

On the possibility of development of a photochemical unit based on an NH₃ laser with an intracavity reactor

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Abstract. The possibility of developing a photochemical setup on the basis of an optically pumped ammonia laser with an intracavity photoreactor is proved. The obtained values of the cavity ‘implication’ factor γ are comparable with those of intracavity systems based on a CO₂ laser. The conditions for achieving the maximum energy in the focusing cavity are determined and the ways to control the shape of its caustic are indicated.

Keywords: NH₃ laser, ammonia, cavity, reactor, energy.

1. Introduction

An NH₃ laser pumped by high-power pulses from a TEA CO₂ laser is one of the most efficient and powerful pulsed lasers emitting in the mid-IR range (11–13 μm). Lasing in ammonia molecules was first obtained in [1]. New working mixtures were found in subsequent investigations; in particular, the decisive influence of nitrogen as a buffer gas on the energy, efficiency and tuning range of the NH₃ laser was proved in [2–5]. The efficient optical schemes have been developed for pumping and lasing [3, 6–9], and a repetitively pulsed regime has been realised both in a nonselective [4] and a selective [10] cavity. The characteristics of the developed NH₃ lasers became close to those of gas-discharge molecular lasers (like CO₂ lasers) in the mid-IR range, the output pulse energy amounting to 1.5 J for a pump efficiency up to 20 % [8].

Along with a broad tuning range (740–890 cm^{-1} [8]) and a high average output power in the repetitively pulsed regime (tens of watts [4, 10]), such parameters have initiated research on a number of practical applications of NH₃ lasers, in particular, in the field of laser isotope separation (LIS). For example, isotope-selective IR multiphoton dissociation (IR MPD) of CCl₄ produced by NH₃ laser

radiation at a frequency of 780.5 cm^{-1} , which is close to the ν_3 mode frequency of the CCl₄ molecule (775 cm^{-1}), was observed in [11]. In this case, the dissociation yield was much higher than in the case of dissociation of CCl₄ by a CO₂ laser pulse [12] upon excitation via the combination band $\nu_1 + \nu_2 + \nu_4$. NH₃ lasers were also used in experiments on LIS of selenium upon dissociation of SeF₆ molecules [13]. The possibility of hydrogen isotope separation (H/D/T) using NH₃ laser radiation was demonstrated for CHCl₃ [14, 15], CHF₃ [16], and CF₃CHBrF [17] molecules. The MPD selectivity $\alpha(\text{D/T}) > 15000$ was achieved in [15].

The efficient use of laser radiation is one of the main tasks facing the practical realisation of any LIS process. For example, in the case of LIS of carbon by IR MPD of CF₂HCl molecules by CO₂ laser radiation, this problem was solved by placing a photochemical reactor inside the laser cavity [18]. The intracavity irradiation scheme allows the formation of extended caustics with a high fluence of laser radiation (tens of J cm^{-2} and higher) and provides a high efficiency of using laser radiation for a reasonable length (~ 1 m) of the interaction region. The high efficiency of the LIS process for carbon with the use of an intracavity photochemical reactor was demonstrated in [18–20]. This method was used in the commercial LIS technology for separation of carbon isotopes [21].

The aim of this paper is to analyse the possibility of using the intracavity irradiation scheme in an NH₃ laser and to determine the optimal conditions for its realisation.

2. Experimental setup

An NH₃ laser is usually pumped by the 9R16 or 9R30 line of the CO₂ laser with frequencies close to those of the aR(6, k) and sR(5, k) transitions, respectively, of ammonia molecules [8]. It was shown in [2–5] that in the latter case, the NH₃ laser efficiency is higher and the laser can be tuned in a broad range. For this reason, we pumped ammonia by the 9R30 line at 9.22 μm . A pulsed CO₂ TEA laser based on the CO₂ : N₂ : He = 1 : 1 : 4 mixture at a total pressure up to 650 Torr and tuned to the 9R30 line was used for pumping. The pump pulse had the standard shape and consisted of a peak of the FWHM of ~ 90 ns and a tail of duration ~ 1.6 μs . The output energy was 2.3–2.6 J, the tail containing less than 50 % of the total pulse energy. The NH₃ laser cell was made of a stainless steel tube of length 180 or 100 cm. The radiation parameters of the NH₃ laser were studied at the frequencies 828 cm^{-1} ($\lambda = 12.08$ μm) and 816 cm^{-1} ($\lambda = 12.25$ μm). The laser pulse energy was measured by using fast calorimeters and the temporal

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characteristics were recorded by using a 50-MHz-band photodetector and a Tektronix TDS-200 oscilloscope.

Various possible pumping schemes for the NH_3 laser were studied experimentally at the first stage of investigations. These include a scheme with partially combined cavities of CO_2 and NH_3 lasers, a scheme with the NH_3 cell inserted into the cavity of a CO_2 laser, and a scheme with separate cavities.

In a scheme with *partially combined cavities* [3], whose main advantage is the maximum utilisation of the pump energy, the efficiency of pump radiation conversion to the 12.08- μm region was quite low. This is apparently due to a low Q factor of the NH_3 laser resonator, which is determined in our case by the presence in the resonator of a diffraction grating with parameters providing the maximum pump energy. In addition, the NH_3 laser radiation passes through the active medium of the CO_2 laser, which may lead to additional losses [8]. The optimisation of this scheme involves considerable technical difficulties and hence we consider it to be unpromising.

In analogy with the results obtained in [18], it can be assumed that *the insertion of the NH_3 cell into the cavity of a CO_2 laser* in the scheme with intracavity pumping of the NH_3 laser will increase the pump energy and hence the output energy. In this case, the cavity of the CO_2 laser was formed by a spherical mirror and a diffraction grating (100 lines mm^{-1} , reflectivity to the first and zeroth diffraction orders is 65% and 24%, respectively) mounted in the autocollimation regime (Littrow scheme) for the chosen pump frequency (9R30 line of the CO_2 laser). The NH_3 laser cell was placed in the cavity of the CO_2 laser.

In the absence of ammonia in the cell (empty resonator), the pump radiation energy in the resonator was ~ 5.5 J. The pump radiation frequency was determined by using an NH_3 -filled opto-acoustic detector placed in the path of the radiation diffracted in the zero order of the grating. The level of the signal from the detector at the frequency of the

9R30 line was more than two orders of magnitude higher than the signal level at the adjacent lines. It was found that the signal from the detector decreased by a factor of 50 and more after filling the NH_3 laser cell with a mixture of ammonia and nitrogen at a pressure of 5–10 Torr and higher. The intracavity energy decreased in this case only by a factor of 2–3, depending on the composition and pressure of the mixture. We believe that this is due to a change in the pump pulse spectrum in the presence of a strongly absorbing gas in the resonator. This means that the modes that are in good resonance with the absorption lines of ammonia molecules are suppressed and hence efficient pumping of ammonia does not occur. The highest output energy (2.5 mJ pulse^{-1}) was obtained for the $\text{NH}_3 : \text{N}_2 = 1 : 150$ mixture at a total pressure of 45 Torr.

An increase in the CO_2 laser pulse energy to ~ 8.8 J in an empty resonator did not improve the situation due to a strong self-focusing of pump radiation in the nonlinear absorbing gas when the ammonia–nitrogen mixture was introduced into the NH_3 laser cell.

These results lead to the conclusion that schemes with the intracavity NH_3 cell in a CO_2 laser are also unpromising. This is due, first, to complications associated with a simultaneous optimisation of operating conditions of two (CO_2 and NH_3) laser systems and, second, to the nonlinear phenomena occurring in the common laser resonator (self-focusing).

Scheme with separate cavities. Generally speaking, different versions are possible for implementing schemes with separate cavities for NH_3 and CO_2 lasers. Having analysed the situation, we chose a version close to that proposed in [6] (Fig. 1). In this scheme, the pump radiation emerging from the zero order of diffraction grating (2) (100 lines mm^{-1} , the reflectivities to the first and zeroth diffraction orders are approximately the same) falls on beamsplitter diffraction grating (3) (75 lines mm^{-1} , the blaze angle $\alpha = 37^\circ 30'$, reflectivity to the first diffraction order is 86% for

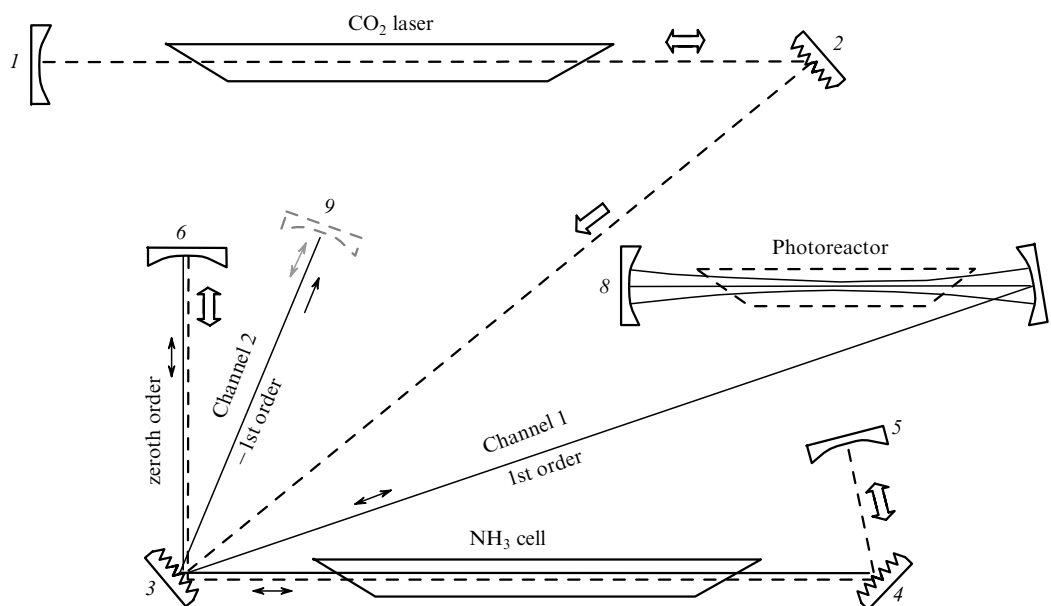


Figure 1. Scheme of a photochemical unit with separate resonators of CO_2 and NH_3 lasers and normal incidence of pump radiation on the beamsplitter grating: (1, 5–9) mirrors; (2–4) diffraction gratings; broad and narrow arrows indicate the directions of propagation of CO_2 and NH_3 laser radiation, respectively.

$\lambda = 16 \mu\text{m}$) along a direction close to the normal to its surface. The pump radiation reflected in the first order at $9.22 \mu\text{m}$ is directed to the NH_3 cell. The NH_3 laser resonator is formed by diffraction grating (4) ($100 \text{ lines mm}^{-1}$, reflectivity to the first diffraction order is $\sim 90\%$) and mirror (6), which are coupled through the zero order of grating (3). Mirror (5) blocks the pump radiation that was not absorbed in the cell. The NH_3 laser radiation at $12.08 \mu\text{m}$ incident from the side of grating (4) (channel 1) and mirror (6) (channel 2) is extracted through the 1st and -1st orders of grating (3), respectively. Mirror (9), which blocks channel 2, makes it possible to concentrate the entire radiation at $12.08 \mu\text{m}$ in channel 1 and also increases the Q factor of the NH_3 laser resonator. All mirrors have a radius of curvature $R = -10 \text{ m}$.

Because this scheme was used in subsequent experiments, Fig. 1 also shows a hypothetical photoreactor which, together with the focusing optical elements [mirrors (7) and (8)] is located in channel 1.

A version of the scheme presented in Fig. 1, in which the pump radiation is incident on grating (3) not along the normal, but at an angle close to the autocollimation angle for $\lambda = 9.22 \mu\text{m}$, seems to be more attractive for maximum possible utilisation of the pump radiation. This scheme increases the pump efficiency compared to the scheme presented in Fig. 1 due to an increase in the pump radiation reflection to the first order (see, for example [22]).

To compare the potentialities of these schemes, we measured the reflectivities of radiation at $9.22 \mu\text{m}$ to the 1st and -1st diffraction orders of grating (3) for various angles of incidence of the pump radiation on it. The results are shown in Fig. 2. On the whole, the shape of the curves shown in Fig. 2 corresponds to the predicted behaviour [22]. However, one can see that the upper curve becomes nonmonotonic at an angle of 0° corresponding to the normal incidence of radiation on the grating. Apparently, a part of the radiation pulse reflected specularly from grating (3) falls into the active medium of the CO_2 laser and is reflected back after amplification, which leads to a noticeable increase in the energy of radiation incident on grating (3) and directed to the 1st order. In addition, for the scheme shown in Fig. 1, the contribution to the 1st order

is made both by radiation incident directly on the grating and by radiation reflected from the -1st order by mirror (6) (about 50% of radiation returning from the -1st order fell into the 1st diffraction order).

Thus, the difference in the energies directed to the cell containing NH_3 is not very large for the cases of normal incidence of radiation on the grating (taking into account the above mentioned effect of additional amplification) and incidence at an angle close to the autocollimation angle. On the other hand, the latter scheme is hard to realise in view of certain difficulties encountered during installation of its elements (the cell containing NH_3 , photoreactor, and mirrors). Moreover, the adjustment of this scheme is more complicated.

In view of the above arguments, we chose for further investigations a scheme with normal incidence of pump radiation on beamsplitter grating (3) (see Fig. 1). In this scheme, the pump energy incident on a cell with NH_3 was $1.6\text{--}1.7 \text{ J}$. The pump fluence was $\sim 1.1 \text{ J cm}^{-2}$ for an area $S \sim 1.5 \text{ cm}^2$ of the radiation spot on the cell.

3. Experimental results and discussion

Recall that the main purpose of our study was to explore the possibility of realising in principle the scheme in which a photoreactor is inserted in the cavity of an NH_3 laser and to determine the optimal conditions for its operation.

In order to find the optimal parameters of the active medium of the NH_3 laser, we studied the dependence of the output energy on the pressure of the $\text{NH}_3\text{--N}_2$ mixture for various ratios of the component concentrations (Fig. 3). These dependences correspond on the whole to the results obtained earlier (see, for example, [8, 23]). The maximum energy of the radiation emerging through channel 1 ($180 \text{ mJ pulse}^{-1}$) was achieved at a frequency of 816 cm^{-1} for the $\text{NH}_3 : \text{N}_2 = 1 : 70$ mixture at a total pressure of about 80 Torr and an NH_3 laser cell of length 100 cm. In experiments with the focusing resonator (see below), the energy of the radiation emerging through channel 1 was maintained at a level of $120\text{--}130 \text{ mJ}$.

It is known that the mechanism of NH_3 laser operation is quite complicated. The output pulse can be formed due to

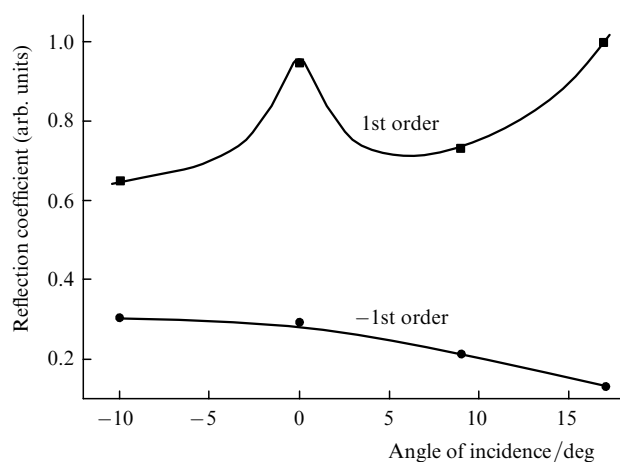


Figure 2. Reflection coefficients of a diffraction grating for various angles of incidence of pump radiation (0° corresponds to normal incidence; the autocollimation angle is $\sim 20^\circ$).

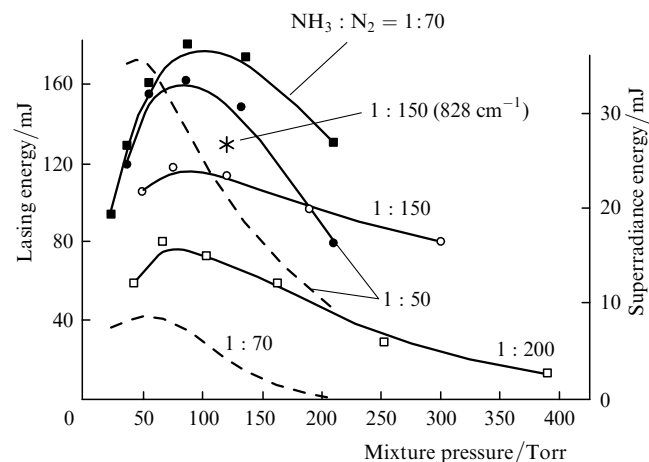


Figure 3. Dependence of the output energy of the NH_3 laser in channel 1 (see Fig. 1) (solid curves) and superradiance energy (dashed curves) on the $\text{NH}_3\text{--N}_2$ mixture pressure; the NH_3 cell length is $l_q = 100 \text{ cm}$ and the lasing frequency is 816 cm^{-1} .

stimulated Raman scattering of CO₂ laser radiation and lasing related to the population inversion in a three-level system [23, 24]. In this case, the active medium is characterised by a high gain, which may lead to stimulated emission (henceforth referred to as superradiance) in some cases even in the absence of one of the resonator mirrors. It was found that the contribution from superradiance increases with increasing partial concentration of ammonia and decreasing pressure of the mixture (dashed curves in Fig. 3). The contribution from superradiance is the highest for a lasing frequency of 828 cm⁻¹ (12.08 μm). This contribution is also found to be higher for a longer cell. Superradiance was virtually not observed for a short cell and for mixtures NH₃ : N₂ = 1 : (150 and more) at a working pressure of 70–80 Torr and higher. It will be shown below that superradiance adversely affects the parameters of the photochemical unit as a whole.

A focusing optical resonator containing the hypothetical photoreactor (see Fig. 1) was placed in the generation channel 1 of the NH₃ laser and consisted of metal mirrors (7) and (8) with focal lengths f_1 and f_2 , respectively. The parameters of the resonator were calculated by using a model based on calculations of the eigenmodes of the resonator in the Gaussian beam approximation (see, for example, [25]). The focal lengths f_1 and f_2 , the total length L_{res} of the resonator [grating (4) – mirror (8)], the distance l_1 between grating (3) and mirror (7), as well as the distance l_2 between mirrors (7) and (8), were varied in the experiment and calculations.

The energy of the field in the focusing resonator was measured using 2D-scanning of a preliminarily calibrated microprobe (of characteristic size 100 μm) according to a specially developed technique.

The effect of ‘implication’ of an additional focusing resonator was detected in direct measurement of energy in the resonator with the help of the microprobe, as well as from the radiation emerging through channel 2 (see Fig. 1).

In the former case, the energy E of radiation emerging through channel 1 was first measured with a calorimeter. Obviously, the energy circulating in channel 1 must be equal to $2E$ in the case when the radiation is simply reflected by mirror (8) without inclusion of the additional resonator. If we now take for E_{res} the energy measured by the microprobe in the space between mirrors (7) and (8) after including the additional resonator, the resonator ‘implication’ factor γ can be defined as $\gamma = E_{\text{res}}/2E$. Thus, the parameter γ is a quantitative characteristic of the energy increase in the resonator of the NH₃ laser compared to the case of single reflection of radiation.

In the latter case, we calculated the coefficient $\gamma_{\text{las}} = E_{6,8}/(E_6 + E_8)$, where E_6 and E_8 are the energies of radiation emerging through channel 2 after reflection from mirror (6) or (8) (see Fig. 1), and $E_{6,8}$ is the energy in channel 2 with simultaneously operating mirrors (6) and (8). The dependences of γ and γ_{las} on the experimental conditions are similar. However, the derivation of the qualitative dependence using γ_{las} was found to be more convenient since the procedure used for determining this parameter was much less cumbersome. Table 1 shows the parameters of the focusing resonators used in this study.

Figure 4 shows the cross-sectional areas S of the caustics calculated using the above model for various versions of focusing resonators, as well as the experimental points obtained using the microprobe. One can see that the model

Table 1. Parameters of focusing resonators.

Resonator version	f_1/cm	f_2/cm	L_{res}/cm
1	80	25	442
1a	80	25	368
2	50	35	489
2a	50	35	402
3	35	25	336

provides a satisfactory agreement with the experiment to within the measuring error of the apparatus. In all cases, a multimode oscillation of the NH₃ laser was observed. The transverse mode index n could vary between 2 and 6. The same figure shows the effective volumes V of the caustics calculated using this model. The effective volume was taken equal to double the volume of a truncated cone with a ratio $R/r = \sqrt{2}$ of radii of base and the top. As expected, the volume of the caustic decreases as we pass to a resonator with short-focal-length elements. Conversely, the effective volume of the caustic increases with the number n of the resonator mode. It is quite significant that according to the computations for a given n , the volume of the caustic increases noticeably upon a decrease in the total length of the resonator (Fig. 4d).

A decrease in the value of S at a certain fixed distance from the waist was observed upon an increase in the pressure of the NH₃–N₂ mixture in NH₃ laser cell for all versions of the resonator (see also the corresponding dependence in Fig. 6). Apparently, the observed effect is due to a variation in the mode composition of radiation upon a change in the pump and lasing conditions with the pressure in the working mixture. Further investigations are required to clarify this situation.

To determine the ways for achieving the maximum values of the resonator effect γ , we obtained the pressure dependences of γ and γ_{las} in a wide range of experimental conditions. Figure 5 shows the pressure dependence of γ_{las} in the working mixtures of various composition in the NH₃-filled cell. One can see that γ_{las} increases with increasing the total pressure of the mixture or decreasing the partial concentration of ammonia in the cell. The dependences for the coefficient γ are similar (see Figs 6 and 7). Such a behaviour of γ and γ_{las} can be attributed to a competition between two mechanisms of the development of generation in the NH₃ laser (see above). Apparently, the presence of a component associated with superradiance in the laser pulse has a considerable effect on the value of γ which decreases in this case. It should be recalled that the contribution of superradiance to the total pulse energy of the NH₃ laser increases when ammonia-enriched working mixtures are used or when the gas pressure in the cell containing NH₃ decreases (see Fig. 3).

We also studied the effect of the total length L_{res} of the focusing resonator on the parameters of the photochemical unit. The resonator versions 2 ($L_{\text{res}} = 489$ cm) and 2a ($L_{\text{res}} = 402$ cm) were compared (see Table 1). In all cases, the value of γ for version 2 did not exceed 1.4. Figure 7 shows the results for version 2a. A comparison of Figs 6 and 7 shows that an increase in the Q factor of the focusing resonator as a result of a decrease in its length L_{res} leads to a considerable increase in the value of γ (right up to $\gamma = 2.7$) and hence in the value of E_{res} as well. In turn, a comparison of Figs 3 and 7 shows that E_{res} achieves maximum values at

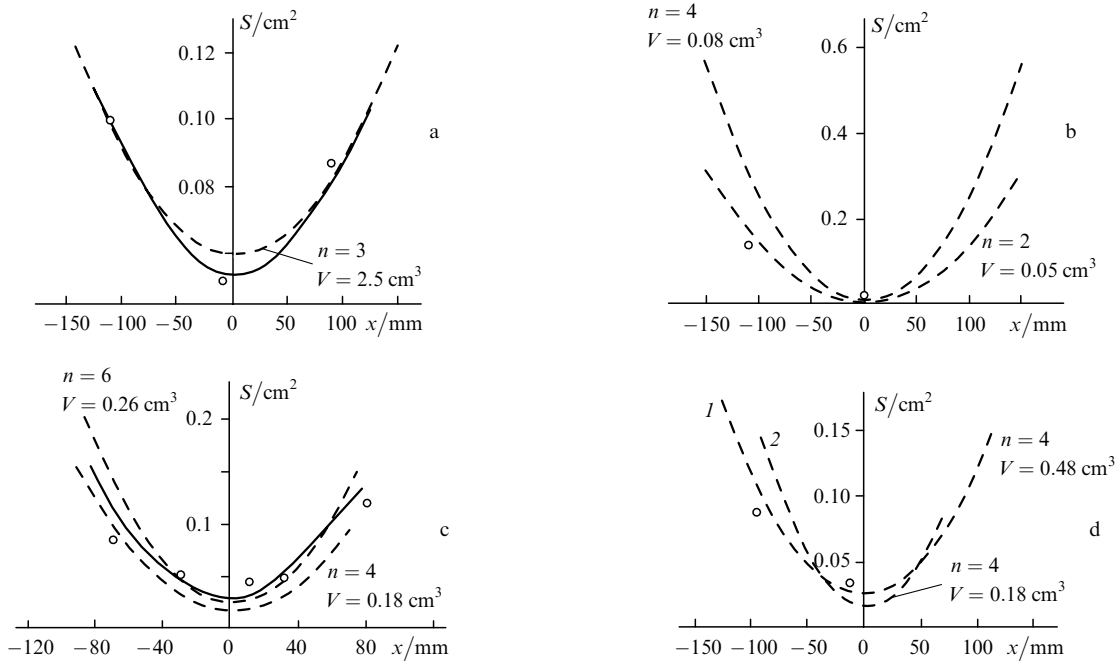


Figure 4. Dependence of the cross-sectional area S of the focusing resonator caustic on the separation x from the waist for various versions of the cavity: the NH_3 cell length is $l_q = 180$ cm, $f_1 = 80$ cm, $f_2 = 25$ cm, $L_{\text{res}} = 442$ cm (a); $l_q = 100$ cm, $f_1 = 50$ cm, $f_2 = 35$ cm, $L_{\text{res}} = 489$ cm (b); $l_q = 100$ cm, $f_1 = 35$ cm, $f_2 = 25$ cm, $L_{\text{res}} = 489$ cm (c) and $l_q = 100$ cm, $f_1 = 50$ cm, $f_2 = 35$ cm, $L_{\text{res}} = 402$ cm [circles and curve (1)] and 489 cm [curve (2)] (d). Circles correspond to the experiment, the solid curves were obtained as a result of approximation of experimental points, while the dashed curves correspond to calculations made by the adopted model for various values of n . The effective volume of the caustic was calculated for the given value of n .

a higher pressure of the working mixture than E . This is due to different forms of the dependences $E(p)$ and $\gamma(p)$. A comparison of Figs 6 and 7 shows once again that a certain decrease in the length of the cell containing NH_3 must lead to an increase in the efficiency of the photochemical unit as a whole (due to higher values of γ in our case). For the same reason, the working mixtures with a component ratio $\text{NH}_3:\text{N}_2$ close to 1 : 150 (rather than 1 : 70 as shown in Fig. 3) are optimal.

Some of the results obtained in this section are presented in Table 2. The conditions under which the fluence Φ achieves its highest value in the waist of the resonator of

the photoreactor for lasing at a frequency of 816 cm^{-1} are specified. For a frequency of 828 cm^{-1} , the results are nearly the same. The parameter V in Table 2 is the effective caustic volume calculated using the calculation model for $n = 4$. The highest values of $E_{\text{res}} = 440\text{ mJ}$ and $\Phi = 19\text{ J cm}^{-2}$ were obtained for version 3 of the resonator with the strongest beam compression ($f_1 = 35\text{ cm}$, $f_2 = 25\text{ cm}$, $L_{\text{res}} = 336\text{ cm}$). The results were obtained for the

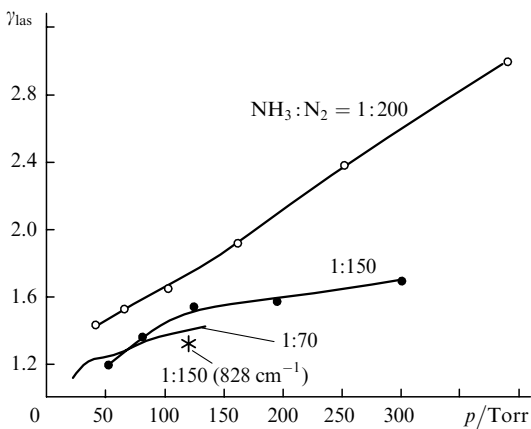


Figure 5. Dependence of the parameter γ_{las} on pressure p of the $\text{NH}_3 - \text{N}_2$ mixture for $l_q = 100$ cm, $f_1 = 80$ cm, $f_2 = 25$ cm, $L_{\text{res}} = 368$ cm and a lasing frequency of 816 cm^{-1} .

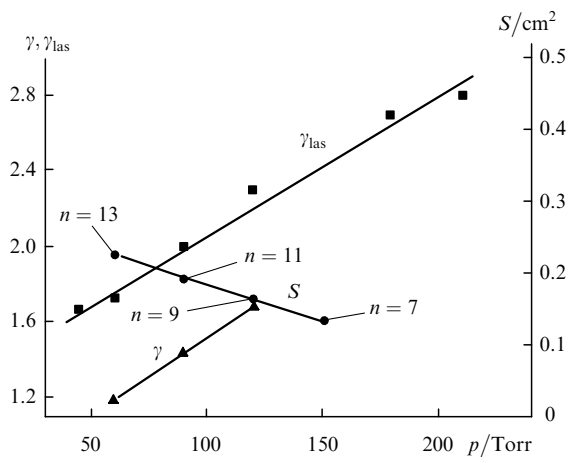


Figure 6. Dependence of the parameter γ , γ_{las} and the cross-sectional area S of the caustic waist on pressure p of the mixture $\text{NH}_3 - \text{N}_2 = 1 : 250$ for $l_q = 180$ cm, $L_{\text{res}} = 442$ cm, $f_1 = 80$ cm, $f_2 = 25$ cm. Symbols on the dependence $S(p)$ correspond to various values of the transverse mode index n obtained for the adopted model and ensuring the best agreement between theory and experiment.

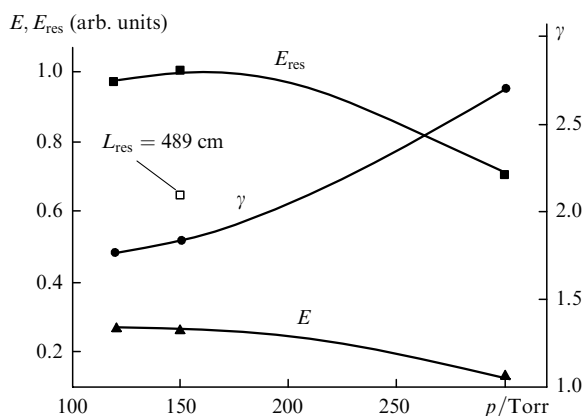


Figure 7. Dependence of the output energy E in channel 1, energy E_{res} in the focusing resonator, and the resonator effect γ on the pressure p of the $\text{NH}_3:\text{N}_2 = 1:150$ mixture for $l_q = 100$ cm, $L_{res} = 402$ cm, $f_1 = 50$ cm and $f_2 = 35$ cm. The light square corresponds to E_{res} for a resonator of length 489 cm.

$\text{NH}_3:\text{N}_2 = 1:150$ mixture at a total pressure of 120 Torr for a 100-cm-long cell.

Thus, the highest value of γ obtained in this work is 2.7. It follows from the experiment that the Q factor of the resonator of the NH_3 laser is one of the main parameters determining the value of γ for a given gain in the medium. It should be interesting to compare the value of γ for the NH_3 laser with the analogous parameter for the CO_2 laser by defining it as $\gamma_{\text{CO}_2} = E_{res}/2E$ (E is the lasing energy in the standard mode (Fig. 1) and E_{res} is the energy in the resonator operating in accordance with the scheme in [18]). In our experiment, the replacement of the output grating (2) (see Fig. 1) in the resonator of the CO_2 laser by a highly reflecting grating at the 9R30 line led to the value $\gamma_{\text{CO}_2} = 1.8 - 2.1$. In principle, γ_{CO_2} may attain even higher values at other lines. However, it is obvious that the values of γ obtained for the NH_3 laser are quite compatible with the values of this parameter for the CO_2 laser. Note that a further increase in the value of γ is possible. For example, in the scheme shown in Fig. 1, the parameters of beamsplitter diffraction grating (3) have a considerable influence on the operation of the entire system, including the Q factor of the NH_3 laser resonator. An appropriate choice of these parameters is a separate problem; in our experiment, the values of these parameters were not optimal. We believe that optimisation of the beamsplitter grating will considerably increase the value of the parameter γ .

Table 2. Experimental and calculation (V) results.

Resonator version	$\Phi/\text{J cm}^{-2}$	S/cm^2	γ	E_{res}/mJ	V/cm^3
1		0.05			3.2
1a	6.7	0.06	1.2	400	4.2
2	6.0	0.04	1.2	240	0.18
2a	8.3	0.04	1.8	330	0.48
3	19	0.02	1.7	440	0.08

Note. The results were obtained for the $\text{NH}_3:\text{N}_2 = 1:150$ mixture at a total pressure of 120 Torr for a 100-cm-long cell and a lasing frequency of 816 cm^{-1} .

4. Conclusions

The main conclusions following from the results obtained in this work can be formulated as follows.

(i) The basic possibility of constructing a photochemical unit based on an NH_3 laser with an intracavity photoreactor is demonstrated.

(ii) The scheme of a photochemical unit with a nearly normal incidence of pump radiation on the beamsplitter grating is found to be optimal.

(iii) The conditions for obtaining the highest lasing energy inside the resonator are different from the conditions under which highest lasing energy is achieved in the NH_3 laser. The presence of a component associated with super-radiance in the lasing pulse lowers the 'implication' factor γ of the additional resonator.

(iv) The highest value of γ achieved in this study is 2.7, which is comparable with the value of the analogous parameter for the intracavity system based on the CO_2 laser. The value of parameter γ can be increased further by optimising the resonator parameters.

Note also that the development of an NH_3 laser with the output energy of 1 J and higher is quite possible. In this case, the value of E_{res} in the resonator inside the photo-reactor must be about 4–5 J, and a fluence $\Phi \sim 20\text{ J cm}^{-2}$ can be achieved in a caustic with an effective volume V equal to tens of cubic centimetres. Considering further that the repetitively pulsed regime of operation of the NH_3 laser can be realised without any serious problems [4, 10], such a laser can be certainly used in large-scale photochemical devices.

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