Detailed studies of yttrium thin films deposited by laser radiation of different pulse durations

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Abstract

Parametric studies have been performed on yttrium films deposited by laser ablation deposition at three different laser pulse durations. Morphological investigations of the film surface have confirmed the strong influence of pulse durations on the density, size and shape of the deposited droplets. The average ablation rate and deposition rate in ns regime resulted to be one order of magnitude higher than those obtained in ps and sub-ps regimes. Present interest in the deposition of Y thin films by laser ablation technique starts from the well known photoemission properties of this metal.

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1. INTRODUCTION

Pulsed laser ablation deposition (PLAD) technique is a very successful and versatile technique that is ever more used for growing extremely pure thin films and complex structure films as multilayers [1]. However, the presence of particulates on the deposited surface and the inhomogeneity of the film thickness still represent the major limits of this deposition technique. In spite of that, recent examples show also successful PLAD implementation in manufacturing [2]. Droplets on the deposited films at the present cannot be completely eliminated. Strong efforts have been done to decrease their density by optimizing the deposition conditions, such as the use of high-density targets [3], special deposition geometry [4], various laser target scanning systems [5], dual laser beam method (6) and velocity filters [7]. However, the use of the above techniques results in a drastic decrease of the deposition rate. It is

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also well known that key laser parameters determining the quality of the deposited films, in terms of size and density of droplets, are the laser fluence and the pulse duration. In this context, we studied the surface morphology of Y films deposited by different laser pulse durations after preliminary experiments performed to find out the optimal laser fluence per all pulse durations.

2. EXPERIMENTAL PROCEDURE

Laser ablation and deposition of Y was performed in high vacuum steel chamber evacuated by a turbomolecular pump (figure 1). All experiments were carried out at room temperature and at a base pressure of $10^{-5}$ Pa. The radiation source consisted of a KrF* excimer laser operating at 248 nm with pulse durations of 0.5 ps, 5 ps and 25 ns. The laser radiation entered through a quartz window and impinged on the target surface with an angle of incidence of 45°. The Si substrate was placed in the plume cone at 4 cm distance from the target. Series of 20,000 laser pulses were directed on the yttrium target which was rotated with frequency of 3 Hz to avoid drilling and to obtain irradiation conditions as uniform as possible.

Preliminary and parametric experiments were carried out for three different laser pulse durations. Careful morphological studies of both the films surface and ablated areas were carried out by scanning electron microscope (SEM, mod. JEOL-JSM-6480LV).

In order to measure the thickness value of the deposited films and hence the deposition rate, silver paste droplets were placed on the surface of each Si substrate. After the deposition, the silver paste was removed by ultrasonic bath in acetone and a profilometer (Tencor Alphastep) was used to measure the thickness of films scanning through these areas. The ablation rate was deduced by weighing the target before and after the laser ablation process.

![Figure 1. PLAD experimental set up. MS: mass spectrometer; L: lens; T: target; S: substrate; W: laser window; M: mirror.](image)

3. RESULTS

Several films were deposited during the preliminary studies. Different laser fluences were applied on the target surface positioned at different distance from the substrate surface. After these preliminary investigations, a target-substrate distance of 4 cm was chosen. It was a good compromise between a reasonable deposition rate and acceptable droplets density. Therefore, only one specimen was sampled out for each of three pulse durations used for the present investigations. In table 1, the
laser parameters, the optimal deposition conditions, the ablation rate and deposition rate of the three films chosen for this study are reported.

Table 1. Laser parameters, deposition conditions, ablation and deposition rates

<table>
<thead>
<tr>
<th>Sample</th>
<th>Laser pulse duration</th>
<th>Laser fluence (J/cm²)</th>
<th>Laser density (GW/cm²)</th>
<th>Total number of pulses</th>
<th>Target-substrate distance (cm)</th>
<th>Ablation rate (µg/pulse)</th>
<th>Thickness of film (nm)</th>
<th>Deposition rate (Å/pulse)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>25 ns</td>
<td>5.0</td>
<td>0.2</td>
<td>20,000</td>
<td>4</td>
<td>1.50</td>
<td>577</td>
<td>0.29</td>
</tr>
<tr>
<td>B</td>
<td>5 ps</td>
<td>4.0</td>
<td>800</td>
<td>20,000</td>
<td>4</td>
<td>0.76</td>
<td>78</td>
<td>0.04</td>
</tr>
<tr>
<td>C</td>
<td>0.5 ps</td>
<td>1.3</td>
<td>2600</td>
<td>20,000</td>
<td>4</td>
<td>0.013</td>
<td>23</td>
<td>0.01</td>
</tr>
</tbody>
</table>

Figure 2 shows the surface morphology of the samples (A, B and C) at two different magnitudes (100 and 500). The droplets shown in fig. 2 have a round micrometric shape indicating that they were, at least partially, molten during the impact with the substrate surface. The number density of the droplets varies strongly with the laser pulse duration.

Figure 2. SEM micrographs at low magnification of the film surfaces obtained with laser pulses of a) 25 ns, b) 5 ps and c) 0.5 ps. The same surfaces at higher magnification are given in d) 25 ns, e) 5 ps and f) 0.5 ps.

It is believed that the explosive nature of the laser-solid interactions may be responsible of the expulsion of the droplets from the target surface and deposition of these droplets on the film. According to Singh et al. [8], the subsurface superheating is the main mechanism from the ejection of droplets from the target. It is evident that the internal superheating responsible for the expulsion of droplets is higher during laser irradiation in ps and sub-ps regimes. The lower degree of subsurface superheating during ns laser ablation will result into much less expulsion of droplets as shown in fig. 2. As the majority of droplets seem to arise from explosive nature of the laser-target interactions, the morphology of the surface irradiated by laser with different pulse durations was carefully studied. Figure 3 shows the SEM images of ablated
areas produced by laser irradiation at two different magnifications. The targets irradiated by ps and sub-ps laser pulses exhibit a typical cone-like structure, while the target irradiated in ns regime shows a ripple-like structure. The latter exhibits a much smoother morphology: more resistant to the ejection of droplets.

![Figure 3. SEM micrographs at low magnification of the ablated areas obtained with laser pulses of a) 25 ns, b) 5 ps and c) 0.5 ps. The same surfaces at higher magnification are given in d) 25 ns, e) 5 ps and f) 0.5 ps.](image)

The surface coverage of the films by the droplets, the range of droplets sizes, the weighted average size (deduced as \( d = \sum d_i N_i / \sum N_i \)), and the average density of the droplets per unit film thickness, for all the laser pulse durations used in these experiments, are reported table 2.

**Table 2. Data on the deposited droplets deduced by analyses of the SEM micrographs**

<table>
<thead>
<tr>
<th>Sample</th>
<th>Number density (cm(^{-2}))</th>
<th>Surface coverage (%)</th>
<th>Range ((\mu m))</th>
<th>Weighted average size ((\mu m))</th>
<th>Average density (cm(^{-2}) Å(^{-1}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>2.8x10(^5)</td>
<td>2.6</td>
<td>(0.3-16)</td>
<td>2.4</td>
<td>48.5</td>
</tr>
<tr>
<td>B</td>
<td>4.3x10(^7)</td>
<td>13</td>
<td>(0.2-11)</td>
<td>0.5</td>
<td>5.5x10(^4)</td>
</tr>
<tr>
<td>C</td>
<td>4.5x10(^7)</td>
<td>6</td>
<td>(0.2- 5)</td>
<td>0.4</td>
<td>3.6x10(^5)</td>
</tr>
</tbody>
</table>

From the data reported in Table 2, it is clear that the ablation and deposition processes with ns laser pulse duration are more effective than those obtained by ultra short pulses. Moreover, the surface of the deposited films is less covered of droplets.

**4. CONCLUSIONS**
These studies confirm the strong influence of laser pulse duration on the density, size and shape of the deposited droplets. The subsurface superheating seems to be the main mechanism responsible of the ejection of the droplets from the target surface. The lower is the laser pulse duration, the higher is the density of droplets deposited on the film. It was found out, in the light of these results, that the laser ablation deposition of yttrium in the ns regime should be by far more appropriate than ps and sub-ps regimes. We assume that the optimal experimental conditions were found out to deposit a Y thin film on a copper substrate to be used as photocathode in a RF photoinjector.

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