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Modeling of High-Pressure 12-μm NH_3 Lasers

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Abstract. Experimental measurements of small-signal gain in an optically-pumped NH_3 amplifier are carried out at pressures ranging from 40 Torr to 760 Torr, and the results are used to validate a rate-equation model describing the amplifier dynamics. The gain measurements show that dilute mixtures of $<0.5\%$ NH_3 in N_2 are required to minimize the problems of gas heating due to pump absorption. The model is used to extrapolate the results to gas pressures of several atmospheres, and to demonstrate the potential for high-pressure operation of optically-pumped NH_3 lasers. For a pump intensity of 100 MW/cm^2, calculations indicate that operation of an NH_3-N_2 laser is feasible up to a pressure of 10 atm, which would provide a maximum continuous tuning range of 4 cm^{-1}. High-resolution spectroscopy reveals that gain on a few NH_3 transitions is eliminated at high pressures due to the presence of overlapping absorptions in other NH_3 bands.

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For many mid-infrared applications it is desirable to have a high-power laser source which can be tuned continuously in frequency and can produce pulses of sub-nanosecond duration. The general requirement for developing these characteristics is a broad gain bandwidth, which is often accomplished in gas lasers by operating at high pressure. Pulsed line-tunable NH_3 lasers that employ dilute mixtures of NH_3 in a buffer gas at pressures up to 100 Torr are highly efficient sources of radiation for more than 30 transitions in the 11–13 μm region [1, 2]. More recent studies have discussed an NH_3-N_2 laser which can operate at pressures up to 3.2 atm [3] (1 atm = 760 Torr = 101.3 kPa), and has been tuned continuously over a range of ~0.25 cm^{-1} (7.5 GHz) at a pressure of 1.1 atm [4]. Another report examines the use of high-pressure NH_3-He mixtures to amplify the emission of a tunable diode laser over a continuous range of 20 GHz [5]. In this paper, we use a rate-equation model to calculate the effects of pressures up to ~13 atm on the small-signal gain in optically-pumped, line-tunable NH_3 lasers. The model is validated by comparison with experimental measurements over a pressure range of 0.05 to 1 atm.

Increasing the pressure in any gas laser can increase the tuning range in two ways. First, the linewidth of each transition broadens, and second, the broadened profiles of adjacent lines can overlap. Unlike the even distribution of CO_2 laser lines, NH_3 laser transitions are spaced rather irregularly. Furthermore, there is a large variation in the maximum gain coefficient even for adjacent NH_3 lines chiefly due to the difference in statistical weight between ortho-NH_3 ($K=3n$) and para-NH_3 ($K=3n\pm1$). To evaluate the possible overlaps of NH_3 lines, let us consider a dilute mixture of NH_3 in N_2 at a pressure of 10 atm, which would give a pressure-broadened linewidth of ~25 GHz half-width at half maximum (HWHM) on the individual transitions. This width is sufficient to merge all the transitions within each $sP(J, K)$ multiplet from $J=3$ to 8. However, the spacings between $aP(J, K)$ transitions are much larger: ~25 GHz, ~40 GHz, and ~58 GHz between $K=1$ and 2, $K=2$ and 3, and $K=3$ and 4, respectively [6]. Thus, at a pressure of 10 atm, continuous tuning would likely be possible only up to $K=3$ within each $aP(J, K)$ multiplet, which would provide a maximum tuning range of ~120 GHz. The separation between successive values of J is generally

greater than 100 GHz, and is much too large to allow continuous tuning between multiplets at 10 atm.

As mentioned above, the possibility of generating short pulses is another reason for having a broad gain bandwidth. An estimate of the minimum pulse duration Δt that can be supported by a gain bandwidth $\Delta\nu$ (full-width) is given approximately by $\Delta\nu\Delta t \sim 1$. Thus, from the example above, a tuning range of 120 GHz is a sufficient bandwidth to support pulses of ~ 8 ps duration. Techniques for creating short pulses are well developed. Passive mode-locking of an NH_3 laser has produced pulses as short as 7 ns in pressures less than 100 Torr [7]. Synchronous pumping with a mode-locked source has generated pulses ~ 1 ns in duration [8, 9]. Applications of these techniques to NH_3 lasers operating at pressures of a few atmospheres should yield frequency-tunable pulses in the picosecond range.

In this paper, experimental and theoretical results are presented to determine the factors which limit high-pressure operation. Experimental measurements of small-signal gain are obtained by pumping $NH_3 - N_2$ mixtures at pressures up to 760 Torr with the $9R(30)$ CO_2 line. Theoretical calculations with a rate-equation model extend the investigations of small-signal gain to higher pressures, up to 10^4 Torr (~ 13 atm). The experimental technique and the theory for the rate-equation model are summarized in Sect. 1 and 2, respectively. The results presented in Sect. 3.1 show that the concentration of NH_3 strongly affects the maximum pressure attainable. As the pressure is increased, gas heating due to pump absorption reduces the gain coefficients. Section 3.2 examines the problem of absorption lines in the $2\nu_2 - \nu_2$ hot bands, which overlap the ν_2 laser transitions as the pressure is increased. The absorption on the $2\nu_2$ transitions is enhanced by optical pumping of the ν_2 band, and thus can strongly interfere with the gain on adjacent ν_2 transitions.

1. Experimental Technique

The apparatus and techniques employed to measure small-signal gain coefficients in NH_3 have been described in detail elsewhere [10, 11]. Only a brief description is provided here. A single-mode hybrid CO_2 laser tuned to $9R(30)$ is used to pump a grating-tunable NH_3 oscillator, as shown in Fig. 1. The $9R(30)$ line pumps the $sR(5, K)$ multiplet of NH_3 with pump offset frequencies ranging from 190 MHz for $sR(5, 0)$ [12] to a maximum value of 1.6 GHz [13]. Laser emission is obtained from most of the P-branch transitions and from several lines in the Q and R-branches. The output of the NH_3 laser is combined with part of the CO_2 pump beam to probe a separate amplifier cell. To facilitate tuning the NH_3 emission wavelength, the NH_3 laser is pumped through a dichroic mirror M_1. A second dichroic mirror M_2 transmits the $9R(30)$ pump pulse and directs the NH_3 laser output to the amplifier cell. Gain measurements are performed in the amplifier cell at the line center of the NH_3 transitions by fine tuning the length of the NH_3 oscillator with a piezoelectric translator (PZT). The $9R(30)$ pump pulse consists of an initial spike of ~ 250 ns full-width at half-maximum (FWHM), followed by a tail which lasts several microseconds. Gain measurements are taken during the period 0.3 to 1.2 μs after the peak of the pump pulse, as the pump intensity in that period is almost constant at about one-quarter of the peak intensity.

2. Theory

The rate-equation model that is employed for the calculations presented in this paper has been described previously [10], and is briefly outlined here. The model, which is used to evaluate the population dynamics in optically-pumped NH_3, is based on two main assumptions. First, it assumes that the rotational populations are always thermalized, except for the two levels of the pumped transition. The second assumption is that the two different species of NH_3, ortho-NH_3 or para-NH_3 are not coupled. These assumptions are justified by the very fast rotational relaxation rate relative to the vibration-translation (V − T) relaxation rate in $NH_3 - N_2$ mixtures, and the lack of collisional interconversion between ortho-NH_3

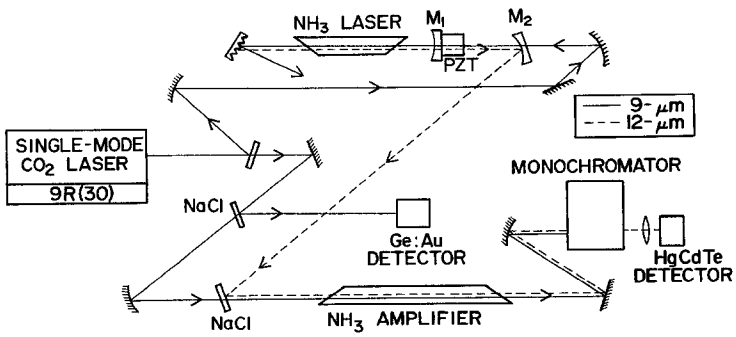

Fig. 1. Schematic diagram of the experimental apparatus for oscillator-amplifier gain measurements in NH_3

and para-NH_3 [10, 14]. Thus, the gain spectrum is completely determined by the vibrational population distribution of the pumped species. In the model the total population is assumed to lie in the ground, first and second excited states of the v_2 vibrational mode. The pump and emission transitions of interest in this paper all occur between the $v_2=0$ and $v_2=1$ levels. A numerical solution of the rate-equation model is used to evaluate the vibrational populations as a function of time for a specific set of pumping conditions. The majority of the molecular constants and relaxation rates used in the rate-equation model are derived from the best values reported in [10]. However, two relaxation rates are treated as variable parameters, as no consistent values have been reported. These variable rates are the rotational relaxation rate of the pumped levels, and the vibration-vibration (V−V) relaxation rate. The V−V relaxation mechanism allows population to be transferred from the $v_2=1$ to $v_2=2$ level in NH_3. The values employed in the present calculations for these two variable rate constants are taken from [11]. A separate program takes the vibrational populations at a given time, together with the NH_3 partial pressure, the total pressure broadening due to a buffer gas, and the rotational temperature, to calculate the gain on the NH_3 transitions of interest. This gain calculation accounts for the splitting of all the vibrational states into symmetric (s) and anti-symmetric (a) levels due to the inversion doubling of the NH_3 molecule. Detailed predictions of the rate-equation model have been validated for gas pressures up to ~100 Torr over a wide range of pump intensities and gas mixtures by the results obtained from previous experiments [10, 11].

3. Results and Discussion

3.1. Small-Signal Gain as a Function of Pressure

The pressure of a gas mixture does not directly affect the small-signal gain at line center on a transition, as long as the transition lineshape is pressure-broadened. For NH_3-N_2 mixtures, the NH_3 transitions are pressure-broadened at pressures greater than 50 Torr. The gas pressure does affect the small-signal gain indirectly through the relaxation rates and their magnitudes relative to the pumping rate. The rotational relaxation rate $1/\tau$ ($=2\pi\Delta\nu$, where $\Delta\nu$ is the pressure-broadened linewidth HWHM) determines the pump saturation intensity $I_s=h\nu_p/2\sigma\tau$, where ν_p is the pump frequency, and σ is the absorption cross-section. In another set of experiments [11], we found that gain was independent of pump intensity for intensities greater than I_s. Thus, for a given NH_3 concentration, the small-signal gain is independent of pressure, provided the pump intensity exceeds I_s. As the pump saturation intensity scales with the square of the pressure, the maximum useful gas pressure is determined by the available pump intensity. A practical upper limit to the pump intensity is defined by the optical breakdown threshold of the gases and materials used in an NH_3 laser. In NH_3-N_2 mixtures that limit is ~1 GW/cm^2 [3], and a similar value holds for crystals of NaCl [15], a common window material. Several manufacturers of high-power reflective optics quote optical damage thresholds of between 0.1 and 1 GW/cm^2. For this study, we will take 100 MW/cm^2 as a conservative upper limit for the pump intensity.

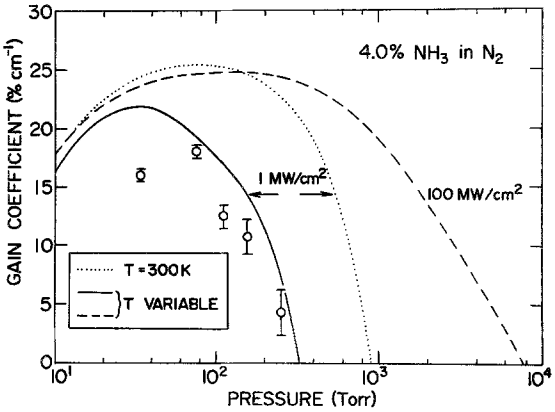

Fig. 2. Comparison between theory and experiment for small-signal gain coefficients of a$P(4,0)$ as a function of pressure in a mixture of 4% NH_3 in N_2. The experimental points were measured for a pump intensity of ~1 MW/cm^2, 0.6 μs after the peak of the pump pulse. The solid curve is calculated with the gas temperature allowed to rise due to the energy absorbed from the pump. The dotted curve is calculated assuming that the gas temperature remains constant at 300 K. Both curves are calculated for the pump-pulse shape used in the experiment. The dashed curve is a variable-temperature calculation for a peak pump intensity of 100 MW/cm^2 in a Gaussian-shaped pulse of duration 100 ns FWHM

We carried out measurements of small-signal gain coefficients as a function of pressure to test our model of the gain dynamics. The CO_2 pump beam was tightly focused into an 11-cm long amplifier cell to produce pump intensities of ~1 MW/cm^2 during the measurement period. The a$P(4, 0)$ transition was chosen for the gain measurements as it generally has the highest gain coefficients. At pressures less than 100 Torr we find that a concentration of 4% NH_3 in N_2 is the optimum mixture for small-signal gain [11]. Thus, a 4% NH_3 mixture was employed for the initial measurements of gain as a function of pressure, and the results are shown in Fig. 2. The gain coefficients are greater than 15% cm^{-1} for pressures up to 80 Torr, but fall rapidly with increasing pressure to ~4% cm^{-1} at 250 Torr.

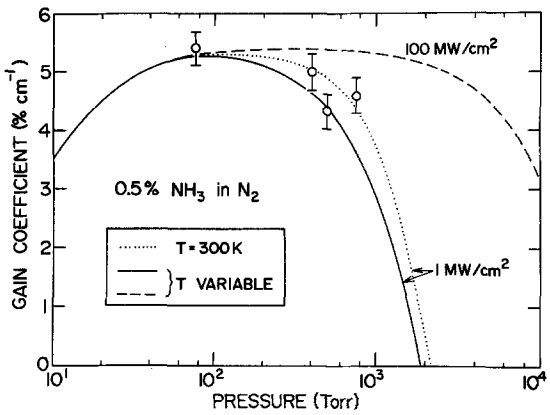

Fig. 3. Comparison between theory and experiment for small-signal gain coefficients of a $P(4,0)$ as a function of pressure in a mixture of 0.5% NH_3 in N_2. See caption of Fig. 2 for full details

Previous experiments show that the gain is reduced in high concentration mixtures by increases in the gas temperature due to pump absorption [11]. Accordingly, the solid curve in Fig. 2 is the pressure dependence of the gain calculated with the ambient gas temperature allowed to increase during the pump pulse [16]. The dotted curve shows the predicted gain coefficients for a constant gas temperature of 300 K. Clearly, the solid curve is in much better agreement with the experimental measurements. At 300 Torr the variable-temperature calculation predicts a temperature rise of ~ 180 K at the time of the gain measurement. The main point illustrated by Fig. 2 is that for a pump intensity of ~ 1 MW/cm^2 in a 4% NH_3 mixture, the maximum useful pressure is ~ 300 Torr. To obtain useful gain coefficients in higher pressures of this mixture will require much higher pumping intensities. A shorter pump pulse of 100 ns duration, rather than the 1 μs pulse used for the experimental measurements, will also help to limit the rise in temperature. The dashed curve in Fig. 2 is a calculation of gain coefficients for a Gaussian-shaped pulse of 100 ns FWHM and peak intensity of 100 MW/cm^2. In the absence of temperature increases, the factor of 100 increase in pump intensity provides a factor of 10 increase in the maximum useful pressure. The calculation for the dashed curve includes temperature changes. Gain coefficients $>5\%$ cm^{-1} are predicted up to 5,000 Torr, but only at the expense of very high gas temperatures. At 5,000 Torr the calculated temperature at the peak of the pump pulse is 570 K, a rise of 270 K. In addition to the reduction in the gain coefficients, a major problem with such a large and rapid temperature increase is distortion of the optical beam if the entire gas is not heated uniformly.

Consequently, we examined mixtures of lower NH_3 concentration where there is less pump absorption and consequently less heating. Figure 3 shows the experimental and predicted results for a mixture of 0.5% NH_3 in N_2. In this case there is very little difference between the constant-temperature and variable-temperature calculations for the pump intensity of ~ 1 MW/cm^2. Both calculations predict gain coefficients in good agreement with the experimental measurements. Although the gain coefficients are generally lower than for the 4% NH_3 mixture, it is possible to maintain significant gains up to much higher pressures. The predicted temperature rise is less than 30 K for all pressures and the gain coefficient remains above 3% cm^{-1} out to 1,000 Torr. Higher pump intensities are required to achieve similar gain coefficients at higher pressures. The dashed curve is a variable-temperature calculation with the Gaussian-shaped 100-ns pulse and a peak intensity of 100 MW/cm^2. At 10^4 Torr the calculated temperature rise at the peak of the pulse is less than 30 K. This temperature rise is insignificant, while the gain coefficient is higher than that predicted at 10^4 Torr from a 4% NH_3 mixture. There should be no need to use mixtures of less than 0.5% NH_3 in practical lasers, unless the rise in temperature must be limited still further. The magnitude of the gain coefficients shown in Fig. 3 indicate that a 10-atm NH_3 laser is feasible. From the calculations in the introduction to this paper, we can predict the fractional tuning range covered by such a laser operating on the P-branch transitions. We estimate that emission from a tunable 10-atm NH_3 laser will cover $\sim 36\%$ of the wavelength range from 10.5 to 13.8 μm.

Synchronous-pumping with a train of short mode-locked pulses is another technique which may be successful in achieving laser operation at pressures greater than 1 atm. As the V–T relaxation rate is very fast at such high pressures it is necessary to have sufficient pump intensity in each pulse to produce gain; there can be no buildup of gain by a succession of pulses. Very high gain coefficients would be necessary to achieve a rapid buildup of an NH_3 laser pulse and minimize the number of pump pulses required. Calculations for a 4% mixture at 5,000 Torr and a Gaussian-shaped pump pulse of 1 ns FWHM in duration indicate that a peak intensity of 100 MW/cm^2 is insufficient to create gain. For a peak intensity of 1 GW/cm^2 the model predicts a peak gain coefficient of 22% cm^{-1} with gain lasting for more than 2 ns. The model also predicts a temperature rise of ~ 12 K. The temperature rise is still a problem as the heat could not be extracted during a train of mode-locked pump pulses, typically 10–20 in number.

All of the previous discussion has concerned pumping NH_3 with the $9R(30)$ line of CO_2. The $9R(16)$ line is the only other conventional CO_2 laser line which is suitable for pumping high-pressure NH_3 lasers to

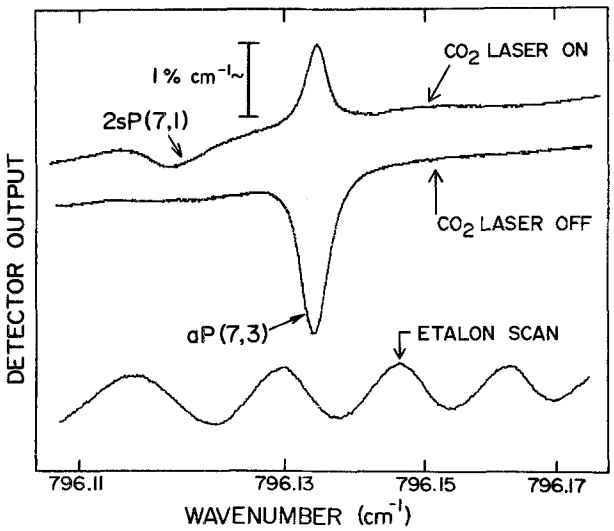

Fig. 4. High-resolution frequency scan of the region near a$P(7,3)$ taken with a tunable diode laser probing a CO_2 laser-pumped sample cell containing 1% NH_3 in N_2 at a total pressure of 5 Torr. The middle trace shows that there is only absorption on a$P(7,3)$ when the CO_2 laser is off. The upper trace shows that gain is produced on a$P(7,3)$ when the CO_2 laser is on, and simultaneously an absorption feature, identified as 2s$P(7,1)$, appears 460 MHz away. The bottom trace is a frequency calibration taken with an etalon having a free spectral range of 0.01627 cm^{-1}

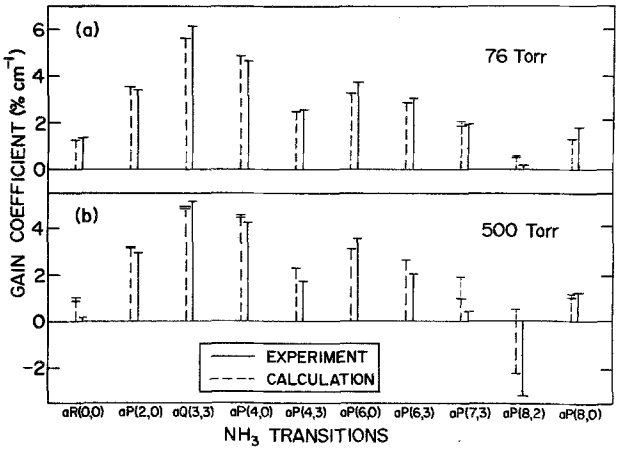

Fig. 5. Comparison between measured and calculated small-signal gain coefficients for a range of transitions in a mixture of 0.46% NH_3 with N_2. The experimental values were obtained at a pump intensity of ~1 MW/cm^2, 0.6 μs after the peak of the pump pulse. Two sets of calculations are displayed. The thick (thin) cross bars indicate the calculations which include (exclude) all the known overlapping gain and absorption contributions due to v_2 and $2v_2$ NH_3 transitions within ±30 GHz (=1.0 cm^{-1}) of each transition listed. (At 500 Torr, 30 GHz corresponds to ~18 half-widths)

obtain wide-range line-tunability. This CO_2 line is offset by 1.35 GHz from a$R(6,0)$ in NH_3, and by 4.94 GHz from a$R(6,1)$ [17]. An advantage of pumping with 9$R(16)$, instead of with 9$R(30)$, is that it provides greater pulse power from any given CO_2 laser. The main disadvantage is the much larger pump offset of the para-NH_3 absorption, i.e. a$R(6,1)$. Using the 9$R(16)$ line to pump NH_3-N_2 mixtures up to ~100 Torr has produced line-tunable emission only on ortho-NH_3 transitions [10]. Efficient pumping of para-NH_3 with 9$R(16)$ requires NH_3-N_2 mixtures at pressures greater than 4 atm, but for such pressures pumping with 9$R(16)$ should be comparable to pumping with 9$R(30)$.

3.2. *Overlapping Absorptions*

A situation that occurs in many applications of molecular gases is an overlap in frequency of transitions in one band by transitions in another band, usually in the same mode. In most optically-pumped NH_3 lasers, only one vibrational level is excited, the $v_2=1$ level. Thus, the selective pumping increases the absorption on all transitions whose lower level is the $v_2=1$ level, such as the $2v_2$ band transitions which overlap the v_2 band transitions. Any $2v_2$ transitions which lie close in frequency to v_2 transitions will tend to reduce the gain on those v_2 transitions. Pressure broadening will increase the interference, and tends to reduce tunability of the laser emission rather than increase it. High-resolution spectroscopy is a valuable aid for revealing transitions where overlaps occur. Figure 4 displays frequency scans taken with a tunable diode laser (TDL) probing the region near the a$P(7,3)$ transition. A sweep-integration technique was employed [18, 19], and the scans shown are an average of four sweeps. The TDL was used to probe a cell containing an NH_3 mixture which could be pumped by a cw CO_2 laser. With the CO_2 laser off there is only absorption on the aP(7,3) transition. Turning the CO_2 laser on creates gain on a$P(7,3)$ and simultaneously creates absorption on 2s$P(7,1)$, 460 MHz away from a$P(7,3)$. At pressures over 100 Torr, absorption on the 2s$P(7,1)$ transition will significantly reduce the gain coefficient at the line center of a$P(7,3)$.

The available spectroscopic data are not sufficiently accurate to evaluate the overlapping absorptions for all the v_2 transitions [20–22]. A more direct method is to measure the gain coefficients as a function of pressure for the v_2 transitions of interest. Measurements of gain coefficients at 76 and 500 Torr are displayed in Fig. 5 for a range of v_2 transitions. As predicted from Fig. 4, the a$P(7,3)$ gain coefficient is much reduced at the higher pressure. The gain coeffi-

cient for one other transition, a$P(8,2)$, changes from a small positive value at 76 Torr to a large negative value at 500 Torr. This dramatic change is due to interference by 2a$R(6,3)$, a very strong absorption ~ 1.7 GHz away. This result is particularly interesting as the *unpumped* absorption on a$P(8,2)$ in 500 Torr was one-third of the absorption in the presence of the pump. The large increase in absorption with pumping supports our conclusion that a $2\nu_2$ absorption is the source of interference. Most of the other transitions show much less change in their gain coefficients [23]. In some cases, the contributions due to overlapping inverted ν_2 transitions increase the gain coefficients. All other overlapping bands, such as $\nu_4+\nu_2$ to ν_2, were neglected. The overall agreement between theory and experiment is very good for both pressures despite the uncertainties of frequency and linestrength for the $2\nu_2$ transitions. Thus we are confident that several transitions will remain useful up to a few atmospheres of pressure.

4. Conclusions

In this paper we have examined the potential for operating line-tunable NH_3 lasers at pressures of several atmospheres. The motivation for developing high-pressure NH_3 laser operation is to increase the tunability of the emission and produce subnanosecond pulses. Experimental measurements of small-signal gain coefficients illustrate that NH_3 concentrations of 0.5% or less must be employed to minimize problems of gas heating. Theoretical calculations indicate that a pump intensity of 100 MW/cm^2, a reasonable upper limit for preventing optical breakdown in the laser, creates sufficient gain for laser action in a 0.5% NH_3 in N_2 mixture up to 10 atm (7,600 Torr). The maximum continuous tuning range at a pressure of 10 atm is estimated to be ~ 120 GHz (4 cm^{-1}), which is sufficient gain bandwidth to support pulses less than 10 ps in duration. Most transitions will give 30–50 GHz tunability at 10 atm. Obvious exceptions are the a$P(7,3)$ and a$P(8,2)$ transitions, which have their gain coefficients drastically reduced due to overlapping absorption from $2\nu_2$ band transitions. Measurements indicate that many interference-free transitions do exist, but there remains a need for high-resolution spectroscopy to help identify such transitions. If we neglect overlapping absorption, then our calculations predict that a 10-atm NH_3 laser operating on the P-branch transitions will be tunable across $\sim 36\%$ of the wavelength range from 10.5 to 13.8 μm.

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