Gas dynamics and film profiles in pulsed-laser deposition of materials

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Film-thickness profiles obtained in pulsed-laser deposition are calculated by using the well-known solution of the gas-dynamic equations which describes the expansion of the plasma plume in vacuum. The time for plasma formation is supposed to be short compared with the time of expansion. The film profile depends on the initial dimensions of the plume and on the adiabatic exponent of the vapor.

I. INTRODUCTION

During the last two decades, laser-matter interactions within the intensity range 10^6 W/cm$^2 \leq I \leq 10^9$ W/cm^2 have become of increasing interest with respect to both technological applications and the elucidation of the fundamental mechanisms involved. Among the technological applications are laser machining, laser surface processing, and laser chemical processing for microfabrication and thin-film formation.[1,2]

The main processes involved in laser-matter interactions with intensities $I < 10^9$ W/cm^2 are the following: absorption and reflection of the incident laser light by the condensed phase, ablation of the condensed phase, absorption of the laser light within the expanding plume, expansion of the vapor plasma, and in the case of pulsed-laser deposition, the interaction of ablated species with the substrate surface.

With the laser beam intensities and pulse widths under consideration, the temperature of the target is considerably lower than its thermodynamic critical temperature T_c. As a consequence, there is a sharp boundary between the gaseous and the condensed phase. The thickness of this boundary is of the order of a few interatomic distances. Ablation is often considered as a surface process (sometimes without sufficient reasons). In this approximation, the vaporization kinetics is determined by the surface temperature which, in turn, depends on the spatial and temporal distribution of the laser-induced temperature within the target. This temperature distribution is determined by the laser parameters and the relaxation of the excitation energy, including electron-phonon interactions, phase transitions, and chemical changes. On this basis, a mesoscopic description of the heating dynamics is quite complicated and will be discussed elsewhere.

The expansion of the vapor-plasma plume into a vacuum has been studied in one dimension for the case of low optical absorption.[3] For ultraviolet (UV) laser ablation, absorption of the laser light within the plasma plume affects both the vapor flow and the laser-induced temperature distribution within the target.[4] However, with the parameters typically employed in pulsed laser deposition (PLD), the characteristic time of the gas-dynamic expansion is much longer than the duration of the laser pulse. This permits a separate consideration of the formation and the expansion of the plasma plume. With this condition, relatively simple analytical solutions of the vapor expansion problem can be obtained, even for the three-dimensional case. The understanding of the (three-dimensional) expansion of the plasma plume is a prerequisite for the analysis of film thickness profiles in PLD. Experiments have revealed that near the axis of the plasma plume the angular distribution of the flux of species is $\approx \cos^n\theta$ with $n \gg 1$.[5,6] This strong forward direction is caused by strong differences in pressure gradients in axial and radial directions.

The problem of the angular distribution of the mass flow in plasma expansion was recently investigated.[7] These authors used the isothermal solution of the gas-dynamical equations[8,9] with Gaussian pressure and density profiles which have been considered already in Refs. 10 and 11. Although these results explain correctly the fast expansion of the plasma in the direction of the maximum pressure gradients, the neglection of spatial temperature gradients is inadequate for the description of pulsed-laser ablation. Indeed, both experiments[12] and numerical calculations[13] reveal considerable temperature gradients inside the plasma plume. These gradients are generated during the laser-pulse action and they become more pronounced in the subsequent free expansion of the plume. In this situation, it is more realistic to consider an adiabatic expansion of the plume. Clearly, in reality the initial state of the plume is neither isothermal nor isentropic. However, in the frame of hydrodynamics, an adiabatic motion is more physical since there is no mechanism which sustains a finite temperature at the outer edge of the plume. This physical inadequacy of the isothermal solution is well known and is discussed in detail in Ref. 14.

In this paper we study the adiabatic expansion of the plasma plume on the basis of analytical solutions described in Refs. 8 and 9. These investigations give a more physical picture on the temperature distribution within the expanding plume. The results are important for a more detailed understanding of the interactions between the plasma plume and the substrate surface.

II. THEORETICAL MODEL

The process under consideration can be described as follows. The laser beam with a pulse width of, typically, several tens of ns, produces a vapor-plasma plume on the target surface which is located at $z=0$ (see Fig. 1). The detailed structure of the plume is not considered any further, since in the present approach only the initial dimensions of the plume, R_0 and Z_0, its mass M, and the initial energy E are required. As already mentioned, the formation of the plume is a very complicated problem.

The radius of the plume R_0 can be approximated by the radius of the laser spot. The height of the plume in the z direction is about $Z_0 \approx v_s \tau_l$, where v_s is the velocity of sound and τ_l is the laser-pulse duration. A rough estimation yields $v_s \approx \sqrt{E/M}$. In the present problem we assume $Z_0 \ll R_0$. Typical values of R_0 are $R_0 \approx 0.1-0.01$ cm. Values of Z_0 are $Z_0 \approx 0.01 \times [T/A]^{1/2}$ cm, where T is the plasma temperature in eV and A is the atomic mass number.

The expansion of the plasma plume can be described by the gas-dynamic equations

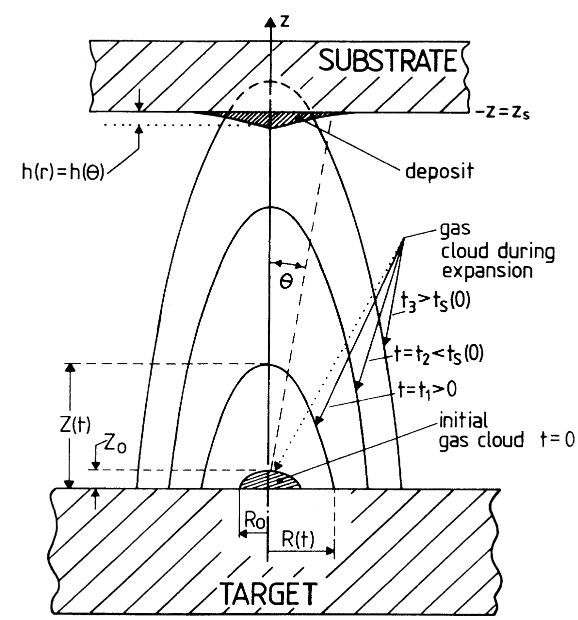

FIG. 1. Schematic for the gas cloud expansion and the deposition of a thin film. The initial gas cloud was created at $t=0$ (after the end of the laser pulse) near the target surface. The gas cloud remains elliptical during its expansion for $t>0$ (see text). The ablated material which reaches the substrate at $z=z_s$ condenses and thereby forms the thin film. The profile of the film is $h(r,t)=h(\theta,t)$, where r is the radial coordinate and θ the radial angle, $\theta = \arctan r/z_s$.

$$\begin{aligned} &\frac{\partial \rho}{\partial t} + \mathrm{div}(\rho \mathbf{v}) = 0 , \\ &\frac{\partial \mathbf{v}}{\partial t} + (\mathbf{v}\cdot\nabla)\mathbf{v} + \frac{1}{\rho}\nabla p = 0 , \\ &\frac{\partial S}{\partial t} + (\mathbf{v}\cdot\nabla) S = 0 , \end{aligned} \tag{1}$$

where ρ, p, $\mathbf{v}$, and S are the density, pressure, velocity, and entropy, respectively. In addition, we need the equation of state. We suppose that the evaporated material can be described by the equation for ideal gases with a constant adiabatic exponent $\gamma = c_p/c_v$.

As already mentioned, a special solution of (1) described in Ref. 8 will be employed. This solution exists due to the invariance of the gas-dynamic equations with respect to some Lie group transforms (see details in Refs. 8, 9, and 14). With this solution, the characteristics of the expanding plasma plume remain constant at ellipsoidal surfaces. The coordinates of a fluid particle r_i ($i=x,y,z$) can be represented in the form[8]

$$r_i(t) = \sum_k F_{ik}(t) r_k(0) , \tag{2}$$

where $r_i(0)$ are the initial coordinates. Equation (2) describes the motion of a fluid particle from $x(0)$, $y(0)$, $z(0)$ to $x(t)$, $y(t)$, $z(t)$. This motion includes the expansion (compression) and rotation of fluid elements. For the particular problem under consideration, we can ignore, in many cases, the rotation and reduce the matrix $F_{ik}(t)$ to the diagonal form

$$F_{ik}(t) = \begin{bmatrix} X(t)/X_0 & 0 & 0 \\ 0 & Y(t)/Y_0 & 0 \\ 0 & 0 & Z(t)/Z_0 \end{bmatrix} , \tag{3}$$

where X_0, Y_0, Z_0 are the initial values of $X(t)$, $Y(t)$, $Z(t)$, respectively.

A further reduction can be obtained by taking into account the symmetry of the plume with respect to the z axis. It is more convenient, however, to make this transformation in the final equations.

Substituting (2) into the set of gas-dynamical equations (1), we obtain a set of ordinary differential equations for the elements of the matrix F_{ik}. This transformation can be carried out, however, only for some particular density and pressure profiles.[8,9] For an adiabatic gas flow these profiles have the form

$$\begin{aligned} \rho(x,y,z,t) &= \frac{M}{I_1(\gamma) XYZ} \left[1 - \frac{x^2}{X^2} - \frac{y^2}{Y^2} - \frac{z^2}{Z^2} \right]^{1/(\gamma-1)} , \\ p(x,y,z,t) &= \frac{E}{I_2(\gamma) XYZ} \left[\frac{X_0 Y_0 Z_0}{XYZ} \right]^{\gamma-1} \\ &\quad \times \left[1 - \frac{x^2}{X^2} - \frac{y^2}{Y^2} - \frac{z^2}{Z^2} \right]^{\gamma/(\gamma-1)} . \end{aligned} \tag{4}$$

For the profiles (4), the density and pressure are constants on the ellipsoidal surfaces $x^2/X^2 + y^2/Y^2 + z^2/Z^2 = \mathrm{const}$. The values of M and E in formula (4) are defined as the integrals over the volume of the plume

$$M=\int_V \rho(x,y,z,t)dV \; ,$$
$$E=\frac{1}{\gamma-1}\int_V p(x,y,z,0)dV \; . \tag{5}$$

The values $I_1(\gamma)$ and $I_2(\gamma)$ are defined as

$$I_1(\gamma)=2\pi\int_0^1 s^2(1-s^2)^{1/\gamma-1}ds \; ,$$
$$I_2(\gamma)=\frac{2\pi}{\gamma-1}\int_0^1 s^2(1-s^2)^{\gamma/\gamma-1}ds \; . \tag{6}$$

Note that the density and pressure profiles (4) describe a plume with a sharp external edge. This is the consequence of entropy conservation, which leads to the well-known relation $T\propto\rho^{\gamma-1}$. Thus, at the front, where the density is equal to zero, the temperature and the velocity of sound approach zero as well. This results in the formation of a sharp front—in contrast to an isothermal expansion, where the velocity of sound at the outer edge of the plume remains finite and generates density and pressure tails.[7,10,11]

According to (2) the velocity at the point **r** is proportional to the radius vector of this point. Thus,

$$v_x=x\frac{\dot{X}}{X}\; , \quad v_y=\frac{\dot{Y}}{Y}\; , \quad v_z=z\frac{\dot{Z}}{Z}\; , \tag{7}$$

where $\dot{X}=dX/dt$, etc. Substituting (2) and (3) into (1) and taking into account (4), we obtain a set of ordinary differential equations which, formally, can be written as the equations of motion of a point in classical mechanics

$$\ddot{X}=-\frac{\partial\tilde{U}}{\partial X}\; , \quad \ddot{Y}=-\frac{\partial\tilde{U}}{\partial Y}\; , \quad \ddot{Z}=-\frac{\partial\tilde{U}}{\partial Z}\; , \tag{8}$$

where

$$\tilde{U}=\frac{\beta(\gamma)}{\gamma-1}\left[\frac{X_0Y_0Z_0}{XYZ}\right]^{\gamma-1}$$

and

$$\beta(\gamma)=\frac{2\gamma}{\gamma-1}\frac{I_1(\gamma)}{I_2(\gamma)}\frac{E}{M}\; .$$

It should be noted that, in the general case when rotations are taken into account, the set of equations for the matrix F_{ik} has a form similar to (8),[10,11]

$$\ddot{F}_{ik}=-\frac{\partial\tilde{U}}{\partial F_{ik}}$$

with

$$\tilde{U}=\mu(\det F_{ik})^{1-\gamma}\; ,$$

where $\mu=$const. Henceforth, we consider a plasma plume which is at rest at $t=0$, i.e., we put $\dot{X}(0)=\dot{Y}(0)=\dot{Z}(0)=0$. Usually, this is a good approximation since the kinetic energy of the vapor flow near the target surface is considerably smaller than the thermal energy of the vapor.[3] In this approximation the initial energy of the plasma plume is

$$E=E_a-M\Delta H_v\; ,$$

where $E_a=I_a\tau_l$ is the laser-light energy absorbed and ΔH_v is the enthalpy of vaporization.

The integrals (6) can be expressed in terms of the Γ function[15]

$$I_1(\gamma)=\pi\Gamma\left[\frac{\gamma}{\gamma-1}\right]\Gamma(\tfrac{3}{2})\Big/\Gamma\left[\frac{\gamma}{\gamma-1}+\frac{3}{2}\right]\; ,$$
$$I_2(\gamma)=\pi\Gamma\left[\frac{\gamma}{\gamma-1}+1\right]\Gamma(\tfrac{3}{2})\Big/(\gamma-1)\Gamma\left[\frac{\gamma}{\gamma-1}+\frac{5}{2}\right]\; .$$

These follow from Euler's integrals of the second kind,

$$B(m,n)=\int_0^1 x^{m-1}(1-x)^{n-1}dx=\Gamma(m)\Gamma(n)/\Gamma(m+n)\; .$$

From standard calculations we get for $\beta(\gamma)$ the following equation:

$$\beta=(5\gamma-3)\frac{E}{M}\; .$$

Finally, we introduce the dimensionless variables

$$\tau=\frac{t}{t_0}\; , \quad \xi(\tau)=\frac{X(t)}{R_0}\; , \quad \eta(\tau)=\frac{Z(t)}{R_0}\; , \quad \sigma=\frac{Z_0}{R_0}\; ,$$

where $t_0=R_0/\sqrt{\beta}$. When taking into account the axial symmetry of the plasma plume, $X(t)=Y(t)$, we obtain the set of equations

$$\xi\ddot{\xi}=\eta\ddot{\eta}=\left[\frac{\sigma}{\xi^2\eta}\right]^{\gamma-1} \tag{9}$$

with

$$\ddot{\xi}=\frac{d^2\xi}{d\tau^2}\; , \quad \ddot{\eta}=\frac{d^2\eta}{d\tau^2}\; .$$

The initial conditions are

$$\xi(0)=1\; , \quad \eta(0)=\sigma\; , \quad \dot{\xi}(0)=\dot{\eta}(0)=0\; .$$

Thus, the evolution of the plume in variables ξ and η is determined by two parameters, σ and γ.

Note that the characteristic time scale for expansion is $t_0=R_0/\sqrt{\beta}\approx(R_0/z_0)\tau_l\gg\tau_l$. This time is much longer than the laser-pulse duration τ_l.

In general, Eqs. (9) can be solved numerically only. It is convenient to use the first integral (in terms of classical mechanics the integral of energy) for a check of the accuracy of calculations. A standard procedure leads to the following relation:

$$\dot{\xi}^2+\tfrac{1}{2}\dot{\eta}^2+U(\xi,\eta)=\varepsilon=\text{const}\; , \tag{10}$$

where U is the "potential energy"

$$U(\xi,\eta)=\frac{\tilde{U}}{\beta}=\frac{1}{\gamma-1}\left[\frac{\sigma}{\xi^2\eta}\right]^{\gamma-1}\; .$$

If we disregard the initial gas velocity within the plume, we obtain for the "total energy"

$$\varepsilon=U[\xi(0),\eta(0)]=\frac{1}{\gamma-1}\; .$$

For the special case $\gamma=\frac{5}{3}$, there exists an additional integral of (9). This integral has been derived in Ref. 11 and it can be written as

$$2\xi^2+\eta^2=2\varepsilon\tau^2+\sigma^2+2 \ . \tag{11}$$

The solution of (10) and (11) can be written as

$$\arctan\left[\frac{\tau\sqrt{3}}{\sqrt{2+\sigma^2}}\right]=\pm\int_{\theta}^{\theta_0}\frac{d\omega}{\left[1-\left[\frac{\sin^2\theta_0\cos\theta_0}{\sin^2\omega\cos\omega}\right]^{2/3}\right]^{1/2}} ,$$

where

$$\xi=(3\tau^2+\sigma^2+2)^{1/2}\frac{\sin\theta}{\sqrt{2}} ,$$

$$\eta=(3\tau^2+\sigma^2+2)^{1/2}\cos\theta ,$$

$$\sin\theta_0=\left[\frac{2}{2+\sigma^2}\right]^{1/2} , \quad \cos\theta_0=\frac{\sigma}{\sqrt{2+\sigma^2}} .$$

This (exact) solution can be used to check the accuracy of numerical calculations and to investigate the asymptotic behavior of the solutions of (9).

Equations (9) have been solved numerically for a wide range of parameters σ and γ. In particular, we have been interested in the behavior of the solutions for long times τ. It can be shown readily that $\xi(\tau)$ and $\eta(\tau)$ are both linear functions of τ when $\tau\rightarrow\infty$. This means that the expansion of the vapor plume becomes inertial, as the pressure gradients tend to zero. The limiting shape of the expanding plume has been calculated for each pair (γ,σ). It is interesting to note that for $\gamma<\frac{5}{3}$ the ratio $k(\tau)=\eta(\tau)/\xi(\tau)$, which describes the shape of the plume, reaches its maximum at $\tau\approx 10-100$ (for the region of practical interest $1.1\leq\gamma\leq 1.4$) and then decreases slowly; it reaches its limiting value at $\tau\approx 10^4$. This can be seen in Fig. 2 which shows that the dependence of k on η for different values of γ [$\eta(\tau)$ is a monotonic function of time τ]. For $\gamma\geq\frac{5}{3}$ the ratio $\eta(\tau)/\xi(\tau)$ is a monotonic function of τ. Table I summarizes some information on the dynamics of the deformation of the plume for large values of τ; here, the values of k are given for $\tau\approx 100$ and $\tau\rightarrow\infty$ for a set of parameters σ and γ. We find that even for $\tau\approx 10^2$ the asymptotic relation between $\eta(\tau)$ and $\xi(\tau)$ can be employed for calculating deposition profiles. An example for the dependences of $\xi(\tau)$, $\eta(\tau)$, and $k(\tau)$ is shown in Fig. 3 for $\gamma=\frac{5}{3}$.

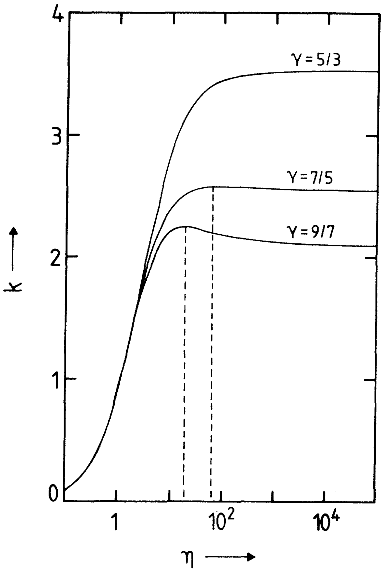

FIG. 2. k as a function of η for $\sigma=0.1$ and various values of γ.

From the numerical calculations we find that with increasing γ, the time required to reach the asymptotic regime of k becomes shorter. The asymptotic and maximum values of k are listed in Table II for different values of parameters γ and σ.

We now calculate the thickness profile, $h(\theta)$, of deposited films. From (4) and (7) we find for the (mass) flux normal to the substrate surface $z=z_s$ (see Fig. 1)

$$j(r,z_s,t)=\rho(r,z_s,t)v_z(z_s,t)=\begin{cases}\dfrac{Mz_s\dot{Z}(t)}{I_1(\gamma)X^2(t)Z^2(t)}\left[1-\dfrac{r^2}{X^2(t)}-\dfrac{z_s^2}{Z^2(t)}\right]^{1/(\gamma-1)} , & \text{with } t\geq t_s(r)\\ 0 \text{ with } t<t_s(r) . \end{cases} \tag{12}$$

Here, $t_s(r)$ is the time when the edge of the expanding plume reaches the substrate surface $z=z_s$ at a given r. This time can be derived from

$$\frac{r^2}{X^2(t_s)}+\frac{z_s^2}{Z^2(t_s)}=1 .$$

After integration of the mass flux $j(r,z_s,t)$ over the time t from t_s to ∞ and dividing the result by the density of the deposited material ρ_s, we obtain for the thickness profile

TABLE I. Comparison of values of $k(\tau)$ at $\tau=100$ and $\tau=\infty$.

σ	0.01	0.03	0.1	0.3
γ		$k(100)$; $k(\infty)$		
$\frac{9}{7}$	3.70;	2.87;	2.16;	1.55;
	3.61	2.85	2.12	1.53
$\frac{7}{5}$	5.21;	3.79;	2.56;	1.69;
	5.20	3.77	2.55	1.69
$\frac{5}{3}$	10.9;	6.37;	3.49;	1.97;
	11.3	6.52	3.53	1.98

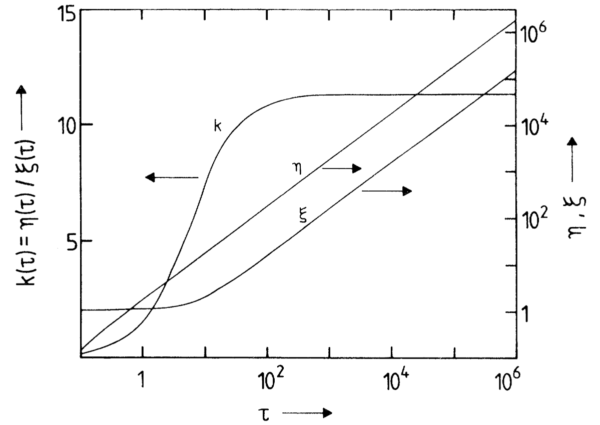

FIG. 3. ξ, η, and k as a function of τ for $\gamma=\frac{5}{3}$ and $\sigma=0.01$.

$$h(\theta)=\frac{Mz_s}{\rho_s I_1(\gamma)} \times \int_{t_s}^{\infty} \frac{\dot{Z}\,dt}{X^2Z^2}\left[1-z_s^2\left[\frac{\tan^2\theta}{X^2}+\frac{1}{Z^2}\right]\right]^{1/(\gamma-1)}, \tag{13}$$

where $\tan\theta=r/z$.

In the general case, the integration in (13) can be performed numerically by using the relation between $X(t)$ and $Z(t)$ as obtained from the (numerical) solution of (9). A simple analytical expression for $h(\theta)$ can be obtained, if $z_s \gg R_0$ which is satisfied in many experimental situations. From this inequality it follows that $t_s \gg t_0$, and we can use the asymptotic relation between $X(t)$ and $Z(t)$ for calculating the integral (13). As already mentioned, the numerical solution of (9) shows that for $z_s/R_0>100$ the ratio $Z(t)/X(t)$ is equal to its asymptotic value $k=\eta(\infty)/\xi(\infty)=\text{const}$ within an accuracy of better than 4%. Substituting $Z(t)=kX(t)$ into (13) we obtain the thickness profile of the form

$$h(\theta)=\frac{Mk^2}{2\pi\rho_s z_s^2}(1+k^2\tan^2\theta)^{-3/2}\ . \tag{14}$$

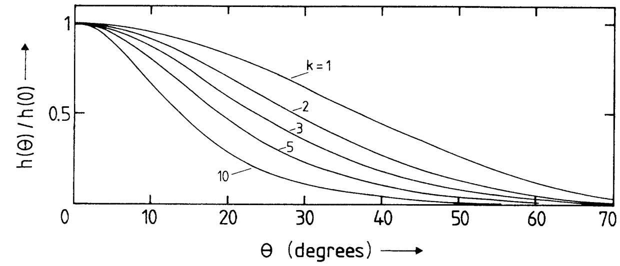

FIG. 4. Stationary profile of the deposited film for various values of k.

The function $h(\theta)$ given by (14) differs, in general, from the usual approximation $h(\theta)\propto\cos^n\theta$. However, with small angles, $\theta \ll \arctan(1/k)$, these two profiles have the same series expansion if $n=3k^2$. In particular, the case of a spherical expansion of the plume (14) has the form

$$h(\theta)=\frac{M}{2\pi\rho_s z_s^2}\cos^3\theta\ .$$

Note that this result agrees with the dependence $h(\theta)$ which holds for a steady-state point source. It can be shown directly from the law of mass conservation that

$$h(\theta)=\frac{\dot{M}t}{2\pi\rho_s z_s^2}\cos^3\theta\ .$$

Here, $\dot{M}$ is the mass production rate.

The dependence $h(\theta)$ which follows from Eq. (14) is presented in Fig. 4. We emphasize that the profile $h(\theta)$ can easily be calculated for the general case where the ratio z_s/R is of the order of 1–10. For this purpose only a simple numerical integration has to be carried out in (13). For a qualitative estimation, the simple equation (14) can be employed.

III. CONCLUSION

We have calculated the profile of a film produced by pulsed-laser deposition (PLD) from a solid target. The analysis is based on the solution of gas-dynamical equa-

TABLE II. The maximum, k_{max}, and asymptotic, $k(\infty)$, values of $k=k(\tau)$ for different values of parameters γ and σ. For $\gamma \geq \frac{5}{3}$ only $k_{max}=k(\infty)$ values are given.

σ	0.001	0.003	0.01	0.03	0.1	0.3
γ			k_{max}; $k(\infty)$			
1.1				2.417 93;	1.907 98;	1.444 64;
				1.609 54	1.396 04	1.208 20
1.2	4.890 24;	4.150 55;	3.396 91;	2.747 77;	2.074 18;	1.511 29;
	3.693 92	3.185 58	2.675 9	2.240 49	1.787 17	1.392 60
$\frac{9}{7}$	6.408 70;	5.182 21;	4.030 79;	3.119 73;	2.252 87;	1.580 42;
	5.604 84	4.571 03	3.607 19	2.847 90	2.116 92	1.531 70
1.4	9.901 155;	7.394 15;	5.293 29;	3.817 57;	2.566 65;	1.695 61;
	9.664 782	7.234 91	5.200 32	3.769 59	2.550 05	1.692 05
$\frac{5}{3}$	35.841 1	20.686 7	11.318 7	6.515 26	3.532 01	1.983 34
2.0	166.127	66.330 8	25.206 8	10.925 3	4.650 97	2.239 61
3.0	636.633	212.275	63.835 7	21.550 1	6.873 29	2.656 77

tions assuming an adiabatic expansion of the plasma plume into vacuum. A simple analytical equation is obtained for the range of parameters typically employed in PLD. This equation shows that, with small angle θ, the film thickness is proportional to $\cos^n\theta$ with $n = 3k^2$. For typical experimental conditions n is within the range of several units to several tens (Table II) which is in qualitative agreement with the experimental results.

The solution can also be used for an estimation of the kinetic energy (temperature) of the species near the substrate surface. This is an important quantity for the interpretation of epitaxial film growth experiments.

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