XLVIth Zakopane School of Physics, International Symposium Breaking Frontiers, Zakopane, Poland, May 16–21, 2011

Pulsed Laser Interference Patterning of Metallic Thin Films

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Pulsed laser interference is applied to metallic and semiconductor thin films in the thickness range of 40–100 nm. At intensities which induce local melting we can observe local retraction of the molten material towards the unmolten areas due to dewetting. Thus micropatterning of surface gets feasible. Although this dewetting induced retraction should be a common behaviour of metals on oxide surfaces, two groups of materials can be distinguished. In the first group the former molten areas get completely blank of metal while in the second group a material droplet remains in the center of the molten area. We show that this behaviour can be attributed to a distinctly different way of liquid movement upon local melting.

PACS: 68.55.−a, 68.08.Bc, 81.16.−c

1. Introduction

The patterning of metals has recently gained additional importance due to the growing field of plasmonics. The commonly used electron beam lithography is ideal for experimental purposes where the variation of parameters is of importance for the understanding of the size-dependence of the function of plasmonic devices. For applications less expensive fabrication methods would be mandantory as e.g. laser patterning of surfaces. Nanosecond pulsed laser interference patterning allows preparing microstructured surfaces in a single shot on areas of mm^2. This has been shown in several publications for direct patterning of surfaces [1–8] or for the generation of surface energy patterns [9]. Nevertheless for a controlled manufacturing process the understanding of the involved mechanims is essential.

We show here that in addition to already known dewetting phenomena which can be induced by ns-pulsed laser interference patterning a new regime of liquid movement can be observed upon melting of certain materials which manifests itself in an unexpected material distribution in the treated areas. These findings show the importance of inertia on the movement of liquids on the nanoscale not only for the annealing of nanostructures [10] but also for inhomogeneous illuminated thin films.

2. Experimental

Metallic thin films of different thickness are evaporated onto glass or Si substrates in a thermal evaporation process at a base pressure of 10^{-6} mbar. The evaporated thickness is controlled by a quartz crystal microbalance. In the case of the Au-layer an adhesion layer of 2 nm Cr was additionally evaporated below the Au-layer. The samples are then illuminated by a single laser pulse interference pattern of a frequency doubled Nd:YAG laser of 12 ns pulse duration. After illumination the samples are analysed by optical, electron and scanning force microscopy. We show here results for three and two beam interference patterns at different angles. We do not observe principle differences between the behaviour of the thin films neither on the two different substrates (Si and glass) nor on the period used.

3. Results and discussion

First we show the results of the group of materials that behave similar to former results by other groups [6, 8]. For these materials (Si, Bi, Ge) we found that the dewetting leads to areas which are free of metal (Fig. 1). One can give an easy interpretation of these results. Due to the nonwetting behaviour of those materials the thin films would like to minimize the contact area towards the substrate. Thus upon local melting the retreatment gets feasible starting in the molten areas. In this way empty rings are formed with an elevated rim, which consists of the retracted material. This gets even clearer in patterns which were formed in the slope of the spatial Gaussian intensity distribution of the Nd:YAG laser pulse: the laterally changing intensity can be translated into a time-evolution of the process as higher intensities lead to longer melting times.

There it can be observed (Figs. 2 and 3) that not all of the rings are identically developed. Some of them are smaller, some of them already bigger in size. This implies that the nucleation of a void towards the substrate is involved either at inhomogeneities in the liquid thickness or by homogeneous nucleation. For the subject discussed

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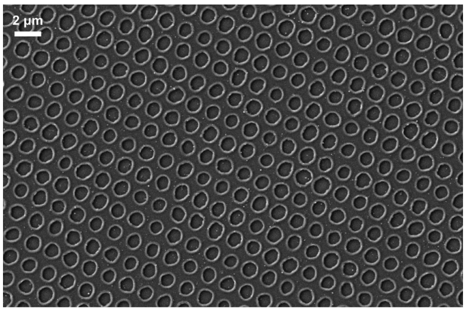

Fig. 1. SEM image of a 40 nm silicon film treated with 3 interfering laser beams.

here the main point is that the empty areas get larger with increasing energy density, respectively melting time.

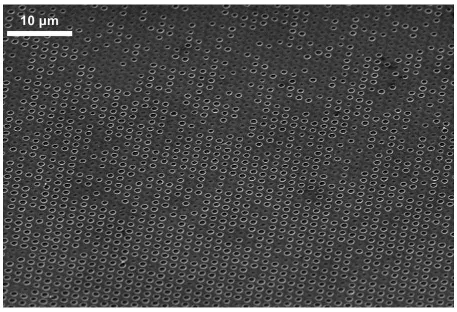

Fig. 2. SEM image of Si sample of Fig. 1. The picture was taken in the gradient of the laser beam with increasing intensity from the upper center towards the lower center.

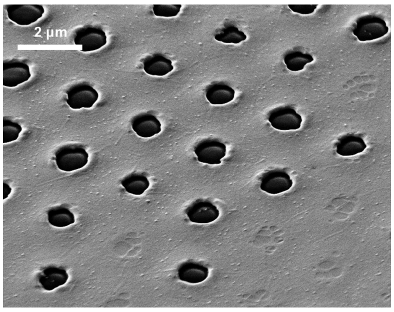

Fig. 3. SEM image of a 70 nm bismuth film treated with 3 interfering laser beams. The picture was taken in the gradient of the laser beam with increasing intensity towards the upper left corner.

This simple picture cannot explain the appearance of several other metal surfaces (Au, Ta, Cu) after annealing with the same interference pattern. Still, an uncovered area with a rim can be observed, but now a metallic particle is found inside (Fig. 4). At first glance one might argue that this process could still be similar, if somehow the contact to a part of the liquid might get lost during retraction. But the appearance at lower intensity shows that a completely different scenario is appearing. First a hollow membrane is formed, which then breaks as can be seen in Fig. 5.

This formation of a liquid membrane gets even clearer after fabrication of a cross section through such a membrane by a focussed ion beam (Fig. 6). Obviously the Ta-film is lifted off the surface for a distance of about 100 nm. Furthermore it can be observed that the membrane is thicker in the center. Thus the chance that the membrane breaks is higher in the thinner connections to the surrounding, which is confirmed by the final particle position which for most of the time is in the center of the finally empty area (Fig. 4).

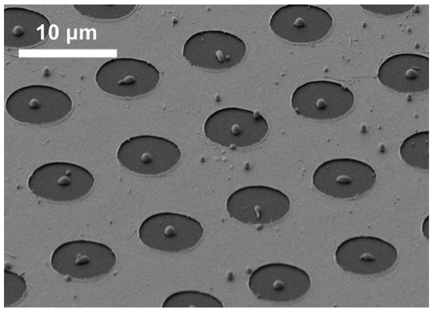

Fig. 4. SEM image of a 50 nm tantalum film irradiated with 3 interfering laser beams after the dewetting process has taken place.

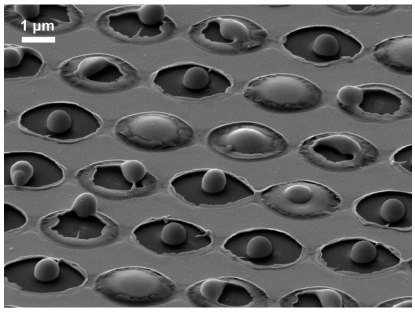

Fig. 5. SEM image of a 50 nm tantalum film irradiated with 3 interfering laser beams. The rupture of the membrane and the subsequent formation of the central metal sphere can be seen.

Similar observation can be made for 2D interference patterns. Here hollow lines are formed as can be seen after focussed ion beam cutting of the surface (Fig. 7). These hollow lines break finally into metal stripes in the center of the emptied area (Fig. 8). One can easily distinguish between the metallic lines formed by dewetting and the stripes which are formed from the hollow lines. While the former are quite straight the latter are subjected to instabilities and therefore appear twisting.

As we pointed out earlier [8] the formation of the membrane is connected to the expansion of the liquid upon melting. If the expansion upon melting cannot be

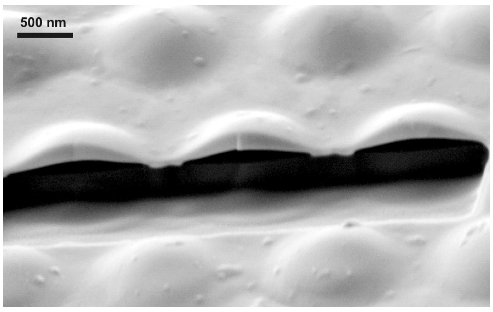

Fig. 6. SEM image of a 100 nm tantalum film irradiated with 3 interfering beams. Here a focused ion beam is used to cut a cross-section through the modulated film.

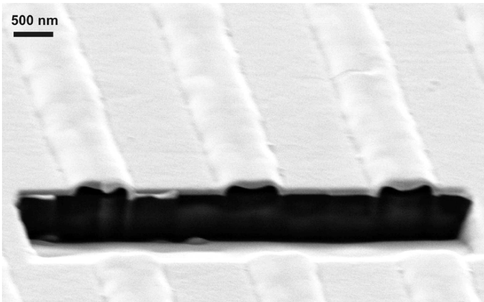

Fig. 7. FIB treated SEM image of a 100 nm tantalum film irradiated with 2 interfering beams.

neglected the metal gains vertical interia upon melting which might lead to detachment from the surface in the case of low adhesion. Additionally the vertical movement of the liquid induces flow towards the center by cohesion, which leads to an increased film thickness in the center. If we now compare the group of materials which show this behaviour with the metals of the first group we clearly find a correlation. While the materials in the first group show a clear density increase from room temperature to the melting point (e.g. Si $+10\%$), the second group shows the opposite behaviour (e.g. Au, -10%). Thus upon melting the center of mass in the second group is accelerated upwards which allows the detachment of a liquid membrane at low adhesion.

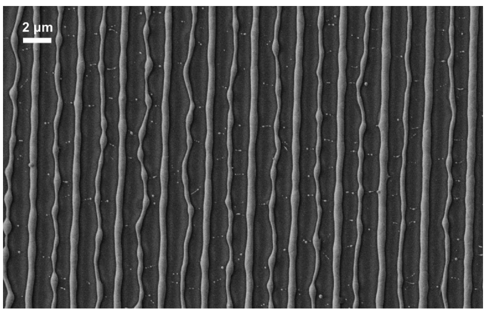

Fig. 8. SEM image of a 50 nm gold film irradiated with 2 interfering beams. The period of the interference pattern was 3.5 μm but at high intensities a bisection of the period can be observed.

4. Summary

In conclusion, we studied the appearance of thin films after illumination with interference pattern of ns-pulsed Nd:YAG laser. While for materials with density increase upon melting, direct dewetting from the heated areas can be observed, another scenario is found in the opposite case. There a liquid membrane can be formed, which finally leads to an incomplete removal of the material from the heated area.

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