Pulsed Laser Deposition: From the Laboratory to the Industry

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ABSTRACT

Simple and inexpensive methods of obtaining large area uniform in thickness and composition thin films on large and very large rotating substrates and moving ribbons through pulsed laser deposition have been proposed. Thin films of different composition were deposited with these methods. The thickness uniformity of films obtained with these methods was preserved within the limits of ±3% on up to 300 mm diameter substrates. Also a method of creating a laser spot of a certain configuration on the target is proposed, allowing almost full utilization of the target material.

Keywords: pulsed laser deposition, large area films, uniform, thickness, composition

1 INTRODUCTION

The wide application of thin films in newest technologies has revealed the need of developing simple methods of obtaining large area thin films. One of these methods is provided by pulsed laser deposition (PLD). It equally provides high speed of deposition, good ratio between the compositions of the target and the film, alteration possibilities in a wide area of gas pressure in the deposition chamber. Main methods of large area film laser deposition have a common shortcoming, namely, significant complication of deposition apparatus [1, 2]. Most of them employ mutual motion of the laser beam, target and substrate. The proposed methods against this background stand out because of particular easiness and reliability.

2 DEPOSITIONS ON ROTATING SUBSTRATE

Three relatively simple methods of laser deposition of large area thin films are proposed.

The first method employs controlled tilting of the target around the axis parallel with the substrate plane, with the respective positions of the laser beam, focal spot, and substrate being kept constant. The peculiarity of the second and third methods is laser deposition of the compound upon a substrate through a mask. The possibilities of different configurations of the slit in the mask are considered. One method uses a mask with a slit in the form of a sector, which is symmetrical with regard to the substrate radius, with various angular dimensions at different distances from the rotation center of the substrate. Another method uses a mask with two slits in the form of a bent sector, the symmetry axis of which coincides with the line of equal velocity of mass transfer of the deposited compound onto the substrate.

2.1 The method of tilting target

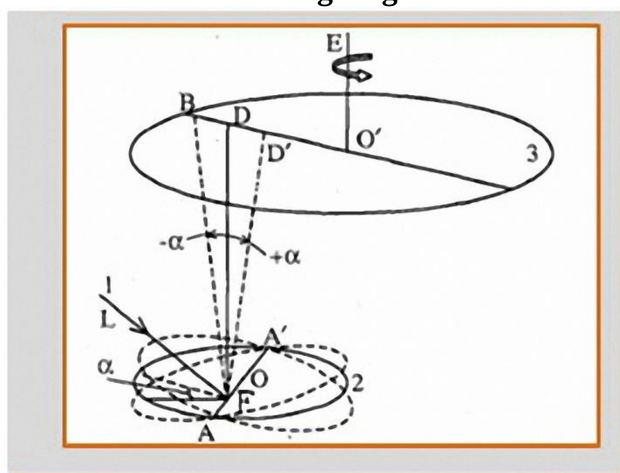

Fig. 1. Geometry of tilting target method: 1- laser beam, 2 – target, 3 – substrate.

In the proposed method the axis of a plume is directed to various radial sections of the substrate [3]. Fig.1 sketches the essence of the method proposed and some of its features. The laser beam falls upon the target which is tilted around the axis perpendicular to its plane and passes through the center of the target O. Focal spot F is on the AOA´ axis, around which the target can be tilted. The AOA´ axis is parallel with the substrate plane, which rotates around the axis O´E. When target plane is parallel with the substrate plane, the normal, which is the plume axis, to the target in the point F, crosses the substrate plane in point D on the radius of the substrate O´B. At target's slating from the position parallel to the substrate by tilting around the axis AOA´, point D travels along the radius O´B.

The angular distribution of ablated material depends on many factors. Thus, in our experiments, we tried to change only one parameter, leaving others intact. As variable parameters we chose the target slant angle (*a*) and the laser beam diameter (d) in the target plane. Fig.2 shows the change of the relative film thickness along the substrate

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radius during different deposition processes. It follows from the data in Fig. 2a,b that the tilting target method allows achieving more intensive deposition both on the edge of the substrate and on its center. Thus, by using of superposition of two angles of depositions, it is possible to obtain film thickness uniformity (Fig. 2c). The thickness nonuniformity in this case did not exceed ±3.3%.

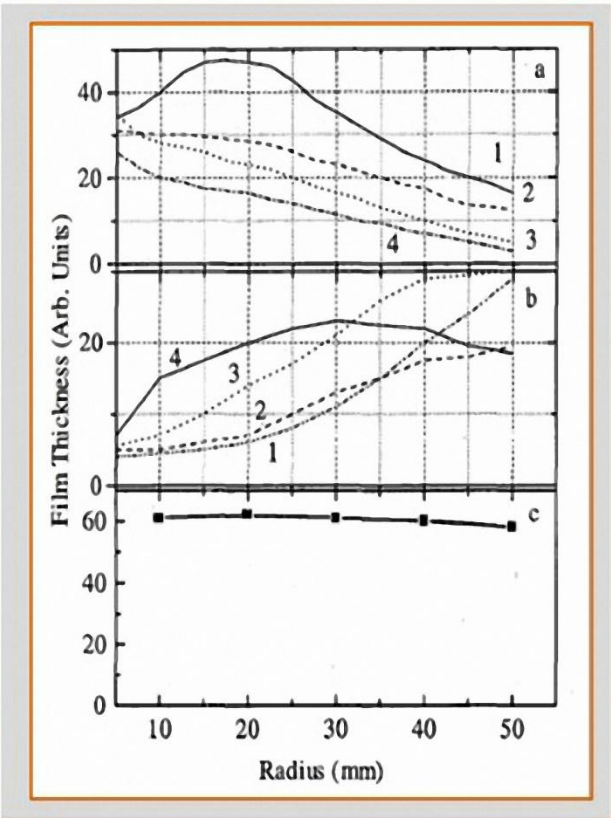

Fig. 2. Dependence of relative film thickness on substrate radius.
a) α=+18°: d = 0.8mm (1); 0.65mm (2); 0.45mm (3); 0.3mm (4);
b) α= –30°, d=1.05mm (1); α= –24°, d=1.05mm (2); α= –30°, d=0.8mm (3); α= –24°, d=0.65mm (4);
c) α= –24°, d=1.05mm and α=+18°, d=0.45mm.

2.2 Mask method of deposition

We have proposed two principally different solutions to the task of obtaining large area films on a rotating substrate [4, 5]. The first solution is based on PLD through the slit in the mask with a certain angular size for each radius. Slit configuration is calculated on the basis of angular dependence of the mass transfer so that to secure the thickness uniformity of the deposited film. The second solution employs sector-shaped slit(s) with a distorted symmetry axis coinciding with the thickness uniformity line of the film deposited on stationary substrate. Fig. 3 shows the geometry of the mask method. It can be seen that it differs from the usual "Off Axis" deposition (when the center of rotating substrate is located at a fixed distance from the axes of the plasma plume) in that the deposition is done through a mask placed in the direct vicinity of the substrate.

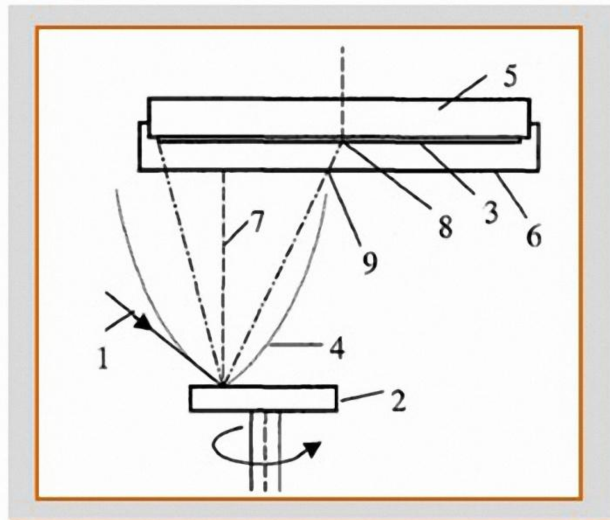

Fig. 3. Geometry of mask deposition on a rotating substrate:
1 – laser beam, 2 – target, 3 – substrate, 4 – plasma plume, 5 – substrate holder, 6 – mask, 7 – plasma plume axis, 8 – substrate rotation center, 9 – intersection point of the line connecting the laser spot and the center of substrate with the mask surface.

2.2.1 Sector-Shaped Slit With Varying Angular Dimensions

The present method implies deposition done through a mask with a symmetrical sector-shaped slit with various angular dimensions at various distances from the substrate rotation axis. The vertex of the sector must be in the intersection point of the line connecting the laser spot and the center of substrate rotation with the mask surface. The slit symmetry axis is parallel to the substrate radius. The plume axis is perpendicular to the slit symmetry axis. Fig. 4 shows variation of the relative thickness of films along the substrate radius for different deposition processes. Curve 1 shows the case where the mask and substrate rotations are absent. Curve 2 corresponds to the deposition through a sector slit on the rotating substrate. Predictably, in this case we obtain thickness nonuniformity, too. Curve 2, however, shows where the sector must be narrowed, and where, conversely, widened. Having made several depositions with different masks, we have succeeded in finding the mask configuration (Fig. 4b), which provides for thickness uniformity of films (curve 3).

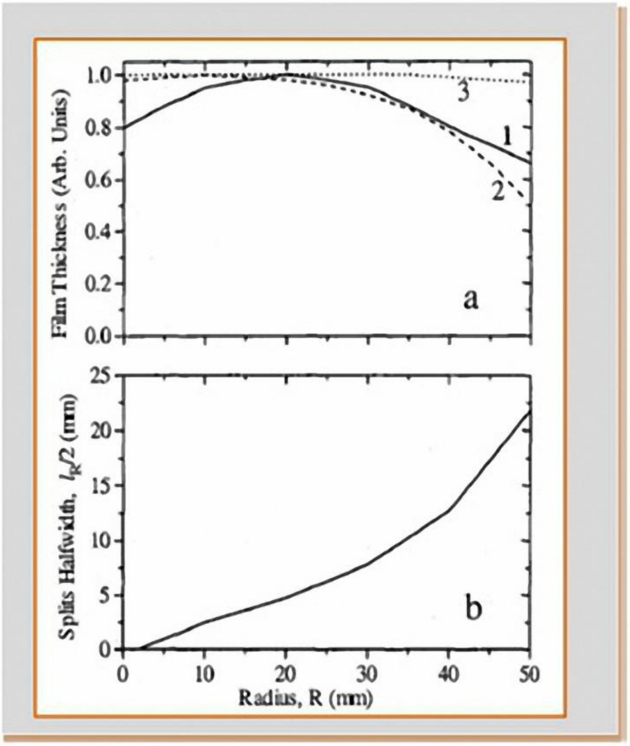

Fig. 4. Radial distribution of film thickness (a), slit configuration which provided film thickness uniformity (b)

2.2.2 Distorted Symmetry Axis Sector Slit

This method is based on the following fact: in order to obtain a film of uniform thickness on a rotating substrate, the mask slit must be in the form of a sector. This is true for a uniform stream of deposit substance. On the other hand, in the case of PLD on a stationary substrate the film's uniform thickness line is usually an ellipse.

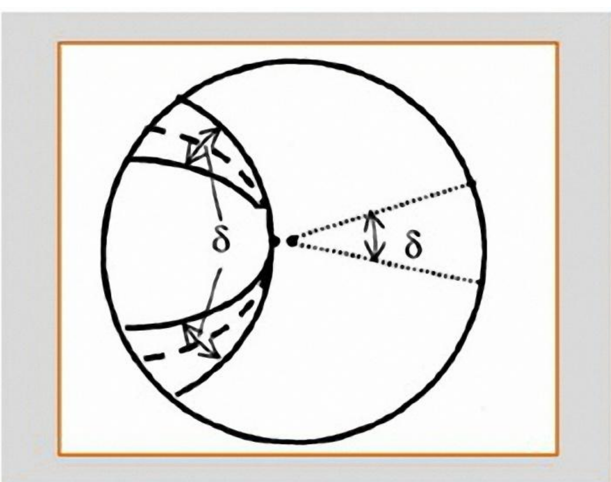

Fig. 5. Distortion of sectorial slit. The mask slit curved along the uniform thickness ellipse (dashed line) provides both thickness and composition uniformity of deposited films.

2.3 Deposition of very large size films

The new methods for fabricating large-area thin films of uniform thickness on a rotating substrate are proposed [6-8]. 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 and selecting beams with identical and maximum mass-transfer rate from the plasma plume. The methods proposed make it possible to obtain thin films of uniform thickness on substrates with sizes limited by only the deposition chamber size.

3 DEPOSITION ONTO RIBBON

The simple method of laser deposition of large area uniform-thickness films onto a moving ribbon of width up to 100 mm is proposed [9]. The peculiarity of the method is the laser deposition of compound upon a substrate through a mask placed in immediate proximity of the substrate. Various configurations of mask slits that provide thickness uniformity of deposited films are considered. The precise sizes of slits in the masks are calculated using the data of an angular distribution of mass transfer of a deposited compound in a plasma plume.

4 EFFECTIVE UTILIZATION OF THE TARGET MATERIAL

A simple solution to the problem of effective utilization of the target material for pulsed laser deposition of thin films is proposed [10]. The new device is suggested and developed for obtaining thin films by the method of laser deposition, which is specific in the employment of a simple optical system mounted outside a deposition chamber that comprises two lenses and the diaphragm and focuses the laser beam onto a target in the form of a sector-like spot. Thin films of CuO and Y-Ba-Cu-O were deposited with this device. Several deposition cycles revealed that the target material is consumed uniformly from the entire surface of the target. A maximal spread of the target thickness was not greater than ±2 % both prior to deposition and after it. The device designed provides a high coefficient of the target material utilisation efficiency.

5 CONCLUSIONS

Implementation of the both mask methods of deposition onto a rotating substrate has revealed a possibility of providing for thickness uniformity for both CuO and Y-Ba-Cu-0 films within ±3.3% on substrates with the diameter of 150 mm. The uniformity is an important feature of large area films and in usual deposition methods is within ±8% even after optimization. In our case it can even have better than ±3% range of values at the appropriate choice of the slit opening, though the deposition time becomes an issue. The Y-Ba-Cu-O films had high value of critical temperature of a superconducting transition (T_c=91K) regardless of the distance from the substrate rotation center, which indirectly indicates radial uniformity of the composition of the deposited films. The above relates more to the distorted sector slit mask method. While PLD may provide films with correct stoichiometry in more degree detailed investigations show that the film stoichiometry varies with the deposition angle θ. This is a serious problem for complex compounds PLD. Film stoichiometry is often satisfactorily uniform within a smaller range of θ. The distorted symmetry axis sector slit method provides that this requirement is met. Another advantage of this method is that the central part of plasma plume is cut off which decreases the density of micron size particles, typical for laser deposition, on the film surface. Thus we can conclude that the thickness uniformity of films obtained with these three methods was preserved within the limits of ± 3.5% on 150 mm diameter substrates.

The new device for laser deposition of thin films is suggested, substantiated, and experimentally tested, which simply and cardinally overcomes the main drawback of the PLD method, namely, a low coefficient of the target material utilization efficiency.

A more detailed description of the proposed methods can be found in articles [11-14].

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