# Study of the residual fundamental gain of a cw HF chemical laser

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## ABSTRACT

The main objective of this work was to determine the extent to which lasing on the overtone suppresses the gain on the fundamental transitions $P_1(4\text{-}9)$ and $P_2(4\text{-}9)$ as a function of media saturation on the overtone. This was accomplished by a comparison of the residual fundamental gain (RFG) data obtained at three different levels of media saturation with the corresponding zero power gain (ZPG) data.

Comparison of the residual fundamental amplification ratio (RF-AR) data with the zero power amplification ratio (ZP-AR) data indicated that the gains of the low J lines $P_1(4\text{-}6)$ and $P_2(4\text{-}6)$ were suppressed more than the gains of the high J lines even though their upper or lower levels were not directly involved in overtone lasing.

Analysis of the HF mole/mass ratios calculated by a rotational nonequilibrium computer 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. The second mechanism is the "rotational relaxation effect" that reduces the rate at which the low J v=2 states are populated and increases the rate at which the low J v=0 states are populated. The third mechanism is the "collisional deactivation effect" that reduces the rates at which the HF(0,J) and the HF(1,J) states that 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 (that are involved in overtone lasing) to these lower energy states. Further analysis of the HF(v,J) concentrations and the ZPG and RFG calculations indicated that rotational relaxation is the primary mechanism responsible for the suppression of the low J lines whose upper or lower levels are not involved in overtone lasing.

Keywords: HF chemical laser, residual fundamental gain, zero power gain, rotational relaxation

## 1. INTRODUCTION

The gains of the HF fundamental 2→1 and 1→0 transitions are almost two orders of magnitude larger than the gains of the 2→0 overtone transitions. Because of this, for the overtone to lase, the fundamental must be suppressed. The fundamental transitions are suppressed through the use of selective mirror coatings, i.e., the mirrors are less than 1% reflective at the fundamental wavelengths and greater than 99% reflective at the overtone wavelengths. When the overtone laser is scaled to large gain lengths, even though the fundamental mirror reflectivities are less than 1%, the fundamental transitions may start to lase and/or amplified spontaneous emission (ASE) may reach levels that would substantially limit the performance of the device.[1] The occurrence of these phenomena will depend on the magnitude of the fundamental gains while lasing on the overtone, called the RFG. Thus, the magnitude of the RFG may present a scaling limitation of the overtone laser.

The fundamental gain suppression due to overtone lasing is determined by comparing the RFG with the ZPG. The SSL RFG's were measured for the peak gain lines $P_1(4\text{-}9)$ and $P_2(4\text{-}9)$ with a pair of 99.7% / 99.7% nominally reflective overtone mirrors, Section 2. These were the mirrors that gave the best overtone efficiency.[2] The gains of the low J lines $P_1(4\text{-}6)$ and $P_2(4\text{-}6)$ whose upper or lower levels were not directly involved in overtone lasing were suppressed more than the gains of the high J $P_1(7\text{-}9)$ and $P_2(7\text{-}9)$ lines whose upper or lower levels were directly involved in overtone lasing.

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To determine the effect of media saturation, the residual fundamental gains were measured for two additional levels of media saturation. Experiments were performed at low media saturation with 98.0% / 99.7% reflective overtone mirrors and at high media saturation with 99.8% / 99.86% reflective overtone mirrors, Section 2.

A detailed rotational nonequilibrium model (ORNECL)[3] of the laser was used 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 residual fundamental gains as a function of media saturation. ORNECL calculations with and without rotational relaxation (RR) in the model indicated that the primary mechanism responsible for the suppression of the low J fundamental gains is a change in the effective rate at which rotational relaxation populates the upper and lower levels of the low J lines due to overtone lasing, Section 3.

ORNECL calculations performed with the original RR rate predicted well the fundamental gain suppression ($\Delta\alpha$) of both the high J and low J lines, Section 3. Several concluding remarks are presented in Section 4.

## 2. RESIDUAL FUNDAMENTAL GAIN MEASUREMENTS AS A FUNCTION OF MEDIA SATURATION

Since the overtone $2\rightarrow0$ transitions share the same upper levels with the fundamental $2\rightarrow1$ transitions and the same lower levels with the fundamental $1\rightarrow0$ transitions, lasing on the overtone should result in a decrease of the fundamental gains. To determine the extent to which this occurs, the supersonic laser RF-AR's were measured for the peak zero power gain lines $P_1(4\text{-}9)$ and $P_2(4\text{-}9)$ while the laser was operating on the overtone. These measurements were performed with 99.7% nominally reflective overtone mirrors that gave the largest overtone power and efficiency.[3] With the CLI off, the overtone output power was 10.26 Watts with lines $P_{20}(7\text{-}10)$ lasing. With the CLI on, the overtone output power was 7.49 Watts with lines $P_{20}(8\text{-}11)$ lasing. Data were taken at 0.5, 2, 4, 6, 8, 10 and 11 mm downstream from the nozzle exit plane as a function of vertical position in the flow channel. The layout of the residual fundamental gain experiment is shown in Fig. 1.

A comparison of the vertically averaged RF-AR data with the corresponding ZP-AR data is presented in Fig. 2 (dark symbols). These figures show that the $P_1(7\text{-}9)$ lines are generally suppressed more than the $P_2(7\text{-}9)$ lines and that the low J lines, $P_1(4\text{-}6)$ and $P_2(4\text{-}6)$, are suppressed more than the high J lines, $P_1(7\text{-}9)$ and $P_2(7\text{-}9)$.

Analysis of these data showed that the low J fundamental gains are suppressed between 41% and 84% and the high J fundamental gains are suppressed between 3% and 43%. For both low and high J lines, the maximum suppression occurred between 2 and 6 mm downstream from the nozzle exit plane. Since the measured overtone beam diameter was about 9.0 mm with its upstream edge at the nozzle exit plane, the maximum suppression of the fundamental gain occurred near the center of the overtone beam.

To determine the variation of RFG with media saturation, the RF-AR's were measured for a range of overtone reflectivities. The data generated by these experiments will provide the database needed to check computer model predictions of the residual fundamental gain profiles and fundamental gain suppression ($\Delta\alpha$).

The residual fundamental amplification ratio measurements with an overtone resonator of low reflectivity were performed first. Computer simulation of RFG vs $\sqrt{R_1 \times R_2}$ suggested that an overtone resonator with mirror reflectivities of 99.7% and 98.0% would provide minimal suppression of the fundamental gains.[4] Mirrors with 99.7% and 98.0% reflectivity were used to perform these low reflectivity RF-AR experiments.

With the CLI off, the maximum overtone output power was 5.1 Watts with lines $P_{20}(6\text{-}8)$ lasing. With the CLI on, the maximum output power was 0.62 Watts with lines $P_{20}(7,8)$ lasing. Residual fundamental amplification ratio data were obtained at this low level of media saturation for axial positions of 0.5, 2, 4, 6, 8, 10 and 11 mm downstream from the nozzle exit plane as a function of vertical position in the flow channel.

Comparison of the ZP-AR and RF-AR data indicated weak suppression in the cases of $P_1(7)$, $P_2(5)$ and $P_2(6)$ between 2 and 6 mm downstream from the nozzle exit plane, while there is no clear indication of suppression in the case of the other lines. This supports the computer results of residual fundamental gain vs. media saturation that showed that the fundamental gain suppression would be minimal in the SSL with overtone mirrors of $R_1$=0.98 and $R_2$=0.997.[4]

Computer calculations[4] of residual fundamental gain and amplification ratio as a function of media saturation indicated that above a reflectivity of 99.7%, the RF-AR does not change significantly with increased reflectivity even when the gain length is increased by a factor of two. To check the computer predictions and to determine the relation between media saturation and residual fundamental gain, RF-AR data were obtained at an increased level of media saturation. New overtone mirrors with a nominal reflectivity of 99.9% were obtained to perform these experiments. The reflectivity and transmissivity of these mirrors were measured by Helios in both the fundamental and the overtone.[4] The mirrors had fundamental reflectivities of less than 1% and fundamental transmissivities of more than 85%; these numbers are typical for all of our overtone mirrors. The power spectral distribution obtained with these mirrors contained lines $P_{20}(7\text{-}12)$, indicating that they have a higher reflectivity than the nominally 99.7% reflective mirrors.[2,4]

These new high reflectivity mirrors resulted in an intracavity flux that is about 55% larger than that obtained with the 99.7% nominally reflective mirrors. The ratio of $\alpha_{sat}$ obtained with these high reflectivity mirrors to $\alpha_{sat}$ obtained with the two 99.7% nominally reflective mirrors is 0.566. The higher reflectivity mirrors result in an $\alpha_{sat}$ that is 43.4% smaller than that of the 99.7% nominally reflective mirrors.

With these high reflectivity mirrors and the CLI off, lasing was observed on lines $P_{20}(7\text{-}12)$, and the output power was 11.9 Watts. With the CLI on, the SSL lased on lines $P_{20}(8\text{-}12)$, and the total output power was 8.5 Watts. Residual fundamental amplification ratio data were obtained at 0.5, 2.0, 4.0, 6.0, 8.0 and 10.0 mm downstream from the nozzle exit plane. These data were plotted along with the corresponding average data obtained with the two 99.7% nominally reflective mirrors, Fig. 2. The curves with the dark symbols show RF-AR data obtained with the nominally 99.7/99.7% reflective mirrors (dark squares), and the corresponding ZP-AR data (dark circles). The curves with the open symbols show RF-AR data obtained with the 99.8/99.86% reflective mirrors (open squares), and the corresponding ZP-AR data (open circles and triangles). These figures show that the suppression obtained with the 99.8/99.86% reflective mirrors and with the two nominally 99.7% reflective mirrors are about the same for all lines. This indicates that the computer prediction that, above a reflectivity of 99.7%, the RF-AR does not change significantly with increased reflectivity[4] agrees with experiment.

## 3. SIMULATION OF RESIDUAL FUNDAMENTAL GAIN

Comparisons between average ZP-AR data and RF-AR data obtained with the two 99.7% nominally reflective mirrors indicated that the 1→0 lines are generally suppressed more than the 2→1 lines, especially at high J, and that the low J $P_1(4\text{-}6)$ and $P_2(4\text{-}6)$ lines are suppressed more than the high J $P_1(7\text{-}9)$ and $P_2(7\text{-}9)$ lines.[4] The SSL was lasing on lines $P_{20}(8\text{-}11)$ while the RF-AR measurements were performed.[4] 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 / gm of mixture ) for the vibrational and rotational levels associated with fundamental and overtone lasing were calculated using ORNECL for zero power and overtone lasing.[4] The overtone lasing calculations were performed with overtone mirror reflectivities of 99.78/99.67%. Since the model predicts lasing on $P_{20}(3\text{-}12)$ while experimentally lasing occurred on

$P_{20}$(8-11), a calculation was made with $SF_x$ absorption[4] 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, Figs. 3-5.

Polanyi and Woodall[5] showed that the pumping reaction populates HF(1,4-13), HF(2,2-13) and HF(3,0-6) and that rotational relaxation was the major collisional process responsible for the transfer of population from the high J states 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 overtone lasing on $P_{20}$(7-12) depopulates the peak of the pumping distribution in v=2, and populates v=0, J=7-12, overtone lasing may have a major effect on the subsequent transfer of populations to the low J states in v=2 and v=0, and thus would be the cause of the suppression of the low J lines. To determine the extent to which the rotational relaxation reactions affect the population of the low J states, the rotational relaxation rate constant was set equal to zero and the zero power and overtone lasing calculations were repeated.

Elimination of the rotational relaxation 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, Figs. 3-8. The concentrations calculated for states HF(0,6-10), HF(1,6-10) and HF(2,5-10) with no rotational relaxation are higher than those calculated with rotational relaxation for both zero power and overtone lasing conditions, Figs. 3-8. These results indicate that rotational relaxation 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.[5] When rotational relaxation is minimized, the HF(v,J) populations tend to stay in the states populated by the pumping reaction, Figs. 6-8.

Analysis of the HF(2,J) zero power and overtone lasing concentration profiles with and without rotational relaxation, Figs. 5 and 8, shows that overtone lasing depopulates the high J v=2 states, which in turn results in decreased rotational relaxation rates that populate the low J v=2 states, which causes the gains of the low J $2 \rightarrow 1$ lines to decrease. The same effect was seen in the ground state, Figs. 3 and 6, where overtone lasing populates the high J v=0 states, which in turn results in increased rotational relaxation rates that populate the low J v=0 states, which causes the gains of the low J $1 \rightarrow 0$ lines to decrease. This effect is termed the "rotational relaxation effect".

Comparison of the difference between the zero power and overtone lasing concentration profiles of the HF(1,J) states with and without rotational relaxation, Figs. 4, 7, indicated no appreciable change in this difference due to the removal of the rotational relaxation reactions from the model. This shows 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". It should be noted that, with no rotational relaxation, the other collisional deactivation processes result in a double peaked relaxation of HF(1,J).

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 "rotational relaxation effect" that reduces the rate at which the low J v=2 states are populated and increases the rate at which the low J v=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 that 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 that are involved in overtone lasing to these lower energy states.

To evaluate the computer model as a tool for the study of residual fundamental gain, three calculations were performed with overtone mirror reflectivities of 99.7/98.0%, 99.78/99.67% and

99.8/99.86%.[4] At all three levels of media saturation, the model over predicted 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 under predicted the suppression in the case of $P_1(4)$ and $P_2(4,5)$. The model predicted the suppression ($\Delta\alpha$) for the $P_1(5\text{-}7)$ and $P_2(6,7)$ lines at the three levels of media saturation reasonably well, but even for these lines, there is disagreement between the calculated and the experimental RFG profiles.[4] Comparison of the calculated and the experimental power spectral distributions for these three levels of media saturation indicates that the predicted and measured spectra are in reasonable agreement.[4]

Since it was shown that rotational relaxation 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.[3] To investigate the effect of RR on fundamental gain suppression, two ORNECL calculations with higher RR rate 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. The higher rotational relaxation rates resulted in better agreement between the calculated and experimental suppression 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 for both the high J and the low J lines was in reasonable agreement with data. In addition, for this case, the model predicted higher suppression on the $1\rightarrow0$ lines than on the $2\rightarrow1$ lines, in agreement with data.[4]

## 4. CONCLUDING REMARKS

The main objective of this work was to determine the extent to which lasing on the overtone suppresses the gain on the fundamental transitions $P_1(4\text{-}9)$ and $P_2(4\text{-}9)$ as a function of media saturation on the overtone. This was accomplished by a comparison of the RFG data obtained at three different levels of media saturation with the corresponding ZPG data.

The first set of RF-AR data was obtained at a relatively high media saturation with two nominally 99.7% reflective mirrors. The gains of the low J lines $P_1(4\text{-}6)$ and $P_2(4\text{-}6)$ were suppressed between 41% and 84% and the gains of the high J lines $P_1(7\text{-}9)$ and $P_2(7\text{-}9)$ were suppressed between 3% and 43%. The $1\rightarrow0$ lines were suppressed more than the $2\rightarrow1$ 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 surprising result was 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.

Residual fundamental gain measurements at low media saturation with overtone mirrors of 99.7/98.0% reflectivity 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. There was no suppression measured for any of the other lines. These results verified the computer model's prediction of minimal fundamental gain suppression at this low level of media saturation.[4]

Residual fundamental gain was measured at an increased level of media saturation with overtone mirrors of 99.8/99.86% reflectivity. The intracavity flux in this case was 55% higher and $\alpha_{sat}$ was 43.4% smaller than that of the two 99.7% nominally reflective mirrors. The suppression obtained with the 99.8/99.86% reflective mirrors was essentially the same as that obtained with the 99.7/99.7% mirrors. This result agrees with the computer model prediction that, above a reflectivity of 99.7%, the RF-AR does not change significantly with increased reflectivity.[4]

Since the maximum overtone device gain length will be determined by the ability of a fully saturated overtone laser to suppress the fundamental gains and reduce amplified spontaneous emission, RFG experiments should be performed with a longer device to experimentally determine the fundamental gain suppression as a function of gain length. The high J $P_2(7\text{-}9)$ lines whose upper states were directly involved in overtone lasing showed minimal suppression in the case of the 30 cm device. If this is also the case at longer gain lengths (higher media saturation), the gains of these 2→1 lines may present a significant scaling limitation for the overtone laser.

Analysis of the HF mole/mass ratios calculated by 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 "rotational relaxation effect" that reduces the rate at which the low J v=2 states are populated and increases the rate at which the low J v=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 that 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 (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 residual fundamental gain as a function of media saturation. The model over predicted 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 under predicted 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 $P_1(5\text{-}7)$ and $P_2(6,7)$ lines at these three levels of media saturation was in reasonable agreement with data, but even for these lines, there was disagreement between the calculated and experimental RFG profiles.

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 rotational relaxation. 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, 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 rotational relaxation rate was increased by a factor of ten 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.

Analysis of the HF(v,J) concentrations and the ZPG and RFG calculations indicate that rotational relaxation is the primary mechanism responsible for the suppression of the low J lines whose upper or lower levels are not involved in overtone lasing. Measurement of the gain on non-lasing transitions while lasing on related transitions has provided a tool for probing the effect of specific kinetic processes on the overall system.

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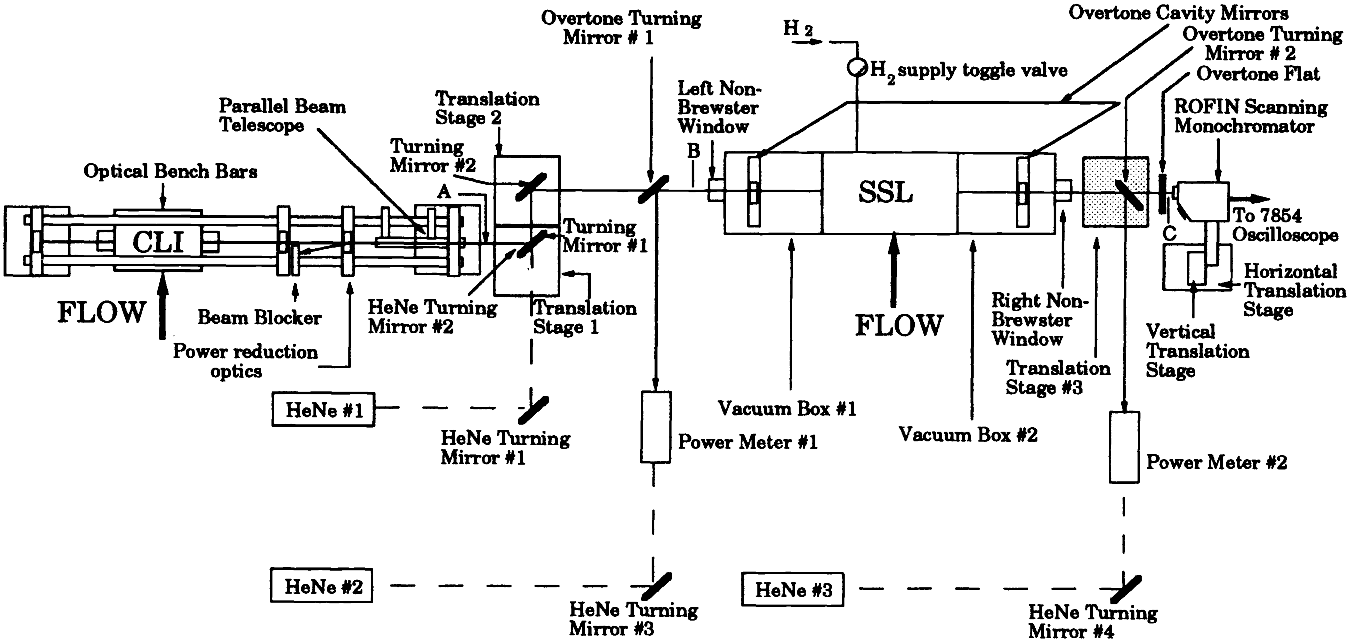

Note: HeNe Turning Mirror #2 removed during experiment

Figure 1. Schematic of the experimental layout for the zero power and residual fundamental gain studies.

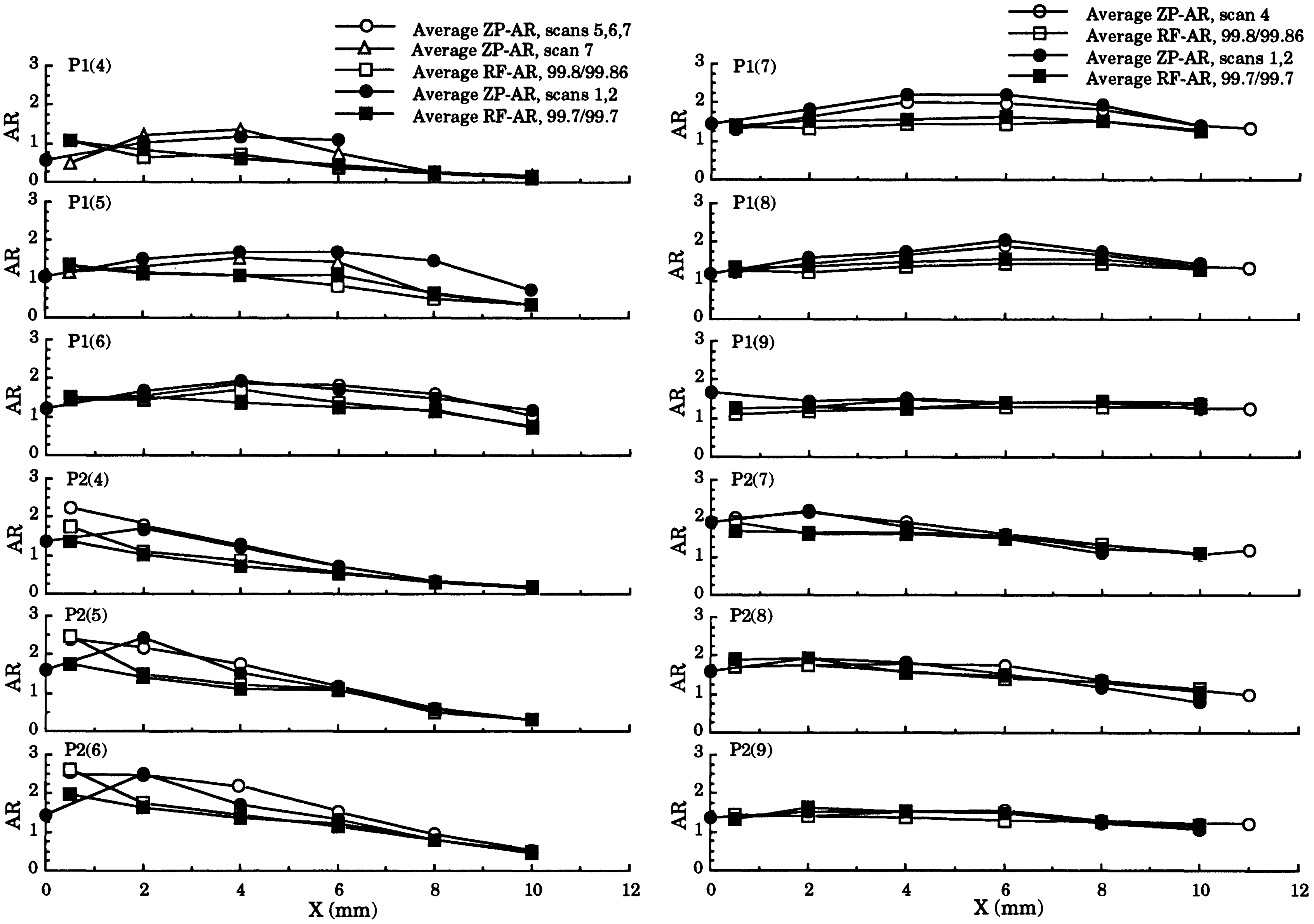

Figure 2. Comparison of ZP-AR and RF-AR data as a function of x and media saturation. The dark symbols represent RF-AR data obtained with 99.7/99.7% reflective mirrors and the corresponding ZP-AR data. The open symbols represent RF-AR data obtained with 99.8/99.86% reflective mirrors and the corresponding ZP-AR data.

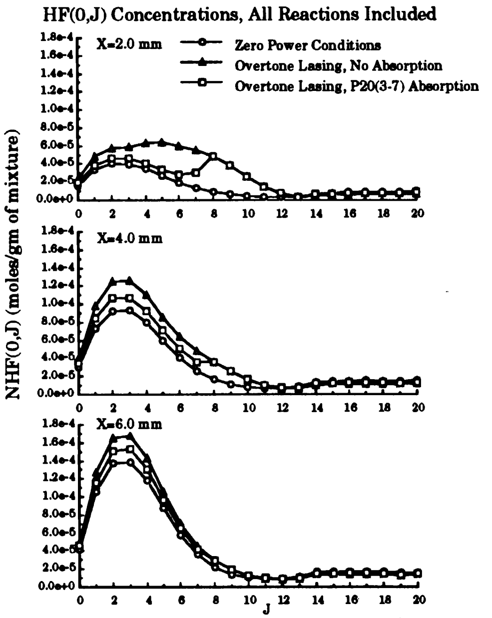

Figure 3. Comparison of ORNECL HF(0,J) concentrations for zero power and overtone lasing conditions. These calculations were performed with the complete reaction set.

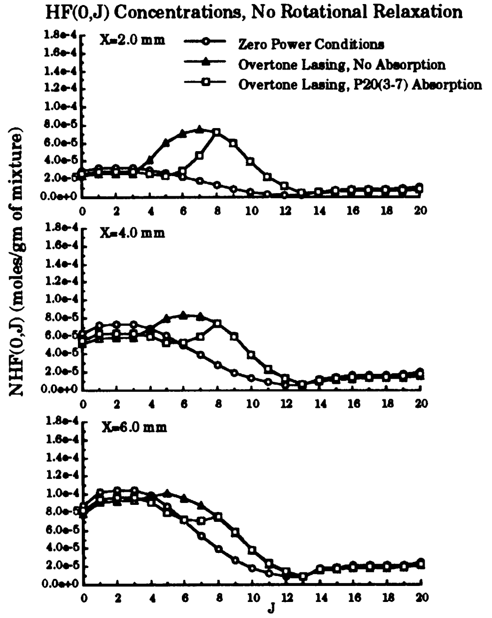

Figure 6. Comparison of ORNECL HF(0,J) concentrations for zero power and overtone lasing conditions. These calculations were performed with no rotational relaxation in the model.

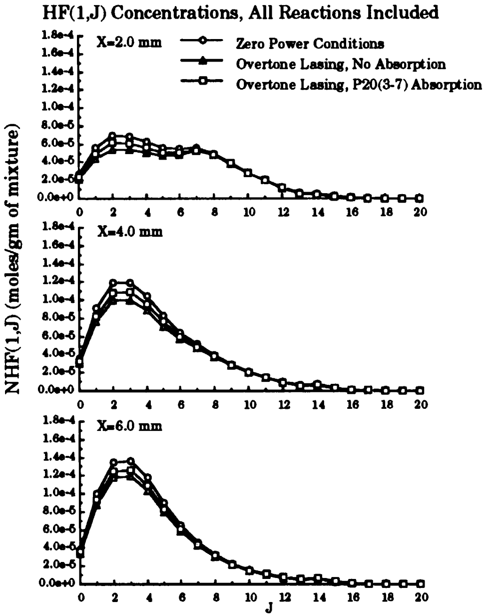

Figure 4. Comparison of ORNECL HF(1,J) concentrations for zero power and overtone lasing conditions. These calculations were performed with the complete reaction set.

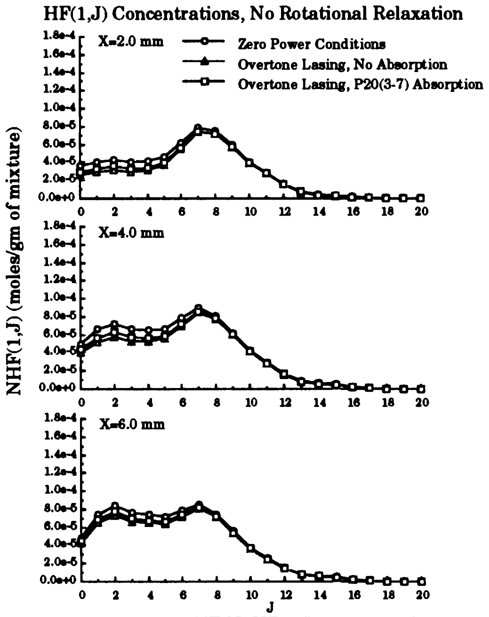

Figure 7. Comparison of ORNECL HF(1,J) concentrations for zero power and overtone lasing conditions. These calculations were performed with no rotational relaxation in the model.

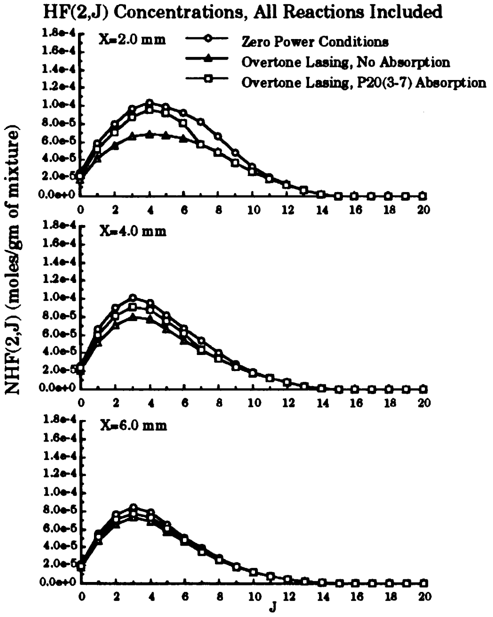

Figure 5. Comparison of ORNECL HF(2,J) concentrations for zero power and overtone lasing conditions. These calculations were performed with the complete reaction set.

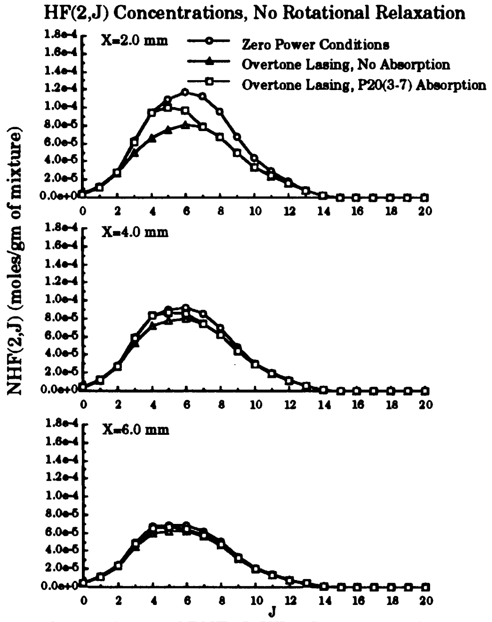

Figure 8. Comparison of ORNECL HF(2,J) concentrations for zero power and overtone lasing conditions. These calculations were performed with no rotational relaxation in the model.