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Power Enhancement of the cw 12.08 μm NH_3 Raman Laser with the Addition of H_2/He Buffer Gases*

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Abstract. The effects of H_2, He, and N_2 buffer gases on the efficiency of the cw 12.08 μm NH_3 Raman laser are studied experimentally. The laser output power is increased by nearly 60% with the addition of H_2 or He, which we essentially attribute to the high thermal conductivity of these buffer gases. In the optimum conditions (NH_3/H_2 : 1/1 mixture with 0.35 Torr partial pressure of NH_3) 3.3 W output power at 12.08 μm is obtained which corresponds to 11% power conversion efficiency.

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The optimization of the CO_2-pumped 12.08 μm NH_3 laser appears to be a determinant step for the future development of powerful continuous-wave sources in the 12–20 μm medium infrared (mir) region [1]. Since the first results reported by Rolland et al. [2], one order-of-magnitude increase in output power and efficiency has been recently obtained through a simultaneous improvement of the mir cavity configuration and of the pump intensity inside the resonator [3]. In this paper, we report on a new progress in the 12.08 μm NH_3 laser efficiency by the addition of buffer gases.

At a given pump power, the power conversion efficiency is presently increased by nearly 60% with the addition of H_2 or He. In the optimum conditions of mixture and pressure we obtain up to 3.3 W output power at 12.08 μm for 30 W 9.22 μm pump power injected into the mir cavity. This is the highest power reported to date for a cw ir laser-pumped molecular laser. In contrast with pulsed experiments [4] the addition of N_2 is always found to be detrimental to the laser efficiency.

The experimental results are well qualitatively interpreted from the theoretical expressions of the local mir gain and pump absorption. Discussion of the different influences of buffer molecules in the cw 12.08 μm laser pumping cycle emphasizes the beneficial effect of light molecules having a large thermal conductivity.

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1. Experiments

1.1. Optical Scheme

The pumping set-up is shown on Fig. 1. The basic elements of the mir cavity have been recently described [3]. The 1 m long mir ring resonator is composed of two gold-coated mirrors and of a dichroic ZnSe output coupler. The whole system is constructed inside a vacuum box. 90% of the 34 W CO_2 pump beam is coupled into the box through a ZnSe plate with antireflecting coating and is closely matched to the 12 μm cavity mode (~3 mm average beam spot size). The same ZnSe plate is also used for the output coupling of the mir and residual CO_2 radiations. The 9.22 and 12.08 μm output beams are separately detected after being diffracted by a 133 l/mm gold-coated grating. The total external losses at 12 μm including transmission of the ZnSe window and 1st order grating efficiency have been estimated to be ~10%.

The main advantages of this new mir cavity configuration with internal optics result both from its

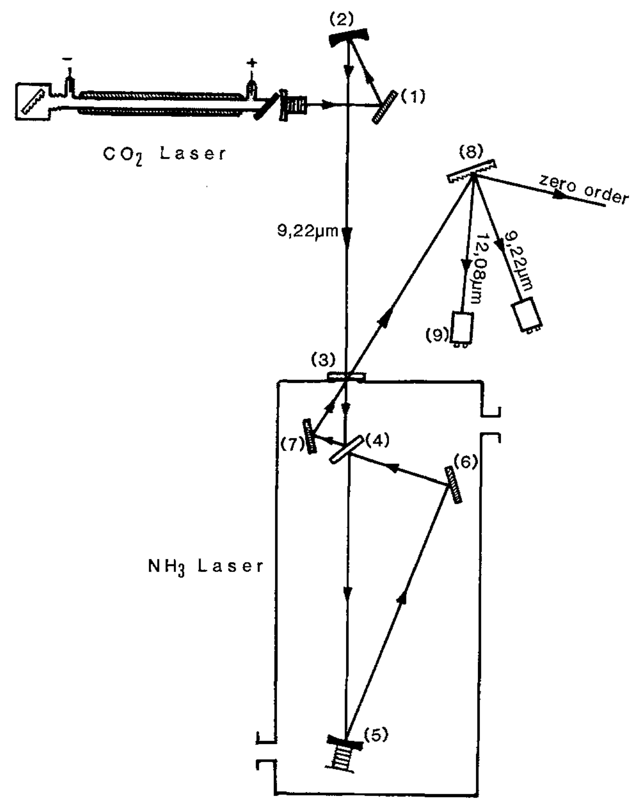

Fig. 1. Optical pumping set-up: *1, 2:* injection mirrors; *3:* ZnSe window; *4:* dichroic ZnSe mirror; *5–7:* gold-coated mirrors; *8:* diffraction grating; *9:* radiation detectors in the first order of the grating *8*

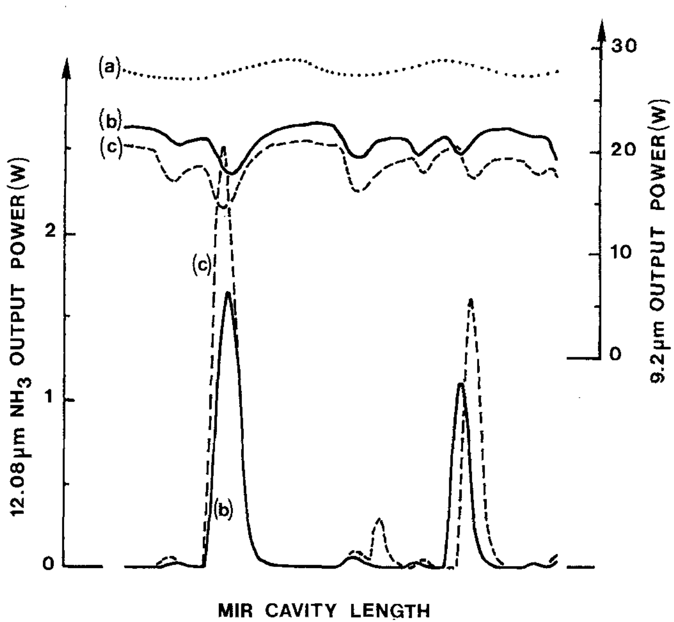

Fig. 2. Simultaneous recordings of the 9.2 µm output signal (top curves) and of the 12.8 µm output power (bottom curves) as functions of the mir resonator length, showing the periodic resonances at these two wavelengths. Curves *(a–c)*, repectively, correspond to $p_{NH_3}=0$, $p_{NH_3}=330$ m Torr, $p_{NH_3-He}=660$ m Torr for a NH_3–He:1/1 mixture

versatility and from the absence of Brewster windows along the active optical path.

For a correct analysis of the pressure-dependence of the laser gain, care has been taken to avoid the mir cavity resonances at the pump wavelength[1]. The angle of the incident beam onto the dichroic mirror has been choosen for maximum transmission at 9.22 µm ($\gtrsim 95\%$). Under this condition, the coupling coefficient at 12.08 µm has been measured to be about 7%. This value is not too far from the optimum value which is theoretically predicted for 250 W/cm^2 pump intensity [1].

1.2. Experimental Results

Typical interferograms of the mir cavity at the pump and mir frequencies are shown on Fig. 2. In all the cases, the 9*R*30 CO_2 laser is tuned for maximum power, i.e. close to its own emission line center, which corresponds to the best efficiency of the 12.08 µm NH_3 Raman laser [3]. The simultaneous recordings of the 9.2 µm output (top curves) and of the 12.08 µm output (bottom curves) are reported, respectively, for $p_{NH_3}=0$ (a), $p_{NH_3}=330$ mTorr (b), and for a NH_3/He : 1/1 mixture at 660 mTorr total pressure (c). It may be emphasized from Curve (*a*) that the mir cavity just exhibits weak resonances at the pump frequency. These weak resonances still slightly affect the peak intensities of the 12.08 µm output (TEM_{00} mode); the most intense mir peaks of Curves (*b*) and (*c*) correspond to the maximum pump energy stored in the mir cavity. The comparison between the three CO_2 curves reveals the contribution of the one-photon and two-photon processes to the pump absorption. The $\sim 18\%$ absorption observed in the absence of 12 µm lasing (Curve *b* for pure NH_3) is in good agreement with the measurement reported in [5]. This value is slightly lower than the calculated one [1], which may be attributed in part to the presence of thermal effects in the laser medium (Sect. 2).

The beneficial effect of He on the NH_3 laser efficiency is already illustrated from Curve (*c*). The detailed experimental study of the H_2, He, N_2 buffer gas effects has been performed in the following conditions:

i) The buffer gas is added at a constant partial pressure of NH_3.

[1] Even for a moderate ir reflectivity of the dichroic mirror ($\gtrsim 20\%$), the interferogram of the mir cavity reveals the presence of sharp 12.08 µm maxima when the resonance condition is simultaneously satisfied at the pump and mir wavelengths. Since the refractive index of NH_3 at these two wavelengths changes differently with pressure, the double-resonance condition cannot be maintained for a given TEM_{00} mode at any pressure. In these conditions, the evolution of the mode intensity does not reflect correctly the pressure-dependence of the laser gain [3]

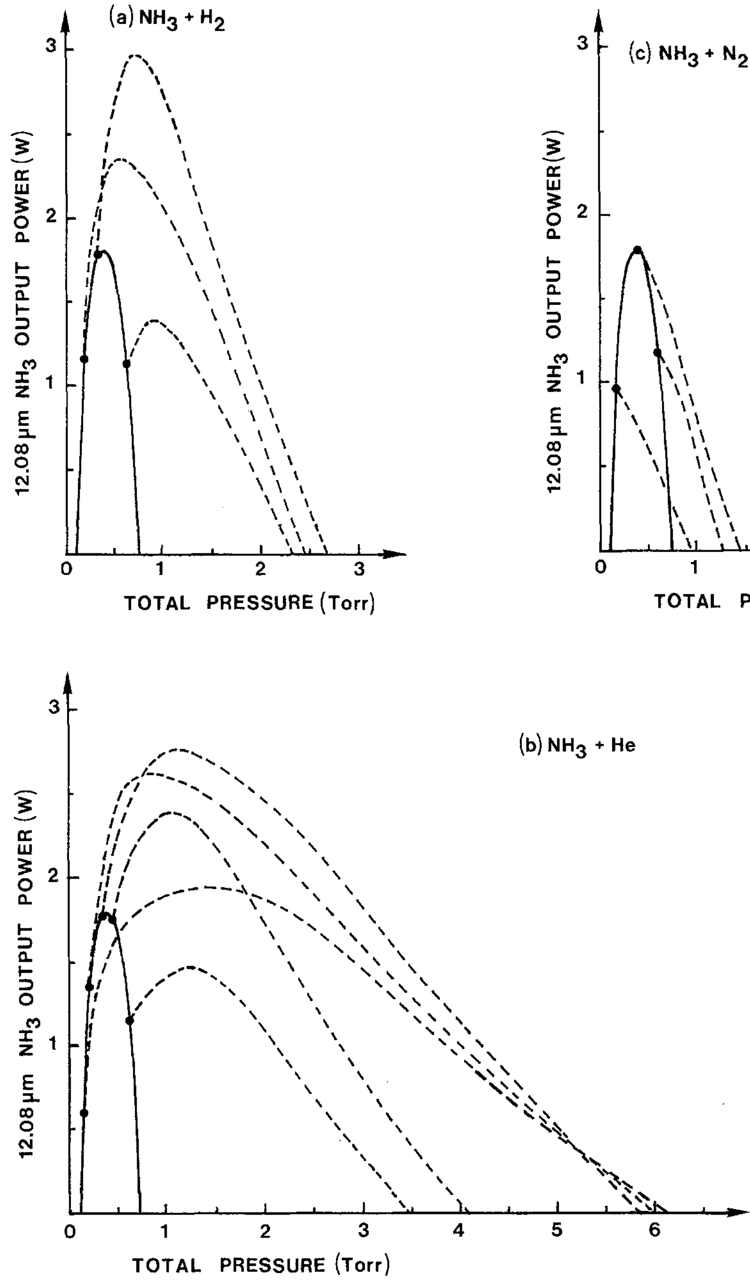

Fig. 3a–c. 12.08 μm output power as a function of the total gas pressure. In each figure, the solid curve represents the pure NH_3 case. The dashed curves show the effect of adding H_2 (a), He (b), N_2 (c) to a constant partial pressure of NH_3

ii) The NH_3/buffer gas laser is operated under sealed-off conditions.
iii) As in Fig. 2, the CO_2 laser is tuned for maximum power (~34 W).
iv) The mir cavity length is adjusted for maximum 12.08 μm power at each pressure (main peak of the interferogram of Fig. 2).

The experimental results are reported in Fig. 3a–c, respectively, for H_2, He, N_2. The 12.08 μm laser output power is depicted as a function of the total pressure measured with an absolute capacitive gauge. The solid curves in Fig. 3 represent the pure NH_3 case. The dashed curves show the effect of adding the buffer gas on the 12.08 μm output for different partial pressures of NH_3.

With pure NH_3, the operating pressures scale from 0.1 Torr up to 0.75 Torr and the maximum output power (1.75 W) is obtained at 0.4 Torr. The wide range of operating pressures of the 12.08 μm NH_3 Raman laser has been explained by the insensitivity of the Raman gain to thermalizing collisions [1, 3].

The addition of light buffer molecules (H_2, He) is found to be beneficial to the laser efficiency at any partial pressure of NH_3. The best power results (3 W in Fig. 3a, 2.8 W in Fig. 3b) are, respectively, obtained for NH_3/H_2 : 1/1 and NH_3/He : 1/2 mixtures with

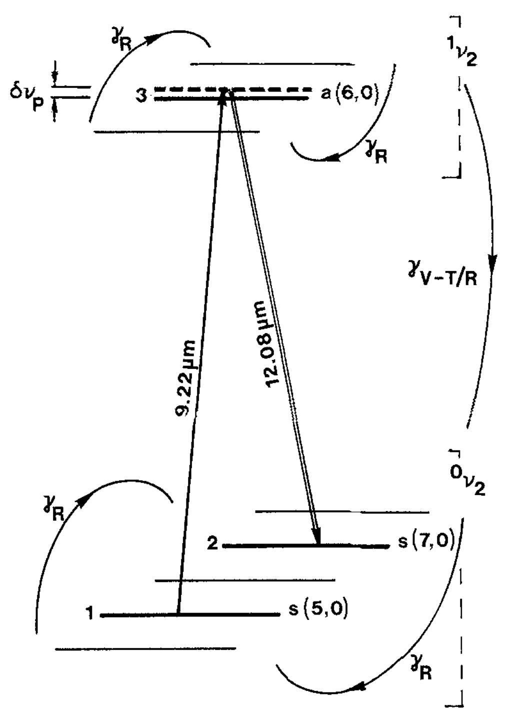

Fig. 4. Partial energy-level diagram for the 12.08 μm NH_3 Raman laser showing pump and mir transitions

0.35 Torr partial pressure of NH_3. It must be noted here that these results are not corrected for the mir losses due to the optical elements between the mir cavity output and the detector (Sect. 1.1). Taking them into account, the maximum 12.08 μm power available at the dichronic-mirror output has been evaluated to be 3.3 W which corresponds to 11% power conversion efficiency.

In contrast, the cw 12.08 μm output is always dramatically decreased when adding N_2 (Fig. 3c). This behavior has been observed for any buffer gas heavier than NH_3 (for instance, Ar and buffer gases used in [6, 7]).

2. Discussion

The basic pumping scheme of the 12.08 μm NH_3 laser is shown in Fig. 4. CO_2 pump and mir emission, respectively, correspond to the $sR(5,0)$ and $sP(7,0)$ rovibrational transitions of the same ν_2-fundamental band of NH_3 [8]. Pumping with the $9R30$ CO_2 line occurs 185 MHz off-resonance ($\sim$4 Doppler widths) and the gain arises from two-photon processes [1, 2]. The vibrational relaxation of the ν_2-state to the vibrational ground state results from $V-T/R$ processes. The corresponding relaxation rate, $\gamma_{V-T/R}$, is comparable to the rotational one [9]. The high value of $\gamma_{V-T/R}$ ($\gtrsim 10^6\,s^{-1}\,Torr^{-1}$), the off-resonant nature of the pump/mir fields and the moderate pump intensities of the cw regime ($I_P, I_{mir} \lesssim 500\,W/cm^2$) simultaneously concur to a very weak saturation of the one-photon absorptions (i.e., weak vibrational bottleneck [2]). Based on this consideration, a two-wave three-level quantomechanical model has been recently developed for the 12.08 μm NH_3 laser [1]. The simplified analytical expressions of the local mir gain and pump absorption are recalled in the Appendix and are presently used for a qualitative analysis of the effects of buffer gases in the laser pumping cycle.

Buffer molecules may essentially influence the laser efficiency in two ways:

i) By changing the rotational lifetimes of levels 1, 2, 3 (labels of Fig. 4). The addition of any buffer gas to the system invariably increases the homogeneous linewidths of the ir/mir transitions;

$$\Delta\nu_H^0 \to \Delta\nu_H^0(1 + x_B \cdot p_B/p_{NH_3}),$$

where $\Delta\nu_H^0$ is the homogeneous linewidth for pure ammonia, p_B and p_{NH_3} are the partial pressures and x_B is the relative contribution of the buffer gas to the homogeneous linewidth.

ii) By changing the ground state populations (i.e., by changing the translational/rotational temperature of the active molecules). The influence on the two-photon gain and on the one-photon mir/pump absorptions, respectively, occurs via the terms $(f_1^e - f_2^e)/\Delta\omega_D$, $\sim f_2^e\Delta\nu_H$ and $\sim f_1^e\Delta\nu_H$ (appendix). All these terms decrease with gas temperature by approximately the same amount (for instance, α_R is decreased by $\sim$1.4 when T increases from 293 to 393 K). The ground state excitation of active molecules has two origins. First it results from $V-T/R$ transfers and is indirectly related to the one-photon pump/mir absorptions. Secondly, it is due to the Raman conversion itself, which directly populates the lower level of the mir transition. Because of the high Raman conversion efficiency in NH_3 this latter excitation is not negligible.

The effect (i) is always negative since the one-photon absorption coefficients are proportional to $\Delta\nu_H$ and the two-photon gain is insensitive to thermalizing collisions (appendix). The decrease of the average pump intensity along the mir resonator and the increase mir self-absorption both tend to decrease the laser efficiency.

The effect (ii) may be negative or positive. A good buffer molecule must be capable to deactivate efficiently the NH_3 molecules and to transport rapidly the excess

[2] Neglecting $V-V$ transfers to higher excited vibrational states, the ν_2-state fractional population, f_{ν_2}, is given by $\alpha_P I_P/(N \cdot h\nu_P \cdot \gamma_{V-T/R})$, where α_P is the pump absorption coefficient and N is the density of active molecules. Taking α_P from [1] and $\gamma_{V-T/R}$ from [9], we obtain $f_{\nu_2} = 0.73\%$ for $I_P = 300\,W/cm^2$

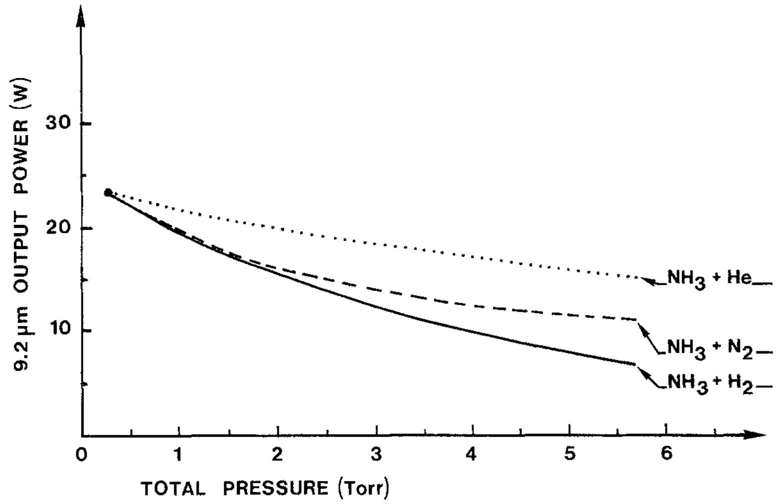

Fig. 5. 9.2 μm signal at the NH_3 cell output as a function of the total pressure. The experimental procedure is the same as in Fig. 3 except that the dichroic mirror is removed from the mir cavity. The buffer gases (respectively, He, N_2, H_2) are added at a constant 330 mTorr partial pressure of NH_3. The round dot (starting point) corresponds to the absorption of pure NH_3

energy to the resonator walls. Let us consider separately two different cases of buffer molecules:

Simple molecules (monoatomic or diatomic gases) play a minor role in the direct vibrational deactivation of NH_3 by $V-T/R$ transfers [9]. They rather contribute to the direct ground state deexcitation. Thus, their influence may be simply interpreted in term of thermal conductivity. Only the molecules lighter than NH_3 (i.e., with a larger thermal conductivity [10]) can have a positive effect. This explains the good results obtained for H_2 and He (Fig. 3a and b) and the detrimental effect of N_2 (Fig. 3c). The high performances reported previously for the NH_3/N_2 laser in pulsed operation [4] may be attributed to the different nature of the lasing processes in this regime.[3]

Complex non-polar molecules having a large number of vibrational modes (i.e., with large vibrational heat content) may a priori contribute more efficiently to the direct vibrational deactivation of NH_3 molecules. As for cw fir optically pumped lasers [6, 7], a positive influence could be obtained provided that; – they remove the vibrational energy from the active molecules faster than do the $V-T/R$ transfers of NH_3, – they are sufficiently mobile to transport the energy to the walls. However these conditions cannot be easily satisfied in NH_3. Moreover, these complex molecules are also probably less efficient to remove the excess rotational ground state energy caused by the Raman conversion. This explains our fruitless tentatives with such buffer molecules.

[3] Both due to the much higher pump intensities ($\gtrsim 1\,MW/cm^2$) and to the higher operating pressures (up to 50 Torr), the 12 μm emissions are rather based on population inversion mechanisms. Resonant transfers between rotational levels of NH_3 and N_2 can strongly enhance the population inversion between the lasing levels and thus, the laser efficiency

The pressure evolutions reported in Fig. 3 for simple buffer molecules can be readily interpreted from the discussion above. The increased 12.08 μm output with H_2/He partial pressures (Curves 3a, b) reflects the increase of the thermal conductivity with pressure, the gas temperature approaching better the ambient value. In contrast, the decreasing output with pressure is associated with the decrease of the rotational lifetimes due to collisions.

The higher thermal conductivity of H_2 as compared to He [10] explains the better efficiency of this buffer gas at the optimum. However, the direct rotational deactivation of NH_3 is more important for molecular gases than for atomic ones [11], which explains the smaller range of operating pressures with the H_2–NH_3 mixture. The cut-off pressures for the NH_3–He and NH_3–H_2 mixtures can be quantitatively evaluated from (A. 1–3). We assume that at these points the gas temperature is close to the ambient value. Typical values for the homogeneous linewidths are taken from [11]. The cut-off pressures calculated at 330 mTorr partial pressure of NH_3 and 250 W/cm^2 injected pump intensity are, respectively, $p_1 = 3.5$ Torr and $p_2 = 8.1$ Torr for the NH_3–H_2 and NH_3–He mixtures.

These values are found to be in relatively good agreement with the experimental ones reported in Fig. 3. In order to bring a second confirmation of the interpretations above, separate measurements of the one-photon ir absorption have been carried out in the presence of buffer gas. The procedure was the same as in Fig. 3. The dichroic mirror was removed for these experiments. The experimental results are reported in Fig. 5 for H_2, He, N_2 with 330 mTorr partial pressure of NH_3. At 5.5 Torr total pressure, the absorption with the H_2-mixture is found to be 2.5 times higher than with the He-mixture. This gives evidence of the high efficiency of H_2 in the direct rotational deactivation of the NH_3 molecules. On the other hand, the weaker absorption of the NH_3–N_2 mixture compared to that of the NH_3–H_2 mixture still reflects the different thermal conductivities of the two diatomic gases.

The nearly 60% increase of the NH_3 laser efficiency with light buffer gases gives account of the importance of thermal effects in this new cw optically pumped laser. This is likely related to the high ir/mir intensities circulating inside the mir cavity ($\gtrsim 200\,\mathrm{W/cm^2}$). For instance, the field intensities are typically twenty times larger than in the case of cw fir lasers. The presence of strong thermal effects also suggests that further increase in the laser efficiency should be obtained by using mir resonators with small transverse sizes. However this assumes that all the advantages of the mir ring resonator described in Sect. 1 could be kept. The present geometry eliminates feedback to the pump laser [2], takes advantage of the strong mir gain anisotropy, reduces spatial hole-burning effects and enables us to change easily the parameters for output power optimization (active medium length, ir/mir reflectivities of the output coupler).

Besides, an increase of the Raman conversion efficiency will increase, in turn, the ground-state excitation of the active molecules. Both due to this counterbalancing effect and to the transversally non-uniform pump excitation, the addition of light buffer molecules is expected to bring about similar improvements in any configuration of the mir resonator.

3. Conclusion

In conclusion, we have reported on a $\sim 60\%$ increase in the 12.08 μm NH_3 Raman laser efficiency by addition of He–H_2 buffer gases. The 11% power conversion efficiency obtained with the NH_3/H_2 : 1/1 mixture at a total pressure of 0.7 Torr confirms the high performances predicted for this laser [1]. Experimental investigations and a qualitative analysis of the influence of buffer gases in the laser pumping cycle show that the beneficial effect is essentially to remove the excess rotational/translational energy from the active molecules in the ground state.

The results of the study are directly applicable to other potential ir-laser pumped mir transitions in NH_3 and in its isotopic varieties. The simultaneous use of He/H_2 buffer molecules and of mir resonators with small transverse sizes is promised to be a determinant step for the development of this new generation of cw infrared lasers.

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Appendix

In the perturbation limit, the mir gain be expressed as [1]

$$g_{\mathrm{mir}} \simeq N\sigma_{32}\left[\frac{\left(f_3^e - \frac{g_3}{g_2} f_2^e\right)\gamma_{32}}{\delta\omega_P^2} + b_2 \frac{\bar{\Lambda}_P^2}{\delta\omega_P^2}\left(f_1^e - \frac{g_1}{g_2}\cdot f_2^e\right) F\right], \quad \text{(A.1)}$$

where f_i^e $(i=1,2,3)$ are the equilibrium fractional populations [labels 1, 2, 3 are referred to the m-sublevels of levels $s(5,0)$, $s(7,0)$, and $a(6,0)$, respectively], N is the density of active molecules, σ_{32} is the m-average integrated mir cross-section, $\bar{\Lambda}_P$ is the m-average absorption Rabi frequency, γ_{32} is the one-photon polarization decay rate related to mir, $\delta\omega_P$ is the pump frequency detuning, $g_K = 2J_K + 1$, $b_2 = 2/5$ and F is the real part of a complex error function [1]. At the exact Raman resonance ($\delta\omega_P = \delta\omega_{\mathrm{mir}}$, i.e. maximum laser efficiency), the factor F is well approximated by $\sim \sqrt{\pi}\,\lambda_P/\Delta\omega_D(\lambda_P - \lambda_{\mathrm{mir}})$ where $\Delta\omega_D$ is the mir Doppler linewidth.

The first term in the brackets of (A.1) is related to one-photon mir absorption, whereas the second one corresponds to the two-photon gain. Equation (A.1) may be rewritten in condensed form

$$g_{\mathrm{mir}} = -\alpha_{\mathrm{mir}} + \alpha_R \cdot I_P . \quad \text{(A.1')}$$

The detailed expression for the pump absorption is similar to expression (A.1). The condensed form may be also written

$$\alpha_P = \alpha'_P + \alpha_R \cdot I_{\mathrm{mir}} = -1/I_P \cdot dI_P/dz , \quad \text{(A.2)}$$

where $\alpha'_P = -N\sigma_{31}\left(f_3^e - \frac{g_3}{g_1}\cdot f_1^e\right)\gamma_{31}/\delta\omega_P^2$ is the one-photon pump absorption.

α_{mir}, α'_P, α_R have been calculated in [1] from spectroscopic and relaxational data given in the litterature. The γ_{32}, γ_{31} terms are deduced from the ir/mir homogeneous linewidths $\Delta\nu_H = 18$ MHz/Torr. At 293 K, $\alpha_{\mathrm{mir}} = 0.57\,\mathrm{m^{-1}\,Torr^{-2}}$, $\alpha'_P = 2.42\,\mathrm{m^{-1}\,Torr^{-2}}$ and $\alpha_r = 1.58 \times 10^{-22}\,\mathrm{Torr^{-1}\,cm^2\,s\,photon^{-1}}$.

The cut-off pressures for the NH_3/H_2 and NH_3/He mixtures may be evaluated from (A.1, 2) as following. We assume that the gas temperature is close to the ambient value and the presence of buffer gas is simply introduced in α'_P and α'_{mir} via the homogeneous ir/mir linewidths. Thus, we may write: $\alpha_R = \alpha_R^0$, $\alpha_{\mathrm{mir}} = \alpha_{\mathrm{mir}}^0(1 + x_B \cdot p_B/p_{NH_3})$, and $\alpha'_P = \alpha_P'^0(1 + x_B \cdot p_B/p_{NH_3})$ where the zero index refers to the values in pure ammonia ($p_B = 0$) given above.

The average contribution of the buffer gas to homogeneous linewidth is deduced from [11].

At the cut-off pressure, the threshold condition may be written.

$$L(-\alpha_{\mathrm{mir}} + \alpha_R \cdot \bar{I}_P) = -\mathrm{Log}\,R . \quad \text{(A.3)}$$

In (A.3), L is the active medium length, $\bar{I}_P$ is the average circulating pump intensity and R synthesizes all the mir cavity losses at the mir wavelength (presently, $R \simeq 85\%$). The average pump intensity may be deduced from (A.2) for $I_{\mathrm{mir}} = 0$; $\bar{I}_P = \xi I(0)\cdot[1 - \exp(-\alpha'_P L)]/\alpha'_P L$, where the loss term ξ includes dichroic-mirror transmission, pump beam expansion and losses due to the other cavity mirrors (presently, $\xi = 87\%$) and $I(0)$ is the injected pump intensity.

As a first remark, it is shown that, for a given partial pressure of NH_3 and a given pump intensity, the partial pressures of H_2 and He at cut-off are simply related by: $p_{H_2}/p_{He} = x_{He}/x_{H_2}$. From [11] the latter ratio is 0.43, which well agrees with the results of Fig. 3.

A direct evaluation of p_{He} and p_{H_2} made for $p_{NH_3} = 330$ mTorr and $I_P(0) = 250\,\mathrm{W/cm^2}$ gives, respectively: $p_{He} = 8.1$ Torr and $p_{H_2} = 3.5$ Torr. The relatively small discrepancies with the experimental results (Fig. 3) may be attributed both to the assumptions made above and to the uncertainties in the measurements (influence of residual CO_2 resonances of the mir cavity).

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