Models of the formation of roughness on the surface of materials and the movement of molten flows under the laser melting processes

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Abstract. We used the nonlinear hydrodynamic model of the motion of molten flows to determine the basic geometric characteristics of the resulting coatings in a laser experiment. To test the capabilities of the developed algorithm, a series of calculations was performed for various values of Reynolds numbers (Re) and certain sizes of the computational domains with several cavities with different configurations. The influence of the channel geometry is clearly noticeable in the case of a deep depression. The direction of motion of different structures (e.g. the vortex) was predicted. Using the convection model, we estimate the temperature field and velocity distribution in the system under consideration for various initial conditions and experimental regimes selected. The profile of the arising island on the surface was modeled by a scaling computational domain with the number of particles \( N = pS \), where the parameter \( p \) – determined the viscosity of melted material, and \( S \) – the area of the computational domain. The influences of the two key parameters, i.e. probability of adhesion (sc) and viscosity (p), on the obtained surface profile of materials can be estimated by our approach, and result in different deposition images, being finally principal for functional characteristics of the samples under the specific regimes of laser treatment.

Introduction
Today, new unique materials, structured by nanoparticles into the main matrix of the substance and / or onto the surface, form the basis for the development of promising materials with predetermined properties that can be widely used in various fields of nanoelectronics and photonics as a new element base, for example, information storage elements and energy, thermoelectric sensors, detectors, radiation sources, solar panels, etc. The physicochemical properties of such materials substantially depend on the location of structural features on the surface, which makes it possible to control their properties. In this regard, the development of methods for controlling the placement of nanoscale elements on a solid substrate becomes a very important task. Existing methods of substance transfer are technologically complex and expensive, and so impose significant restrictions on the choice of the working material used.

For these reasons, laser-controlled synthesis of nanostructured surfaces is one of the fastest growing tools of modern micro-nanoelectronics and photonics, which allows one to obtain a wide class of nanostructured materials with the required surface topology due to the controlled motion of the laser beam.
Moreover, the use of substrates with complex geometry allows the synthesis of complex topological systems - periodic and/or random.

A large number of studies have been carried out in this area (see, for example, works of a survey type [1-5]), but clear models for the implementation of technologies for the targeted synthesis of given structures in a laser experiment, as well as descriptions of their relief features, have not yet been developed. This article discusses one of the possible approaches along this path with respect to fractal island metal nanofilms.

**Results of experiment**

To create nanofilms, we used the method of thermal diffusion deposition onto glass of a substance from a colloidal solution (Fig. 1), Obtained by intensively mixing noble metal nanoparticles (Ag, Au, and/or Ag/Au mixtures, with an average size of 100 nm) with glycerin [6]. The experimental scheme is presented in Fig. 1.

![Figure 1. Schematic diagram of the experiment on laser deposition of metal nanoparticles from a colloidal system: 1 - laser radiation, 2 - cuvette with a colloidal solution, 3 - substrate, 4 - track deposition of a nanostructured surface when the laser beam is displaced.](image)

As a result of local laser exposure to the colloidal solution, sintered noble metal nanoparticles of the above composition were deposited on the substrate surface along the path of the laser beam with the formation of island structures. Their parameters depended on the number of passes of the laser during the deposition process. We used a YAG: Nd3+ laser (λ = 1.06 μm) with a laser pulse duration of 100 ns, an average radiation power of 2.5 W, and a pulse repetition rate of 20 kHz; the diameter of the laser beam on the surface of the substrate on which the formation of nanostructures took place was 50 μm. The laser beam moved along the surface of the substrate at a speed of 0.4 mm/s to 5 mm/s. The obtained nanostructures were studied using a Quanta 200 3D scanning electron microscope and an Integra-Aura probe nanolaboratory (Fig. 2)[7].

![Figure 2. Images of deposited structures in rarefied island films: Ag/Au (a); Au (b); Ag (c) obtained by electron microscope.](image)

![Figure 3. Images of the surface of a layer deposited on a periodic substrate.](image)

During deposition, cluster films with an average height of 10 to 50 nm were formed. When colloids are deposited from pure gold and silver, various fractal cluster structures are formed, for example, dendrites (Fig. 2 a, b) or stars (Fig. 2 c). In agglomerates, individual particles 30–100 nm in size are well distinguished. Their values exceed the particle diameters in the initial colloid, which indicates thermal diffusion and particle aggregation prior to deposition. Fig. 3 shows the image obtained using...
an atomic force microscope of a surface of the deposited layer using a substrate with a periodic relief formed by a system of rectangular depressions. Thus, it is obvious that the use of substrates with a periodic system of depressions makes it possible to order the deposited structures [8].

**Molten Flow Model**

The distribution of fusion products is conveniently presented in the hydrodynamic approximation [9]. Calculation of the hydrodynamic model as a first approximation allows us to determine areas with concentrations of laser products sufficient for the formation of well-formed islands. In the framework of this model, we consider the stationary flow of a viscous incompressible fluid across the channel structure in their infinite length with upper boundaries in the form of horizontal surfaces. The channel geometries were considered where in the computational region of the lower surface there is an inhomogeneity in the form of an infinite rectangular (Fig. 4 a) and triangular (Fig. 4 b) depression.

![Figure 4. The computational domain with heterogeneity in the form of rectangular (a) and triangular (b) troughs.](image)

The hydrodynamic model is described by the stationary Navier-Stokes equations for the plane geometry problem [10-11]. The mathematical model represents a system of dimensionless equations for the stream and vortex functions [12]. The boundary conditions on the left and right transverse boundaries of the channel were set based on the Poiseuille flow and adhesion conditions for the lower and upper walls; the vortex was specified by the Woods formula [13]. The equations for the model used were discretized on a uniform network, using a conservative scheme with a five-point pattern. [14] The system of equations for the solution values at the grid nodes was solved by the relaxation method [15].

**Model of temperature field and velocity distribution**

It is convenient to take into account the influence of thermal factors in the framework of the Rayleigh-Benard convection model for plane geometry in the Boussinesq approximation in a dimensionless form [16]. The system of Boussinesq equations is solved by the method of lattice Boltzmann equations [17] in the Batnagar-Gross-Crook approximation (BGK) [18]. The Lattice Boltzmann method is implemented on a two-dimensional lattice with nine velocity channels for the Navier-Stokes equations and five for the heat equation [19].

**Island Profile and Roughness Model**

To describe the structure of island film aggregates, a diffusion limited aggregation (DLA) model was chosen [20] taking into account the viscosity of the medium and the probability of particle adhesion to the boundary. In the simulation, the initial concentration of aggregation centers (center and lower boundary — Figs. 5 a, b) and particle coalescence probabilities within the two-dimensional von Neumann neighborhood of order 1 [21] (Fig. 5(c)) were varied.

Scheme Fig. 5a allows you to simulate nanofilms - a top view - and evaluate its length, width, and the diagram in Fig. 5b - as a side view - to assess the roughness - the thickness and height of the inhomogeneities.
Figure 5. Island model: DLA scheme (a) - formation of a computational domain from the center, diffuse limited aggregation scheme; (b) - formation from the lower boundary; (c) - a von Neumann neighborhood of order 1.

Simulation results
The hydrodynamic model and deposition were used to determine the basic geometric characteristics of the coating obtained by us in the experiments. To test the capabilities of the developed algorithm, a series of calculations was performed for various values of Reynolds numbers (Re) and the characteristic dimensions of the computational domain with several troughs of the same type.

Figure 6. Channel flow: with three triangular depressions at Re = 10 (a), Re = 100 (b), Re = 1000 (c); with one rectangular long shallow hollow Re = 1000 (d); with four rectangular depressions Re = 1000 (e); with one rectangular Re = 100000 (f).

Calculations showed that for small Reynolds numbers, the main flow captures the cavity (Fig. 6a), while moderate - in addition to the main flow, a vortex forms in the channel in the channel (Fig. 6b, c). The motion in this vortex is carried out clockwise, i.e. in the area of contact of these currents, they are directed in the same way. An analysis of the flow results in the presence of a “long cavity” shows that the vortex occupies the entire cavity, however its center shifts to the right boundary (in the direction of the main current) (Fig. 6d.) With an increase in the Reynolds number, the vortex seems to expand, occupying the entire depression and “displacing” the main flow into the channel (Fig. 6c). The influence of the channel geometry is clearly noticeable in the presence of a deep depression - another one forms under the main vortex (Fig. 6e, f). The direction of motion in this vortex is opposite to the direction of motion of the first vortex.

Applying the convection model, we will estimate the temperature field of the system under consideration under various initial conditions. The initial temperature was calculated as the average of the temperatures of the upper and lower boundaries of the computational domain. The Prandtl number
was chosen equal to 1, the Rayleigh number equal to 2000, which corresponds to the conditions for the occurrence of convection (Fig. 7). Below are the graphs of the temperature field and relative velocities for the case of heating the lower wall to 5000 °C (Fig. 7 a, b) and 3000 °C (Fig. 7 c, d).

![Graphs](image)

**Figure 7.** Graphs of the temperature field (left) and the velocity field (right) when heating the lower boundary to 5000 °C (a), up to 3000 °C (b).

The presence of vortices in the velocity field, which are localized in the center of the computational domain, was revealed. The central vortices are elongated vertically, and the nature of the vortices is symmetrical about the horizontal axis. In addition, two central vortices have extensions in the upper right part; In the lower part of the computational domain, there are two small vortices, which indicates the presence of instability near the heated substrate.

Thus, by analyzing the graphs of the stream function, the temperature field, and the velocity field, it can be assumed that deposition and formation of film islands is most likely in the depressions of the substrate (Fig. 8). In this case, the deposition zones are marked by broken lines ABCD (Fig. 8 a) for the rectangular depression and EFG for the triangular (Fig. 8 b).

![Area of deposition](image)

**Figure 8.** Area of deposition: rectangular (a), triangular (b) hollows.

Fig. 4 shows images of fractals of the maximum, average and minimum rates of innovative development, with a single center of aggregation in the middle of the computational domain. From fig. 4 we can conclude that at rates above the average and average, a jump can be achieved due to the large coverage of the computational domain with innovation, on the other hand, with general trends below the average, the jump will be difficult to achieve (Fig. 4 g, h).

Let us model islands in these regions in the DLA approximation. Figure 9 shows the island profile for a shallow rectangular depression in which the nanostructure is deposited only on the lower boundary. In the framework of the model for such a case, the lower boundary of the computational domain is chosen as the seed structure (Fig. 5 b). The profile of the island of the film was modeled on a rectangular computational domain of 100 rel. units, the number of particles was set as \( N = p \times S \), where the parameter \( p \) - determined the viscosity, and \( S \) - the area of the computational domain. The influence of the probability of adhesion (sc) and viscosity (p) on the film profile can be estimated from fig. 9.
Figure 9. The center of aggregation is the line at the lower boundary of the computational domain: a) \( p = 0.01, \text{sc} = 0.1 \); b) \( p = 0.1, \text{sc} = 0.5 \); c) \( p = 0.5, \text{sc} = 1 \); d) \( p = 0.8, \text{sc} = 1 \).

With this deposition of particles, the resulting structure resembles “sticky fingers” - clusters grow from the surface of the substrate into the bulk of the colloidal system. The distortion of the structure is due to the fact that at the height corresponding to the area of the particle throw-in, "gaps" are formed. With an increase in the probability of adhesion, the film profile becomes smoother and stretches in the horizontal direction, and the computational region becomes more filled in the vertical direction, which indicates an intensification of the island formation process. At low values of the probability of adhesion, the profile of the island is more heterogeneous, the computational domain is weakly filled, which is confirmed by the values of fractal dimension. Figure 10 shows the island profile for the case of a deep depression, in which the nanostructure is deposited on the lateral and lower boundaries. In Fig. 10 e, the deposition of the film on the border of a triangular depression is shown; 10 e - deposition in the corner of a triangular depression.

Figure 10. Aggregation centers - a line on the lateral and lower boundaries of the computational domain: a) \( p = 0.1, \text{sc} = 0.1 \); b) \( p = 0.1, \text{sc} = 0.5 \); c) \( p = 0.5, \text{sc} = 1 \); d) \( p = 0.8, \text{sc} = 1 \); e) \( p = 0.5, \text{sc} = 1 \).

Similar structures are also observed here - viscous fingers, and the film is generated faster and the viscosity coefficient characterizing the particle velocity is mainly influencing this process. In fig. 11 is a plan view of simulated films.

Figure 11. Model films top view: 5 islands, \( p = 1, \text{sc} = 0.1 \) (a); 2 islands, \( p = 0.1, \text{sc} = 1 \) (b); 2 islands, \( p = 0.1, \text{sc} = 0.1 \) (c); 5 islands, \( p = 0.5, \text{sc} = 1 \) (d).

With a decrease in the probability of adhesion, the resulting fractal structure becomes almost uniformly distributed at the edges. In the case of growth of aggregates from an initially homogeneous
system (Fig. 11), several clusters arise simultaneously, and their growth is realized due to the absorption of small particles, as well as the adhesion of already formed structures to each other. Such conditions make it possible to obtain well formed island films at the final stages of experimental studies.

**Conclusion**

The proposed models of the motion of laser interaction products in the hydrodynamic approximation and the models of product deposition within the framework of diffusely limited aggregation make it possible, as a first approximation, to assess areas where the formation of nanofilm islands at the “general structural level” is possible. The island profile model in the DLA approximation complements the algorithms mentioned above and illustrates the film structure at the “level of behavior of an individual particle”.

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