Applied Surface Science 302 (2014) 109–113

Contents lists available at ScienceDirect

Applied Surface Science

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

Picosecond laser induced periodic surface structure on copper thin films

Thi Trang Dai Huynh, Agnès Petit, Nadjib Semmar*

GREMI-UMR 6606, CNRS/Université d'Orléans, 14 rue d'Issoudun, BP 6744, 45067 Orléans cedex2, France

ARTICLE INFO

Article history:
Received 19 June 2013
Received in revised form 28 October 2013
Accepted 29 October 2013
Available online 18 November 2013

Keywords:
LIPSS
Spikes
Metallic thin films
Picosecond laser beam

ABSTRACT

LIPSS (Laser Induced Periodic Surface Structure) formation on copper thin films induced by a picosecond laser beam (Nd:YAG laser at 266 nm, 42 ps and 10 Hz) was studied experimentally. Copper thin films were deposited on glass and silicon substrates by magnetron sputtering. The surface modifications of irradiated zones were analyzed by scanning electron microscopy. Two distinct types of LIPSS were identified with respect to the laser fluence (F), number of laser shots (N) and substrate material. Namely, with a number of laser shots ($1000 < N < 10{,}000$) and a fluence of ($200\ \mathrm{mJ/cm^2} < F < 500\ \mathrm{mJ/cm^2}$), Low Spatial Frequency LIPSS (LSFL with a spatial period of $\Lambda \sim 260$ nm and an orientation perpendicular to polarization) and High Spatial Frequency LIPSS (HSFL with a spatial period of $\Lambda \sim 130$ nm and an orientation parallel to the polarization) were observed. The regime of regular spikes formation was determined for $N \geq 1000$. Moreover, the 2D-map of the relationship among LIPSS formation, laser fluence and number of laser shots on copper thin film with two different substrates was established. A physics interpretation of regular spikes and LIPSS formation on copper thin film induced by ps laser with overlapping multi-shots is proposed based on experimental data and the theory of Plateau-Rayleigh instability.

1. Introduction

In recent years, Laser Induced Periodic Surface Structures (LIPSS) have attracted widespread research attention because of their applications in micro and nanoscale technology fields such as lithography, high density data storage, micromachining for microelectronics and micro-electromechanical systems (MEMS) [3–6]. LIPSS have been induced on different materials by ultrashort lasers in the nanosecond (ns) to femtosecond (fs) regimes [1–12]. Two distinct types of LIPSS have been identified: Low Spatial Frequency LIPSS (LSFL) and High Spatial Frequency LIPSS (HSFL). Their orientations can be perpendicular or parallel to the polarization of the incident laser radiation, depending on laser parameters and material properties. LSFL have a spatial period close to the irradiation laser wavelength. Their formation is attributed to the interference between the incident and scattered waves [14] or Surface Plasmon Polaritons (SPP) [15]. HSFL have a spatial period that is much smaller than the laser wavelength. Their formation is still debated and several theories have been proposed: second harmonic generation (SHG) [11–13], self-organization [16], or interference with modification of the optical properties during laser processing [17]. These types of LIPSS have generally been identified in the femtosecond laser regime on semiconductors [8,9], bulk metals [10,18] and dielectrics [7,18]. However, they have seldom been investigated in the picosecond regime on such materials.

Among the metallic films used in LIPSS formation, copper is particularly interesting due to its notable characteristics such as higher electrical and thermal conductivity, a higher melting temperature and correspondingly lower rates of diffusivity, and high strengths [19]. Moreover, the formation of LIPSS by pulsed laser achieves highly controlled nanostructures, allowing an easier tunability of the conductivity. LIPSS formation on copper thin film has thus attracted the interest of many investigators [20,21]. To date, however, this process has mainly been studied in the infrared wavelength with femtosecond laser beams.

In order to understand the mechanism of LIPSS formation in the case of copper thin films, we report here new results in the picosecond regime (intermediate between the nano- and femtosecond) using the fourth harmonic of an Nd:YAG Gaussian-like laser beam (~266 nm). The laser beam parameters varied from 20 to 500 mJ/cm^2 for the fluence employed, and from 10 to 10.000 shots.

2. Experimental details

Copper thin films with a thickness of 500 nm were deposited on silicon and glass substrates by magnetron sputtering. The

* Corresponding author. Tel.: +33 238 494 885; fax: +33 238 417 154.
E-mail addresses: nadjib.semmar@univ-orleans.fr, nadjib.semmar@orange.fr (N. Semmar).

0169-4332/$ – see front matter © 2013 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.apsusc.2013.10.172

typical coating parameters were background pressure of 0.015 mTorr, working pressure of 3 mTorr, and deposition duration of 20 min. Before deposition, the silicon and glass substrates were ultrasonically cleaned in acetone and rinsed in ethanol for 10 min. No intermediate layers were used to increase the adhesion of copper to the substrate.

A picosecond Nd: YAG laser was used to generate linearly polarized laser pulses at a wavelength of 266 nm with a pulse duration of 42 ps and a repetition rate of 10 Hz. The laser beam was perpendicular to the sample surface. It was focused into an almost circular spot of 400 μm diameter by a Plano-convex lens (75 mm focal length). The pulse energy used in this experiment was set below and over the ablation threshold value (from 30 μJ to 550 μJ), corresponding to the laser fluence of 20–500 mJ/cm^2. It was measured by a powermeter placed just before the sample surface. The number of laser shots used was N = 10–10,000. All experiments were carried out in ambient air. The surface morphologies of the irradiated regions were examined by scanning electron microscope (SEM: Zeiss Supra 40).

3. Results

Due to the influence of the Gaussian-like shape of the laser beam profile, the surface morphologies of the copper thin film after irradiation by the ps laser with an increasing fluence and N = 10–10,000 shots can be located at: (i) The center of the irradiated zone, which corresponds to the highest local fluence, where easy removal of the surface layer from the substrate is achieved; (ii) The rim of the irradiated zone, which corresponds to a lower local fluence, leading to easy identification of LIPSS and the formation of regular spikes at N = 1000–10,000 shots.

Fig. 1 shows surface morphologies of Cu (500 nm)/glass at the center of the irradiated zone after 1000–5000 shots for the lowest fluence of 24 mJ/cm^2. This fluence was not sufficient to produce any visible surface change with N < 1000. On increasing the number of laser shots to 1000, long spikes were formed as shown in Fig. 1(a). Their spatial period is close to 100 nm. For N = 2000 shots, these long spikes tend to shrink to circular spikes with a diameter around 200 nm on the grooves [Fig. 1(b)]. The distance between

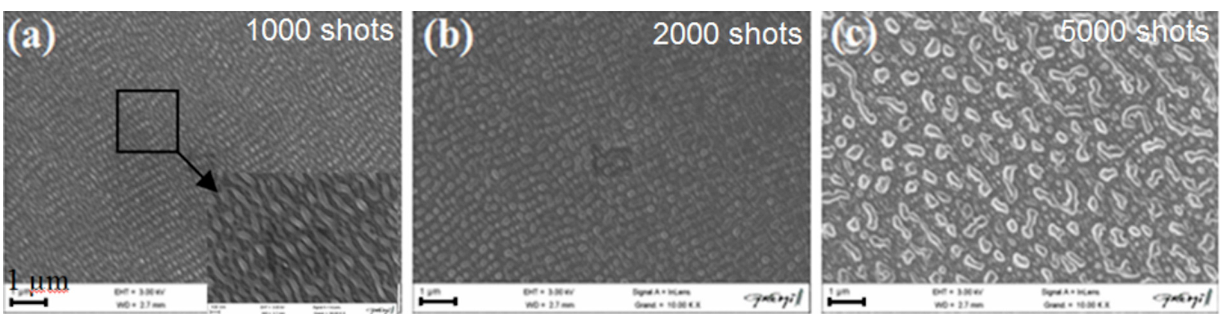

Fig. 1. Formation of long and circular spikes of Cu/glass surface at the center of the irradiated zone induced by the ps laser at the lowest fluence F = 24 mJ/cm^2 with (a) 1000, (b) 2000 and (c) 5000 shots.

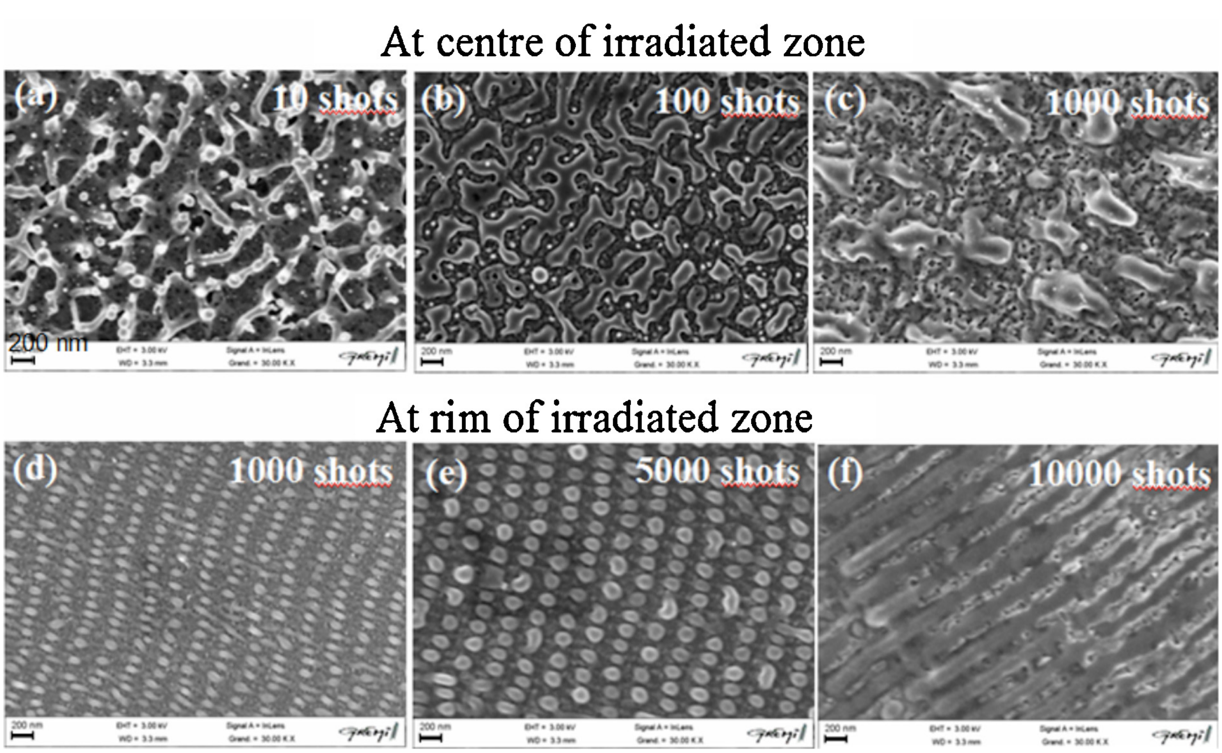

Fig. 2. Regular spikes of Cu/glass surface induced by the ps laser at the laser fluence F = 199 mJ/cm^2 with N = 10–10,000 shots.