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A New Dynamics Expansion Mechanism for Plasma during Pulsed Laser Deposition *

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The dynamics expansion mechanisms for plasma plume generated by pulsed laser radiation are studied in detail, taking account of plasma ionization effect. Based on the consideration of local conservations of mass, momentum, collected as the assumption that plasma can be viewed as compressible ideal fluid and high temperature-high pressure ideal gas, we develop a new dynamics expansion mechanism for plasma produced by pulsed laser radiation. Using the analytical method, the space number density and pressure evolvement of plasma in cylindrical coordinate are obtained, the dynamics evolvement equations are also derived. The results from the present model indicate that the plasma dynamic expansion behaviour can be evidently influenced by the ionization fraction η. Its effect is similar to a new dynamic source for plasma expansion and increases the expansion acceleration in all directions. The predictions of the expansion of the plasma is affected by the temperature, the average atoms mass and the ionization degree of the plasma are consistent with the experimental results.

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In the last decades, plasma expansion processes during the pulsed laser deposition has received a great deal of attention because they are helpful for researchers to understand the natural mechanism of plasma expansion and to prepare more good-quality film materials [1−4] since the quality of deposited films is critically dependent on the range and profile of kinetic energy and density of the ablated plasma.[5−9] The simple one-dimensional hydrokinetics model was used to study the velocity evolvement at low laser energy fluence.[5−7] A two-dimensional distributions of the plume number density was investigated in detail by Ardehali *et al.*[7] and Nossov *et al.*[8] Singh *et al.*[4] developed a three-dimensional self-similar model(S-N model) of plasma flow in a chamber based on the whole conservations of mass and linear momentum. All these models were begun with an assumed self-similar profile of plasma velocity and considered the plasma velocity near the target surface irradiated by laser as zero. It can explain the plasma expansion preferentially normal to the target surface quantitatively, however, the assumption about velocity is not consistent with the experimental results.[10−12] that is, the velocity of the plasma near the target surface is not zero, but large. Thus Chen *et al.*[5] developed an accelerated expansion one-dimension model. For studying the laser radiation effect, an assumed profile of plume velocity

$$v = v_m[a + (1 - a)\xi], \tag{1}$$

was used. Here v_m is the maximum expansion velocity, a is a constant and $\xi = x/V_m t$. It is a pity that this model is only partially consistent with facts and it is difficult to extend to two- or three-dimensional conditions. Furthermore, experiments show that the ionization degree of plasma affects the velocity of plasma markedly.[14] However, little attention was paid on the ionization effect to plasma kinetic behaviour generated by laser ablation in previous works.[4−9]

In this Letter, a numerical hydrodynamic model is used to study the accelerated expansion mechanism. Different from the previous models, the energy absorption of target from the laser radiation is regarded as a dynamic source, which accelerates expansion of ablated material occurring near the solid surface. Local mass and momentum conservations which are more catholical are used, the ionization effect to plasma expansion is considered in our model as well. The processes are introduced in our model as three parts. A brief resume of pulsed laser deposition is first given. Then, considering the plasma plume as non-viscose gas, the plasma number density and pressure are studied. Using analytical method, the hydrokinetics equations are deduced. Lastly, the corresponding novel physical results from our model are derived.

We assume that a laser beam with radius R_0 and pulse duration τ interacts with a plane target ($z < 0$). At the initial stage of nanosecond laser ablation (laser pulse duration is in the order of 10^{-8} s), the ablated material recession velocity is roughly about $10^3 \sim 10^4$ m/s,[5] the axial expansion of the ablation plume

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for this period is estimated to be $10^{-2} \sim 10^{-1}$ mm. This value is much less than the laser spot size (typically about 1 mm), so we consider temporal evolution of the ablation plume as two stages: At the initial stage the basic processes are the laser radiation absorption, the target heating, melting, evaporation, and 1D axial expansion of the vapour. This stage is responsible for the ablation plume formation too. The second stage can be regarded as 2D axial and radial expansion of gas-dynamic stage and the plasma behaves as a continuum fluid.[15,16] We assume that the plasma propagates in the direction normal to the target surface, the axis lies parallel to the target surface and the laser-irradiation centre is located at the origin of cylindrical coordinate. We give a sketching of the plasma formation on laser radiation in Fig. 1.

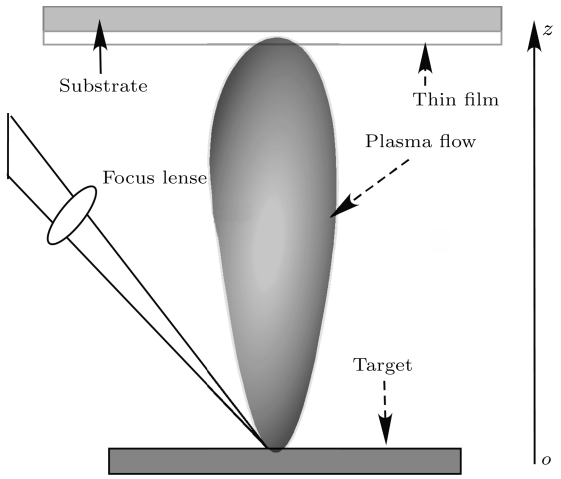


Fig. 1. Schematic diagram of the plasma flow in the PLD process.

During the stage of laser radiation, a mixture of vapour and multiform particles will sputter from the target surface,[2,3,17] and the initial expansion stage is almost out of order,[6] namely this stage is unsteady and nonadiabatic near the target surface. The system may develop to be a Gaussian profile.[18,19] Thus the density of the plasma in cylindrical coordinates can be given by (the plume expands along the z axis):

$$n(z,r,t) = A\exp\Big[-\frac{z^2}{2Z(t)^2} - \frac{r^2}{2R(t)^2}\Big], \qquad (2)$$

where z and r represent the position of particles, $Z(t)$ and $R(t)$ are the position of plasma corresponding to the distance where the density of the plasma reduces to 60.65% in the z and r directions, respectively; S is the area irradiated by the laser. A is a variable for time t, namely

$$A = f(t). \qquad (3)$$

During laser radiation time t, according to the principle of mass balance, the mass for evaporation must be equal to the ablated material for the target, i.e.

$$\int_0^{2\pi}\int_0^t\int_0^\infty\int_0^\infty n(z,r,t)rdrdzd\theta dt = \alpha S\nu_z t. \qquad (4)$$

Assume that the plasma behaves as an ideal gas, and then the pressure $P(z,r,t)$ in the plasma related to its density by the ideal gas equation can be expressed as

$$P = n(1+\eta)k_BT, \qquad (5)$$

where k_B is the Boltzmann constant, T is the isothermal temperature of the plasma and η is the ionization fraction of the plasma.

Substituting Eqs. (1) and (2) into Eqs. (3) and (4) separately yields

$$f(t) = \frac{\alpha S\nu_z}{2\pi Z(t)R(t)^2}, \qquad (6)$$

$$n(z,r,t) = \frac{\alpha S\nu_z}{2\pi Z(t)R(t)^2}\exp\Big[-\frac{Z}{Z(t)} - \frac{r^2}{2R(t)^2}\Big], \qquad (7)$$

$$P(z,r,t) = \frac{\alpha S\nu_z k_BT}{2\pi Z(t)R(t)^2}\exp\Big[-\frac{Z}{Z(t)} - \frac{r^2}{2R(t)^2}\Big]. \qquad (8)$$

It only needs several picoseconds for electron to thermal equilibrium with ions and neutrons, so we can consider the different particles in plasma plume containing atoms, ions, and electrons as a non-viscous vapour and on local thermal equilibrium. Under this condition, the ionization fraction can be determined by the Saha equation,[19]

$$\frac{\eta^2}{1-\eta} = \frac{2u_+}{nu_0}\Big(\frac{2\pi m_e k_BT}{h^2}\Big)^{\frac{3}{2}}\exp\Big(-\frac{U_i}{k_BT}\Big), \qquad (9)$$

where U_i is the ionization energy, u_+ and u_- the partition function of ionized atoms and neutral atoms respectively, m_e the electron mass, and h Planck's constant. Based on fluid-dynamics theory, the equation of the mass conservation laws in 2D cylindrical frame (r,z) can be written as

$$\frac{\partial\rho}{\partial t} + \frac{\rho u_r}{r} + \frac{\partial(\rho u_r)}{\partial r} + \frac{\partial(\rho u_z)}{\partial z} = 0, \qquad (10)$$

where $\rho = nm$, m is the average mass of atomic species, u_r and u_z are the radial and longitudinal velocity respectively.

Substituting Eq. (7) into Eq. (10), we can obtain

$$\frac{\partial u_r}{\partial r} + \frac{u_r}{r} - \frac{r}{R^2(t)}u_r + \frac{r^2}{R^3(t)}\frac{dR(t)}{dt} = -\frac{\partial u_z}{\partial z} + \frac{z}{Z^2(t)}u_r - \frac{z^2}{Z^3(t)}\frac{dZ(t)}{dt} - \frac{1}{t}. \qquad (11)$$

Assuming that u_z is independent of r, and u_r is independent of z, we obtain

$$\frac{\partial u_r}{\partial r}+\frac{u_r}{r}-\frac{r}{R^2(t)}u_r+\frac{r^2}{R^3(t)}\frac{dR(t)}{dt}=f(t), \tag{12}$$

$$\frac{\partial u_z}{\partial z}-\frac{z}{Z^2(t)}u_z+\frac{z^2}{Z^3(t)}\frac{dZ(t)}{dt}+\frac{1}{t}=-f(t), \tag{13}$$

where the function $f(t)$ is to be determined, the boundary conditions read

$$u_r|_{r=0}=0, \quad u_r|_{r=R(t)}=\frac{dR(t)}{dt}, \tag{14}$$

$$u_z|_{z=Z(t)}=\frac{dZ(t)}{dt}. \tag{15}$$

Integrating Eqs. (12) and (13) over r and z respectively, and applying the boundary conditions (14) and (15) yield

$$f(t)=0, \tag{16}$$

$$u_r=\frac{r}{R(t)}\frac{dR(t)}{dt}, \tag{17}$$

$$u_z=\frac{1}{t}\exp\Big(\frac{z^2}{2Z^2(t)}\Big)\int_z^Z \exp\Big(-\frac{s^2}{2Z^2(t)}\Big)ds + \frac{z}{Z(t)}\frac{dZ(t)}{dt}, \tag{18}$$

namely

$$u_z\approx\frac{Z(t)}{t}+\Big[\frac{dZ(t)}{dt}-\frac{Z(t)}{t}\Big]\frac{z}{Z(t)}. \tag{19}$$

Meanwhile, the equation of the conservation of linear momentum or the equation of motion can be expressed as[20−21]

$$\rho\Big[\frac{\partial u_r}{\partial t}+u_r\frac{\partial u_r}{\partial r}\Big]=-\frac{1}{r}\frac{\partial(rP_r)}{\partial r}, \tag{20}$$

$$\rho\Big[\frac{\partial u_z}{\partial t}+v_z\frac{\partial u_z}{\partial z}\Big]=-\frac{1}{r}\frac{\partial(rP_z)}{\partial z}, \tag{21}$$

where P_r and P_z are the pressures in the r and z directions. Substituting Eqs. (8), (17) and (18) into Eqs. (20,21), we can obtain the new dynamics evolvement equations for plasma expansion during pulsed laser deposition,

$$Z(t)\frac{d^2Z(t)}{dt^2}=\frac{(1+\eta)k_BT}{m}, \tag{22}$$

$$R(t)\frac{d^2R(t)}{dt^2}=\frac{(1+\eta)k_BT}{m}. \tag{23}$$

So far, we have obtained a set of dynamic equations (17), (18), (22) and (23) for the plasma.

The above theoretical results show interesting physics mechanism. We can see during the laser ablation, the inner velocity of the plasma is self-similar in the radial direction, that is, $u_r=\frac{r}{R(t)}\frac{dR(t)}{dt}$, which is consistent with the assumption in the literature.[5,8,19] Different from the assumption, it is a nature derivation result from our model. From Eq. (19), we can see that the velocity in the longitudinal direction is different from u_r, this owes to the existence of dynamic source in normal direction. If we suppose $v_m=\frac{dZ(t)}{dt}$,and $a=\frac{Z(t)}{t}/\frac{dZ(t)}{dt}$, then $u_z=v_m\Big[a+(1-a)\frac{z}{Z(t)}\Big]$, this is similar to the assumption in Refs. [5,8] presented by Eq. (1). Different from the assumption of Refs. [5,8], the expression of u_z is also a nature derivation result in our model. The experimental results in Refs. [22,23] tell us that during the plasma expansion, the slow component is the heavier particles and the fast component is the lighter particles. These experimental facts are consistent with our results completely.

In summary, based on the consideration of local conservations of mass, momentum and particle ionization of plasma, collected as the assumption that plasma can be viewed as compressible ideal fluid and high temperature-high pressure ideal gas, we have developed a new dynamics expansion mechanism for plasma produced by pulsed laser radiation. Using analytical method, the space number density and pressure evolvement of plasma in cylindrical coordinate are obtained, the dynamics evolvement equations are also derived. The results from the present model indicate that the plasma dynamic expansion behaviour can be evidently influenced by the ionization fraction η. Its effect is similar to a new dynamic source for plasma expansion and to increase the expansion acceleration in all directions. The inner velocity of the plasma is self-similar in the radial direction. The velocity in the longitudinal direction is different from u_r because of the existence of dynamic source in the normal direction. The predictions of the expansion of the plasma is affected by the average atoms mass are consistent with the experimental observation. This verifies the validity of the present model for laser-induced plasma expansion.

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