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Laser induced transparency during nanosecond laser ablation

S.N. Andreev^{*a}, A.A. Samokhin^a, I.N. Kartashov^b, V.I. Mazhukin^c, D. Grevey^d

^aGeneral Physics Institute RAS, Moscow, Russia

^bMoscow State University, Moscow, Russia

^cIMM RAS, Moscow, Russia

^dLab. Traitement des Materiaux, France

ABSTRACT

Steady state laser vaporization regime of condensed matter is investigated in the case of laser-induced transparency in irradiated target. Vaporization front stability problem is also considered taking into account bulk absorption in the target and different Mach number in evaporation plume.

Keywords: laser ablation, phase transition, superheated liquid, explosive boiling

1. INTRODUCTION

For overwhelming majority of metals, at temperatures not exceeding critical temperature, the equilibrium degree of vapor ionization is rather small because of great value of ionization potential in comparison with critical temperature T_c . That is why at quaziequilibrium evaporation of metals liquid-vapor phase transition is accompanied by metal-dielectric transition.

At increase of temperature the density of metal decreases and metal-dielectric transition can take place, in principle, in condensed phase at temperature $T_{md} < T_c$. Obviously, such transition will be accompanied by abrupt reduction of the absorption coefficient of optical radiation¹.

Such laser induced transparency (LIT) was discussed^{2, 3, 4} in connection with results, obtained in work², where the action of millisecond pulses of neodim laser on lead and bismuth targets were investigated.

The problem of the appearance of LIT in experiments² is stayed unclear due to some causes (space-temporal heterogeneity of laser pulse, absence of data about recoil pressure behavior during irradiation of the target, etc.) that complicate interpretation of experimental results². As far as we know, experimental results for millisecond laser action of a similar sort were not reported any more.

In recent paper⁵, at action of laser radiation ($\lambda = 266$ nm, $I_0 = 10^9 - 10^{11}$ W/cm²) on silicon, it was found abrupt (more than one order of magnitude) increase of crater depth at intensity $I > 2.2 \cdot 10^{10}$ W/cm². The most part of mass removal takes place ~ 300 ns later after laser pulse with duration $\tau = 3$ ns.

This effect was interpreted by authors⁵ as LIT in the irradiated target providing deep penetration of laser radiation and then explosive boiling of overheated liquid. Some aspects of this interpretation were discussed also^{6, 7}.

The abrupt increasing of crater depth (ten times) observed in work⁵ is a serious argument for the benefit of the interpretations offered by the authors⁵. In this connection more detailed theoretical research of features of a laser evaporation regime with LIT in irradiated substance is of interest because their experimental observation permits to get additional information about processes in irradiated substance. This is the main purpose of our paper.

* E-mail: andreev@nccom.ru

2. ANALYSIS

Schematic diagram of the target during laser-induced transparency is shown in Fig. 1. In work ⁵ it is actually supposed that when the transparency temperature T_b is reached the absorption factor in transparent media becomes vanishingly small.

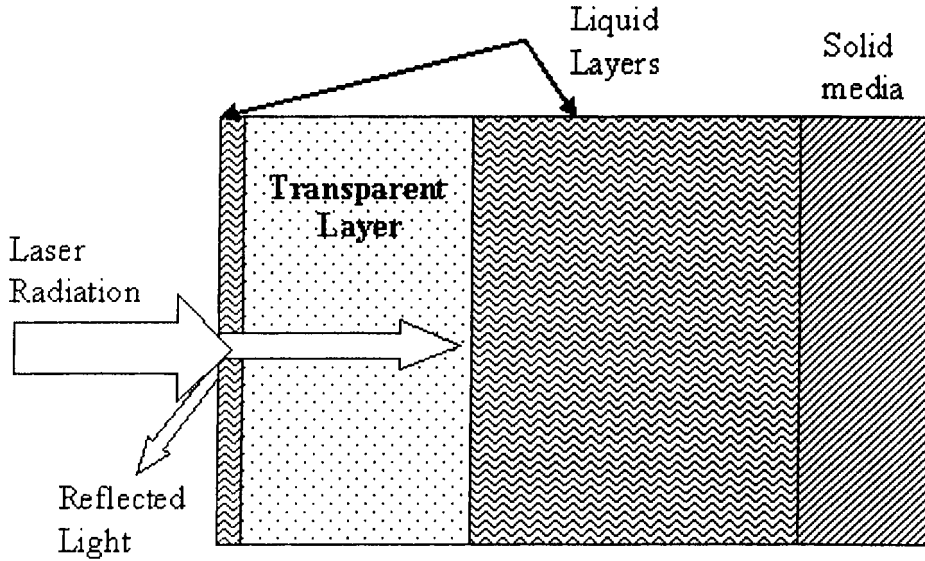


Figure 1: Schematic diagram of the target during laser-induced transparency.

In that case, usual stationary vaporization regime with time independent temperature profile actually can not be reached in one-dimensional model, because, the transparency front can penetrate infinitely deep into substance because of absence of absorption in the transparent layer. If laser intensity is great enough the transparency front velocity v_t can be a lot more greater than vaporization front velocity v , which remains a constant due to laser radiation absorption in the thin non-transparent layer, directly adjoining to vaporization boundary. The thickness of this layer can be defined using the formula:

$$h = \frac{l}{\alpha - q} \ln \left(\frac{\alpha I}{\alpha l - (\alpha - q) \rho L_v v} \right), \quad (1)$$

where $q = v/\chi$, χ – temperature conductivity, $I = \eta(1-R)I_0$ – absorbed laser intensity, R – reflectivity factor of media and factor $\eta < 1$ describes weakening of laser radiation inside the near-surface plasma plume. Note, that factor R is various for the transparent and non-transparent media and depends on position of transparent layer boundaries.

The formula (1) can be obtained from solving of stationary heat transfer equation in evaporation front frame with Bouguer absorption and evaporation boundary condition $\chi(\partial T/\partial z) = vL_v/(cp)$ on free surface with temperature T_s , where c – specific heat, L_v – latent evaporation heat, $T = T_b$ and $(\partial T/\partial z) = 0$ are conditions on transparency boundary.

Evaporation front velocity v and vaporization pressure p_v are given by formulas:

$$v = \sqrt{\frac{m}{2\pi k T_s} \frac{p_s}{\rho} (1 - r^8)}, \quad p_v = \frac{p_s}{2} (1 + r^9), \quad (2)$$

where $\rho = 2.52 \text{ g/cm}^3$ – mass density of silicon, k – Boltzmann constant, $m = 4.67 \cdot 10^{-23} \text{ g}$ – mass of molecule of silicon and value r - depends on Mach number M in vapor plume. At $M=1$ value r is equal to 0.78 and $p_v = 0.56 p_s$. Saturation pressure p_s is approximated by formula

$$p_s = p_0 \exp(A(1 - T_v / T_s)), \quad (3)$$

where $T_v = 3076 \text{ K}$ – boiling temperature of silicon at normal pressure $p_0 = 1 \text{ Bar}$ and $A = 13.2$.

At rather great values of laser intensity I the thickness of non-transparent near-surface layer becomes too small for macroscopic description. Nevertheless, absorption inside the near-surface layer is necessary to support this regime, because, temperature profile $T = \text{const}$ inside the transparent layer does not provide the heat supply necessary for maintenance this vaporization regime. In work⁵ absorption inside near-surface non-transparent layer is taken into account in general equation of energy balance (eq. (7)).

At stationary vaporization regime without LIT, vaporization pressure contribution to recoil pressure, dependent from surface temperature T_s , appears significantly greater than the contribution from change of density of the heated layer of the condensed substance, which can be estimated under the formula $\Delta p \sim v^2 \Delta \rho$, where $\Delta \rho = (\rho_\infty - \rho(T_s))$ the difference between densities on a surface and in depth of substance⁹. In the case of LIT at sufficiently great values of laser intensity, the transparency front and connected melting front can move at non-stationary stage with velocity v_1 , much greater than evaporation front velocity v . Transparency front velocity, which can be estimated⁵ as crater depth $H = 30 \text{ }\mu\text{m}$ divided by laser pulse duration $\tau = 3 \text{ ns}$, is of order of sound velocity in condensed matter $v_1 \sim 10^6 \text{ cm/s}$, i.e. additional pressure Δp can exceed vaporization pressure even at rather small $\Delta \rho$ ⁹.

Vaporization pressure as function of time, calculated by the code LasteC 1.1¹⁰, with parameters of silicon from work⁵ and laser intensity $I = 1.6 \cdot 10^9 \text{ W/cm}^2$ is shown in Fig. 2. The change in absorbed intensity in the beginning of the pulse is due to reflection factor change after the melting. As follows from calculations, when the surface temperature reaches value T_b the vaporization pressure ceases to grow and remains practically constant $p \sim 3000 \text{ Bar}$ despite of continued growth of laser intensity. Pressure begins to decrease after $\sim 2 \text{ ns}$ after a maximum of a laser pulse. Dependence on time of a full signal of the pressure including vaporization pressure and additional pressure, connected with change of density at heating in condensed matter is shown on Fig. 3. Additional pressure, which is due to fast movement of transparency front, includes positive and negative parts with maximum amplitude $\sim 1000 \text{ Bar}$.

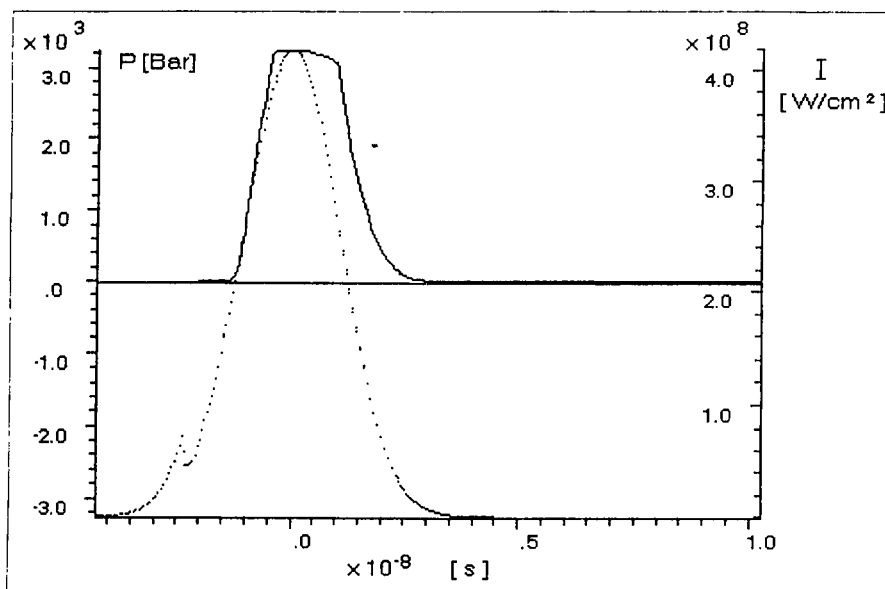


Figure 2: Vaporization pressure (solid line) and absorbed intensity (dotted line) as function of time.

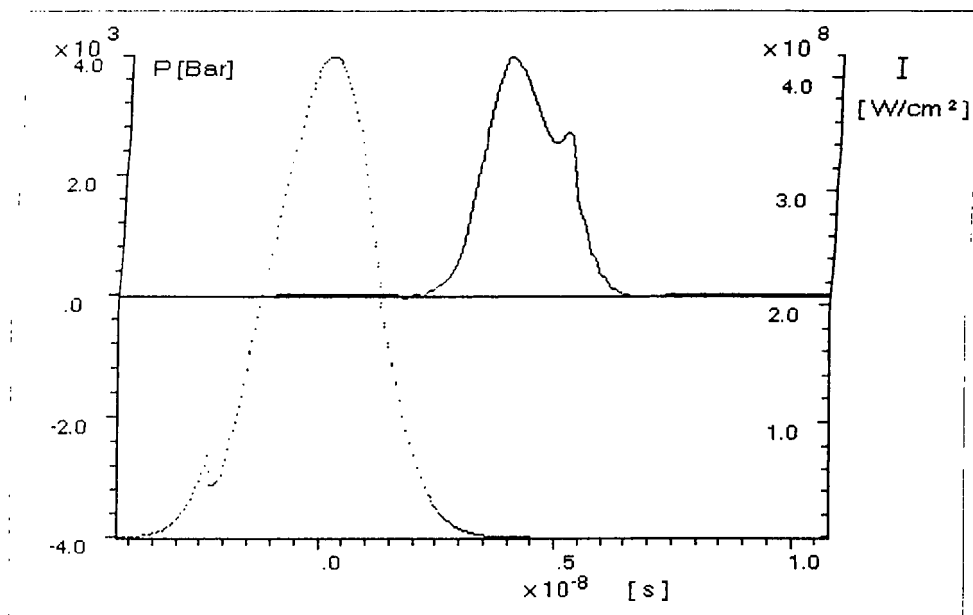


Figure 3: Full pressure signal (solid line) and absorbed intensity (dotted line) as function of time.

The value $\Delta\rho$ can be positive or negative depending on a proportion of contributions from change of density at thermal expansion and melting, because after melting silicon becomes metallic and its density increases. Such a metallization of silicon gives, in principle, opportunity to register the melting front velocity with the help of probing infra-red radiation, which goes on front of melting from back side of the target and than is reflected from it.

Using the results of experiment⁵ it is possible to estimate the upper limit of absorption factor α in the transparent media. Obviously, the maximum value of absorption factor α can not exceed inverse value of crater depth H , i.e. $\alpha H < 1$. At $H=20 \mu\text{m}$, the value of absorption coefficient is $\alpha < 500 \text{ cm}^{-1}$. On the other hand, during action of a laser pulse τ temperature change of the transparent layer $\Delta T \sim \alpha I \tau / (\rho c)$ should not exceed $T_1 - T_b = 0.2 T_1$, where T_1 - limit overheating temperature, which is less than critical temperature T_c , (usually, $T_1 - T_c \leq 0.1 T_c$)¹¹. Such estimation is based on the assumption, that lifetime of metastable state at T_1 is much less than duration of a laser pulse. At the values of absorbed laser intensity $I \geq 10^{10} \text{ W/cm}^2$, specific heat $c \sim 1 \text{ J/(g K)}$, $\Delta T = 0.2 T_c = 1600 \text{ K}$, one can find $\alpha < 130 \text{ cm}^{-1}$. It is supposed that $T_b = 0.8 T_c$ and $T_c = 8000 \text{ K}$.

At nonzero values α velocity of transparency front will decrease and becomes equal to the velocity of vaporization front. Thus, the temperature profile will not depend on time in accompanying reference frame. Assuming that thermophysical parameters are constants in transparent and non-transparent layers, one can find temperature profile from stationary heat transfer equation

$$-v \frac{\partial T}{\partial z} = \chi \frac{\partial^2 T}{\partial z^2} + \frac{\alpha I}{c\rho} \exp(-\alpha z) \quad (4)$$

inside each layers, solution in which are sewed on their boundaries by conditions of a continuity of temperatures and heat flows, and on free boundary it is used vaporization boundary condition^{3,12}.

Characteristic temperature profile at $\alpha = 100 \text{ cm}^{-1}$ and $I = 3.197 \cdot 10^7 \text{ W/cm}^2$ is shown in Fig. 4. In contrast to model considered in work⁵, where the temperature of transparent layer is constant, in this case the maximum of temperature takes place inside the transparent layer. Value and position of the temperature maximum depend on parameters of

media. At laser intensity $I_1=3.1964 \cdot 10^7 \text{ W/cm}^2$ the depth of transparent layer H is equal to zero, whereas the depth of non-transparent layer is maximal $h=0.84 \cdot 10^{-6} \text{ cm}$, at the same time the temperature maximum coincides with temperature of transparency T_b . With growth of laser intensity I the depth H is increased, and h tends to zero. At intensity $I_1=3.1976 \cdot 10^7 \text{ W/cm}^2$ the maximum of a temperature profile reaches T_c , while $h=0.7 \cdot 10^{-6} \text{ cm}$ and $H=20 \text{ }\mu\text{m}$. Disappearance of subsurface non-transparent layer occurs at $I_1=3.273 \cdot 10^7 \text{ W/cm}^2$, at which the temperature profile maximum exceeds value of critical temperature in 2.3 times, that leaves the framework of applicability of model with constant factors.

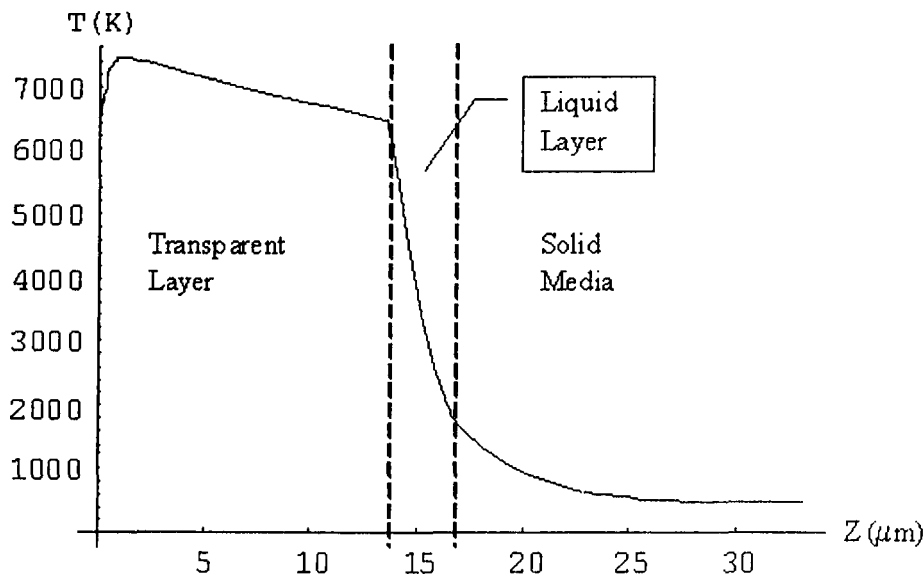


Figure 4: Stationary temperature profile at presence of laser-induced transparency in irradiated substance.

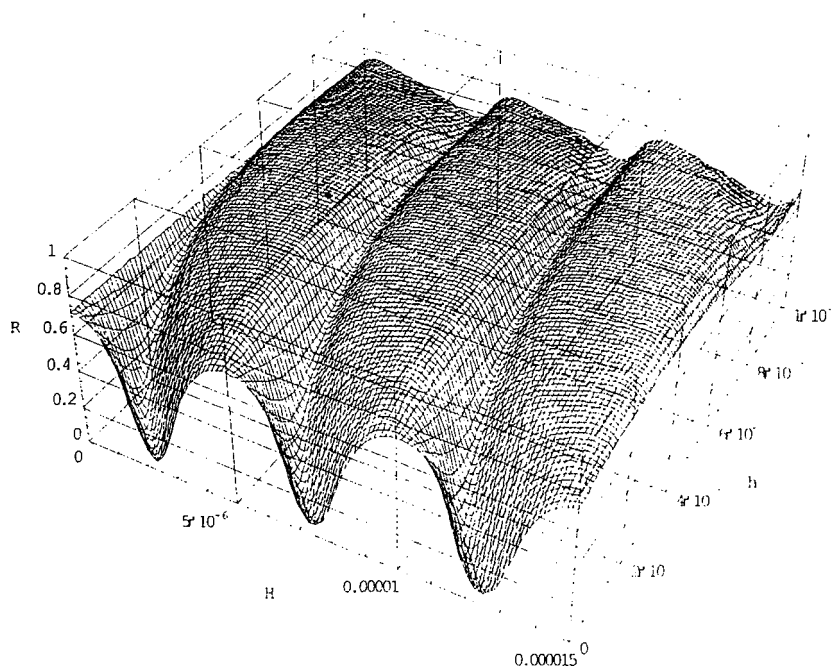


Figure 5: Reflection factor dependence on thickness of transparent H and non-transparent h layers.

Such narrow interval of intensity in this case is due to significant modification of temperature distribution when α decreases during LIT and relatively small temperature interval between T_b and T_c .

At sufficient stability of metastable phase, for the description of its behavior it is possible to use model of surface evaporation taking into account special features of behavior of heat conductivity and a thermal capacity close to spinodal¹³. Otherwise the model of periodic boiling up¹⁴ can be realized.

It is necessary to note, that for maintenance of a stationary regime of evaporation considerably smaller values of laser intensity, than used in work⁵ are required. At such rather small intensity the plasma plume can not arise, i.e. $\eta=1$ and difference between incident and absorbed intensity values is defined only by reflection factor.

If boundaries of transition metal-dielectric are sharp enough and the condensed phase is stable enough, occurrence of LIT will be accompanied by the phenomena of interference typical for a thin-film situation. Fig. 5 shows dependence of reflection factor on thickness of transparent H and non-transparent h layers for wave length $\lambda=266$ nm. Real and imaginary parts of factor of refraction of transparent and non-transparent media are $n=2.62$, $\xi=0.0002$, $n_0=11.7$, $\xi_0=2.39$, that corresponds to values of absorption $\alpha=100$ cm⁻¹, $\alpha_0=1.13 \cdot 10^6$ cm⁻¹ and reflection $R=0.2$, $R_0=0.72$ factors of semi-infinite transparent and non-transparent media.

In case of small excess of intensity over value I_1 , the transparent layer arises on depth of the order of $h=0.8 \cdot 10^{-6}$ cm. Depending on thickness of the transparent layer, the factor of reflection changes in a range from 0.65 up to 0.84. In a stationary regime the factor of reflection as well as other parameters of media, depend only on laser intensity. In a non-stationary case additional explicit dependence on time occurs.

Obviously, presence of a plasma plume can interfere with direct observation of such a behavior of factor of reflection. From this point of view it is preferable to use near stationary regimes of evaporation with LIT, for which are required smaller, than in work⁵, values of laser intensity.

Plasma formation time dependence on incident laser intensity for different initial temperatures and concentrations of the vapor on external boundary of Knudsen layer, calculated by code described in work¹⁵, is shown in Fig. 6. At Mach number $M=1$ the temperature and concentration of the vapor on external boundary of Knudsen layer are expressed from surface temperature T_s and saturated vapor concentration $n_s = p_s / (k T_s)$ as $0.63 T_s$ and $0.32 n_s$. Fig. 6 shows, that threshold value of laser intensity for plasma formation exceeds significantly intensities characteristic for stationary regime. That is why plasma can not arise in the considered stationary regime of laser vaporization.

The assumption of existence of one-dimensional stationary regime of evaporation at presence of LIT is connected not only to a question of thermodynamic stability of a overheated metastable phase, but also with a problem of morphological stability of flat front of evaporation^{9,16}.

In case of bulk absorption of laser radiation with constant factor $\alpha = 100$ cm⁻¹ and at free evaporation to the vacuum¹⁶, the solution of the dispersion equation for small perturbation of flat evaporation front (at $T_s = 6400$ K and other parameters from⁵) gives the maximum value of increment $\gamma_{max} \sim 1$ ns⁻¹ at $k_{max} \sim 20$ μm^{-1} . Such values of k correspond to wavelengths, which are small, in comparison with distance from a surface up to a temperature maximum position, i.e. in this case, features of behavior of temperature near to a maximum do not affect significantly increment behavior. Presence of the non transparent layer near to a surface of evaporation most likely will not increase presented values of γ_{max} , though this question demands additional research.

If evaporation is not free, for example, because of occurrence of absorbing plasma near to an irradiated surface, then Mach number of the vapor plume on external boundary of Knudsen layer can be less than unity. As the Mach number is decreased from unity to zero, the maximum of increment increases, reaches the maximum value $\gamma_{max} = 1.5$ ns⁻¹ at the point $k_{max} = 32$ μm^{-1} and then tends to zero when $M \rightarrow 0$. The corresponding value of k_{max} then moves to long wavelength region. Values of γ_{max} and k_{max} as function of Mach number are presented in Fig. 7.

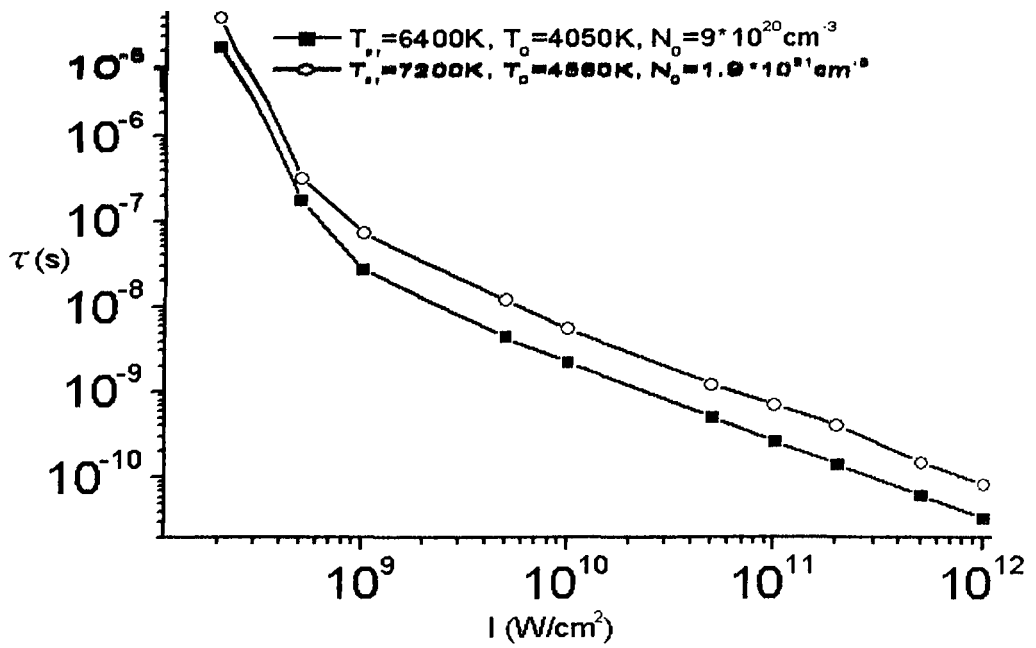


Figure 6: Plasma formation time dependence on incident laser intensity.

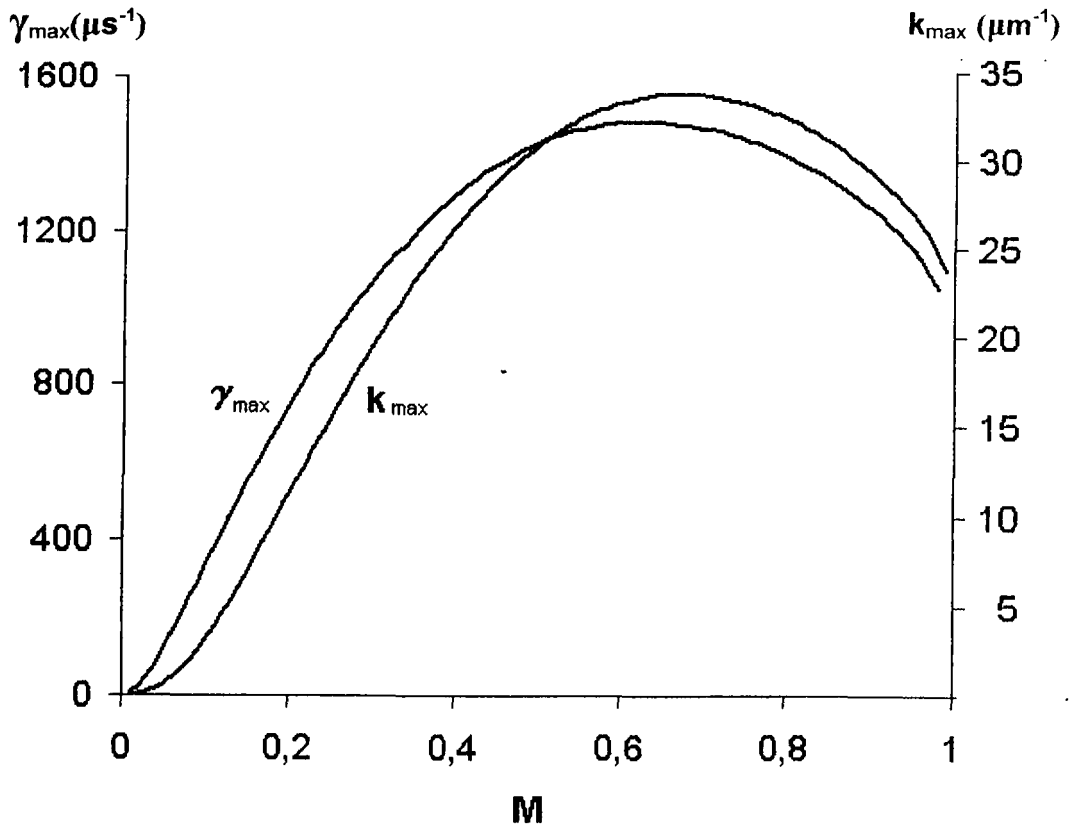


Figure 7: γ_{\max} and k_{\max} as function of Mach number.

3. CONCLUSION

Thus, if the metastable transparent liquid is sufficiently stable, the stationary regime of developed vaporization with the flat front can exist on times of order of 10^{-8} s. Change of factor of reflection at occurrence of transparent layer is not so expressed, even if the plasma plume is not presented. Induced transparency can manifest itself more essentially in photoacoustic effect, which is usually registered by the gauge of pressure on the back side of a target⁹. The increase of pressure connected to the beginning of explosive boiling can be registered by the same way.

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