Applied Surface Science xxx (2017) xxx–xxx

Contents lists available at ScienceDirect

Applied Surface Science

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

Full Length Article

Inducing subwavelength periodic nanostructures on multilayer NiPd thin film by low-fluence femtosecond laser beam

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ARTICLE INFO

Article history:
Received 28 October 2016
Received in revised form 10 March 2017
Accepted 15 March 2017
Available online xxx

Keywords:
Periodic surface structures
Femtosecond beam
Multilayered thin film
Surface modifications

ABSTRACT

During femtosecond interaction with surfaces, the processes of liquid and solid-state dewetting could be responsible for the generation and regrouping of nanoparticles and nanoparticle clusters. The occurrence of surface plasmon polariton most probably induces the LIPSS arrangement. We have used low-fluence scanning femtosecond beam to generate sub-wavelength periodic structures on multilayer Ni/Pd thin films on Si. The spatial period of LIPSS increases with the change of scanning directions in respect to the polarization direction due to the phase difference increase between the incoming and induced oscillations.

1. Introduction

One of the inherent phenomena related to the irradiation of solid surfaces by ultrashort-pulse laser is the emergence of periodic grating structures on the surface (laser induced periodic surface structures, LIPSS). Being the topic of research for a long time, the phenomenon has been approached from the points of view of material types, modes of operations, beam parameters, and possible applications [1–12]. LIPSS formation has interest both form the fundamental point on view and also considering the change in properties of the nanostructured surfaces.

Two types of LIPSS are reported, low spatial frequency LIPSS (LSFL) and high spatial frequency LIPSS (HSFL) [13]. Named after their size (magnitude of spatial frequency), their orientation in respect to the polarization direction is not yet fully understood. While for LSFL it is dependent on the dielectric permittivity – it seems that LSFL orientation is perpendicular to polarization for metals and semiconductors and parallel for dielectrics ($|\varepsilon'| < 1$) and sometimes perpendicular – for HSFL is not well explained [2,14–16].

The most probable causes of the LIPSS emergence are the surface plasmon polaritons (SPP) generated on the material surface in the irradiation area (leading to spatial periodic distribution of energy over the surface) or self-organization of the material upon the pulse impact [17,18].

The applications of thin films play an important role in many fields, like semiconductor technology, optics, chemistry, mechanics, magnetics, electricity. Various types of coatings for protection, diffusion barriers, filtering, reflection/antireflection, sensing, waveguiding, decorative and other purposes are just some to mention. Structuring of thin films can enhance their characteristics. The interaction of femtosecond laser beam with thin films can generate LIPSS. Thin films and alloys based on Ni and Pd have specific physico-chemical as well as mechanical characteristics, like high corrosion resistance, durability and high tensile strength and due to its mechanical characteristics and the catalytic activity, the applications range from catalyst and hydrogen storage material to holography [19]. In this work, we have demonstrated the generation of sub-wavelength periodic structures on the multilayer Ni/Pd thin films by the scanning low-fluence femtosecond laser beam. The structures have been identified as HSFL probably caused by the SPP. The influence of the scanning direction to the spatial period of HSFL is seen in the phase difference increase between the incoming and induced oscillations for scanning direction approaching the perpendicular to the polarization direction.

2. Experimental setup

The experimental setup is based on a femtosecond laser (Coherent Mira 900). The laser beam was focused by a GF Panachromium

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http://dx.doi.org/10.1016/j.apsusc.2017.03.141
0169-4332/

objective (40x/0.65) on the specimen. The wavelength of the beam was monitored with the fiber coupled spectrometer (Ocean Optics HR2000CG-UV-NIR). Irradiated samples were Ni/Pd bilayers, with five layers of each metal, deposited on the silicon (100) wafer. The depositions were performed by the Balzers Sputron II apparatus using 1.3 keV argon ions and with 99.9% of Ni and Pd targets purity. Each layer (Ni or Pd) was of ~13 nm thickness reaching the total thickness of bilayer group of ~130 nm. The irradiations of the top (Ni) layer of the Ni/Pd bilayer system were performed in air with focused femtosecond laser beam under normal incidence. The laser beam properties were: wavelength 760–880 nm, pulse duration ~100 fs, repetition rate 76 MHz, power of 175–195 mW, linear polarization in the horizontal plane, Gaussian-like profile, spot diameter ~200–1000 nm. Femtosecond laser interactions have been performed in two modes, with laser beam being static or scanning. The results of the interactions have been analyzed by scanning electron microscopy (SEM) – the TESCAN MIRA3 system.

3. Results and discussion

In order to examine the response of the material to irradiations in both static and dynamic modes, the beam was moved from point to point over the surface of the sample by computer-controlled pair of galvo-scanning mirrors. When irradiating the samples, two time intervals have been used as experimental parameters: the "dwell time", or the time the beam is at the same point, and the "flight time" or the time for travelling between two points. The pattern drawn by the scan of the beam is a matrix, consisted of 80 points in the 8 by 10 scheme (Fig. 1).

The beam was positioned in the center of the area – central hole in Fig. 1, where it dwelled for predefined dwell time. It then scanned during flight time to the top left point (top left hole) and waited for dwell time. Point by point, the beam scanned and dwelled, thus engraving the pattern in the predefined area. The distance between two points was 7.5 μm in the direction parallel to the polarization direction and 8.5 μm in the direction perpendicular to the polarization direction.

The sample has been irradiated with femtosecond beams of single-pulse fluences below 146 mJ/cm^2 which is single-pulse ablation threshold for 5x(Ni/Pd)/Si multilayer system [20]. For single-pulse fluences below the ablation threshold and shorter expositions, the LIPSS are typically formed. For longer expositions, the accumulation of pulses would lead to melting in some materials, but in some materials the LIPSS would remain stable.

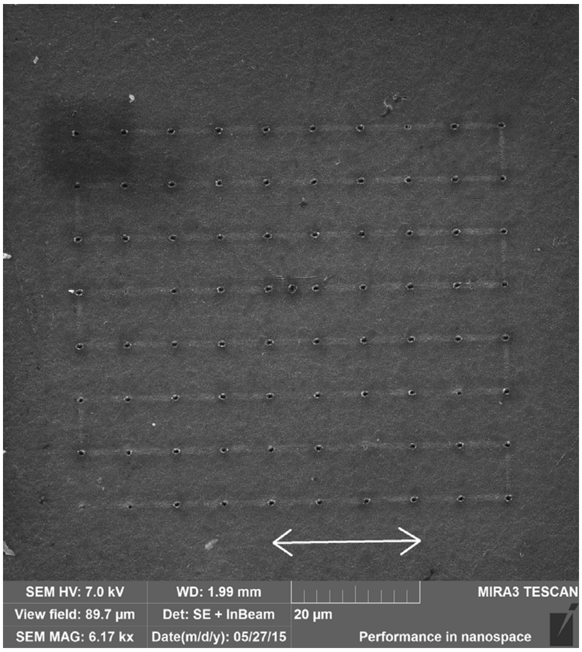
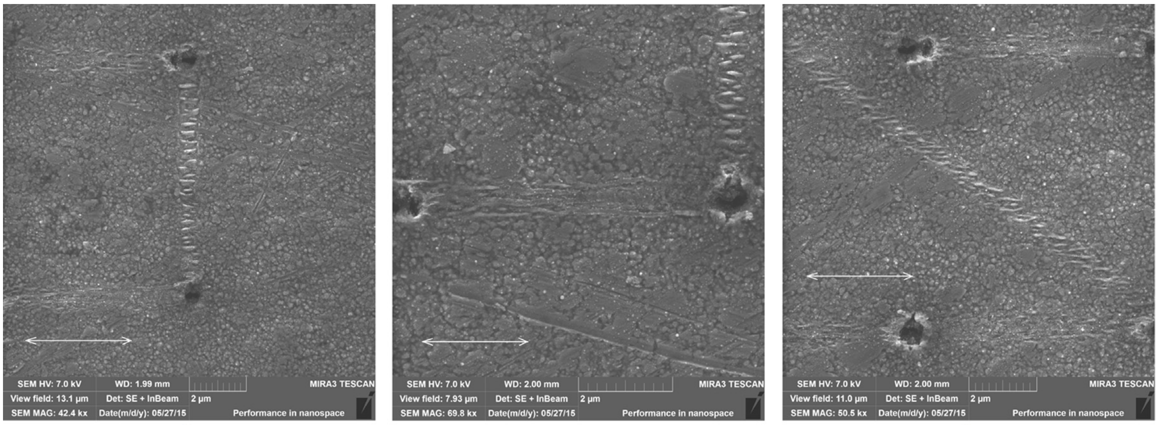


Fig. 1. Upper left part of the pattern (double arrow indicates polarization direction); wavelength 880 nm, power 175 mW, fluence 135 mJ/cm^2, dwell time 20 ms, flight time 20 ms.

The main objective was to investigate the appearance of LIPSS (HSFL) during scanning. During static irradiation, the number of pulses delivered to the irradiated area (the spot) is dependent on the exposition time. During scanning, the number of pulses that irradiate the area of the same size as in static irradiation (the spot) depends on the scanning rate.

For examining the influence of the scanning direction to the spatial frequency of the induced HSFL, the sample was irradiated in similar pattern, with inter-point distances of 7 μm (direction parallel to polarization) and of 8.3 μm (direction perpendicular to polarization). The laser beam parameters were: wavelength 880 nm, power 175 mW, single pulse fluence ~142 mJ/cm^2, dwell time 10 ms, flight time 10 ms. Scanning rate was 830 μm/s (direction parallel to polarization), 700 μm/s (direction perpendicular to polarization) and 4015 μm/s (direction oblique to polarization, ~38°). Static irradiation led to hole drilling, while scanning modified the material surface.



a) b) c)

Fig. 2. SEM micrographs of irradiated areas (wavelength 880 nm, power 175 mW, dwell time 10 ms, flight time 10 ms, double arrow shows polarization direction): a) scanning perpendicular to polarization direction; b) scanning parallel to polarization direction; c) scanning oblique to polarization direction.

Please cite this article in press as: A.G. Kovačević, et al., Inducing subwavelength periodic nanostructures on multilayer NiPd thin film by low-fluence femtosecond laser beam, Appl. Surf. Sci. (2017), http://dx.doi.org/10.1016/j.apsusc.2017.03.141