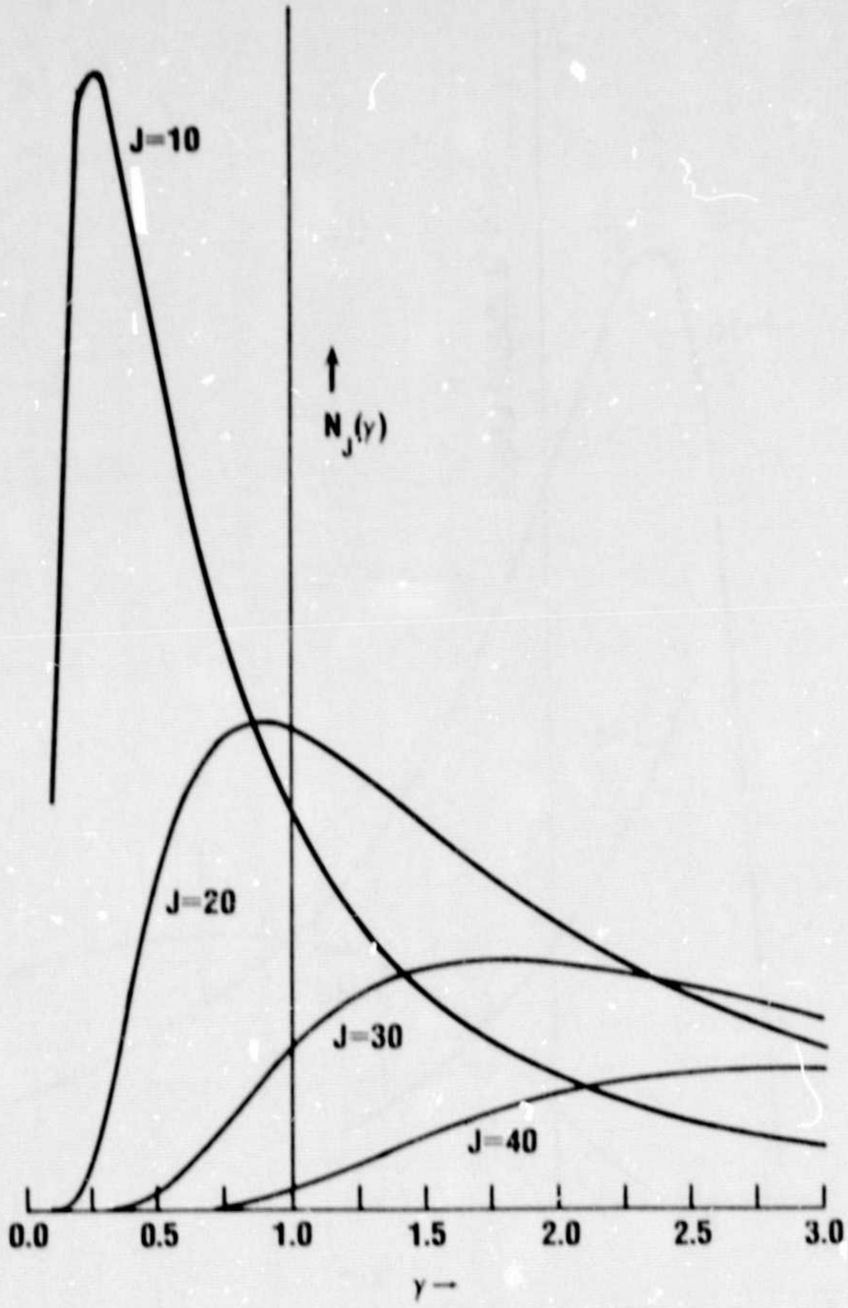

Figure 3. Population Density of 0-State in the Absence of Pumping (constant pressure)

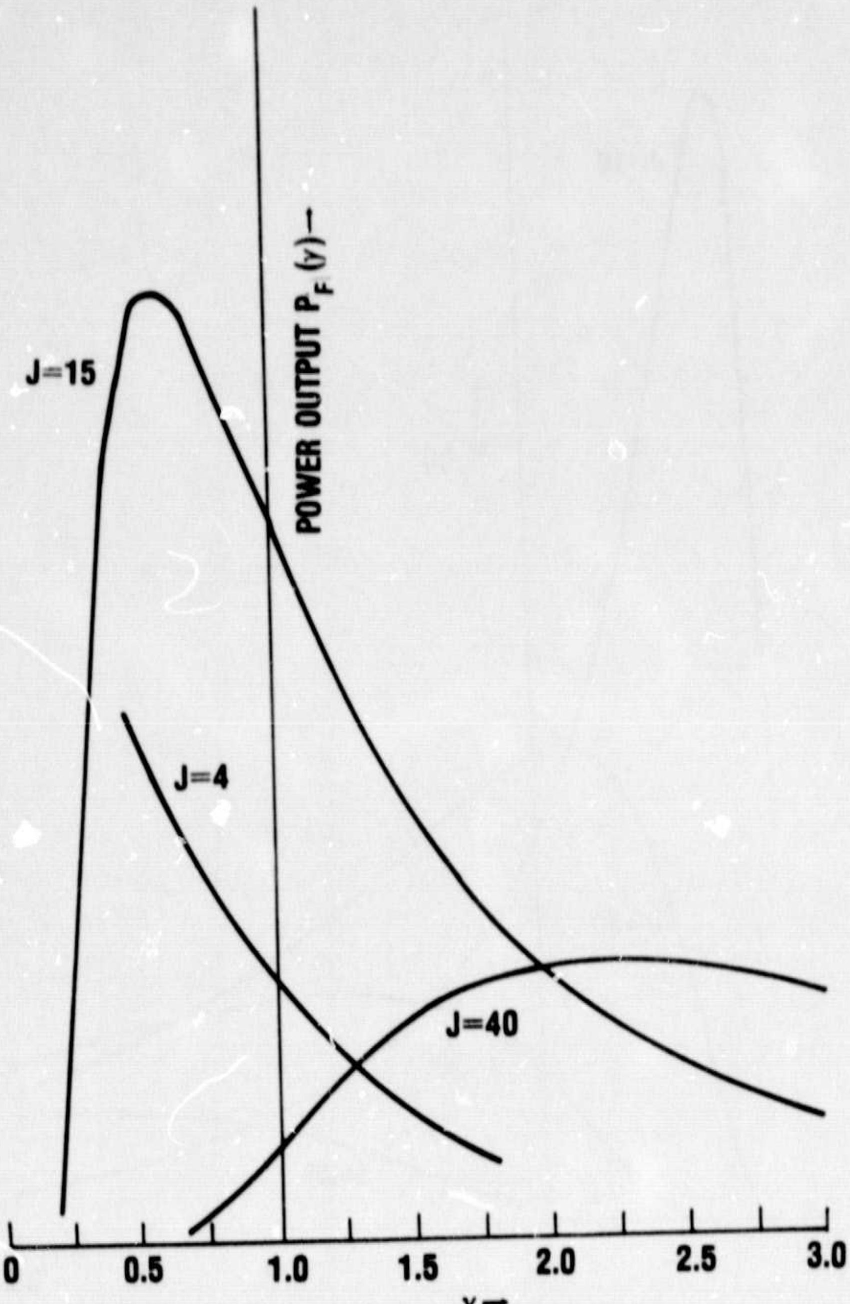

Figure 4. Power Output at Constant Pressure

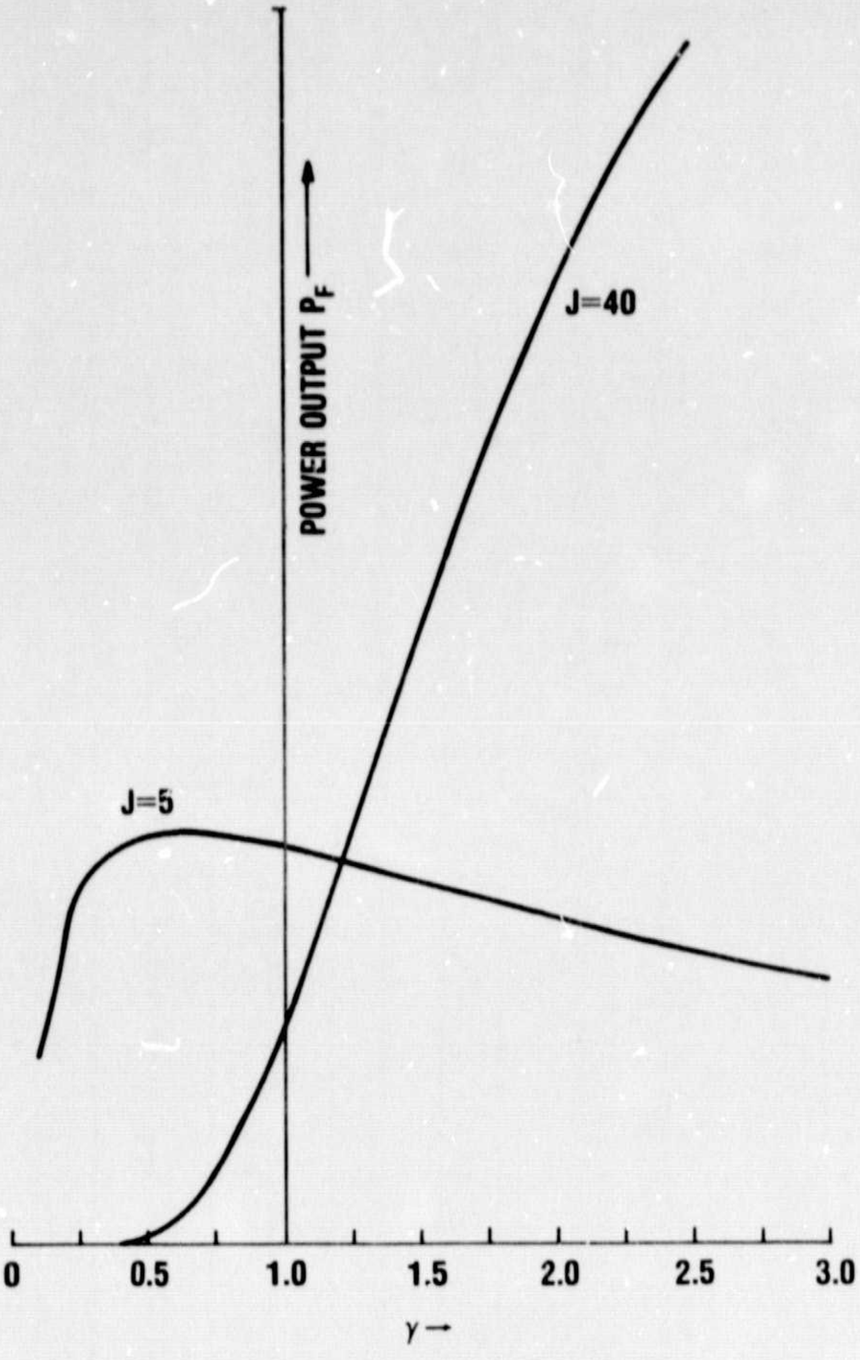

Figure 5. Power Output at Constant Density

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APPENDIX

In this appendix, the temperature dependences of the three rates, W_p, W_r, and W_v, are derived.

Rate of Pumping

The rate of pumping, W_p, is given by:

$$W_p = \frac{c^2 I(\nu_p)}{8\pi h \nu^3 t_{sp}} g(\nu) \quad ,$$

where $I(\nu_p)$ is the intensity at the pumping region, and $g(\nu)$ is a normalized Gaussian that describes the 0 → 2 transition, which is Doppler-broadened. $g(\nu)$ is given by:

$$2\left(\frac{\ln(2)}{\pi}\right)^{1/2}\left(\frac{1}{\Delta\nu_D}\right)\exp\left\{-\left[4\ln(2)(\nu_p-\nu_p^\circ)^2/\Delta\nu_D^2\right]\right\} \quad ,$$

where

$$\Delta\nu_D = 2\nu_p^\circ\sqrt{\frac{2kT}{Mc^2}\ln(2)} \quad .$$

If it is assumed that temperature changes do not affect the average pump intensity inside the FIR cavity, then only the line shape function changes with temperature. In particular, the Doppler width, $\Delta\nu_D$, and the exponential are the varying quantities. This results

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in a temperature dependence given by:

$$W_p(\gamma) = \frac{W_p^\circ}{\sqrt{\gamma}} \exp\left[Z\left(1 - \frac{1}{\gamma}\right)\right] ,$$

where Z is a constant defined in terms of room-temperature values.

Homogeneous Linewith

It was stated previously that the rotational relaxation rate is proportional to the homogeneous linewidth of the FIR transition, $\Delta\nu_N$. The effects of temperature changes on $\Delta\nu_N$ result from the velocity dependence of the collision cross section between two molecules. Intermolecular interactions may be treated by perturbation theory with interaction Hamiltonians of the form, r^{-n}. Because most FIR molecules either are symmetric tops or can be approximated as such (for example, CH_3 OH), their strongest interaction is that of two dipoles. This interaction has the form, $V(r) = [\mu_1 \cdot \mu_2 - 3(\mu_1 \cdot r)(\mu_2 \cdot r)/r^2)]\ r^{-3}$, where μ_1 and μ_2 are dipole moment vectors, and r is the vector that separates them. The r^{-3} term is the first-order term because symmetric tops have a dipole-moment component that maintains its orientation relative to the angular momentum vector.*

From considerations involving the matrix elements for the r^{-n} interaction and total cross sections for collision, it can be shown that the homogeneous linewidth is proportional to:

$$\rho V^{1-2/(n-1)} ,$$

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*C. H. Townes and A. L. Schawlow, *Microwave Spectroscopy*. McGraw-Hill. New York, 1955, pp. 359-360.

where ρ is the density of molecules, and V is the molecular velocity. Moreover, because $\rho \propto T^{-1}$, $V \propto T^{1/2}$, and $n = 3$ for symmetric tops, $\Delta\nu_N = \Delta\nu_N^o/\gamma$ for constant pressure and $\Delta\nu_N = \Delta\nu_N^o$ for constant density.

Vibrational Relaxation Rate

The vibrational relaxation rate, W_v, is the rate at which molecules return to the ground vibrational manifold. This rate has been attributed predominantly to the collisions of vibrationally excited molecules with the container walls. It is assumed that each collision with the wall results in the nonradiative deexcitation of the molecule. Therefore, the rate of wall collisions is approximately given by the reciprocal of the time it takes for a molecule to diffuse from the center of the tube to the wall. This is given by:

$$\frac{8D}{d^2} \quad ,$$

where D is the self-diffusion coefficient, and d is the laser-tube diameter. Simple billiard-ball diffusion theory gives:

$$D = V\lambda/3 \quad ,$$

where V is the mean velocity of a molecule, and λ is the mean free path. This results in:

$$D \propto \frac{\sqrt{\gamma}}{\rho} \quad .$$

This relationship leads to:

$$W_v(\gamma) = W_v^{o}\,\gamma^{3/2} \quad \text{(constant pressure)},$$

$$W_v(\gamma) = W_v^{o}\,\gamma^{1/2} \quad \text{(constant density)}.$$