G Model
APSUSC-31324; No. of Pages 4
ARTICLE IN PRESS
Applied Surface Science xxx (2015) xxx–xxx
Contents lists available at ScienceDirect
Applied Surface Science
journal homepage: www.elsevier.com/locate/apsusc

Importance of layer thermal conductivity on the sharpness of patterns produced by laser interference

R.J. Peláez[a,*], C.N. Afonso[a], M. Škereň[b], J. Bulíř[c]

[a] Laser Processing Group, Instituto de Optica, CSIC, Serrano 121, 28006 Madrid, Spain
[b] Faculty of Nuclear Sciences and Physical Engineering, Czech Technical University in Prague, Brehova 7, 115 19 Prague 1, Czech Republic
[c] Institute of Physics, ASCR, v.v.i., Na Slovance 2, Prague, Czech Republic

ARTICLE INFO

Article history:
Received 1 June 2015
Received in revised form
10 September 2015
Accepted 13 September 2015
Available online xxx

Keywords:
Laser interference
Metal layers
Thermal conductivity
Nanoparticles
Dewetting

ABSTRACT

In this work, we compare patterns produced in Ag layers having similar thickness in the range 8.3–10.8 nm but having different initial nanostructure, i.e. behaving either as discontinuous or continuous layers and thus having very different thermal conductivities. The patterns are produced by exposing a phase mask to an excimer laser operating at 193 nm and using a projection optics that leads to similar fringed patterns with periods in the range 6.3–6.7 μm. The layer breaks up into isolated NPs due to laser induced melting at the regions around the intensity maxima sites. The resulting fringes have sharp interfaces in the case of discontinuous layers while a variety of regions across the pattern with no sharp interfaces are produced in the case of continuous layers. The results show that while the temperature distribution across the pattern matches almost perfectly the laser beam intensity profile for the former case, it becomes smeared due to lateral heat flow for the latter case. These results provide evidences for significant heating at the intensity minima sites that lead to solid-state dewetting and will eventually limit the minimum period achievable in the case of continuous metal layers or thermally conducting layers.

1. Introduction

Laser interference is a versatile technique for producing periodically patterned surfaces that are being very attractive due to their unique optical, electrical and chemical properties. Interesting fields of applications include photonic or bio devices as well as a variety of sensors [1,2]. When applied to metal layers, the metal dewets from the substrate in the areas exposed to intensity maxima where formation of nanoparticles (NPs) and/or metal transport is generally reported [3–6]. The combination of metal NPs that have a plasmonic response with the diffractive properties due to their organisation [7,8] offers several possibilities for sensing [2,9].

Laser interference of 2–4 beams is the most widely used approach that has the advantage to produce patterns with periods smaller than the laser wavelength and has the potential for producing large area patterns (>mm^2) with single pulses. The use of amplitude or phase masks is an alternative approach that is particularly suited for producing large patterned regions with micron or sub-micron periods that has much less been used [10]. The most generally discussed mechanism is mass transport due to thermocapillary or hydrodynamic flow [3]. In spite of a general agreement exists on that the transformation and mechanism depend on thickness, the results reported for similar thicknesses and same metal look different (see Ref. [8] and references therein). The differences appear to be related to different experimental conditions such as laser wavelength, fluence, number of pulses or different beam profile. However, the role of the initial nanostructure of the layer has little been discussed in spite of the importance of the initial layer nanostructure on solid-state dewetting is known since very long [11]. The aim of this work is to show that the initial nanostructure of the metal layer plays an essential role on the features of the periodic patterns produced by laser interference and becomes a key parameter limiting the range of achievable periods.

2. Experimental

Silver layers with three different thicknesses have been prepared at room temperature on fused silica substrates by magnetron sputtering. Two of them, having thickness of 8.3 nm and 10.8 nm, were prepared in an Ar operating pressure of 3×10^{-2} mbar [12]. One layer having an intermediate thickness (9.5 nm) was prepared in an Ar/N_2 mixture having a 3.5% of N_2 at similar operating pressure. The addition of nitrogen into the gas mixture prevents dewetting of the Ag during the deposition process thus making the

* Corresponding author.
E-mail address: rpelaez@io.cfmac.csic.es (R.J. Peláez).

http://dx.doi.org/10.1016/j.apsusc.2015.09.110
0169-4332/

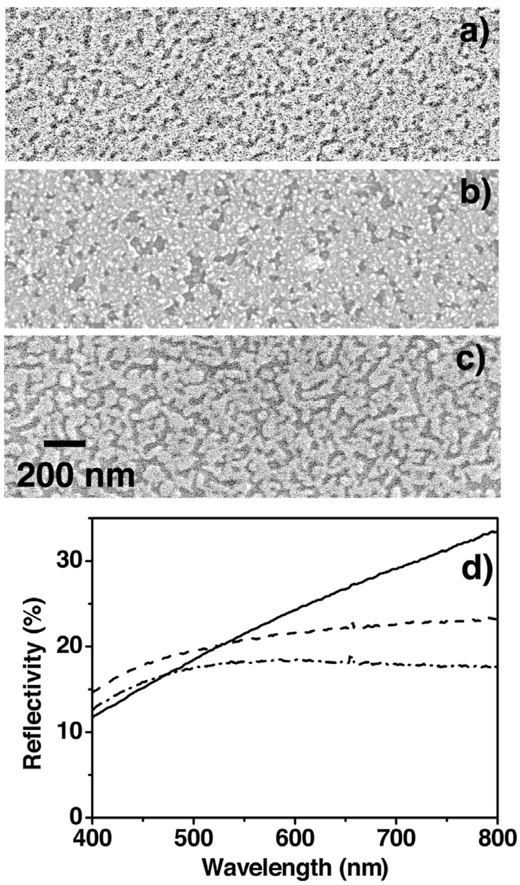

Fig. 1. (a–c) SEM images and (d) reflectivity spectra of the (a) 10.8 nm (dashed line), (b) 9.5 nm (full line) and (c) 8.3 nm (dashed-dot line) thick layers.

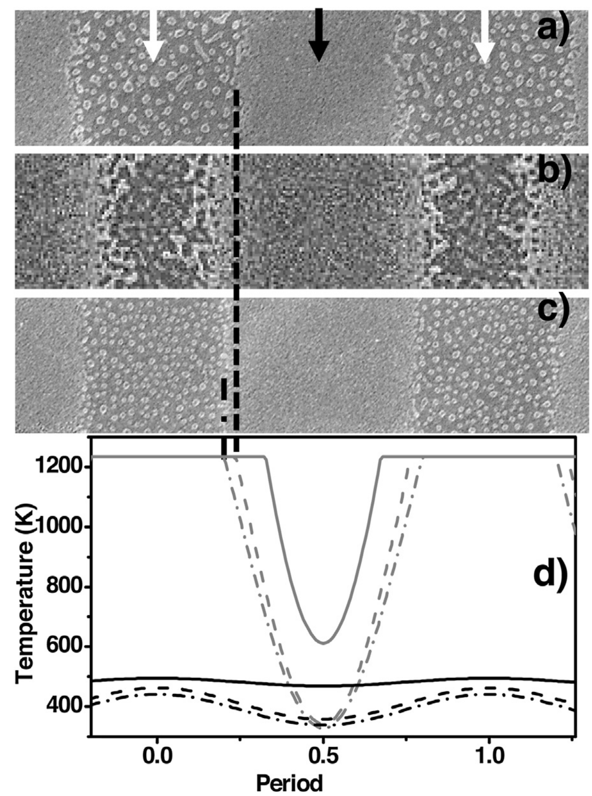

Fig. 2. SEM images of one period of patterns recorded in the (a) 10.8 nm, (b) 9.5 nm and (c) 8.3 nm thick layers showing (white arrows) two transformed regions around intensity maxima sites and (black arrow) a region around intensity minimum site. (d) Temperature profiles calculated (grey lines) at the time at which the extension of the molten region was maximum and (black lines) at 600 ns after the laser pulse maximum for the (dashed lines) 10.8 nm, (full lines) 9.5 nm and (dashed-dot lines) 8.3 nm thick layers. The distance between the two white arrows is the pattern period that is (a, c) 6.3 μm and (b) 6.7 μm.

layers smoother and more continuous than those produced in pure Ar as reported elsewhere [13].

We have used an excimer laser working at 193 nm and delivering 20 ns laser pulses to expose a phase mask manufactured by Laser Lab. Gottingen on SiO_2 plates. It has parallel lines with a period of 20 μm. It has been optimised for high efficiency in the ±1st diffraction order at 193 nm that were forced to interfere at the sample surface. The projection optics leads to fringed patterns of 6.7 μm and 6.3 μm periods in the continuous and discontinuous layers, respectively. The sample surface becomes exposed to a modulated intensity formed by the maxima and minima of interference and the fluence at the sample site becomes modulated between 0 and 2***F*** where ***F*** is the average fluence measured at the sample site. In this work, ***F*** was 90 mJ cm^{-2} and 52 mJ cm^{-2} for the continuous and discontinuous layers respectively. These fluences were selected in order to produce patterns with similar fringe width ratio. Further details can be found elsewhere [7,8,14].

The reflectivity of as-grown layers was measured at nearly normal incidence using a spectrometer USB4000 and a light source DT-MINI-2-GS from Ocean Optics. The experimentally recorded reflectivity spectra from the samples were corrected by the known spectral reflectivity of a polished silicon substrate. Finally, as grown and patterned layer morphology and structure were characterised by field emission scanning electron microscopy (SEM) using a JEOL JSM-7500F microscope.

3. Results

Fig. 1 shows SEM images of the as grown layers where it can be seen that the layers produced in Ar (Fig. 1a and c) are discontinuous although the thickest one (Fig. 1a) is around the percolation limit. The layer produced in an Ar/N_2 mixture (Fig. 1b) is instead continuous but contains some holes. Overall, the metal coverage of the two thicker layers (Fig. 1a and b, 80 ± 5% and 89 ± 2%) is significantly higher than that of the thinnest layer (Fig. 1c, 63 ± 5%). Fig. 1d shows the reflectivity spectra measured in the three studied samples. The one recorded from the 9.5 nm thick sample further confirms its continuous character since it is similar to that of bulk silver [14]. While the spectrum of the 8.3 nm thick layer shows a broad maximum around ≈550 nm that arises from the plasmonic response of the irregular nanostructures, the spectrum of the 10.8 nm thick layer shows similar wavelength dependence for short wavelengths while it remains increasing for wavelengths >600 nm confirming further the layer is around the percolation limit.

Fig. 2 shows SEM images showing one period of the patterns produced in the three studied layers where the centres of the regions around the intensity maxima and minima sites are marked with arrows. The patterns in the discontinuous layers (Fig. 2a and c) are formed by alternate regions whose nanostructure is similar to that of the as grown layers (compare to Fig. 1a and c) and regions in which the layer has broken-up into NPs, each located respectively at intensity minima and maxima sites. Instead, a sequence of regions can be identified in the pattern produced on the continuous layer (Fig. 2b). Starting at the intensity maxima sites (at left white arrow) where the layer breaks into isolated NPs and fingers, a bright contrast region containing holes is first seen when moving right in Fig. 2b. The contrast decreases and the holes become smaller until a darker region is eventually seen around intensity minima sites. The different features are better appreciated in Fig. 3 where magnified