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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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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

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

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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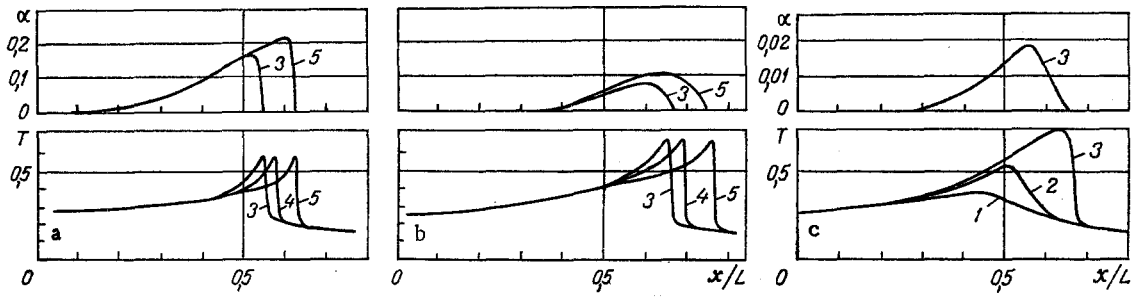

Fig. 1. Structure of the absorption and dissociation waves with q_0 = 3.1 MW/cm²; $C_p = 1.5 \cdot 10^5$ (a); $1.5 \cdot 10^4$ (b), and $1.5 \cdot 10^3$ (c); 1-5) t = 77; 102; 179; 205; 256 μsec, respectively. T, 10^4°K.

$\frac{3}{4}\left(\frac{k}{ze}\right)^2 U\sigma$ can be obtained between the thermal conductivity and the ionic conductivity σ, and in addition the bond rupture energy U plays the role of the temperature. This dependence holds near the maximum of the energy liberation function, where the temperature gradient is relatively small, for silicate melts we know only of the work [6], in which a direct proportionality between the thermal conductivity and the cationic conductivity was observed experimentally. Estimates show that at T ~ 4000-5000K in multicomponent silicate glasses the "ionic" thermal conductivity can reach, in order of magnitude the values of 10^{-2}-10^{-1} W/cm•deg, required for explaining the experimental data [1-3]. This type of thermal conductivity is very likely to occur in fused silicate glasses, which exhibit cationic conductivity with a very small electronic component.

We point out that although there is no experimental proof for the predominant action of any of the mechanisms of thermal conductivity — electronic or ionic — in multicomponent glasses at T ~ 5000 K, in any case, the coefficient of thermal conductivity is an exponential function of the temperature.

We shall study the motion of the absorption wave in the volume of the glass. The pressure in the region occupied by theoptical discharge is ~10^4 atm. At temperatures below the critical temperature (for silicate glass T_{cr} ~ 5000°K) the medium is in a condensed state, and above T_{cr} the glass transforms into a dense nonideal plasma in which a chemical reaction of the decomposition type $SiO_2 \rightleftarrows SiO + 1/2\ O_2$ occurs [7]. For the temperatures and concentrations of reacting particles characteristic for an absorption wave in transparent dielectrics the vibrational excitation rapidly relaxes into translational energy and the energy extracted from the laser radiation causes uniform heating, i.e., the thermal mechanism of the process is realized, and the reaction rate depends on the local temperature. This is not inconsistent with the fact that in the case of a traveling discharge in a condensed medium the temperature distribution is nonuniform over the volume of the reacting system. For an isolated system in which a chemical reaction occurs the problem is determined by a small number of thermodynamic parameters: the temperature, the pressure, the volume, and the chemical-equilibrium constant. In the case when there is an external source, when heat is transferred or work is performed, the reacting system is not in thermodynamic equilibrium and the kinetic equation must be used to describe its evolution.

In order to take into account the chemical decomposition of SiO_2 molecules in fused glass and in the dense plasma we shall use the model equation of chemical kinetics.

$$\frac{d\alpha}{dt} = -C_p[\alpha^3 - (1-\alpha)^2 K_p]. \qquad (2)$$

Here α is the degree of dissociation, defined as $\alpha = 1 - N_{SiO_2}/N^\circ_{SiO_2}$, where N_{SiO_2} and $N^\circ SiO_2$ are the instantaneous and initial concentrations of the structural elements of SiO_2; $K_p = AT^{-3/2}\exp(-E_\alpha/T)$ is the equilibrium reaction constant.

The theory of chemical kinetics in nonideal gases and plasma with strong interparticle interaction is at the present time far from complete, and in practice the quantities K_p and C_p are parameters which must be found from experiment; in addition, it should be noted that a stronger dependence of the parameter C_p on the density of the medium should be expected in an isothermal process.

The initial and boundary conditions (cylindrical symmetry of the problem) are imposed at the bounded region of the dielectric heated by focused laser radiation:

$$T(x, r, 0) = \frac{T_0 \exp(-r^2/a_0^2)}{1 + x^2/L^2}; \quad q(x_b) = q_0 \exp(-r^2/a_0^2), \tag{3}$$

where q_0 is the power density of the laser beam incident on the sample and x_b is the position of the boundary of the sample.

The system of equations (1)-(3) was solved numerically. The equations (1) were solved separately, and the change in the temperature owing to the effects of thermal conductivity and absorption of the laser radiation in some time interval Δt was determined. Then in this interval Δt the kinetic equation (2) and the contribution of $Id\alpha/dt$ to the temperature were calculated. The overall scheme is constructed based on the principle of componentwise splitting and is of first-order accuracy in time and second-order accuracy in the spatial coordinates.

Figure 1 shows the structure of the one-dimensional absorption wave as a function of the ratio of C_p and the inverse transit time of the absorption wave $t_v = \chi/v^2$ (v is the velocity of the absorption wave), while Fig. 2 shows the isotherms of the temperature field of the absorption wave at different times. The values of the parameters $A = 2 \cdot 10^7$; $E_\alpha = 2.7 \cdot 10^4$ and $I = E_\alpha$ are chosen by fitting the computed temperature profiles to the experimental profiles (see item 2 below).

We shall study different cases of the ratios of C_p and t_v:

a) For $C_p \ll 1/t_v$ the dissociation wave develops behind the front of the absorption wave, whose velocity is independent of the value of C_p;

b) For $C_p \lesssim 1/t_v$ the dissociation wave overtakes the absorption wave, and when $C_p \sim 0.1/t_v$ the equilibrium degree of dissociation is reached at distances of 50-100 μm behind the absorption front; the temperature drop owing to dissociation forms the characteristic peak behind the absorption front, whose width at half-height is equal to 15-25 μm;

c) when $C_p > 1/t_v$ the front of the dissociation wave moves synchronously with the front of the absorption wave, whose velocity depends strongly on C_p.

Comparison of the calculations based on the dissociation model with the experimental facts [1-3] shows the following.

1. The recombination rate constant for silicate glass $C_p \sim 1 \cdot 10^5$ sec^{-1}. For higher values of C_p the computed dependence of the velocity of the absorption wave on the radiation intensity does not agree with the experimental dependence.

2. When $C_p \sim 1.10^5$ sec^{-1} the temperature peak behind the front becomes smeared within a characteristic time $\tau_x \lesssim 10$ μsec. In the experiment the surface layer of the optical discharge decays in less than 10 μsec. The experimental values of the temperatures of the surface layer (6600°K) and of the region lying behind it (4500°K) are in agreement with the computed values (see Fig. 1b) with the selected values of the constants A and E_α.

3. The computed temperature of the plateaulike section of the interior region with $C_p \sim 10^5$ sec^{-1} is virtually independent of the incident radiation intensity. No such dependence on the laser intensity was observed in the experiments.

4. The characteristic thickness of the layer in the calculations — 15-25 μm — is in satisfactory agreement with the experimental thickness of the strata — 30 μm.

5. The overall contour of the absorption wave (Fig. 2) is in good agreement with the experimental form of the "head" part of the optical discharge [1]. In order to describe more accurately the experimental form of the optical discharge in glasses, where aside from the head part propagating toward the laser radiation there is also a narrow part moving in the direction of propagation of the laser beam, the hydrodynamic flow processes forming a cumulative jet in a conical geometry of the optical discharge must be taken into account. The irregular nature of the surface layer of the optical discharge with a characteristic distance of ~30 μm between the strata cannot be explained on the basis of the model studied in this work. The strata apparently appear because of the instability of the hydrodynamic flow of matter from the surface into the volume of the optical discharge — an instability of the Benard effect type [8]. In this case it may be expected that the characteristic scale of the instability will coincide in order of magnitude with the thickness of the temperature peak behind the front of the absorption wave.

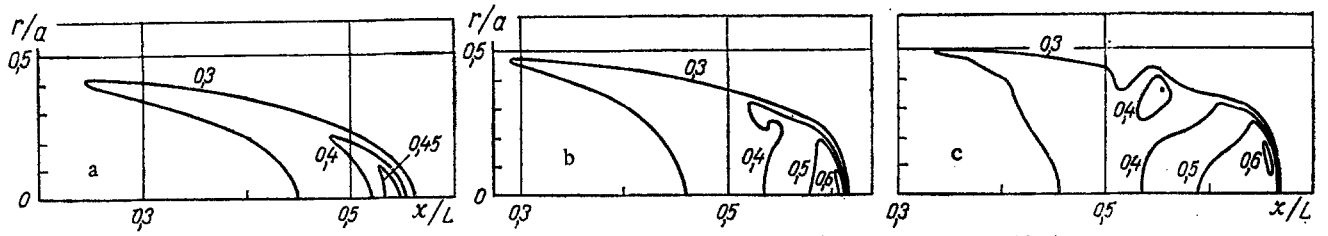

Fig. 2. Structure of the isotherms of the absorption wave (q_0 = 3.1 MW/cm^2; $C_p = 5 \cdot 10^4$ sec^{-1}): a) t = 256 μsec; b) 384; c) 512. The numbers of the isotherms are the values of the temperature in 10^4°K.

NOTATION

k, Boltzmann's constant; ze, ion charge; C, heat capacity of the medium; T, absolute temperature; a_0, radius of the neck of the caustic; L, length of the focal region of the focusing system; $\varkappa(T)$, coefficient of thermal conductivity of the medium; $\kappa(T)$, coefficient of absorption of optical radiation; and $\chi(T)$, coefficient of thermal diffusivity.

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