Rapid communication

Microdeposition of metal and oxide structures using ultrashort laser pulses


Foundation for Research and Technology-Hellas, Institute of Electronic Structure and Laser, FORTH-IESL, P.O. Box 1527, 71110, Hellas (Fax: +30-81/391318, E-mail: vainos@iesl.forth.gr)

Received: 4 February 1998/Accepted: 9 February 1998

Abstract. Microdeposition of chromium metal and indium oxide microstructures via femtosecond KrF excimer laser (248 nm) ablation in a forward-transfer mode has been studied. The short pulse length, the short absorption length, and the consequently limited thermal diffusion, lower the ablation threshold and enable the deposition of high-definition features. Experiments carried out in a low-vacuum (0.1 Torr) environment result in highly reproducible, well-adhered structures of submicron size. Microdeposition of Cr and polycrystalline In$_2$O$_3$ on glass and silicon substrates is performed. The superior quality of the results allows the direct, one-step fabrication of binary-amplitude and multilevel optical diffractive structures.

PACS: 81.15.F; 07.10.C; 42.40.J

The laser-induced forward transfer (LIFT) technique [1] utilises pulsed lasers to remove thin film material from a transparent support and deposit it onto a suitable substrate. The thin film which is deposited onto a quartz plate is transferred by using a single laser pulse onto the receiving substrate usually placed parallel to the source thin film. The LIFT process was first shown by Bohandy et al. [1, 2] to produce direct writing of 50-µm-wide Cu lines by using single ns excimer laser pulses (193 nm) under high vacuum (10$^{-5}$ mbar). Fogarassy et al. [3, 4] have also reported 100-µm-width patterns of superconducting thin films using ns ArF and Nd:YAG lasers. The deposition of diamond-like carbon films by the LIFT technique using a copper laser and a KrF laser was also reported [5]. Several other studies [6–9] have resulted in direct writing of 20-µm-wide metal lines under air or helium ambient conditions also using ns laser pulses. The interface study of gold and aluminium deposition was also reported [10]. The dynamics of the laser ablation transfer (LAT) of 1-µm-thick coatings effected by near-IR ($\lambda = 1064$ nm) 23 ps laser pulses was studied by Sandy Lee et al. [11] via an optical microscope with ps temporal resolution and showed that the velocity of the ejected material is Mach 0.75 and that the use of ps optical pulses results in a reduction of the laser fluence threshold by one order of magnitude, compared to that of 100-ns-long pulses.

The ability to deposit patterns, spots, and lines, with sub-µm resolution may have applications in microelectronics as well as in the optoelectronics fabrication industries. Artificial texturing and engineering of sensitive parts is an effective approach for overcoming stiction problems in micro-electromechanical system (MEMS) surfaces, thereby improving tribological performance and component lifetime. Conventional methods of surface patterning are chemical vapour deposition (CVD), plasma CVD and sputtering, which by nature have no spatial selectivity.

In this work we demonstrate, for the first time to our knowledge, direct microdeposition of high-quality patterns with sub-µm features. Metal and oxide structures are transferred using fs ultraviolet radiation. The present approach exploits all advantages over conventional methods including simplicity in terms of vacuum handling, deposition purity, position selectivity, and high-accuracy sub-µm pattern transfer.

1 Experimental technique

The materials used as the “target surface” in the microdeposition experiments were thin chromium and indium oxide films deposited on transparent quartz wafers. Cr films of 400, 800, and 2000 Å thickness were prepared by sputtering and e-beam evaporation and were uniform and well adhering. Thin films of In$_2$O$_3$ of 500 Å to 4500 Å in thickness were prepared by reactive pulsed laser deposition [12]. Glass (Corning type 7059) and Si(100) were used as “receiver surfaces”. The distance between the target and the receiver surfaces was variable from near-contact to 1000 µm with a 5-µm accuracy. The target–receiver pair was placed in a miniature vacuum cell, under a pressure of 10$^{-3}$ mbar, driven by a rotary pump. The miniature cell was fixed onto a computer-controlled x−y translation stage, allowing a maximum 25 mm × 25 mm
movement, by means of piezoelectric motors (Burleigh Inchworm type), having a 50 nm positioning resolution determined by optical encoders. Serial writing of metal lines and isolated dots is achieved, as well as the fabrication of complicated computer-generated optical diffractive structures.

A schematic diagram of the excimer laser microdeposition setup is depicted in Fig. 1. The ablation laser was a distributed-feedback dye-laser-based fs excimer laser system (248 nm, 13 mJ pulse energy, 500 fs pulse duration (FWHM), 1–10 pps repetition rate, 30 mm × 10 mm beam size). The laser beam was focused onto the target surface through a high-power image projection micromachining system [13], based on the inverse microscope principle performing mask projection on a large-reduction basis (×30) onto the target. The optical system dispersion is seen to produce negligible pulse broadening. The estimated depth of focus of the system is 2 μm. The spot size of the laser was varied between 1 and 64 μm. The energy density of the laser was varied between 50 mJ/cm² and 550 mJ/cm² on target. During the deposition process the target area was viewed through an imaging system, including a CCD camera and microscope ocular lenses. The surface characterisation of the deposited samples was performed by using optical microscopy and scanning electron microscopy. The stoichiometry of the oxide films was deduced by X-ray diffraction analysis. The film thickness of the pre-coated and the transferred films were measured by surface profilometry (Perphometer) and atomic force microscopy.

2 Experimental results and discussion

The optical absorption in the source film affects the threshold laser fluence above which a single pulse leads to film removal and transfer onto the receiver surface. The absorption depth of Cr at 248 nm is 220 Å, implying that the transmission of the excimer laser light through even the thinner source film (400 Å) is negligible.

Various features spots, lines, and patterns were deposited. Figure 2, shows the scanning electron micrograph of Cr dots deposited on glass by fs laser microdeposition. The target source was 400 Å Cr. The UV-illuminated area was 4 μm × 4 μm and the energy density was 100 mJ/cm² as, because of the laser beam profile, only a portion of the focused laser spot is above the ablation threshold. The adhesion of the deposited patterns was satisfactory as this was tested by the Scotch Tape tests.

Figure 3 shows the deposited feature width of Cr dots as a function of the laser energy fluence using different thickness target sources (400 Å and 800 Å). The forward ablation threshold was defined as the single-pulse energy-density value at which complete thin film material removal enabling transfer in the direction of propagation of the laser beam is performed. This threshold was influenced by the thickness of the source film. No deposition from the 2000-Å-thick Cr source film was obtained by a single excimer laser pulse even at the highest energy density (500 mJ/cm²) available. The forward ablation thresholds of the 800-Å and 400-Å Cr source films were, respectively 150 mJ/cm² (±20%) and 100 mJ/cm² (±20%), whereas the best quality of the de-
posited dots in terms of uniformity was obtained for the thinner Cr source film, thus minimising thermal effects and the extent of the damaged (melted) area.

The spread of the ablated material was also studied by varying the distance between the source and target surfaces from near-contact to 500 µm. Figure 4 shows the deposited feature spread size of Cr lines as a function of the distance for a 4 µm × 4 µm UV-illuminated area of the source Cr film at 156 and 260 mJ/cm² energy fluence.

Complicated diffractive optics such as binary-amplitude computer-generated holograms were fabricated for that purpose, demonstrating the potential of this technique. The microdeposition was done either by serial writing (pixel-by-pixel) of the diffractive pattern or by directly projecting a master hologram mask on the target source film. Figure 5 depicts a hologram pattern produced by the deposition from a 400-Å-thick Cr target source on silicon at an energy density of 339 mJ/cm² having a pixel size of 3 µm × 3 µm. Figure 6 depicts a multilevel computer-generated holographic structure on glass, comprising three layers of Cr selectively deposited on appropriate areas on a glass plate. The energy density was 372 mJ/cm².

It is worth noting here that, for comparison purposes, a series of identical experiments was carried out using the same optical system with a conventional KrF excimer laser emitting pulses of 20 ns duration. The results obtained with the ns excimer laser as a source for ablation did not yield deposition features of comparable quality to the fs laser in terms of morphology and resolution. The re-melting of the transferred material by the trailing part of the laser pulse is often encountered in experiments with ns, and longer, laser pulses. Recent studies [14] have shown that ultrafast laser pulses have precise breakdown thresholds and minimum thermal diffusion, thus offering advantages in precision micromachining and microdeposition applications. Micrometer and submicrometer feature sizes have been achieved in micromachining experiments with fs lasers.

Indium oxide exhibits interesting electrical and optical properties including the demonstration of holographic recording [15]. Indium oxide patterns were also grown in a forward-transfer mode. Figure 7 depicts a hologram pattern produced by the deposition from a 200-nm-thick In₂O₃ target source on glass at an energy density of 150 mJ/cm² having a pixel size of 4 µm × 4 µm. The crystallinity of the deposited features was studied by X-ray diffraction. The X-ray diffraction patterns from the PLD films on quartz substrates shown in Fig. 8a, exhibited discrete diffraction peaks superimposed on a continuous background, indicating the presence of crystalline grains embedded in an amorphous matrix. All the XRD peaks could be assigned to the cubic bixbyte structure of In₂O₃ without any metallic In peaks. The intensity ratio of (004) to (222) peaks was 0.15, which was half the corre-

![Fig. 4. Plot of the deposited feature spread size as a function of the distance between the target film and the receiver surfaces. The energy densities were 156 and 260 mJ/cm²](image)

![Fig. 5. Scanning electron micrograph of a computer-generated holographic pattern produced by Cr microdeposition on silicon (100). The target source was 400 Å Cr. The pattern consists of 64 × 64 pixels with pixel size 3 µm × 3 µm](image)

![Fig. 6. Scanning electron micrograph of a computer-generated multilevel structure of Cr on glass. The pattern pixel size was 5 µm × 5 µm](image)
Fig. 7. Scanning electron micrograph of a computer-generated holographic pattern produced by In$_2$O$_3$ microdeposition of glass. The target source was 200 nm In$_2$O$_3$. The pattern consists of 64 x 64 pixels with pixel size 4 µm x 4 µm.

The X-ray pattern of the as-transferred material on a Corning glass substrate is also shown in Fig. 8b. It should be mentioned that a direct comparison of the peak intensities between indium oxide/quartz and indium oxide/glass systems cannot be facilitated from the spectra in Figs. 8a and 8b, since the two films did not cover the same area. Again, all the diffraction lines could be assigned to the In$_2$O$_3$ phase. In this case the intensity ratio of the (400) peak to the (222) peak was 0.26 indicating that the indium oxide/glass consisted of crystalline grains with almost random orientation. The indium oxide/glass film had a dark appearance suggesting an oxygen deficiency and/or oxygen displacement in the atomic matrix. Annealing in flowing oxygen at 350 °C improved the transparency and the crystalline quality as evidenced from the enhanced diffraction intensities also shown in Fig. 8c.

3 Conclusion

High-quality Cr and In$_2$O$_3$ microdeposition on glass and silicon using a fs KrF excimer laser was achieved. Serial writing of well-defined lines and isolated dots with sub-µm resolution was performed. In this first demonstration of the method, holographic single and multilevel structures are fabricated verifying the promising potential and a prospective application of the presented technique. The microdeposition of indium oxide features indicates the potential of the scheme in the growth of miniature crystalline structures. Investigation is also focused on the fundamental physical mechanisms of ablation, film removal and transfer, and also on the controllable desorption of materials for the formation of 3-D microstructures.

Acknowledgements. This work was supported by the EU Large Installations Plan, “Ultraviolet Laser Facility” at the Foundation for Research and Technology-Hellas and the NATO Collaborative Research Grant # 950146 and Hellenic GSRT via PENED No 754.

References