Gain and lasing of an N_2O-N_2(CO)-He mixture in a supersonic stream

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Results are presented of an experimental investigation of the dependence of the gain of an N_2O-N_2-He mixture on the partial composition, stagnation temperature, and distance from the nozzle slit at a wavelength 10.9μ. The measurements are performed for a shock-heated mixture flowing out of a wedge-like nozzle with a divergence angle 30° and an area ratio $A_{noz}/A_{cr} = 15$. Maximum gain is attained in a mixture containing 0.1 N_2O - 0.2 N_2 - 0.7 He at a distance 15.5 mm from the nozzle slit. The laws governing the escape of the supersonic gas are determined, and used to determine the gain per unit length. The effect of substitution of carbon monoxide for nitrogen and of addition of various amounts of carbon monoxide to the optimal mixture on the gain is assessed. The gain is measured in mixtures containing CO at a wavelength of 10.9μ (N_2O) as well as at 10.6μ for chemically produced CO_2. The lasing power is measured for the optimal mixture 0.1 N_2O - 0.2 N_2 - 0.7 He. The experimental plots are based on the maximal values of the gain.

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The method of obtaining inverted population by expanding a mixture of gases in nozzles and jets has by now found extensive use. The development of gasdynamic lasers (GDL), based on this principle, particularly using the CO_2 molecule, has provided unprecedented power levels in cw operation. Besides GDL, intensive studies are being made of chemical lasers and it is proposed to develop their combination, namely a chemical GDL. In this respect, interest attaches to the nitrous-oxide molecule (N_2O) which has a vibrational-level system analogous to CO_2.

Lasing in N_2O was first registered in an electric-discharge system.[1] Energywise, however, the electric-discharge N_2O lasers turn out to be several times less effective than CO_2 lasers.[2] If there is decreased interest in further investigation of the laser properties of working media based on the N_2O molecule, and the number of publications on nitrous oxides has since that time been extremely limited. The lowering of the population inversion on the N_2O molecule in the electric discharge is determined mainly by the following processes: When an N_2O molecule collides with electrons, permanent population of the lower laser level takes place ($\sigma_{10^00}/\sigma_{00^01} = 1.2$)[3]; intense dissociation of the N_2O molecule takes place as a result of electron-molecule collisions[4]

$$N_2O+e \rightarrow N_2O^+ + e + e, \quad (1)$$
$$N_2O^+ \rightarrow NO^+ + N, \quad (2)$$
$$N_2O+e \rightarrow N_2O^- \rightarrow N_2 + O^-, \quad (3)$$
$$N_2O+O^- \rightarrow NO^- + NO, \quad (4)$$
$$NO^- + M \rightarrow NO + M + e. \quad (5)$$

In the case of reaction (1), a positive ion N_2O^+ is produced with a lifetime on the order of 10^{-6} sec, followed by dissociation in accordance with the reaction (2).[4] As a result of the electron sticking (3), an unstable intermediate complex[5] N_2O^- is produced, the decay of which is followed by the reactions (4) and (5).[6] Excitation of the electronic energy levels also leads to the decay of N_2O. These causes can be reduced to a minimum in a pulsed gas dynamic regime, which can be conveniently realized in a shock tube in conjunction with a planar supersonic nozzle. A gain close to that realized with CO_2 mixtures by using the mixture N_2O–N_2–He in a GDL was reported earlier.[7]

The present paper is devoted to an assessment of the feasibility of GDL with N_2O–N_2–He and of a chemical GDL with N_2O–CO–He, as well as to the dynamics of the escape of a jet from a planar nozzle. An analysis of the vibrational spectra of the molecules N_2O, N_2, and CO shows that the energy defect between the antisymmetrical vibration of N_2O and the first vibrational levels of N_2 and CO amounts to 107 and 81 cm^{-1}, respectively, and experimental investigations[6] confirm the presence of almost resonant exchange. The duration of the existence of the active laser medium in the shock tube is determined by the stagnation parameters and by the processes of interaction of the reflected shock wave with the contact surface.

A rigorous determination of the inverted population in a pulsed supersonic stream of N_2O from a planar nozzle is made difficult by the very scanty information on the relaxation times and their temperature dependences, as well as by the insufficient study of the laws governing the behavior of a pulsed jet. An analysis of the published data on the relaxation of the vibrational energy of the molecule working levels 00^01 and 10^00 has shown that an experimental investigation of the laser properties is best carried out with N_2O–N_2(CO)–He mixtures. In this case, to calculate the parameters in the nozzle it suffices to confine oneself to the known approximate relations for the temperature and density in terms of the Mach number (M) and the adiabatic constant (γ):

$$T = T_{cr}\left(\frac{\gamma+1}{2}\right)\left(1+\frac{\gamma-1}{2}M^2\right)^{-1}, \quad \rho = \rho_{cr}\left(\frac{T}{T_{cr}}\right)^{1/(\gamma-1)},$$
$$T_{cr} = T_2\frac{2}{\gamma+1}, \quad \rho_{cr} = \rho_2\left(\frac{2}{\gamma+1}\right)^{\gamma/(\gamma-1)}.$$

The value of M along the nozzle and the relaxation time τ were determined from the expressions

$$\frac{A}{A_{cr}}=\frac{1}{M}\left[\frac{2+(\gamma-1)M^2}{\gamma+1}\right]^{\eta},\quad \eta=\frac{\gamma+1}{2(\gamma-1)}, \tag{6}$$

$$\frac{1}{\tau}=\frac{1-X_M}{\tau_{N_2O-N_2O}}+\frac{X_M}{\tau_{N_2O-M}}, \tag{7}$$

where X is the molar fraction of the M component.

To take into account the influence of the temperature and pressure on the relaxation time of the upper level we used the relations

$$\tau_{N_2O-N_2O}=\frac{\exp(10.9/T^{1/3})}{25.7p},$$

$$\tau_{N_2O-N_2}=\frac{\exp(20.4/T^{1/3})}{p},\quad \tau_{N_2O-He}=\frac{\exp(10.74/T^{1/3})}{p};$$

the experimental values of the relaxation time of the symmetrical vibration were obtained mainly at $T=293$ °K, and we assumed a relaxation time $\tau_{10^00}=0.165$ sec-atm, which is much less than the expansion time for any section of the supersonic part of the nozzle.

To estimate the upper limit of the amplifying properties, we consider the flow of the mixture 0.1 N_2O–0.2 N_2–0.7 He through a wedgelike nozzle (aperture angle 30°, height of critical section $h=1.3$ mm, ratio of areas $A_{noz}/A_{cr}=15$). The stagnation parameters were calculated with allowance for the excitation of the rotational and vibrational degrees of freedom. For $T=1400$ °K and $p=7.8$ atm, the inversion at the slit of the nozzle amounts to 1.2×10^{-2} of the total concentration of the N_2O molecules.

We use the expression for the weak-signal again α in the case of expansion due to collisions

$$\alpha=\frac{\lambda^2(2J+1+2\Delta J)}{8\pi\tau\nu_c Q}\left\{N_{00^01}\exp\left[-\frac{F(J+\Delta J)hc}{kT}\right]-N_{10^00}\exp\left[-\frac{F(J)hc}{kT}\right]\right\}, \tag{8}$$

where λ is the wavelength, τ is the radiative lifetime of the 00^01–10^00 transition; ν_c is the frequency of molecular collisions; Q is the rotational partition function; $\Delta J=\pm1$; B is the rotational constant; J is the rotational quantum number; $F(J)=BJ(J+1)$; N_{00^01} and $N_{10^00}=\sum N_J$ are the populations of upper and lower vibrational levels; for the most intense line, $P(20)$, the value of

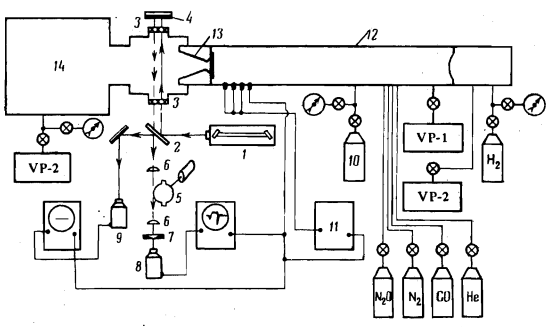

FIG. 1. Experimental setup: 1—probing laser (N_2O, CO_2), 2—semi-transparent mirror, 3—KCl windows, 4—rotating mirror, 5—shutter, 6—lenses, 7—filter, 8—signal receiver, 9—control receiver, 10—mixture, 11—meter for incident shock-wave velocity, 12—shock tube, 13—nozzle, 14—receiver.

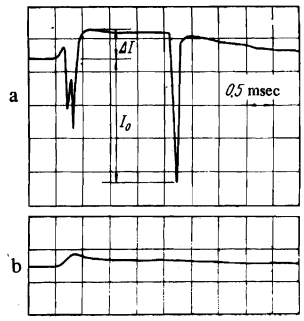

FIG. 2. Oscillograms of signals: a—probing-laser signal passing through the supersonic stream; b—spontaneous-emission signal.

α obtained at the nozzle slit is of the order of 1.7% cm^{-1}, and c is the speed of light.

To verify the foregoing, we have performed experiments on the amplifying properties of the gas mixtures N_2O–N_2(CO)–He, N_2O–N_2–CO–He, and pulsed planar flow. The experimental setup is illustrated in Fig. 1. The installation consisted of a shock tube 12 of length 900 cm, at the end of which was placed a supersonic planar wedge-shaped nozzle 13 with aperture angle 30°, critical cross section area 1.3×90 mm^2, and length 36 mm, a receiver 14, and an assembly for measuring the gain. The receiver and the low- and high-pressure chambers (LPC, HPC) of the shock tubes were separately evacuated, and for this purpose diaphragms were placed at the entrance to the nozzle and between the LPC and the HPC. The laser gas was mixed beforehand and injected into the LPC to a pressure 147 Torr. Application of the propelling gas broke the diaphragm and caused compression and heating of the laser mixture. When the incident wave was reflected from the end, the second diaphragm broke and the heated mixture escaped through the nozzle.

To determine the gain of the active medium we used a probing electric-discharge N_2O (or CO_2) laser 1 with wavelength 10.9 (or 10.6) μ, the beam of which was incident on a semitransparent tiltable mirror 2. The beam then proceeded to the laser unit through KCl window 3, where it crossed twice the escaping jet at right angles and was reflected from the tiltable mirror 4. To fix the zero level, the beam was modulated with a shutter 5 at a frequency 200 Hz, the duration of the calibration pulse being 0.11 msec and the laser power 0.5 W. To cut off the spontaneous emission at $\lambda=4.5$ μ, an InSb filter 7 was placed in front of the receiver 8 (GeAu). The fluctuations of the probing laser were monitored with receiver 9. The working mixture was prepared in a mixing flask at a pressure 2 atm. A mass-spectrometric control analysis of the batches of the injected mixture in five successive experiments has shown that the composition of the mixture in the LPC remains constant within not more than 5%.

The gain was determined from the ratio $\Delta I/I_0$, where I_0 is the intensity of the probing laser and ΔI is the intensity increment following the passage of the beam through the inverted section of the jet. As seen from Fig. 2(a), the gain takes place at a time on the order of 3.5 msec, with an abrupt increase and a prolonged smooth decrease. The decrease of the gain is apparently due to dissociation processes and not to a change in

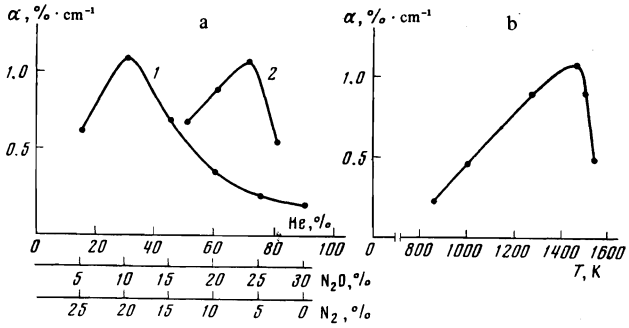

FIG. 3. Plots of the gain at a distance 15.5 mm from the nozzle slit at $p_0 = 147$ Torr: a—in the mixture $N_2O - N_2 - He$ vs the partial composition (curve 1—for He = 70% = const., curve 2 for $N_2O/N_2 = 1/2$); b—in the optimal mixture 0.1 N_2O – 0.2 N_2 – 0.7 He vs the stagnation temperature.

the gas parameters upon interaction of the reflected shock wave with the contact surface, inasmuch as no decrease was observed in analogous measurements made on the CO_2–N_2–He mixture.

An investigation of the dependence of the gain on the stagnation parameters and on the ratio of the mixture components makes it possible to determine the optimal conditions for obtaining maximum inverted population. The dependence of the gain on the partial composition of the mixture was investigated by varying the content in the nitrous oxide and nitrogen at constant helium content (70%), and the helium content was varied at a fixed ratio $N_2O/N_2 = 1/2$. An analysis of the experimental results shown in Fig. 3(a) indicates that the largest gain is realized in the mixture 0.1 N_2O–0.2 N_2–0.7 He. In the temperature interval up to 1400 °K, the gain increases smoothly with increasing derivative as the temperature rises above 1200 °K, has a maximum in the region of 1400 °K, and decreases sharply with further increase of temperature (~ 1500 °K), in contrast to mixtures with CO_2 (Fig. 3(b)), owing to the cascade character of the N_2O molecule dissociation.

When the mixture is diluted with an inert gas (He), the density of the N_2O molecules decreases together with the heat released in the decomposition process, so that the dissociation is smoother and the inversion is preserved at higher temperatures. Since the gain builds up within a finite time reckoned from the instant of reflection of the shock wave from the end of the nozzle, we have estimated the degree of thermal decomposition of the nitrous oxide. At stagnation parameters $T_2 = 800$–1600 °K and $p_2 < 10$ atm, corresponding to a maximum particle concentration 7×10^{-5} mole/cm^3, the monomolecular reaction of the decomposition of N_2O is of second order and the change in the N_2O concentration can be expressed by the following system of equations:

$$\frac{d[N_2O]}{dt} = -k_1[N_2O][M] - (k_2 + k_3)[N_2O][O], \quad (9)$$

$$\frac{d[O]}{dt} = k_1[N_2O][M] - (k_2 + k_3)[N_2O][O], \quad (10)$$

where $[M]$ is the total particle concentration, and the rate constants are[9,10] $k_1 = 1.38 \cdot 10^{15} e^{-57.0/RT}$, $k_2 = k_3 = 1.6 \cdot 10^{13} e^{-25.5/RT}$ cm^3/mole-sec. The calculations show that for the instant of time when the gain reaches a maximum the number of dissociated N_2O molecules does not exceed 20%. As seen from a comparison with the CO_2 molecule, the characteristics of the amplification are significantly shifted towards lower temperatures, an important factor in practical applications.

To determine the inversion zone and the location of the maximum of the gain, we measured the gain in the range 3–96 mm from the nozzle slit for the optimal mixture 0.1 N_2O–0.2 N_2–0.7 He. The maximum gain occurred at a distance 15.5 mm from the nozzle slit in the region of the escape of the jet into the free space. Extrapolation of the $\alpha = f(l)$ curve towards larger distances from the nozzle orifice shows that the inversion can be observed at distances more than 200 mm.

With N_2 replaced by CO, the amplification properties were investigated for mixtures of composition 0.1 N_2O–0.1 N_2O–0.2 CO–0.7 He and 0.15 N_2O–0.15 CO–0.7 He (initial pressure $p_0 = 147$ Torr). CO_2 can be produced in a mixture N_2O–CO–He as a result of the chemical processes[11,12]

$$N_2O + M \rightarrow N_2 + O, \quad O + CO + M \rightarrow CO_2 + M,$$
$$N_2O + CO \rightarrow N_2 + CO_2. \quad (11)$$

We note that the CO dissociation at temperatures below 1500 °K is very insignificant.

In view of the presence of two active laser molecules N_2O and CO_2 in the heated gas mixture, the gain was measured at the wavelengths 10.9 and 10.6 μ, respectively, and the results are shown in Fig. 4. At temperatures $T_2 < 900$ °K there is no gain at 10.6 μ, and the calculated amount of CO_2 produced by the time the maximum gain is reached does not exceed 0.2%. It is seen that when gain appears at 10.6 μ (CO_2), the gain at 10.9 μ decreases. The reason is the decrease in the concentration of the working molecules (N_2O) and the decreased lifetime of the upper level 00^01 of N_2O in the presence of the CO_2.[13] The smooth decrease of the gain with increasing temperature shows that no cascade decomposition of N_2O takes place. The gain in the mixture 0.1 N_2O–0.2 CO–0.7 He is approximately half as large than in the mixture with the same N_2 content at the same stagnation parameters. This points to a lower efficiency of carbon monoxide as a vibrational-energy reservoir for N_2O, possibly as a result of excitation of the symmetrical and deformation types of vibrations.

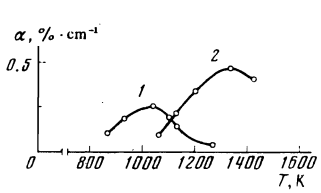

FIG. 4. Dependence of the gain in the mixture 0.1 N_2O – 0.2 CO – 0.7 He at a distance 15.5 mm from the nozzle orifice at p_0 at 147 Torr on the stagnation temperature (curve 1—at 10.9 μ wavelength, curve 2—at 10.6 μ).

The statement[14] that CO is less effective by two orders of magnitude than N_2 is apparently in error. The reason for this error may be the difference in the amount of vibrational energy stored in the N_2 and CO when excited by electric discharge. Our experiments in which 10% CO was added to the optimal mixture 0.1 N_2O–0.2 N_2–0.7 He have shown that, in contrast to the electric-discharge system,[2] no improvement takes place in the parameters of the active medium.

The main characteristic of the laser medium is the gain per unit length; to determine it, it is necessary to know the laws governing a jet escaping from a planar nozzle. Investigations of planar flows have revealed a number of singularities in the escape from a planar nozzle as compared with an axisymmetric one. It was established that the distance h to the Riemann wave (this is the customary name of the analog of the Mach disk in flat jets) does not depend on $\gamma = c_p/c_v$ and is proportional to $n = p_{noz}/p_\infty$ (p_{noz} is the pressure at the nozzle slit, p_∞ is the pressure in the surrounding space), whereas for an axisymmetric jet h is proportional to $\sqrt{\gamma}$ and $\sqrt{n}$.[15] In real jets escaping from flat nozzles, h is proportional to n^α, where $0.5 < \alpha < 1$, depending on the ratio of the nozzle dimensions.

So far there have been few investigations of jets escaping from non-axisymmetrical nozzles. Thus, Ivanov, Kraĭko, and Nazarov[16] note in their theoretical paper that such jets have a rather complicated spatial structure. We have investigated the formation of the three-dimensional wave structure of a jet escaping from planar sonic and supersonic nozzles, using a shock tube of 40×40 mm cross section. The picture of the flow was visualized by the IAD-451 shadow instrument. Measurements with an acoustic nozzle (critical cross section 1.5×40 mm) of nitrogen ($n = 25$) escaping into a pressure chamber has shown that the most intense expansion of the stream takes place in the direction of the minor axis of the nozzle and that near the stationary position of the Riemann wave the major axis of the jet is perpendicular to the major axis of the nozzle. This phenomenon must be taken into account when producing inverted population in streams escaping with sonic velocity.

Experimental investigations of GDL are carried out as a rule with flat non-axisymmetric supersonic nozzles at aperture angles on the order of 30°. In our study the supersonic jet was formed by a planar nozzle with half-aperture angle 15° and with critical and exit sections 1.3×40 and 20×40 mm, respectively. The placement of the major axis of the nozzle perpendicular to the IAB-451 axis has made it possible to determine the maximum jet dimensions in the plane of the major axis. From an analysis of the rarefaction waves it follows, as confirmed also by experiment, that the maximum dimension of the jet escaping from a planar finite nozzle into free space should be smaller in the plane of the minor axis of the nozzle than the corresponding dimension obtained in the two-dimension model.

Figure 5(a) shows the variation of the position of the jet boundary in the plane of the major axis as a function of time for a section located $l = 50$ mm from the nozzle

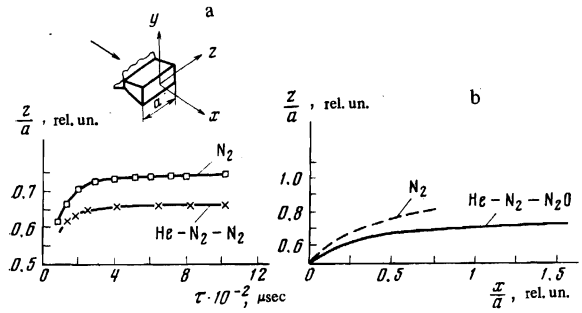

FIG. 5. Ratio of the half-width of the jet z to the width of the nozzle a in the plane of the major axis of the nozzle: a—at a distance 15 mm from the orifice of the nozzle vs the time (the lower curve is for the He—N_2—N_2O mixture; b—vs the distance downstream.

slit. The position of the jet boundary is given along the ordinate axis in units of the nozzle width. It is seen that the geometric dimensions of the jet become practically constant approximately 250 μsec after the start of the escape. Figure 5(b) shows the dependence of the geometric dimensions of the jet on the distance along the escaping jet. By varying the intensity of the incident shock wave and the pressures in the shock tube and in the pressure chamber, we varied n in the range 15–70, but this exerted no noticeable influence on the width of the jet in the plane of the major axis.

Experiments on lasing of N_2O–N_2 (CO)–He mixtures were performed with a non-selective resonator having mirrors of 44 mm diameter. To observe separately the lasing at the wavelengths 10.9 and 10.6 μ we used a prism monochromator. Investigations of the dependence of the maximum energy in the lasing regime for the optimal mixture 0.1 N_2O–0.2 N_2–0.7 He on the dimensions of the exit aperture have shown that the maximum power is ~15 W at a ratio $R_{apert}/R_{mirror} = 0.18$. In the mixture 0.1 N_2O–0.2 CO–0.7 He, lasing was registered at both 10.9 and 10.6 μ. The lasing pulse energy at 10.9 μ was lower than in the N_2O–N_2–He mixture, in the same regime, owing to the smaller gain.

It should be noted in conclusion that even in a pulsed shock tube it is impossible to exclude completely the influence of dissociation on the amplifying properties of laser media containing N_2O. To eliminate the influence of the dissociation of the N_2O molecule and to raise the vibrational temperature of the N_2 or CO_2 molecules used as the reservoir of vibrational energy, it is advantageous to heat the N_2 or CO molecules, to mix them with cold N_2O as they escape through the nozzle. As a result of the mixing, collisions with the previously heated N_2 or CO will then cause effective vibrational excitation of the N_2O. It appears that in this regime we can expect an increase of the gain by an approximate factor 1.5–2.

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Parametric generation of coherent radiation in a spatially incoherent pumping field

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Parametric generation in a spatially incoherent pumping field (radiation emission by a multimode ruby laser) is investigated experimentally. It is shown that at a low ratio l_r/l_n (l_r and l_n are the correlation lengths for waves at resonance and not at resonance with respect to the pumping wave), the incoherence of the nonreasonant waves has little effect on the spectral and spatial characteristics of resonant wave emission. The dependence of the self-excitation threshold and of the transformation coefficient on the correlation length l_n and on the mirror transparency is investigated for parametric generation of light. Transformation of spatially incoherent pumping into frequency-tunable emission with a divergence angle close to the diffraction value (which is smaller by approximately one order of magnitude than the pump divergence) and a transformation coefficient close to that for coherent pumping (up to 20% of the number of photons in the resonance wave and 40% of the photon flux) is obtained.

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A theoretical analysis of the processes of parametric amplification and generation[1–4] shows that under certain conditions either amplification or generation of nearly-coherent radiation can occur in an incoherent pumping field. The incoherence of the pump radiation has the least effect on the degree of coherence of this radiation (we shall call this the signal or resonant-wave radiation) if the correlation length of the pump wave with the resonant wave (l_r) is much less than either the length over which noticeable amplification takes place, or the correlation length of the pump with the idling (or nonresonant) waves (l_n).[4,5] The condition under which the incoherence of the pump has little effect on the energy characteristics of the amplification or self-excitation of the oscillations in the case of resonant-wave feedback can be determined from the expression for the spatial increment p_+ for the average field of this wave. If the synchronism conditions are satisfied and $l_r \ll l_n$, this expression can be written in the form

$$p_+ = -(\alpha_r+\alpha_n+l_n^{-1})/2+[(\alpha_n+l_n^{-1}-\alpha_r)^2/4+\gamma_0^2]^{1/2}. \quad (1)$$

Here α_r and α_n are the absorption coefficients of the resonant and nonresonant wave; γ_0 is the self-action coefficient[1)]; the correlation length of the nonresonant wave and the pump wave can be estimated from the formula

$$l_n=(l_{n\tau}^{-2}+l_{nd}^{-2}+l_{na}^{-2})^{-1/2}. \quad (2a)$$

The quantities $l_{n\tau}$, l_{nd}, and l_{na} in (2a) are respectively the temporal, diffraction, and aperture correlation lengths and are defined by the relations

$$l_{n\tau}=\tau_3/|v_{n3}|, \quad l_{nd}=k_3\rho_3^2/|1-k_3k_n^{-1}|, \quad l_{na}=\rho_3/\beta_n, \quad (2b)$$

where $\tau_3 = 2(\Delta\Omega_3)^{-1}$ is the characteristic time scale determined by the width of the pump frequency spectrum $2\Delta\Omega_3$; $v_{n3} = v_n^{-1} - v_3^{-1}$ is the mismatch of the longitudinal components of the group velocities v_n and v_3 of the nonresonant wave and the pump wave (group mismatch); k_n and k_3 are the wave numbers of the same waves in a nonlinear crystal; $\rho_3 = 2/k_3\Delta\theta_3$ is the characteristic scale of the spatial inhomogeneities of a pump wave with divergence $2\Delta\theta_3$ (at the e^{-1} level); β_n is the angle between the directions of the group velocities of the nonresonant wave and the pump wave. The length l_r is determined by expressions analogous to (2).

It is seen from (1) that two limiting regions can be separated in the dependence of the growth rate p_+ on the correlation length l_n and on the pump power density, namely, the region where $\gamma_0 l_n \ll 1$ and the region of quasi-coherent interaction, when $\gamma_0 l_n \gtrsim 1$.

In the quasi-coherent interaction region, when besides the condition