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Laser deposition of large-area thin films

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***Abstract*. A new method for fabricating large-area thin films of uniform thickness on a rotating substrate is proposed. Its distinctive features are (i) the presence of a diaphragm, partially transmitting the evaporated material, between the target and substrate and (ii) the translatory motion of the rotating substrate with respect to the target at a certain velocity. The method proposed makes it possible to obtain thin films of uniform thickness on substrates with sizes limited by only the deposition chamber size. The method is experimentally verified by depositing thin CuO films on silicon substrates placed over the radius of a disk 300 mm in diameter. The deviation of the film thickness from the average value does not exceed ±3 % throughout the entire radius, which confirms good prospects of this method for microelectronics, optical industry, and other modern technologies.**

Keywords: *pulsed laser deposition, large-area films, uniform thickness.*

1. Introduction

Pulsed laser deposition (PLD) is used to grow thin films of metals, oxides, polymers, biocompatible materials, etc. It provides simultaneously a high deposition rate, a good correspondence between the target and film compositions, and a possibility of changing pressure in the deposition chamber in a wide range; moreover, it allows one to fabricate ultrathin epitaxial single-crystal, polycrystalline, and amorphous films; heterostructures; and nanocrystalline coatings [1 – 3]. PLD is especially valuable for obtaining films of multicomponent compounds, which are difficult to synthesise by other methods. In particular, this method was applied to synthesise first films of high-temperature superconductor $YBa_2Cu_3O_{7-x}$. PLD is simple in application and, therefore, is widely used in research laboratories. However, it is also promising for various commercial applications, in particular, growth of large-area films.

Films of uniform thickness on large-diameter substrates are necessary for many applications in microelectronics, optical industry, and other modern technologies. Wide use of PLD in the growth of large-area films is impeded by the following circumstance: the angular distribution of the mass-transfer rate in the plasma plume formed by laser radiation is nonuniform. Therefore, using conventional laser deposition, one cannot obtain films of uniform thickness on substrates more than 30 mm in diameter. In this study we describe some existing solutions to this problem and propose a new technique for depositing thin films of uniform thickness on substrates whose sizes are limited by only the deposition chamber dimensions.

All versions of PLD of large-area films are based on the fact that the angular distribution of the mass-transfer rate in a plasma plume is set by the function $F(\theta) = A\cos^m\theta$ [1], where θ is the angle of deviation from the perpendicular to the target plane. The plasma plume axis, i.e., the direction in which the mass-transfer rate is maximum, is perpendicular to the target surface in a wide range of variation in the angle of incidence of laser beam on the target. Knowing the angular distribution of the mass-transfer rate of the material evaporated from the target, one can arrange the mutual position and motion of the target and substrate so as to provide identical amount of evaporated material per substrate unit area throughout the substrate surface and thus grow a film of uniform thickness.

2. Techniques for depositing large-area films

Let us briefly consider the main existing ways for growing large-area films. In one of them a laser beam is incident on a rotating target, before which a rotating substrate is located parallel to the target [1]. The substrate axis is shifted with respect to the plasma plume axis by some distance d (Fig. 1a). This distance is determined by the width of the gap between the target and substrate, the substrate diameter, and the angular distribution of evaporated particles in the plasma plume. Thus, the part of the plasma plume that is characterised by a higher mass transfer rate of evaporated material arrives at the substrate edge, where a larger area must be coated for the same time; as a result, a film of uniform thickness grows.

A modification of this technique is the version schematically shown in Fig. 1b. Here, the substrate simultaneously rotates and moves in the horizontal plane. A computer controls the horizontal displacement velocity of the substrate in such a way that the plasma plume axis is directed toward the substrate edge for a longer period as compared to the centre.

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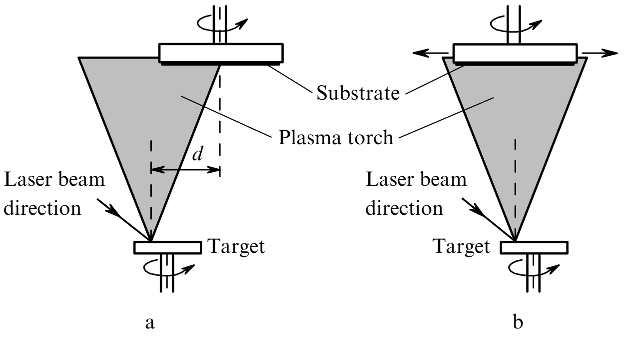

Figure 1. Schematics of the main techniques for depositing large-area films: (a) 'off-axis' deposition and (b) deposition on substrate that simultaneously rotates and moves in the horizontal direction.

The drawback of the first version (Fig. 1a) is the spatial confinement of the ablation plasma plume. Substrates whose diameter exceeds some limiting size are not overlapped completely by the plume region where the mass-transfer rate is sufficiently high. This drawback can be compensated for by mounting the substrate at a larger distance from the target; however, the larger the substrate – target distance, the lower the deposition rate. In addition, the stoichiometry of multicomponent films can be preserved only in a certain range of variation in the substrate – target distance.

The drawback of the second version (Fig. 1b) is the necessity of preliminary analysis of the angular distribution of the mass-transfer rate in the plasma plume in specific geometry and under specific deposition conditions. In turn, the angular distribution of the mass-transfer rate may vary during deposition, because it depends on several parameters, which may also vary during long-term deposition [1, 4].

There are many modifications of the above-described techniques. We proposed two relatively simple and inexpensive PLD modifications for films up to 150 mm in diameter: 'mask' and 'sweeping target' techniques [5 – 8]. The geometry of the 'mask' technique [6, 7] is shown in Fig. 2. A laser beam is incident on a rotating target to form a plasma plume, as in the conventional PLD method. Before reaching the substrate, which rotates around the vertical axis, the evaporated material must pass through a slit of peculiar configuration in a mask located in the immediate vicinity of the substrate.

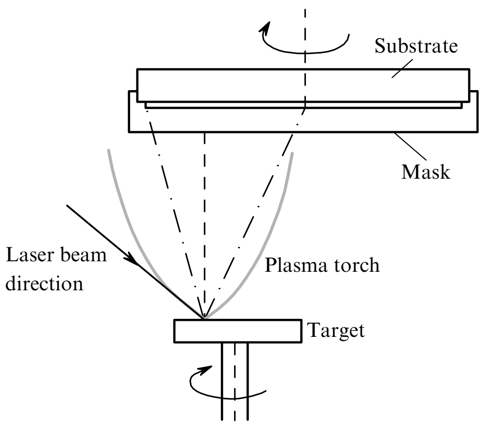

Figure 2. Geometry of the 'mask' technique.

Different slit configurations, aimed at depositing films of uniform thickness, were considered in [5 – 7]. The data on the mass transfer angular distribution in a plasma plume were used to calculate the exact sizes of the slits in masks.

A specific feature of the 'sweeping target' technique is the programmable target rocking around the axis parallel to the substrate plane, whereas the laser beam, focal spot, and substrate are immobile with respect to each other [8]. As shown in Fig. 3a the laser beam is incident on a target, which rocks around the AA' axis passing through the target centre (point O). As a result, the plasma plume axis can be scanned over the substrate along its radius. The substrate rotates around the E axis. The laser beam propagation direction is perpendicular to the AA' axis. Controlling the time during which the target remains in this oblique position, one can control the amount of the material deposited on each specific portion along the substrate radius $O'B$.

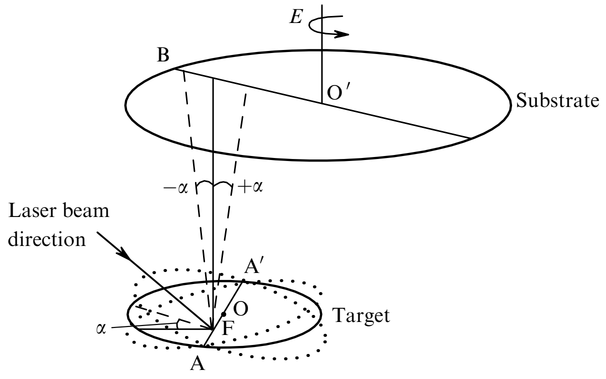

Figure 3. Geometry of the 'sweeping target' technique.

The latest achievements in growing large-area films by PLD were described in [9]. As previously [1], the main technique is based on scanning from a large target.

Generalising the above-described methods, we can state that none of them allows one to obtain large-area films whose sizes would be limited by only the deposition chamber dimensions. To solve this problem, we propose a new PLD version.

3. Technique for depositing thin films of arbitrary size

Figure 4 shows schematically the deposition geometry for large-area thin uniform films, whose transverse sizes are limited by only the deposition chamber dimensions [10].

A laser beam is incident on a rotating target placed in a vacuum chamber. A diaphragm with a cut slit is located in the path of evaporated material between the target and substrate to select the plasma plume region where the mass-transfer rate is constant and maximum. Specifically this part of the plasma plume falls on the uniformly rotating substrate. It can be seen in Fig. 4 that the substrate undergoes translatory motion with respect to the target. Obviously, in this geometry the substrate motion can be replaced by the joint motion of the target and diaphragm.

To grow a film of uniform thickness over the entire substrate surface, it is necessary to determine the motion law for the substrate. Figure 5 shows the dependences of the film

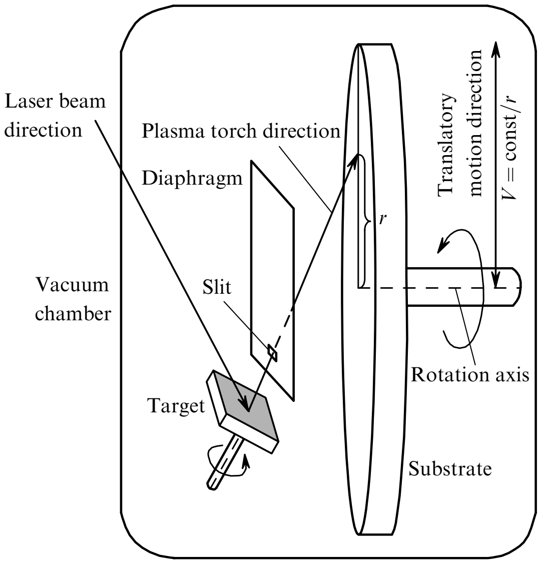

Figure 4. Schematic of the new technique for depositing thin films of arbitrary sizes.

thickness h on the distance r from the substrate centre to the point at which the evaporated material arrives at a given instant, calculated for different laws of change in the substrate velocity V. It can be seen that the films are uniform over thickness at $V = \mathrm{const}/r$. This means that the arrival time of the flux of evaporated target material for the substrate points located closer to the centre is shorter than for the more remote points (the larger the distance r from the substrate centre, the larger the area that must be coated for the same time).

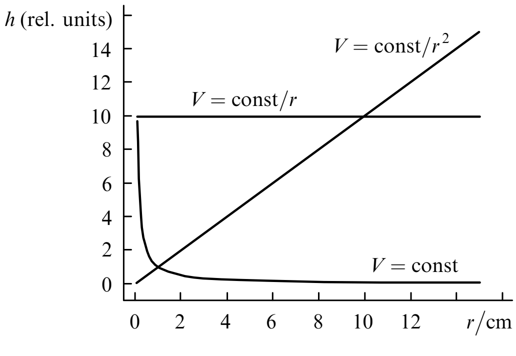

Figure 5. Calculated dependences of the film thickness h on the distance r from the substrate centre to the point at which the evaporated material arrives at a given instant for different laws of change in the substrate velocity.

The calculation results were experimentally checked by depositing thin CuO films on silicon substrates 30 mm long, located over the radius of a disk 300 mm in diameter. The target and substrate rotation speeds were 37 and 2 rpm, respectively. The deposition was performed using the third harmonic of a Nd^{3+} :YAG laser ($\lambda = 355$ nm) with the following pulse characteristics: energy, 15 mJ; width, 10 ns; and repetition rate, 20 Hz. The angles between the target plane, laser beam axis, and diaphragm plane were chosen so as to make the diaphragm select a particle beam oriented perpendicular to the target from the plasma plume. At any changes in the deposition parameters during a long-term process in the chosen geometry the mass-transfer rate of the target material to the substrate will be maximum and the deposition time of a film of specified thickness will be minimum.

The surface profile (AMBIOS XP-1 profilometer) of a CuO film deposited on a substrate transferred with a velocity $V = \mathrm{const}/r$ is shown in Fig. 6. The average film thickness is 85 nm. The thickness deviation from the average value did not exceed ± 3 % over the entire substrate surface.

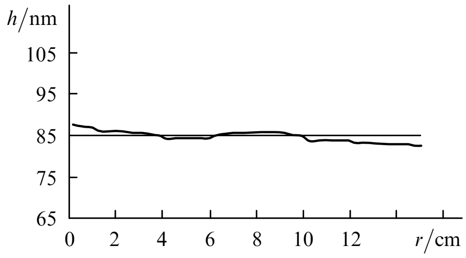

Figure 6. Surface profile of a CuO film deposited on a substrate transferred at a velocity $V = \mathrm{const}/r$.

Due to the translatory substrate motion; specific mutual position of the target, substrate, and laser beam; and the use of a diaphragm, this method imposes no limitations on the film size and makes it possible to deposit films on substrates of arbitrary diameter.

4. Technique for depositing films of very large size

To reduce the deposition time for films on substrates of very large size, one can modify the above-described way as follows: use several targets instead of one and scan a laser beam over them, applying target after target for deposition. If the laser pulse repetition rate increases in correspondence with the number of targets, the deposition from each target will be performed at the same frequency as in the case of one target. Each target must have its own diaphragm, moving simultaneously with the target at a velocity $V = \mathrm{const}/r$.

Suppose we are going to use n targets to deposit a film on a substrate with a radius R. Obviously, the targets must be located with respect to each other and with respect to the substrate in such a way as, having started deposition from all targets simultaneously and moving each target at a velocity $V = \mathrm{const}/r$, to deposit a film on the entire substrate surface and finish deposition from all targets simultaneously. It can be shown that, to deposit a film of uniform thickness on a substrate of radius R, the n targets must be initially located so as to provide a distance $R\sqrt{l-1}/\sqrt{n}$ from the substrate centre to the point of intersection of the perpendicular dropped from the first target with the substrate plane.

The use of n targets will reduce the deposition time of a film of specified thickness by a factor of n.

5. Conclusions

A new version of PLD of large-area films of uniform thickness is proposed. Its main feature is the possibility for growing thin films on substrates of extremely large size. It is implemented using translatory substrate motion with respect to the target in a certain velocity regime and selecting beams with identical and maximum mass-transfer rate from the plasma plume. Although the latter circumstance somewhat decreases the deposition efficiency, it allows one to make this process more controlled and less dependent on possible changes in the deposition parameters during the long-term process. The deposition time can be reduced using several targets.

The technique proposed is novel and rather simple. We expect the area of its application in various new high-tech fields, along with other latest achievements in the laser deposition of thin films, to constantly increase.

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