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On the approach to the analysis of the growth of epitaxial layers by pulsed laser deposition

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Abstract

This paper considers an analytical approach for the prognosis of mass and heat transport during the growth of epitaxial layers by means of pulsed laser deposition. The approach provides the opportunity to make a prognosis which takes into account the spatial and temporal variations of their parameters and, at the same time, the nonlinearity of these processes. Based on this approach, the influence of the variation of several parameters on the growth process is investigated.

Keywords: pulsed laser deposition, mass and heat transport, analytical modelling

1. Introduction

One of the most promising modern methods for producing epitaxial layers is pulsed laser deposition. This method provides the opportunity to grow special materials (metals, carbides, etc.) on the surface of the considered parts, helping to restore geometry, increase surface strength and improve corrosion resistance, etc. (see Abe et al., 2005; Bonse et al., 2001, 2005; Borowiec & Haugen 2003; Chelnokov et al., 2006; Couillard et al., 2007; Nutsch et al., 1998; Shen & Kwok, 1994; Zherikhin et al., 2003; Zhvavyi et al., 2006). This work considers mass and heat transfer in the reaction chamber during the growth of an epitaxial layer under pulsed laser deposition. An analytical approach to analyzing the considered processes was introduced, allowing their nonlinearity and changes of parameters in space and time to be taken into account.

2. Method of solution

To solve the issue, mass and heat transfer in the direction perpendicular to the source of the material evaporated during laser deposition is considered by the second Fourier law:

$$c_p\rho\left[\frac{\partial T(x,t)}{\partial t}-u(t)\frac{\partial T(x,t)}{\partial x}\right]=\frac{\partial}{\partial x}\left[\lambda(T)\frac{\partial T(x,t)}{\partial x}\right]+p(x,t) \tag{1}$$

where ρ is the density of the evaporated material; c_p is the specific heat at constant pressure; $\lambda(T)$ is the thermal conductivity; $p(x, t)$ is the power density of laser radiation; x and t are the current coordinate and time; $T(x, t)$ is the heating temperature of the material. The temperature dependence of the thermal conductivity

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coefficient in the desired temperature range can be approximated as follows: $\lambda(T) = \lambda ass\{1 + \mu[Td/T(x, t)]\varphi\}$ (see, for example, Shalimova, 1985), λ_{ass} is the asymptotic value of thermal conductivity at large values of temperature, T_d is the Debye temperature, parameters μ and φ were used for increasing of exactness of approximation of experimental data by the considered function; $\alpha(T) = \lambda(T)/c(T)$ is the thermal diffusivity. The speed of movement of the evaporation boundary is determined by the flows J_i of particles evaporated from the surface: $u(t)=\sum_i J_i/\rho_i$, where i means the material used during growth. The boundary and initial conditions are defined in the following form:

$$\lambda(T)\frac{\partial T(x,t)}{\partial x}\bigg|_{x=0} = Q_p \cdot u(t)$$
$$T(\infty, t) = T_r,\ T(x, 0) = T_r \tag{2}$$

where T_r is the equilibrium temperature equals room temperature; Q_p is the heat of vaporization.

The transfer of the growth components is described by Fick's second law in the following form:

$$\frac{\partial C(x,t)}{\partial t} - u(t)\frac{\partial C(x,t)}{\partial x} = \frac{\partial}{\partial x}\left[D_C \frac{\partial C(x,t)}{\partial x}\right] \tag{3}$$

with boundary and initial conditions:

$$C(0, t) = C_0,\ \frac{\partial C(x,t)}{\partial x}\bigg|_{x=0} = 0$$
$$C(0, 0) = C_0,\ C(x > 0, 0) = 0 \tag{4}$$

where: $C(x, t)$ – the concentration of vaporized material; D_C – the diffusion coefficient of this material.

Next, Equations (1) and (3) can be transformed to the following forms:

$$c_p\rho \frac{\partial T(x,t)}{\partial t} = c_p\rho\, u(t)\frac{\partial T(x,t)}{\partial x} +$$
$$\lambda_{ass}\frac{\partial^2 T(x,t)}{\partial x^2} - (\phi+1)\frac{\mu\lambda_{ass}T_d^{\phi}}{T^{\phi+1}(x,t)}\left[\frac{\partial T(x,t)}{\partial x}\right]^2 + \tag{5}$$
$$\frac{\mu\lambda_{ass}T_d^{\phi}}{T^{\phi}(x,t)}\frac{\partial^2 T(x,t)}{\partial x^2} + p(x,t)$$

$$\frac{\partial C(x,t)}{\partial t} = \frac{\partial}{\partial x}\left[D_C\frac{\partial C(x,t)}{\partial x}\right] + u(t)\frac{\partial C(x,t)}{\partial x} \tag{6}$$

Now Equations (5) and (6) are solved with the method of averaging functional corrections (see, for example, Sokolov, 1955). Within the framework of this method, one replaces the not yet known functions $T(x, t)$ and $C(x, t)$ by their unknown average values α_{1T} and α_{1C} in the right-hand sides of the considered equations. Then the equations for the first approximations of the desired functions $T_1(x, t)$ and $C_1(x, t)$ are obtained:

$$T_1(x,t) = \int_0^t \frac{p(x,\tau)}{c_p\rho}d\tau + T_r$$
$$C_1(x, t) = C_0 \tag{7}$$

The unknown average values of α_{1T} and α_{1C} are determined using standard relations (see, for example, Sokolov, 1955):

$$\alpha_{1T} = \frac{1}{\Theta L}\int_0^{\Theta}\int_0^{L} T_1(x,t)\,dx\,dt$$
$$\alpha_{1C} = \frac{1}{\Theta L}\int_0^{\Theta}\int_0^{L} C_1(x,t)\,dx\,dt \tag{8}$$

where L is the distance between the source of growth material and the grown layer.

Substitution Equations (7) into (8) and calculating of the appropriate integrals leads to relations for average values of α_{1T} and α_{1C}:

$$\alpha_{1T} = \frac{1}{\Theta L}\int_0^{\Theta}(\Theta - t)\int_0^{L}\frac{p(x,t)}{c_p\rho}\,dx\,dt + T_r$$
$$\alpha_{1C} = C_0 \tag{9}$$

The second-order approximations of the required functions $T(x, t)$ and $C(x, t)$ have been determined within the framework of the standard procedure (see, for example, Sokolov, 1955), i.e. by replacing these functions with the following sums: $T(x, t) \to \alpha_{2T} + T_1(x, t)$ and $C(x, t) \to \alpha_{2C} + C_1(x, t)$ on the right side of Equations (4), where α_{2T} and α_{2C} are the average values of the second-order approximations of the considered temperature $T_2(x, t)$ and concentration $C_2(x, t)$. Higher-order approximations are calculated in a similar fashion, with a corresponding increase in the summation indices indicating the order of approximation. The relations for the second-order approximations of the considered functions after the substitution take the following form:

$$T_2(x,t) = \int_0^t u(\tau)\frac{\partial T_1(x,\tau)}{\partial x}d\tau + \frac{\lambda_{ass}}{c_p\rho}\int_0^t\frac{\partial^2 T_1(x,\tau)}{\partial x^2}d\tau -$$
$$\frac{1}{c_p\rho}\int_0^t\frac{\mu\lambda_{ass}T_d^{\phi}(\phi+1)}{\left[\alpha_{2C}+T_1(x,\tau)\right]^{\phi+1}}\left[\frac{\partial T_1(x,\tau)}{\partial x}\right]^2 d\tau + \tag{10}$$
$$\frac{\mu T_d^{\phi}}{c_p\rho}\int_0^t\frac{\lambda_{ass}}{\left[\alpha_{2C}+T_1(x,\tau)\right]^{\phi}}\frac{\partial^2 T_1(x,\tau)}{\partial x^2}d\tau + \int_0^t\frac{p(x,\tau)}{c_p\rho}d\tau + T_r$$

$$C_2(x,t) = \frac{\partial}{\partial x}\left[\int_0^t D_C \frac{\partial C_1(x,\tau)}{\partial x} d\tau\right] + \int_0^t u(\tau)\frac{\partial C_1(x,\tau)}{\partial x} d\tau + C_0 \quad (11)$$

Calculation of the considered average values of the second-order approximations of the required functions α_{2T} and α_{2C} is carried out using standard relations (see, for example, Sokolov, 1955):

$$\alpha_{2T} = \frac{1}{\Theta L}\int_0^\Theta\int_0^L \left[T_2(x,t) - T_1(x,t)\right] dx\,dt$$
$$\alpha_{2C} = \frac{1}{\Theta L}\int_0^\Theta\int_0^L \left[C_2(x,t) - C_1(x,t)\right] dx\,dt \quad (12)$$

This leads to the following equations for the considered parameters:

$$\alpha_{2T} = \frac{1}{\Theta L}\int_0^\Theta (\Theta - t)u(t)\left[T_1(x,t) - T_r\right] dx\,dt + \frac{\lambda_{ass}}{\Theta L c_p \rho}\int_0^\Theta (\Theta - t)\int_0^L \left[Q_p \cdot u(t) - \frac{\partial T_1(x,t)}{\partial x}\right] dx\,dt - \frac{\varphi+1}{c_p \rho \Theta L}\int_0^\Theta\int_0^L \frac{\mu \lambda_{ass} T_d^\varphi}{\left[\alpha_{2T} + T_1(x,t)\right]^{\varphi+1}}\left[\frac{\partial T_1(x,t)}{\partial x}\right]^2 dx\,dt + \frac{\lambda_{ass}\mu T_d^\varphi}{c_p \rho\, \Theta L}\int_0^\Theta\int_0^L \frac{\partial^2 T_1(x,t)}{\partial x^2}\frac{dx\,dt}{\left[\alpha_{2T} + T_1(x,t)\right]^\varphi} \quad (13)$$

$$\alpha_{2C} = \frac{C_0}{\Theta L}\int_0^\Theta (\Theta - t)\, u(t)\, dt \quad (14)$$

The average value of α_{2T} depends on the value of the parameter φ and is calculated with the account of available empirical data. In this paper, the spatio-temporal distributions of the concentration of the growth component and temperature was carried out analytically by using the second-order approximation framework and the method of averaging functional corrections. The approximation is usually sufficient to make qualitative analysis and obtain some quantitative results. The results of analytical calculations were verified by comparing them with the results of direct numerical modelling. For the numerical simulation of mass and heat transport, a direct solutions of Equations (1) and (3) with account appropriate boundary and initial conditions were used with an explicit difference scheme.

3. Discussion

The spatio-temporal distribution of the concentration of the growth component is considered as a case study. Figure 1 shows the dependence of the concentration of the growth component on equilibrium temperature. The increasing number of the curve corresponds to a decrease in the pulse power. Increasing pulse power leads to increasing the quantity of the material which should grow in the considered epitaxial layer. In this situation, the considered concentration of growth material increases. Figure 2 shows the dependences of the concentration of the growth component on the density of the grown material. Increasing number of curve corresponds to increasing of the pulse continuance. Increasing pulse continuance corresponds to the increase of energy, which is the source of material growth. In this situation, a larger quantity of the material leaves the

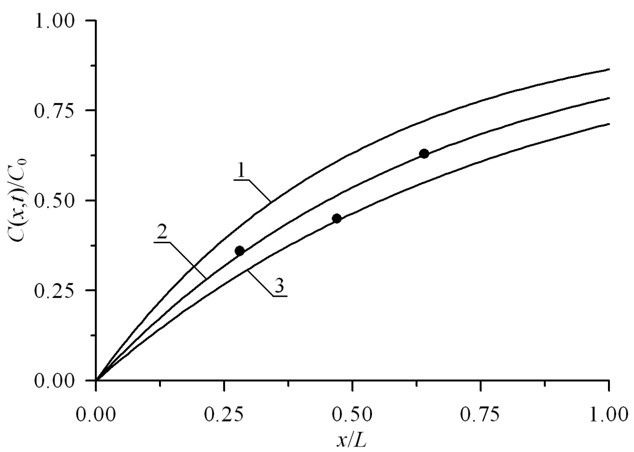

Fig. 1. Distributions of the concentration of the growth component at different values of equilibrium temperature. Increasing of number of curve corresponds to decreasing pulse power. Solid lines are the calculated results. Points are the experimental results from references Ivanov & Smirnov, 2012; Zherikhin et al., 2003

considered source at a fixed value of the growth chamber. In this situation, the considered concentration of growth material increases. Figure 3 shows the dependences of the concentration of the growth component on the heat of the vaporization of the density of the material in the reaction chamber. Increasing the number of the curve corresponds to increasing the distance between the source of the growth material and the growth layer. Increasing the considered distance at the fixed value of growth material corresponds to decreasing the concentration of the considered material.

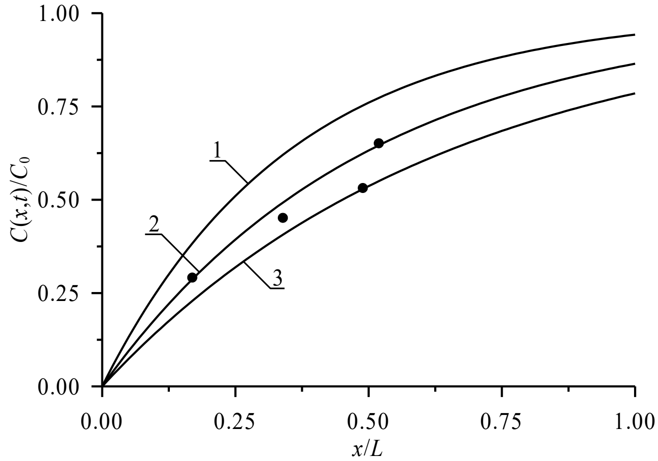

Fig. 2. Distributions of the concentration of the growth component at different values of density of the growth material. Increasing of number of curve corresponds to increasing of the pulse continuance. Solid lines are the calculated results. Points are the experimental results from references Ivanov & Smirnov, 2012; Zherikhin et al., 2003

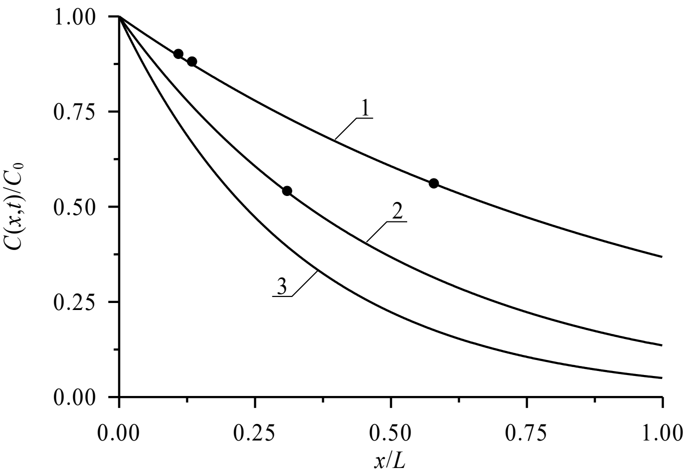

Fig. 3. Distributions of the concentration of the growth component at different values of heat of vaporization of density of material in growth chamber. Increasing the number of the curve corresponds to increasing the distance between the source of the growth material and the growth layer. Solid lines are the calculated results. Points are the experimental results from references Ivanov & Smirnov, 2012; Zherikhin et al., 2003

4. Conclusion

An analytical approach (based on the solution of partial differential equations) was developed for the prognosis of mass and heat transport during the growth of epitaxial layers under pulsed laser deposition. The approach provides the opportunity to make a considered prognosis with a spatial account and, at the same time, temporal variations of their parameters as well as the nonlinearity of the considered process. Based on the approach, the influence of the variation of several parameters on the growth process for the improvement of properties of epitaxial layers was disscused.

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