Applied Surface Science 257 (2011) 2775–2778

Contents lists available at ScienceDirect

Applied Surface Science

journal homepage: www.elsevier.com/locate/apsusc

Effects of phase explosion in pulsed laser deposition of nickel thin film and sub-micron droplets

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ARTICLE INFO

Article history:
Received 16 February 2010
Received in revised form 14 May 2010
Accepted 15 October 2010

Keywords:
Pulsed laser deposition
Phase explosion
Droplets
Melt ejection

ABSTRACT

Nickel (Ni) thin films were deposited on glass substrates in high vacuum and at room temperature with third-harmonic or 355-nm output from a nanosecond Nd:YAG laser. At low laser fluence of 1 J/cm^2, the deposition rate was about 0.0016 nm/shot which increased linearly until 4 J/cm^2. Above 4 J/cm^2, the onset of phase explosion in the ablation abruptly increased the optical emission intensity from laser-produced Ni plume as well as thin-film deposition rate by about 6×. The phase explosion also shifted the size distribution and number density of Ni droplets on its thin-film surface. On the other hand, the surface structures of the ablated Ni targets were compared between the scan-mode and the fixed-mode ablations, which may suggest that droplets observed on the thin-film surface were caused by direct laser-induced splashing of molten Ni rather than vapour-to-cluster condensation during the plume propagation.

1. Introduction

Pulsed laser deposition (PLD) is a common deposition technique for many types of thin films as compared to other conventional deposition methods [1,2]. One of the main advantage lies in laser ablation of almost any material, be it organics or refractory oxides, and transform into a self-propagating plume with sufficient kinetic energy which enables surface coating or thin-film deposition. On the other hand, one of the major disadvantages of PLD is the presence of particulates on the thin film [3–8], which may include droplet formation from laser-induced molten surface on the target, debris expulsion due to laser-induced damage on target, gas-phase clustering of nanoparticles, etc. Although these particulates are usually undesirable for high-quality thin film with low roughness, and some techniques have been implemented to reduce the particulates [1,6,9,10], the generation of nanoparticles via laser ablation has attracted considerable interest in recent years. The advent of femtosecond laser has enhanced such a simple technique for nanoparticle generation [11–15].

For laser ablation with nanosecond Nd:YAG and excimer lasers [16,17], three kinds of thermal processes during the laser-matter interactions may lead to material removal from the target: (1) vapourization, (2) normal boiling, and (3) explosive boiling. The third process is also referred to as the phase explosion above a certain threshold of laser fluence (J/cm^2), which has been explained by superheating in the sub-surface region when the temperature breached a thermodynamic critical point [18,19]. The term "explosive boiling" in silicon ablation [20] has been coiled to describe the effect of phase explosion in sub-surface. The sub-surface boiling would convert part of the laser-induced melt into vapour due to superheating effect, hence increasing the plume density [21]. In addition, a delayed ejection of particles from the ablation target was reported [21–23] due to laser-induced splashing of molten layer.

The onset of phase explosion in nickel (Ni) ablation by an excimer laser [24] was investigated by a pressure sensor at the backside of Ni target, which was correlated to the etch rate or depth. In contrast, a simpler technique is used to detect the onset of phase explosion in this work. An optical multichannel analyzer (OMA) is employed for measuring the optical emission from Ni plasma by pulsed Nd:YAG laser ablation. The effects of phase explosion in Ni ablation, as detected by the OMA, are correlated to the deposition rate of Ni thin film as well as the number density and size distribution of sub-micron droplets. These correlations between the onset of phase explosion and thin-film deposition, to the best of our literature, have hardly been reported.

2. Experimental setup for thin film deposition

A pulsed Nd:YAG laser (EKSPLA, NL301) with the third-harmonic output at 355 nm, 10 Hz and 4 ns of pulse duration was used

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doi:10.1016/j.apsusc.2010.10.061

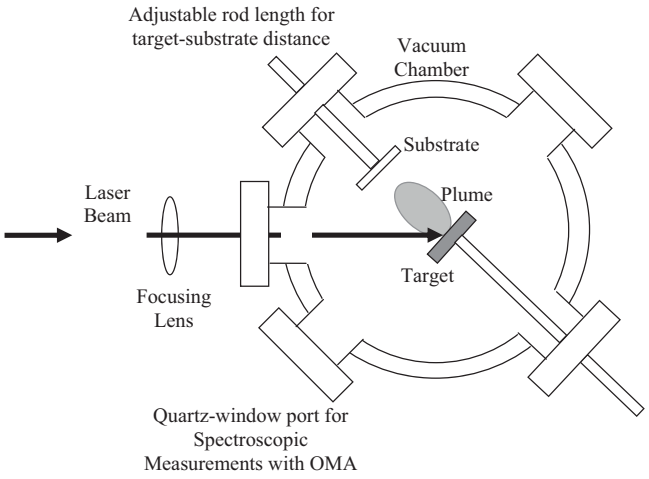

Fig. 1. Schematic of experimental setup for Ni ablation and thin film deposition by pulsed Nd:YAG laser with output at 355 nm and pulse duration of 4 ns.

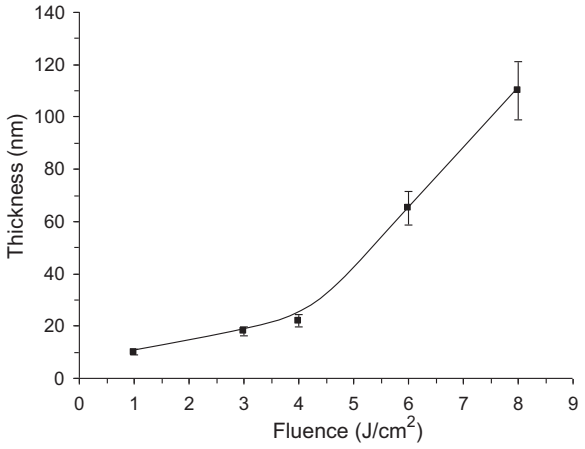

Fig. 3. Thickness of Ni thin-film as a function of the laser fluence: a sudden jump to different deposition rate, increased by about 6×, is observed after 4 J/cm^2.

for the Ni ablation and thin-film deposition in high vacuum at room temperature. Fig. 1 shows the schematic of the experimental setup, which was reported previously for depositions of diamond-like carbon and indium tin oxide [25,26]. The deposition chamber was evacuated to a base pressure of 5×10^{-6} mbar. The laser beam was controlled by a $X-Y$ scanning mirror and was focused by a quartz lens with $f = 50$ cm on the Ni target with spot size of about 0.5 mm in diameter. The laser fluence was varied between (1–10) J/cm^2 by changing the flashlamp voltage while each ablated spot on Ni target would overlap with adjacent spots by about 30%.

For normal PLD, glass substrates are usually placed at 10 cm from the target. In contrast, our glass substrates were placed at 5 cm in order to collect sufficient Ni droplets for the analysis of size distribution and number density as a function of laser fluence. The duration for PLD of Ni thin film was fixed at 15 min, equivalent to 9000 laser shots. Each thin-film sample was deposited from a new area on the Ni target as the ablated surface showed periodic structures consisting of ripples, as shown in Fig. 2(a). These ripples may represent the secondary, laser-induced surface roughness which

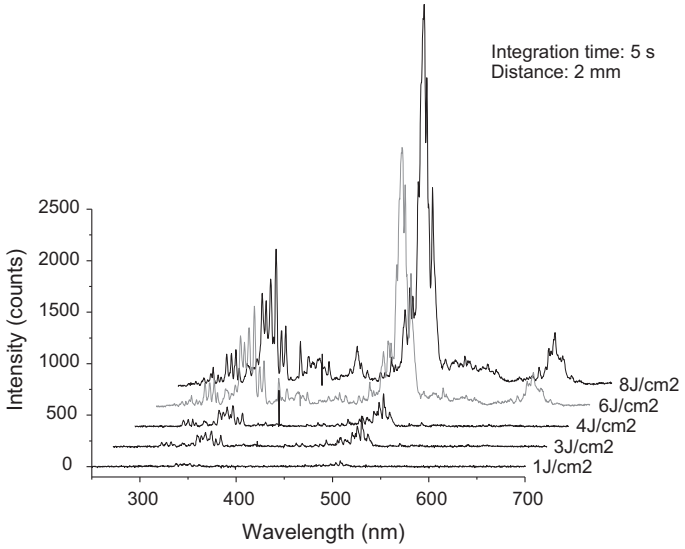

Fig. 4. Intensity change in the optical emission spectra (OES) from the Ni plasma as a function of laser fluence.

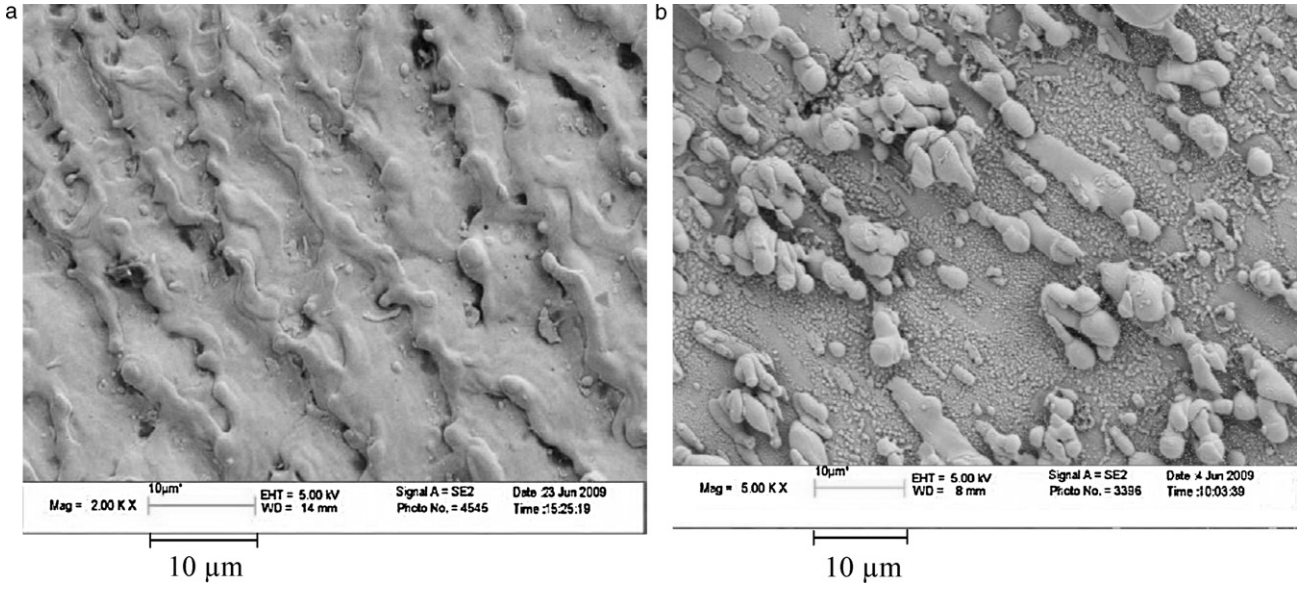

Fig. 2. (a) Periodic structures (ripples) on ablated Ni target surface formed after multiple passes by scan-mode laser ablation. (b) Fixed-point laser ablation which shows radial splashing of molten Ni droplets of either spherical or elongated shapes, as well as agglomerated.