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Numerical Simulation of Plume Dynamics in Pulsed Laser Ablation for Deposition of Diamond-Like Carbon Films

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Pulsed laser deposition (PLD) is not a new way of forming high-quality thin coatings materials with extraordinary characteristics [1]. PLD is fairly straightforward. A high-intensity, pulsed laser beam is focussed on a target in a chamber that is either evacuated or filled with a specific gas. The laser beam causes the target material to vaporize (or ablate) into the chamber. A substrate to be coated is placed in the path of the laser-produced plume, and the vapor clings to its surface, forming a thin layer of the ablated target material. It is possible to build films of specific thickness by using appropriate characteristics of laser pulses (laser wavelength, laser irradiance, pulse length, gas environment, etc.).
PLD can produce diamond-like coatings that make a surface nearly diamond hard, or high-temperature superconducting films that may pave the way for practical superconducting devices. It has the highest instantaneous deposition rate among all other known deposition methods (such as electron-beam deposition or magnetron sputtering).

Dynamics of a carbon ablation plasma plume when preparing diamond-like carbon films by pulsed laser deposition was investigated using computational simulation. Numerical simulation results on dynamics of a plume under the action of Nd:YAG laser pulses (duration ~50 ns) with laser irradiance in maximum ~10-100 W/cm^2 are given.

The models for laser–solid interaction [2] are based on thermal processes: heating of a solid, followed by melting and evaporation. They describe the laser–solid interaction on a macroscopic scale, i.e. by the heat conduction equation. This assumption is justified for ns-pulsed laser interaction, especially for metals. The plume was investigated by the hydrodynamic model for the case of expansion in a low pressure background gas (up to a few 10^{-6} atm). For computer simulation the data on the equations of state and thermodynamic functions, thermal conductivity and optical characteristics are necessary. The equation of state for carbon plasma is calculated and tabulated in the ranges of temperatures $T = 10^3 - 10^6$ K and densities $\rho = 10^{-9} - 10^{-2}$ g/cm^3. The composition of plasma representing an intermixture of molecules, atoms and ions of different-degree ionization is defined according to “the chemical model” with usage of the relevant equilibrium constants. The optical characteristics (absorption coefficients for laser radiation) are determined taking into account the main mechanisms of laser radiation absorption in the plasma (bremsstrahlung, photoionization and selective absorption) [3].

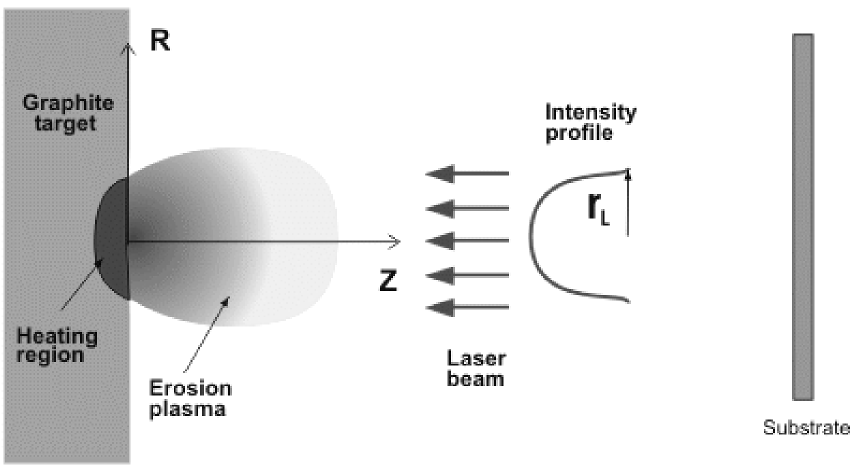

Figure 1. The geometrical scheme of pulsed laser deposition.

Taking into account the fact that beams acting on the surface have, as a rule, an axial symmetry, we will consider the corresponding system of equations that describe the dynamics of action, in two-dimensional cylindrical geometry (Fig.1). The laser pulse is characterized by the radiation wavelength λ_L, the diameter of the irradiation spot r_L, the distribution of the power density over the spot, and by the time pulse shape $q(r,t)$. The laser beam propagates along the symmetry axis z.

The dynamics of the target heating is described by the nonstationary heat conduction equation

$$\frac{\partial \rho_c c_p T}{\partial t} + \operatorname{div} \vec{W} = 0, \qquad \vec{W} = -\chi \cdot gradT ,$$

where T is the temperature; $\vec{W}$ is the heat flux; ρ_c is the density of the condensed substance; c_p is the specific heat; χ is the thermal conductivity coefficient. The boundary condition on the surface (the interface of the condensed and gaseous phase) for the heat flux component z has the form

$$W_Z(r, z=0, t) = \begin{cases} -\chi \left.\dfrac{\partial T}{\partial z}\right|_{z=0} = (1-R)\cdot q(r,t) & r < r_L \\ 0 & r > r_L \end{cases}$$

where R is the reflection coefficient. The initial temperature of the entire region is constant and equal to T_0. The propagation of the front of melting and crystallization is described by the Stefan condition

$$\rho_c \lambda_m V_m = W_l - W_s = \chi_s \left.\frac{\partial T}{\partial x}\right|_{+} - \chi_l \left.\frac{\partial T}{\partial x}\right|_{-}, \qquad T(x = x_m, t) = T_m$$

where x_m is the coordinate of the melting front; T_m is the melting point; V_m is the velocity of melting (crystallization) of a wave; W_l and W_s are the heat fluxes at the melting front on the side of the melted and crystalline phases.

From the onset of evaporation (when the pressure of saturated vapors at the surface temperature exceeds that of the ambient atmosphere) the density of the laser radiation flux, absorbed by the surface, decreases due to evaporation expenditures. Then the boundary condition on the target surface takes the form

$$W_z(r, z=0, t) = (1-R)\cdot q(r,t) - \rho V_{sub} \Delta H$$

where V_{sub} is the propagation velocity of the evaporation front in the depth of the target; ΔH is the specific enthalpy of evaporation of the target material. The evaporation velocity of the wave is determined from the solution of the external gas dynamics problem and conditions for conservation of mass fluxes, momentum, and energy in a Knudsen layer.

The gas dynamics problem. The products of damage scattering from the surface form an erosion laser torch in the environment. The expansion of the vapors of the target material leads to their cooling, whereas the absorption of incident radiation increases their heat energy. The competition of these mechanisms depending on the action conditions (the parameters of the laser pulse, the thermophysical properties of the target and environment, the degree of transparency of vapors for laser radiation) determines the dynamics of formation and development of the erosion plume. At large spots of irradiation of the target or at the initial moments of the action the motion is of one-dimensional (plane) character.

The side spreading becomes noticeable. The motion of the vapors turns out to be two-dimensional. The dynamics of expansion of the axially symmetric laser plume in a cylindrical system of coordinates is described by a set of equations of conservation of mass, momentum, and energy

$$\frac{\partial\rho}{\partial t}+\mathrm{div}\left(\rho\vec{V}\right)=0\,;$$
$$\frac{\partial\rho u}{\partial t}+\mathrm{div}\left(\rho u\vec{V}\right)+\frac{\partial P}{\partial z}=0\,;$$
$$\frac{\partial\rho v}{\partial t}+\mathrm{div}\left(\rho v\vec{V}\right)+\frac{\partial P}{\partial r}=0\,;$$
$$\frac{\partial\rho E}{\partial t}+\mathrm{div}\left(\rho E\,\vec{V}\right)+\mathrm{div}\left(P\,\vec{V}\right)+\frac{\partial q_+}{\partial z}-\frac{\partial q_-}{\partial z}=0\,;$$

Here u and v are the azimuth and radial components of the velocity vector $\vec{V}$; P is the pressure; $E=\varepsilon+\vec{V}^2/2$ is the total specific energy of the plasma; ε is the internal energy. The intensity of the laser radiation directed to the target surface and reflected from it is determined by the expressions

$$q_-(r,z,t)=q(r,\infty,t)\cdot\exp\left(-\int_z^\infty \kappa_L\,dz'\right),\qquad q_+(r,z,t)=R(T_{sur})\cdot q_-(r,0,t)\cdot\exp\left(-\int_0^z \kappa_L\,dz'\right),$$

where κ_L is the absorption coefficient of the plasma at the laser radiation frequency; $R(T_{sur})$ is the reflection coefficient of the target surface which depends generally on its temperature. The system of gas dynamics equations is closed by the equations of state

$$T=T(\rho,\varepsilon)\,,\qquad P=P(\rho,\varepsilon)\,,$$

prescribed in table form. The absorption coefficients of the laser radiation required for calculation of

$$\kappa_L=\kappa_L(t,\rho,E_L),$$

are also prescribed in table form.

The boundary conditions on the target surface with the onset of evaporation (when the pressure of the saturated vapors of the target material $P_s=P_s(T_{sur})$ exceeds that of the ambient atmosphere P_0) are determined from consideration of a gas-kinetic problem in a Knudsen layer. The asymptotic (in the kinetic sense) values of the density and temperature obtained as a result of solving the kinetic equation are taken as boundary conditions to solve a problem of the gas flow in the hydrodynamic region. The relation of the parameters on the Knudsen layers boundary with surface temperature T_s and vapor density ρ_s is determined by the expressions:

$$\frac{T_g}{T_s}=\left[\sqrt{1+\pi\left(\frac{\gamma-1}{\gamma+1}\frac{m}{2}\right)^2}-\sqrt{\pi}\,\frac{\gamma-1}{\gamma+1}\frac{m}{2}\right]^2,\qquad P_g=\rho RT_g\,,\qquad m=\frac{V_g}{\sqrt{2kT_g/A}}\,,$$

$$\frac{\rho_g}{\rho_s}=\sqrt{\frac{T_s}{T_g}}\left[\left(m^2+\frac{1}{2}\right)\exp(m^2)\mathrm{erfc}(m)-\frac{m}{\sqrt{\pi}}\right]+\frac{1}{2}\frac{T_s}{T_g}\left[1-\sqrt{\pi}m\exp(m^2)\mathrm{erfc}(m)\right].$$

The subscript g corresponds to the values on the gasdynamic boundary of the Knudsen layer, s – on the phase interface; A is the atomic weight of target material vapors, $\rho_g=\rho_g(T_s)$ is the density of saturated vapors at the surface temperature, γ is the adiabatic exponent.

The evaporation velocity of the wave V_{sub}, propagating in the depth of the target over the condensed substance of density ρ_{sol}, is related to the condition for conservation of the mass flow of vapors entering into the external gasdynamic region $\rho_{sol}V_{sub}=\rho_g V_g$. Thus, the solution of the internal heat problem by means of boundary conditions is associated with boundary conditions and flow regimes in the external gasdynamic region.

The modeling was carried out for the conditions of experiments [4] (wavelength of 1064 nm, , 50 ns pulse duration half-width with a “tail” up to 250 ns, the ambient pressure of a gas equal to 10^{-6} atm). Among the main parameters, laser pulse energy and also power density have been changed in the study. A detailed study has been performed of a space-time development of the laser ablation plume on a graphite sample at the following conditions: 2 - 8 J laser pulse energy uniformly distributed with a 9 mm characteristic radius. It corresponds to maximum irradiance of 6.5 - 65 MW/cm^2. The time profile of the laser pulse (normalized to irradiance in maximum) is given in fig. 2. A substrate to be coated is placed opposite to the graphite target at a distance of 10 cm. The thermophysical properties and optical parameters of graphite (with a density of 2.2 g/cm^3) necessary for numerical simulation were set in view of the temperature dependence according to the literature data [5,6].

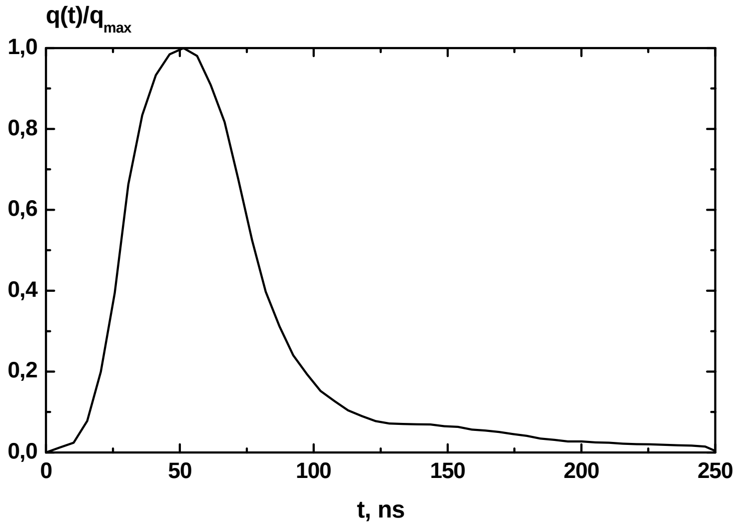

Fig.2 Laser intensity-time profile assumed in the model.

Dynamics of temperature of the graphite target and thickness of the evaporation layer for different laser energy is given in fig. 3. Estimated critical irradiance for the erosion is observed at ~20 MW/cm^2). For all investigated laser irradiance (20<q<70 MW/cm^2) the target evaporation comes to the end to the moment of time ~100 ns. Let us notice that for this irradiance the laser plume remains transparent and the shielding of the target is not observed.

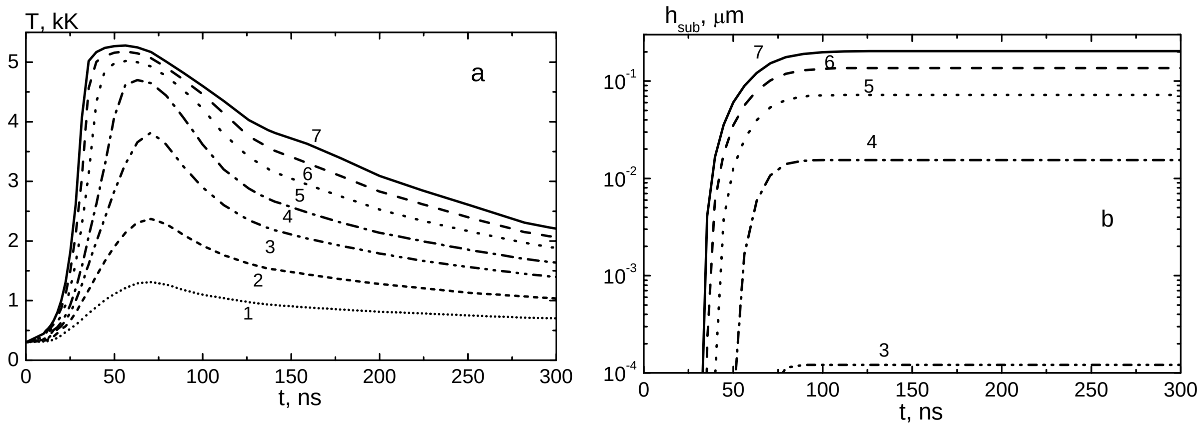

Fig. 3. Calculated temperature at the target surface (a) and evaporation depth (d) vs. time for pulse laser energy: 1 – 1 (6.5), 2 – 2 (13), 3 – 3 (20), 4 – 4 (26), 5 – 6 (40), 6 – 8 (52), 7 – 10 J (65.5 MW/cm^2). Maximum irradiance is given in brackets.

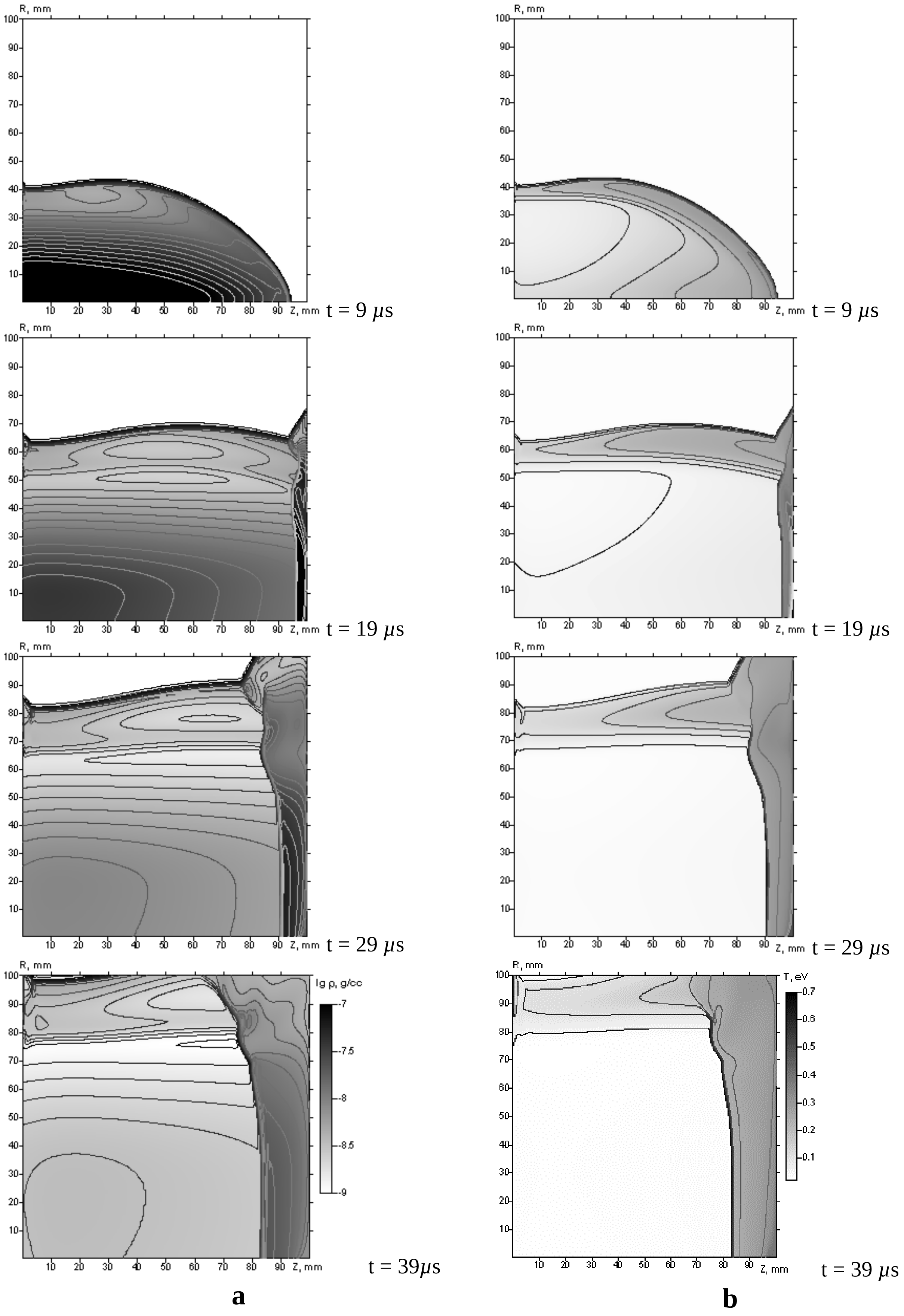

Fig. 4 Carbon plume expansion. Energy of the laser pulse 10 J. Distribution of the density (a) and temperature (b).

Dynamics of development of a laser plume is shown in fig. 4 for the energy laser impulse of 10 J (q_{max} = 65,5 MW/cm^2). Figs. 4a and 4b depict density and temperature profiles at times: 9, 19, 29,39 μs. At ~10μs, the expanding plasma front hits the substrate wall and reflects back. The reflected wave recompresses the carbon plume (see Fig. 5) and squeezes the plume causing the temperature rise to its maximum approximately at 10μs.

The time dependence of the plasma density (a) and temperature (b) close by substrate at different laser pulse energy is presented in fig. 4.

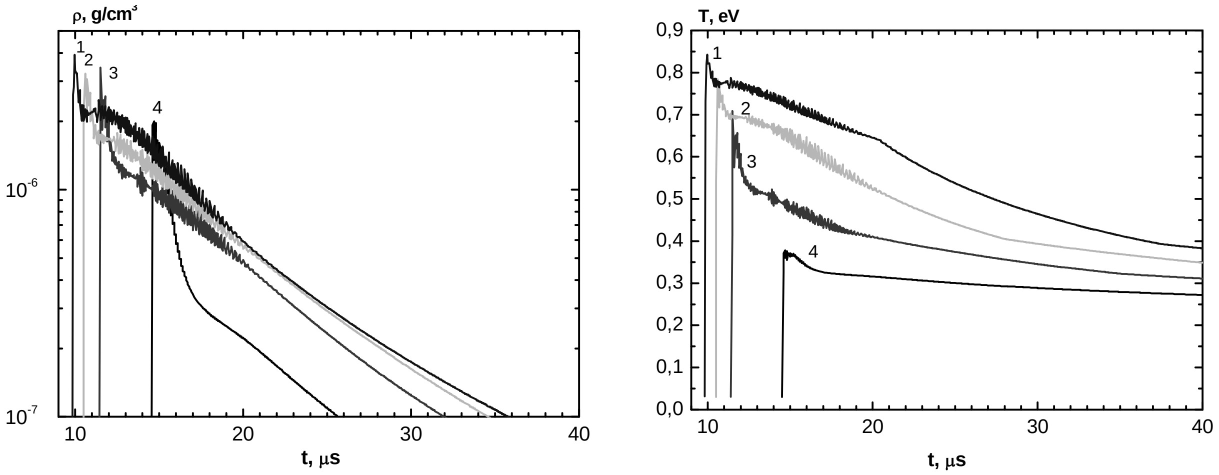

Fig. 5 Calculated carbon plasma density (a) and temperature near the substrate (b) vs. time for energy of the laser pulse: 1 – 10; 2 – 8; 3 –6; 4 – 4 J.

Numerical simulation enables one to receive the information on plume parameters at all stages of the process of pulsed laser deposition.

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