LASER DEPOSITION

PACS numbers: 81.15.Fq
DOI: 10.1070/QE2013v043n12ABEH015194

A simple solution to the problem of effective utilisation of the target material for pulsed laser deposition of thin films

A.S. Kuzanyan, A.A. Kuzanyan, V.A. Petrosyan, S.Kh. Pilosyan, A.Z. Grasiuk

Abstract. **The factors determining the efficiency of the target material utilisation for pulsed laser deposition of films are considered. The target volume is calculated, which is evaporated in the ablation process by the focused laser radiation having a rectangular form. 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 YBaCuO 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.**

Keywords: *pulsed laser deposition, laser beam, focal spot, target.*

1. Introduction

The method of pulsed laser deposition (PLD) is widely used for obtaining thin films of various compositions, which is related with certain advantages over other deposition methods [1–3]. A commercial employment of PLD is hindered by its few drawbacks, main among those are a nonuniform thickness of the deposited film and a low coefficient of effective utilisation of the target material. A series of improvements have been performed to overcome the first drawback of the PLD method, such as mutual movements of the laser beam, target and substrate; an employment of simple or complicated additional constructions [4, 5]. The improvements suggested allow one to deposit the films of arbitrary size, which are uniform in thickness and composition. One may assert that the first drawback of PLD is successfully overcome.

Consider the second drawback in more details. In this method, the films are deposited due to ablation of the target material by a laser beam. Obviously, if both the target and laser beam are immovable, then a crater arises on the target in a certain time interval. The crater affects the angular distribution of the evaporated material, that is, the thickness of the film deposited per unit time on a certain area of the substrate. One more undesirable result is overheating of the target and distortion of its composition. The target is also overheated in the case when the laser beam is focused onto the entire surface of the target. In this geometry, no crater is formed in the target; however, the target undergoes overheating. Thus, in the case of PLD with immovable both the target and the laser beam, the thickness and composition of the film deposited would vary in time. There is a simple solution to the problem – rotation of the target, which substantially facilitates the situation preventing the target from overheating and prolonging the duration of deposition without disturbing the film composition and angular distribution of the evaporated material. However, such a geometry does not solve the problem yet. A groove arises in the rotating target (Fig. 1) and the angular distribution of the evaporated material changes with time [6, 7]. After several deposition cycles with a variation in the distance of the laser beam from the target centre, concentric grooves arise on the surface of the target and the latter becomes unsuitable for further employment. In the conventional geometry, utilisation efficiency of PLD (the fraction of its evaporated volume) is very poor (0.010.02). It is not important if the target material is cheap. However, if the films of rare metals and its alloys are deposited or the target is made of very-high-purity chemicals or isotopes, then such a low efficiency is inappropriate.

2. Ways of increasing the target utilisation efficiency

There are several known solutions to the problem for improving the target utilisation efficiency in PLD of thin films. The targets employed for deposition may have polished surfaces. In this case, up to 75% of the target material may be lost [8]. There are cardinal approaches, which, however, substantially complicate the deposition installation suggesting the computer-controlled scanning of the large target surface by a laser beam (Fig. 2a) or moving the target in two perpendicular directions relative to the fixed laser beam (Fig. 2b). In this case, the material ablation occurs from 90% of the target surface providing in this way a noticeable improvement in the utilisation efficiency [9].

The suggestion in [10] is simple to realise: it is assumed that the laser beam is split into four parts, each of them performing material ablation from a certain area of the rotating target. Thus, a wider plasma jet of the evaporated material is formed (which facilitates the uniformity of the film thickness)

A.S. Kuzanyan, A.A. Kuzanyan, V.A. Petrosyan Institute of Physics Research, National Academy of Sciences of Armenia, 0203 Ashtarak, Armenia; e-mail: akuzan@ipr.sci.am, kuzanian@yahoo.com, vahagn.petrosyan@gmail.com;
S.Kh. Pilosyan, A.Z. Grasiuk P.N. Lebedev Physics Institute, Russian Academy of Sciences, Leninsky prosp. 53, 119991 Moscow, Russia; e-mail: sp-93@mail.ru, Grasiuk2008@yandex.ru

Received 31 March 2013; revision received 1 November 2013
Kvantovaya Elektronika **43** (12) 1170–1174 (2013)
Translated by N.A. Raspopov

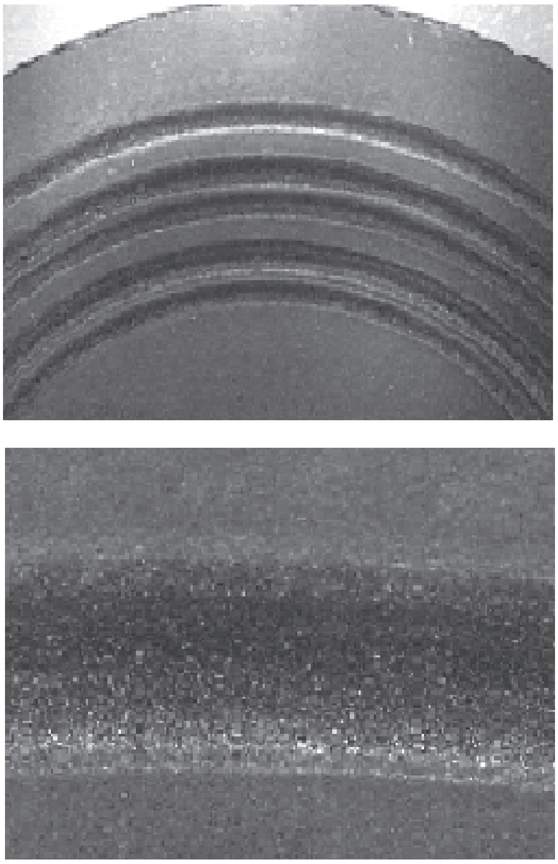

Figure 1. Target made of $YBa_2Cu_3O_{7-\delta}$ of diameter 50 mm after five deposition cycles performed by the conventional PLD method at magnification (a) $10^{\times}$ and (b) $60^{\times}$.

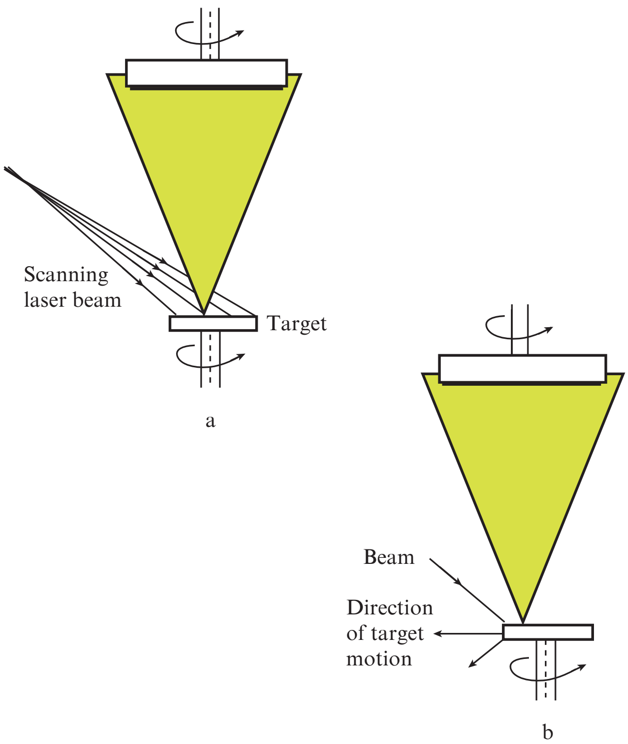

Figure 2. PLD schemes with the laser beam screening the surface of target (a) and target motion relative to the laser beam (b).

and ablation occurs from a larger surface of the target. From the viewpoint of enhancing the target utilisation efficiency, this approach is simply an increase in the focal spot dimensions. However, the grooves on the rotating target will still be produced with the following negative consequences.

A more reasonable suggestion was patented [11, 12]. The authors complicated the mechanism of target rotation in such a way that the target rotates around two parallel axes. In this case, the laser beam circumscribes a cycloid rather than a circle over the target surface, which will increase the target utilisation efficiency. Unfortunately, this approach has also a serious disadvantage (see below), which hinders obtaining high values of target utilisation.

A most simple solution is, evidently, a uniform distribution of laser radiation over the entire surface of a fixed target. Such a distribution can be obtained, for example, with raster focusing systems [13, 14]. Nevertheless, as mentioned above, such a scheme cannot be used for depositing multi-component compounds, because overheating of the target will affect the composition of the latter.

Thus, the known methods of enhancing a PLD process aimed at increasing the target utilisation efficiency either noticeably complicate the installation or do not solve the problem completely. In the present work we suggest a simple solution to the problem for attaining a maximal utilisation of the target material, capable of increasing the utilisation efficiency actually to unity.

3. Rectangular focal spot arranged along the target radius

One possible solution to the problem for increasing the target utilisation efficiency is a rectangular spot of the laser beam arranged along the radius of the rotating target, where the focal spot of width L has the length of at least the target radius (Fig. 3). The centre of one focal spot side coincides with the target rotation centre O, and the axis of symmetry coincides with the target radius (Fig. 4). At a first glance, it may appear that it is a simple and effective solution, because ablation of the target material will occur from the entire surface. But the quantity of the substance evaporated from a unit area is proportional to the energy passed on it. The sites of focal spot closer to the target centre will affect (per single round) a smaller area than those residing far from the centre. Consequently, the target material at its centre will be consumed faster than at periphery.

Let a laser beam fall onto the rotating target, which has the form of a disk with radius R and thickness h_0, forming the

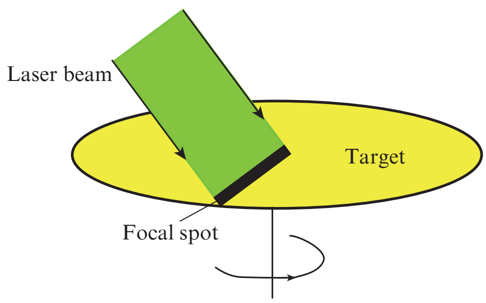

Figure 3. Geometry of the PLD method with the rectangular focal spot along the radius of the rotating target.

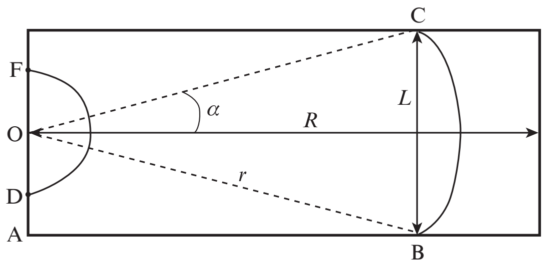

Figure 4. Scheme of the rectangular focal spot directed along the radius of the rotating target.

rectangular focal spot of width L arranged along the target radius. In time t_0 the target executes N revolutions and the laser burns a hole at the centre of the target. How much of target volume is ablated in this case?

According to Fig. 4, one should consider two domains of the target surface: inside the circle of radius $L/2$ and outside it. When the target rotates at a constant angular velocity ω, all the points inside the circle of radius OA = $L/2$ are exposed to laser radiation during half the process duration ($t_0/2$), because for these points the focal laser spot is a semicircle. In other words, inside the rectangular focal spot of width L the trajectory of any point residing closer than $L/2$ to the centre of rotation (point O) is a semicircle. For example, point D on the target surface is subjected to laser radiation as long as it follows semicircle DF (Fig. 4). Point B residing at a distance longer than $L/2$ is subjected to laser radiation until it reaches point C having passed the arced path BC.

By denoting the rate of target evaporation σ (the thickness evaporated per unit time), from the condition of burning the target to a throughout hole we may write

$$h_0 = \sigma t_0/2, \quad \sigma = 2h_0/t_0. \tag{1}$$

The points of the target surface outside the circle of radius $L/2$ at a distance r from the centre are exposed to laser radiation during the time interval Δt (a single revolution of target) so that after N revolutions the target thickness reduces by the value

$$h = \sigma \Delta t N = \sigma N|\overset{\frown}{BC}|/v = \sigma N|\overset{\frown}{BC}|/(\omega r), \tag{2}$$

where v is the linear speed of point B; $|\overset{\frown}{BC}| = 2ra = 2r \times \arcsin(L/2r)$ is the length of arc BC; and $\omega = 2\pi N/t_0$.

Then at $r \geqslant L/2$ the reduction of the target thickness will be

$$h = \frac{2h_0 \arcsin(L/2r)}{\pi}. \tag{3}$$

Dependences of $h(r)$ for various L are given in Fig. 5. For the points $0 < r \leqslant L/2$ we obtain $h = h_0$.

The volume of the ablated target material is determined by the rotation of curve $h(r)$ around axis h:

$$V = \pi h_1 R^2 + \pi \int_{h_1}^{h_2} \left[\frac{L}{2\sin(\pi h/2h_0)}\right]^2 \mathrm{d}h, \tag{4}$$

where $h_1 = [2h_0 \arcsin(L/2R)]/\pi$, and $h_2 = h_0$. This entails

$$V = 2h_0 R^2 \arcsin\left(\frac{L}{2R}\right) + h_0 LR\left(1 - \frac{L^2}{4R^2}\right)^{1/2}. \tag{5}$$

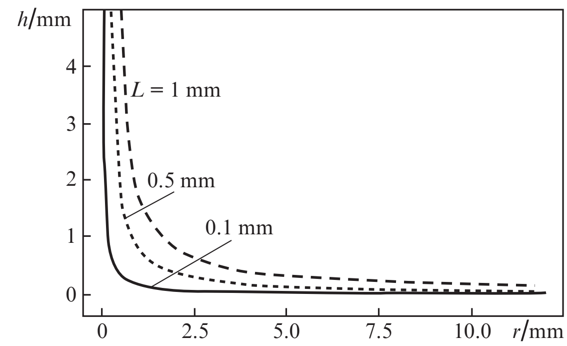

Figure 5. Target thickness h vs. radius r under PLD in the rectangular focal spot geometry.

One can see that the volume depends on three parameters: the target height h_0, target radius R, and width of the laser beam L.

Because the value of the target material utilisation efficiency is $\eta = V/V_0$, where $V_0 = \pi h_0 R^2$ is the target volume prior to ablation, in view of (5) we obtain

$$\eta = \left[2\arcsin\left(\frac{L}{2R}\right) + LR^{-1}\left(1 - \frac{L^2}{4R^2}\right)^{1/2}\right]\frac{1}{\pi}. \tag{6}$$

The value of η depends only on two parameters L and R. Dependences of $\eta(R)$ for various values of L are shown in Fig. 6. One can see that at a fixed parameter R the volume of the ablated part of the target is greater at longer L.

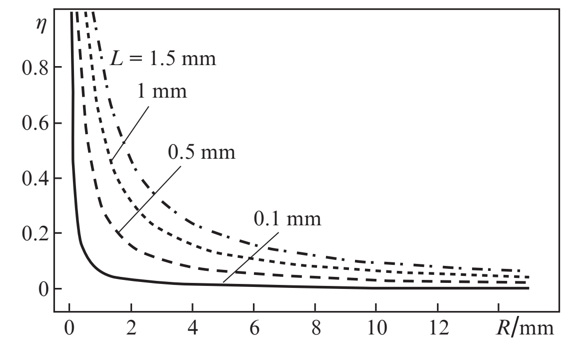

Figure 6. Target utilisation efficiency vs. target radius at various laser beam spot widths.

The expression for the target material utilisation efficiency may be simplified. By introducing the parameter $k = L/R$ we may write Eqn (6) in the form

$$\eta = [2\arcsin(k/2) + k(1 - k^2/4)^{1/2}]\pi^{-1}. \tag{7}$$

Recall that the calculations are performed for the case $R \geqslant L/2$. At $0 < R < L/2$ the equality $\eta = 1$.

Figure 7 presents the dependence $\eta(k)$. One can see that at $k < 1$ it is well approximated by the straight line according to the formula

$$\eta = 0.004 + 0.617k \approx 0.6k. \tag{8}$$

Thus, we have the simple expression for target utilisation efficiency with a sufficiently good approximation. Obviously, for

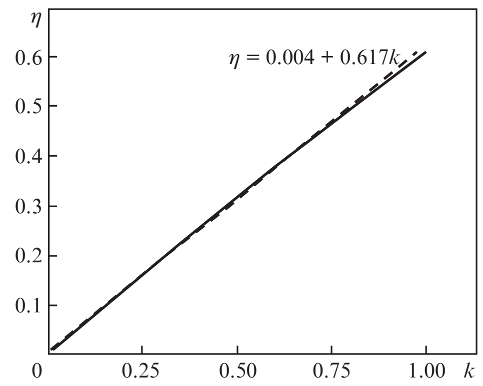

Figure 7. Target utilisation efficiency η vs. k. The dashed line refers to approximation.

experimentally actual values of L and R where k $\ll$ 1 the value of η will not be greater than 0.5.

Note that the method suggested in patents [11, 12] has a similar drawback. The cycloidal trajectory described by the focal laser spot on the surface of the target arising due to the rotation around two parallel axes will also result in more intense material evaporation from the central part of the target and reduced utilisation efficiency.

4. Focal spot in the form of a sector coinciding with a surface sector of the rotating target

The consideration of the problem stated above suggests its cardinal solution. In the range $0 < R < L/2$ where the focal spot is a semicircle, the equality $\eta = 1$ holds. But a semicircle is the particular case of a sector with the angle of 180°.

The scheme of the modified PLD method is shown in Fig. 8, which simply and cardinally solves the problem on the maximal utilisation of the target material. The laser deposition installation is suggested, which differs from ordinary devices by a simple optical system placed outside the deposition chamber. It comprises two lenses and a diaphragm and provides the focal spot in the form of a sector on the target surface. If such a focal spot coincides with a sector-shape area on the target surface and the density of energy is uniform over the focal spot, then the surface of the uniformly rotating target will be uniformly irradiated, which will provide a uniform material ablation from the surface.

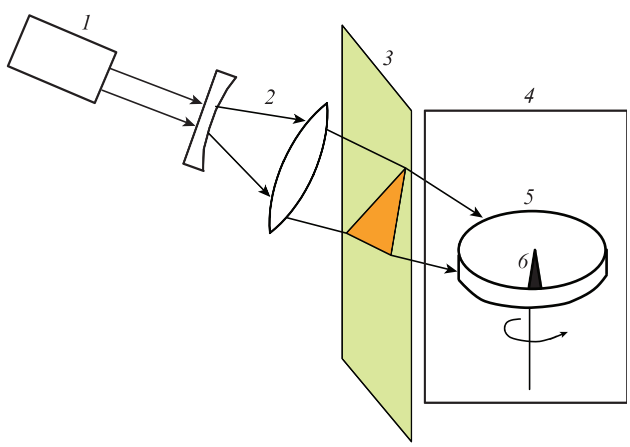

Figure 8. Geometry of the PLD method with the focal spot in the form of a sector:
(*1*) laser; (*2*) optical system; (*3*) diaphragm with a hole; (*4*) deposition chamber; (*5*) target; (*6*) laser focal spot in the form of the sector coinciding with the sector of the rotating target surface.

The device suggested was employed for depositing CuO and $YBa_2Cu_3O_{7-\delta}$ films from the targets 10-mm in diameter by the pulses of second harmonic radiation of the Nd^{3+} : YAG laser with the repetition rate of 20 Hz. The speed of the substrate rotation was 2 rounds min^{-1} and target rotation speed was 37 rounds min^{-1}. The optical system provided the focal spot of laser radiation on the target in the form of the sector with an angle of 60° and the energy density of 4 J cm^{-2}. Variations in the target thickness were within $\pm 2\%$ both before deposition and after 5 deposition cycles lasting for 45 min. One may assert that the device suggested enhances the target material utilisation efficiency up to $\eta = 1$.

5. Conclusions

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 utilisation efficiency. The device comprises two lenses and the diaphragm with a hole and produces a sector-like focal spot on the surface of the target, which provides a uniform rate of material ablation from the entire surface of the rotating target. The device is placed outside the deposition chamber, requires no additional mechanical movements complicating a conventional PLD installation and allows one to increase the target utilisation efficiency actually to unity. It is particularly important in depositing the films from expensive materials (rare and noble metals and the corresponding alloys, pure oxides and multicomponent compounds on their basis, isotopes and so on). The device suggested is interesting by its novelty, simplicity and reliability. In view of the fact that existing solutions either noticeably complicate the deposition installation or do not solve the problem of enhancing the target utilisation efficiency completely, the suggested solution combined with the improvements in the PLD method [5] for obtaining thin films on theoretically infinite substrates may be widely employed in research laboratories or high-technological commercial production.

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