Fundamental Gain Suppression Mechanisms in a Continuous Wave Hydrogen Fluoride Overtone Laser

P. T. Theodoropoulos* and L. H. Sentman[†] *University of Illinois at Urbana–Champaign, Urbana, Illinois 61801*

When lasing occurs on the overtone, a rotational nonequilibrium computer model showed that the fundamental gains are determined by three independent mechanisms. First, overtone lasing decreases the gains of the $P_1(J)$ and $P_2(J)$ lines whose upper or lower levels are directly involved in $P_{20}(J)$ overtone lasing. Second, overtone lasing reduces the rate at which the low J v=2 states are populated by rotational relaxation and increases the rate at which the low J v=0 states are populated by rotational relaxation, resulting in suppression of the low J fundamental gains whose upper or lower levels are not directly involved in overtone lasing. Third, overtone lasing reduces the rate at which the HF(0, J) and HF(1, J) states are populated by the various collisional deactivation processes. The computer model gave reasonable agreement with the measured fundamental zero power gain profiles, Fabry-Perot power, and spectra. The model overpredicted the fundamental gain suppression ($\Delta \alpha$) for the P_1 (8, 9) and P_2 (8, 9) lines whose upper or lower levels were directly involved in overtone lasing and underpredicted the suppression for lines P_1 (4) and P_2 (4, 5). The model predicted the suppression for lines P_1 (5–7) and P_2 (6, 7) reasonably well. When the rotational relaxation rate was increased by a factor of 10, the model was in reasonable agreement with the measured suppression, $\Delta \alpha$, of all P_1 (4–9) and P_2 (4–9) lines. However, with the increased rotational relaxation rate, the model's prediction of the experimental zero power gain and residual fundamental gain profiles was not adequate.

I. Introduction

▼ OMPARISON of residual fundamental amplification ratio (RF-AR) data obtained at relatively high medium saturation with two 99.7% reflective mirrors ($\alpha_{\rm sat} = 0.00010015$ and $L_{\rm g} =$ 30 cm) with the zero power amplification ratio (ZP-AR) data indicated that the gains of the low J lines $P_1(4-6)$ and $P_2(4-6)$ were suppressed between 41 and 96% and the gains of the high J lines $P_1(7-$ 9) and $P_2(7-9)$ were suppressed between 3 and 44%. The 1 \rightarrow 0 lines were suppressed more than the $2 \rightarrow 1$ lines. The maximum suppression occurred between 2 and 6 mm downstream from the nozzle exit plane, near the center of the 9-mm overtone beam. There was minimal suppression of lines $P_2(8, 9)$. The low J fundamental gains were suppressed more than the high J fundamental gains even though their upper or lower levels were not directly involved in overtone lasing. Residual fundamental gain (RFG) measurements at low medium saturation with overtone mirrors of 99.7/98.0% reflectivity ($\alpha_{sat} = 0.000386787$) showed weak suppression on lines $P_1(7)$, $P_2(5)$, and $P_2(6)$ at axial positions between 2 and 6 mm downstream from the nozzle exit plane. 1,2 There was no suppression measured for any of the other lines. RFG measurements performed at an increased level of medium saturation with 99.8/99.86% reflective overtone mirrors ($\alpha_{\text{sat}} = 0.000056716$) resulted in essentially the same suppression obtained with the 99.7/99.7% mirrors. 1,2

The objectives of this work were to determine why the low *J* fundamental gains were suppressed more than the high *J* fundamental gains even though their upper or lower levels were not directly involved in overtone lasing and to predict the RFG as a function of medium saturation. Since there are many kinetic processes involved in the population and depopulation of the upper and lower levels of the fundamental transitions, a detailed rotational nonequilibrium model (ORNECL)^{3,4} of the laser flow was used. When ORNECL was baselined to ZP-AR data, it was necessary to decrease the rotational relaxation (RR) rate^{5,6} by a factor of 10 and to include the multiquantum HF-H₂ vibration-to-vibration (VV) transfer reactions.

ment with the data; see Sec. II.B. Several concluding remarks are presented in Sec. III.

The ZP-AR profiles predicted by ORNECL are in good agreement with the vertically averaged data except for $P_1(4)$, which overpre-

With ORNECL baselined to the ZP-AR data, power predictions

were in good agreement with the measured fundamental Fabry-

Perot power and spectra as a function of reflectivity.7 With this

agreement of the ORNECL computer model with the fundamental

Fabry-Perot power, spectra, and zero power gain (ZPG) data, the model can be used to study the suppression of the fundamental gains

ORNECL calculations indicated that the primary mechanism re-

sponsible for the suppression of the low J fundamental gains is a

change, which is caused by overtone lasing, in the effective rate at

which RR populates the upper and lower levels of the fundamental

low J lines; see Sec. II.A. When the RR rate was increased by a

factor of 10 (to its original value), the fundamental gain suppression $(\Delta \alpha)$ of both the high J and low J lines was in reasonable agree-

dicts the data, and $P_1(7)$, which underpredicts the data.⁴

by lasing on the overtone $2 \rightarrow 0$ transitions.

II. Simulation of Residual Fundamental Gain

A. Low J Line Gain Suppression

Comparisons between average ZP-AR data and RF-AR data obtained with the two 99.7% reflective mirrors indicated that the $1 \to 0$ lines are generally suppressed more than the $2 \to 1$ lines, especially at high J, and that the low J, $P_1(4-6)$ and $P_2(4-6)$, lines are suppressed more than the high J, $P_1(7-9)$ and $P_2(7-9)$, lines. The supersonic laser was lasing on lines $P_{20}(8-11)$ while the RF-AR measurements were performed. Since the upper and lower levels for the $P_{20}(8,9)$ lasing transitions are also the upper or lower levels for the $P_1(8,9)$ and $P_2(8,9)$ transitions, it is reasonable that the gains of these high J lines would be suppressed. The effect that overtone lasing has on the gains of the high J fundamental lines whose upper or lower levels are directly involved in overtone lasing is termed direct lasing effect.

The question that needs to be answered is why the low *J* fundamental lines were suppressed since their upper or lower levels were not directly involved in overtone lasing. To answer this question, the mole/mass ratios of HF (number of moles/gram of mixture) for the vibrational and rotational levels associated with fundamental and overtone lasing were calculated using ORNECL for zero power and overtone lasing. The overtone lasing calculations were performed with overtone mirror reflectivities of 99.78/99.67%. Since the model

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^{*}Research Assistant, Aeronautical and Astronautical Engineering Department

[†]Professor, Aeronautical and Astronautical Engineering Department. Associate Fellow AIAA.

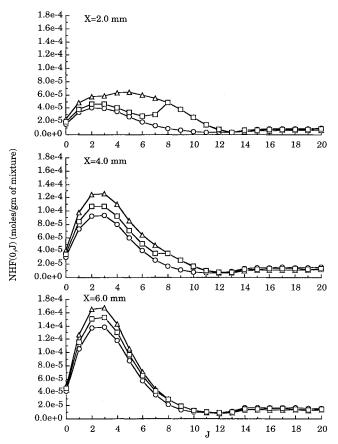


Fig. 1 Comparison of ORNECL HF(0, J) concentrations for zero power and overtone lasing conditions. These calculations were performed with the complete reaction set. The profiles are plotted at 2, 4, and 6 mm downstream from the nozzle exit plane: —○—, zero power conditions; —△—, overtone lasing, no absorption; —□—, overtone lasing, $P_{20}(3-7)$ absorption.

predicts lasing on $P_{20}(3-12)$ while experimentally lasing occurred on $P_{20}(8-11)$ a calculation was made with SF_x absorption^{7,8} on lines $P_{20}(3-7)$ to bring the calculated spectra into agreement with the data and to determine the effect of suppressing lasing on these lines on the HF(v, J) populations; see Figs. 1–3.

Polanyi and Woodall⁹ showed that the pumping reaction populates HF(1, 4-13), HF(2, 2-14), and HF(3, 0-6) with the peaks at (1, 7, 8), (2, 6, 7), and (3, 2) and that RR was the major collisional process responsible for the transfer of population from the high Jstates populated by the pumping reaction to the low J 0-4 states of v = 1 and 2. This occurred through a double peaked relaxation process. Since the pumping reaction does not populate HF(0, J), the only way HF(0, J) states are populated is through collisional deactivation processes or lasing. Overtone lasing on the $P_{20}(7-12)$ transitions rapidly depopulates the peak of the pumping distribution in v = 2 and rapidly populates the v = 0, J = 7-12 states. Because of the significant change in the populations of these states, overtone lasing may have a major effect on the subsequent transfer of populations to the low J states in v = 2 and 0 and thus would be the cause of the suppression of the low J lines. To determine the extent to which the RR reactions affect the population of the low J states, the RR rate constant was set equal to zero and the zero power and overtone lasing calculations were repeated; see Figs. 4–6. The relative magnitudes of the concentrations calculated with and without RR were examined to determine to gross effects of RR on the populations of the various HF states. The overtone lasing and zero power concentrations calculated with no RR were compared to determine the extent to which RR is responsible for low J line suppression during overtone lasing.

Elimination of the RR reactions from the model resulted in lower concentrations for the low J states of HF(0, 3–5), HF(1, 3–5), and HF(2, 3, 4) for both overtone lasing and zero power conditions; see Figs. 1–6. The concentrations calculated for states HF(0, 6–10),

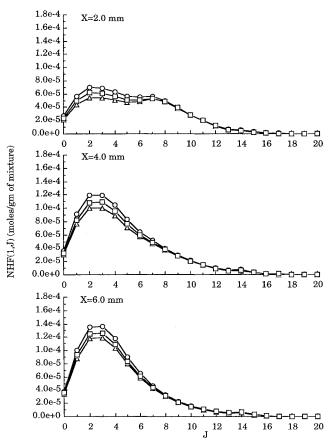


Fig. 2 Comparison of ORNECL HF(1, J) concentrations for zero power and overtone lasing conditions. These calculations were performed with the complete reaction set. The profiles are plotted at 2, 4, and 6 mm downstream from the nozzle exit plane: — \bigcirc —, zero power conditions; — \bigcirc —, overtone lasing, $P_{20}(3-7)$ absorption.

HF(1, 6–10), and HF(2, 5–10) with no RR are larger than those calculated with RR for both zero power and overtone lasing conditions; see Figs. 1–6. These results indicate that RR populates the low J HF(0, 3–5), HF(1, 3–5), and HF(2, 3, 4) states at the expense of the high J HF(0, 6–10), HF(1, 6–10), and HF(2, 5–10) states, respectively. This effect was observed in the experiments by Polanyi and Woodall. When RR is minimized, the HF(v, J) populations tend to stay in the states populated by the pumping reaction; see Figs. 4–6.

To determine the extent to which RR is responsible for the low J line suppression measured during overtone lasing, the difference between the zero power and overtone lasing concentration profiles calculated with and without RR was compared for states HF(v, 3-10) in Figs. 1-6. Figure 6 shows that, with no RR, the populations of HF(2, 0-4) are the same with and without overtone lasing. Since Fig. 3 shows that the populations of HF(2, 0-4) are decreased when overtone lasing occurs, comparison of Figs. 3 and 6 demonstrates that the removal of population from HF(2, 6-11) by overtone lasing reduces the transfer in population to the low J states by RR.

Comparison of Figs. 1 and 4 shows two effects of overtone lasing on the populations of HF(0, 3–5). The first point to be noted is that RR is the major cause of population transfer from HF(0, 6–12) to HF(0, 3–5). Second, when there is no RR, overtone lasing results in a decrease in HF(0, 3–5) populations below their zero power values. This is a consequence of the depletion of the HF(2, 6–11) states by overtone lasing, which results in decreased deactivation of these levels to the HF(0, 3–5) states. When RR is included, overtone lasing results in an increase in the populations of the HF(0, 3–5) states compared with their zero power values. This results from an increase in the rate of rotational transfer from the HF(0, 6–12) states to the HF(0, 3–5) states due to the increase in populations of the HF(0, 6–12) states by overtone lasing.

Comparison of the difference between the zero power and overtone lasing concentration profiles of the HF(1, J) states with and

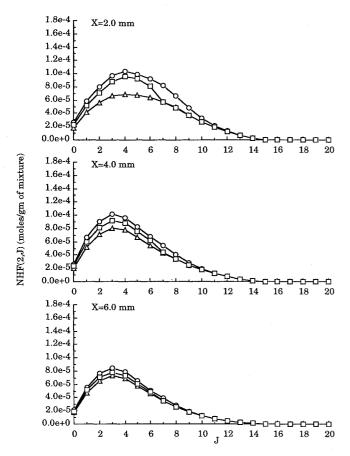
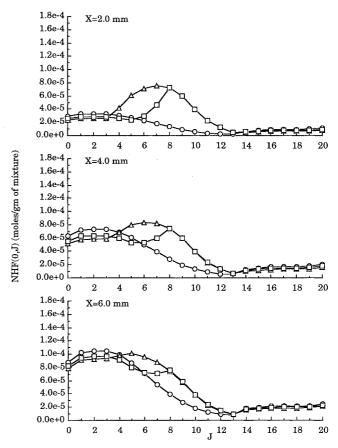


Fig. 3 Comparison of ORNECL HF(2, J) concentrations for zero power and overtone lasing conditions. These calculations were performed with the complete reaction set. The profiles are plotted at 2, 4, and 6 mm downstream from the nozzle exit plane: $-\bigcirc$, zero power conditions; $-\triangle$, overtone lasing, no absorption; $-\Box$, overtone lasing, $P_{20}(3-7)$ absorption.

without RR (Figs. 2 and 5) shows no appreciable change in this difference due to the removal of the RR reactions from the model. This indicates that the lower HF(1, 3–10) power-on concentrations are due to a decrease in the rate at which these levels are populated by the other collisional deactivation processes that transfer molecules from the HF(2, 6–11) states to the HF(1, 3–10) states. These rates are decreased because the populations of the HF(2, 6–11) states are decreased by overtone lasing. This decrease in effective collisional deactivation rates due to overtone lasing is termed the collisional deactivation effect. Note that with no RR, the other collisional deactivation processes result in a double peaked relaxation of HF(1, J).

Comparison of the HF(2, J) zero power and overtone lasing concentration profiles with and without RR, Figs. 3 and 6, shows that the elimination of RR resulted in practically no difference between the zero power and overtone lasing concentrations for HF(2, 3) and HF(2, 4). The HF(2, 5) state was not involved in overtone lasing in the case of the complete reaction set, Fig. 3, but it was involved in overtone lasing when RR was zeroed, Fig. 6; i.e., $P_{20}(6)$ lased. Even though the difference between the zero power and overtone lasing concentration profiles with and without RR for the HF(2, 5) state is about the same, it is due to two different causes. In the case of the complete reaction set, this difference is due to a decrease in the rate of RR that results from a decrease in population of the HF(2, 6-11) states by overtone lasing. In the case of no RR, this difference is due to overtone lasing on $P_{20}(6)$, which directly depopulates HF(2, 5). The states that are directly involved in overtone lasing, HF(2, 6-10), showed a significant increase in the difference between zero power and overtone lasing concentrations when the RR reactions were removed from the model, particularly at positions near the nozzle exit plane; see Figs. 3 and 6.

The preceding analysis shows that overtone lasing depopulates the high Jv = 2 states, which results in decreased RR rates that



populate the low Jv = 2 states, which causes the gains of the low $J2 \rightarrow 1$ lines to decrease. The same effect was seen in the ground state, where overtone lasing populates the high Jv = 0 states, which results in increased RR rates that populate the low Jv = 0 states, which causes the gains of the low $J1 \rightarrow 0$ lines to decrease. This effect is termed the RR effect.

This analysis showed that the fundamental gains are determined by three independent mechanisms when lasing occurs on the overtone. The first is the direct lasing effect that depopulates the v=2 states and populates the v=0 states that are directly involved in overtone lasing. This effect decreases the gains of the $P_1(J)$ and $P_2(J)$ lines whose upper or lower states are directly involved in $P_{20}(J)$ overtone lasing.

The second mechanism that affects the fundamental gains during overtone lasing is the RR effect that reduces the rate at which the low Jv = 2 states are populated and increases the rate at which the low Jv = 0 states are populated, resulting in suppression of the low J fundamental gains whose upper or lower levels are not directly involved in overtone lasing.

The third mechanism that affects the fundamental gains during overtone lasing is the collisional deactivation effect that reduces the rates at which the HF(0, J) and the HF(1, J) states, which are not directly involved in overtone lasing, are populated by the various collisional deactivation processes that transfer molecules from the high J v = 2 states (which are involved in overtone lasing) to these lower energy states.

There are two other effects noticeable in Figs. 1 and 2 that are the result of certain kinetic processes. The first effect is an increase in the HF(0, J) concentrations for J > 13. The zero power HF(v, J) concentrations were calculated with the complete reaction set and with the HF-H₂ VV transfer reaction⁴:

$$H_2(1) + HF(0, J''') \Leftrightarrow H_2(0) + HF(2, J')$$

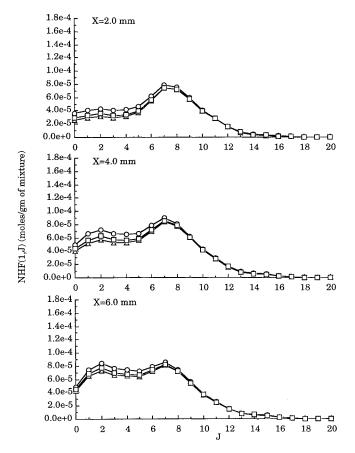


Fig. 5 Comparison of ORNECL HF(1, J) concentrations for zero power and overtone lasing conditions. These calculations were performed with no RR in the model. The profiles are plotted at 2, 4, and 6 mm downstream from the nozzle exit plane: $-\bigcirc$, zero power conditions; $-\triangle$, overtone lasing, no absorption; $-\Box$, overtone lasing, $P_{20}(3-7)$ absorption.

removed from the model. The results of these calculations indicated that this increase in the HF(0, J > 13) concentrations is due to the above HF-H₂ VV transfer reaction that populates the high J v = 0 states through a near resonant energy transfer from low J v = 2 states. The second effect is the small bump in the concentration of HF(1, J) at HF(1, 14), shown in Fig. 2 at axial positions of 4 and 6 mm downstream from the nozzle exit plane. This small increase in the concentration of HF(1, 14) is due to a near resonant collisional deactivation process from v = 2, J = 3, 4 to v = 1, J = 14.

Although the preceding analysis has explained why the fundamental gains of the low J lines whose upper or lower levels are not directly involved in overtone lasing are suppressed, it has not explained why the gains of these lines are suppressed more than the gains of the high J lines whose upper or lower levels are directly involved in overtone lasing.

B. Comparison of Calculated and Experimental RFG Suppression as a Function of Medium Saturation and RR Rate

To evaluate the computer model as a tool for the study of RFG, calculations were performed with overtone mirror reflectivities of 99.7/98.0% ($\alpha_{\rm sat}=0.000386787$), 99.78/99.67% ($\alpha_{\rm sat}=0.000091798$, $L_g=30$ cm), and 99.8/99.86% ($\alpha_{\rm sat}=0.000056716$). For all three levels of saturation, the model overpredicts the fundamental gain suppression ($\Delta\alpha$) for the $P_1(8,9)$ and $P_2(8,9)$ lines whose upper or lower levels are directly involved in overtone lasing and underpredicts the suppression in the case of $P_1(4)$ and $P_2(4,5)$. The model predicts the suppression ($\Delta\alpha$) for the $P_1(5-7)$ and $P_2(6,7)$ lines at the three levels of medium saturation reasonably well, but even for these lines, there is disagreement between the calculated and the experimental RFG profiles. The experiments show that the highest suppression occurred between 2 and 6 mm downstream from the nozzle exit plane. The model predicts that the highest suppression occurs between 1 and 2 mm downstream from

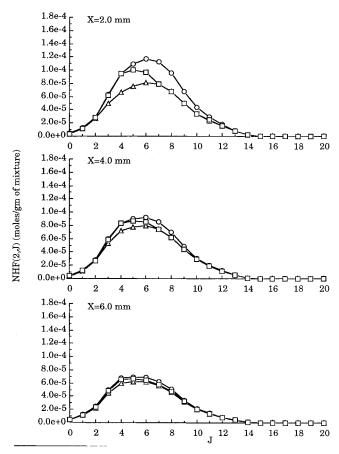


Fig. 6 Comparison of ORNECL HF(2, J) concentrations for zero power and overtone lasing conditions. These calculations were performed with no RR in the model. The profiles are plotted at 2, 4, and 6 mm downstream from the nozzle exit plane: — \bigcirc —, zero power conditions; — \bigcirc —, overtone lasing, no absorption; — \square —, overtone lasing, $P_{20}(3-7)$ absorption.

the nozzle exit plane. Comparison of the calculated and the experimental power spectral distributions for these three levels of medium saturation indicates that the predicted and measured spectra are in reasonable agreement.¹

The disagreement between the calculated and the experimental RFG profiles is in part due to the Fabry-Perot resonator optics model employed by ORNECL and the fact that the RFG experiments were performed with 4-m concave mirrors that form a stable resonator. A Fabry-Perot resonator results in a disproportionately high intracavity intensity near the nozzle exit plane, as opposed to a stable resonator. This is due to the lack of upstream-downstream coupling of the radiation field in the Fabry-Perot resonator. The peak intracavity intensities predicted by the ORNECL Fabry-Perot model for the three levels of medium saturation¹ (from the lowest to the highest) are 2.15, 54, and 98 kW/cm². The peak intracavity intensities estimated from the experimental intracavity power and overtone beam intensity profiles for these three levels of medium saturation¹ are 0.243, 21, and 32 kW/cm², respectively. This means that the Fabry-Perot model predicted peak intracavity intensities that are 8.8, 2.6, and 3.1 times larger than the corresponding peak intracavity intensities obtained from the experiments. The intracavity intensities predicted by the Fabry-Perot model peaked at axial positions between 0.5 and 1.5 mm downstream from the nozzle exit plane, whereas the experimental intracavity intensity distribution has a relatively flat profile.1 The high peak intracavity intensities predicted by the Fabry-Perot model at axial positions of 0.5-1.5 mm downstream from the nozzle exit plane result in high fundamental gain suppression at these positions for the lines whose upper or lower levels are directly involved in overtone lasing. This effect is demonstrated by the sharp decrease in the RFG profiles near the nozzle exit plane.

In an effort to improve the agreement between the experimental and calculated RFG profiles, the fluid dynamic, kinetic, stable resonator model, ORNECL-SR, was run with overtone mirror

reflectivities of 99.78/99.67%. ORNECL-SR uses a geometric optics stable resonator model to simulate the upstream-downstream coupling of the radiation field. This coupling has a strong effect on the intracavity intensity distribution. The peak intracavity intensity predicted by ORNECL-SR for the 99.78/99.67% case is 26 kW/cm². The peak intracavity intensity estimated from the experimental intracavity power and overtone beam intensity profile for these mirrors is 21 kW/cm². The ORNECL-SR peak intensity is in reasonable agreement with that estimated from the data. Agreement between the experimental and theoretical intracavity intensity distributions is one of the main requirements for a model to correctly predict the RFG profiles.

The improved intracavity intensity distribution predicted by ORNECL-SR resulted in better agreement between the calculated and experimental RFG profiles for lines $P_1(8, 9)$ and $P_2(7-9)$. The sharp decrease in the RFG profiles of these lines predicted by the Fabry-Perot model was eliminated, and maximum suppression occurred between 1.5 and 4.0 mm downstream from the nozzle exit plane, which is in better agreement with data. The more realistic intensity distribution predicted by ORNECL-SR resulted in improved agreement between the calculated and experimental suppression ($\Delta \alpha$) of the gains for lines $P_1(8,9)$ and $P_2(7-9)$ whose upper or lower levels were directly involved in overtone lasing. The RFG profiles and the fundamental gain suppression $(\Delta \alpha)$ of the low J lines whose upper or lower levels were not directly involved in overtone lasing were not improved by the use of the stable resonator model. The stale resonator model still overpredicts the fundamental gain suppression of the high J lines and underpredicts the fundamental gain suppression of the low J lines.

Since the improved optics model in ORNECL-SR did not eliminate the discrepancy between the calculated and experimental low J and high J line suppression and since it was shown in Sec. II.A that RR is the primary mechanism responsible for the suppression of the low J lines, calculations were performed to determine the effect of the RR rate on suppression of the fundamental gains. When ORNECL was baselined to fundamental data, the RR rate was reduced by a factor of 10 to obtain better agreement between the experimental and calculated ZPG profiles. To investigate the effect of RR on fundamental gain suppression, two ORNECL calculations with larger RR rates were performed.

The first set of ORNECL calculations was performed with 50% of the original RR rate and the second set was performed with the original RR rate. 5,6 The calculated ZPG and RFG profiles obtained at 0.1, 0.5, and 1.0 RR were compared with the experimental ZPG and RFG profiles. These comparisons showed that, for the 0.5 and 1.0 RR cases, the model overpredicts the $P_1(4-6)$ and $P_2(4-7)$ ZPG, underpredicts the $P_1(7-9)$ ZPG, and gives good agreement between the experimental and calculated $P_2(8, 9)$ ZPG profiles. In terms of the RFG profiles, the 0.5 and 1.0 RR models overpredict the $P_1(4-6)$ and $P_2(4-7)$ RFG and underpredict the $P_1(7-9)$ and $P_2(8, 9)$ RFG. ¹ These comparisons showed that the increase in RR resulted in worse agreement between the experimental and calculated ZPG and RFG profiles for all lines, except for the $P_2(8, 9)$ ZPG profiles, where the higher RR resulted in better agreement between the calculations and the experiment. The higher RR rates resulted in better agreement¹ between the calculated and experimental suppression $(\Delta \alpha)$ of the high J and low J lines. The best agreement of fundamental gain suppression $(\Delta \alpha)$ with data was obtained with the original RR rate. For this case, the predicted suppression $(\Delta \alpha)$ for both the high J and the low J lines was in reasonable agreement with data as illustrated in Figs. 7 and 8. In addition, for this case, the model predicted higher suppression on the $1 \rightarrow 0$ lines than on the $2 \rightarrow 1$ lines, in agreement with data.1

These calculations indicate that the ORNECL prediction of the suppression $(\Delta \alpha)$ of the fundamental gains on both the low J and the high J lines due to overtone lasing is in reasonable agreement with data when the original RR rate is used. With 10% of the original RR rate, the model is in reasonable agreement with the ZPG profiles, Fabry-Perot power, and spectra. To resolve this dichotomy, a comprehensive study of the fluid dynamic model and the effect of all of the reactions in the kinetic model on the ZPG and RFG profiles should be undertaken. A resolution of this dichotomy will

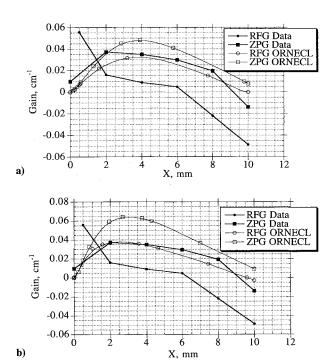


Fig. 7 Comparison of $P_1(5)$ experimental and ORNECL ZPG and RFG profiles (99.78/99.67% mirrors) obtained with RR rates of a) 0.1 RR and b) 1.0 RR.

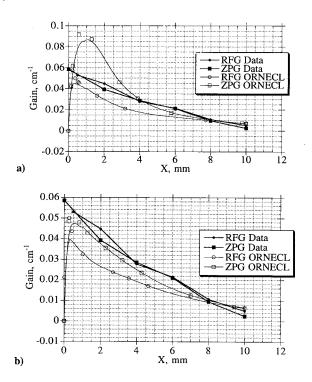


Fig. 8 Comparison of $P_2(9)$ experimental and ORNECL ZPG and RFG profiles (99.78/99.67% mirrors) obtained with RR rates of a) 0.1 RR and b) 1.0 RR.

not affect the calculation (Sec. II.A) that alteration of the RR rate by overtone lasing is the mechanism responsible for suppression of the low J gains whose upper or lower states are not directly involved in overtone lasing. For larger RR rates, this effect would be even more pronounced.

III. Concluding Remarks

The first set of RF-AR data was obtained at a relatively high medium saturation with two nominally 99.7% reflective mirrors. These data indicated that the low J fundamental gains were

suppressed more than the high J fundamental gains even though their upper or lower levels were not directly involved in overtone lasing.

Analysis of the HF mole/mass ratios calculated by the rotational nonequilibrium model ORNECL showed that the fundamental gains are determined by three independent mechanisms when lasing occurs on the overtone. The first mechanism is the direct lasing effect that depopulates the v=2 states and populates the v=0 states that are directly involved in overtone lasing. This effect decreases the gains of the $P_1(J)$ and $P_2(J)$ lines whose upper or lower levels are directly involved in $P_{20}(J)$ overtone lasing.

The second mechanism that affects the fundamental gains during overtone lasing is the RR effect that reduces the rate at which the low Jv = 2 states are populated and increases the rate at which the low Jv = 0 states are populated, resulting in suppression of the low J fundamental gains whose upper or lower levels are not directly involved in overtone lasing.

The third mechanism that affects the fundamental gains during overtone lasing is the collisional deactivation effect that reduces the rates at which the HF(0, J) and the HF(1, J) states, which are not directly involved in overtone lasing, are populated by the various collisional deactivation processes that transfer molecules from the high Jv=2 states (that are involved in overtone lasing) to these lower energy states.

The ORNECL computer model was used to perform a ZPG calculation and RFG calculations for overtone mirror reflectivities of 99.7/98.0, 99.78/99.67, and 99.8/99.86% to determine the ability of the model to predict RFG as a function of medium saturation. The model overpredicted the fundamental gain suppression ($\Delta \alpha$) for the $P_1(8, 9)$ and $P_2(8, 9)$ lines whose upper or lower levels are involved in overtone lasing and underpredicted the suppression ($\Delta \alpha$) on the $P_1(4)$ and $P_2(4, 5)$ lines. The model's prediction of the suppression $(\Delta \alpha)$ for the $P_1(5-7)$ and $P_2(6,7)$ lines at these three levels of medium saturation was in reasonable agreement with data, but even for these lines, there was disagreement between the calculated and experimental RFG profiles. This disagreement between the calculated and experimental RFG profiles is in part due to the Fabry-Perot resonator model employed by ORNECL. A Fabry-Perot resonator results in a disproportionately high intracavity intensity and high fundamental gain suppression near the nozzle exit plane compared with the stable resonator used to perform the RFG experiments.

To improve the agreement between the calculated and experimental RFG profiles, the stable resonator version of ORNECL, ORNECL-SR, was used to calculate the RFG for overtone mirror reflectivities of 99.78/99.67%. The stable resonator model resulted in a more realistic intracavity intensity distribution and in improved agreement between the calculated and experimental RFG profiles for lines $P_1(8,9)$ and $P_2(7-9)$. The stable resonator model overpredicted the fundamental gain suppression $(\Delta\alpha)$ for the $P_1(8,9)$ and $P_2(8,9)$ lines and underpredicted the fundamental gain suppression $(\Delta\alpha)$ for the low J lines whose upper or lower levels were not directly involved in overtone lasing.

The peak intracavity intensity predicted by ORNECL-SR was in good agreement with that estimated from the data. The power spectral distributions predicted by ORNECL and ORNECL-SR were in good agreement with the experimental power spectral distribution.

Since it was shown that RR was the major cause of the suppression of the low J lines, in an effort to determine why the low J lines are suppressed more than the high J lines, two ORNECL calculations were performed with higher rates of RR. When ORNECL was baselined to fundamental data, the RR rate was reduced by a factor of 10 to obtain better agreement between the calculated and experimental ZPG profiles, Fabry-Perot power, and spectra. The first set

of ORNECL calculations was performed with 50% of the original RR rate and the second set was performed with the original RR rate.

When the RR rate was increased by a factor of 10 to the original RR rate, the model's prediction of the fundamental gain suppression $(\Delta \alpha)$ of both the low J and the high J lines was in reasonable agreement with the data and the model predicted greater suppression on the low J lines than on the high J lines in agreement with the data.

With 10% of the original RR rate, the model was in reasonable agreement with the measured fundamental ZPG profiles, Fabry–Perot power, and spectra. With the original RR rate, the model was in reasonable agreement with the measured fundamental gain suppression ($\Delta\alpha$) of both the high J and low J lines but did not agree with the ZPG or RFG profiles. To resolve this dichotomy, a comprehensive study of the fluid dynamic model and the effect of all of the reactions in the kinetic model on the ZPG and RFG profiles should be undertaken.

The analyses of the HF(v, J) concentrations (Sec. II.A) and the ZPG and RFG profiles (Sec. II.B) indicate that RR is the primary mechanism responsible for the suppression of the low J lines whose upper or lower levels are not involved in overtone lasing. RR, however, may not be the only mechanism responsible for the suppression of the low J lines. The double peaked relaxation of the HF(1, J) concentration profile calculated with no RR suggests that other processes, such as collisional deactivation, may play an important role in the suppression of the low J lines when lasing on the overtone. Further studies are necessary to identify these chemical kinetic processes and determine their effects.

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