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Laser induced patterning of Bi thin films

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ABSTRACT

Laser irradiation of Bismuth thin films through a diffractive mask was investigated. The thin films were composed of nano and microcrystals Bismuth with sizes ranging from 20 to 500 nm. Upon laser irradiation (λ=355 nm) the structured illumination field locally modified the material. In the high intensity regions the surface was transformed whilst low intensity areas were left intact. The modification mechanism was melting followed by coalescence of the nanocrystals giving rise to a more uniform structure. The laser irradiated area was characterized by scanning electron microscopy and atomic force microscopy. The patterns were computed by Fresnel diffraction theory and the agreement between the theory and the experiments was very good.

INTRODUCTION

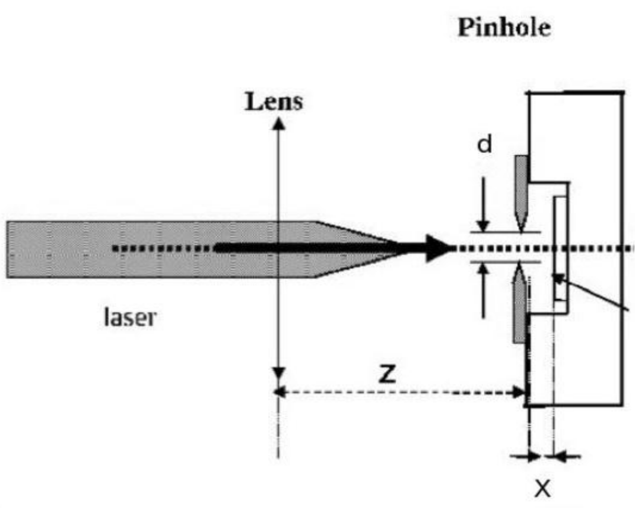
Pulsed or continuous laser irradiation effects on metallic thin films deposited on various types of substrates have been studied extensively. One of the most important potential applications in the early eighties was the laser annealing of ion-implanted semiconductors [1]. Laser processing of materials is also a very active field of study [2]. Furthermore, several effects can be observed depending on the characteristics of the laser used, such as, wavelength, pulse duration, laser energy density etc…[3-4]. More recently, it has also been demonstrated that it is possible, by laser irradiation and the use of masks, to locally modify the size and morphology of nanostructured thin films [5]. This technique has been referred to as the diffraction-assisted method (DAM) [6]. We should also mention that, additional to nanopatterning created by the irradiation of films made up of nano or micro sized particles, it is also possible to obtain ordered micron-sized features on semiconductor surfaces by irradiating them near to the laser ablation regime with a diffractive element [7]. Furthermore, specific micro and nano patterns can be generated on the metal surface using one or a few laser pulses.

In this work we present the results of bismuth patterning. Bi based thin-films have been intensively studied in particular because of their special thermoelectrical properties. Superconductivity and quantum-size phenomena have been also reported [8]. Nano and micro patterning of surfaces has several applications such as for memory devices, data encryption and sensors.

A variety of different techniques have been employed to print patterns on material surfaces, such as nanolithography and bottom-up techniques. Recently, we have shown that it is possible to simultaneously change the morphology and atomic arrangement of quasi percolated silver thin films by pulsed laser irradiation. Different patterns can be designed and created. The procedure to achieve this, is to use first a quasi-percolated thin film and, second to use a diffractive mask on the irradiation path. If no diffraction mask is used, the irradiation process results only in a change of morphology [9]. In this work we show how the morphology in a bismuth thin film changes and a pattern is generated when irradiated through a single slit. Furthermore, using a simple Fresnel model, we can explain the observed results.

PREPARATION AND CHARACTERIZATION OF THE SAMPLES

The bismuth thin films were prepared by thermal deposition on glass substrates and had thicknesses between 500 and 1000 nm. The bismuth thin film were irradiated with 10 pulses of the third harmonic (λ = 355 nm) of a Q-switched Nd:YAG laser, providing pulses of 10 ns of duration. The experimental set-up is shown in Figure 1. In this figure, a TEM microphotograph of the original bismuth morphology film is presented. In order to pattern the incident light field, we have used a slit; the width of the slit was 45 μm and the sample was placed at different distances behind it, and irradiated with different fluences, firstly at 115 μm with 0.8 mJ and secondly at 525 μm with 0.5 mJ respectively. The morphological changes were investigated by scanning electron microscopy (SEM).



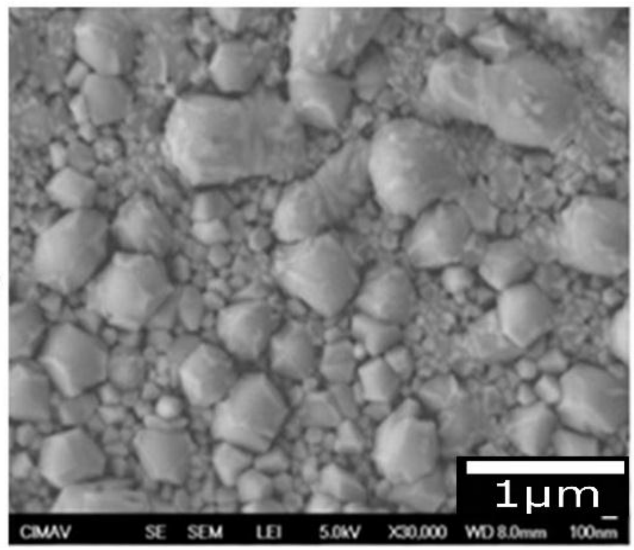


Figure 1. a) left Experimental set-up used to induce the transformations on bismuth thin films. Typical distances; X is varied, Z = 15 cm, d = 45 μm. b) right side, the SEM photograph of the bismuth microcrystals (up to 500 nm) and nanocrystals (down to 10 nm) is presented.

RESULTS AND DISCUSSION

After a single laser shot with a slit having a width of 45 μm, the sample is analyzed using an optical microscope with a 100 X objective. The corresponding optical micrograph is presented in Figure 2. The transformations are evident since multiple parallel bands are clearly distinguishable. The separation and the width of the bands vary. As revealed by SEM outside these bands, the film is composed of the original micro and nano crystals shown in Figure 1. From the results shown in figure 3 one can evaluate the widths of the narrower band and confirm that these are in the nanometer range.

These results are in close correspondence with those reported before [10] on nanostructured silver thin films. However due to the original morphology of the Bi thin films the effects after pulsed laser irradiation are quite different. In the nanostructured silver thin films the bands are composed of nanoparticles having a quasi spherical shape.

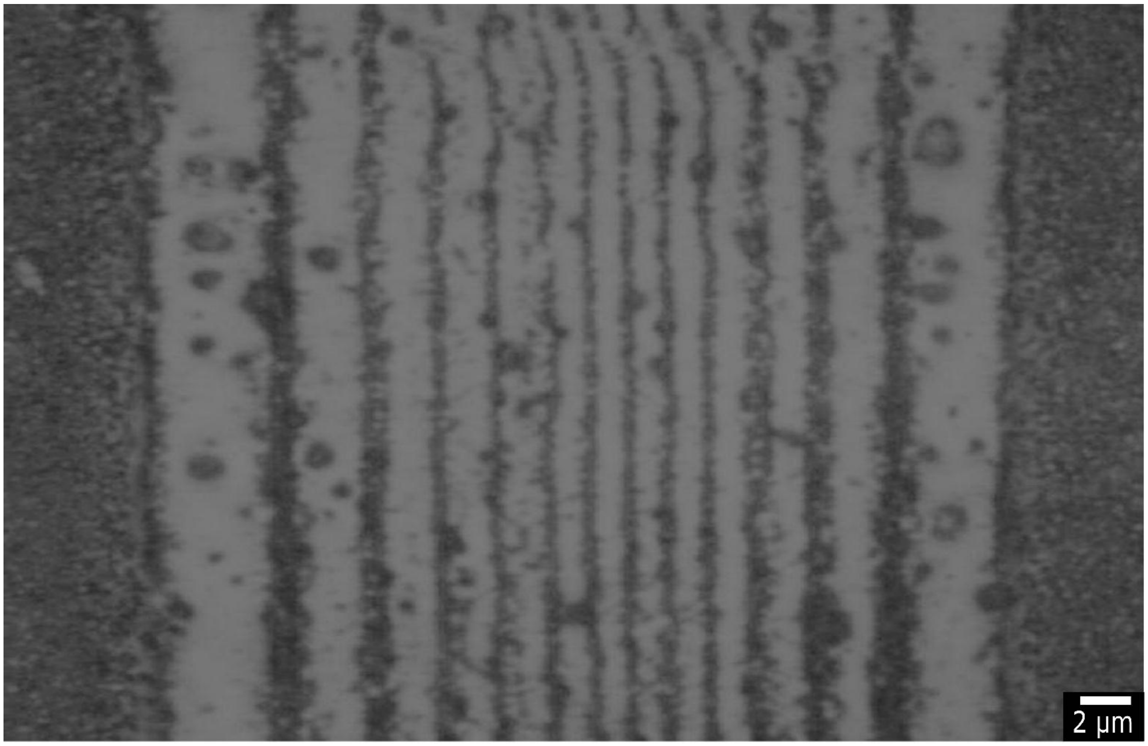


Figure 2. Photograph of the bismuth thin film after UV(355 nm) irradiation through 45µm wide slit at a distance of 115 µm with 10 pulses at a fluence of 0.8 mJ. The bands, where the microcrystals have been transformed to a smooth surface, are clearly observable in light grey.

In the present case, inside the bands which, as we shall see later correspond to the high intensity peaks of the light pattern, the energy is enough to melt the bismuth nano and micro crystals and to generate a much smoother surface this has been confirmed by atomic force microscopy (results not shown here). This type of effect has been observed previously by real time reflectivity measurements [10]. Furthermore we have checked by Raman spectroscopy measurements in the different bands that the material is pure bismuth this indicates that if oxidation occurs only a very thin layer of the material is involved.

In order to analyze the obtained results, we have used a Fresnel model for a single slit. The analysis is equivalent to the one presented in reference [11]. The intensity for a slit of width d in the y direction that is at a distance X from the plane of observation is given by

$$I(Y) = [C_p(Y) - C_q(Y)]^2 + [S_p(Y) - S_q(Y)]^2 \qquad \textbf{(1)}$$

where Y is the horizontal coordinate in the plane of observation and

$$C_q(Y) = \int_0^{q(Y)} \cos(\frac{\pi}{2}\eta^2)d\eta \qquad C_p(Y) = \int_0^{p(Y)} \cos(\frac{\pi}{2}\eta^2)d\eta$$

$$S_q(Y) = \int_0^{q(Y)} \sin(\frac{\pi}{2}\eta^2)d\eta \qquad S_p(Y) = \int_0^{p(Y)} \sin(\frac{\pi}{2}\eta^2)d\eta$$

are the Fresnel integrals with

$$\eta = (Y - y)\sqrt{\frac{2}{\lambda X}}$$

The dependence on y of the integral upper limits is given by

$$q(Y) = (Y + \frac{d}{2})\sqrt{\frac{2}{\lambda X}} \qquad\qquad p(Y) = (Y - \frac{d}{2})\sqrt{\frac{2}{\lambda X}}$$

In Figure 3, the bands are presented together with the intensity I(Y) profile obtained from Equation (1). For this calculation, we have used the following parameters: laser wavelength =355 nm. The distance from the slit to the sample in the first film is X=115 μm and in the second is X= 525 μm.

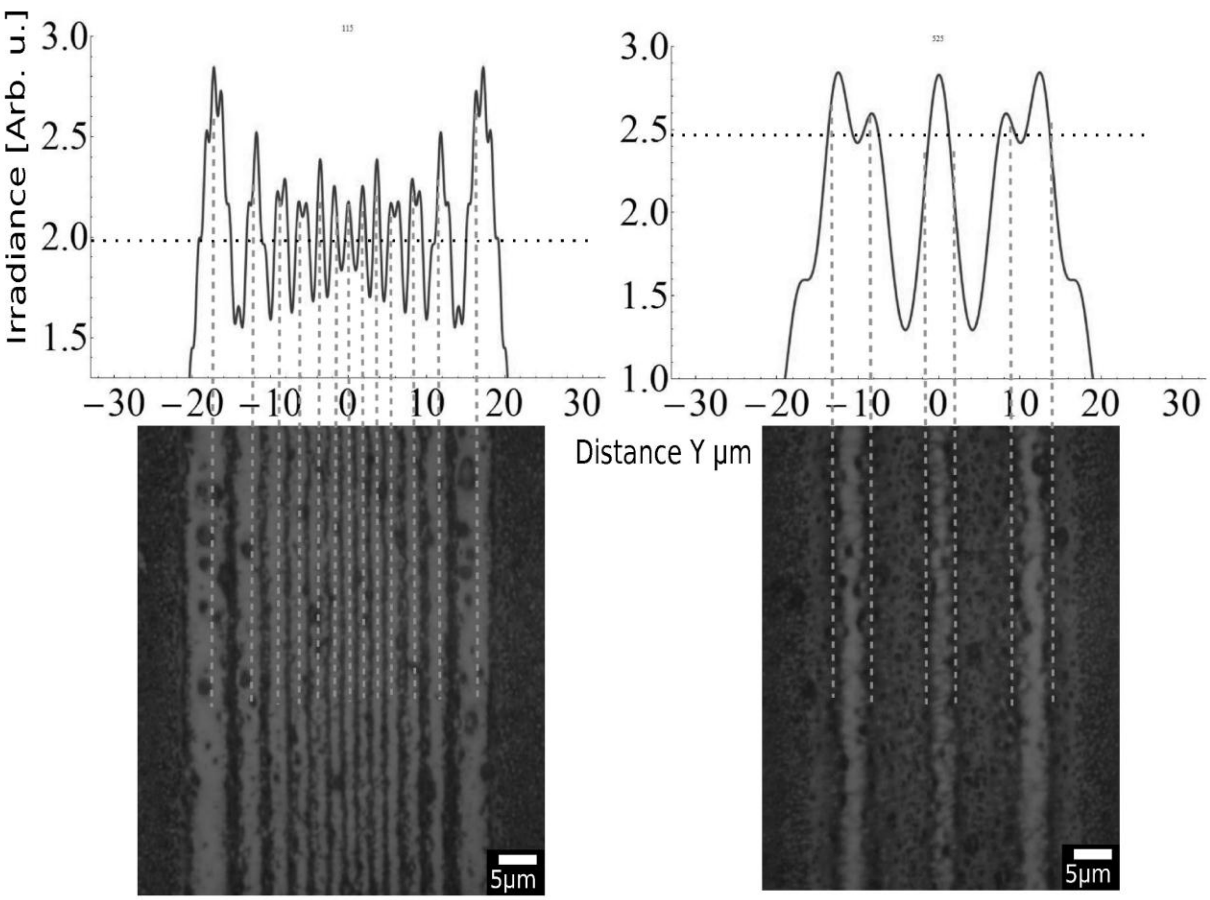


Figure 3. Photograph of the bismuth thin film with the transformed bands. On the left-hand side, the irradiation was made through a slit placed at a distance X=115 μm and formed a diffraction pattern with 13 bands. On the right-hand side, the slit was placed at a distance X=525 μm and formed a pattern with 3 bands. The result of the intensity spatial distribution in the plane of the sample using Fresnel diffraction theory is plotted as a solid curve for both cases and is described in the text.

The dotted horizontal line shown in Figure 3 has been determined using the imprinted pattern in the surface of the sample and is related to the energy density needed to produce the corresponding changes in the morphology of the original Bi thin film. In the regions having an energy density above this irradiance threshold, the Bi nano and micro crystals are melted and coalesce before cooling and generate a smooth surface. In the low irradiance regions just below this threshold, the nano and micro crystals are not affected. As one can observe from figure 3 the agreement between theory and experiment is very good.

CONCLUSIONS

In this work, we have shown that by laser irradiation through a mask it is possible to induce transformations on microstructured bismuth thin films. The imprinted pattern matches closely the spatial intensity distribution obtained by Fresnel diffraction theory. Furthermore the patterning process, which most likely is produced by melting and re-nucleation of the material in the zones of constructive interference of the diffraction pattern, is achieved in a few tenths of nanoseconds.

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