Inversion of vibrational-level population by mixing streams of nonequilibrium and carbon dioxide aerosol

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Two-phase mixing of streams of vibrational excited nitrogen and carbon dioxide aerosol is suggested as a method of increasing the inversion density and improving the energy characteristics and homogeneity of an active medium. The mixing, sublimation, and vibrational-relaxation processes are analyzed. Conditions for the realization of the model of instantaneous mixing of gas streams are obtained, and the inversion density ($\sim 10^{17}$ cm^{-3}), efficiency ($\sim 3\%$), and energy output (~ 120 J/g) are calculated. The possibility of using two-phase mixing in electric-discharge and chemical lasers is suggested.

1. From the energy point of view, gasdynamic lasers (GDL) constitute systems in which the reserve of vibrational energy produced in the adiabatic fast cooling of the relaxing gas stream is converted into radiation. The reserve of vibrational energy is determined by the initial gas temperature and by the rates of the $V-T$ relaxation. For example, for expanded heated nitrogen it can be large, $\sim 600 J/g$ at $T_N^0 \approx 4000\,°K$, since the rates of the collisional deactivation are low.[1] To convert the vibrational energy into radiation it is necessary to add to the nitrogen stream impurity molecules, on which inversion of the population of the energy levels is produced as a result of transfer of vibrational excitation. Typical examples are carbon dioxide molecules, in which the inversion takes place between the vibrational levels 00^01 and 10^00, 02^00.[2] In this case the energy reserve is determined by the rates of the $V-T$ relaxation of the gas mixture, which are functions of the composition, temperature, and pressure in the stream. In a heated gas mixture, in the region of the critical section, the $V-T$ relaxation is rapid, and under optimal gasdynamic parameters the inversion density is $\Delta N \lesssim 5 \times 10^{15}$ cm^{-3}, the efficiency is $\eta \lesssim 1\%$, and the energy delivered by one gram of mixture is $W \lesssim 10-20 J/g$.[3,4] These energy characteristics can be improved by mixing streams of cooling nitrogen and cold carbon dioxide,[5,6] in view of the slower $V-T$ relaxation. The main problem in this case is to realize molecular mixing in sufficiently large volume and ensure at the same time optical homogeneity of the active medium. In the case of diffuse mixing of the co-moving streams, the mixing zone is small. Turbulent mixing makes it possible to increase the zone, but the molecule-concentration field turns out to be inhomogeneous.

In the present study, to increase the volume of the active medium and the inversion density, we propose to use two-phase mixing of streams of vibrationally-excited nitrogen and carbon dioxide aerosol. In accordance with the phase-transition diagram,[7] the aerosol consists of a gas phase and a solid (cluster) phase, the mass ratio of which depends on the temperature and on the pressure. In principle it is possible to ensure conditions under which the main flow is determined by the solid phase and the principal role is played by the mechanical mixing of the streams of the clusters and of the vibrational-excited nitrogen, as well as diffusion of the CO_2 molecules in the region between the particles.

The heat of sublimation of the clusters, which ensures additional lowering of the temperature of the gas mixture, makes it possible, at a given degree of expansion, to raise the stagnation temperature of the nitrogen and consequently leads to an increase of ΔN both as a result of increasing the vibrational temperature of the upper level and as a result of the lower temperature of the lower levels. The smallness of the geometrical dimensions of the clusters in comparison with the wavelength of the radiation should contribute to an improvement of the optical homogeneity of the active medium.

2. We consider the interaction of supersonic streams of carbon-dioxide aerosol with temperature T_1 and nitrogen flowing in a channel of constant cross section with velocity v_N and having a translational temperature T_2 and a vibrational temperature T_N. The clusters in the streams sublimate, and a stream of a gas mixture of CO_2 and N_2 is formed.

From the equations for the conservation of the mass flows, the energy flux, and the momentum flux[8,9] before and after the mixing, neglecting the loss of the stored vibrational energy and disregarding the energy in the lower levels of CO_2, we obtain the following expression for the temperature of the gas mixture:

$$T = \frac{2}{7}\frac{1}{(1+\varkappa)k}\left[\frac{(v_N-v_{Ct})^2}{2(1/\varkappa M_N+1/M_C)}+\frac{1}{2}M_C v_{C_n}^2+c_p M_C T_1+\frac{7}{2}\varkappa k T_2-q M_C\right],$$

(1)

where $\varkappa = N/N_C$; N, N_C, M_N, and M_C are the concentrations and masses of the molecules N_2 and CO_2; v_{Ct} and v_{C_n} are the tangential and normal components of the aerosol velocity, c_p is the specific heat of the aerosol, and q is the specific heat of sublimation of the solid carbon dioxide. The temperature of the gas mixture increases with increasing T_1, T_2, v_{C_n} and the difference of the velocity of the nitrogen and the tangential component of the velocity of the aerosol, and is decreased as a result of the latent energy of sublimation of the clusters.

3. Let us analyze the dynamics of the mechanical mixing of the gas and aerosol streams, assuming that the heat drawn from the ambient goes only to evaporation of the clusters. The depth of penetration of the clusters and the time of evaporation can be obtained by solving the system of energy-conservation and motion equations:

$$dm/dt = -Q/q, \quad mdu/dt = -F, \tag{2}$$

where m is the mass of the cluster, u is the velocity of the cluster relative to the gas stream, Q is the heat-flux power, and F is the resistance force.

The character of the interaction of the cluster with the gas stream is determined by the Knudsen number $Kn = \bar{\lambda}/l$,[10] where $\bar{\lambda}$ is the average mean free path of the gas molecules and l is the characteristic dimension of the cluster. In our problem, interest attaches to the case when the cluster dimension is small, i.e., $Kn \gtrsim 1$, and free molecular flow around it takes place. In other words, no boundary layer is produced on the surface of the cluster.

The expression for Q and F is obtained from the molecular-kinetic theory.[19] For clusters of spherical shape with radius R we can write with sufficient accuracy

$$Q = \pi R^2 N k T_2 u_p (s + \pi^{-1/2})(s^2 + 2(T_2 - T_1)/T_2), \tag{3}$$
$$F = \tfrac{1}{2}\pi R^2 N M_N u_p^2 s(s + \pi^{-1/2}),$$

where $u_p = (2kT_2/M_N)^{1/2}$ is the most probable thermal velocity of the nitrogen molecules, and $s = u/u_p$ is the dimensionless velocity.

The system of Eqs. (2) with right-hand side (3) has in general no analytic solution. However, for estimates it is advantageous to introduce certain simplifications. The first integral is of the form

$$R = R_0 \left[\left(\frac{s}{s_0}\right)^{2(1-T_1/T_2)} \exp\left(\frac{s^3 - s_0^3}{3}\right) \right]^\alpha, \quad \alpha = \frac{2}{3}\frac{kT_2}{qM_N}, \tag{4}$$

where R_0 and s_0 are the initial radius and the dimension-less velocity of the cluster. Numerical integration of the system (2) shows that the function $s(t)$ at initial instants of time decreases exponentially with time, and then the decrease of the velocity slows down. It is assumed further that the decrease of the velocity is exponential:

$$s_1 = s_0 e^{-pAt}, \tag{5}$$

where

$$A = \frac{3}{8}\frac{NM_N u_p}{\rho R_0}\left[s_0^{2(1-T_1/T_2)} \exp\left(\frac{s_0^3}{3}\right) \right]^\alpha$$

is the density of solid carbon dioxide, and p is determined from the condition $|ds_1/dt| \geqslant |ds/dt|$. In this case the lower limit of the depth of penetration of the clusters and the flux $L(t)$ are determined from the expression

$$L(t) = \frac{s_0 u_p}{pA}(1 - e^{-pAt}). \tag{6}$$

The volume mixing is due to the motion of the clusters across the gas stream. If $v_{ct} = V_N$, then the mixing time t_1 can be calculated by assuming $L(t_1) = h_1/2$, where h_1 is the transverse dimension of the gas stream, when

$$t_1 = -\frac{1}{pA}\ln\left(1 - \frac{h_1 pA}{2s_0 u_p}\right). \tag{7}$$

The characteristic time of evaporation of the clusters t_2 can be obtained by substituting (5) in (4) and assuming, for example, that during this time the volume of the clusters decreases by a factor 10^2:

$$t_2 = \frac{2.3qM_N/kT_2 - s_0^3/3}{2pA(1 - T_1/T_2)}, \tag{8}$$

The molecular mixing of the nitrogen and of the carbon dioxide is the result of diffusion to half the distance between the clusters. In a wide interval of parameters, the diffusion time is $\tau_D \lesssim 10^{-6}$ sec.

4. Assuming rapid exchange of the vibrational energy between the nitrogen and the asymmetrical mode of CO_2, and assuming equilibrium of the deformational and symmetrical modes with the translational degrees of freedom, the time dependence of the population of the upper level N_3 of the CO_2 molecules is determined after the mixing by the expression

$$N_3 = \varepsilon_3 N_C \approx \frac{\varepsilon N}{1+\varkappa}\exp\left[-\frac{t}{(1+\varkappa)\tau_V}\right], \tag{9}$$

where ε_3 and ε are the average numbers of quanta per CO_2 and N_2 molecules $\tau_V = (W_{CC}N_C + W_{CN}N)^{-1}$ is the time constant of the relaxation, while W_{CC} and W_{CN} are the probabilities of the collisional deactivation of the 00^01 level of the CO_2 molecules by the CO_2 and N_2 molecules. The temperature dependence of these probabilities are given in[11].

The use of the model of instantaneous mixing of the nitrogen and carbon dioxide is justified if the mechanical and diffusion mixing, and also the sublimation and the passage of the active medium through the resonator region (2_3) takes place within a time shorter than the effective relaxation time $\tau_{eff} = (1 + \varkappa)\tau_V$, i.e.,

$$\tau_D, \ t_1, \ t_2, \ t_3 \leqslant \tau_{eff} \tag{10}$$

The population of the lower vibrational levels of the CO_2 molecules at the initial instant is determined by the translational temperature T, which is low, while the conversion density ΔN is determined completely by the population of the upper level.

In the general case, the requirements imposed on the composition can be satisfied if the stream contains an admixture of other gases, for example CO. To accelerate the relaxation of the lower level in the stream it is necessary to add He, H_2, or water vapor. Addition of H_2O to carbon-dioxide aerosol causes an increase of q, and consequently additional cooling of the gas stream, and also facilitates the condensation conditions when the aerosol is produced.

5. Let us estimate the inversion density produced by mixing of streams of vibrationally excited nitrogen and carbon-dioxide aerosol. We consider first an example in which the gas dynamic parameters are similar to those known for GDL. Let nitrogen heated to $T_N^0 = 2000$ K at a pressure $p_0 = 15$ atm be cooled on escaping through the nozzle to $T_2 = 300\,°K$. In a channel of constant cross section, the pressure is $p_N = 15$ mm Hg, $v_N = 1.9 \times 10^5$ cm/sec, and the Mach number is M = 5.3. Since the relaxation of the nitrogen in the stream can be neglected, we put $T_N = 2000\,°K$, in which case $\varepsilon = 0.2$. In accordance with relation (1), we assume that the gas mixture is not heated, $T = T_2$ at $v_{Ct} = v_N$. Assuming for the sake of argument $t_3 = 10^{-4}$ sec, we obtain $\varkappa \gtrsim 0.4$ from condition (10). Assuming on the other hand a single utilization of the CO_2 molecules, we have $\varkappa = 5$, and consequently we can neglect the relaxation of the upper laser level. Then $\Delta N = N_3 = 1.5 \cdot 10^{16}$ cm^{-3} and increases with decreasing $\varkappa$.

The maximum efficiency of the considered system is determined by the relation

$$\eta = \varepsilon \varkappa h\nu \left[3.5k(T_c^0 + \varkappa T_N^0) + h\nu_N \varepsilon(T_N^0)\varkappa + \sum_{i=1}^{3} h\nu_i \varepsilon_i(T_c^0) \right]^{-1}. \quad (11)$$

where ν, ν_N, ν_i are frequencies of the radiation and of the vibrations of the N_2 and of the i-th mode of CO_2, ε_i is the average number of quanta per CO_2 molecule in the i-th mode, and T_C^0 is the initial temperature of the carbon dioxide from which the aerosol is made. To ensure the condition $v_{Ct} \sim v_N$ it is necessary that T_C^0 be large, and we assume it equal to $2400\,°K$; this temperature is attainable in a flame of carbon monoxide. In the case considered here $\eta \approx 2\%$.

The energy output is obtained from the relation

$$W = \varepsilon \varkappa h\nu / (M_c + \varkappa M_N). \quad (12)$$

In this case $W \approx 60$ J/g. We note that in ordinary GDL the energy characteristics are worse and cannot be increased as a result of the relaxation losses. [3,4]

Let us determine the aerosol parameters which make it possible to obtain the calculated energy characteristics. From relation (1) at $T_1 = 153\,°K$, $q = 596$ J/g, [7] $c_p = 1$ J/g-deg, [12] and $v_{Ct} = v_N$ we obtain $v_{Cn} = 1.1 \times 10^5$ cm/sec. Calculating the constants and substituting them in (7) and (8), we obtain $t_1 = 2.7R_0$ and $t_2 = 11R_0$. At $R_0 \lesssim 10^{-5}$ cm, the depth of penetration of the clusters into the nitrogen stream is $L \approx 1$ cm, and the sublimation time is 10^{-4} sec. A rigorous solution of the system (2) yields for $R_0 = 10^{-5}$ cm a value $L = 1.8$ cm after 0.8×10^{-4} sec, in which case the radius of the cluster decreases by more than a factor of three. Since the relaxation losses in the nitrogen stream are small, the system under consideration makes it possible to increase considerably the pressure p_0 and the stagnation temperature T_N^0.

Let us examine the result of increasing the pressure. At $t_3 = 10^{-4}$ sec and $p_0 = 150$ atm we obtain $p_N = 150$ mm Hg. In this case the inversion is $\Delta N = 1.5 \times 10^{17}$ cm^{-3}. An increase in the stagnation pressure leads to an increase of the inversion density but, as seen from (11) and (12), leaves η and W unchanged. The depth of penetration of clusters with $R_0 = 10^{-4}$ cm amounts to 1.7 cm. With increasing temperature to $T_N^0 = 4000\,°K$ and at $p_0 = 150$ atm, $p_N = 13$ mm Hg, $\varkappa = 1.7$, $N = 4 \times 10^{17}$ cm^{-3}, $\Delta N = 9 \times 10^{16}$ cm^{-3}, $\eta \approx 3.3\%$, and $W \approx 120$ J/g. Consequently, an increase of the temperature T_N^0 leads simultaneously to an increase of the inversion density, of the efficiency, and of the energy output.

It should be noted in conclusion that the calculation of the energy characteristics was carried out under the assumption $v_{Ct} = v_N$, which is stronger than might turn out in practice. The choice of the optimal temperature of the mixture makes it possible to decrease T_C^0 substantially, which leads to an increase of η. In principle, mixing of the quasi-co-moving streams is possible, so that $v_{Cn} \ll v_N$. In the case of multiple utilization of the CO_2 molecules, the energy output tends to a limiting value equal to the product of the initial reserve of vibrational energy in the nitrogen and the quantity yield (240 J/g at $T_N^0 = 4000\,°K$).

6. Let us consider the possible sources of excitation losses. At $t_2 \approx t_3$ the clusters move out into the resonator region, and this leads to losses of three types: to deactivation of the N_2 and CO_2 molecules on account of collisions with clusters, to absorption, and to scattering of the laser radiation. The excited molecule losses δN due to collisions with clusters (concentration N_t) are determined by the expression

$$\delta N = 2\pi^{-1/2} \xi \varepsilon N N_c \langle \sigma_c u_p \rangle t_3, \quad (13)$$

where ξ is the fraction of the quenching collisions and σ_C is the cross section of the cluster. The maximum value of δN at $\xi = 1$ (all the collisions are quenching) reaches at the characteristic parameters a value $\delta N_{max} = \varepsilon N/3$. However, the experimental data[13] show that $\xi \ll 1$ when the excited molecules collide with a number of materials, and sublimation of the clusters leads furthermore to a decrease of δN. The maximum coefficient for absorption by clusters, α_t, is obtained by assuming that all the CO_2 in one cm^3 is condensed:

$$\alpha_c = \alpha_s M_c N_c / \rho, \quad (14)$$

where $\alpha_s \lesssim$ cm^{-1} is the absorption coefficient of solid carbon dioxide. For the characteristic values N_C we have $\alpha_c \sim 10^{-5}$ cm^{-1}.

The scattered radiation can lead to considerable losses in systems with large linear dimensions of the active medium, inasmuch as in this case conditions can be produced wherein the losses decrease to the level of the saturation density. In the system considered here the concentration of the scattering centers (clusters) is low, their dimension is much less than the radiation wavelength, and their total elimination is possible.

7. The increase of the inversion density in GDL is based on obtaining a maximum possible vibrational disequilibrium in the gas stream. In this case, when a gas mixture is being heated, ΔN is limited by relaxation

processes and cannot be increased by raising the temperature or pressure (the particle density). An appreciable increase (by more than one order) of ΔN is possible in the model of instantaneous mixing of the vibrationally excited nitrogen with cold carbon dioxide. The large vibrational disequilibrium of the nitrogen, obtained in an electric discharge or by high-temperature heating, is transferred to the CO_2 molecules, the lower levels of which remain empty as a result of the low translational temperature.

After effecting the instantaneous molecular mixing, the active medium decays with a characteristic time τ_{eff}, the order of magnitude of which should be not less than the time of flight of the medium through the resonator; this imposes a lower limit on the stream velocity. The principal mechanisms of gas-stream mixing are diffusion and turbulent mixing, in which individual macroscopic regions containing one gas are spread out in final analysis also as a result of diffusion. These processes impose an upper bound on the velocity. The use of diffusion mixing is ineffective because of the small depths of diffusion within the time t_3. Turbulent mixing is faster, for in this case it is necessary that the diffusion occur over the characteristic turbulence scale. Thus, the inversion density in the case of mixing of gas streams depends substantially on the geometry of the system, on the ratio of the stream velocities, and on the characteristic turbulence scale.

To obtain laser radiation with controllable mode composition it is necessary to satisfy the additional requirement that the active medium be optically highly homogeneous. Turbulent mixing violates this requirement to some degree and is the cause of additional losses in the resonator.

Let us analyze a variant of a system in which the injection of the cold CO_2 gas takes place in the region of the critical section of the nitrogen nozzle. In this case the relative stream velocity is large and should lead to stagnation and consequently to heating of the mixture and to population of the lower level. Losses to relaxation depend on turbulence scale. If this scale is small, rapid molecular mixing takes place and the relaxation losses in the critical region will be large. If the scale is large, then the losses are smaller, but no molecular mixing takes place, and this causes the excited particles to leave the resonator. As shown by experiment, [5,6] in this case there is a gain in the increase of ΔN in comparison with the usual GDL, but this gain is less than predicted by the instantaneous-mixing model, apparently for the reasons indicated above.

The system proposed in the present paper for the mixing of streams of excited nitrogen and carbon-dioxide aerosol makes it possible to realize in the best manner the model of instantaneous mixing and to attain the largest possible inversion density typical of flow-through gas-discharge lasers. Because of the large depth of penetration of the aerosol particles in the gas stream, the mixing volume reaches a practically acceptable value within short time intervals, so that large inverted-medium volumes can be obtained. The presence of clusters in the resonator can ensure, at a low level of

losses to absorption and at sufficient optical homogeneity in the resonator, an equalization of the field of the inversion density as a result of the change in the composition of the mixture. The model of instantaneous mixing is realized, since we can neglect the relaxation losses prior to a mixing that takes place in a time shorter than the relaxation time. This makes it possible to increase appreciably the pressure and the stagnation temperature of the vibrationally excited gas, which are determined by the technical means and not by the losses to relaxation as in classical GDL. This makes it possible to increase the pressure in the resonator and to relax the requirements on the ejection of the gas to the atmosphere. By attaining complete evaporation of the clusters at the entry into the resonator, we can increase the optical homogeneity of the gas-mixture stream.

It should be noted that a margin for further improvement of the energy characteristics of the system is provided by additional non-equilibrium excitation of the nitrogen in the region of the critical cross section, for example, by an electric discharge[14] or with the aid of nonequilibrium chemical reactions.

The main obstacle on the road to realization of the proposed system is the formation of a stream of carbon-dioxide aerosol with a specified average cluster dimension, with the largest specific consumption of the solid phase, and with a velocity $\sim 10^5$ cm/sec. The known experimental results[15-18] point to the feasibility in principle of solving the problem by a gasdynamic method. One of the possibilities of obtaining a stream of clusters with high velocity is apparently gasdynamic expansion prior to condensation of the combustion products of the carbon monoxide at high pressure in the combustion chamber. It is possible also to use acceleration and sorting of the previously ionized clusters in an electric field.

The principle of mixing gas streams with aerosols can find practical application also in the development of other types of lasers, such as electric-discharge CO_2 lasers, CO lasers in which CO aerosol is injected, and chemical lasers, for example with injection of fluorine aerosol.

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Nonresonant excitation of atomic fluorescence in alkali-metal vapor

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The mechanism of excitation of atomic fluorescence by radiation having a frequency ω_1 that differs considerably (by about 2000 cm^{-1}) from the atomic-transition frequency ω_0 is investigated. It is shown that the observed atomic emission is due to direct excitation of atomic electron transitions by nonresonant radiation in particle pair collisions. The process is interpreted in terms of an instantaneous change in the interaction potential of the atoms at the instant of absorption of a light quantum. A nonlinear dependence of the atomic-emission intensity on the exciting-radiation intensity is observed at high intensities of the nonresonant excitation. This dependence is ascribed to saturation effects of the electron-translational colliding atoms in the vicinity of the "intersection-turning" points.

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1. We report here the results of an experimental investigation of atomic fluorescence of alkali-metal vapors excited by laser radiation at a frequency ω_1 that is not at resonance with the frequency ω_0 of the atomic transition. The stimulus for this study was the interest in interactions between a radiation field and colliding atoms and of the influence of radiation on the dynamics of particle collision.

It is known that, in the two-level approximation, scattering of light by a system of non-interacting atoms is an elastic process, so that the emission is only at the frequency of the exciting radiation.[1] The interaction between the atoms causes the appearance of emission at the frequency ω_0 of the atomic transition at nonresonant excitation. Without going into details of the character of the collisions, the phenomenon of nonresonant excitation of atomic fluorescence can be described within the framework of the model of phase relaxation of an atomic system.[2] With such a description, the trajectories of the colliding atoms are assumed to be given and, as a rule, straight lines, while the action of the collisions reduces only to an instantaneous change of the phase of the state of the excited atom; it is important here that the characteristics of the collisions are assumed to be independent of the parameters of the irradiation field (its frequency and intensity). These approximations are violated at large detunings from resonance $\omega_1 - \omega_0$, when the difference between the energies of the incident and scattered photons is of the order of

or larger than the average kinetic energy of the colliding particles. In this case an appreciable change in the particle energy takes place in the active photons scattering by the atoms, and this leads to a noticeable change in the character of their motion. This influence of the radiation on the dynamics of the particle collisions can be interpreted as a result of an instantaneous change in the potential of the interaction between the atoms at the instant when they absorb a light quantum with energy $\hbar\omega_i$, a fact that can change not only the magnitude but also the sign of the force of interaction between the atoms. Since the duration of the atom-collision act ($\sim 10^{-12}$ sec) is incomparably shorter than the lifetime of the optical excitation ($\sim 10^{-8}$ sec), it follows that the quantum $\hbar\omega_1$ absorbed at the instant of collision will be emitted in the overwhelming majority of cases by the free atom at the resonant frequency ω_0.

Of course, in order for such an elastic process to take place, the electronic energy levels of the incident particles must be displaced at the instant of the collision strongly enough, so that the trajectories of the atoms can no longer be regarded as straight lines, and the process of excitation of the atomic fluorescence at such deviations from resonance should be described by the value of the radiation field quantum (the nonresonance).

Questions concerning the effect of a radiation field on the characteristics of broadening collisions has already