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Lorentz Broadening in the Modeling of the HF Chemical Laser

Aerophysics Laboratory
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El Segundo, Calif. 90245

8 June 1977

Interim Report

Prepared for

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This technical report has been reviewed and is approved for publication.

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The results of a survey of pressure-broadening data in the literature, pertaining to species present in H_2-F_2 chemical lasers are described. Pressure-broadened linewidths of the HF vibration/rotation transitions were found to depend strongly on the vibrational and rotational quantum numbers, as well as on the perturbing species. The experimental data were incorporated into a rate equation model of the pulsed H_2-F_2 laser; where experimental data were unavailable, theoretical values were used. The calculations made from

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this model demonstrate the importance of including such detailed pressure-broadening information. Recent experimental results of Kwok and Cohen for HF V → R, T deactivation were also included in the model and were taken to be a multiquantum process. Comparisons of model predictions for pulse energy and pulse duration with several atmospheric-pressure laser measurements showed good agreement.

SUMMARY

The results of a survey of pressure broadening data in the literature pertaining to species present in the H_2-F_2 chemical laser are presented. The HF pressure broadened linewidths are shown to have strong dependences on the vibrational and rotational quantum numbers as well as on the nature of the species that serve as diluent in the gas mixture.

Results of calculations from the $H_2 + F_2$ laser code SPIKE, which included a detailed model for line-broadening, indicate that the incorporation of the detailed v, J and species dependence is important for achieving accurate predictions of laser performance. In addition to significant effects on total laser energy (increases or decreases, depending on species present), the detailed broadening model predicts a larger fraction of the output to be in the higher rotational transitions, and a longer pulse duration.

The recent experimental results of Kwok, Cohen, and Wilkins [68] for the V-dependence of the HF-HF V → R, T reaction are considered from the viewpoint of its effect on laser performance, both as a single-quantum and a multiquantum process. Because of the large scaling factor, the multiquantum process is deemed more likely. It also yields better model agreement with experiments.

Comparisons of SPIKE predictions of pulse length and pulse energy with three recent independent experiments over a broad range of operating conditions show generally good agreement.

PREFACE

The author is grateful to the following colleagues for valuable discussions and suggestions: Drs. R. Hofland, M. Kwok, and J. S. Whittier of The Aerospace Corporation; Dr. R. E. Meredith of Science Applications, Inc.; Prof. R. M. Herman of Pennsylvania State University; and Dr. G. Emanuel of Los Alamos Scientific Laboratory. The author also thanks J. S. Lesser and J. V. Patterson for their assistance in making the calculations.

CONTENTS

FIGURES

TABLES

I. INTRODUCTION

Interest in the potential of pulsed HF lasers, pumped by the $H_2 + F_2$ chain reaction, to deliver large specific power densities has motivated the development of several theoretical models for the prediction of laser performance [1]-[6]. However, these have had only limited success in their prediction of actual experimental results. Where model calculations of the H_2-F_2 laser have been compared with experimental data, some have shown fairly good agreement in the prediction of pulse shape [4]-[6]. In general, however, predictions of laser energy over broad ranges of operating conditions have not shown good agreement. Furthermore, substantial discrepancies remain between predicted and measured spectral content of the laser output.

Pulsed HF lasers typically operate at cavity pressures of ≥ 50 torr, where linewidths are dominated by pressure broadening. This broadening varies widely with the transitions and the nature of the perturbing species. The above models all assumed pressure-broadened linewidths by foreign molecules to be independent of J, the rotational quantum number of the transitions. The foreign-gas-broadened linewidths typically used in [1]-[5] were obtained from calculations based on billiard-ball-like interactions. These linewidths included no dependence on J and made no distinctions in the identities of the perturbing molecules. We show here that these numbers, in some cases, varied by several hundred percent from the experimentally measured values. The linewidths used in [6] accounted for the

distinct collision partners and were obtained from the calculations of Spellicy et al. [7]. For simplicity in the model calculations, however, these linewidths were averaged over all Js and were assumed to be the same for all transitions. In addition, self-broadening was not treated separately, as was done in [1]-[5], but was also included in this average linewidth.

Resonant self-broadened linewidths of the HF molecules used in the laser models of [1]-[5] were calculated from the resonant-dipole billiard-ball model (RDBBM) [8], [9], which is in excellent agreement with experimental values in the 1-0 band. In the higher vibrational level transitions, as will be seen, this model fails to account for the very important resonant interactions with the ground level ($v = 0$) molecules, and the predicted linewidth could be off by several hundred percent in some cases.

Whereas the major impetus for the present work stemmed from the study of the pulsed HF laser, much of the resulting data may be significant in the study of the cw HF laser also. The original cw laser measurements were made at pressures of $\sim$5 torr, where pressure broadening is not important. Recent cw experiments, however, have involved cavity pressures in excess of 20 Torr [10]. In this regime, both Doppler and pressure broadening are generally prevalent. Several computer models for the cw HF laser have been developed [1], [2], [11]-[19]; but these models have either neglected pressure broadening entirely or treated it in the same manner as was done in [1]-[6].

In the present work, a survey of available data for Lorentz broadening parameters of interest to the modeling of the $H_2 + F_2$ chemical laser was performed. Graphic representations of the relevant experimental data found in the literature are presented. For those cases where such data were not found, available theoretical values are shown. Collision broadening in the HF chemical laser has been reviewed by Emanuel [20], who indicated the necessity for inclusion of additional broadening data in the existing laser models, and by Meredith [21]-[23], who calculated the HF resonant self-broadened linewidths used in the present model. The present work is, in part, an extension of these prior studies and draws extensively from them. In addition to assembling these line-broadening data, a primary objective in this paper is to assess the importance of incorporating this detailed information in the simulation of the HF laser. It is shown that the specific information regarding the v and J dependence of the broadening cross sections, and the distinguishing broadening characteristics of various species in the gas, must be included in theoretical calculations in order to achieve good agreement with experiments, both in the prediction of total energy and in the discrimination of spectral content.

In addition to the revision of the Lorentz broadening functions, recent experimental results of Kwok and Cohen [24], for the v-dependence of the self-deactivation rate of HF were incorporated in the present model. Calculations made with the revised model were then compared with experimental measurements from several sources.

II. BROADENING THEORY

A brief summary of the theoretical background for pressure broadening is given in this section. Many detailed treatments for various aspects of this topic have been reported (see [25], for example).

For a fundamental transition $(v + 1, J_u) \rightarrow (v, J_l)$, the frequency is given by

$$\begin{aligned} \nu_{vm} &= \frac{E_u - E_l}{hc} \\ &= \nu_v + (B_v + B_{v+1})\, m + (B_v - B_{v+1})\, m^2 \\ &\quad + 2(D_v + D_{v+1})\, m^3 + (D_v - D_{v+1})\, m^4 \end{aligned} \tag{1}$$

where $m = J_u$ for a R-branch transition, and $m = -J_l$ for a P-branch transition. B_u and D_v are rotational constants. Measured values of these constants for HF are given in [26].

The Lorentz (or pressure) broadened half-width is

$$\gamma_m = \frac{n\bar{v}}{2\pi c} \sum_{J_p} \rho_{J_p} \sigma_{J_p} \tag{2}$$

where

n = density of perturbing molecules

$\bar{v}$ = mean relative collision velocity

ρ_{J_p} = population distribution of perturbing molecule's rotational states

σ_{J_p} = collision cross section of each rotational state J_p of the perturbing molecule.

The sum is taken over the rotational states of the perturbing molecule J_p.

Typically, the perturbing species are in rotational equilibrium and, thus, have a population distribution given by

$$\rho_{J_p} = \frac{(2J_p + 1) \exp(-E_{J_p}/kT)}{\sum_{J_p} (2J_p + 1) \exp(-E_{J_p}/kT)} \tag{3}$$

In the case of the HF chemical laser, however, the rotational equilibrium assumption may not be valid [5], [14], [15], [27]. For conditions of highly non-Boltzmann rotational distributions, the cross sections σ_{J_p} in (2) would be weighted very differently than in the case of rotational equilibrium. The result is an altered value for γ_m and a different optical gain coefficient for the corresponding laser transition. Such behavior may have a significant effect on predicted laser performance and is the subject of another study.

A theory was developed by Anderson [28] in 1949, and subsequently amplified by Tsao and Curnutte [29] in 1954, for calculation of the cross section σ_{J_p}. This theory resulted from detailed quantum mechanical calculations of the probabilities of nonradiative transitions induced by the combined effects of all the intermolecular forces in the system, consisting of the radiating

molecules and the perturbing molecules. Many calculations for the broadening of HF and HCl have been reported in which this theory was applied. These calculations used various forms of approximations for the interaction potential and the collision trajectories [21], [30]-[33]. As pointed out by Meredith, the Anderson theory has shown generally good agreement with experimental results for situations in which long-range dipole-dipole forces are dominant, as is the case for much of HF self broadening [21]-[23]. For conditions involving close collisions, however, where short-range forces become important, this theory is less satisfactory [34]-[36]. In [21]-[23] the Fiutak-Kranendonk [37], [38] approximation was used together with the Anderson theory to treat close collisions with some success. HF linewidths broadened by H_2 and N_2 calculated with this modified theory showed reasonable agreement with experimental results, especially for $m \lesssim 6$ (within ~30%, much less for lower values of m). However, HF linewidths broadened by D_2 and CO_2, calculated with the same approximation, remained in poor agreement with experiment.

Among other theories [39]-[44] that differ in basic approach from the Anderson theory and have been tested against experimental data for collisions not dominated by long-range dipole-dipole forces, the most extensive comparisons were made by Tipping and Herman [42], [43] for HCl lines and, more recently, by Jarecki and Herman [44] for HF lines. Their treatments included detailed approximations of the intermolecular potential and collision trajectories and velocities and were carried out using the Kolb-Griem-Baranger theory [45], [46] as compared with the perturbation technique

used in the Anderson theory. The calculated linewidths of [42] for HCl broadened by rare gases, and of [44] for HF broadened by rare gases, were generally in good agreement with experiment. The predictions for most lines in the fundamental band fell within 25% of the corresponding typical experimental values and showed approximately the same type of dependence on the rotational quantum number as that observed in experiments. These calculations, however, are limited to broadening by rare gases only and cannot be applied to diatomics such as H_2, F_2 and N_2, which are of interest in the present work. Comparisons of calculated line broadening with experimental values were also made [39] for a few selected lines of HCl, CO, and OCS broadened by He and Ar. These also showed good agreement (within 20% for the cases shown), but no comparisons were made for broadening by diatomics.

From the above discussion, it appears that the Anderson theory may be used with some confidence for predicting HF self broadening. It also appears that a theory similar to that of [39] or [44] may be used for foreign broadening of HF by rare gases. However, for HF lines broadened by several other species present in the typical HF laser medium, such as F_2, F, and H, no accurate theoretical predictions have yet been obtained.

III. PRESSURE-BROADENED HF LINEWIDTHS

A. HF SELF BROADENING

A considerable body of experimental data exists concerning the shapes and widths of self-broadened lines in the 1-0 band of HF [31], [47]-[51]. A graphic summary of the measured values of HF linewidths is given in Fig. 1. In the present notation, γ is the half linewidth at half maximum in units of 1/cm-atm. The vertical bars represent the scatter in the data from five independent experiments [31], [47]-[50] over a wide range of pressures (21.5 Torr to ~5 atm) and at ~100°C; the temperatures were not given in [47] and [50], but are presumed here to be >70°C since HF becomes highly polymerized at lower temperatures [31], [48]. Although the half-width γ for HF self broadening is believed to have a nonlinear relationship with pressure [21]-[23], these experimental data show that, within the pressure ranges considered (0.03 to 5 atm), the nonlinear effect is not large and may be ignored for the purposes of this study.

As discussed in [21]-[23], [52], and [53], the variation of the linewidth with temperature is complex, and no conclusive experimental data for this relationship are available. The temperature dependence used in the present work for HF self broadening consists of functional fits to calculations based on the Anderson theory completed by Meredith et al. [21]-[23] at several temperatures.

Figure 1 also shows the theoretical values for γ (HF 1-0 band) previously used in the chemical laser models of this laboratory, including RESALE [2]. The agreement between the theoretical values and the experimental data is seen to be excellent. These theoretical values were derived from the RDBBM of Benedict et al. [8], [9], which is a specialized version of the Anderson theory. This model, however, fails when applied to upper vibrational level transitions, as shown in the following discussion.

No experimental measurements of HF self-broadened linewidths have been made for the upper vibrational level $\Delta v = 1$ transitions, i.e., for the bands $1 \rightarrow 2$, $2 \rightarrow 3$, $3 \rightarrow 4$, $\cdots$. However, calculations have been performed by Meredith et al. [21]-[23], based on the Anderson theory, for these transitions up to the $4 \rightarrow 5$ band. These computations were carried out at 300, 600 and 900 K. No distinction was made between the P and R branches in these calculations; experimental measurements have shown that differences between the branches are generally not significant. Their results are presented as sets of coefficients resulting from least-squares fits to the calculated results of the linear combination of exponential functions (5):

$$\gamma(m) = c_1 + c_2 e^{-m/4} + c_3 m e^{-m/4} + c_4 m^2 e^{-m/2} + c_5 m e^{-m^2/8} + c_6 m^2 e^{-m^2/16} + c_7 e^{-m^2/8} \qquad (5)$$

Self-broadened linewidths for the HF (0-1) band calculated from these coefficients are also plotted in Fig. 1. The predictions appear to be in satisfactory agreement with experiment. Similarly calculated values for the

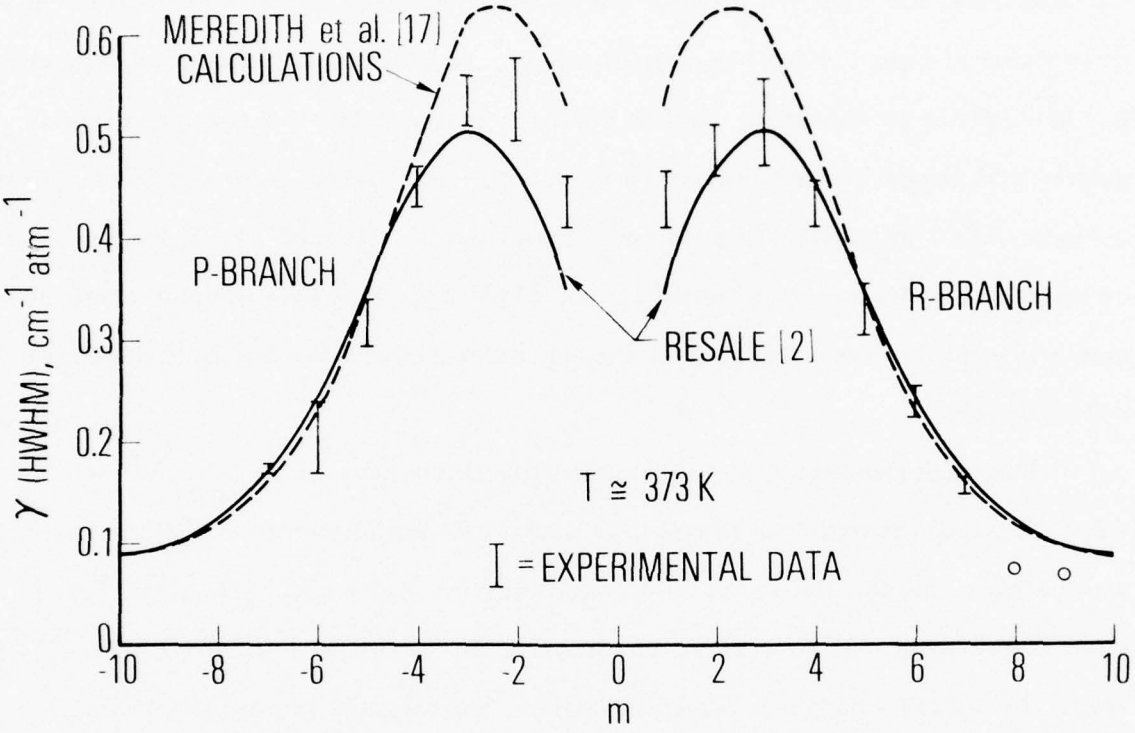


Fig. 1. Experimental and theoretical values for HF self-broadened linewidths; (0-1) band. Experimental data are from [31], [47]-[50].

higher vibrational level $\Delta v = 1$ transitions, at two temperatures, are shown in Figs. 2 and 3, along with the values presently used in laser modeling, calculated from the RDBBM model. Because of the small upper vibrational level population at equilibrium, the resonant-dipole contribution became negligible in the RDBBM formulation, which does not account for the resonant interaction with large ground-state ($v = 0$) population. Thus, only the hard sphere, billiard-ball interaction is evident in this model. It is clear, from the more complete calculation of Meredith et al. [21]-[23], that this ground-state resonance is quite appreciable. It is therefore considered in the present laser calculations.

The experiments that determined the linewidths of self-broadened HF were carried out with the sample HF gas at thermodynamic *equilibrium*, when almost all the HF is in the ground-vibrational state. Thus, the pressure broadening measured was essentially the result of collisions with HF(0) only. In the HF chemical laser, however, significant fractions of the HF population are expected to be found in the upper vibrational states. Because experimental broadening data for HF perturbed by HF molecules in the vibrationally excited states are not available, one is forced again to depend entirely on calculated values, as was necessary in the case of upper vibrational level $\Delta v = 1$ transitions. Results of these calculations for the cases of HF broadened by HF(1) and HF(2) can be found in [21]-[23] and are plotted here in Fig. 4.

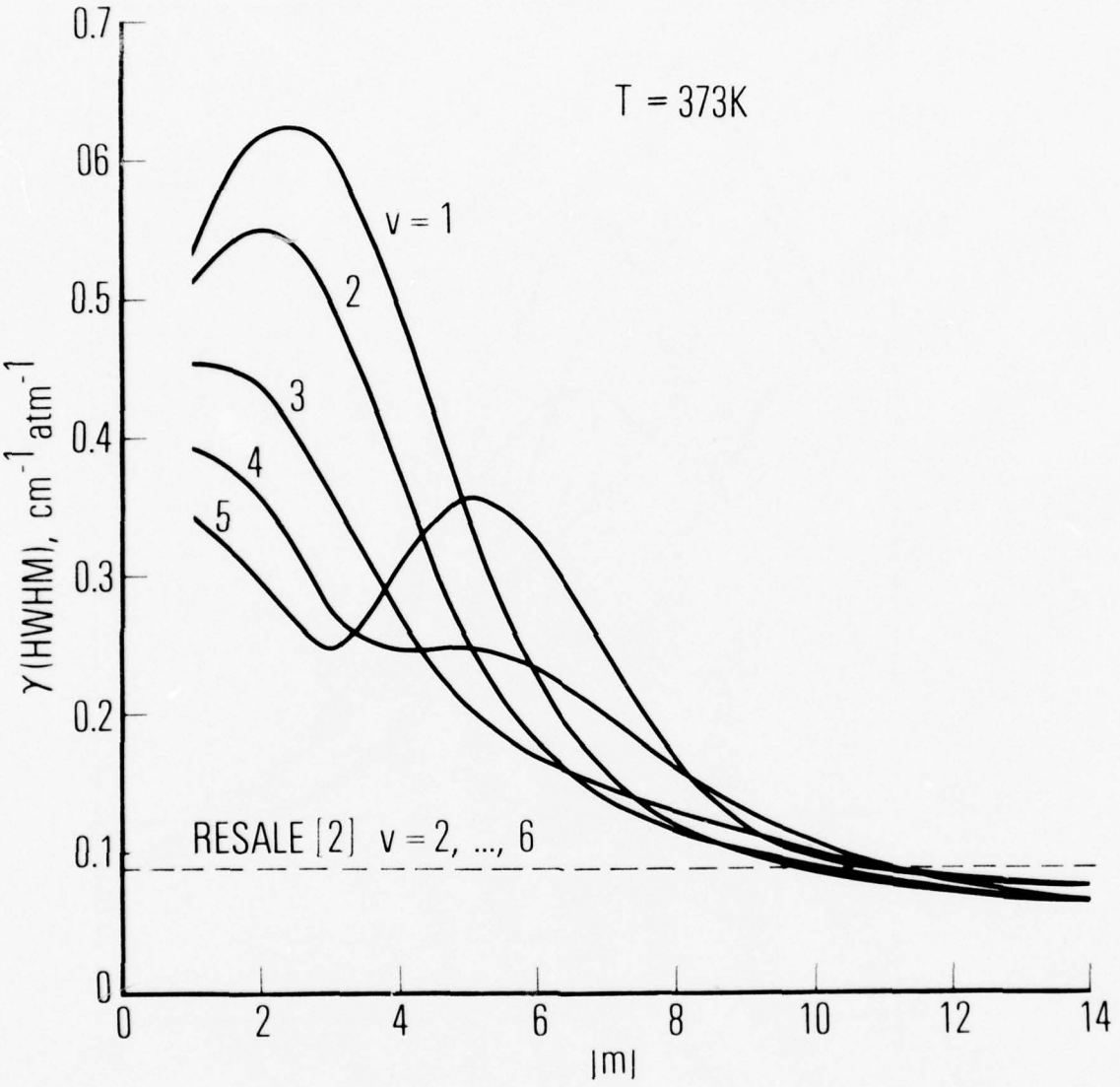


Fig. 2. Collision broadening of HF(v → v - 1) lines by HF(0), at T = 373 K. These curves were calculated based on the coefficients given in [21].

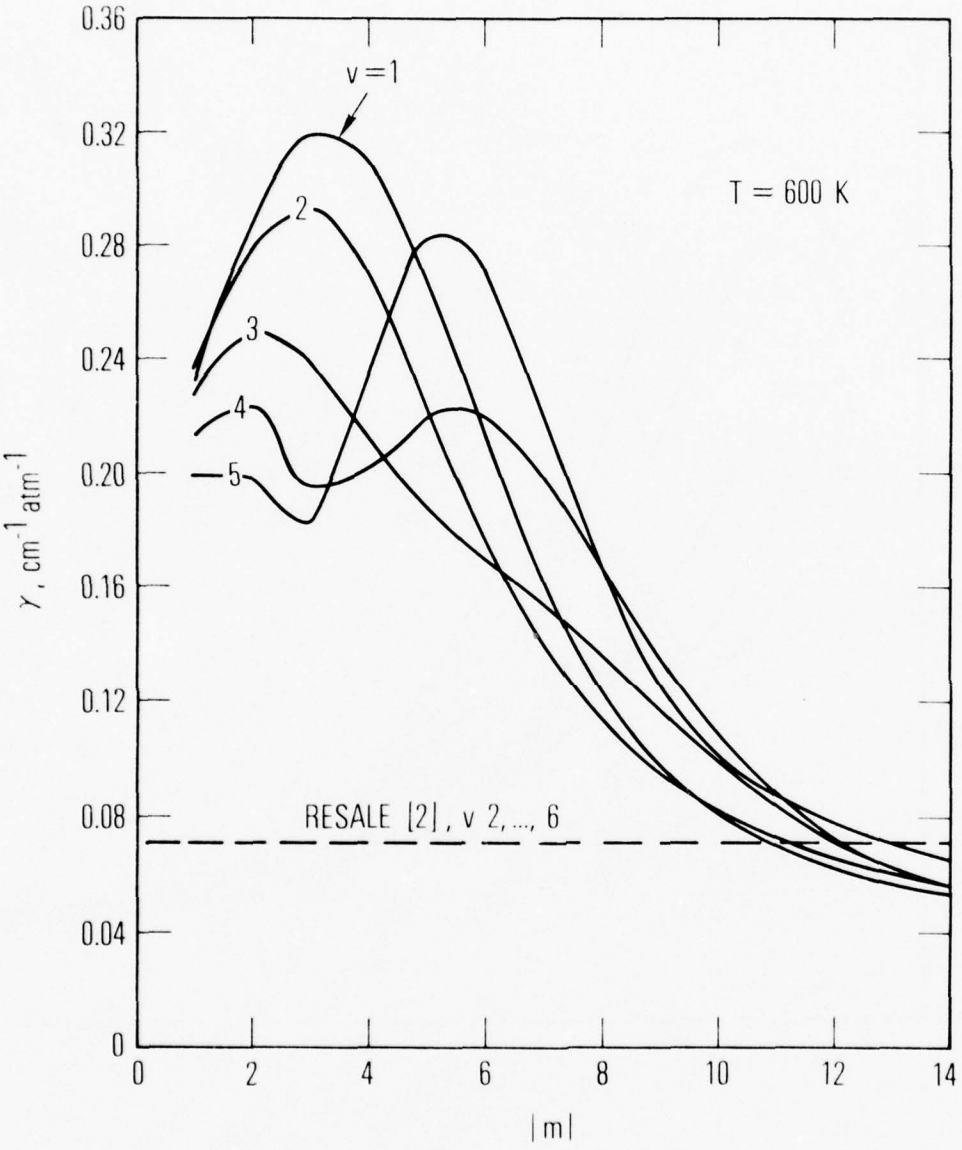


Fig. 3. Collision broadening of HF(v → v - 1) lines by HF(0), at T = 600 K. These curves were calculated based on the coefficients given in [21].

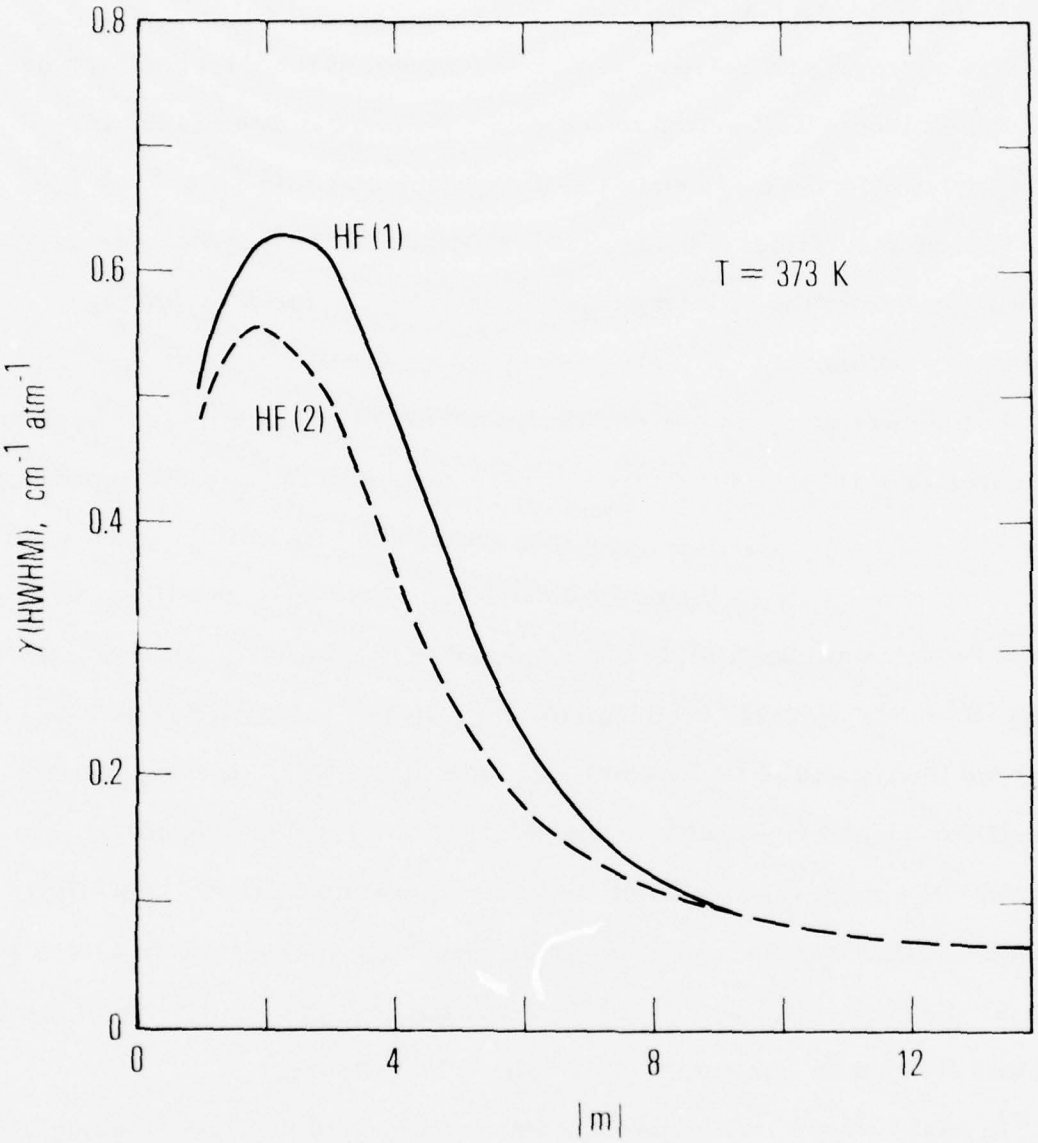


Fig. 4. Collision broadening of HF(0-1) lines by HF(v), at 373 K. These curves were calculated based on the coefficients given in [21].

B. FOREIGN GAS BROADENING OF HF

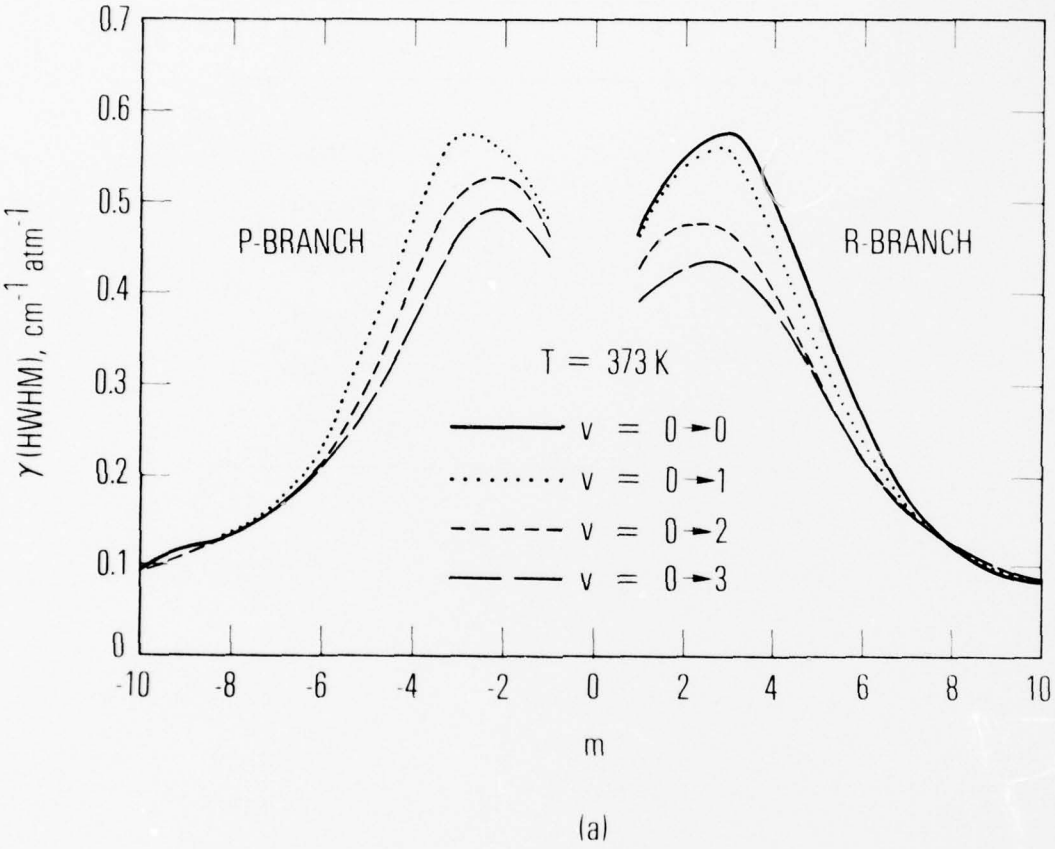
The strength and relatively wide separation of the HF spectral lines make the molecule well suited to the study of spectral line shapes [49]. Accordingly, a fairly large quantity of experimental [21]-[23], [32], [47], [54]-[58] and theoretical [7], [21]-[23], [30]-[32], [59]-[62] data are available for the linewidths of foreign-gas-broadened HF for a variety of perturbing species.

In the case of collisions dominated by dipole-dipole forces, the primary parameter that governs the degree of resonance and, hence, the broadening is the rotational constant B_v. For HF, the values of B_v at $v = 0$ and $v = 1$ are quite close (see Table I); thus, the linewidths of the pure-rotational band (0-0) and the fundamental band (0-1) are expected to be similar. This is illustrated in Fig. 5 for HF-HF self broadening: Fig. 5(a) is calculated from the Anderson theory and is taken from Meredith et al. [21]. It compares the HF self-broadened linewidths as a function of the rotational quantum number m for the pure-rotation band and several overtone bands. Fig. 5(b) shows a similar comparison of the experimentally measured linewidths (from [31]) for the pure-rotation and fundamental bands. The similarity of the (0-0) band and (0-1) band linewidths is evident in both figures.

In cases where intermolecular interactions are not dominated by dipole-dipole forces, the similarity of linewidths for the (0-0) and (0-1) bands usually still exists. Figure 6 shows the experimental data taken from [54]-[56] for HF (0-1) and HF (0-0) bands broadened by argon. The fundamental band data and the pure-rotation band data are comparable.

Table I. Rotational constants for the HF molecule [26].

v	B_v, cm^{-1}
0	20.5596
1	19.7872
2	19.0328
3	18.2995
4	17.5829
5	16.8792
6	16.1895



(a)

Fig. 5a. Comparison of self-broadened linewidths for the (0-0), (0-1), and overtone bands of HF. Calculations of [21], based on the Anderson Theory. This figure taken directly from Fig. 1-2 of [21].

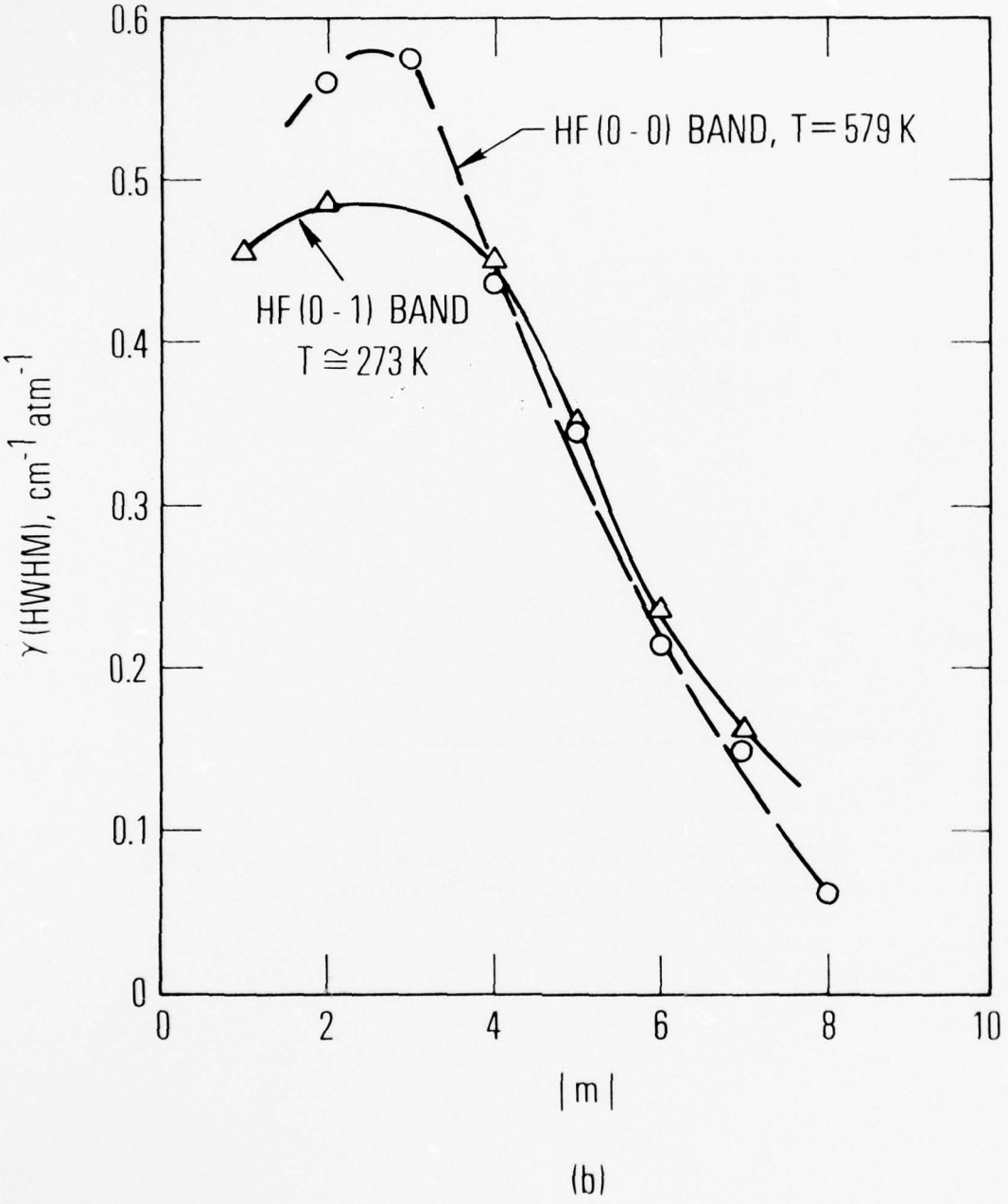


Fig. 5b. Comparison of self-broadened linewidths for the (0-0), (0-1), and overtone bands of HF. Experimental data from [31].

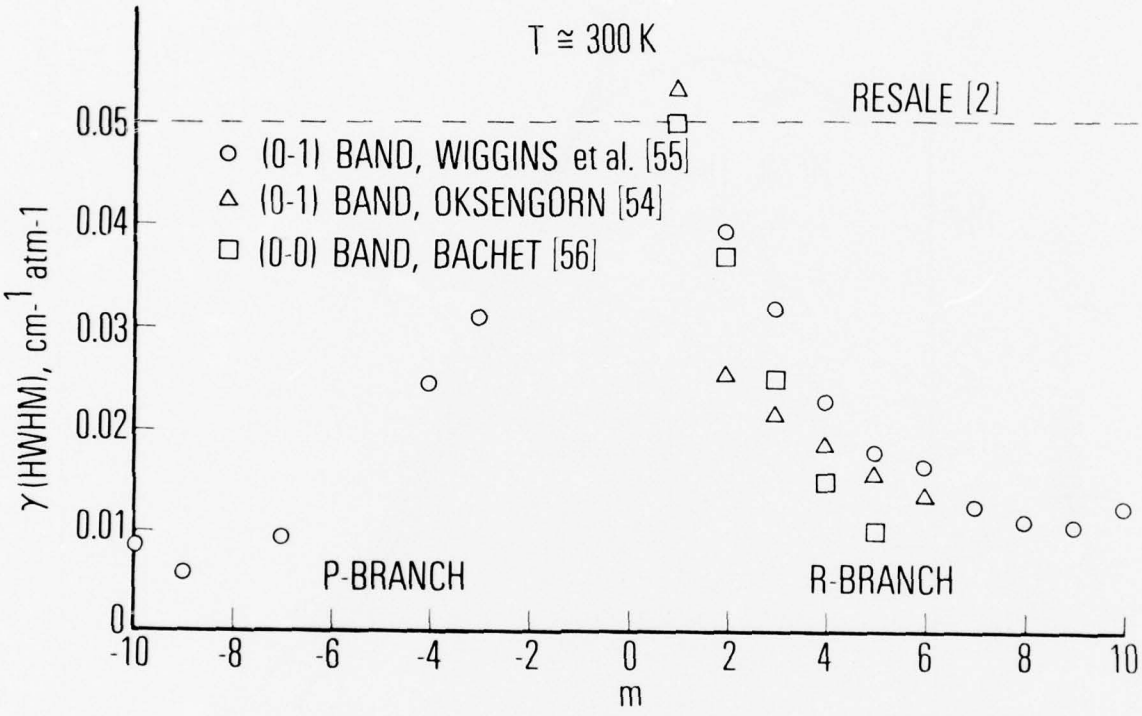


Fig. 6. Collision broadening of (0-0) and (0-1) bands of HF by Ar. Experimental data are from [54]-[56].

Because of this anticipated similarity, where experimental data for the (0-1) band are not available, the corresponding pure-rotational band data are presented instead. This is the case for HF broadened by N_2, H_2, and He. Line broadening of the pure-rotational band of HF, in collisions with N_2, H_2, and He, are reported in [32] and [56] and are shown in Fig. 7. Only one data point is available for the He case (atm = 1); however, because of the small mass of the He atom and its symmetry, it is expected that only small differences will be seen at larger values of m. This supposition is supported by observations of HCl and CO broadened by He [63], [64], where the linewidths displayed only minor variations with the value of m.

Neither experimental data nor calculated values are available on pressure broadening of HF lines with F_2 as the perturbing molecule; therefore, the data for Cl_2 as the perturbing molecule [58] are presented in Fig. 8 to illustrate the possible pattern and to provide a first order estimate of the HF linewidth as broadened by F_2.

Unlike HF self broadening, foreign-gas-broadened HF lines were shown in [21]-[23] to have the expected linear dependence on pressure. However, like the HF self-broadening case, the temperature dependence is inconclusive. No experimental data pertaining to temperature dependence were found during this study. Of the theoretical work [39], [42]-[44] that showed reasonably good agreement with experimental data in the prediction of foreign-gas-broadened linewidths, only [39] discussed temperature dependence. For all cases considered in [39], the broadening cross sections showed very slow increase with temperature, i.e.,

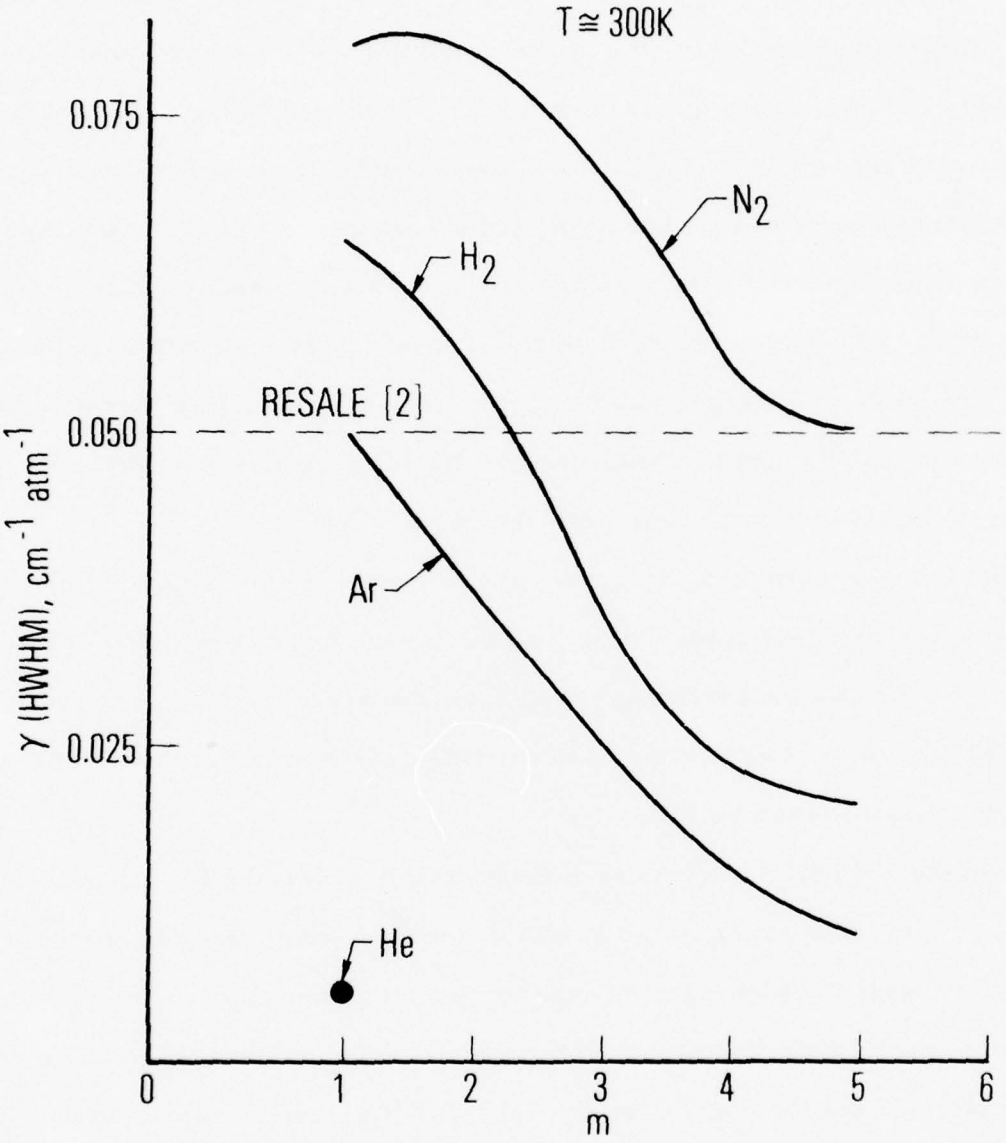


Fig. 7. Foreign gas broadening of the pure-rotational band of HF. Experimental data are from [32], [55], [56].

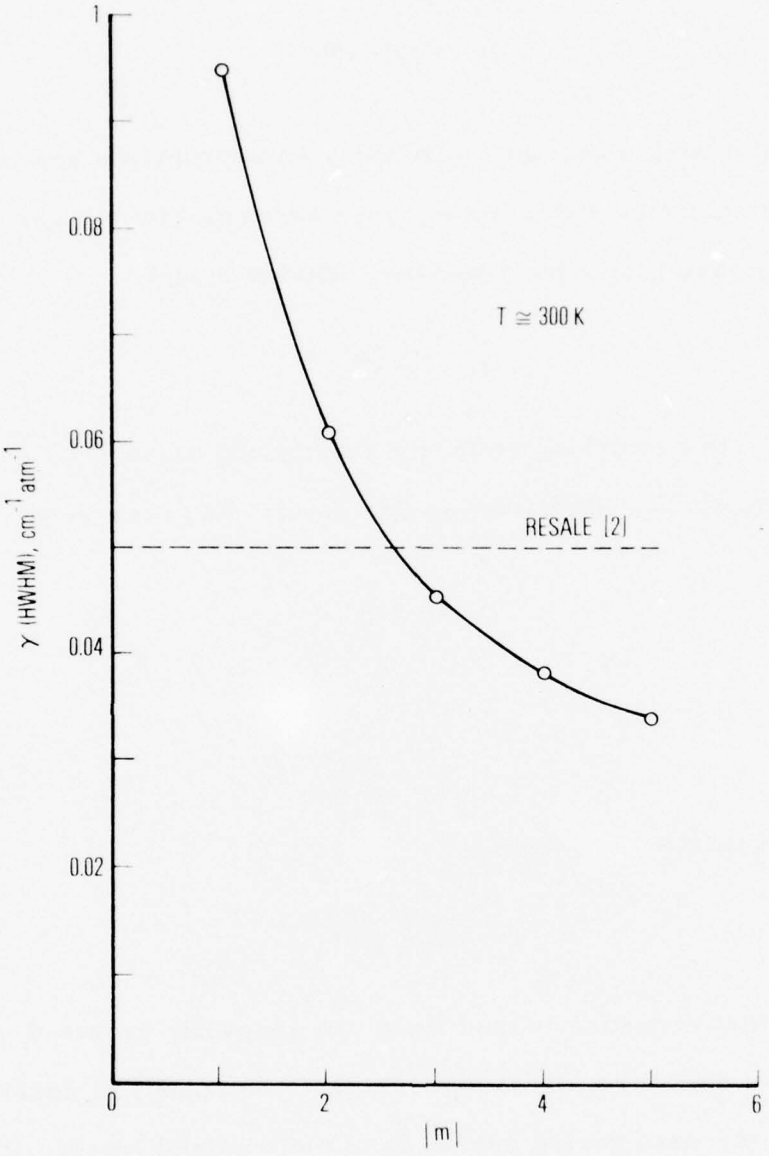


Fig. 8. Collision broadening of (0-1) band of HF by Cl_2. Experimental data are from [58].

$$0 \leq \frac{d\sigma_J}{dT}_p \leq 0.3 \tag{6}$$

Until more information becomes available, an appropriate approximation, consistent with the slow variation of cross sections with temperature, may be obtained by assuming a hard-sphere collision model

$$\gamma \propto n\bar{v} \tag{7}$$

where n and $\bar{v}$ are perturbing molecule density and mean relative collision velocity, respectively, as defined in Section II. At pressure p

$$n \propto \frac{1}{T} \tag{8}$$

and

$$v \propto \sqrt{T} \tag{9}$$

therefore, the relation becomes

$$\gamma \propto \frac{1}{\sqrt{T}} \tag{10}$$

Thus, at constant pressure, broad lines are generally expected to become narrower as temperature increases. On the other hand, at constant number density, as is the case during operation of most pulsed lasers, it is expected that linewidths will increase with temperature.

C. PRESSURE-BROADENING DATA INCORPORATED IN THE HF LASER MODEL

Because of incomplete experimental data and the apparent success of the Anderson theory, HF self-broadened linewidths are modeled with the calculated coefficients of [21]-[23] and (5). The calculations of [21]-[23] were carried out at 300, 600, and 900 K; hence, the temperature dependence in the model is obtained by interpolating between the results at these three temperatures. For foreign gas broadening of HF, the experimental data of Figs. 6-8 are used directly; here, the hard-sphere temperature dependence is used. Data for pressure broadening cross sections at large values of m are rare and are assumed to be constants equal to the cross sections of the largest m's (typically $m_{largest} \simeq 5$) for which data are available. Because of a similar lack of experimental data, foreign gas broadening of the upper vibrational band transitions is assumed to be the same as for the (0-1) band. The model assumes the pressure dependence to be linear for both foreign- and self-broadened linewidths and makes no distinction between the P and R branches.

IV. LASER MODEL WITH DETAILED BROADENING

The gain coefficient α, in units of cm^{-1}, is

$$\alpha(v,J,m) = \left(\frac{hN_A}{4\pi}\right) \omega_c(v,J,m)\, \phi_c\, B(v,J,m) \times \left[\frac{2J+1}{2J+1+2m}\, n_i(v+1,\ J+m) - n_i(v,J)\right] \tag{11}$$

where v and J refer to the lower level quantum numbers of the laser transition. The wave number of the transition is $\omega_c(v,J)$, $B(v,J)$ is the Einstein isotropic absorption coefficient based on intensity, and n_i represents the species concentrations of the upper and lower rotational populations. Only Doppler and Lorentz broadening, which are used in the Voigt profile ϕ_c, are considered in this formulation, The subscript c indicates that ϕ is being evaluated at line center, where the gain is at maximum. In the limit of pure Doppler broadening [2],

$$\phi_c \xrightarrow{\text{Doppler}} \frac{(\ln 2/\pi)^{1/2}}{\gamma_{Dp}} \tag{12}$$

where γ_{Dp} is the Doppler half-half width. This is the usual situation at the low pressures ($\sim$5 Torr) at which cw lasers operate. At higher pressures ($\gtrsim$50 Torr), where pulsed lasers typically operate, pressure broadening is most important. Here,

$$\phi_c \xrightarrow{\text{Lorentz}} \frac{1}{\pi\gamma_{LR}} \tag{13}$$

where γ_{LR} is the Lorentz half-half width. It is evident from (11) through (13) that, in the pure (Doppler or Lorentz) broadening regimes

$$\alpha(v, J, m) \propto \frac{1}{\gamma} \tag{14}$$

Thus, the unsaturated gain of the laser medium decreases as linewidth increases.

RESALE[2] is a well-known computer model that is currently in wide use for the prediction of HF laser performance. It is a comprehensive rate equation computer simulation of the H_2-F_2 chemical laser that incorporates up to 150 of the kinetic reactions within the laser medium, which is assumed to be homogeneous and contained in a Fabry-Perot optical cavity. For each vibrational level within the active species, the rotational states are assumed to be in equilibrium at the translational temperature. Lasing on each vibrational band is taken to be at line center of the transition having maximum gain. Once threshold is reached, gain is assumed to remain constant at the threshold level. When pumping reactions are no longer able to maintain the gain at threshold, lasing ceases. The present status of the Lorentz linewidth parameters, as applied to the calculation of HF chemical laser performance with this code, can be summarized as follows:

1. Self broadening of HF 1-0 band transition lines by the HF(0) molecules is well modeled; however, broadening of lines in the higher bands ($v \rightarrow v - 1$, $v \geq 2$) is not. Specifically, the resonance interactions with the heavily populated lower vibrational states are not adequately accounted for in such bands.

In laser mixtures containing large concentrations of reactants, this could cause errors in the determination of linewidth and, subsequently, small-signal gain, by as much as a factor of ~4 in some lines.

2. The same number (calculated from billiard-ball-type interactions) is currently used to represent the foreign-gas-broadened linewidth of all HF transitions. This lack of structure in the rotational quantum number J and the lack of dependence on the nature of the perturbing species is clearly unrealistic. The largest error introduced by this approximation is a factor of ~10 (for He diluents). In laser mixtures of large diluent ratios, this translates to approximately the same proportionate error in small-signal gain.

The status delineated above also applies to the models of [1], [3]-[5]. The linewidths used in the model of [6] do account for the differences between the various foreign-gas-broadening species. However, the foreign-gas-broadening cross sections used were theoretical and were averaged over all J transitions for computational simplicity. Moreover, resonant self broadening was not treated separately, as in [1]-[5], but was included in the average linewidth.

In order to evaluate the effect of the more detailed line-broadening parameters on the predicted HF laser performance, the calculated HF self-broadening linewidths of Meredith [21], as well as the available experimental data for broadening by foreign gases, were incorporated into the model SPIKE. This is a comprehensive model similar to RESALE that incorporates

up to 198 kinetic reactions and is capable of predicting transients and relaxation oscillations within the laser cavity. Hitherto, pressure broadening was modeled in SPIKE exactly as in RESALE. A detailed description of SPIKE is given in [5].

A comparison of predictions by codes in which the new and old Lorentz-broadening models were used is shown in Fig. 9 and Tables II and III for an approximately atmospheric 19 cm H_2 - F_2 chain laser. The conditions simulated here are from the experiments of [65], in which a 6% F_2 and 3% H_2 reactant composition was used with 37% Ar and 54% He as diluents for the balance of the gas mixture. The figure shows the calculated laser output energy for a typical range of initiation levels obtained in these experiments. As one would expect from the decreased linewidth, which resulted from the new broadening parameters, the predicted laser output energy shows a substantial increase. The effect is most significant near low levels of initiation, where small-signal gains do not rise much above threshold, and laser performance is most sensitive to threshold conditions. At $(F/F_2)_i = 0.3\%$, for example, the laser output energy predicted by the present model, with detailed broadening, is 300% of that predicted by the same model with use of the old broadening parameters. This difference between the predictions of the two models becomes much larger for gas mixtures with pure helium as diluent, as was done in the experiments of [6] and [66].

The predicted spectral contents of the calculations made with the two Lorentz-broadening models are compared in Tables II and III. Although it is

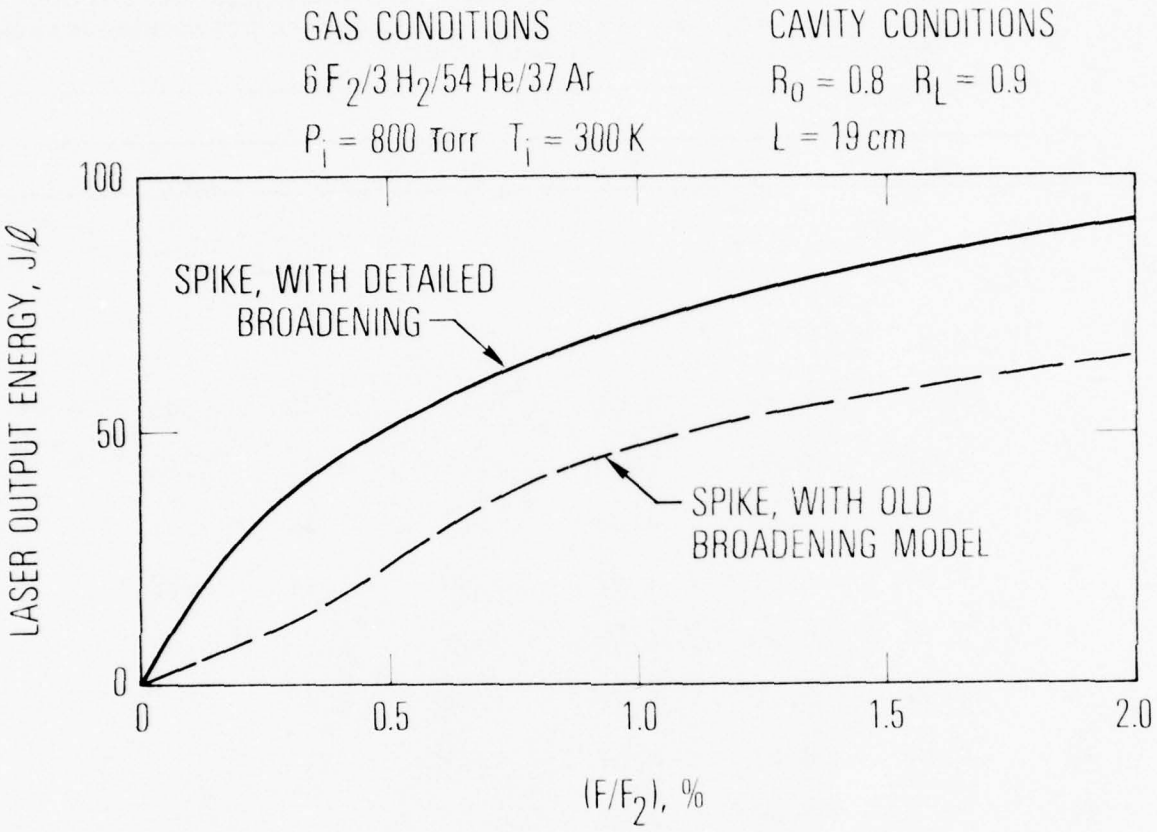


Fig. 9. Comparison of laser output energies as predicted by SPIKE, utilizing the present detailed broadening model and the old broadening model, as a function of the initiation level.

Table II. Distribution of laser energy among the individual transitions, calculated with use of the improved pressure broadening model, expressed in percent of total output.[a]

	v						
J	0	1	2	3	4	5	6
1							
2							
3	0.01						
4	0.31	0.24	0.08	0.11	0.09	0.03	0.01
5	0.54	0.85	0.78	1.02	2.21	2.55	0.10
6	1.09	2.03	2.38	3.13	3.36	1.31	
7	2.60	4.31	4.51	3.50	2.41	0.88	
8	3.98	7.90	4.62	3.11	1.99	0.07	
9	1.94	9.18	5.11	1.94	0.42		
10	0.14	8.07	3.67	0.35			
11		3.69	2.11				
12		0.76	0.48				
Total	10.61	37.03	23.74	13.16	10.48	4.84	0.11

[a](v, J) is the lower level of the transition.

Table III. Distribution of laser energy among the individual transitions, calculated with use of the old pressure broadening model, expressed in percent of total output.[a]

	v						
J	0	1	2	3	4	5	6
1							
2	0.01						
3	0.63	0.42	0.04	0.07	0.09	0.01	
4	0.87	1.17	0.80	0.77	1.32	1.22	
5	1.81	3.07	3.06	3.42	4.57	2.09	
6	3.00	5.72	5.72	4.42	2.17	0.01	
7	3.62	9.93	5.41	2.68	0.95		
8	1.02	10.64	5.62	0.99			
9		6.95	3.16				
10		1.64	0.88				
11							
12							
Total	10.96	39.54	24.69	12.35	9.10	3.33	

[a](v, J) is the lower level of the transition.

expected that, because of such limiting approximations as rotational equilibrium assumed in these calculations, the resulting predictions cannot accurately reflect experimental measurements, the comparison is still instructive in delineating the trends obtained from changes in the linewidth formulation within the laser model. As shown in the tables, the incorporation of rotational structure has resulted in a redistribution of the predicted laser output energy within each vibrational band, from the lower J to the higher J transitions. Furthermore, because these high J transitions dominate during the latter portion of the laser pulse due to the increased gas temperature (the gas temperature at pulse termination for the present case is ~900 K), the larger gain predicted for these transitions by the detailed broadening model results in an increased pulse length. For the present case, pulse length is increased 25% from 3 to 3.75 μsec.

The kinetic rate data used in the present calculations were essentially those compiled by Cohen [67], with the exception of the HF - HF V → T, R reactions. Recent experimental data of Kwok and Cohen [24] and Kwok and Wilkins [68] for the self-deactivation reaction

$$HF(v) + M \xrightarrow{k_v} HF(v') + M, \quad M = HF(v), \quad v = 0, \ldots, 8, \quad v' < v \qquad (15)$$

have resulted in a revised scheme for the scaling of the rate coefficient k_v with the vibrational quantum number v. The rate was determined from experiments performed in a large diameter, medium pressure flow tube facility and found to scale empirically as $v^{2.3}$. It is not yet clear what the

deactivation mechanism is or whether it is a single-quantum or multiquantum process. Calculations were carried out with the model by using each assumption to compare the effects of single- and multiquantum deactivation on laser performance. Multiquantum deactivation was modeled with the assumption that the reaction proceeds with equal probability through all channels with $\Delta v \geq 1$ and that the net deactivation rate k_v of reaction (15) scales according to $v^{2.3}$, i.e.,

$$k_v = \sum_{v'} k_{vv'} = k_1 \times v^{2.3} \tag{16a}$$

$$k_{vv'} = k_{vv''} \qquad v', v'' < v \tag{16b}$$

One such comparison is shown in Fig. 10, where the assumption of the multiquantum deactivation process decreased the laser output by >28% compared with the case with single-quantum deactivation. Most of the energy loss occurred in the second half of the laser pulse after the HF population had reached appreciable values (the total amount of HF produced at the end of the pulse for this case is 5.3% of the gas mixture with 48% of the F_2 consumed). The multiquantum deactivation also resulted in a reduction of the pulse duration by ~10%. From the magnitude of the self-deactivation rate coefficient scaling factor, it appears that the multiquantum process is more likely because it implies a larger number of transfer channels for deactivation with increasing v. This possibility is further substantiated by the fact that, with the assumption of a multiquantum deactivation mechanism, coupled with

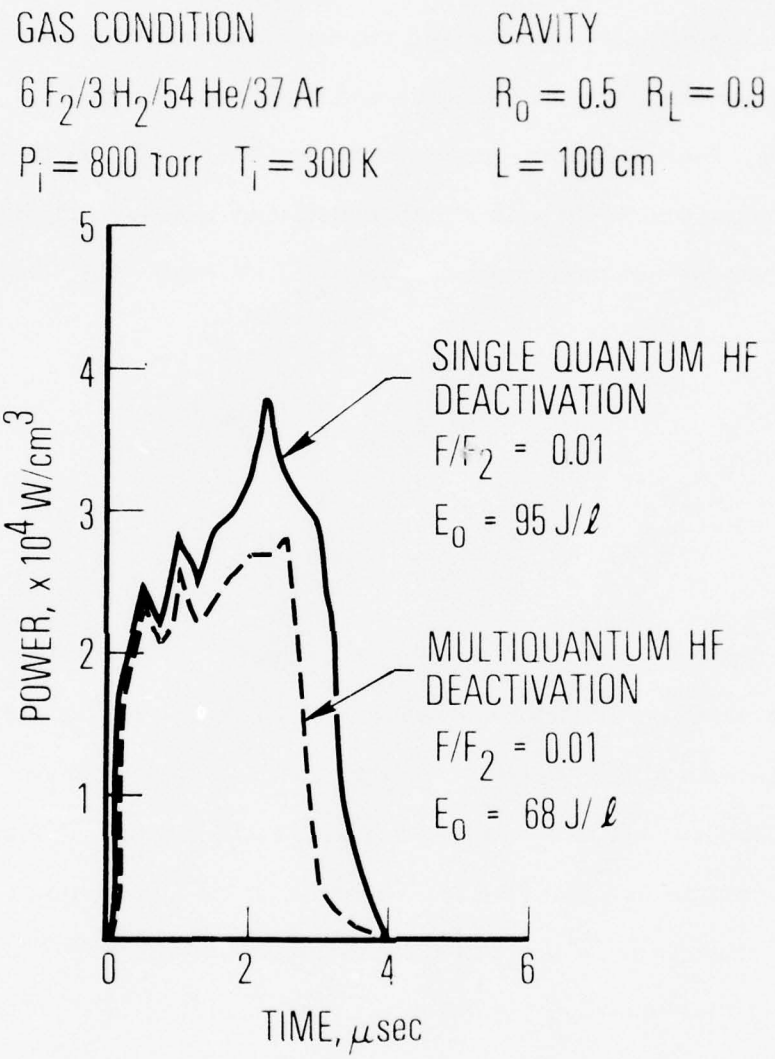


Fig. 10. Effect of multiquantum deactivation on predicted laser output

the more detailed Lorentz-broadening model, good agreement between predicted laser output and experimental results was obtained. The comparison of model predictions with experimental data is discussed in Section V.

V. COMPARISON OF MODEL PREDICTIONS WITH EXPERIMENT

In this section, a comparison is presented of calculated laser performance with several recent experimental measurements. In all the following calculations, the model assumes the multiquantum deactivation mechanism for the HF V → R, T reaction and uses the detailed pressure-broadening model.

In Figs. 11 and 12, predicted pulse outputs are compared with two independent experiments. The experimental pulse in Fig. 11 is from [65], in which the H_2-F_2 laser was initiated by an electron beam irradiated discharge; the attendant experimental conditions are as described in the discussion for Fig. 9. Figure 12 shows the experimental result from [6] for a H_2-F_2 laser initiated by flash photolysis; here, the gas mixture (consisting of 4% F_2, 4% H_2, and 92% He) is retained in a cavity that formed a 50 cm gain length, but is significantly more "lossy" than the preceding case (86% output coupler transmission compared with 20% in the cavity of [65]. In both cases the predicted pulse length and pulse energy show good agreement with the experimental pulses. The predicted pulse shapes, however, show a great deal of oscillation. We speculate that this is due, in part, to the restriction of lasing to line center (which is equivalent to operating a mode-limited device, where the lasing modes are restricted to those with frequency near line center). This condition results in large gain overshoots and is manifested in the subsequent "relaxation oscillations." Additional discussions on this

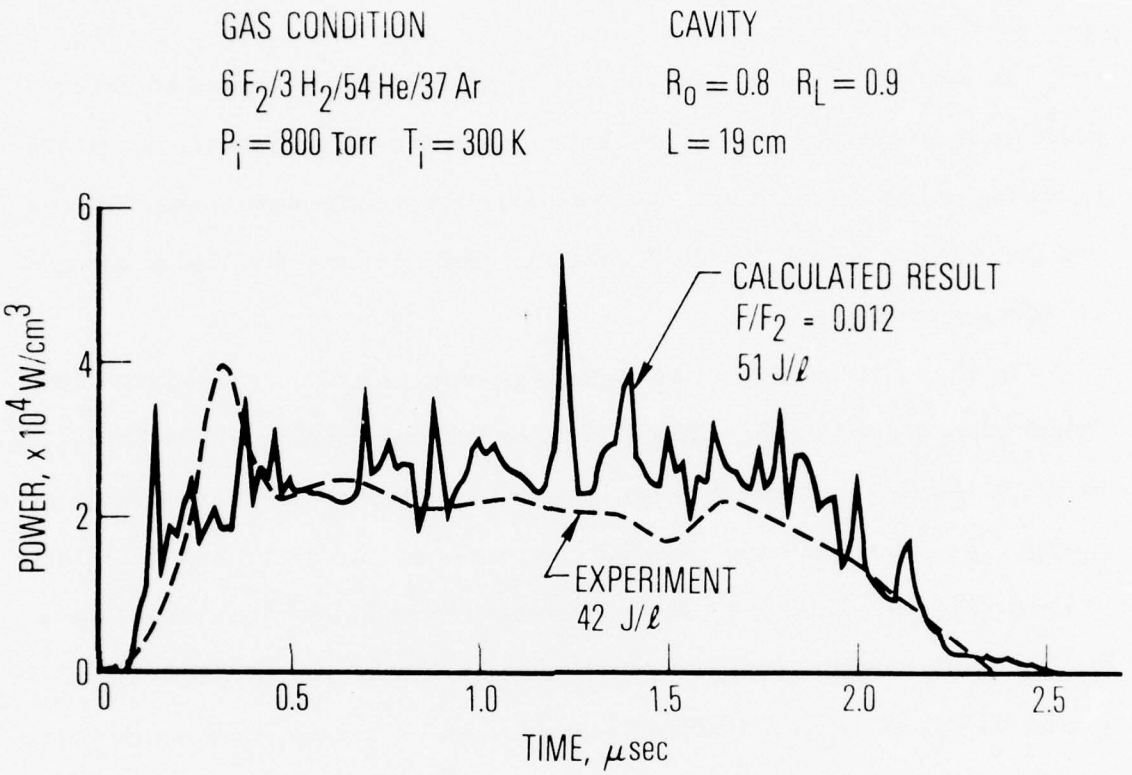


Fig. 11. Comparison of predicted laser pulse with the experiment of Hofland et al. [65]

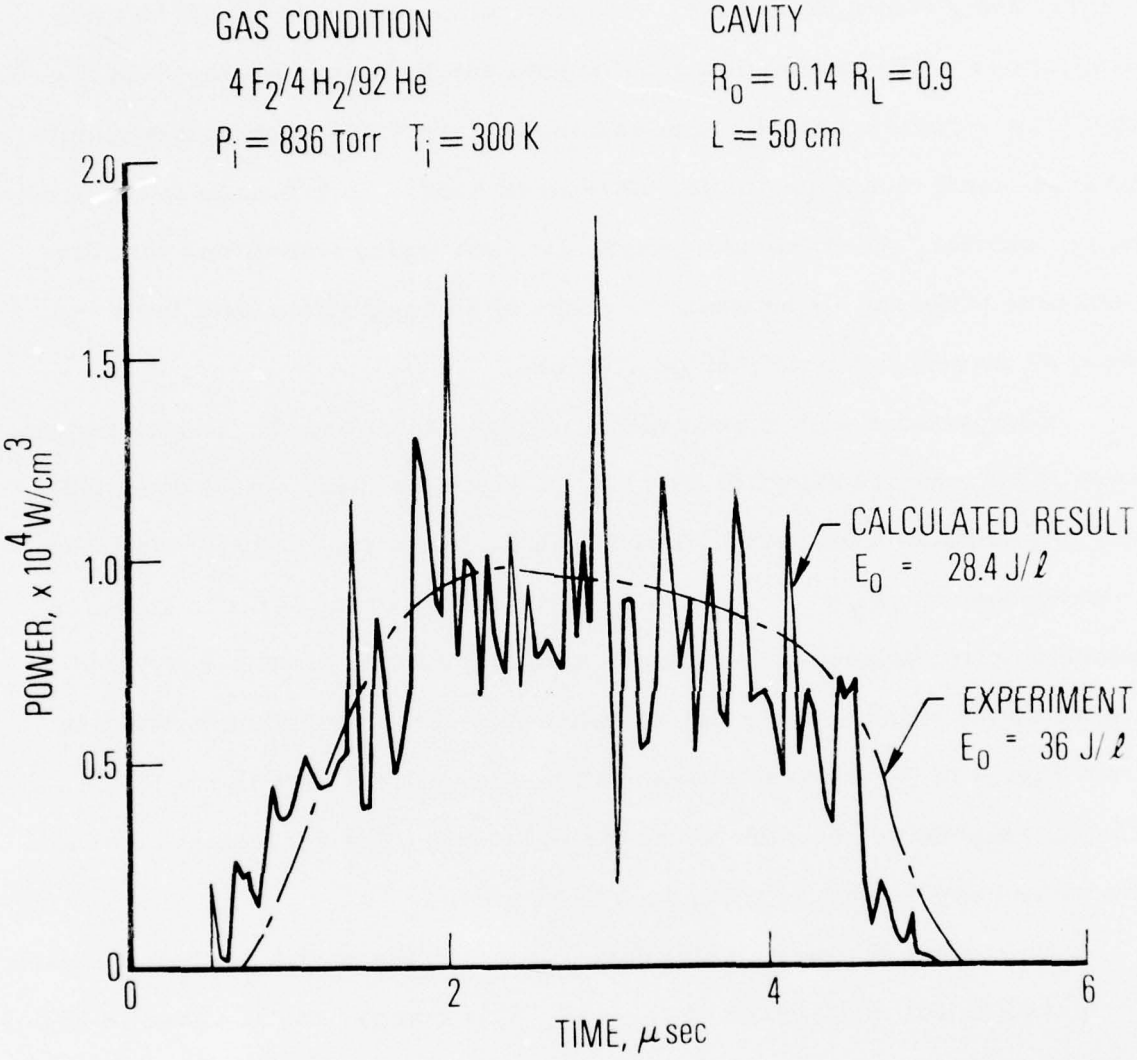


Fig. 12. Comparison of predicted laser pulse with the experiment of Chen et al. [6]

subject are given in [5] and [69]. Another factor that contributes to these oscillations is the assumption of rotational equilibrium. As shown in Fig. 8 of [5], the rotational equilibrium assumption precludes appreciable simultaneous lasing on any particular vibrational band. This results in the prediction of shorter, more intense, pulses for each lasing transition, with distinct time differentials between the peaks of adjacent (timewise) lines and, thus, an increase in predicted oscillations.

Comparisons with experimental results over a broader range of operating conditions are shown in Fig. 13, in which the laser output energy density is plotted as a function of percentage F_2 in the gas mixture. In these calculations, the F_2 ratio is varied from 0 to 10% of the laser mixture, a stiochiometric balance is maintained with H_2, and the diluent is He, which makes up the balance of the gas composition. The experimental data are from Fig. 4 of [6] and are represented by the vertical bars in the figure. The corresponding reactant ratios are indicated. The agreement between theory and experiment is again seen to be close.

A final set of experimental data with which the model predictions were compared is that of Mangano et al. [66]. This comparison is shown in Fig. 14. In these experiments, the H_2-F_2 laser was initiated by a direct e-beam pulse, and the percentage of F_2 in the atmospheric pressure, F_2/H_2/He gas mix was varied from 6 to 30%. The experimental data are represented by the open circles, and the present calculations are indicated by solid circles. The brackets in the figure indicate the percentage of H_2 in the gas. Agreement

- FLASHLAMP INITIATED, $\left[F/F_2\right]_i \sim 0.8\%$
- 50 cm ACTIVE LENGTH, SAPPHIRE OUTPUT COUPLER
- GAS MIXTURE: $xH_2/xF_2/(1-2x)$ He, 836 Torr

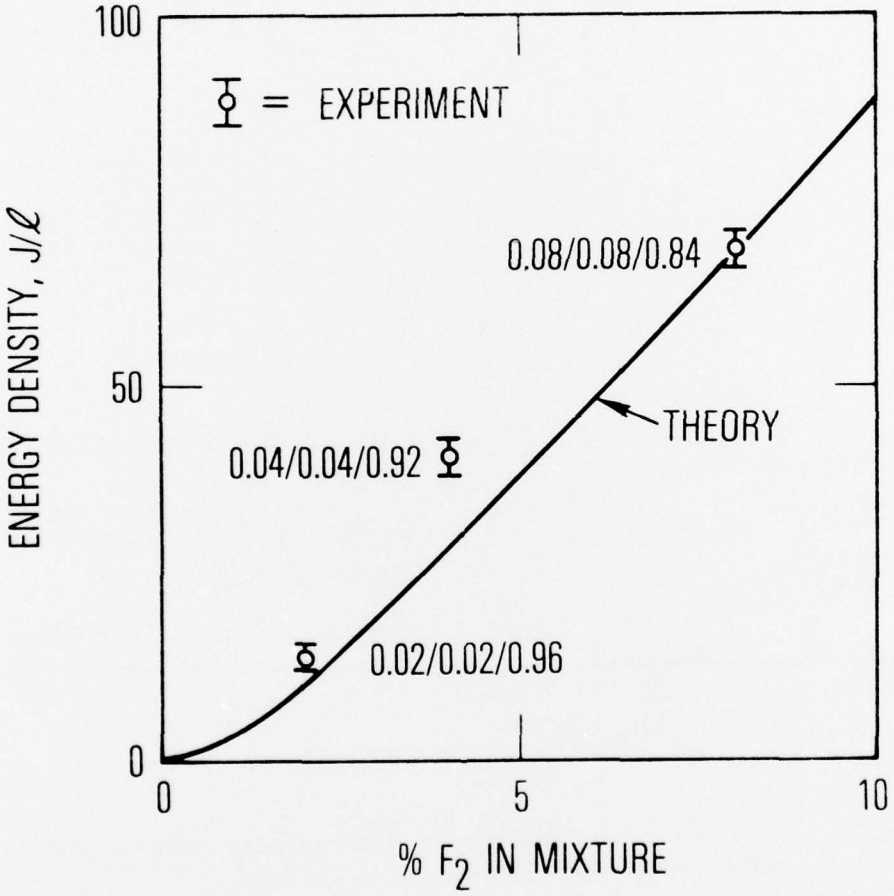


Fig. 13. Comparison of model predictions with experimental measurements of Chen et al. [6]

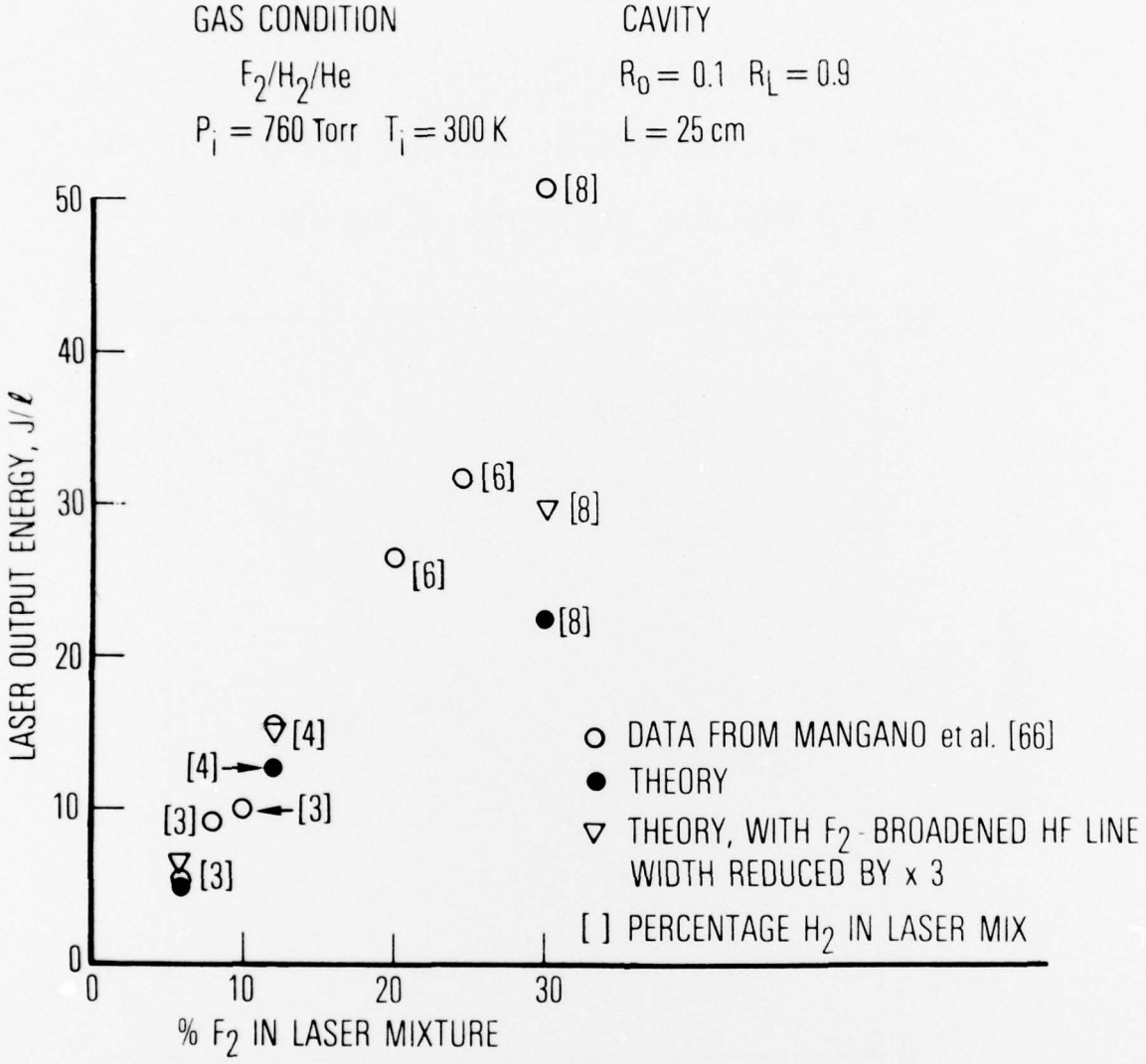


Fig. 14. Comparison of model predictions with experimental measurements of Mangano et al. [66]

between theory and experiment is good for the low F_2 cases. The 30% F_2 case, on the other hand, shows a large divergence between the measured and predicted results.

Since the F_2-broadened HF linewidths (γ_{F_2}) used in this model were estimated from Cl_2 results, the above calculations were repeated with different values for γ_{F_2} to assess the sensitivity of the model predictions to this parameter. The case of γ_{F_2} reduced by a factor of 3 is shown in Fig. 14. As anticipated, the effect of this change becomes more appreciable with increasing F_2 ratio. Predicted laser output energy increased by 33% in the 30% F_2 case. This, however, still falls far short of the Mangano measurement. The initiation levels used in the calculations of Fig. 14 were based on the estimates of [66], which assumed that every 12 eV of the deposited electron-beam energy contributed to the production of one F atom. It is still not certain, however, if such an approximation of F_2 dissociation may be applied equally to gas mixtures of widely different compositions. If the estimated initiation level is indeed low, the difference between the measured and predicted laser energies for the 30% F_2 case may yet be small. These considerations are still under investigation.

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