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The flip-over effect in pulsed laser deposition: Is it relevant at high background gas pressures?

Alejandro Ojeda-G-P [a], Christof W. Schneider [a,*], Max Döbeli [b], Thomas Lippert [a], Alexander Wokaun [a]

[a] Paul Scherrer Institut, General Energy Research Department, 5232 Villigen-PSI, Switzerland
[b] Ion Beam Physics, ETH Zurich, CH-8093 Zurich, Switzerland

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ABSTRACT

In pulsed laser deposition the use of a rectangular or elliptical beam spot with a non 1:1 aspect ratio leads to the so called flip-over effect. Here, the longest dimension of the laser spot results in the shortest direction of plasma plume expansion. This effect has been mainly reported for vacuum depositions of single element targets and is particularly noticeable when the aspect ratio of the beam spot is large.

We investigate the flip-over effect in vacuum and at three relevant background-gas pressures for pulsed laser deposition using a $La_{0.4}Ca_{0.6}MnO_3$ target by measuring the thickness dependence of the deposited material as a function of angle. The film thicknesses and compositions are determined by Rutherford backscattering and argon is used to reduce the influence of additional chemical reactions in the plasma. The results show the prevalence of the flip-over effect for all pressures except for the highest, i.e. 1×10^{-1} mbar, where the film thickness is constant for all angles. The composition profiles show noticeable compositional variations of up to 30% with respect to the target material depending on the background gas pressure, the angular location, and the laser spot dimensions.

1. Introduction

In pulsed laser deposition (PLD) the choice of the laser spot shape and size is not trivial and has a strong influence on the angular distribution of film thickness and composition [1–9]. One of the most fascinating effects of spot choice is the so-called flip-over effect. It is a specific laser ablation phenomenon, which occurs when using a non-circular laser spot. The longest direction of the laser spot results in the shortest direction of the deposition pattern and vice versa. For example, for an elliptical laser spot the plasma plume expansion "appears" to be turned by 90° with respect to the laser spot orientation since the plasma plume appears broader in the shorter dimension of the laser spot and shorter in the longer direction. Thus the name, flip-over effect. Nevertheless, the flip-over effect is a purely gas-dynamical phenomenon and the apparent turn is not necessarily restricted to 90° [1]. It originates from the fact that the narrowing of the deposition profile increases with the number of intra-plume collisions, which increase roughly as the ratio of the initial plume dimensions (given by the spot size) to the atoms' Knudsen layer mean free path (MFP) [2].

The flip-over effect has been reported frequently when investigating the angular distribution of species ablated in vacuum conditions. Very often the analysis has been done for elliptical laser spots and by looking at the thickness of the deposited pattern [3,9–11]. In other cases, the temporal evolution of the expanding plasma has been studied. An example is shown in reference [4] where a rectangular laser spot of 1 mm × 2.4 mm and time-resolved imaging was used to probe the plasma plume in 3 dimensions to provide a visualization of the effect. Mostly, the flip-over effect is studied using single element targets whereas the resulting film composition when using multi-element targets is little explored [5]. Additionally, modelling the plasma expansion by adiabatic expansion or isothermal expansion equations have all shown the prediction of the flip-over effect [1,3,6,7,9].

Although not strictly related to the plasma expansion analysis, Kelly and Miotello [1] explored the deposition of debris around the craters of ablated polyimide targets for different spot shapes in air (Fig. 1). These patterns show turns of 60°, 45° or even new shapes. Although not explained in their article these results can be understood by referring to the original definition of the flip–over effect: the longest direction of laser spot results in the shortest direction of deposition. Assuming that everything expands from the centre outwards and drawing on the picture an inscribed circle with the shortest dimension (in blue) and a circumscribed circle with the

* Corresponding author at: OFLB/U111, CH-5252 Villigen-PSI, Switzerland.
E-mail address: christof.schneider@psi.ch (C.W. Schneider).

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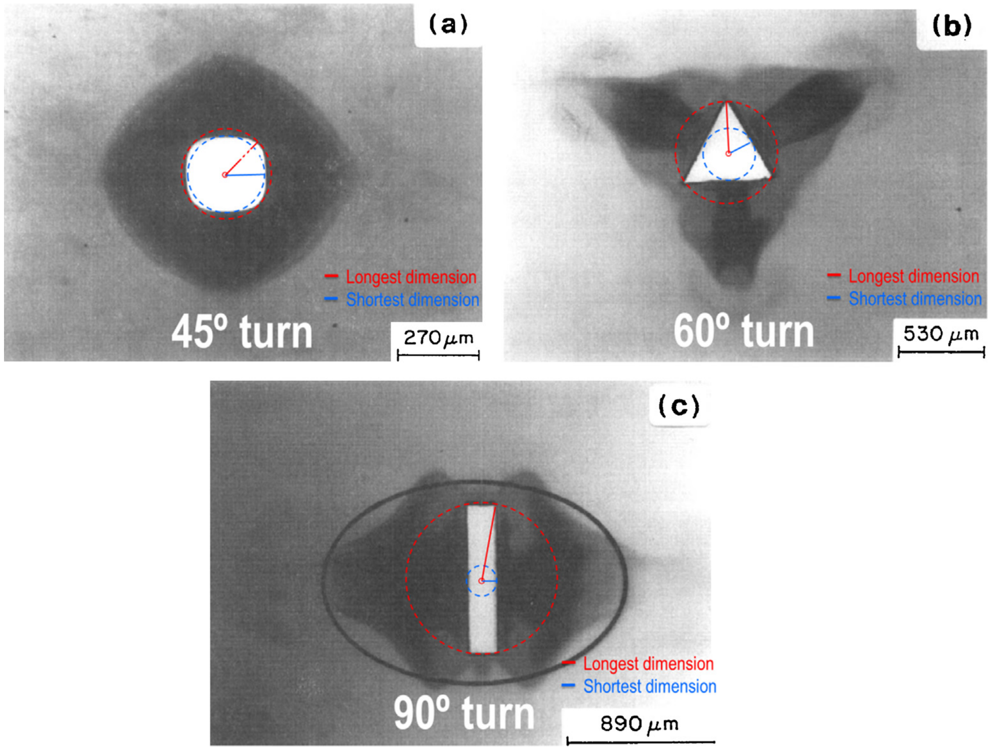

Fig. 1. Modified images from [1] of deposited material on the surface of a polyimide target exposed to 50 laser pulses in air for different laser spot shapes (308 nm, ~20 ns, ~0.25 J/cm^2). An explanation in red and blue colours has been added to the images to visually explain the debris patterns (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article).

longest dimension (in red). One can see that the deposited debris expands more in the shortest dimension and less in the longest. It is even more interesting for Fig. 1c as the transition from the long dimensions to the shorter happens in a non-linear fashion as one moves along the angle. The shortening of the long dimension is initially very fast, explaining the abrupt deposited pattern recorded. Such clear images for different laser spot shapes provide a better visualization of the flip-over effect than the previously mentioned studies with rectangular or elliptical laser spots, as the complexity of the spot shapes reveals the particularities of the plasma plume expansion results.

Despite the interest in the flip-over effect, most studies are limited to vacuum ablation and single elemental targets. The more commonly used depositing pressure range of ~0.1 mbar and ablation from multi-elemental targets are less explored. Nevertheless, in [8] the effect is studied at 5×10^{-2} mbar argon and a broadening of the plasma plume with the higher background gas pressure is reported. Unfortunately, the research was limited to one single pressure and the ablation of Ag. Therefore, a detailed understanding of the flip-over effect at higher background gas pressures and using multi-element targets is very relevant for PLD.

The present work studies the flip-over effect for the ablation of the multi-element oxide $La_{0.4}Ca_{0.6}MnO_3$ and analyses the influence of the flip-over effect with respect to film thickness and composition for four relevant background pressures (vacuum (1×10^{-6} mbar), 1×10^{-3} mbar, 1×10^{-2} mbar and 1×10^{-1} mbar). The corresponding angular distribution of the ablated species and thickness measurements of the deposited films have been determined using Rutherford backscattering spectrometry (RBS). The implications of these experiments are not limited to the flip-over effect solely, but provide guidance to PLD users attempting large area depositions by varying the spot dimensions in combination with laser beam rastering.

2. Materials and methods

To capture the evolution of the flip-over effect with pressure the ablation experiments have been conducted in a UHV chamber (base pressure 1×10^{-9} mbar) using a KrF excimer laser (Lambda Physik LPX 300, 20 ns, λ = 248 nm) with a laser fluence of 2 J/cm^2 and a repetition rate of 3 Hz for 40 min. A mask was used to ensure a flat-top beam profile with a rectangular laser spot of 1 mm × 2.6 mm on the ablated target. The laser pulses arrived at the target with a 45° angle with respect to the target to substrate axis and in the same horizontal plane. A sintered, polycrystalline cylindrical $La_{0.4}Ca_{0.6}MnO_3$ target with a diameter of 12 mm and a length of 30 mm was used for the depositions. In order to avoid cratering during the ablation the target was continuously rotated and displaced along its main axis. Prior to these experiments the target composition was verified using RBS.

The films were deposited onto two aluminium foils with 99.999% purity and 20 μm thickness. These Al-substrates were placed perpendicular to each other in a vertical and horizontal position on a special substrate holder named the "semi-sphere" holder (Fig. 2). The holder keeps a constant target-to-substrate distance of 40 mm due to its intrinsic spherical geometry and has an angular range of ±86°. The choice to use high purity aluminium foils is to avoid substrate channelling effects in RBS and to simplify the measuring process itself due to its continuous and flexible nature [12].

All the depositions were performed at room temperature and at four different background pressures: vacuum (1×10^{-6} mbar), 1×10^{-3} mbar, 1×10^{-2} mbar and 1×10^{-1} mbar with argon as background gas to reduce possible chemical reactions with the background environment in order to simplify the interpretation of the measurements. Except vacuum, the other pressures were chosen to capture the evolution from a free expansion of the plasma plume (<1×10^{-3} mbar) with hardly any interactions with