EFFECTS OF ARGON DILUTION ON THE SUPEREQUILIBRIUM PUMPING OF A CO_2 GDL BASED ON CO + N_2O

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The effects of an inert diluent (argon) are examined on the superequilibrium pumping in a CO_2 GDL based on reacting mixtures of initial compositions 5% N_2O + 25% CO + (0.1% H_2) + 70% Ar and 5% N_2O + 5% CO + 90% Ar.

Introduction. Individual exothermic stages in the overall reactions of CO + O_2 and CO + N_2O occur in a thermodynamically nonequilibrium gas flow expanding in the supersonic nozzle of a gas-dynamic laser GDL, and the energy deposition in the asymmetric mode of CO_2 raises the gain on the $00^01 \rightarrow 10^00$ CO_2 transition [1, 2]. This combination of chemical and gas-dynamic methods of producing a population inversion has been called superequilibrium pumping SEP in a GDL [2].

The SEP mechanism in the overall reaction of CO + N_2O has not been finally elucidated, which hinders the practical use of this effect in raising the power and efficienty of CO_2 GDL. In [3], a five-stage scheme was used for the overall reaction of CO + N_2O [4] to show that SEP can be due to the recombination $CO + O + M \rightarrow CO_2 + M$ in the critical section of the nozzle. Later studies [5] have shown that temperatures in the range 800-1100°K, which correspond to ignition in a CO + N_2O mixture under GDL conditions, cause the overall CO + N_2O reaction to involve a chain mechanism dependent on traces of hydrogen or water. In [6], a kinetic scheme was devised for this reaction on the basis of a chain mechanism for the conditions of a CO_2 GDL with superequilibrium pumping, and in [7], evidence was presented for the effects of hydrogeneous components on the SEP.

In [6, 7], there is a joint analysis of the kinetics of the overall CO + N_2O reaction and the subsequent relaxation in the vibrational energy, which shows that the SEP in a CO_2 GDL with the gain K_0 increased by 0.1-0.3 m^{-1} may be due to energy deposition in the vibrational degrees of freedom in the following elementary reactions:

$$CO + N_2O \rightarrow CO_2 + N_2 + 87{,}3 \text{ kcal/mole}, \quad (1)$$

$$H + N_2O \rightarrow N_2 + OH + 62 \text{ kcal/mole}, \quad (2)$$

$$OH + CO \rightarrow CO_2 + H + 25 \text{ kcal/mole}. \quad (3)$$

It has been found [8] that the conditions corresponding to the maximum SEP effect in a CO_2 GDL [2] involve the elementary reactions of CO + N_2O being characterized by strong vibrational disequilibrium in the reactants due to the formation of excited particles, which cause the reaction to be self-accelerated because energy is transferred from the vibrationally excited molecules to the initial reactants. The rate of the CO + N_2O reaction under nonequilibrium conditions is substantially dependent on the inert diluent, which determines the vibrational relaxation, because the vibrational excitation of the reagents overcomes the activation barrier in the reaction [8, 9].

One assumes that the rates of the elementary stages in CO + N_2O reaction in which energy is supplied to the vibrational degrees of freedom are largely determined by the production of the nonequilibrium vibrational-energy distribution. Therefore, it is important to examine the effects of inert components on the SEP. Previously, the SEP effect has been observed and studied for mixtures where the inert diluent was helium [2, 3, 7]. Here we present results on the SEP effect in CO + N_2O mixtures diluted with argon, which substantially affects the vibrational energy exchange and thus the kinetics of the individual stages.

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TABLE 1. Initial Compositions and Numbers of Mixtures Used, mol.%

Substance	Mixture No.									
	I	I'	II	II'	III	III'	IV	IV'	V	V'
CO_2	—	5	—	5	—	5	—	5	—	5
CO	25	20	25	20	5	—	5	—	25	20
N_2O	5	—	5	0,1	5	—	5	0,1	5	—
N_2	—	5	—	5	—	5	—	5	—	5
H_2	<0,025	<0,025	0,1	0,1	<0,01	<0,01	0,1	0,1	<0,025	<0,025
He	—	—	—	—	—	—	—	—	70	70
Ar	70	70	69,9	69,8	90	90	89,9	89,8	—	—

We used a quasistationary GDL based on a shock tube ST. We measured K_0 on the P20 line for the $00^01 \rightarrow 10^00$ transition of the CO_2 molecule and the intensity I of the IR emission at 4.3 μm from CO_2 in the range $\lambda = 4.37 \pm 0.11$ μm. The measured K_0 and I were used with the method of [10] to recover the vibrational temperatures T_3 of the unsymmetric mode and T_2 of the combined (symmetric and deformation) mode of CO_2. The SEP was determined by a comparative method [2] based on comparing the vibrational disequilibrium parameters: K_0 together with T_3 and T_2 in reacting CO + N_2O mixtures and in comparison mixtures simulating the final composition of the products.

Methods. Table 1 gives the initial compositions of mixtures I-V and those of the corresponding comparison mixtures I'-V'; mixtures I-IV and I'-IV', where argon is the diluent, were examined here. Mixtures V and V', which were diluted with helium, have been examined previously [3, 6, 7]. We give the results for V and V' in order to examine the effects of inert diluents on SEP.

The experiments were performed with two types of reacting mixtures. Mixtures I and II had carbon monoxide contents initially larger than the stoichiometric value, $\xi_{CO} = 0.25$, while III and IV were stoichiometric, $\xi_{CO} = \xi_{N_2O}$. Traces of hydrogen and water had marked effects on the CO + N_2O reaction kinetics [5]. Mass spectrometry for mixtures I and III showed that the amounts of hydrogen were not more than 0.025 and 0.01%, correspondingly. In mixtures II and IV, molecular hydrogen was introduced in amounts of 0.1%.

The basis has been given in [6] for choosing the reference mixtures to simulate the compositions of the products from CO + N_2O in order to determine the lower bound to the superequilibrium pumping effect. In that paper, a numerical calculation was performed on the product composition in CO + N_2O and an analysis of the effects of trace components in the CO_2 GDL media such as NO, CO, and O_2, as well as of the intermediate particles O, H, OH, NO_2, HNO_2, as regards the population-inversion kinetics.

To examine mixtures II and IV, namely CO + N_2O with added H_2, it is important to simulate the water vapor contents in the corresponding comparison mixtures. On the direct addition of water vapor to a comparison mixture, it is difficult to maintain the concentration with the required accuracy because H_2O condenses on the ST walls. Calculations [6] show that the water-vapor content in a reacted CO + N_2O + H_2 mixture is 70-90% of the initial hydrogen content, so we added H_2 and N_2O in the stoichiometric relation $\xi_{H_2} = \xi_{N_2O}$ to mixtures II' and IV' in amounts equal to the initial contents of hydrogen in II and IV. The overall reaction $H_2 + N_2O \rightarrow N_2 + H_2O$ produced the required amount of water vapor. With characteristic temperatures ahead of the nozzle in the comparison mixtures $T_0 > 2.2$°kK, this reaction goes to completion in less than 20-40 μsec, i.e., before completion of the quasistationary flow state in the nozzle.

We recovered T_3 and T_2 from the measured K_0 and IR intensity I by numerical solution of the following equations [10]:

$$I = A\int C(\omega)[I_{4,3}(\xi_{CO_2}, T_3, T_2, \omega) + I_{4,7}(\xi_{CO}, T_5, \omega)]\,d\omega, \tag{4}$$

$$K_0 = K(T_3, T_2), \tag{5}$$

where A is a constant determining the sensitivity, $C(\omega)$ is a function describing the spectral transmission of the IR recording system, $I_{4.3}$ and $I_{4.7}$ being the corresponding intensities

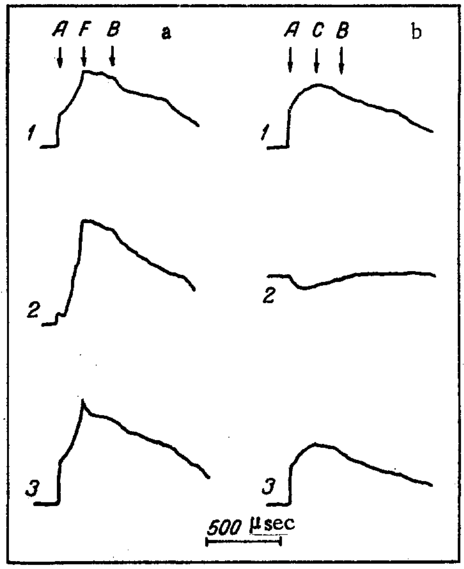

Fig. 1

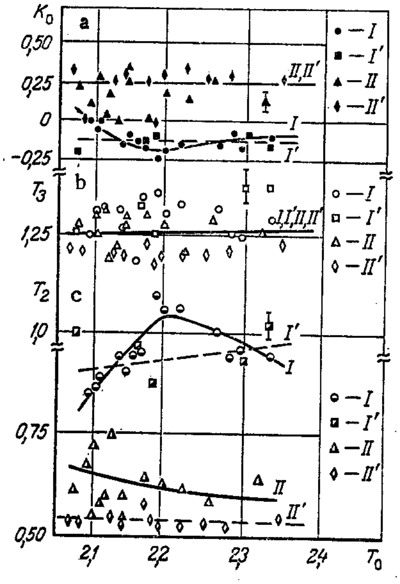

Fig. 2

Fig. 1. Typical waveforms: a) mixture V with initial gas parameters ahead of nozzle: P_i = 10.2 atm, T_i = 1180°K; b) mixture I with P_i = 11.0 atm and T_i = 1350°K; 1 and 3) IR intensities ahead of the nozzle and in flow correspondingly; 2) change in intensity ΔI in probe laser beam.

Fig. 2. Dependence of gain K_0 (m^{-1}) (a) and vibrational temperatures of asymmetric mode T_3 (10^3°K) (b) and combined mode T_2 (10^3°K) (c) for CO_2 in reacting mixtures I and II and in comparision mixtures I' and II' as influenced by the gas temperature ahead of the nozzle T_0 (10^3°K) for P_0 = 10.0 ± 1.0 atm. The curve numbering corresponds to the mixture symbols in Table 1.

emitted in the 4.3 μm band of CO_2 and the 4.7 μm band of CO averaged over the rotational structures and calculated by the method described in [10]. Under the conditions characteristic of CO_2 GDL cavities, the 4.3-μm CO_2 band and the 4.7-μm CO one overlap considerably [10]. Estimates show that the transmission function of the optical channel used here was such that λ = 4.37 ± 0.11 μm as used in recording the central part of the 4.3-μm band of CO_2 could have a contribution from carbon monoxide to the total emission for ξ_{CO} = 20% of 20-30%. This is important and was incorporated into (4) in recovering T_3 and T_2.

The constant A was determined in calibration experiments on the equilibrium emission from carbon dioxide in a 20% CO_2 + 80% He mixture heated to 800-1000°K behind a shock wave, which enabled us to calibrate the appratus and measure the intensity in the GDL without changing the optical system. Also, C(ω) was determined by the filter used; this was recovered by means of a spectrograph.

It was assumed in solving (4)-(5) that the vibrational temperatures of CO, namely T_5, and the asymmetric mode of CO_2, namely T_3, were the same. In [6], it has been shown that the error in this approximation in determining T_3 does not exceed +50 and −30°K for analogous conditions, while that for T_2 is +30 and −20°K.

We used a shock tube with a nozzle at the end described in detail in [10]. The reactions occurred in a low-pressure section behind the shock wave reflected from the end of the tube. The mixture expanded in a planar profiled nozzle of minimal length having a height h_* = 1.0 mm for the critical section and a degree of flow expansion h/h_* = 23.

The gas state ahead of the nozzle was monitored from the pressure and the IR intensity in the range λ = 4.75 ± 0.8 μm. It has been shown [6, 7] that the IR intensity in this range for a reacting CO + N_2O mixture is determined mainly by the gas temperature, because most of the radiation is due to the 4.3 μm band of CO_2 and the 4.5 μm band for N_2O, which are similar in intensity, and the total concentration of CO_2 and N_2O varies little during the reaction. Consequently, such measurements enable one to monitor the reaction from the gas temperature.

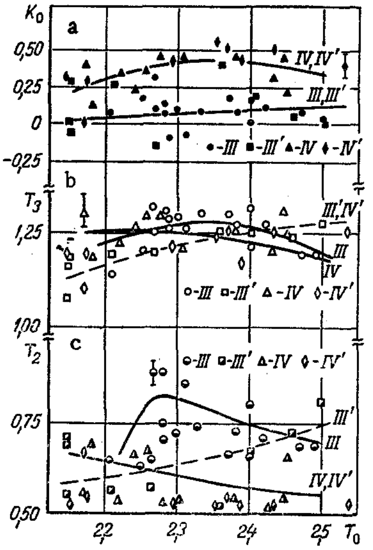

Fig. 3. Dependence of the gain K_0 (m^{-1}) and the vibrational temperatures of the asymmetric mode T_3 (10^3°K) (b) and coupled mode T_2 (10^3°K) (c) of CO_2 for reaction mixtures III and IV and comparison mixtures III' and IV' on gas temperature ahead of the nozzle T_0 (10^3°K) at $P_0 = 10.0 \pm 1.0$ atm; the numbering on the curves corresponds to that of the mixture given in Table 1.

The initial temperature T_i and pressure P_i behind the reflected shock wave (without allowance for the heating by reaction) were determined numerically from the measured incident shock-wave speed, which was measured on two baselines by piezoelectric sensors. The temperature T_0 in the reacting mixture behind the reflected shock wave after ignition was calculated with allowance for the heating due to the reaction on the assumption that the pressure behind the reflected shock wave is constant. The temperature rise did not exceed 20°K for the comparison mixtures, so we assumed $T_0 \approx T_i$; the pressure measurements showed that the rise during ignition was not more than 10-20%. The gas purities were as follows: nitrogen 99.9%, helium and argon 99.99%, hydrogen electrolytic 99.8%, and carbon monoxide and carbon dioxide of technical grade purified by triple distillation and carefully dried 99.7%. The traces of hydrogen were determined by mass spectrometry and did not exceed 0.01% in the CO_2, He, and N_2 or 0.1% in the CO. There were traces of water vapor due to H_2O desorption from the shock tube during the preparations. Mass spectrometry indicated that the water level did not exceed 0.05%.

Results. Figure 1 shows characteristic waveforms for reacting mixture V diluted with helium as obtained in [6, 7] (Fig. 1a) and for mixture I diluted with argon in this study (Fig. 1b), which enables one to examine the effects of the inert diluent on the CO + N_2O ignition kinetics and the SEP. The initial conditions ahead of the nozzle correspond to approximately equal periods A-F (Fig. 1a) and A-C (Fig. 1b) of about 150 μsec, which was required to attain 70-90% reaction.

The arrows in Fig. 1 show the instants corresponding to the start of each of the following processes: A shock-wave reflection from the end of the tube, F ignition (for mixture V), C completion of reaction (for mixture I), and B destruction of the hot-gas region ahead of the nozzle by negative-pressure waves reflected from the opposite end of the shock tube.

At the end of period A-F, mixture V showed explosive self-accelerating ignition, which was accompanied by a considerable rise in the IR intensity ahead of the nozzle (curve 1 in Fig. 1a) and in the supersonic flow (curve 3 in Fig. 1a). The gas pressure ahead of the nozzle increased by not more than 20-30%. Mixture I showed slow combustion, with the reaction rate decreasing as the reagents burned (period A-C) (curve 1 in Fib. 1b).

To provide identical periods for reactions, the initial temperature in mixture I should be about 200°K higher than in V, so the inert diluent has a substantial effect on the CO + N_2O reaction rate.

The intensity from the 4.3-μm CO_2 band in the supersonic flow characterizes the excitation of the unsymmetric mode in carbon dioxide [10], and in mixture I it was lower by about a factor 1.5-2 than in V (curve 3 in Fig. 1), so the diluent also determines the vibrational excitation of the CO_2 formed in the reactions.

Curves 2 in parts a and b of Fig. 1 show that mixture I differs from V in that the laser beam is absorbed, which may be due not only to pumping of the vibrational degrees of freedom by the reactions but also to substantial differences in the conditions for vibrational relaxation on expansion.

Figure 2 shows measurements on K_0 and on T_3 and T_2 for CO_2 in mixtures I and II containing different amounts of hydrogen together with those for I' and II'. Mixture I not containing hydrogen and correspondingly I' gave negative K_0 (I and I' in Fig. 2a); in I, K_0 had a minimum of about $-0.25\ m^{-1}$ at $T_0 = 2.2$°kK. The gains for I and I' were similar. On adding 0.1% hydrogen (II and II' mixtures), K_0 increased to about $0.3\ m^{-1}$ (II and II' in Fig. 2a). In mixtures II and II', the gains were virtually identical and varied little with T_0 in the range 2.1-2.4°kK.

T_3 was in the range 1.2-1.3°kK for mixtures I and II as well as for I' and II' and was almost independent of T_0 or the hydrogen content (Fig. 2b). Measurements on T_2 are given in Fig. 2c. T_2 for mixture I was high, attaining about 1.1°kK for T_0 = 2.15-2.25°kK (I in Fig. 2c). This is higher by about 200-250°K than for mixture I'. The additional hydrogen reduced the excitation of the combined mode considerably; T_2 for mixture II fell to 0.6-0.7°kK but remained 100-150°K higher than in mixture II' (II and II' in Fig. 2c). Therefore, $CO + N_2O$ mixtures containing excess carbon monoxide and diluted with argon show SEP only in the combined mode of the carbon dioxide, which raises its temperature by 100-250°K.

The main factors determining the absence of SEP in mixtures I and II from mixtures of type V diluted with helium [3, 6, 7] are as follows: reduction in T_3 by 300-400°K and substantially higher T_2 of about 1.1°kK, which are responsible for negative gain in mixtures I and II, whereas V showed a high gain of $K_0 \approx 0.7\ m^{-1}$ [6, 7].

Figure 3 shows the results for K_0, T_3, and T_2 for stoichiometric mixtures III and IV and the corresponding reference mixtures III' and IV'; the gain in III, which did not contain hydrogen, was close to zero (III in Fig. 3a), and the same K_0 were recorded for mixture III' (III' in Fig. 3a). There was a considerable spread in the measured gains for III and III' mixtures, evidently due to variations in the traces of water desorbed from the ST. Adding 0.1% hydrogen increased K_0 to $0.5\ m^{-1}$ for mixture IV (IV in Fig. 3a). Similar gains were recorded for IV' (IV' in Fig. 3a).

T_3 for mixtures III and IV and for the comparison mixtures III' and IV' were only slightly dependent on T_0 and on the addition of H_2 at a level of 0.1% (III, III', IV, and IV' in Fig. 3b). For $T_0 \leq 2.3$°kK, the T_3 in mixtures III and IV were somewhat higher than in the corresponding reference ones. On the whole, the T_3 for the stoichiometric mixtures (Fig. 3b) were approximately the same as those for mixtures containing excess carbon monoxide (Fig. 2b).

T_2 for III had a maximum of about 0.75°kK for $T_0 \approx$ 2.2-2.4°kK (III in Fig. 3c); T_2 for mixture III' was lower by more than 150-200°K for this T_0 range (III' in Fig. 3c). Adding hydrogen reduced T_2 approximately to 550°K for mixtures IV and IV' (IV and IV' in Fig. 3c). Therefore, stoichiometric reacting $CO + N_2O$ mixtures diluted with argon (III and IV) show SEP due to increases in T_3 in mixtures III and IV at $T_0 \leq 2.3$°kK and considerable increase in T_2 in mixture III, which did not contain any hydrogen.

These results and the data of [3, 6, 7] indicate that the SEP effect and the influence on the pumping of CO_2 modes differ considerably as between mixtures I-IV diluted with argon and that with helium V. In the first, there is no rise in vibrational temperature for the unsymmetric mode, or else it is small, whereas mixtures I-III show marked SEP for the combined mode. It is important that high vibrational temperatures of $T_2 \approx 1.1$°kK can be attained in the expanding products from the $CO + N_2O$ reaction diluted with argon, which is important to the development of CO_2 GDL on transitions between levels of the combined mode of CO_2 [11], where this reaction can be used to produce and heat the working mixture; it is also possible to improve the efficiency of such lasers in this way by additional excitation of the combined mode of CO_2 by superequilibrium pumping. This feature of the $CO + N_2O$ reaction is unique, since reacting systems in GDL usually show substantial reductions in the vibrational excitation in the expanding products, particularly in the softest combined mode, because of the strong deactivation produced by certain final products and intermediate particles [12].

The changes in SEP in mixtures diluted with argon are evidently determined by the kinetics of the overall $CO + N_2O$ reaction under nonequilibrium conditions. In the mixtures containing argon, the ignition temperatures are 200-250°K higher than for ones containing helium, which shows in particular that vibrational excitation in the reagents is important to the overall $CO + N_2O$ reaction. Therefore, the reactions in the expanding flow of a mixture diluted with argon occur at higher gas temperatures and thus differ appreciably in vibrational relaxation conditions, which may influence the kinetics of the elementary stages in the reaction and thus the SEP.

Carbon monoxide plays an important part in the vibrational energy exchange in these mixtures. Mixture I contained about 20% CO in the products, and the maximal T_2 were more than 300°K higher than for mixture III, where the carbon monixide content in the products was low (I in Fig. 2c and III in Fig. 3c). The same regularity occurs for the inert mixtures I' and III', where the difference is about 200°K (I' in Fig. 2c and III' in Fig. 3c). Carbon monoxide produces an additional channel for energy exchange between the asymmetric mode of CO_2 and nitrogen and between carbon monoxide and the deformation mode of carbon dioxide. This channel includes fast V—V' exchange for $CO_2(00^01) \rightleftarrows N_2(1) \rightleftarrows CO(1)$ and subsequent quasiresonant interaction between the molecules $CO_2(00^00)$ and $CO(1)$

$$CO(1)+CO_2(00^00)\rightleftharpoons CO(0)+\underset{03^10}{CO_2(11^10)}+\Delta E, \qquad (6)$$

where $\Delta E \approx 65$ cm^{-1} is the vibrational energy deposited in the translational degrees of freedom. It is important to consider (6) for a CO_2 GDL based on mixtures containing carbon monoxide and working on the $00^01 \rightarrow 10^00$ CO_2 transition [10]. Our results indicate that adding carbon monoxide can increase T_2 substantially and thus raise the efficiency in a CO_2 GDL working on transitions between levels of the combined mode.

NOTATION

λ, Wavelength, μm; K_0, measured weak-signal gain, m^{-1}; K, calculated gain, m^{-1}; T_3, T_2, vibrational temperatures of asymmetric and coupled CO_2 modes, respectively; T_5, vibrational temperature of carbon monoxide (CO), °K; T_i, initial temperature of gas in chamber, °K; T_0, temperature of gas after reaction, °K; I, IR intensity in the 4.3 ± 0.11-μm band; $I_{4.3}$, $I_{4.7}$, intensities of 4.3-μm CO_2 and 4.7-μm CO bands calculated from vibrational temperatures; ξ_i, molar concentration of component i of mixture; ω, wave number, cm^{-1}; $C(\omega)$, spectral transmission of filter; h_*, height of critical nozzle section, mm; ΔE, vibrational energy deficiency, cm^{-1}.

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STRUCTURE OF A LASER ABSORPTION WAVE IN THE PRESENCE OF THERMAL DISSOCIATION OF A TRANSPARENT CONDENSED MEDIUM

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UDC 621.378.385

The structures of absorption and dissociation waves are studied based on the numerical solution of the nonstationary equation of heat conduction with a nonlinear source and a model equation for the kinetics of thermal dissociation of inorganic glass.

The experimental characteristics of an absorption wave in multicomponent silicate glasses are presented in [1-3] for incident laser radiation intensities q_0 in the range 10^6-10^7 W/cm^2. In this range of intensities the geometric-optics approximation for the radiation field can be used [4, 5], and therefore in order to find the temperature profile and the profile of the optical radiation flux density q in this work we used the system of equations

$$\begin{cases} \dfrac{\partial T}{\partial t} = \operatorname{div}\left(\dfrac{\varkappa(T)}{C}\nabla T\right) + \dfrac{1}{C}\kappa(T)\,q - I\dfrac{d\alpha}{dt}, \\ \dfrac{dq}{dx} = -\kappa(T)\,q, \end{cases} \qquad (1)$$

where I is the temperature effect of the endothermal chemical decomposition reaction of silica. For dielectrics the temperature dependence of κ and k can be represented in the form

$$\varkappa(T) = \varkappa_p + \varkappa_0 \exp(-E_1/T) \text{ and } \kappa(T) = \kappa_l + \kappa_0 \exp(-E_2/T),$$

where $\varkappa_p$, $\varkappa_0$, κ_l, and κ_0 are constants of the material, whose numerical values are taken from [4].

Nonlinear absorption could be associated with the thermal population of the energy band of free states (free–free transitions for absorption of the incident radiation) or levels from which a resonance transition to an upper-lying level followed by nonradiative dissipation of the excitation energy (bound–free and bound–bound transitions) can occur. In the general case $E_1 \neq E_2$, but in this work we neglect this difference which is unimportant in our case.

In models of radiation-absorption waves in transparent dielectrics used thus far the nonlinear part of the coefficient of thermal diffusivity is associated with the free electrons and (or) holes. But heat can also be transferred by means of diffusion of atoms and ions, when the temperature gradient gives rise to directed motion of ions.

$$\mathbf{J} = -\frac{D_T}{T}\nabla T,$$

where D_T is the coefficient of thermal diffusion. In this case, since to coefficient of diffusion depends exponentially on the temperature (the concentration of mobile ions and their mobility vary in an activation manner), nonlinear thermal conductivity is also observed. Using the Nernst–Einstein equation for ions, a relationship of the Wiedeman–franz law type $\varkappa =$

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